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Van der Waals forces in the context of non-relativistic quantum electrodynamics

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Part I

Introduction, mathematical setup and main results

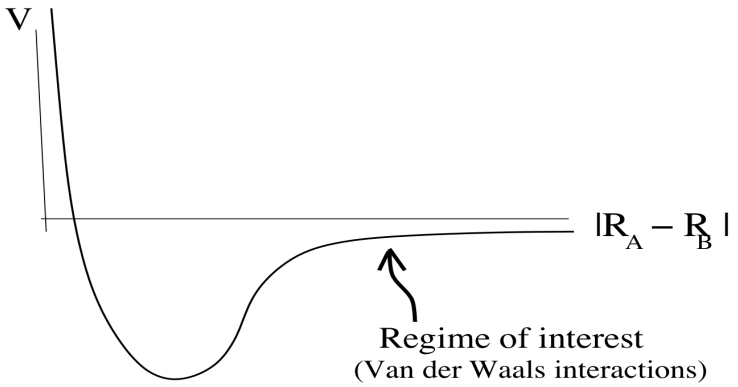
Chapter 1

Introduction

Van der Waals forces between atoms and molecules play a fundamental role in many fields of chemistry, biology, nanotechnology and condensed matter physics (for a survey, see e.g. [KMM09]), [BMM01]). Loosely, the term 'Van der Waals forces' refers to weak long-range forces, as opposed to strong short-range forces such as covalent and ionic bonds.
Van der Waals forces can be grouped into two classes. The first class consists of forces due to interactions of permanent dipoles with induced dipoles. These are sometimes referred to as Debye forces. The second class, which will be in the focus of the present work, contains the so-called dispersion forces. This form of interaction originates purely from fluctuations in the charge distributions and cannot be easily understood in classical terms. It occurs in particular between atoms and molecules without any permanent dipoles in their charge distributions, e.g. noble gases or any systems with spherically symmetric ground state electron distributions. In the case of noble gases, dispersion forces are among the main interactions present between different atoms, and the existence of a liquid phase for these elements is a manifestation of their effect.
Heuristically, dispersion forces are often explained by the following mechanism: the moving electrons at one atom dynamically polarize the charge distribution, thereby creating fluctuating multipole moments which dynamically induce multipole moments in the charge distributions of the other atoms. These moments then act back on the electrons of the first atom, and on average this process leads to an attractive interaction. For more details and further discussion, we refer to [Sto97].
Although as indicated above, dispersion forces constitute only a subclass of the more general Van der Waals forces, we will use the terms 'dispersion forces/interactions' and 'Van der Waals forces/interactions' interchangeably.
In a quantum-theoretical approach to interatomic forces, the mathematical object describing the interaction is the Born-Oppenheimer potential energy surface

$$V(\mathbf{R}) := \inf \operatorname{spec}(H(\mathbf{R})) - \lim_{R\to\infty} [\inf \operatorname{spec}(H(\mathbf{R}))].$$

Here $H(\mathbf{R})$ is a Hamiltonian describing the electrons (and, in a more complete treatment, photons) in the two-atom system with interatomic distance $\mathbf{R} = \mathbf{R}_B - \mathbf{R}_A \in \mathbb{R}^3$. Customarily, the nuclei (at $\mathbf{R}_A$ and $\mathbf{R}_B$) are treated as classical particles appearing parametrically in the Hamiltonian. The large $|\mathbf{R}|$-behaviour of $V(\mathbf{R})$ then captures the Van der Waals forces.



The foundation for a quantum theory of dispersion forces was laid in [EL30]. In this seminal work a Schrödinger operator describing two non-interacting neutral hydrogen atoms with spherically symmetric ground states situated at a distance $\mathbf{R} \in \mathbb{R}^3$ from one another is considered, i.e.

$$H_0 = H_A + H_B = -\frac{\hbar^2}{2m}\Delta_{\mathbf{x}_1} - \frac{e^2}{|\mathbf{x}_1|} - \frac{\hbar^2}{2m}\Delta_{\mathbf{x}_2} - \frac{e^2}{|\mathbf{x}_2|}.$$

Here $\mathbf{x}_1, \mathbf{x}_2 \in \mathbb{R}^3$ are the coordinates of the two electrons, e is the elementary charge, m is the electron mass and $\hbar$ denotes Planck's constant. The operator

$$H' := \frac{e^2}{|\mathbf{R}|^3}\Big((1 - 3\hat{\mathbf{R}} \otimes \hat{\mathbf{R}})\mathbf{x}_1 \cdot \mathbf{x}_2\Big), \tag{1.0.1}$$

which is the lowest-order contribution to the (formally) multipole-expanded interatomic Coulomb interaction

$$\tilde{Q}_R(\mathbf{x}_1, \mathbf{x}_2) := \frac{e^2}{|\mathbf{R}|} + \frac{e^2}{|\mathbf{x}_2 - \mathbf{x}_1 + \mathbf{R}|} - \frac{e^2}{|\mathbf{x}_1 - \mathbf{R}|} - \frac{e^2}{|\mathbf{x}_2 + \mathbf{R}|}, \tag{1.0.2}$$

is treated as a perturbation of the system. Calculation of the matrix element

$$-\langle H'(\Psi_A^0 \otimes \Psi_B^0)|((H_0 - E_0)_{|\{(\Psi_A^0 \otimes \Psi_B^0)\}^\perp})^{-1}|H'(\Psi_A^0 \otimes \Psi_B^0)\rangle \tag{1.0.3}$$

occurring in second-order Rayleigh-Schrödinger perturbation theory then yields the interaction potential

$$V_{VdW}(\mathbf{R}) = -\frac{c_6}{|\mathbf{R}|^6} + \mathcal{O}(1/|\mathbf{R}|^8), \tag{1.0.4}$$

where

$$c_6 := |\mathbf{R}|^6\langle H'(\Psi_A^0 \otimes \Psi_B^0)|((H_0 - E_0)_{|\{\Psi_A^0 \otimes \Psi_B^0\}^\perp})^{-1}|H'(\Psi_A^0 \otimes \Psi_B^0)\rangle.$$

Here Ψ_A^0 and Ψ_B^0 are the ground states of H_A and H_B corresponding to ground state energies E_0^A and E_0^B, respectively, and $E_0 = E_0^A + E_0^B$.
However, there are several mathematical issues regarding this approach (which the authors were actually aware of). Firstly, the perturbation operator H' is not relatively bounded with respect to H_0, and the Hamiltonian $H_0 + H'$, even if it was realized as a self-adjoint

operator, could not be bounded from below, which is a necessary condition for a ground state to exist. Furthermore, the formal perturbation series involving H' diverges. These problems have recently been resolved in [Fri], which contains a rigorous proof of (1.0.4). A crucial ingredient, which in particular justifies the multipole expansion of $\tilde{Q}_R$, is the proof that in the ground state of the full system described by $H_0 + \tilde{Q}_R$, the electrons are exponentially localized at 'their' respective nuclei. In particular, this result states that ionic configurations (in which both electrons are localized near one of the nuclei) are 'exponentially unlikely' as the interatomic distance $R = |\mathbf{R}|$ increases. This assertion is stronger than the standard results about exponential decay of ground state wave functions, since the latter only establish exponential smallness outside regions containing both the nuclei, and thus do not distinguish ionic electron configurations from neutral ones.
Earlier works on the on $1/R^6$-decay of Van der Waals interactions in the mathematics literature include [LT86], in which a universal upper bound proportional to $-1/R^6$ on the interaction potential is deduced using the Rayleigh-Ritz variational principle and carefully chosen test functions, and [MS80], in which it is proved that asymptotically non-degenerate energy levels of $H_0 + \tilde{Q}_R$ possess an asymptotic series expansion in powers of $1/R$. However, the coefficients of that expansion are not identified explicitly.
Heuristically, the theoretically predicted lowest-order ($\sim 1/R^6$)-contribution to the interaction energy between atoms with spherically symmetric ground states is expected to break down in the regime of roughly 100 Bohr radii. This can be attributed to the electromagnetic interaction having a finite speed of propagation, namely the speed of light. Since it takes the information about the motion of the electrons at one atom a finite amount of time to reach another atom, and since at an interatomic separation of around 100 atomic diameters, the time of travel of light between the atoms is the same as the average 'circulation time' of electrons around their nuclei, the correlation between the motion of the electrons at the different nuclei partially breaks down. This mechanism of retardation should effectively lower the strength of the interaction. Note, however, that this heuristic argument does not indicate the strength of the retarded interaction.

The first theoretical investigation of the retarded Van der Waals interaction between atoms and molecules in the physics literature goes back to Casimir and Polder [CP48]. In this work the authors investigate a model of two non-interacting neutral atoms (with nuclear charges Z_A and Z_B) which are coupled to a quantized radiation field. The interatomic electrostatic interaction is modelled by a dipole operator as in (1.0.3).
The Hamiltonians H_A and H_B describing the non-interacting atoms are assumed to have purely discrete spectra and spherically symmetric ground states, and the photon momenta live on a lattice. The latter is achieved by means of a 'box quantization', i.e. by enclosing the electromagnetic field in a finite volume, the size of which is sent to infinity in the course of the calculation, so that sums over photon momenta are replaced by integrals. Treating the coupling to the field and this electrostatic interaction as perturbations of the non-interacting system, the authors employ a method which combines elements of fourth-order Rayleigh-Schrödinger perturbation theory with a calculus dubbed 'Heisenberg's method', which involves first perturbing one atom by the quantized radiation field, and then coupling the resulting vector potential to the second atom. For the difference of the energy corrections for the system with finite interatomic distance R and those for the

infinitely separated atoms, the authors derive the formula

$$\Delta E(R) = -\frac{2}{\pi\hbar c}\sum_{l,m}\int_0^\infty \frac{k_l k_m u^4 du}{(k_l^2+u^2)(k_m^2+u^2)}\frac{e^{-2uR}}{R^2} \tag{1.0.5}$$
$$\times\left\{\left((q_l^x q_m^x)^2+(q_l^y q_m^y)^2\right)\left(1+\frac{1}{uR}+\frac{1}{u^2R^2}\right)^2+4(q_l^z q_m^z)^2\left(\frac{1}{uR}+\frac{1}{u^2R^2}\right)^2\right\},$$

where l, m label the eigenvalues E_l, E_m of the atomic Hamiltonians H_A and H_B, respectively, $k_l = E_l/(c\hbar)$, and

$$q_l^\alpha := e^2\langle\psi_A^0|\sum_{i=1}^{Z_A}\mathbf{x}_i^\alpha|\psi_A^l\rangle, \quad q_m^\alpha := e^2\langle\psi_B^0|\sum_{i=1}^{Z_B}\mathbf{x}_i^\alpha|\psi_B^l\rangle, \quad \alpha\in\{1,2,3\}$$

are matrix elements of the electric dipole operators.
By formal arguments, the R-asymptotics of (1.0.5) are then deduced to be

$$-\frac{23\hbar c}{4\pi}\frac{1}{R^7}\alpha_E^A\alpha_E^B \tag{1.0.6}$$

if $R >> \lambda_l = \frac{2\pi}{k_l}$, $R >> \lambda_m$, and

$$-\frac{1}{R^6}\sum_{l,m}\frac{(q_l^x q_m^x)^2+(q_l^y q_m^y)^2+4(q_l^z q_m^z)^2}{k_l+k_m},$$

if $R << \lambda_l$, $R << \lambda_m$, respectively. α_E^A and α_E^B are the so-called static polarizabilities of the atoms A and B, which are defined by

$$\alpha_E^A := \frac{1}{3}e^2\sum_{\alpha=1}^3\langle\sum_{i=1}^{Z_A}\mathbf{x}_i^\alpha\Psi_A^0|((H_A-E_0)_{|\{\Psi_A^0\otimes\Psi_B^0\}^\perp})^{-1}|\sum_{j=1}^{Z_A}\mathbf{x}_j^\alpha\Psi_A^0\rangle, \tag{1.0.7}$$

and correspondingly for atom B.
At a certain point in the derivation of (1.0.5), an ultraviolet-cutoff is inserted ad hoc into some of the encountered integrals over photon momenta in order to handle divergences, and a number of terms are extracted from residues at finite points in the domain of integration. However, it remains somewhat unclear how and at what point the ultraviolet-cutoff is removed and in what respect the final results depend on its choice. Furthermore, it is not completely apparent whether or not the dipole-approximation (see below for an explanation) is used for the quantized radiation field.
From a mathematical point of view, the Casimir-Polder result, as well as many of the subsequent perturbative approaches in the physics literature (see Section 1.4 below), are questionable in several respects. Firstly, it should be emphasized that any perturbation calculation remains formal unless a suitable underlying operator-theoretic model (e.g. a self-adjoint Hamiltonian with spectral gap, perturbed by a relatively bounded symmetric operator) is specified. Secondly, if an ultraviolet-cutoff is incorporated into the calculations, which, as we will see below, is necessary to realize the corresponding models as semi-bounded, self-adjoint Hamiltonians, this has to be done in a systematic way. In particular, a careful investigation of the final ($R\to\infty$)-asymptotics and its dependence on

the chosen cutoff function has to be carried out. Thirdly, the above-mentioned problems related to replacing the interatomic Coulomb potential by a dipole operator in the Hamiltonian also occur in the model including the radiation field.
The aim of the present work is to reinvestigate the perturbative approach of Casimir and Polder to the derivation of the retarded Van der Waals interaction within a mathematically rigorous context and show how these gaps and loose ends can be accounted for. In particular, we wish to remove as many of the restricting assumptions (e.g. discrete or even finite atomic spectra) as possible.
In recent years the quantum-mechanical description of non-relativistic matter coupled to the quantized radiation field (sometimes termed non-relativistic quantum electrodynamics, or simply NRQED) has been given a rigorous mathematical foundation, in particular by the works of Bach, Fröhlich and Sigal ([BFS98], [BFS99]; see also the references therein for earlier work in this field). One of the most important models considered in these works describes a system of N non-relativistic spinless particles which are coupled to the quantized radiation field via the canonical momentum, and is given by the Hamiltonian

$$H_{\mathrm{PF}} = \sum_{j=1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_j} - \frac{e}{c}\mathbf{A}(\rho, \mathbf{x}_j) \right)^2 + e^2\tilde{Q} + H_f. \tag{1.0.8}$$

Following, e.g. [Spo04], we will call Hamiltonians of this form Pauli-Fierz Hamiltonians in this work, although it should be mentioned that this terminology is not used universally. $\mathbf{A}(\rho, \mathbf{x})$ is the vector potential of the quantized radiation field in Coulomb gauge, $\tilde{Q}$ is a scalar potential (typically a Coulomb potential) describing all interactions involving the nuclei and the electrons, and H_f is the Hamiltonian of the free quantized radiation field. H_{PF} field acts on the space $\mathrm{L}^2_{anti}(\mathbb{R}^{3N}) \otimes \mathcal{F}$, where $\mathcal{F}$ is the bosonic Fock space over the Hilbert space $W = L^2(\mathbb{R}^3) \oplus L^2(\mathbb{R}^3)$, the two summands accounting for the two polarization degrees of freedom of the photons.
Among the fundamental results about H_{PF} (with varying assumptions on $\tilde{Q}$) are self-adjointness on $D(-\Delta) \otimes D(H_f)$, essential self-adjointness on many convenient subspaces, semi-boundedness and the existence of a ground states, see [BFS98], [BFS99], [GLL01] and the references therein; for a proof of self-adjointness for arbitrary values of the coupling strength e, see [Hir02].
Furthermore, many interesting results about the spectrum of H_{PF} have been obtained, in particular concerning the connection between excited states of the corresponding atomic (respectively molecular) Schrödinger operator (not involving the radiation field) and so-called resonances of H_{PF}. The physical fact that photons are massless particles and can thus acquire arbitrarily small amounts of energy manifests itself mathematically in the spectrum of H_f consisting of a single non-degenerate eigenvalue corresponding to the vacuum state of the field, which lies at the bottom of a stretch of continuous spectrum extending from 0 to ∞. Thus the spectrum of the non-interacting Hamiltonian (with the interaction between electrons and photons switched off) consists of the countable set of eigenvalues of the atomic (respectively molecular) Hamiltonian, all of which are embedded into the continuous spectrum. In particular, the non-interacting Hamiltonian is lacking a spectral gap above its ground state energy, prohibiting the 'naive' use of perturbation theory. We will have to deal with this fact, which is related to the well-known infrared problem of quantum electrodynamics, in the course of this work, since we want to extract information from the system using perturbation theory nevertheless. This will be accom-

plished by introducing an additional infrared cutoff into the interaction between electrons and photons, producing a spectral gap, which is then removed in the final results.
Once the interaction is switched on, one expects all the excited levels of the atom (respectively molecule) to become 'dissolved into the continuum', turning into resonances, while only the ground state survives. This picture has been well-established mathematically by now, see e.g. [BFS98], [BFS99], [BFSS99], [FGS08].
Another crucial feature of H_{PF} is that the vector potential contains an ultraviolet-cutoff ρ which suppresses the interaction of the electrons with photons above a certain energy scale Λ. The introduction of the ultraviolet-cutoff serves as a mathematical means for realizing H_{PF} as a semi-bounded, self-adjoint Hamiltonian. So far, this seems to be the only available method accomplishing this.
As we have pointed out above and as we will argue in the review of the physics literature below, so far no perturbative analysis of the retarded Van der Waals interaction which starts out from a well-defined, semi-bounded self-adjoint Hamiltonian containing an ultraviolet-cutoff has been carried out, and it is one of the aims of the present work to fill this gap.
Of course, any property of H_{PF} will a priori depend on the choice of the cutoff ρ, and so far, the $(\Lambda \to \infty)$-behaviour of H_{PF} and related quantities (like for instance its ground state and ground state energy) are not well-understood mathematically (see e.g. [Spo04], Chapter 19). From a physical point of view the dependence on the ultraviolet-cutoff may seem quite unsatisfactory: obviously many measurable phenomena in nature happen exactly the way they do, with unambiguous values of certain parameters and measurement outcomes. On the other hand, one has to bear in mind that the model itself is designed with the limitation to phenomena below certain energy scales in mind. After all, it is this modelling assumption which allows for the non-relativistic description of the electrons in the first place. Taking this into account, there might actually be situations, in particular in the low-energy regime, in which the dependence on the ultraviolet-cutoff turns out to be marginal. In fact, as we will prove in the present work, the (asymptotic) $1/R^7$-coefficient in the interaction potential considered by us actually has a well-defined $(\Lambda \to \infty)$-limit.
The model which will be the starting-point of the considerations in the present work belongs to a subclass of Hamiltonians of the form (1.0.8) and is described by the Hamiltonian

$$H_{\mathrm{QA}} = \sum_{j=1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_j} - \frac{e}{c}\mathbf{A}(\rho, \mathbf{x}_j) \right)^2 + e^2 Q_\psi + H_f. \tag{1.0.9}$$

Its crucial feature, whose importance and physical origin was emphasized by Spohn ([Spo04]), is that instead of a general scalar potential $\tilde{Q}$ as in (1.0.8), it contains the 'smeared' Coulomb potential Q_ψ, which is obtained by convolving the electrostatic charge density of an array of point charges with a form factor ψ (see below for the definition). The reason for this choice is the observation that the classical system of the (point-like) particles and the radiation field, considered as a joint dynamical system, constitutes an ill-posed initial value problem for a system of ODEs, which is due to singularities of the evolution equations on the trajectory of the charged particles. This problem can be circumvented by using smeared charge densities instead, which leads to a model sometimes called the Abraham model. The quantization of this regularized classical system then yields (1.0.9).
A natural additional structure, which arises from first regularizing the classical system and then carrying out the quantization, is that that the form factor ψ and the ultraviolet-cutoff

function ρ occurring in H_{QA} are coupled by the relation

$$\rho = \widehat{\psi}, \tag{1.0.10}$$

the hat denoting the Fourier transform on $\mathbb{R}^3$ (see below for the normalization convention used by us). For an introduction to and an extensive discussion of both the regularized classical dynamical system and H_{QA}, we refer to [Spo04].
The relationship (1.0.10), while originally coming from physical arguments, has interesting mathematical implications for the perturbative analysis of the Van der Waals interaction we will carry out in this work. In fact, it has been one of the reasons why we decided to use H_{QA} as a starting point instead of the Hamiltonian (1.0.8). Firstly, by the behaviour of the Fourier transform under dilations, (1.0.8) automatically couples the spatial scale describing the extension of the charge density and the energy scale at which the ultraviolet-cutoff comes into effect. This fact will turn out to be extremely useful when considering the large-R-asymptotics of Fourier integrals of the form (1.4.2) (but with the ultraviolet-cutoff present), since (1.0.8) then automatically provides a dual and simpler picture of the investigated sequence of integrands in real space. In particular, it will lead to a convergent ultraviolet behaviour of the $1/R^7$-interaction coefficient.
Secondly, as it will turn out, the fact that the function ρ appears not only in the field operators, but also - via its Fourier transform - in the electrostatic potential, is crucial for the different contributions to the energy corrections to be comparable while the interaction with the radiation field is still cut off at a finite momentum: in the 'non-smeared' electrostatic Coulomb potential Q, the limit of an infinitely large ultraviolet-cutoff (i.e. an infinitely small amount of smearing) has already been taken in a sense, while the scale of the ultraviolet-cutoff in the vector potential $\mathbf{A}(\rho, \mathbf{x})$ is still finite. In particular, as will be discussed in Section 6.6, the (asymptotic) cancellation of the 'London term' (1.0.3) by contributions involving the field operators, which is crucial for the emergence of the asymptotic $1/R^7$ behaviour, breaks down if Q instead of Q_ψ is used in the Hamiltonian, see the discussion in that section.
It is clear why this subtlety did not show up in the physics literature reviewed above (see Section 1.4 for more details): if one omits the ultraviolet-cutoff altogether, then, although this renders the underlying Hamiltonian meaningless, on a formal level the contributions from the electrostatic and the field interactions match and do not cause discrepancies in the calculation.
In the remainder of this introduction we

- Describe the mathematical setup of the present work in more detail (Section 1.1)
- State the main results (Section 1.2)
- Explain the strategy of their proofs (Section 1.3)
- Continue the discussion of the physics literature on the subject (Section 1.4)
- Summarize the history and status quo of experiments measuring dispersion forces (Section 1.5)

1.1 Outline I: Mathematical setup

In Chapter 2 we introduce the model used in this work. The starting point will be to consider Hamiltonians of the form (1.0.9) for three different physical systems: the first one, denoted by $H_{\mathrm{QA}}(\mathbf{R})$, describes a molecule consisting of two neutral atoms with nuclear charges Z_A and Z_B, respectively, with the nuclei located at the points 0 and $\mathbf{R} \in \mathbb{R}^3$.
The other two Hamiltonians H^A_{QA} and H^B_{QA} describe two individual atoms A and B, located at 0 and $\mathbf{R}$, respectively, each of which is coupled to the quantized radiation field. This corresponds to the situation of the two atoms in the molecule being infinitely separated.
We then employ two common approximations: firstly, we use a dipole-approximated vector potential. This approximation amounts to 'pinning down' the vector potential $\mathbf{A}(\rho, \mathbf{x})$ at the nuclei by letting the first Z_A electrons interact with the radiation field at the point 0, and the remaining one with the field at $\mathbf{R}$. This approximation will turn out to greatly simplify the perturbation calculations later on.
Secondly, for the two-atom system described by $H_{\mathrm{QA}}(\mathbf{R})$, we neglect antisymmetry of the electrons of atom A and B, retaining only the antisymmetry of the first Z_A and the last Z_B electrons among each other. Mathematically, this is implemented by choosing $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}$ as an underlying Hilbert space. The motivation for these alterations is discussed in Section 2.2.
The two modifications result in three new Hamiltonians $H(\mathbf{R})$, H^A, and H^B, and the main goal of the present work is is to understand the long-range behaviour of the Born-Oppenheimer potential energy surface for the model just introduced. This interaction potential is defined as the difference between the ground state energy

$$E(\mathbf{R}) := \inf \mathrm{spec}(H(\mathbf{R}))$$

of the system consisting of the two atoms A and B at a finite distance $R := |\mathbf{R}|$ from each other, interacting via Coulomb interaction and the radiation field, and the limit of this ground state energy as the atoms are moved infinitely apart. In mathematical terms,

$$\tilde{V}(\mathbf{R}) := E(\mathbf{R}) - \lim_{R\to\infty} E(\mathbf{R}). \tag{1.1.1}$$

It is strongly conjectured (see [MS09]) that

$$\lim_{R\to\infty} E(\mathbf{R}) = E_A + E_B,$$

where $E_A = \inf \mathrm{spec}(H^A)$, and analogously for E_B. The analogous result in the Schrödinger case, i.e. without the radiation field, was proved by Morgan and Simon ([MS80]) and can be viewed as a clamped-nuclei variant of the HVZ theorem.
However, since we would like to apply perturbative methods to the analysis of $\tilde{V}(\mathbf{R})$, which a priori are not available due to the lack of spectral gaps of the operators involved in $\tilde{V}(\mathbf{R})$, we will consider an infrared-regularized version of (1.1.1), see below.
In Section 2.3 we implement this infrared regularization for the Hamiltonians, $H(\mathbf{R})$, H^A and H^B, which will endow them with spectral gaps and thus make perturbation theory applicable. There are different possibilities available for doing so, e.g. by introducing a photon mass, which creates a gap at the bottom of the spectrum of the free field Hamiltonian H_f, or by constraining the photons to a momentum lattice, rendering the spectrum of H_f discrete and creating a gap at its bottom. The method of choice for the present

work is to use an infrared-cutoff (parametrized by a small number $\sigma > 0$) in the coupling function occurring in $\mathbf{A}(\rho, \mathbf{x})$. As we will see below, this amounts to the introduction of new coordinates in photon momentum space, distinguishing between low- and high-energy contributions to photon wave functions, and induces a splitting of the photon Fock space into a tensor product $\mathcal{F}_\sigma \otimes \tilde{\mathcal{F}}_\sigma$. The corresponding decompositions of the Hamiltonians each consist of a part which acts trivially, and a new Hamiltonian (denoted by $H_\sigma(\mathbf{R})$, H_σ^A and H_σ^B, respectively) of the form 1.0.9, which has a finite spectral gap above its ground state energy and which acts on a Fock space built from the restrained single-photon momentum space $L^2(\mathbb{R}^3 \setminus B_{\sigma/c}(0)) \oplus L^2(\mathbb{R}^3 \setminus B_{\sigma/c}(0))$. It is these latter Hamiltonians that will be further analyzed. An important remark is that the infrared regularization is well-controlled in the sense that the ground state energies of the regularized Hamiltonians converge to those of the original ones as the regularization parameter tends to zero, see Lemma 2.3.1.

In Section 2.4 we set up the perturbation-theoretic framework for the analysis of the ground state energies of $H_\sigma(\mathbf{R})$, H_σ^A and H_σ^B. It turns out that these three operators can be rewritten as

$$H_\sigma(e, \mathbf{R}) = H_0^\sigma + eH'_\sigma + e^2 H''_\sigma, \tag{1.1.2}$$

$$H_\sigma^A(e) = (H_A + H_{f\geq\sigma}) + eH'_{\sigma,A} + e^2 H''_{\sigma,A}, \tag{1.1.3}$$

$$H_\sigma^B(e) = (H_B + H_{f\geq\sigma}) + eH'_{\sigma,B} + e^2 H''_{\sigma,B}, \tag{1.1.4}$$

where $H_0^\sigma = H_A + H_B + H_{f\geq\sigma}$ describes the non-interacting system of the two atoms and the radiation field, H_A and H_B are the Hamiltonians of the two atoms A and B, $H_{f\geq\sigma}$ is the infrared-regularized Hamiltonian of the free field, and the remaining operators contain the interactions between the electrons and the field, as well as the interatomic Coulomb interaction (in the case of H''_σ).

Although H_0^σ, $H_A + H_{f\geq\sigma}$ and $H_B + H_{f\geq\sigma}$ also depend on the electronic charge e via the atomic Coulomb potentials, we suppress this dependence and keep the (true, non-zero) value of e fixed for these operators. This amounts to treating e as an independent mathematical parameter and will lead to expansions of ground states and ground state energies which are partial expansions with respect to this parameter, with coefficients that still depend on the physical value of the electronic charge entering H_0^σ, $H_A + H_{f\geq\sigma}$ and $H_B + H_{f\geq\sigma}$. In light of this, each of (1.1.2), (1.1.3), (1.1.4) is a quadratic operator family parametrized by the coupling strength e. See Section 2.4 for more details on this concept.

We establish that the operators H_A and H_B converge to their Schrödinger counterparts $\tilde{H}_A$ and $\tilde{H}_B$ (containing the non-smeared Coulomb potential) in norm resolvent sense as the ultraviolet-cutoff parameter Λ tends to infinity, and assume non-degeneracy of the ground states of the latter Hamiltonians. If follows that for large enough values of Λ the assumption of non-degeneracy carries over to the ground states Ψ_A^0 and Ψ_B^0 of H_A and H_B, which exist by standard results on atomic Schrödinger operators, see Proposition 2.5.1.

In Section 2.6 we prove that the unperturbed operators H_0^σ, $H_A + H_{f\geq\sigma}$, and $H_B + H_{f\geq\sigma}$, viewed as operators on the Hilbert spaces $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}$, $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes \mathcal{F}$ and $L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}$, respectively (recall the above remarks about neglecting 'interatomic' antisymmetry), have self-adjoint realizations, which in turn possess the non-degenerate ground states $\Psi_A^0 \otimes \Psi_B^0 \otimes \Omega$, $\Psi_A^0 \otimes \Omega$ and $\Psi_B^0 \otimes \Omega$, respectively. Here Ω denotes the vacuum vector of Fock space.

The corresponding ground state energies are $E_0 = E_A^0 + E_B^0$, E_A^0 and E_B^0, where E_A^0 and E_A^0

are the ground state energies of H_A and H_B. Due to the above infrared regularization, the operators H_0^σ, $H_A + H_{f\geq\sigma}$ and $H_B + H_{f\geq\sigma}$ have spectral gaps of sizes $\min\{\Delta_A, \Delta_B, \hbar\sigma\}$, $\min\{\Delta_A, \hbar\sigma\}$ and $\min\{\Delta_B, \hbar\sigma\}$, respectively, above their ground state energies. Here Δ_A and Δ_B are the spectral gaps of H_A and H_B, which are independent of σ. Under suitable assumptions on the ultraviolet-cutoff function ρ the above families are found to be analytic of type (A) in the sense of Kato, so that standard results of analytic perturbation theory imply that for small enough values of the coupling constant e, the operators $H_\sigma^A(e)$, $H_\sigma^B(e)$ and $H_\sigma(e, \mathbf{R})$ have non-degenerate ground states, with the corresponding energy levels given by the series expansions

$$E_A^\sigma(e) = \sum_{i=0}^{\infty} e^i \, E_{i,A}^\sigma, \quad E_B^\sigma(e) = \sum_{i=0}^{\infty} e^i \, E_{i,B}^\sigma, \tag{1.1.5}$$

$$E^\sigma(e, \mathbf{R}) = \sum_{i=0}^{\infty} e^i \, E_i^\sigma(\mathbf{R}), \tag{1.1.6}$$

see Lemma 2.7.1. As discussed above, these expansions are partial with respect to the parameter e, in the sense that the coefficients $E_{i,A}^\sigma$, $E_{i,B}^\sigma$ and $E_i^\sigma(\mathbf{R})$ still depend on the physical value of e via the Coulomb potentials in the atomic Hamiltonians H_A and H_B. Note that the radii of convergence of these series depend on the size of the infrared-cutoff parameter σ, and decrease to zero as $\sigma \to 0$. However, this issue does not pose a problem because we will work with a simplified model (see (1.2.1) below) which will turn out to have a well-defined $(\sigma \to 0)$-limit. See also Remark 2.7.2.
In Section 2.8 the regularized interaction potential

$$V^\sigma(e, \Lambda, \mathbf{R}) := E^\sigma(e, \mathbf{R}) - (E_A^\sigma(e) + E_B^\sigma(e))$$

is defined. Here we have stressed its dependence on the ultraviolet-cutoff parameter Λ, which it inherits from the operators $H_\sigma^A(e)$, $H_\sigma^B(e)$ and $H_\sigma(e, \mathbf{R})$. It is established that by (1.1.5) and (1.1.6), $V^\sigma(e, \Lambda, \mathbf{R})$ has the series expansion

$$V^\sigma(e, \Lambda, \mathbf{R}) = \sum_{i=1}^{\infty} e^i \left(E_i^\sigma(\mathbf{R}) - (E_{i,A}^\sigma + E_{i,B}^\sigma) \right) =: \sum_{i=1}^{\infty} e^i \, V_i^\sigma(\Lambda, \mathbf{R}). \tag{1.1.7}$$

In particular, all partial derivatives with respect to e at $(e = 0)$ exist, and

$$\frac{\partial^i}{\partial e^i} V^\sigma(0, \Lambda, \mathbf{R}) = (i!) V_i^\sigma(\Lambda, \mathbf{R}).$$

1.2 Statement of the main results

We are now in a position to state the main results of the present work, which concern the first four coefficients in the above series expansion. The first result is the assertion that these coefficients have a well-defined limit as the infrared regularization is removed.

Theorem 1.2.1. *Assume (A1) and (A2) (see below) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Then for $i = 1, 2, 3, 4$,*

$$V_i(\Lambda, \mathbf{R}) := \lim_{\sigma \to 0} \left(\frac{1}{i!} \frac{\partial^i}{\partial e^i} V^\sigma(0, \Lambda, \mathbf{R}) \right) = \lim_{\sigma \to 0} \left(V_i^\sigma(\Lambda, \mathbf{R}) \right)$$

exists.

The following two theorems concern the large-R-asymptotics of the interaction potential

$$V(\Lambda,\mathbf{R}) := \sum_{i=1}^{4} e^i V_i(\Lambda,\mathbf{R}) = \sum_{i=1}^{4} \frac{e^i}{i!}\left(\lim_{\sigma\to 0}\left(\frac{\partial^i}{\partial e^i}V^\sigma(0,\Lambda,\mathbf{R})\right)\right), \tag{1.2.1}$$

which will serve as an approximate model for the 'true' interaction potential $\tilde{V}(\mathbf{R})$ from (1.1.1).

Theorem 1.2.2 ($1/R^7$-law for ultraviolet-cutoff system)**.** *Assume (A1) and (A2) (see below) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Then*

$$\lim_{R\to\infty}\Big(R^k V(\Lambda,\mathbf{R})\Big) = 0$$

for any $0 \leq k < 7$, and

$$c_7(\Lambda) := \lim_{R\to\infty}\big(R^7 V(\Lambda,\mathbf{R})\big) = -e^4\frac{23}{2}(2\pi)^{-3}\frac{\hbar c}{9}\alpha_E^A(0)\alpha_E^B(0), \tag{1.2.2}$$

where

$$\alpha_E^A(\mathbf{k}) = \Big\langle\sum_{i=1}^{Z_A}\mathbf{x}_i\Psi_A^0\Big|\Big((H_A - E_A^0 + \hbar\omega(\mathbf{k}))_{\{\Psi_A^0\}^\perp}\Big)^{-1}\Big|\langle\sum_{j=1}^{Z_A}\mathbf{x}_j\Psi_A^0\Big\rangle,$$
$$\alpha_E^B(\mathbf{k}) = \Big\langle\sum_{i=1}^{Z_B}\mathbf{x}_i\Psi_B^0\Big|\Big((H_B - E_B^0 + \hbar\omega(\mathbf{k}))_{\{\Psi_B^0\}^\perp}\Big)^{-1}\Big|\langle\sum_{j=1}^{Z_B}\mathbf{x}_j\Psi_B^0\Big\rangle$$

are the dynamic polarizabilities of the systems described by H_A and H_B.

Note that the interaction coefficient $c_7(\Lambda)$ still depends on the ultraviolet-cutoff via $\alpha_E^A(0)$ and $\alpha_E^B(0)$: their definition involves the operators H_A and H_B, their ground states and their lowest eigenvalues, all of which are Λ-dependent via the smeared Coulomb potential. The third main result states that $c_7(\Lambda)$ has a well-defined limit as the ultraviolet-cutoff is removed.

Theorem 1.2.3 (Ultraviolet convergence and universality of $1/R^7$-law)**.** *Assume the hypotheses of Theorem 2.8.4. Then $c_7(\Lambda)$ has a well-defined limit as the ultraviolet-cutoff Λ is removed, which is given by*

$$\lim_{\Lambda\to\infty} c_7(\Lambda) = -e^4\frac{23}{2}(2\pi)^{-3}\frac{\hbar c}{9}\tilde{\alpha}_E^A(0)\tilde{\alpha}_E^B(0), \tag{1.2.3}$$

where $\tilde{\alpha}_E^A(\mathbf{k})$ and $\tilde{\alpha}_E^B(\mathbf{k})$ are the dynamic polarizabilities of the corresponding atomic Schrödinger operators $\tilde{H}_A$ and $\tilde{H}_B$ incorporating non-smeared Coulomb potentials, see (2.5.3).

Remarks:
i) (1.2.3) reproduces the formula (1.0.6) found by Casimir and Polder ([CP48]), the only difference being the additional factor $1/(2\pi)^2$. This factor is due to our usage of units, in which the vacuum permittivity ε_0 is set equal to one, as opposed to the convention $4\pi\varepsilon_0 = 1$ used in [CP48].

ii) The connection between the approximate model $V(\Lambda, \mathbf{R})$ and the full model $\tilde{V}(\mathbf{R})$ is not entirely clear: although we will show that

$$\lim_{\sigma \to 0} V^\sigma(e, \Lambda, \mathbf{R}) = \tilde{V}(\mathbf{R})$$

for e in an interval $[0, e_0]$ (see Lemma 2.3.1), it is an open problem whether every individual term in the series (1.1.7) has a well-defined ($\sigma \to 0$)-limit, let alone whether $\tilde{V}(\mathbf{R})$ is four (or more) times differentiable, with i-th derivative equalling $V_i(\Lambda, \mathbf{R})$. Note that if this was the case, then the $V_i(\Lambda, \mathbf{R})$ would correspond to the coefficients of the Taylor series of $\tilde{V}(\mathbf{R})$ at zero. However, recent results by Bach, Fröhlich and Pizzo ([BFP09], on asymptotic expansions of the ground state energy of Pauli-Fierz Hamiltonians in terms of the fine structure constant $\alpha = e^2/(\hbar c)$) and by Griesemer and Hasler ([GH09], on analytic expansions of the same quantity with respect to $\alpha^{3/2}$ when the dipole approximation is used) suggest that the limiting objects $V_i(\Lambda, \mathbf{R}) = \lim_{\sigma \to 0} (V_i^\sigma(\Lambda, \mathbf{R}))$ considered by us may have a more than formal significance even for an expansion of $\tilde{V}(\mathbf{R})$ up to arbitrary order in e.
iii) As will become clear in the proofs of the main results, in principle the method for analyzing the interaction coefficients $V_i(\Lambda, \mathbf{R})$ presented in this work could be carried out up to any desired (finite) order of e in (1.1.7). But as Theorems 1.2.2 and 1.2.3 show, the cancellation of the $1/R^6$-term corresponding to (1.0.3) by contributions caused by the radiation field, and thus the emergence of the $1/R^7$-behaviour at long range, already occurs within the first four terms.

1.3 Outline II: Strategy of the proofs

Part II of this thesis is devoted to the proof of Theorems 1.2.1, 1.2.2 and 1.2.3.
In Chapter 4 we prove Theorem 3.0.6, which states that $\sum_{i=1}^4 e^i V_i^\sigma(\Lambda, \mathbf{R})$ can be converted into a sum of terms with a structure that makes them a lot more suitable for the following investigation of the large $\mathbf{R}$-asymptotics. The proof uses the following ingredients: First of all, explicit formulas for the energy corrections up to fourth order (in e) in terms of matrix elements involving the reduced resolvent T^σ of H_0^σ are derived in Section 4.1. After an extensive analysis of the properties of T^σ in Section 4.2, it is shown that $\sum_{i=1}^4 e^i V_i^\sigma(\Lambda, \mathbf{R})$ further simplifies due the general structure of the perturbation problem and the fact that many terms occur both in the case of finite and infinite interatomic separation, see also Section 4.3. The remaining terms can then be grouped according to whether they are generated by both the interaction with the field and the interatomic Coulomb interaction, or whether they originate purely from either of them.
In Section 4.4 the terms containing only the field interaction are processed further by converting them into integrals over the photon momenta. This is done using invariance

properties and fiber decompositions of the reduced resolvent T^σ, commutator and other operator identities as well as symmetries involving the polarization vectors $\mathbf{e}(\mathbf{k}, \lambda)$ of the photons. Furthermore, a number of $\mathbf{R}$-dependent as well as some $\mathbf{R}$-independent terms are shown to cancel in the course of the calculation. This observation was partially inspired by the formal manipulations carried out in the physics literature, see Section 1.4 below.
A guiding principle in grouping and comparing the terms encountered during the processing is the homogeneity of (parts of) the integrands that occur. As mentioned above, the converted terms have the structure of Fourier integrals, and thus - motivated by well-known properties of the distributional Fourier transform - homogeneity can serve as a first hint towards which terms will contribute to which power of $1/|\mathbf{R}|$ in the final result. Following this idea, we group the terms originating purely from the field interaction into three terms denoted by $F_6(\mathbf{R}, \sigma)$, $F_7(\mathbf{R}, \sigma)$, $F_8(\mathbf{R}, \sigma)$. As is already suggested by the notation, we expect these terms to decay asymptotically as $1/|\mathbf{R}|^6$, $1/|\mathbf{R}|^7$ and $1/|\mathbf{R}|^k$ (with $k \geq 8$), respectively, after the infrared-cutoff σ is removed.

Chapter 5 deals with the terms in $\sum_{i=1}^4 e^i V_i^\sigma(\Lambda, \mathbf{R})$ which contain the smeared interatomic Coulomb interaction. An important tool for the analysis of these terms is the multipole expansion of the interatomic Coulomb potential Q_R, which is introduced and investigated in Section 5.1. In particular, we derive tail estimates for the series expansion on bounded regions of configuration space and give an estimate for the 'exterior' contribution to expectation values of Q_R on exponentially decaying functions. These estimates are essential in making the arguments used in the physics literature rigorous. As pointed out in the above discussion and in Section 1.4 below, the respective authors incorporate the lowest-order term of the multipole expansion (the 'dipole operator') into the Hamiltonian as a perturbation. Although this formally leads to the same lowest-order (in $1/|\mathbf{R}|$) matrix elements in the energy corrections, this method is flawed, since the resulting expression does not define a semi-bounded Hamiltonian. However, following our approach, one can exploit the exponential localization of the ground state eigenfunctions to rigorously carry out the multipole expansion within the relevant matrix elements.
The results of Section 5.1 are applied to the terms

$$\langle \Psi_0 | Q_R | \Psi_0 \rangle$$

and

$$-\langle Q_R \Psi_0 | T^\sigma | Q_R \Psi_0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}} \tag{1.3.1}$$

in Sections 5.2 and 5.3. In particular, we prove that a version of the London term (1.0.3) involving the smeared Coulomb potential Q_R is the lowest-order contribution (in $1/R$) to (1.3.1). Furthermore, this 'smeared' version of the London term has a representation as an integral over photon momenta, which is intimately connected to the relationship (1.0.10) discussed above.
In Section 5.4, a number of terms from $\sum_{i=1}^4 e^i V_i^\sigma(\Lambda, \mathbf{R})$ which contain both the quantized radiation field and the interatomic Coulomb potential and which are grouped into terms denoted by $M_A(\mathbf{R}, \sigma)$ and $M_B(\mathbf{R}, \sigma)$ are investigated. We prove Theorem 5.4.1, which identifies the contributions to $M_A(\mathbf{R}, \sigma)$ and $M_B(\mathbf{R}, \sigma)$ at orders $1/R^6$ and $1/R^7$ and provides error estimates for the remaining ones, proving them to be $\mathcal{O}(1/R^8)$.
In the course of the proof, which comprises Sections 5.5.2 through 5.5.5, a number of methods is used. In Section 5.5.2 the multipole expansion is applied to Q_R, again introducing

a spatial cutoff and providing error estimates for the 'interior' and 'exterior' contributions to the corresponding matrix elements. The estimates on the 'exterior' contributions are independent of the infrared regularization, and their proof involves results on the conservation of exponential decay under the application of resolvents of atomic Schrödinger operators, see Lemma A.2.2 in the Appendix.
We then use rotation invariance and parity of the atomic ground states to show that a number of terms obtained from replacing Q_R by its series expansion vanish, see Section 5.5.3.
In Section 5.5.4 we establish the existence of the $(\sigma \to 0)$-limits of the 'interior' contributions to $M_A(\mathbf{R}, \sigma)$ and $M_B(\mathbf{R}, \sigma)$ and give estimates on the rate of convergence.
In the remainder of the proof of Theorem 5.4.1 we show that the lowest powers of $1/|\mathbf{R}|$ occurring in these limits are $1/|\mathbf{R}|^6$ and $1/|\mathbf{R}|^7$ and calculate the corresponding coefficients, see Section 5.5.5. To this end, we investigate the occurring Fourier integrals using two different methods. The first one uses an approach involving the distributional Fourier transform of singular functions. It exploits the homogeneity of parts of the integrands as well as the relation (1.0.10) to transform the integrals in question into convolutions with Dirac sequences parametrized by the interatomic distance R. This method is introduced in Section 5.5.5. Its drawback is that one has to be able to compute the distributional Fourier transforms of the integrands explicitly. In the cases where this is not possible, we therefore employ a second method, which involves standard decay estimates for oscillatory integrals involving smooth functions. Fortunately, the latter regularity requirement is met in the cases in which we would like to apply this method, which turns out to be due to properties of the ultraviolet-cutoff and the analyticity of resolvents.

In Chapter 6 we analyze the terms $F_7(\mathbf{R}, \sigma)$, $F_8(\mathbf{R}, \sigma)$ and establish the (asymptotic) cancellation of the $1/|\mathbf{R}|^6$-contributions. We first derive error estimates comparing $F_7(\mathbf{R}, \sigma)$ and $F_8(\mathbf{R}, \sigma)$ to their respective $(\sigma \to 0)$-limits $F_7(\mathbf{R})$ and $F_8(\mathbf{R})$, which turn out to exist since the integrands are sufficiently regular at the origin. Subsequently, in Section 6.5, we first show that the lowest power of $1/|\mathbf{R}|$ that enters in $F_7(\mathbf{R})$ is $1/|\mathbf{R}|^7$ and calculate the (asymptotic) coefficient explicitly. This is done by first rescaling the photon momenta, at which point the homogeneity mentioned above comes into play, and then applying a method for the asymptotic analysis of a certain class of singular Fourier integrals. This method, which is developed in Sections 6.2 and 6.3, uses the fact that the Fourier transform of the (rescaled) ultraviolet-cutoff, which coincides with the (rescaled) smeared charge distribution due to the relation (1.0.10), is a Dirac sequence. Furthermore, it involves an analysis of the regularity of the dynamic polarizabilities $\alpha_E^{A,B}(\mathbf{k})$ and their Fourier transforms, thereby making the formal asymptotics arguments used in the physics literature (see e.g. formulas (1.0.5) and (1.0.6)) rigorous.
The term $F_8(\mathbf{R})$ contains integrands that are more regular than those occurring in $F_7(\mathbf{R})$, and can be shown to be $\mathcal{O}(1/|\mathbf{R}|^8)$ by the standard decay estimates for oscillatory integrals mentioned above.
In Section 6.6 we show how the $1/|\mathbf{R}|^6$-contribution originating from the electrostatic Coulomb interaction is cancelled asymptotically by contributions involving the radiation field, in the sense that their sum decays faster than any inverse power of $|\mathbf{R}|$. As mentioned above, this crucially exploits the relation (1.0.10), which is a consequence of using a smeared Coulomb potential for the interaction of the atomic particles. We also argue why

we expect this mechanism of cancellation to break down if a proper Coulomb potential is used instead.

Finally, in Chapter 7, we collect all the previous results, carry out the proofs of Theorems 1.2.1 and 1.2.2, and prove Theorem 1.2.3 by combining the norm resolvent convergence of the atomic Hamiltonians mentioned above with an argument involving the uniformity of exponential decay estimates for the atomic ground states with respect to the ultraviolet-cutoff parameter Λ.

1.4 Further discussion of the physics literature

In the 1950s and 1960s, research on the retarded Van der Waals interaction in the theoretical physics literature mainly focused on a model described by a Hamiltonian which is formally given by

$$H_1 = H_A + H_B + H_f - e\left(x_1 \cdot \mathbf{E}(0) + x_2 \cdot \mathbf{E}(\mathbf{R})\right),$$

where H_A and H_B are non-relativistic Schrödinger Hamiltonians describing hydrogen atoms located at 0 and $\mathbf{R}$, H_f is the free Hamiltonian of the quantized radiation field, and

$$\mathbf{E}(\mathbf{x}) = \sum_{\lambda=1,2} \int d\mathbf{k} \sqrt{\frac{\hbar\omega(\mathbf{k})}{2}} \frac{i}{(2\pi)^{3/2}} \mathbf{e}(\mathbf{k},\lambda)(\mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{x})} a_\lambda(\mathbf{k}) - \mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{x})} a_\lambda^\dagger(\mathbf{k}))$$

is the transverse electric field. Here $\omega(\mathbf{k}) = c|\mathbf{k}|$ is the photonic dispersion relation, c denotes the speed of light, $\mathbf{e}(\mathbf{k},\lambda)$ are polarization vectors, and $a_\lambda^\dagger(\mathbf{k})$, $a_\lambda(\mathbf{k})$ are creation and annihilation operators (see Section 2.1 below for precise definitions). Note that no ultraviolet-cutoff has been incorporated into the electric field, so that it is not clear whether H_1 is well-defined as a self-adjoint Hamiltonian. Formally, H_1 can be viewed as being obtained from the dipole-approximated Pauli-Fierz Hamiltonian

$$\begin{aligned} H_{dip} =& \frac{1}{2m_e}\left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_1} - \frac{e}{c}\mathbf{A}(\rho,0)\right)^2 + \frac{1}{2m_e}\left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_2} - \frac{e}{c}\mathbf{A}(\rho,\mathbf{R})\right)^2 \\ &+ e^2\tilde{Q}_R(x_1,x_2) + H_f \end{aligned}$$

(with $\tilde{Q}_R$ as in (1.0.2)) via the unitary transformation

$$\mathrm{e}^{-\mathrm{i}((e/(\hbar c)x_1\cdot\mathbf{A}(\rho,0)+(e/(\hbar c)x_2\cdot\mathbf{A}(\rho,\mathbf{R}))},$$

which yields the Hamiltonian

$$\begin{aligned} H_2 =& H_A + \frac{e^2}{3(2\pi)^3}\|\rho\|_{L^2}^2|x_1|^2 + H_B + \frac{e^2}{3(2\pi)^3}\|\rho\|_{L^2}^2|x_2|^2 + H_f \\ &- e\big(x_1\cdot\mathbf{E}_\rho(0) + x_2\cdot\mathbf{E}_\rho(\mathbf{R})\big) + (x_1\cdot x_2)\frac{e^2}{(2\pi)^3}\int d\mathbf{k}|\rho(k)|^2\mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}} \\ &+ e^2\tilde{Q}_R(x_1,x_2) - \frac{e^2}{(2\pi)^3}\int d\mathbf{k}\frac{|\rho(\mathbf{k})|^2}{|k|^2}(x_1\cdot\mathbf{k})(\mathbf{k}\cdot x_2)\mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}}, \end{aligned}$$

where

$$\mathbf{E}_\rho(\mathbf{x}) = \sum_{\lambda=1,2} \int d\mathbf{k} \sqrt{\frac{\hbar\omega(\mathbf{k})}{2}} \frac{\mathrm{i}\rho(\mathbf{k})}{(2\pi)^{3/2}} \mathbf{e}(\mathbf{k},\lambda)(\mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{x})} a_\lambda(\mathbf{k}) - \mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{x})} a_\lambda^\dagger(\mathbf{k})).$$

Note that the coupling function in $\mathbf{E}_\rho(\mathbf{x})$ behaves like $\sqrt{|\mathbf{k}|}$ at $\mathbf{k} = 0$, in contrast to the one occurring in the electromagnetic vector potential $\mathbf{A}(\rho, \mathbf{x})$, which behaves more singular, namely as $1/\sqrt{|\mathbf{k}|}$ at zero. Neglecting the harmonic terms $\frac{e^2}{3(2\pi)^3}\|\rho\|_{L^2}^2 |x_1|^2$, $\frac{e^2}{3(2\pi)^3}\|\rho\|_{L^2}^2 |x_2|^2$ and

$$(x_1 \cdot x_2) \frac{e^2}{(2\pi)^3} \int d\mathbf{k} |\rho(k)|^2 \mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}},$$

setting $\rho \equiv 1$ (this corresponds to a distribution of point charges), and arguing that to first approximation, the dipole contribution from $\tilde{Q}_R$
is cancelled exactly by the term

$$-\frac{1}{(2\pi)^3} \int d\mathbf{k} \frac{1}{|k|^2} (x_1 \cdot \mathbf{k})(\mathbf{k} \cdot x_2) \mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}},$$

which by (formal) Fourier transform is equal to the dipole operator

$$-\frac{1}{4\pi R^3} \left(\mathbf{x}_1 \cdot (1 - 3|\hat{\mathbf{R}}\rangle\langle\hat{\mathbf{R}}|) x_2 \right),$$

one arrives at H_1, the only interaction operator being

$$H' = -e \left(x_1 \cdot \mathbf{E}(0) + x_2 \cdot \mathbf{E}(\mathbf{R}) \right). \tag{1.4.1}$$

Note however that it is not clear a priori how accurate the above approximations are and whether they can be controlled. Furthermore, even though H_2 is unitarily equivalent to H_{dip}, it is not clear whether H_1 - even if properly realized as a self-adjoint Hamiltonian - is unitarily equivalent to any physical model derived from first principles.
The first paper outlining a perturbation calculation using H_1 seems to be [PZ57], in which the result (1.0.5) of [CP48] is re-derived. The authors start out with a box quantization of the radiation field, leading to a discrete spectrum of H_f. The corresponding sums are then later replaced by integrals over photon momenta.
The additional assumption that the spectra of H_A and H_B are finite and consist of a ground state energy and one threefold degenerated excited level is made, which results in modified polarizabilities $\alpha_E^{A,B}$. The resulting integrals over photon momenta are transformed into (1.0.5) by arguments involving residue calculus and principal value integrals with implicit cutoffs at infinity.
In [MP65] the results of [PZ57] are extended to the case of molecules in excited states, resulting in a retarded Van der Waals interaction that can be either attractive or repulsive. Craig and Power ([CP69]) perform a calculation using (1.4.1) as a perturbation operator and arrive at the formula

$$-\alpha_E^A \alpha_E^B \frac{\hbar c}{(2\pi)^4} \int_{\mathbb{R}^6} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\mathbf{k}_1||\mathbf{k}_2|}{|\mathbf{k}_1| + |\mathbf{k}_2|} (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}} \tag{1.4.2}$$

for the lowest-order contribution to the retarded Van der Waals interaction. Here α_E^A and α_E^B are static polarizabilities of two neutral atoms with purely discrete spectra. The

authors then claim that a finite value for the integral can be found by 'explicit evaluation'. Taking into account that this integral is clearly divergent in the sense of Lebesgue, this can only mean that implicitly, a principal value or another limit was computed.
As will be shown in the course of the present work, if an ultraviolet-cutoff is incorporated systematically into the model from the start, terms formally similar to (1.4.2) actually contribute to the $1/R^7$- coefficient in the asymptotic expansion of the interaction potential, and the constant found in [CP69] has a well-defined meaning as a limit. For textbook accounts of the derivations and the methods used in the above works, see [Pow65] and [MK67].
Feinberg and Sucher ([FS68], [FS70]) follow an approach using scattering-theoretic arguments and interaction potentials defined in terms of spectral representation of Feynman amplitudes. They find the formula

$$-\frac{\hbar c}{R^7}\left(\frac{23}{4\pi}(\alpha_E^A\alpha_E^B+\alpha_M^A\alpha_M^B)+\frac{7}{4\pi}(\alpha_E^A\alpha_M^B+\alpha_M^A\alpha_E^B)\right) \tag{1.4.3}$$

for the lowest-order contribution to the retarded Van der Waals interaction. Note that (1.4.3) contains the magnetic polarizabilities

$$\begin{aligned}\alpha_M^A &:= -\frac{e^2}{12}\sum_{\alpha=1}^{3}\Big\langle\sum_{i=1}^{Z_A}\mathbf{x}_i^\alpha\Psi_A^0|\sum_{j=1}^{Z_A}\mathbf{x}_j^\alpha\Psi_A^0\Big\rangle,\\ \alpha_M^B &:= -\frac{e^2}{12}\sum_{\alpha=1}^{3}\Big\langle\sum_{i=1}^{Z_B}\mathbf{x}_i^\alpha\Psi_B^0|\sum_{j=1}^{Z_B}\mathbf{x}_j^\alpha\Psi_B^0\Big\rangle,\end{aligned} \tag{1.4.4}$$

which do not occur in the Casimir-Polder result. In [Boy74], (1.4.3) is re-derived by arguments involving the zero-point energy of the quantized radiation field.
In a recent work ([MS09]) Miyao and Spohn follow a functional-integral approach to the problem. Starting from a self-adjoint, ultraviolet-cutoff, non-dipole-approximated Hamiltonian for an H_2-molecule with 'smeared' electrons coupled to the quantized radiation field (see the Hamiltonian (1.0.9) above), their very insightful (though in parts not completely rigorous) derivation uses the Feynman-Kac formula and an expansion of the second cumulant. The result is a formula for the retarded Van der Waals interaction which is similar to (1.4.3), but which has different coefficients in front of the terms containing the magnetic polarizability. A further difference is that their polarizabilities are defined in terms of the (reduced) resolvent and the ground state of the Pauli-Fierz Hamiltonian of a single hydrogen atom coupled to the radiation field, as opposed to the Schrödinger operators $H_{A,B}$ that were used in the definitions (1.0.7) and (1.4.4).
An interesting future task would be to investigate the relation between these quantities for the two cases with and without radiation field. It might be worth noting that in the course of their derivation, the authors also encounter integrals of the form (1.4.2), so the rigorous method introduced in the present work for asymptotically expanding such integrals in powers of R provides a rigorous foundation of the corresponding steps in [MS09].
For further references to the physics and quantum chemistry literature, we refer to the textbooks [Spo04] and [Mil93].

1.5 Experimental situation

The experimental investigation of Van der Waals forces and the effect of retardation has quite a long history, which dates back to the 1930s and is still an active field today; for extensive reviews of the history and the current status of the experimental situation, see [KMM09] and [BMM01].
However, so far experiments in this area have mainly focussed on measuring a macroscopic manifestation of dispersion forces, the so-called Casimir effect. This effect describes an interaction between macroscopic bodies which is a consequence of the underlying quantum phenomenon. As in the microscopic theory, these macroscopic interactions are predicted to exhibit a crossover in behaviour between a non-retarded short-range regime and a retarded long-range regime, which manifests itself in the relevant potentials obeying different power laws in the respective regimes of separation between the bodies. The scale at which this crossover occurs, as well as the sign, the different power laws and the strength of the interaction itself, sensitively depend on the geometry and the material properties of the macroscopic objects considered.
There are two commonly used theories of the macroscopic Casimir effect, neither of which is based directly on quantum-mechanical Hamiltonians describing atoms and molecules. The first one is a direct macroscopic approach based on the fluctuation-dissipation theorem. This approach goes back to work by Lifshitz [Lif56] and takes into account macroscopic material properties like the (frequency-dependent) dielectric permittivity, as well as atomic properties such as dynamic polarizabilities (see below for a definition). The second theory is based on arguments from quantum field theory and views the presence of macroscopic bodies as imposing boundary conditions which alter the vacuum state of the field. This approach was first used by Casimir ([Cas48]) shortly after the publication of the Casimir-Polder result [CP48]. The two approaches are compatible in that the Lifshitz results can be obtained from the field-theoretic approach, see the references in [KMM09].
It should be noted that even the macroscopic Casimir effect is extremely difficult to measure, and experiments have to obey stringent requirements on equipment, methods and precision, for instance extremely precise determination of the separation distance, circumvention of residual potential differences, and minimizing material roughness and impurity. However, in recent years there have been experiments using atomic force microscopy which achieve accuracies of about one per cent, and which confirm the behaviour of the dispersive interaction between macroscopic bodies as predicted by the Lifshitz theory (and modifications of it which take into account properties of real materials). In particular, the crossover from the non-retarded to the retarded regime (the interaction in the latter is sometimes termed the Casimir force) has been observed for different geometries (e.g. plates, spheres, lenses) and materials (e.g. metals, mica, coated polystyrene). There have also been indirect measurements of the Casimir force between an atom and a plate, which use Bose-Einstein-condensates, and in mesoscopic situations using extremely small material samples. For all the results just mentioned, see [KMM09], [MPR$^+$08] and the references therein.
To our knowledge, so far no direct experimental verification of the microscopic theory of retarded Van-der Waals forces between atoms and molecules, which are the subject of the current work, seems to be available; the only commonly cited 'indirect experimental evidence' being the classic monograph [VO99], parts of which actually inspired the theoretical considerations in [CP48] in the first place. Nevertheless, since the Lifshitz theory

reproduces the microscopic predictions of London and Casimir-Polder in the limit of dilute bodies (see the references in [KMM09] and [BMM01]), experiments verifying predictions about the macroscopic Casimir effect can be considered an indirect verification of the microscopic theory.

Chapter 2

Definition of the model and main results

2.1 Hamiltonians

Consider two neutral atoms A and B consisting of two clamped nuclei located at 0 and $\mathbf{R} \in \mathbb{R}^3$, with nuclear charge Z_A, Z_B respectively, whose (spinless) components interact via smeared Coulomb potentials. The corresponding Hamiltonian (1.0.9) describing the compound system of the molecule (composed A and B) coupled to the quantized radiation field is given by

$$\begin{aligned} &H_{\mathrm{QA}}(\mathbf{R}) \\ &= \sum_{j=1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_j} - \frac{e}{c}\mathbf{A}(\rho, \mathbf{x}_j) \right)^2 + e^2(Q_\psi(\mathbf{R}) \otimes I_{\mathcal{F}}) + I_{\mathcal{H}_A} \otimes I_{\mathcal{H}_B} \otimes H_f, \end{aligned} \tag{2.1.1}$$

and the Hamiltonians of the individual systems consisting of atom A (respectively B) coupled to the quantized radiation field are given by the Hamiltonians

$$H_{\mathrm{QA}}^A = \sum_{j_A=1}^{Z_A} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}(\rho, \mathbf{x}_{j_A}) \right)^2 + e^2 Q_A + H_f, \tag{2.1.2}$$

$$H_{\mathrm{QA}}^B = \sum_{j_B=Z_A+1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}(\rho, \mathbf{x}_{j_B}) \right)^2 + e^2 Q_B(\mathbf{R}) + H_f. \tag{2.1.3}$$

These three operators act on the Hilbert spaces $\mathrm{L}^2_{anti}(\mathbb{R}^{3N}) \otimes \mathcal{F}$, $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes \mathcal{F}$ and $\mathrm{L}^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}$, respectively.

$$\mathcal{F} := \mathcal{F}(W) := \oplus_{n=0}^{\infty} \mathcal{F}^{(n)} := \oplus_{n=0}^{\infty} \otimes_s^n W$$

is the bosonic Fock space over the Hilbert space

$$W := L^2(\mathbb{R}^3) \oplus L^2(\mathbb{R}^3).$$

It describes the quantum states of an unconstrained number of photons with two degrees of polarization. $\otimes_s^n$ denotes the symmetric tensor product of Hilbert spaces, and we have

set $\mathcal{F}^{(0)} := \mathbb{C} = \text{span}\{\Omega\}$, where Ω is the so-called photon vacuum. m_e is the mass of an electron, e denotes the elementary charge, c is the speed of light in the vacuum and $\hbar$ is the reduced Planck constant. $N = Z_A + Z_B$ is the total charge, and

$$\begin{aligned}
Q_\psi(\mathbf{R}) = &\sum_{i_A,j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \Big(\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}} - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{i_A}-\mathbf{R})} - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{j_B}} + \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}-\mathbf{x}_{i_A})} \Big) \\
&+ \sum_{i_A<j_A} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_A}-\mathbf{x}_{i_A})} - Z_A \sum_{i_A} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}} \\
&+ \sum_{i_B<j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}-\mathbf{x}_{i_B})} - Z_B \sum_{i_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{i_B}-\mathbf{R})},
\end{aligned}$$

$$Q_A = \sum_{i_A<j_A} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_A}-\mathbf{x}_{i_A})} - Z_A \sum_{i_A} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}}, \tag{2.1.4}$$

$$Q_B(\mathbf{R}) = \sum_{i_B<j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}-\mathbf{x}_{i_B})} - Z_B \sum_{i_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{i_B}-\mathbf{R})} \tag{2.1.5}$$

are the Coulomb potentials corresponding to charge densities smeared by convolution with the function ψ, which will be specified below. See A.5 in the appendix for a derivation of these formulas for the potentials.

Remark: In the whole of this work, we will adhere to the convention

$$\hat{f}(k) := \frac{1}{(2\pi)^{n/2}} \int f(x) \mathrm{e}^{-\mathrm{i}(k\cdot x)} dx \tag{2.1.6}$$

for the Fourier transform. Using this convention, the electromagnetic vector potential in Coulomb gauge is defined by

$$\mathbf{A}(\rho, \mathbf{x}) := a^\dagger(G^{\mathbf{x}}) + a(G^{\mathbf{x}}). \tag{2.1.7}$$

Here $a^\dagger$ and a are the photon creation and annihilation operators on the Fock space $\mathcal{F}$. They are defined by first prescribing their actions on $\mathcal{F}^{(n)}$, namely

$$(a(f)^\dagger \Psi)^{(n)}(\mathbf{k}_1, \lambda_1, \ldots, \mathbf{k}_n, \lambda_n) := \frac{1}{\sqrt{n}} \sum_{j=1}^{n} f(\mathbf{k}_j, \lambda_j) \Psi^{(n-1)}(\mathbf{k}_1, \lambda_1, \ldots, \widehat{\mathbf{k}_j, \lambda_j}, \ldots, \mathbf{k}_n, \lambda_n),$$

$$(a(f)\Psi)^{(n)}(\mathbf{k}_1, \lambda_1 \ldots, \mathbf{k}_n, \lambda_n) := \sqrt{n+1} \sum_{\lambda=1,2} \int_{\mathbb{R}^3} \overline{f(\mathbf{k}, \lambda)} \Psi^{(n+1)}(\mathbf{k}, \lambda, \mathbf{k}_1, \lambda_1 \ldots, \mathbf{k}_n, \lambda_n),$$

where $f \in W = L^2(\mathbb{R}^3) \oplus L^2(\mathbb{R}^3)$ describes a single (polarized) photon, and then extending this to the dense subspace

$$\mathcal{F}_{fin} := \{\Psi = \{\Psi^{(n)}\}_{n=0}^{\infty} \in \mathcal{F} | \quad \Psi^{(n)} = 0 \text{ for all but finitely many } n\}.$$

It is easily shown (see e.g. [Mer06]) that $a(f)^\dagger$ and $a(f)$ are closable and that their closures (again denoted by the same symbols) satisfy the relation

$$(a(f))^* = a^\dagger(f), \tag{2.1.8}$$

where the asterisk denotes the adjoint operator. Note that on each n-particle level $\mathcal{F}^{(n)}$, these operators are bounded, but their extensions to $\mathcal{F}$ are unbounded operators.
On $\mathcal{F}_{fin}$, the creation and annihilation operators satisfy the canonical commutation relations

$$\begin{aligned}[a(f), a^\dagger(g)] =& \langle f, g\rangle_W,\\ [a(f), a(g)] =& [a^\dagger(f), a^\dagger(g)] = 0.\end{aligned}$$

For any $\mathbf{x} \in \mathbb{R}^3$ the coupling function $G^{\mathbf{x}} \in W$ is defined by

$$G^{\mathbf{x}}(\mathbf{k}, \lambda) := c\,\rho(\mathbf{k})\sqrt{\frac{\hbar}{2\omega(\mathbf{k})}}\mathbf{e}(\mathbf{k}, \lambda)\mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{x})}, \tag{2.1.9}$$

where $\omega(\mathbf{k}) = c\mathbf{k}$ is the photonic dispersion relation. Note that formally, the vector potential is often written as

$$\mathbf{A}(\mathbf{x}) = \sum_{\lambda=1,2}\int d\mathbf{k}\; c\rho(\mathbf{k})\sqrt{\frac{\hbar}{2\omega(\mathbf{k})}}\mathbf{e}(\mathbf{k}, \lambda)(\mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{x})}a^\dagger_\lambda(\mathbf{k}) + \mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{x})}a_\lambda(\mathbf{k})),$$

but to give rigorous mathematical meaning to the objects $a^\dagger(\mathbf{k})$ and $a(\mathbf{k})$ would require the introduction of operator-valued distributions, which we shall not need for our purposes.
The polarization vectors $\mathbf{e}(\mathbf{k}, \lambda)$ satisfy

$$\mathbf{e}(\mathbf{k}, \lambda)\cdot\mathbf{k} = 0, \quad \mathbf{e}(\mathbf{k}, \lambda)\cdot\mathbf{e}(\mathbf{k}, \mu) = \delta_{\lambda\mu}$$

and can be chosen such that

$$\mathbf{e}(\mathbf{k}, \lambda) = \mathbf{e}(-\mathbf{k}, \lambda), \quad \lambda = 1, 2.$$

A typical choice is

$$\begin{aligned}\mathbf{e}(\mathbf{k}, 1) &= \frac{1}{\sqrt{k_1^2 + k_2^2}}(-k_2, k_1, 0),\\ \mathbf{e}(\mathbf{k}, 2) &= \frac{\mathbf{k}}{|k|}\times\mathbf{e}(\mathbf{k}, 1) = \frac{1}{|k|\,\sqrt{k_1^2 + k_2^2}}(k_1k_3, k_2k_3, -k_1^2 - k_2^2).\end{aligned}$$

The ultraviolet-cutoff function ρ satisfies

$$\rho(\cdot) = \hat{\psi}(\cdot) = \Lambda^3\widehat{\psi_0(\Lambda\cdot)} = \rho_0(\cdot/\Lambda),$$

where ψ is the form factor with which the charge distributions are convoluted. Here and in everything that follows, we will always place the following assumption on ψ.

Assumptions (A1):

$$\psi(\mathbf{x}) = \Lambda^3\psi_0(\Lambda\mathbf{x}),$$

where $\Lambda > 0$, $\psi_0 \in C_0^\infty(\mathbb{R}^3)$, $\operatorname{supp}\psi_0 \subset B_1(0)$, $\int\psi_0 = 1$ and ψ is real and invariant under rotations, i.e. $\psi(R^{-1}\mathbf{x}) = \psi(\mathbf{x})$ for any $R \in SO(3)$ and $\mathbf{x} \in \mathbb{R}^3$. In particular, ψ is even, i.e. $\psi(-\mathbf{x}) = \psi(\mathbf{x})$ for all $\mathbf{x} \in \mathbb{R}^3$.

The parameter Λ characterizes the (inverse) length scale over which the point charges are smeared. From the behaviour of the Fourier transform under dilations we conclude

$$\rho(\cdot) = \hat{\psi}(\cdot) = \Lambda^3 \widehat{\psi_0(\Lambda \cdot)} = \rho_0(\cdot/\Lambda), \tag{2.1.10}$$

where $\rho_0 = \widehat{\psi_0}$, so that on the level of the photons, Λ plays the role of an ultraviolet-cutoff. Note that the assumptions on ψ imply that ρ is real, $\rho \in \mathcal{S}(\mathbb{R}^3)$, and that it satisfies $\rho(-\mathbf{k}) = \rho(\mathbf{k})$ for all $\mathbf{k} \in \mathbb{R}^3$. Furthermore, the integral constraint on ψ_0 enforces $\hat{\psi}(0) = 1/(2\pi)^{3/2}$, so that in the definition of the vector potential $\mathbf{A}$, we automatically absorb the factor $1/(2\pi)^{3/2}$ coming from the Fourier decomposition of solutions of the Maxwell equations into normal modes (see [Spo04]).
The relationship (2.1.10) is an inherent feature of the quantization of the Abraham model (for an explanation and further discussion, see [Spo04]), and the fact that the ultraviolet-cutoff ρ is the Fourier transform of a scaled C_0^∞-function (i.e. a Dirac sequence) will have interesting consequences for the investigation of the large R-asymptotics of the fourth-order energy corrections investigated below, see Section 6.5.
The Hamiltonian of the free quantized radiation field is given by

$$H_f := \sum_{\lambda=1,2} \int_{\mathbb{R}^3} \hbar\omega(\mathbf{k}) a_\lambda^\dagger(\mathbf{k}) a_\lambda(\mathbf{k}) d\mathbf{k} := d\Gamma(\hbar\omega(\mathbf{k})),$$

where the right-hand side denotes the second quantization of the multiplication operator $\hbar\omega(\mathbf{k}) \oplus \hbar\omega(\mathbf{k}) = \begin{pmatrix} \hbar\omega(\mathbf{k}) & 0 \\ 0 & \hbar\omega(\mathbf{k}) \end{pmatrix}$ on $\mathcal{F}^{(1)} = W = L^2(\mathbb{R}^3) \oplus L^2(\mathbb{R}^3)$. The latter is self-adjoint on its maximal domain (this is always true for multiplication by real measurable functions), and by construction of the second quantization (see e.g. [RS80], Section VIII.10), H_f is a self-adjoint operator on $\mathcal{F}$ with domain $D(H_f)$.
The following result is a fundamental prerequisite for the mathematically rigorous treatment of the Hamiltonians we have just introduced.

Theorem 2.1.1. *Let the assumptions (A1) on the form factor ψ be satisfied, and assume that $|e| \leq e_0$ for a suitable e_0. Then the operators $H_{QA}(\mathbf{R})$, H_{QA}^A, H_{QA}^B are self-adjoint and bounded from below on $D(-\Delta_{A,B} + H_f)$, $D(-\Delta_A + H_f)$, $D(-\Delta_B + H_f)$, respectively.*

Proof. It can be shown (see e.g. [BFS99], [Spo04]) that the domains of $a(f)$ and $a^\dagger(f)$ contain $D((H_f)^{1/2})$ and that the relative boundedness estimates

$$\|a^\dagger(f)\Psi\|_{\mathcal{F}} \leq \|f/\omega\|_W \| \, \|(H_f)^{1/2}\Psi\|_{\mathcal{F}} + \|f\|_h \, \|\Psi\|, \tag{2.1.11}$$

$$\|a(f)\Psi\|_{\mathcal{F}} \leq \|f/\omega\|_W \| \, \|(H_f)^{1/2}\Psi\|_{\mathcal{F}} \tag{2.1.12}$$

hold for any $f \in W$ and $\Psi \in D((H_f)^{1/2})$. Combining these with (infinitesimal) relative boundedness of the Coulomb potential with respect to the Laplacian and noting that the assumptions (A1) on the form factor ψ imply that

$$\int_{\mathbb{R}^3} d\mathbf{k} |\rho(\mathbf{k})|^2 \frac{1}{\omega(\mathbf{k})^2} < \infty,$$

one deduces that the symmetric operators $e\frac{\mathrm{i}\hbar}{2m_e c} \nabla_{\mathbf{x}_j} \cdot \mathbf{A}(\rho, \mathbf{x})$ and $e^2 \frac{1}{2m_e c^2} \mathbf{A}(\rho, \mathbf{x})^2$ are relatively form-bounded with respect to the operator $-\Delta + H_f$. The smallness assumption

on e and an application of the KLMN theorem (see e.g. [RS75]) yield the assertion.

□

Remark 2.1.2. By functional integral methods it can actually be shown that this result holds true for any value of the coupling strength e, cf. [Hir02]). For more details on the Hamiltonian of the quantized Abraham model, see [Spo04].

Remark 2.1.3. As mentioned in the introduction, the realization of Pauli-Fierz Hamiltonians as semi-bounded, self-adjoint operators (which as such can have ground states with finite energies and define a strongly continuous unitary group describing the time evolution of the quantum system) was pioneered in the works of Bach, Fröhlich and Sigal (see [BFS98], [BFS99]). Without these fundamental results, the rigorous approach to retarded Van der Waals interactions undertaken in the present work would not be possible. Note, however, that the introduction of an ultraviolet-cutoff (which was necessary in order to obtain the bounds (2.1.11)) implies that the operators $H_{QA}(\mathbf{R})$, H_{QA}^{A}, H_{QA}^{B}, as well as all objects and quantities derived from them (such as energy levels, ground states and resolvents), depend on the choice of the cutoff function ψ_0 and the scale Λ at which the ultraviolet-cutoff comes into effect. Furthermore, it is by no means clear (and probably should not be expected at this level of generality) how the ultraviolet-cutoff could be removed in any well-defined way. For instance, note that the right-hand sides of (2.1.11) diverge as $\Lambda \to \infty$ if $f = G^{\mathbf{x}}(\mathbf{k}, \lambda)$). This general situation is in contrast to the case of the asymptotic $1/R^7$-coefficient c_7 (see (1.2.2)), which is part of a description of a very special physical situation (two atoms in their ground states and in equilibrium with the radiation field), and which we find to have a well-defined ($\Lambda \to \infty$)-limit, see Theorem 1.2.3.

2.2 Approximations

Next we implement two common approximations, namely we employ the dipole approximation and neglect 'interatomic' antisymmetry of the electrons.

2.2.1 The dipole approximation

From now on we will use Hamiltonians which are subject to the so-called dipole approximation. This means that instead of using an $\mathbf{x}$-dependent vector potential for the quantized field, we 'fix' the electromagnetic field at the nuclei located at 0 and $\mathbf{R}$. The corresponding Hamiltonians are

$$H(\mathbf{R}) := \sum_{j_A=1}^{Z_A} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}(\rho, 0)\right)^2 + \sum_{j_B=Z_A+1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}(\rho, \mathbf{R})\right)^2 + e^2(Q_\psi(\mathbf{R}) \otimes I_{\mathcal{F}}) + I_{\mathcal{H}_A} \otimes I_{\mathcal{H}_B} \otimes H_f \tag{2.2.1}$$

for the compound system and

$$H^A := \sum_{j_A=1}^{Z_A} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}(\rho,0)\right)^2 + e^2 Q_A + H_f, \tag{2.2.2}$$

$$H^B := \sum_{j_B=Z_A+1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}(\rho,\mathbf{R})\right)^2 + e^2 Q_B(\mathbf{R}) + H_f, \tag{2.2.3}$$

for the individual systems.

2.2.2 Neglecting interatomic antisymmetry

The second approximation consists of neglecting interatomic antisymmetry of the electrons, which is implemented by considering $H(\mathbf{R})$ an operator on the Hilbert space $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}$ instead of $L^2_{anti}(\mathbb{R}^{3N}) \otimes \mathcal{F}$. As before, H^A and H^B act on $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes \mathcal{F}$ and $L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}$, respectively. Note that using the partition of the vector potential in (2.2.1) (i.e. j_A's interact with the field at 0, j_B's with the field at $\mathbf{R}$) in an operator on $L^2_{anti}(\mathbb{R}^{3N}) \otimes \mathcal{F}$ would not be possible to start with: since $H(\mathbf{R})$ is not invariant under permutation of the electronic variables, it does not leave this subspace invariant.
Both of the above approximations are motivated by the observation that as soon as an ionicity avoidance result, i.e. a bound of the form

$$\rho_{\psi_0}(x) \leq C\mathrm{e}^{-c\,\mathrm{dist}(x,\{0,\mathbf{R}\})}$$

on the one-particle density of the (true, antisymmetric) ground state ψ_0 is available, a variational argument (see e.g. [MS80] for the Schrödinger/no-field case) shows that the exchange error, i.e. the difference

$$\inf_{\psi\in A_N(\mathcal{H}_A\otimes\mathcal{H}_B)\otimes\mathcal{F}} \frac{\langle\psi|H(\mathbf{R})|\psi\rangle}{\|\psi\|^2} - \inf_{\psi\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}} \frac{\langle\psi|H(\mathbf{R})|\psi\rangle}{\|\psi\|^2},$$

decays exponentially in the interatomic distance R. Such bounds are known to hold for atoms (see standard results on exponential decay) and molecules (see [Fri]) in the Schrödinger case without field, for atoms in NRQED (see [Gri04]), and they are strongly conjectured to also hold for molecules in NRQED.
In order to define the setup and carry out the calculations on the Hilbert space $L^2_{anti}(\mathbb{R}^N)\otimes\mathcal{F}$, one would then have to use a partition of the (true, antisymmetric) ground state according to regions of configuration space where each of the electrons is localized near one of the two nuclei. Although this is possible in principle, we refrain from implementing it in the present work so as not to overload the notation and to keep the calculations within a reasonable length.
As for the non-dipole-approximated Hamiltonians, one has the following self-adjointness and semi-boundedness result, which is proven in exactly the same manner.

Lemma 2.2.1. *Let the assumptions (A1) on the form factor ψ be satisfied, and assume that $|e| \leq e_0$ for a suitable e_0. Then the operators $H(\mathbf{R})$, H^A, H^B are self-adjoint and bounded from below on*

$$\begin{aligned} D(-\Delta_A - \Delta_B + H_f) &\subset L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}, \\ D(-\Delta_A + H_f) &\subset L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes \mathcal{F}, \\ D(-\Delta_B + H_f) &\subset L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}, \end{aligned}$$

respectively.

Again, this result actually holds true for any value of the coupling strength e, cf. [Hir02].

2.3 Infrared regularization

The quantity of interest in this work is the long-range behaviour of the interaction potential

$$\tilde{V}(\mathbf{R}) := E(\mathbf{R}) - \lim_{R\to\infty} E(\mathbf{R}). \tag{2.3.1}$$

Here

$$E(\mathbf{R}) := \inf \operatorname{spec}(H(\mathbf{R}))$$

is the ground state energy of the system consisting of the two atoms A and B at a finite distance R from each other, interacting via Coulomb interaction and the radiation field, and the second term in (2.3.1) is the limit of this ground state energy as the atoms are moved infinitely apart.
The great challenge in understanding $\tilde{V}(\mathbf{R})$ (whether qualitatively or quantitatively) is that the dependence of the ground state energy on the interatomic distance $\mathbf{R}$ is quite subtle and enters in a very indirect way. This is already the case in the situation without coupling to the radiation field, where the role of $\mathbf{R}$ is that of a parameter in the interatomic Coulomb potential, namely the position of the second nucleus. In the presence of the radiation field, an additional $\mathbf{R}$-dependence is generated, which - as we will see in the course of our analysis - is more subtle, even if the dipole-approximation is used.
As we pointed out in the introduction, perturbation theory is a tool that has been used extensively in the physics literature to tackle this problem in both cases, with the mathe–matical problems indicated above. However, as we also mentioned, the $1/|\mathbf{R}|^6$-asymptotics of $\tilde{V}$ in the case without radiation can be proven rigorously, so in order to explain the crossover to $1/|\mathbf{R}|^7$-behaviour in the presence of the field, one has to understand the mechanism by which contributions from the radiation field suppress the London term

$$-c_6 \frac{1}{|\mathbf{R}|}.$$

Since this term is of perturbative origin, it is natural to try and understand how the perturbative energy corrections obtained formally in the works mentioned above can be given a rigorous mathematical meaning.
Due to the lack of spectral gaps in the operators $H(\mathbf{R})$, H^A and H^B involved in the definition of $\tilde{V}(\mathbf{R})$ (see the discussion in the introduction above), a priori standard perturbation

theory is not available for their treatment. Our approach will therefore be to consider an infrared-regularized version of the interaction potential $\tilde{V}(\mathbf{R})$, the introduction of which we will now begin to prepare.
We incorporate an infrared-cutoff into the coupling function $G^{\mathbf{x}}(\mathbf{k},\lambda)$ by setting

$$\mathbf{G}^{\mathbf{x}}_{\sigma}(\mathbf{k},\lambda) := \chi_\sigma(\mathbf{k})G^{\mathbf{x}}(\mathbf{k},\lambda) = \chi_\sigma(\mathbf{k})\, c\, \rho(\mathbf{k})\sqrt{\frac{\hbar}{2\omega(\mathbf{k})}}\mathbf{e}(\mathbf{k},\lambda)\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}},$$

where $\sigma > 0$, and $\chi_\sigma(\mathbf{k})$ is the characteristic function of the set $\{\mathbf{k}\in\mathbb{R}^3,\, \omega(\mathbf{k})\geq\sigma\}$. The infrared-regularized vector potential is defined as

$$\mathbf{A}_\sigma(\mathbf{x}) := a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma) + a(\mathbf{G}^{\mathbf{x}}_\sigma),$$

and the corresponding infrared-regularized Hamiltonians are

$$\begin{aligned} H^\sigma(\mathbf{R}) := &\sum_{j_A=1}^{Z_A}\frac{1}{2m_e}\left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}_\sigma(0)\right)^2 + \sum_{j_B=Z_A+1}^{N}\frac{1}{2m_e}\left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}_\sigma(\mathbf{R})\right)^2 \\ &+ e^2Q_\psi(\mathbf{R}) + H_f \end{aligned}$$

for the compound system and

$$\begin{aligned} H^\sigma_A &:= \sum_{j_A=1}^{Z_A}\frac{1}{2m_e}\left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}_\sigma(0)\right)^2 + e^2Q_A + H_f, \\ H^\sigma_B &:= \sum_{j_B=Z_A+1}^{N}\frac{1}{2m_e}\left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}_\sigma(\mathbf{R})\right)^2 + e^2Q_B(\mathbf{R}) + H_f, \end{aligned}$$

for the individual systems.
The decomposition

$$L^2(\mathbb{R}^3) = L^2(\{\omega(\mathbf{k})\geq\sigma\})\oplus L^2(\{\omega(\mathbf{k})<\sigma\})$$

carries over to

$$\begin{aligned} W =& L^2(\mathbb{R}^3)\oplus L^2(\mathbb{R}^3) \\ =& \big(L^2(\{\omega(\mathbf{k})\geq\sigma\})\oplus L^2(\{\omega(\mathbf{k})\geq\sigma\})\big)\oplus\big(L^2(\{\omega(\mathbf{k})<\sigma\})\oplus L^2(\{\omega(\mathbf{k})<\sigma\})\big) \\ =& : W_\sigma\oplus W_{<\sigma} \end{aligned}$$

and induces an isomorphism

$$\tilde{U} : \mathcal{F}(W)\to\mathcal{F}(W_\sigma)\otimes\mathcal{F}(W_{<\sigma}) =: \mathcal{F}_\sigma\otimes\tilde{\mathcal{F}}_\sigma$$

on the level of the Fock spaces. Corresponding isomorphisms

$$\begin{aligned} U &:= I_{L^2_{anti}(\mathbb{R}^{3Z_A})\otimes L^2_{anti}(\mathbb{R}^{3Z_B})}\otimes\tilde{U}, \\ U_A &:= I_{L^2_{anti}(\mathbb{R}^{3Z_A})}\otimes\tilde{U}, \\ U_B &:= I_{L^2_{anti}(\mathbb{R}^{3Z_B})}\otimes\tilde{U}, \end{aligned}$$

are induced between the Hilbert spaces, with respect to which the Hamiltonians transform as

$$UH^{\sigma}(\mathbf{R})U^{-1} = H_{\sigma}(\mathbf{R}) \otimes I_{\tilde{\mathcal{F}}_{\sigma}} + I_{\mathcal{F}_{\sigma}} \otimes H_{f<\sigma}, \tag{2.3.2}$$

$$U_A H_A^{\sigma} U_A^{-1} = H_{\sigma}^A \otimes I_{\tilde{\mathcal{F}}_{\sigma}} + I_{\mathcal{F}_{\sigma}} \otimes H_{f<\sigma}, \tag{2.3.3}$$

$$U_B H_B^{\sigma} U_B^{-1} = H_{\sigma}^B \otimes I_{\tilde{\mathcal{F}}_{\sigma}} + I_{\mathcal{F}_{\sigma}} \otimes H_{f<\sigma}, \tag{2.3.4}$$

where

$$\begin{aligned} H_{\sigma}(\mathbf{R}) = &\sum_{j_A=1}^{Z_A} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}_{\sigma}(0)\right)^2 \\ &+ \sum_{j_B=Z_A+1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}_{\sigma}(\mathbf{R})\right)^2 + e^2 Q_{\psi}(\mathbf{R}) + H_{f\geq\sigma}, \end{aligned} \tag{2.3.5}$$

$$H_{\sigma}^A = \sum_{j_A=1}^{Z_A} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_A}} - \frac{e}{c}\mathbf{A}_{\sigma}(0)\right)^2 + e^2 Q_A + H_{f\geq\sigma}, \tag{2.3.6}$$

$$H_{\sigma}^B = \sum_{j_B=Z_A+1}^{N} \frac{1}{2m_e} \left(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{j_B}} - \frac{e}{c}\mathbf{A}_{\sigma}(\mathbf{R})\right)^2 + e^2 Q_B(\mathbf{R}) + H_{f\geq\sigma} \tag{2.3.7}$$

act on $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}_{\sigma}$, $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes \mathcal{F}_{\sigma}$ and $L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}_{\sigma}$, respectively, and

$$H_{f\geq\sigma} = \int_{\omega(\mathbf{k})\geq\sigma} \hbar\omega(\mathbf{k}) a^{\dagger}(\mathbf{k}) a(\mathbf{k}) d\mathbf{k} = d\Gamma(\hbar\omega(\mathbf{k})_{L^2(\{\omega(\mathbf{k})\geq\sigma\})}).$$

Note that the Fock space vacuum sector $\mathcal{F}^{(0)} = \mathbb{C}$ is left unaltered by the above isomorphism. On $\mathcal{F}_{\sigma}^{(1)}$, $H_{f\geq\sigma}$ acts by multiplication with $\hbar\omega(\mathbf{k})$, and this operator is bounded away from zero by construction, since the underlying space is $L^2(\omega(\mathbf{k}) \geq \sigma)$. We have

$$\mathrm{spec}(H_{f|\mathcal{F}_{\sigma}^{(1)}}) = \mathrm{ess\,ran}(\hbar\omega(\mathbf{k})) = [\hbar\sigma, \infty).$$

In particular, $H_{f|\mathcal{F}_{\sigma}^{(1)}}$ is boundedly invertible, with $\|(H_{f|\mathcal{F}_{\sigma}^{(1)}})^{-1}\| = \frac{1}{\hbar\sigma}$ Analogously, $H_{f|\mathcal{F}_{\sigma}^{(2)}}$ is multiplication by $\hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))$, and we have the identities $\mathrm{spec}(H_{f|\mathcal{F}_{\sigma}^{(2)}}) = [2\hbar\sigma, \infty)$ and $\|(H_{f|\mathcal{F}_{\sigma}^{(2)}})^{-1}\| = \frac{1}{2\hbar\sigma}$. As an operator on all of $\mathcal{F}_{\sigma}$, the spectrum of $H_{f\geq\sigma}$ is given by

$$\begin{aligned} \mathrm{spec}(H_{f\geq\sigma}) =& \mathrm{spec}\left(\oplus_{n=0}^{\infty}\hbar(\sum_{i=1}^{n}\omega(\mathbf{k}_i))\right) \\ =& \{0\} \cup_{n=1}^{\infty} [n\hbar\sigma, \infty) \\ =& \{0\} \cup [\hbar\sigma, \infty). \end{aligned}$$

Evidently, $H_{f\geq\sigma}$ has a spectral gap of size $\hbar\sigma$ above its ground state energy. Now note that since the inner product on the Fock space $\mathcal{F}_{\sigma}$ is generated by that on $L^2(\{\omega(\mathbf{k}) \geq \sigma\})$,

all integrations occurring in the calculation of the matrix elements will range over the set $\{\omega(\mathbf{k}) \geq \sigma\}$ respectively $\{\omega(\mathbf{k}_1) \geq \sigma\} \times \{\omega(\mathbf{k}_2) \geq \sigma\}$, and the final $(\sigma \to 0)$-limit can be carried out by replacing these with integrals over $\mathbb{R}^3$ and $\mathbb{R}^6$, respectively.
The next lemma states two facts: firstly, it shows that the infrared regularization is controlled in the sense that if it is removed by carrying out the $(\sigma \to 0)$-limit, we recover the ground state energies of the dipole-approximated Hamiltonians $H(\mathbf{R})$, H^A and H^B we set out to study in the first place from those of the infrared-regularized Hamiltonians $H^\sigma(\mathbf{R})$, H^σ_A and H^σ_A. Secondly, for the study of the ground state energies of the latter, we can restrict ourselves to the study of the operators $H_\sigma(\mathbf{R})$, H^A_σ and H^B_σ, which act on the Hilbert spaces $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}_\sigma$, $L^2_{anti}(\mathbb{R}^{3Z_A}) \otimes \mathcal{F}_\sigma$ and $L^2_{anti}(\mathbb{R}^{3Z_B}) \otimes \mathcal{F}_\sigma$, respectively, and have spectral gaps above their ground state energies. In particular, as we will see below, these latter operators can be treated using analytic perturbation theory.

Lemma 2.3.1. *Let the assumptions (A1) on the form factor ψ be satisfied, and assume that $|e| \leq e_0$ for a suitable e_0. Then*

$$\inf \operatorname{spec}(H^\sigma(\mathbf{R})) = \inf \operatorname{spec}(H_\sigma(\mathbf{R})) =: E^\sigma(\mathbf{R}), \tag{2.3.8}$$

$$\inf \operatorname{spec}(H^\sigma_A) = \inf \operatorname{spec}(H^A_\sigma) =: E^\sigma_A, \tag{2.3.9}$$

$$\inf \operatorname{spec}(H^\sigma_B) = \inf \operatorname{spec}(H^B_\sigma) =: E^\sigma_B. \tag{2.3.10}$$

and

$$\lim_{\sigma \to 0} E^\sigma(\mathbf{R}) = E(\mathbf{R}) = \inf \operatorname{spec}(H(\mathbf{R})), \tag{2.3.11}$$

$$\lim_{\sigma \to 0} E^\sigma_A = E_A := \inf \operatorname{spec}(H^A), \tag{2.3.12}$$

$$\lim_{\sigma \to 0} E^\sigma_B = E_B := \inf \operatorname{spec}(H^B). \tag{2.3.13}$$

Assume in addition that the atomic Hamiltonians H_A, H_B and $H_A + H_B + e^2 Q_R$ (see (2.5.1), (2.5.2) and (2.4.3) below for their definitions) have non-degenerate ground states. Then for e small enough and $\sigma \leq \sigma_0$ for a suitable σ_0 (depending on the spectral gaps of the atomic Hamiltonians), $E^\sigma(\mathbf{R})$, E^σ_A and E^σ_B are isolated eigenvalues of $H_\sigma(\mathbf{R})$, H^A_σ and H^B_σ, respectively, with spectral gaps at least of size σ.

Proof. The identities (2.3.8), (2.3.9) and (2.3.10) are immediate from the decompositions (2.3.2) through (2.3.4) upon noting that $\inf \operatorname{spec} H_{f<\sigma} = 0$. The assertions (2.3.11) through (2.3.13) follow from ([FGS08], Lemma 22) upon noting that all operator estimates used by the authors also hold for the dipole-approximated vector potential. The last assertion is a consequence of ([FGS08], Theorem 18). □

Remark 2.3.2. i) We have included the previous result here since it demonstrates the mechanism of the infrared regularization and the fact that it is well-controlled upon removing the infrared-cutoff. However, note that it concerns ground state energies before employing a perturbation expansion. As already mentioned in the remarks after Theorem 1.2.3, this result does not make any assertions about the $(\sigma \to 0)$-behaviour of individual terms in the perturbation expansion and therefore cannot be used to relate $(\sigma \to 0)$-limits of individual perturbation coefficients to quantities related to the true (i.e. non-infrared-regularized) ground state energies E_A, E_B and $E(\mathbf{R})$.

ii) In Theorem 2.5.2 below we will see that if the ultraviolet-cutoff Λ is chosen large enough, then the non-degeneracy assumption on the ground states of H_A and H_B follows from the assumption that the ground states of the atomic Schrödinger operators $\tilde{H}_A$ and $\tilde{H}_B$ with proper (i.e. non-smeared) Coulomb potential (see 2.5.3) are non-degenerate.

2.4 Perturbation-theoretic setup

In the following we will write $\mathbf{p}_j = -\mathrm{i}\hbar\nabla_{\mathbf{x}_j}$ and use the shorthand $\mathcal{H}_A = L^2_{anti}(\mathbb{R}^{3Z_A})$, $\mathcal{H}_B = L^2_{anti}(\mathbb{R}^{3Z_B})$. Multiplying out the squares in (2.3.5) and switching to the relative coordinates $\{\mathbf{x}_{Z_A+1} + \mathbf{R}, \ldots, \mathbf{x}_N + \mathbf{R}\}$ on $\mathcal{H}_B$ yields the perturbation problem

$$H_\sigma(\mathbf{R}) = H_\sigma(e, \mathbf{R}) := H_0^\sigma + eH'_\sigma + e^2 H''_\sigma, \tag{2.4.1}$$

where

$$H_0^\sigma = H_A + H_B + H_{f\geq\sigma}$$

is the non-interacting Hamiltonian of the two atoms and the free radiation field (see (2.5.1) and (2.5.2) below for the definition of H_A and H_B), and the perturbations are given by

$$H'_\sigma = -\frac{1}{m_e c}\sum_{j_A=1}^{Z_A}(\mathbf{p}_{j_A}\cdot\mathbf{A}_\sigma(0)) - \frac{1}{m_e c}\sum_{j_B=Z_A+1}^{N}(\mathbf{p}_{j_B}\cdot\mathbf{A}_\sigma(\mathbf{R})), \tag{2.4.2}$$

$$\begin{aligned} H''_\sigma =& \frac{1}{2m_e}\sum_{j_A=1}^{Z_A}\frac{1}{c^2}\mathbf{A}_\sigma(0)^2 + \frac{1}{2m_e}\sum_{j_B=Z_A+1}^{N}\frac{1}{c^2}\mathbf{A}_\sigma(\mathbf{R})^2 + Q_R \\ =& \frac{1}{2m_e}\frac{Z_A}{c^2}\mathbf{A}_\sigma(0)^2 + \frac{1}{2m_e}\frac{Z_B}{c^2}\mathbf{A}_\sigma(\mathbf{R})^2 + Q_R, \end{aligned}$$

where

$$Q_R = \sum_{i_A, j_B}\int_{\mathbb{R}^3} d\mathbf{k}\frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2}\Big(\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}} - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{i_A}-\mathbf{R})} - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}+\mathbf{R})} + \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}-\mathbf{x}_{i_A}+\mathbf{R})}\Big) \tag{2.4.3}$$

is the smeared interatomic Coulomb potential in relative coordinates.
Rewriting the operators (2.3.6) and (2.3.7) (which act on $\mathcal{H}_A \otimes \mathcal{F}_\sigma$ and $\mathcal{H}_B \otimes \mathcal{F}_\sigma$, respectively) perturbatively yields

$$H^A_\sigma(e) = (H_A + H_{f\geq\sigma}) + eH'_{\sigma,A} + e^2 H''_{\sigma,A}, \tag{2.4.4}$$

$$H^B_\sigma(e) = (H_B + H_{f\geq\sigma}) + eH'_{\sigma,B} + e^2 H''_{\sigma,B}, \tag{2.4.5}$$

where the perturbations are now given by

$$H'_{\sigma,A} = -\frac{1}{m_e c}\sum_{j_A=1}^{Z_A}(\mathbf{p}_{j_A}\cdot\mathbf{A}_\sigma(0)), \quad H''_{\sigma,A} = \frac{1}{2m_e}\frac{Z_A}{c^2}\mathbf{A}_\sigma(0)^2, \tag{2.4.6}$$

$$H'_{\sigma,B} = -\frac{1}{m_e c}\sum_{j_B=Z_A+1}^{N}(\mathbf{p}_{j_B}\cdot\mathbf{A}_\sigma(\mathbf{R})), \quad H''_{\sigma,B} = \frac{1}{2m_e}\frac{Z_B}{c^2}\mathbf{A}_\sigma(\mathbf{R})^2. \tag{2.4.7}$$

Recall from above that due to the introduction of relative coordinates, the unperturbed operator $H_\sigma^B(0)$ is independent of $\mathbf{R}$, so that the $\mathbf{R}$-dependence of the family $H_\sigma^B(e)$ is caused only by the perturbation operators.

Remark 2.4.1. A few conceptual issues are worth remarking on at this point. The Hamiltonians (2.4.1), (2.4.4) and (2.4.5) depend on the three paramaters e, Λ and $\mathbf{R}$. Our objective is to study the large-$|\mathbf{R}|$-behaviour of the interaction potential $V^\sigma(e,\Lambda,\mathbf{R})$ (see Section 1.1 or Section 2.8 below) derived from these operators, and - if possible - to gain results which are independent of the ultraviolet-cutoff Λ. To obtain a formula for $V^\sigma(e,\Lambda,\mathbf{R})$ which is accesible to further analysis, we first employ perturbation theory with respect to e, resulting in an expansion whose coefficients depend on Λ and $\mathbf{R}$. In a second step, we then analyze the $|\mathbf{R}|$-asymptotics of these coefficients, and later investigate their $(\Lambda\to\infty)$-behaviour.
Note that the Hamiltonians H_A and H_B depend on e via the atomic Coulomb potentials and therefore, strictly speaking, H_0^σ, $H_A+H_{f\geq\sigma}$ and $H_B+H_{f\geq\sigma}$ are not the constant terms (i.e. those resulting upon setting $e=0$) of the families (2.4.1), (2.4.4) and (2.4.5). This issue could be resolved by regarding the fine structure constant $\alpha=e^2/(4\pi\hbar c)$ as the coupling constant: noting that the definition of the coupling function (see (2.1.9)) implies

$$\frac{e}{c}\mathbf{A}_\sigma(\mathbf{x})\sim\frac{e}{c}\sqrt{\hbar c}\sim\frac{e}{\sqrt{c}}\sim\alpha^{1/2},$$

we find

$$\begin{aligned}
eH'_\sigma &= e\left(H'_{\sigma,A}+H'_{\sigma,B}\right)\sim\frac{e}{c}\left(\mathbf{A}_\sigma(0)+\mathbf{A}_\sigma(\mathbf{R})\right)\sim\alpha^{1/2},\\
e^2H''_\sigma &= e^2\left(H''_{\sigma,A}+H''_{\sigma,B}+Q_R\right)\\
&\sim\frac{e^2}{c^2}\left(\frac{Z_A}{2m_e}\mathbf{A}_\sigma(0)^2+\frac{Z_B}{2m_e}\mathbf{A}_\sigma(\mathbf{R})^2\right)+e^2Q_R\\
&\sim\alpha^1+\alpha^1+e^2\alpha^0,
\end{aligned}$$

so that setting $e=1$, we find that the interatomic Coulomb term e^2Q_R (as well as the atomic Hamiltonians H_A and H_B, as can be seen analogously) is of zeroth order with respect to α, while the remaining terms in $e^2H''_\sigma$ are of first order, and the perturbations $eH'_{\sigma,A}$, $eH'_{\sigma,A}$ and eH'_σ are of order $1/2$.
However, ultimately we are interested in the large-$|\mathbf{R}|$-behaviour of $V^\sigma(e,\Lambda,\mathbf{R})$, and we expect that the interatomic Coulomb potential e^2Q_R only makes a small contribution to this if $|\mathbf{R}|$ is large. This is why on the level of perturbation theory with respect to e, we split the interatomic Coulomb potential e^2Q_R from the atomic Coulomb potentials contained in H_A and H_B and (as mentioned in the introduction) suppress the e-dependence of the atomic Hamiltonians H_A and H_B. Mathematically, this can be regarded as treating e as an independent parameter for the families (2.4.1), (2.4.4) and (2.4.5), which we will henceforth view as quadratic operator families with respect to e. In Section 2.7 we apply standard results from analytic perturbation theory to the families (2.4.1), (2.4.4) and (2.4.5), obtaining series expansions for their ground states and ground state energies. The remaining e-dependence in the constant terms H_0^σ, $H_A+H_{f\geq\sigma}$ and $H_B+H_{f\geq\sigma}$ will manifest itself in the fact that these expansion are partial expansions with respect to the parameter e, in the sense that their coefficients still depend on the physical value of the electronic charge contained in these operators.

2.5 The atomic Schrödinger operators

In (2.4.4) and (2.4.5),

$$H_A := -\sum_{i_A=1}^{Z_A} \frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_{i_A}} + e^2 Q_A \tag{2.5.1}$$

and

$$H_B := -\sum_{j_B=Z_A+1}^{Z_A+Z_B} \frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_{j_B}} + e^2 \tilde{Q}_B \tag{2.5.2}$$

are Schrödinger operators describing the atoms A and B, respectively. Q_A is the smeared Coulomb potential from (2.1.4), and $\tilde{Q}_B$ corresponds to $Q_B(\mathbf{R})$ from (2.1.5) after switching to relative coordinates for the electrons of atom B. The operators H_A and H_B act on the Hilbert spaces $\mathcal{H}_A = L^2_{anti}(\mathbb{R}^{3Z_A})$ and $\mathcal{H}_B = L^2_{anti}(\mathbb{R}^{3Z_B})$, respectively. Note that the introduction of relative coordinates has eliminated the $\mathbf{R}$-dependence from the operator H_B. The $\mathbf{R}$-dependence involving the electrons is now solely contained in the interatomic Coulomb potential Q_R.
Next we collect some important properties of the operators H_A and H_B.

Proposition 2.5.1. *Suppose that the form factor ψ satisfies the assumptions (A1). Then*

i. H_A and H_B are self-adjoint operators on $L^2_{anti}(\mathbb{R}^{3Z_A})$ and $L^2_{anti}(\mathbb{R}^{3Z_B})$ with domains $D(H_A) = H^2(\mathbb{R}^{3Z_A}) \cap L^2_{anti}(\mathbb{R}^{3Z_A})$ and $D(H_B) = H^2(\mathbb{R}^{3Z_B}) \cap L^2_{anti}(\mathbb{R}^{3Z_B})$, respectively.

ii. Their spectra are of the form

$$\begin{aligned}\mathrm{spec}(H_A) &= \{E_A^i\}_{i=0}^{\infty} \cup [\Sigma_A, \infty),\\ \mathrm{spec}(H_B) &= \{E_B^i\}_{i=0}^{\infty} \cup [\Sigma_B, \infty),\end{aligned}$$

where the E_A^i and the E_B^i are isolated eigenvalues of finite multiplicity satisfying $E_A^0 \le E_A^1 \le \cdots < \Sigma_A$ and $E_B^0 \le E_B^1 \le \cdots < \Sigma_B$. In particular, H_A and H_B both have a ground state, and the corresponding ground state energies E_A^0 and E_B^0 are separated from the rest of the spectrum by finite gaps $\Delta_A > 0$, $\Delta_B > 0$.

iii. (Rotational and parity invariance) H_A and H_B are rotationally invariant in the sense that

$$U_R H_A = H_A U_R, \quad U_R H_B = H_B U_R \quad \forall R \in SO(3),$$

where the bounded operator U_R, $(U_R\Psi)(x) := \Psi([R^{-1} \times \cdots \times R^{-1}]x)$ belongs to the unitary representation of the diagonal of the group $SO(3)\times\cdots\times SO(3)$. Furthermore, H_A and H_B commute with the parity operators P_A and P_A, where

$$\begin{aligned}P_A\psi(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}) &= \psi(-\mathbf{x}_1, \ldots, -\mathbf{x}_{Z_A}),\\ P_B\psi(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_B}) &= \psi(-\mathbf{x}_1, \ldots, -\mathbf{x}_{Z_B}).\end{aligned}$$

iv. (Exponential decay of eigenfunctions)

(a) (L^p bounds on ground state eigenfunctions) Let Σ_A and Σ_B be the ionization thresholds of H_A and H_B, and let Ψ_A^0 and Ψ_B^0 be eigenfunctions of H_A and H_B corresponding to the lowest eigenvalues E_A^0 and E_B^0, respectively. Then for any $\alpha < 1$,

$$\Psi_A^0 \mathrm{e}^{\alpha\sqrt{\Sigma_A - E_A^0}|x|_A} \in L^\infty(\mathbb{R}^{3Z_A}) \cap L^2(\mathbb{R}^{3Z_A}),$$
$$\Psi_B^0 \mathrm{e}^{\alpha\sqrt{\Sigma_B - E_B^0}|x|_B} \in L^\infty(\mathbb{R}^{3Z_B}) \cap L^2(\mathbb{R}^{3Z_B}),$$

where $|x|_A = \sqrt{\sum_{i=1}^{Z_A} 2m_e(\mathbf{x}_i \cdot \mathbf{x}_i)}$ and $|x|_B = \sqrt{\sum_{i=Z_A+1}^{Z_A+Z_B} 2m_e(\mathbf{x}_i \cdot \mathbf{x}_i)}$.

(b) (Exponential decay of the one-particle densities) Let Ψ_A^0 and Ψ_B^0 be as in (a). Then the corresponding one-particle densities

$$\rho_{\Psi_A^0}(x) := Z_A \int_{\mathbb{R}^{3Z_A-3}} |\Psi_A^0(\mathbf{x}, \mathbf{x}_2, \ldots, \mathbf{x}_{Z_A})|^2 d\mathbf{x}_2 \ldots \mathbf{x}_{Z_A},$$
$$\rho_{\Psi_B^0}(x) := Z_B \int_{\mathbb{R}^{3Z_B-3}} |\Psi_B^0(\mathbf{x}, \mathbf{x}_2, \ldots, \mathbf{x}_{Z_B})|^2 d\mathbf{x}_2 \ldots \mathbf{x}_{Z_B}$$

satisfy

$$\rho_{\Psi_A^0}(\mathbf{x}) \leq C_A \, \mathrm{e}^{-C'_A|\mathbf{x}|},$$
$$\rho_{\Psi_B^0}(\mathbf{x}) \leq C_B \, \mathrm{e}^{-C'_B|\mathbf{x}|}$$

for suitable constants $C_A, C'_A, C_B, C'_B > 0$.

v. Let Ψ_A^0 and Ψ_B^0 be as in iv)(a). Then for any $i_A \in \{1, \ldots, Z_A\}$, $j_B \in \{Z_A + 1, \ldots, Z_A + Z_N\}$ and any $\alpha \in \{1, 2, 3\}$,

$$\mathbf{x}_{i_A}^\alpha \Psi_A^0 \in H^2(\mathbb{R}^{3Z_A}), \quad \mathbf{x}_{j_B}^\alpha \Psi_B^0 \in H^2(\mathbb{R}^{3Z_B}).$$

Proof. i) By rewriting Q_A and $\tilde{Q}_B$ in position space (using the fact that the Fourier transform converts products into convolutions, see A.5), one observes that each summand is of the form

$$W(x) = C \int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\psi(y')}{|x - y + y'|} = C(\psi * \psi) * \left(\frac{1}{|\cdot|}\right)(x),$$

with $x = x_i$ or $x = x_i - x_j$. Note that we have used the assumption that ψ is even. Since $\psi \in C_0^\infty(\mathbb{R}^3)$, so is $\psi * \psi$, and thus we conclude $W \in C^\infty(\mathbb{R}^3)$. In particular, W is bounded on compact subsets of $\mathbb{R}^3$. To estimate its decay at infinity, note that for $|x| \geq 4 \operatorname{diam} \operatorname{supp} \psi$ and $y, y' \in \operatorname{supp} \psi$ we have $|x - y + y'| \geq ||x| - |y - y'|| \geq |x| - 2 \operatorname{diam} \operatorname{supp} \psi \geq 1/2|x|$, and thus

$$|W(x)| \leq \frac{2C}{|x|} \int dy dy' \psi(y)\psi(y') = \frac{2C}{|x|},$$

since $\int \psi = 1$ by assumption. Thus we conclude that at infinity, W decays at least like the non-smeared Coulomb potential. Combining this with the boundedness on compact

subsets, we conclude $W \in C_b^\infty(\mathbb{R}^3)$, its supremum depending on the size of $\operatorname{supp}\psi$. (Note that if we rescale $\psi = \Lambda^3\psi_0(\Lambda\cdot)$ to form a Dirac sequence, then we recover the non-smeared Coulomb potential in the limit $\Lambda \to \infty$, which is of course unbounded near zero.) Thus $Q_A \in C_b^\infty(\mathbb{R}^{3Z_A})$ $\tilde{Q}_B \in C_b^\infty(\mathbb{R}^{3Z_B})$, and their infinitesimal relative boundedness with respect to the $3Z_A$- ($3Z_B$-) dimensional Laplacian is trivial, establishing the self-adjointness of H_A (H_B) on $D(H_A) = H^2(\mathbb{R}^{3Z_A}) \cap L^2_{anti}(\mathbb{R}^{3Z_A})$ ($D(H_B) = H^2(\mathbb{R}^{3Z_B}) \cap L^2_{anti}(\mathbb{R}^{3Z_B})$) via the Kato-Rellich theorem.
ii) This is basically Zhislin's theorem ([Žis60]). In our case of a smeared Coulomb potential, it can for instance be proven by a slight modification of the proof given in [Fri03].
iii) Note that U_R leaves $D(H_A) = H^2(\mathbb{R}^{3Z_A}) \cap L^2_{anti}(\mathbb{R}^{3Z_A})$ and $D(H_B) = H^2(\mathbb{R}^{3Z_B}) \cap L^2_{anti}(\mathbb{R}^{3Z_B})$ invariant. The operators

$$-\sum_{j_A=1}^{Z_A} \frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_{j_A}} \text{ and } -\sum_{j_B=Z_A+1}^{Z_A+Z_B} \frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_{j_B}}$$

commute with U_R, since every term in the sum is a Laplacian on $\mathbb{R}^3$ and thus commutes with any element of the representation of $SO(3)$. By assumption, the smeared charge distribution ψ, and thus also its Fourier transform $\hat{\psi}$, is invariant under rotations. Furthermore, $|R\mathbf{k}| = |\mathbf{k}|$ for any $R \in SO(3)$ and $\mathbf{k} \in \mathbb{R}^3$, so that the first assertion follows by a change of variables in the expressions defining Q_A and $\tilde{Q}_B$. The assertion on the parity operators follows similarly: P_A and P_B commute with $-\sum_{j_A=1}^{Z_A} \frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_{j_A}}$ and $-\sum_{j_B=Z_A+1}^{Z_A+Z_B} \frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_{j_B}}$, respectively (twofold differentiation produces $(-1)^2 = 1$), and $|-\mathbf{k}| = |\mathbf{k}|$, $\hat{\psi}(-\mathbf{k}) = \hat{\psi}(\mathbf{k})$ for all $\mathbf{k} \in \mathbb{R}^3$ (the latter by the above assumption on ψ).
iv)a) The smeared Coulomb potentials Q_A and $\tilde{Q}_B$ satisfy the condition (C3) from [DHSV79]: by the proof of i), each term W in the sums, viewed as a multiplication operator on $L^2(\mathbb{R}^3)$, is decomposable into an element of $L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ (actually the L^2-part can be chosen to be 0, see above) and satisfies $W(\mathbf{x}) \to 0$ as $|\mathbf{x}| \to \infty$. The results now follow from §5 and §7 of [DHSV79].
iv)b) See [AHOHOM81].
v) It suffices to verify that $\sum_{j=1}^{Z_A} \Delta_{\mathbf{x}_j}(\mathbf{x}_{i_A}^\alpha \Psi_A^0) \in L^2(\mathbb{R}^{3Z_A})$. To this end, note that

$$\sum_{j=1}^{Z_A} \Delta_{\mathbf{x}_j}(\mathbf{x}_{i_A}^\alpha \Psi_A^0) = 2\sum_{j=1}^{Z_A} \nabla_{\mathbf{x}_j}(\mathbf{x}_{i_A}^\alpha) \cdot \nabla_{\mathbf{x}_j}\Psi_A^0 + \mathbf{x}_{i_A}^\alpha \sum_{j=1}^{Z_A} \Delta_{\mathbf{x}_j}(\Psi_A^0).$$

Since $\Psi_A^0 \in H^2(\mathbb{R}^{3Z_A})$ is an eigenfunction, we have

$$\sum_{j=1}^{Z_A} \nabla_{\mathbf{x}_j}(\mathbf{x}_{i_A}^\alpha) \cdot \nabla_{\mathbf{x}_j}\Psi_A^0 = (\nabla_{\mathbf{x}_{i_A}}\Psi_A^0)_\alpha \in L^2(\mathbb{R}^{3Z_A}).$$

Using the eigenvalue equation for Ψ_A^0, we find

$$\mathbf{x}_{i_A}^\alpha \sum_{j=1}^{Z_A} \Delta_{\mathbf{x}_j}(\Psi_A^0) = \mathbf{x}_{i_A}^\alpha \left(\frac{2m_e}{\hbar}(e^2 Q_A(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}) - E_A^0)\Psi_A^0\right),$$

and the exponential decay estimates from iv) allow us to conclude $(\mathbf{x}_{i_A}^{\alpha}\Psi_A^0) \in L^2(\mathbb{R}^{3Z_A})$. Furthermore, $Q_A \in C_b^\infty(\mathbb{R}^{3Z_A})$ by the proof of i), so that also

$$Q_A(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})(\mathbf{x}_{i_A}^{\alpha}\Psi_A^0) \in L^2(\mathbb{R}^{3Z_A}),$$

finishing the proof. □

To prepare for the following, consider the standard atomic Schrödinger operators

$$\begin{aligned}\tilde{H}_A &= -\sum_{i=1}^{Z_A}\frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_i} + e^2V_A,\\ \tilde{H}_B &= -\sum_{i=1}^{Z_B}\frac{\hbar^2}{2m_e}\Delta_{\mathbf{x}_i} + e^2V_B\end{aligned} \tag{2.5.3}$$

describing two atoms A and B located at 0 and $\mathbf{R} \in \mathbb{R}^3$, respectively. Here the interaction is given by the Coulomb potentials

$$V_A = -\frac{1}{4\pi}\sum_{i=1}^{Z_A}\frac{Z_A}{|\mathbf{x}_i|} + \frac{1}{4\pi}\sum_{i<j}\frac{1}{|\mathbf{x}_i-\mathbf{x}_j|}$$

and

$$V_B = -\frac{1}{4\pi}\sum_{i=1}^{Z_B}\frac{Z_B}{|\mathbf{x}_i|} + \frac{1}{4\pi}\sum_{i<j}\frac{1}{|\mathbf{x}_i-\mathbf{x}_j|}.$$

Note that here we have already chosen the relative coordinates $\{\mathbf{x}_1+\mathbf{R},\ldots,\mathbf{x}_{Z_B}+\mathbf{R}\}$ for the operator $\tilde{H}_B$. The Hamiltonians $\tilde{H}_A$ and $\tilde{H}_B$ are self-adjoint on the domains $L^2_{anti}(\mathbb{R}^{3Z_A}) \cap H^2(\mathbb{R}^{3Z_A})$ and $L^2_{anti}(\mathbb{R}^{3Z_B}) \cap H^2(\mathbb{R}^{3Z_B})$, respectively (this was first proven in [Kat51]), and their spectra consist of a countable set of eigenvalues of finite multiplicity at the bottom, and a branch of essential spectrum stretching from the ionization threshold to infinity above these (see [Žis60]). In particular, $\tilde{H}_A$ and $\tilde{H}_B$ have ground states, and the corresponding eigenvalues are separated from the rest of the spectrum by finite gaps $\tilde{\Delta}_A$ and $\tilde{\Delta}_B$.

Assumption (A2): The ground states of the Schrödinger operators $\tilde{H}_A$ and $\tilde{H}_B$ (containing non-smeared Coulomb potentials) are non-degenerate.

The next proposition establishes the close connection between the Hamiltonians H_A, H_B (containing smeared Coulomb potentials) and their counterparts $\tilde{H}_A$, $\tilde{H}_B$.

Proposition 2.5.2. *Suppose that assumptions (A1) and (A2) are satisfied. Then there exists $\Lambda_0 > 0$ such that for $\Lambda \geq \Lambda_0$,*

i. The ground states of H_A and H_B are non-degenerate and are spanned by two normalized, antisymmetric wave functions Ψ_A^0 and Ψ_B^0.

ii. Ψ_A^0 and Ψ_B^0 can be chosen to be real functions.

iii. Ψ_A^0 *and* Ψ_B^0 *both have definite parity, i.e. they are eigenfunctions of the parity operators* P_A *and* P_B *(see the preceding proposition) with eigenvalues* $\varepsilon_A, \varepsilon_B \in \{1, -1\}$*, respectively. Furthermore,* Ψ_A^0 *and* Ψ_B^0 *are invariant under the representation of the diagonal of* $SO(3) \times \cdots \times SO(3)$*. Note that by the introduction of relative coordinates for atom* B*, both* Ψ_A^0 *and* Ψ_B^0 *are centered at* 0*.*

iv. The spectral gaps Δ_A *and* Δ_B *are bounded away from zero uniformly in* Λ*.*

Proof. i) and iv) follow from the fact that H_A and H_B converge to $\tilde{H}_A$ and $\tilde{H}_B$, respectively, in norm resolvent sense (see [RS80] for the definition). In particular (see [RS80], Thm. VIII.23), this convergence implies

$$\|P_{(a,b)}(H_A) - P_{(a,b)}(\tilde{H}_A)\| \underset{\Lambda\to\infty}{\to} 0,$$
$$\|P_{(a,b)}(H_B) - P_{(a,b)}(\tilde{H}_B)\| \underset{\Lambda\to\infty}{\to} 0$$

for all spectral projections $P_{(a,b)}$ corresponding to intervals (a, b) with endpoints belonging to the resolvent set of $\tilde{H}_A$ (respectively $\tilde{H}_B$). Choosing (a, b) so that the only point of spec $(\tilde{H}_A)$ (resp. spec $(\tilde{H}_B)$) it contains is an isolated eigenvalue $\tilde{E}$, and letting Λ be large, implies the existence of isolated eigenvalues E_i of H_A (resp. H_B) in the vicinity of $\tilde{E}$, their total multiplicity being equal to that of $\tilde{E}$. In particular, this implies the existence of non-degenerate ground states of H_A (resp. H_B) for large Λ. Furthermore, choosing Λ_0 so large that none of the eigenvalues of H_A and H_B obtained from those of $\tilde{H}_A$ and $\tilde{H}_B$ cross, we obtain uniform lower bounds for the spectral gaps Δ_A and Δ_B as Λ ranges over the interval $[\Lambda_0, \infty)$.
We will prove norm resolvent convergence of H_A to $\tilde{H}_A$, the other case being completely analogous. First note that H_A and $\tilde{H}_A$ have the common domain $H^2(\mathbb{R}^{3Z_A})$, so that by ([RS80], Theorem VIII.25 b)), it is sufficient to show that

$$\sup_{\|\varphi\|_H=1} \|(H_A - \tilde{H}_A)\varphi\|_H \underset{\Lambda\to\infty}{\to} = 0,$$

where $\|\varphi\|_H := \|\varphi\|_{L^2(\mathbb{R}^{3Z_A})} + \|H_A\varphi\|_{L^2(\mathbb{R}^{3Z_A})}$ is the graph norm of H_A, which is equivalent to the $H^2(\mathbb{R}^{3Z_A})$-norm by standard relative boundedness estimates. To this end, we first rewrite Q_A in position space (see A.5), which yields

$$\begin{aligned} &Q_A(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}) \\ &= -\frac{Z_A}{4\pi} \sum_{i_A=1}^{Z_A} \left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}_{i_A}) + \frac{1}{4\pi} \sum_{i_A<j_A} \left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}_{i_A} - \mathbf{x}_{j_A}). \end{aligned}$$

For any $\varphi \in D(H_A) = H^2(\mathbb{R}^{3Z_A})$ with $\|\varphi\|_H = 1$ we have

$$\begin{aligned} &\|(H_A - \tilde{H}_A)\varphi\|^2_{L^2(\mathbb{R}^{3Z_A})} \\ =&\|Q_A - V\|^2_{L^2(\mathbb{R}^{3Z_A})} \\ \leq&\frac{Z_A}{4\pi} \sum_{i_A=1}^{Z_A} \left\| \left(\left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}_{i_A}) - \frac{1}{|\mathbf{x}_{i_A}|} \right) \varphi \right\|^2_{L^2(\mathbb{R}^{3Z_A})} \qquad (2.5.4) \\ &+ \frac{1}{4\pi} \sum_{i_A<j_A} \left\| \left(\frac{1}{4\pi} \left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}_{i_A} - \mathbf{x}_{j_A}) - \frac{1}{|\mathbf{x}_{i_A} - \mathbf{x}_{j_A}|} \right) \varphi \right\|^2_{L^2(\mathbb{R}^{3Z_A})}. \qquad (2.5.5) \end{aligned}$$

Consider a typical term in (2.5.4) and assume without loss of generality that $i_A = 1$. Put

$$f(\mathbf{x}) := \left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}) - \frac{1}{|\mathbf{x}|}$$

and note that $f \in L^2_{loc}(\mathbb{R}^3)$. Recalling that by assumption we have $\psi = \Lambda^3 \psi_0(\Lambda \cdot)$, where $\psi_0 \in C_0^\infty(\mathbb{R}^3)$, $\int \psi_0 = 1$ and $\operatorname{supp} \psi_0 \subset B_1(0)$, we note that

$$\operatorname{supp}(\psi * \psi) \subset 2 \operatorname{supp} \psi \subset B_{2/\Lambda}(0).$$

Now choosing $L > 2/\Lambda$ and using the splitting $L^2(\mathbb{R}^3) = L^2(B_L(0)) \oplus L^2(\mathbb{R}^3 \setminus B_L(0))$ yields

$$\begin{aligned}
&\left\| \left(\left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}_{i_A}) - \frac{1}{|\mathbf{x}_{i_A}|} \right) \varphi \right\|^2_{L^2(\mathbb{R}^{3Z_A})} \\
=& \int_{\mathbb{R}^{3Z_A - 3}} d\mathbf{x}_2 \dots d\mathbf{x}_{Z_A} \int_{\mathbb{R}^3} |f(\mathbf{x}_1)|^2 |\varphi|^2(\mathbf{x}_1, \dots, \mathbf{x}_{Z_A}) d\mathbf{x}_1 \\
=& \int_{\mathbb{R}^{3Z_A - 3}} d\mathbf{x}_2 \dots d\mathbf{x}_{Z_A} \int_{\mathbb{R}^3 \setminus B_L(0)} |f(\mathbf{x}_1)|^2 |\varphi|^2(\mathbf{x}_1, \dots, \mathbf{x}_{Z_A}) d\mathbf{x}_1 \\
&+ \int_{\mathbb{R}^{3Z_A - 3}} d\mathbf{x}_2 \dots d\mathbf{x}_{Z_A} \int_{B_L(0)} |f(\mathbf{x}_1)|^2 |\varphi|^2(\mathbf{x}_1, \dots, \mathbf{x}_{Z_A}) d\mathbf{x}_1.
\end{aligned}$$

Outside $\operatorname{supp}(\psi * \psi)$ we have

$$\left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}) = \frac{1}{|\mathbf{x}|}$$

by Newton's theorem (see [LL97], Theorem 9.7), so that the first integral vanishes. As concerns the second one, we apply the Sobolev inequality (twice) to the function

$$\varphi(\cdot, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A}) \in H^2(\mathbb{R}^3)$$

to conclude that $\varphi(\cdot, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A}) \in L^\infty_{loc}(\mathbb{R}^3)$, with

$$\begin{aligned}
\|\varphi(\cdot, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A})\|_{L^\infty(B_L(0))} \leq& C \|\varphi(\cdot, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A})\|_{H^2(B_L(0))} \\
\leq& C \|\varphi(\cdot, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A})\|_{H^2(\mathbb{R}^3)}
\end{aligned}$$

for a suitable constant C which is independent of $\mathbf{x}_2, \dots, \mathbf{x}_{Z_A}$. This yields the estimate

$$\begin{aligned}
&\int_{\mathbb{R}^{3Z_A - 3}} d\mathbf{x}_2 \dots d\mathbf{x}_{Z_A} \int_{B_L(0)} |f(\mathbf{x}_1)|^2 |\varphi|^2(\mathbf{x}_1, \dots, \mathbf{x}_{Z_A}) d\mathbf{x}_1 \\
\leq& C^2 \int_{\mathbb{R}^{3Z_A - 3}} d\mathbf{x}_2 \dots d\mathbf{x}_{Z_A} \, \|\varphi(\cdot, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A})\|^2_{H^2(\mathbb{R}^3)} \int_{B_L(0)} |f(\mathbf{x}_1)|^2 d\mathbf{x}_1 \\
=& C^2 \|f\|^2_{L^2(B_L(0))} \int_{\mathbb{R}^{3Z_A}} \sum_{|\alpha| \leq 2} |D^\alpha_{\mathbf{z}} \varphi(\mathbf{z}, \mathbf{x}_2, \dots, \mathbf{x}_{Z_A})|^2 \, d\mathbf{z} d\mathbf{x}_2 \dots d\mathbf{x}_{Z_A} \\
\leq& C^2 \|f\|^2_{L^2(B_L(0))} \|\varphi\|^2_{H^2(\mathbb{R}^{3Z_A})} \leq \tilde{C} \|f\|^2_{L^2(B_L(0))} \underbrace{\|\varphi\|^2_H}_{=1},
\end{aligned}$$

where the last inequality follows from the equivalence of the norms $\|\cdot\|_{H^2(\mathbb{R}^{3Z_A})}$ and $\|\cdot\|_H$, see above. By standard results on mollification with C_0^∞-functions (see e.g. [Eva98]), we have $f \to 0$ in $L^2_{loc}(\mathbb{R}^3)$, which shows that

$$\sup_{\|\varphi\|_H=1} (2.5.4) \underset{\Lambda\to\infty}{\to} = 0.$$

The corresponding assertion for the terms in (2.5.5) is proved similarly by introducing relative coordinates (in $\mathbb{R}^{3Z_A}$) for each pair $(\mathbf{x}_{i_A}, \mathbf{x}_{j_A})$.
ii) Since H_A and H_B are real operators (in the sense that they commute with the involution $\psi \mapsto \overline{\psi}$), Ψ_A^0 and Ψ_B^0 can be chosen to be real functions.
iii) Since H_A and H_B are rotationally invariant and commute with the parity operators (see Proposition 2.5.1), the non-degeneracy of E_A^0 and E_B^0 implies that each of Ψ_A^0 and Ψ_B^0 has to lie in a one-dimensional common eigenspace of the Hamiltonian and the generators of the Lie group $\{\underbrace{(R,\ldots,R)}_{Z_{A,B} \text{ times}}, R \in SO(3)\}$, which are the angular momentum operators $\mathbf{L}_\alpha = \sum_{i=1}^{Z_{A,B}} (\mathbf{x}_i \times \mathbf{p}_i)_\alpha$, $\alpha = 1,2,3$. The only one-dimensional eigenspaces of these operators are those corresponding to the eigenvalue $L = 0$, and thus the representation of the operator U_R, $U_R\psi = \psi(R^{-1}\mathbf{x}_1, \ldots, R^{-1}\mathbf{x}_{Z_{A,B}})$ in terms of these generators yields the assertion. □

2.6 Non-degenerate isolated ground state energy and reduced resolvents

In this section we investigate the unperturbed operators $H_\sigma(0,\mathbf{R})$, $H_\sigma^A(0)$ and $H_\sigma^B(0)$ from the quadratic operator families (2.4.1), (2.4.4) and (2.4.5). We will establish essential self-adjointness and (assuming the hypotheses of Proposition 2.5.2) the existence of non-degenerate ground states for the closures, corresponding to the isolated eigenvalues $E_0 = E_A^0 + E_B^0$, E_A and E_B, respectively.
To this end, we first prove some basic results on the tensor product of subspaces and on the relation between reducing subspaces and self-adjoint closures of operators. Following this, we prove an abstract result about the ground state energies and the ground state eigenspaces of (closures of) operators with a 'non-interacting' structure

$$A \otimes I + I \otimes B,$$

where A and B are self-adjoint and possess non-degenerate, isolated eigenvalues at the bottom of their spectra. This result is then applied to the three unperturbed operators corresponding to the compound system and the two individual systems.
We begin with a few definitions and remarks on notation. Throughout, H will denote a Hilbert space. For two (arbitrary) subspaces $U_1 \subset H_1$ and $U_2 \subset H_2$, the algebraic tensor product is defined as

$$U_1 \widehat{\otimes} U_2 := \text{span}\,\{u_1 \otimes u_2,\ u_1 \in U_1, u_2 \in U_2\}.$$

The tensor product $U_1 \otimes U_2$ of two subspaces is defined as the completion of $U_1 \widehat{\otimes} U_2$ with respect to the product norm inherited from $H_1 \widehat{\otimes} H_2$:

$$U_1 \otimes U_2 := \overline{U_1 \widehat{\otimes} U_2}.$$

We will make use of both the $U_1 \otimes U_2$ and the $\overline{U_1 \widehat{\otimes} U_2}$ notation, depending on the situation. Note that even if U_1 and U_2 are both closed, $U_1 \widehat{\otimes} U_2$ need not be closed. However, in the special case that both U_1 and U_2 are closed and at least one of them is finite-dimensional,

$$\overline{U_1 \widehat{\otimes} U_2} = U_1 \widehat{\otimes} U_2,$$

see the proof of Lemma 2.6.1 below.
Given two closable operators $A : D(A) \to H_1$ and $B : D(B) \to H_2$, their tensor product

$$A \otimes B : D(A) \widehat{\otimes} D(B) \to H_1 \otimes H_2$$

is defined by setting $(A \otimes B)(u_1 \otimes u_2) := Au_2 \otimes Bu_2$ and extending linearly. It is a standard fact that $A \otimes B$ is closable (see e.g. [RS80]), its closure being denoted by

$$\overline{A \otimes B}.$$

Analogous to the case of subspaces, $A \otimes B$ need not to be closed, even if A and B are both closed operators.
The orthogonal projection onto a closed subspace U of a Hilbert space H will be denoted by P_U, and I_U will denote the identity operator on the smaller Hilbert space U.
A closed subspace $U \subset H$ is called a reducing subspace for a linear operator $A : D(A) \to H$ if $P_U A \subset A P_U$. In this case the restrictions $A_{|U}$ and $A_{|U^\perp}$ are well-defined.

Lemma 2.6.1. *Let $U_1 \subset H_1$ and $U_2 \subset H_2$ be closed subspaces of the Hilbert spaces H_1 and H_1, and let P_{U_1} and P_{U_2} be the associated orthogonal projections. Then*

i. $P_{\overline{U_1 \widehat{\otimes} U_2}} = \overline{P_{U_1} \otimes P_{U_2}}$.

ii. If in addition U_1 (or U_2) is finite-dimensional, then

$$\mathrm{Ran}(\overline{P_{U_1} \otimes P_{U_2}}) = \overline{U_1 \widehat{\otimes} U_2} = U_1 \widehat{\otimes} U_2 = \mathrm{Ran}(P_{U_1} \otimes P_{U_2}).$$

Proof. i) Let $u \in H_1 \otimes H_2$. Since $\overline{P_{U_1} \otimes P_{U_2}}$ is the closure of the bounded operator $P_{U_1} \otimes P_{U_2}$ with domain $H_1 \widehat{\otimes} H_2$, we can find a sequence $(u_n) \subset H_1 \widehat{\otimes} H_2$ such that $u_n \to u$ and

$$\begin{aligned}(\overline{P_{U_1} \otimes P_{U_2}})^2 u &= \lim_{n\to\infty} \left((P_{U_1} \otimes P_{U_2})^2 u_n\right) \\ &= \lim_{n\to\infty} \left((P_{U_1}^2 \otimes P_{U_2}^2) u_n\right) \\ &= \lim_{n\to\infty} \left((P_{U_1} \otimes P_{U_2}) u_n\right) \\ &= (\overline{P_{U_1} \otimes P_{U_2}}) u,\end{aligned}$$

which shows that $(\overline{P_{U_1} \otimes P_{U_2}})$ is a projection. Self-adjointness follows similarly, and thus $(\overline{P_{U_1} \otimes P_{U_2}})$ is an orthogonal projection. It is left to show that

$$\mathrm{Ran}\,(\overline{P_{U_1} \otimes P_{U_2}}) = \overline{U_1 \widehat{\otimes} U_2}.$$

Noting that

$$\overline{U_1 \widehat{\otimes} U_2} = \overline{\mathrm{Ran}\, P_{U_1} \widehat{\otimes} \mathrm{Ran}\, P_{U_2}} = \overline{\mathrm{Ran}\,(P_{U_1} \otimes P_{U_2})},$$

it suffices to show that

$$\mathrm{Ran}\,(\overline{P_{U_1}\otimes P_{U_2}}) = \overline{\mathrm{Ran}\,(P_{U_1}\otimes P_{U_2})}.$$

To this end, note that trivially, $\mathrm{Ran}\,(P_{U_1}\otimes P_{U_2}) \subset \mathrm{Ran}\,(\overline{P_{U_1}\otimes P_{U_2}})$, the latter being a closed subspace since $\overline{P_{U_1}\otimes P_{U_2}}$ is an orthogonal projection. Since the closure of a set is its smallest closed superset, we conclude

$$\overline{\mathrm{Ran}\,(P_{U_1}\otimes P_{U_2})} \subset \mathrm{Ran}\,(\overline{P_{U_1}\otimes P_{U_2}}).$$

For the converse inclusion let $v \in \mathrm{Ran}\,(\overline{P_{U_1}\otimes P_{U_2}})$, pick a preimage u and choose a sequence $(u_n) \subset H_1\widehat{\otimes}H_2$ with $u_n \to u$, which is possible by the construction of the closure. Then

$$(P_{U_1}\otimes P_{U_2})u_n = \overline{P_{U_1}\otimes P_{U_2}}u_n \to \overline{P_{U_1}\otimes P_{U_2}}u = v,$$

the sequence on the left consisting of elements of $\mathrm{Ran}\,(P_{U_1}\otimes P_{U_2})$. This proves $v \in \overline{\mathrm{Ran}\,(P_{U_1}\otimes P_{U_2})}$ and thus

$$\mathrm{Ran}\,(\overline{P_{U_1}\otimes P_{U_2}}) \subset \overline{\mathrm{Ran}\,(P_{U_1}\otimes P_{U_2})}.$$

ii) The first equality was already shown in i), so it is left to prove the second one. This is a general result for the algebraic tensor product of two closed subspaces, one of which is finite-dimensional, and is proven as follows. Suppose U_1 is finite-dimensional. For an element $u \in \overline{U_1\widehat{\otimes}U_2}$, choose a sequence $(u_n) \subset U_1\widehat{\otimes}U_2$ with $u_n \to u$. Using an orthonormal basis $\{e_i\}_{i=1}^d$ of U_1, any member of the sequence can be written as

$$u_n = \sum_{i=1}^{d} c_i^n(e_i\otimes a_i^n) = \sum_{i=1}^{d}(e_i\otimes c_i^n\, a_i^n)$$

for suitable coefficients $c_i^n \in \mathbb{C}$ and vectors $a_i^n \in U_2$. The Cauchy property, the fact that $\{e_i\}$ is an orthonormal basis and the closedness of U_2 then imply that the sequences $c_i^n\, a_i^n$, $i = 1,\ldots,d$, converge in U_2 (their limits denoted by a_i), which leads to the representation

$$u = \lim_{n\to\infty} u_n = \lim_{n\to\infty}\sum_{i=1}^{d}(e_i\otimes c_i^n\, a_i^n) = \sum_{i=1}^{d}(e_i\otimes a_i),$$

and the latter is evidently an element of $U_1\widehat{\otimes}U_2$. □

Lemma 2.6.2. *Let $A : D(A) \to H$ be densely defined and closable, and let $U \subset H$ be a reducing subspace for A. Then U is a reducing subspace for $\overline{A}$. Furthermore, the restrictions $A_{|U} : D(A)\cap U \to U$ and $A_{|U^\perp} : D(A)\cap U^\perp \to U^\perp$ are densely defined and closable (as operators on the Hilbert spaces U and $U^\perp$), and*

$$\overline{(A_{|U})} = (\overline{A})_{|U}\,,\ \overline{\left(A_{|U^\perp}\right)} = (\overline{A})_{|U^\perp}\,. \tag{2.6.1}$$

Proof. i) To show that U reduces $\overline{A}$ we have to verify $P_U\overline{A} \subset \overline{A}P_U$. To this end, let $u \in D(\overline{A})$. By definition of the closure, there exists a sequence $\{u_n\} \subset D(A)$ such that $u_n \to u$ and $Au_n \to \overline{A}u$. Using that P_U is continuous and that U reduces A, we find

$$P_U\overline{A}u = \lim_{n\to\infty} P_U A u_n = \lim_{n\to\infty} AP_U u_n.$$

Now $P_U u_n \to P_U u$ by continuity, so that the fact that A is closable implies

$$\lim_{n\to\infty} AP_U u_n = \overline{A}\left(\lim_{n\to\infty} P_U u_n\right) = \overline{A}P_U u.$$

This shows that $P_U D(\overline{A}) \subset D(\overline{A})$ and that $P_U\overline{A} = \overline{A}P_U$ on $D(\overline{A}) = D(P_U\overline{A})$, which suffices to prove $P_U\overline{A} \subset \overline{A}P_U$.
ii) The domains $D(A_{|U}) = D(A) \cap U$ and $D(A_{|U^\perp}) = D(A) \cap U^\perp$ are dense in U and $U^\perp$, respectively, since U is a reducing subspace and therefore $U \ominus D(A_{|U}) = (D(A))^\perp \cap U = \{0\}$ and $U^\perp \ominus D(A_{|U^\perp}) = \{0\}$, see [Wei76]. To see that $A_{|U}$ is closable, consider a sequence $u_n \subset D(A_{|U})$ with $u_n \to 0$ and $A_{|U}u_n \to u$ for some u. With respect to the decomposition $H = U \oplus U^\perp$, define $\tilde{u}_n := (u_n, 0) \subset D(A)$, the latter inclusion holding since U is a reducing subspace for A. Obviously, $\tilde{u}_n \to 0$ and

$$A\tilde{u}_n = (A_{|U}u_n, A_{|U^\perp}0) = (A_{|U}u_n, 0) \to (u, 0).$$

Since A is closable, we conclude $u = 0$. The proof that $A_{|U^\perp}$ is closable is analogous.
iii) We prove the first identity in (2.6.1), the proof of the other being completely analogous. Let $u \in D(\overline{(A_{|U})}) \subset U$. By the definition of the closure, there exists a sequence $\{u_n\} \subset D(A_{|U}) \subset U$ such that $u_n \to u$ and

$$\overline{(A_{|U})}u = \lim_{n\to\infty} A_{|U}u_n = \lim_{n\to\infty} Au_n = \overline{A}u = (\overline{A})_{|U}u, \tag{2.6.2}$$

where the second to last equality holds since A is closable. This shows that $u \in D(\overline{A}) \cap U = D((\overline{A})_{|U})$. Conversely, let $u \in D((\overline{A})_{|U})$. By the definition of the closure $\overline{A}$, there exists a sequence $\{\tilde{u}_n\} \subset D(A)$ (not necessarily lying in U), such that $\tilde{u}_n \to u$ and $A\tilde{u}_n \to \overline{A}u$. We have

$$(\overline{A})_{|U}u = \overline{A}u = \lim_{n\to\infty} A\tilde{u}_n = \lim_{n\to\infty} A\left(P_U\tilde{u}_n + P_{U^\perp}\tilde{u}_n\right) = \lim_{n\to\infty} \left(AP_U\tilde{u}_n + AP_{U^\perp}\tilde{u}_n\right). \tag{2.6.3}$$

Since U is a reducing subspace for $\overline{A}$ and A, the spaces U and $U^\perp$ are invariant under these operators, which implies $\overline{A}u \in U$, $AP_U\tilde{u}_n \in U$ and $AP_{U^\perp}\tilde{u}_n \in U^\perp$. Applying $P_{U^\perp}$ and P_U to (2.6.3) and using their continuity now yields

$$P_{U^\perp}(\overline{A}u) = 0 = \lim_{n\to\infty} AP_{U^\perp}\tilde{u}_n$$

and

$$P_U\overline{A}u = \overline{A}u = \lim_{n\to\infty} P_U A P_U \tilde{u}_n = \lim_{n\to\infty} AP_U\tilde{u}_n = \lim_{n\to\infty} A_{|U}P_U\tilde{u}_n. \tag{2.6.4}$$

In particular, the limit on the right-hand side exists. The continuity of P_U implies $P_U\tilde{u}_n \to P_U u = u$, and thus the closability of $A_{|U}$ yields

$$\lim_{n\to\infty} A_{|U}P_U\tilde{u}_n = \overline{(A_{|U})}\left(\lim_{n\to\infty} P_U\tilde{u}_n\right) = \overline{(A_{|U})}u, \tag{2.6.5}$$

which shows that $u \in D(\overline{(A_{|U})})$. Together with the above inclusion, this yields

$$D(\overline{(A_{|U})} = D((\overline{A})_{|U}),$$

and by (2.6.2), (2.6.3) (2.6.4) and (2.6.5), the action of the two operators on this domain coincides, finishing the proof. □

Corollary 2.6.3. *Let $A : D(A) \to H$ be densely defined, symmetric and essentially self-adjoint, and let $U \subset H$ be a reducing subspace for A. Then U is a reducing subspace for $\overline{A}$. Furthermore, the restrictions $A_{|U} : D(A) \cap U \to U$ and $A_{|U^\perp} : D(A) \cap U^\perp \to U^\perp$ are essentially self-adjoint (as operators on the Hilbert spaces U and $U^\perp$), and*

$$\overline{(A_{|U})} = (\overline{A})_{|U}\,,\ \overline{\left(A_{|U^\perp}\right)} = (\overline{A})_{|U^\perp}\,.$$

Proof. The assertion follows from Lemma 2.6.2 and the fact that restrictions of self-adjoint operators to reducing subspaces are self-adjoint. □

Proposition 2.6.4. *Let $A : D(A) \to H_1$ and $B : D(B) \to H_2$ be self-adjoint operators with non-degenerate eigenvalues E_A and E_B at the bottom of their spectra, and suppose that these eigenvalues are separated from the rest of the spectrum by finite gaps $\Delta_A, \Delta_B > 0$. Let Ψ_A^0 and Ψ_B^0 be ground state eigenfunctions corresponding to E_A and E_B, and consider the orthogonal decomposition*

$$\begin{aligned} &H_1 \otimes H_2 = U_1 \oplus U_2 \oplus U_3 \oplus U_4 \\ :=&(\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}) \oplus (\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp) \oplus (\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}) \oplus \overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp} \end{aligned}$$

into closed subspaces. Furthermore, define the operator $C := A \otimes I + I \otimes B$ with domain $D(C) := D(A)\widehat{\otimes}D(B)$. Then

i. The U_i are reducing subspace for both C and its self-adjoint closure $\overline{C}$.

ii. $E_A + E_B$ is a non-degenerate eigenvalue of $\overline{C}$ which is separated from the rest of its spectrum by $\min\{\Delta_A, \Delta_B\}$. The corresponding ground state is $\Psi_A^0 \otimes \Psi_B^0$.

iii.

$$\operatorname{spec}\left((\overline{C} - (E_A + E_B))_{|U_1^\perp}\right) \subset [\min\{\Delta_A, \Delta_B\}, \infty)\,.$$

In particular,

$$\left((\overline{C} - (E_A + E_B))_{|U_1^\perp}\right)^{-1}$$

exists and is bounded .

Proof. i) By [RS80], Section VIII.10, C is essentially self-adjoint. To show that the U_i are reducing subspaces for C, we have to verify

$$P_{U_i} C \subset C P_{U_i}, \tag{2.6.6}$$

which amounts to showing that the inclusion

$$D(P_{U_i} C) = D(C) \subset D(C P_{U_i}) = \{u \in H_1 \otimes H_2 |\, P_{U_i} u \in D(C)\}$$

between their domains holds and that $P_{U_i}C = CP_{U_i}$ on $D(C)$. Since U_1, U_2, U_3 are algebraic tensor products (which are closed nevertheless since $\{\Psi_A^0\}$ and $\{\Psi_B^0\}$ are one-dimensional, see Lemma 2.6.1), the corresponding orthogonal projections are given by $P_{\{\Psi_A^0\}} \otimes P_{\{\Psi_B^0\}}$, $P_{\{\Psi_A^0\}} \otimes P_{\{\Psi_B^0\}^\perp}$ and $P_{\{\Psi_A^0\}^\perp} \otimes P_{\{\Psi_B^0\}}$ (no closures), which comprise spectral projections of A and B, respectively. The fact that self-adjoint operators commute with all their spectral projections now implies (2.6.6) for $i = 1, 2, 3$. As regards U_4, note that on $D(C) = D(A)\widehat{\otimes}D(B)$, the corresponding orthogonal projection $\overline{P_{\{\Psi_A^0\}^\perp} \otimes P_{\{\Psi_B^0\}^\perp}}$ acts as $P_{\{\Psi_A^0\}^\perp} \otimes P_{\{\Psi_B^0\}^\perp}$, so that

$$\overline{P_{\{\Psi_A^0\}^\perp} \otimes P_{\{\Psi_B^0\}^\perp}}[D(C)] \subset D(C)$$

and $(\overline{P_{\{\Psi_A^0\}^\perp} \otimes P_{\{\Psi_B^0\}^\perp}})C = C(\overline{P_{\{\Psi_A^0\}^\perp} \otimes P_{\{\Psi_B^0\}^\perp}})$ on $D(C)$ again follow from the fact that $P_{\{\Psi_A^0\}^\perp}$ and $P_{\{\Psi_B^0\}^\perp}$ are spectral projections of A and B, respectively.
Corollary 2.6.3 implies that the U_i are also reducing subspaces for $\overline{C}$ and that taking the closure and restricting to the U_i commutes:

$$\big(\overline{C}\big)_{|U_i} = \overline{\big(C_{|U_i}\big)}, \quad i = 1, 2, 3, 4. \tag{2.6.7}$$

This finishes the proof of i).
ii), iii) Assume without loss of generality that $E_A = E_B = 0$. By [RS80], Section VIII.10, the spectrum of $\overline{C}$ is given by

$$\mathrm{spec}(\overline{C}) = \overline{\mathrm{spec}(A) + \mathrm{spec}(B)} \subset \{0\} \dot{\cup} [\min\{\Delta_A, \Delta_B\}, \infty). \tag{2.6.8}$$

Obviously, $\Psi_A^0 \otimes \Psi_B^0$ is an eigenvector of C and $\overline{C}$ corresponding to the eigenvalue 0, and the fact that it is separated from the rest of $\mathrm{spec}(\overline{C})$ is apparent from (2.6.8). To show that 0 is a non-degenerate eigenvalue of $\overline{C}$, assume that there exists another eigenvector v corresponding to 0. Without loss of generality, v can be chosen to be orthogonal to $\Psi_A^0 \otimes \Psi_B^0$, which implies $v \in U_2 \oplus U_3 \oplus U_4$. By (2.6.7) and the definition of C, the restriction of $\overline{C}$ to U_2 is given by

$$\big(\overline{C}\big)_{|U_2} = \overline{\big(C_{|U_2}\big)} = \overline{A_{|\{\Psi_A^0\}} \otimes I_{\{\Psi_B^0\}^\perp} + I_{\{\Psi_A^0\}} \otimes B_{|\{\Psi_B^0\}^\perp}}.$$

Noting that $A_{|\{\Psi_A^0\}}$ and $B_{|\{\Psi_B^0\}^\perp}$ are self-adjoint operators on the Hilbert spaces $\{\Psi_A^0\}$ and $\{\Psi_B^0\}^\perp$, respectively, we conclude from [RS80], Section VIII.10, that

$$\mathrm{spec}\left((\overline{C})_{|U_2}\right) \subset [\Delta_B, \infty).$$

Similarly, one deduces

$$\mathrm{spec}\left((\overline{C})_{|U_3}\right) \subset [\Delta_A, \infty)$$

and

$$\mathrm{spec}\left((\overline{C})_{|U_4}\right) \subset [\Delta_A + \Delta_B, \infty).$$

Combining these three inclusions, we conclude that a) they are in contradiction to the assumptions that $v \in U_2 \oplus U_3 \oplus U_4$ is an eigenvector corresponding to the eigenvalue 0, which proves ii), and b) that

$$\mathrm{spec}\left((\overline{C})_{|U_1^\perp}\right) = \mathrm{spec}\left((\overline{C})_{|(U_2 \oplus U_3 \oplus U_4)}\right) \subset [\min\{\Delta_A, \Delta_B\}, \infty)\,,$$

which proves iii). □

Next we will apply the preceding result (or more precisely, its generalization to three operators, which is straightforward) to the operators

$$\begin{aligned} H_\sigma(0,\mathbf{R}) &= H_0^\sigma \\ &= H_A \otimes I_{\mathcal{H}_B} \otimes I_{\mathcal{F}} + I_{\mathcal{H}_A} \otimes H_B \otimes I_{\mathcal{F}} + I_{\mathcal{H}_A} \otimes I_{\mathcal{H}_B} \otimes H_{f\geq\sigma}, \qquad (2.6.9) \\ H_\sigma^A(0) &= H_A \otimes I_{\mathcal{F}} + I_{\mathcal{H}_A} \otimes H_{f\geq\sigma}, \qquad (2.6.10) \\ H_\sigma^B(0) &= H_B \otimes I_{\mathcal{F}} + I_{\mathcal{H}_B} \otimes H_{f\geq\sigma} \qquad (2.6.11) \end{aligned}$$

from (2.4.1), (2.4.4) and (2.4.5), with domains

$$\begin{aligned} D(H_\sigma(0,\mathbf{R})) =& D(H_A)\widehat{\otimes}D(H_B)\widehat{\otimes}D(H_{f\geq\sigma}) \subset \mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}_\sigma, \\ D(H_\sigma^A(0)) =& D(H_A)\widehat{\otimes}D(H_{f\geq\sigma}) \subset \mathcal{H}_A \otimes \mathcal{F}_\sigma, \\ D(H_\sigma^B(0)) =& D(H_B)\widehat{\otimes}D(H_{f\geq\sigma}) \subset \mathcal{H}_B \otimes \mathcal{F}_\sigma, \end{aligned}$$

respectively.

Proposition 2.6.5. *Assume (A1),(A2) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Let Ψ_A^0 and Ψ_B^0 be the non-degenerate ground states of H_A and H_B (which exist by Proposition 2.5.2), and let E_A and E_B be the corresponding atomic ground state energies and Δ_A, Δ_B the corresponding spectral gaps.*
Then the closures of (2.6.9), (2.6.10) and (2.6.11), which we will denote by the same symbols, are self-adjoint and have the non-degenerate eigenvalues $E_0 = E_A^0 + E_B^0$, E_A and E_B at the bottom of their respective spectra. These eigenvalues are separated from the remaining spectra by the finite spectral gaps $\min\{\Delta_A, \Delta_B, \hbar\sigma\}$, $\min\{\Delta_A, \hbar\sigma\}$ and $\min\{\Delta_B, \hbar\sigma\}$, respectively, and the corresponding ground states are $\Psi_A^0 \otimes \Psi_B^0 \otimes \Omega$, $\Psi_A^0 \otimes \Omega$ and $\Psi_B^0 \otimes \Omega$. Furthermore,

$$\begin{aligned} \operatorname{spec}\left((H_0^\sigma - E_0)_{|\{\Psi_0\}^\perp}\right) &= [\min\{\Delta_A, \Delta_B, \hbar\sigma\}, \infty), \qquad (2.6.12) \\ \operatorname{spec}\left((\overline{H_A + H_{f\geq\sigma}} - E_A^0))_{|\{\Psi_A^0\otimes\Omega\}^\perp}\right) &= [\min\{\Delta_A, \hbar\sigma\}, \infty), \qquad (2.6.13) \\ \operatorname{spec}\left((\overline{H_B + H_{f\geq\sigma}} - E_B^0))_{|\{\Psi_B^0\otimes\Omega\}^\perp}\right) &= [\min\{\Delta_B, \hbar\sigma\}, \infty), \qquad (2.6.14) \end{aligned}$$

and in particular the reduced resolvents

$$\begin{aligned} T^\sigma :=& ((H_0^\sigma - E_0)_{|\{\Psi_0\}^\perp})^{-1}, \quad \Psi_0 = \Psi_A^0 \otimes \Psi_B^0 \otimes \Omega, \\ T_A^\sigma :=& ((\overline{H_A + H_{f\geq\sigma}} - E_A^0))_{|\{\Psi_A^0\otimes\Omega\}^\perp})^{-1}, \\ T_B^\sigma :=& ((\overline{H_B + H_{f\geq\sigma}} - E_B^0))_{|\{\Psi_B^0\otimes\Omega\}^\perp})^{-1} \end{aligned}$$

exist as bounded operators.

Proof. By the assumptions and Proposition 2.5.2, the lowest eigenvalues E_A^0 and E_B^0 of the self-adjoint operators H_A and H_B are non-degenerate and have finite spectral gaps Δ_A and Δ_B. Furthermore, $H_{f\geq\sigma}$ is self-adjoint with

$$\operatorname{spec}(H_{f\geq\sigma}) = \{0\}\dot{\cup}[\hbar\sigma, \infty),$$

where 0 is a non-degenerate eigenvalue which is separated from the rest of the spectrum of $H_{f\geq\sigma}$ by the gap $\hbar\sigma$ (see the construction of the infrared regularization in Section 2.3). Thus all the assumptions of Proposition 2.6.4 are fulfilled, and the assertion follows. Note that the additional claim about equality in (2.6.12) through (2.6.14) follows from the fact that $\mathrm{spec}(H_{f\geq\sigma})$ contains the continuous part $[\hbar\sigma,\infty)$. □

Note that both E_0 and Ψ_0 are are independent of σ, since $\inf\mathrm{spec}(H_{f\geq\sigma})=0$, $H_{f\geq\sigma}\Omega=0$ for all $\sigma\geq 0$ (the vacuum sector is left unchanged by the infrared regularization).

2.7 Analyticity of infrared-regularized ground states and eigenvalues

We are now in a situation in which analytic perturbation theory under its standard assumptions (see e.g.[Kat80]) is applicable, and the next proposition collects the ensuing results.

Proposition 2.7.1. *Let assumptions (A1) and (A2) be satisfied and let $\Lambda\geq\Lambda_0$, with Λ_0 as in Proposition 2.5.2. Then $H_\sigma(e,\mathbf{R})$, $H^A_\sigma(e)$ and $H^B_\sigma(e)$ are self-adjoint analytic families (with respect to the parameter e) of type (A) (in the sense of Kato) on $D(H^\sigma_0)\subset L^2(\mathbb{R}^{3N})\otimes\mathcal{F}$, $D(H_A+H_{f\geq\sigma})$ and $D(H_A+H_{f\geq\sigma})$, respectively. For $|e|\leq e_0$ (the latter depending on σ and Λ), the operators $H_\sigma(e,\mathbf{R})$, $H^A_\sigma(e)$ and $H^B_\sigma(e)$ have non-degenerate eigenvalues $E^\sigma(e,\mathbf{R})$, $E^\sigma_A(e)$ and $E^\sigma_B(e)$ at the bottom of their respective spectra. The corresponding eigenvectors are denoted by $\psi^\sigma(e,\mathbf{R})$, $\psi^\sigma_A(e)$ and $\psi^\sigma_B(e)$. In a neighbourhood of 0 (depending on σ), these objects are analytic functions of e, given by the series expansions*

$$E^\sigma(e,\mathbf{R})=\sum_{i=0}^{\infty}e^i\,E^\sigma_i(\mathbf{R}),\quad \psi^\sigma(e,\mathbf{R})=\sum_{i=0}^{\infty}e^i\psi^\sigma_i(\mathbf{R}),\tag{2.7.1}$$

$$E^\sigma_A(e)=\sum_{i=0}^{\infty}e^i\,E^\sigma_{i,A},\quad \psi^\sigma_A(e)=\sum_{i=0}^{\infty}e^i\psi^\sigma_{i,A},\tag{2.7.2}$$

$$E^\sigma_B(e)=\sum_{i=0}^{\infty}e^i\,E^\sigma_{i,B},\quad \psi^\sigma_B(e)=\sum_{i=0}^{\infty}e^i\psi^\sigma_{i,B},\tag{2.7.3}$$

where $E^\sigma_{0,A}=E^0_A$, $E^\sigma_{0,B}=E^0_B$, $\psi^\sigma_{0,A}=\Psi^0_A\otimes\Omega$, $\psi^\sigma_{0,B}=\Psi^0_B\otimes\Omega$, $\psi^\sigma_0(\mathbf{R})=\Psi^0_A\otimes\Psi^0_B\otimes\Omega$ are independent of σ and $E^\sigma_0(R)=E_0=E^0_A+E^0_B$ is independent of both σ and R.

Proof. We will give the proof for $H_\sigma(e,\mathbf{R})$. The other two assertions are proven analogously. H^σ_0 is self-adjoint on $D(H^\sigma_0)\subset\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}$, and H'_σ and H''_σ are symmetric and relatively bounded with respect to H^σ_0. For Q_R, this is proven in Lemma A.1.1, and the relative bounds for the operators involving the vector potential $\mathbf{A}$ were discussed in Section 2.1. Note that the relative bounds for operators involving $\mathbf{A}$ depend on the size of the ultraviolet-cutoff.
Therefore, $H_\sigma(e,\mathbf{R})$ is a quadratic operator family (in the parameter e) whose constant term is a self-adjoint operator and whose non-constant members are H^σ_0-bounded symmetric operators. By [Kat80], Ch.VII. §3, it follows that $H_\sigma(e,\mathbf{R})$ is a self-adjoint analytic

family of type (A) on a (complex) neighbourhood of 0, the size of which depends on both the infrared-cutoff σ and the ultraviolet-cutoff Λ. Since by the assumptions on H_A and H_B and its construction, $H_0^\sigma = H_\sigma(0, \mathbf{R})$ has a non-degenerate eigenvalue at the bottom of its spectrum, the remaining assertions follow from standard results of analytic perturbation theory (see also [Kat80]). □

Remark 2.7.2. As already mentioned in the introduction, the expansions (2.7.1) through (2.7.3) are partial with respect to the parameter e, in the sense that the coefficients $E_{i,A}^\sigma$, $E_{i,B}^\sigma$, $E_i^\sigma(R)$, $\psi_{i,A}^\sigma$, $\psi_{i,B}^\sigma$ and $\psi_i^\sigma(\mathbf{R})$ still depend on the physical value of e via the Coulomb potentials in the atomic Hamiltonians H_A and H_B. Furthermore, as discussed above, the spectral gaps of H_0^σ, $\overline{H_A + H_{f\geq\sigma}}$ and $\overline{H_A + H_{f\geq\sigma}}$ depend on σ and shrink to zero as $\sigma \to 0$. This implies that the radii of convergence of the expansions (2.7.1) through (2.7.3) shrink to zero as $\sigma \to 0$. As mentioned before, this issue does not pose a problem since our investigations concern the simplified model (1.2.1) for the interaction potential, and this quantity will turn out to have a well-defined ($\sigma \to 0$)-limit in the next section.

2.8 Regularized interaction potential and main results

As mentioned in the introduction, it is strongly conjectured (see e.g. [MS09]) that

$$\lim_{R\to\infty} E(\mathbf{R}) = \inf \operatorname{spec}(H^A) + \inf \operatorname{spec}(H^B)$$

and that

$$\lim_{R\to\infty} E^\sigma(e, \mathbf{R}) = E_A^\sigma(e) + E_B^\sigma(e),$$

for the corresponding infrared-regularized Hamiltonians, the corresponding result being a well-known fact in the case of molecular Schrödinger operators without coupling to the radiation field. The following definition of the regularized interaction potential is guided by this.

Definition 2.8.1. Assuming the hypotheses of Proposition 2.7.1, we set

$$E_\infty^\sigma(e) := E_A^\sigma(e) + E_B^\sigma(e)$$

and define the regularized interaction potential

$$V^\sigma(e, \Lambda, \mathbf{R}) := E^\sigma(e, \mathbf{R}) - E_\infty^\sigma(e).$$

For $i = 0, 1, 2 \ldots$, the coefficients $V_i^\sigma(\mathbf{R})$ are defined by

$$V_i^\sigma(\Lambda, \mathbf{R}) := E_i^\sigma(\mathbf{R}) - (E_{i,A}^\sigma + E_{i,B}^\sigma).$$

As already mentioned in the introduction, we stress the dependence on the ultraviolet-cutoff parameter Λ, which $V^\sigma(e, \mathbf{R})$ and $V_i^\sigma(\mathbf{R})$ inherit from the operators $H_\sigma^A(e)$, $H_\sigma^B(e)$ and $H_\sigma(e, \mathbf{R})$. Noting that $V_0^\sigma(\Lambda, \mathbf{R}) = 0$, these definitions and the series expansions from Proposition 2.7.1 immediately yield

Corollary 2.8.2. *Assume the hypotheses of Proposition 2.7.1. Then* $V^\sigma(e,\Lambda,\mathbf{R})$ *is analytic in* e *in a neighbourhood of* 0 *and has the series expansion*

$$V^\sigma(e,\Lambda,\mathbf{R}) = \sum_{i=1}^{\infty} e^i \; V_i^\sigma(\Lambda,\mathbf{R}). \tag{2.8.1}$$

In particular, all partial derivatives with respect to e *at* $(e=0)$ *exist, and*

$$\frac{\partial^i}{\partial e^i} V^\sigma(0,\Lambda,\mathbf{R}) = (i!) V_i^\sigma(\Lambda,\mathbf{R}).$$

In the remainder of this section we restate the three theorems containing the main results in order to streamline the presentation.

Theorem 2.8.3. *Assume (A1) and (A2) and let* $\Lambda \geq \Lambda_0$*, with* Λ_0 *as in Proposition 2.5.2. Then for* $i = 1,2,3,4$*,*

$$V_i(\Lambda,\mathbf{R}) := \lim_{\sigma\to 0}\left(\frac{1}{i!}\frac{\partial^i}{\partial e^i} V^\sigma(0,\Lambda,\mathbf{R})\right) = \lim_{\sigma\to 0}\left(V_i^\sigma(\Lambda,\mathbf{R})\right)$$

exists.

Recall the definition

$$V(\Lambda,\mathbf{R}) := \sum_{i=1}^{4} e^i V_i(\Lambda,\mathbf{R}) = \sum_{i=1}^{4} \frac{e^i}{i!}\left(\lim_{\sigma\to 0}\left(\frac{\partial^i}{\partial e^i} V^\sigma(0,\Lambda,\mathbf{R})\right)\right)$$

of the approximate model (1.2.1) for the full interaction potential $\tilde{V}(\mathbf{R})$ (2.3.1) from Section 1.2. For remarks on this simplification, see Section 1.2 of the introduction.

Theorem 2.8.4 ($1/R^7$-law for ultraviolet-cutoff system)**.** *Assume (A1) and (A2) and let* $\Lambda \geq \Lambda_0$*, with* Λ_0 *as in Proposition 2.5.2. Then*

$$\lim_{R\to\infty}\Big(R^k V(\Lambda,\mathbf{R})\Big) = 0$$

for any $0 \leq k < 7$*, and*

$$c_7(\Lambda) := \lim_{R\to\infty}\big(R^7 V(\Lambda,\mathbf{R})\big) = -e^4 \frac{23}{2}(2\pi)^{-3}\frac{\hbar c}{9}\alpha_E^A(0)\alpha_E^B(0),$$

where

$$\alpha_E^A(\mathbf{k}) = \Big\langle \sum_{i=1}^{Z_A} \mathbf{x}_i \Psi_A^0 \Big| \Big((H_A - E_A^0 + \hbar\omega(\mathbf{k}))_{\{\Psi_A^0\}^\perp}\Big)^{-1} \Big| \sum_{j=1}^{Z_A} \mathbf{x}_j \Psi_A^0 \Big\rangle,$$

$$\alpha_E^B(\mathbf{k}) = \Big\langle \sum_{i=1}^{Z_B} \mathbf{x}_i \Psi_B^0 \Big| \Big((H_B - E_B^0 + \hbar\omega(\mathbf{k}))_{\{\Psi_B^0\}^\perp}\Big)^{-1} \Big| \sum_{j=1}^{Z_B} \mathbf{x}_j \Psi_B^0 \Big\rangle$$

are the dynamic polarizabilities of the systems described by H_A *and* H_B*.*

Note that the interaction coefficient $c_7(\Lambda)$ still depends on the ultraviolet-cutoff via $\alpha_E^A(0)$ and $\alpha_E^B(0)$: their definition involves the operators H_A and H_B, their ground states and their lowest eigenvalues, all of which are Λ-dependent via the smeared Coulomb potential. The third main result states that $c_7(\Lambda)$ has a well-defined limit as the ultraviolet-cutoff is removed.

Theorem 2.8.5 (Ultraviolet-convergence and universality of $1/R^7$-law)**.** *Assume the hypotheses of Theorem 2.8.4. Then $c_7(\Lambda)$ has a well-defined limit as the ultraviolet-cutoff Λ is removed, which is given by*

$$\lim_{\Lambda\to\infty} c_7(\Lambda) = -e^4\frac{23}{2}(2\pi)^{-3}\frac{\hbar c}{9}\tilde{\alpha}_E^A(0)\tilde{\alpha}_E^B(0),$$

where $\tilde{\alpha}_E^A(\mathbf{k})$ and $\tilde{\alpha}_E^B(\mathbf{k})$ are the dynamic polarizabilities of the corresponding atomic Schrödinger operators $\tilde{H}_A$ and $\tilde{H}_B$ incorporating non-smeared Coulomb potentials, see (2.5.3).

Part II

Proof of the main results

Chapter 3

Simplification of terms in the interaction potential

The results of the following theorem constitute the first step in the proof of Theorems 1.2.1 through 1.2.3. It provides a decomposition of the terms $V_1^\sigma(\Lambda, \mathbf{R})$ through $V_4^\sigma(\Lambda, \mathbf{R})$ into terms which possess a structure that makes them more easily accessible to the analysis carried out in the later chapters. One important feature of some of the terms in this decomposition - as will become clear from their definitions below - is that they can be expressed in terms of integrals over the photon momenta, with integrands that involve only atomic quantities.
Before stating the theorem, let us introduce some notational conventions and definitions.

- Restrictions to the subspaces $\{\Psi_A^0\}^\perp$, $\{\Psi_B^0\}^\perp$, $\{\Psi_A^0\}^\perp \otimes \{\Psi_B^0\}^\perp$ etc., which are used in the construction of the reduced resolvents, are always understood.

- We will always write H_A, H_B and H_0^σ instead of $H_A - E_A^0$, $H_B - E_B^0$ and $H_0^\sigma - (E_A^0 + E_B^0)$.

- Whenever there is no risk of confusion, trivial tensor factors occurring in operators are left out, e.g. H_A instead of $H_A \otimes I_{\mathcal{H}_B}$.

- Statements, equations and identities that refer both to atom A and atom B will frequently be summarized using the notation $\Psi_{A,B}^0$, $H_{A,B}$, $\alpha_E^{A,B}$ et cetera.

Define

$$F_6(\mathbf{R}, \sigma) := -\frac{1}{9} L(\infty) \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}},$$

$$
\begin{aligned}
&F_7(\mathbf{R},\sigma)\\
&:=-\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Bigg[(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{-4\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2-6\hbar^3\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\qquad+\left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)+\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg],
\end{aligned}
\tag{3.0.1}
$$

$$
\begin{aligned}
&F_8(\mathbf{R},\sigma)\\
&:=-\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Bigg[2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)+\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2)\\
&\qquad-8\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_5(\mathbf{k}_1,\mathbf{k}_1)+\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_6(\mathbf{k}_1,\mathbf{k}_2)\Bigg].
\end{aligned}
\tag{3.0.2}
$$

Here $\Omega_\sigma := \{\omega(\mathbf{k}) \geq \sigma\} \subset \mathbb{R}^3$ is the restricted region of the one-photon momentum space which arises from the infrared regularization,

$$
C(\mathbf{k}) := \rho(\mathbf{k})\sqrt{\frac{\hbar}{2\omega(\mathbf{k})}} \tag{3.0.3}
$$

describes the coupling function of the electromagnetic vector potential $\mathbf{A}(\rho,\mathbf{x})$, the dynamic polarizabilities (see Theorem 2.8.4) are denoted by $\alpha_E^{A,B}(\mathbf{k})$, and

$$
\mathbf{v}_A := \sum_{i_A=1}^{Z_A}\mathbf{x}_{i_A}\Psi_A^0, \quad \mathbf{v}_B := \sum_{j_B=1}^{Z_B}\mathbf{x}_{j_B}\Psi_B^0 \tag{3.0.4}
$$

are the dipole moments of the ground states of atom A and B, respectively. Furthermore, we have defined

$$
L(\infty) := \sum_{\alpha,\beta=1}^{3}\left\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\right|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}\left|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\right\rangle_{L^2(\mathbb{R}^{3N})}, \tag{3.0.5}
$$

$$
\begin{aligned}
T_4(\mathbf{k}_1,\mathbf{k}_2) :=& \sum_{\alpha=1}^{3}\Big[\Big\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha}\alpha_E^B(\mathbf{k}_1)\\
&+\alpha_E^A(\mathbf{k}_1)\Big\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_B^\alpha}\Big],
\end{aligned}
\tag{3.0.6}
$$

$$T_5(\mathbf{k}_1,\mathbf{k}_2) := \sum_{\alpha,\beta=1}^{3} \Big\langle (\overline{H_A+H_B})^{-1}(H_A+\hbar(\omega(\mathbf{k}_1))^{-1}(H_B+\hbar(\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}, \quad (3.0.7)$$

$$T_6(\mathbf{k}_1,\mathbf{k}_2) := \sum_{\alpha,\beta=1}^{3} \Big[\Big\langle (\overline{H_A+H_B})^{-1}\Big[(H_A+\hbar(\omega(\mathbf{k}_1))^{-1}(H_A+\hbar(\omega(\mathbf{k}_2))^{-1} + (H_B+\hbar(\omega(\mathbf{k}_1))^{-1}(H_B+\hbar(\omega(\mathbf{k}_2))^{-1}\Big]\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\Big]. \quad (3.0.8)$$

Recall that $\mathbf{x}_i\psi_{A,B}^0 \in H^2(\mathbb{R}^{3Z_{A,B}})$ by the remarks in Section 2.5. Furthermore, $\mathbf{x}_i\psi_{A,B} \in \{\psi_{A,B}\}^\perp$ by the assumed non-degeneracy of the atomic ground states, and

$$\sigma(H_{A|\{\Psi_A^0\}^\perp}) \subset [\Delta_A,\infty), \quad \sigma(H_{B|\{\Psi_B^0\}^\perp}) \subset [\Delta_B,\infty),$$

so $\alpha_E^{A,B}(\mathbf{k})$ and the T_i are well-defined. Finally, define

$$\begin{aligned}
&M_B(\mathbf{R},\sigma)\\
&:= \frac{2}{\hbar^2}\mathrm{Re}\Bigg[\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\quad\times\Bigg[-2\hbar\omega(\mathbf{k})\Big\langle \mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B|(\overline{H_A+H_B})^{-1}|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0|Q_R\Big|\\
&\qquad\quad\Big|\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\Big\rangle\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\right)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)\,|(\overline{H_A+H_B})^{-1}|\\
&\qquad\quad\Big|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle\left(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)\,|(\overline{H_A+H_B})^{-1}|\\
&\qquad\quad\Big|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Bigg]\Bigg]
\end{aligned}$$

and

$$
\begin{aligned}
&M_A(\mathbf{R},\sigma)\\
&:=\frac{1}{\hbar^2}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\\
&\quad\times\Bigg[\Bigg\{\Big\langle \mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\qquad-\hbar\omega(\mathbf{k})\Big\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\qquad-\hbar\omega(\mathbf{k})\Big\langle\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\Big|\\
&\qquad\quad\Big|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\Bigg\} \qquad (3.0.9)\\
&\qquad+\Bigg\{\Big\langle\Psi_A^0\otimes\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle\\
&\qquad-\hbar\omega(\mathbf{k})\Big\langle\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle\\
&\qquad-\hbar\omega(\mathbf{k})\Big\langle\Psi_A^0\otimes\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\Big\rangle\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\Big|\\
&\qquad\quad\Big|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\Big\rangle\Bigg\}\Bigg] \qquad (3.0.10)\\
&+2\mathrm{Re}\Bigg[\int_{\Omega_\sigma} d\mathbf{k}\left(\frac{-\mathrm{i}}{m_e\hbar}\right)|C(\mathbf{k})|^2\\
&\quad\times\Bigg[\Bigg\{\Big\langle(H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big((\sum_{j_A}\mathbf{p}_{j_A})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_A\Big)\otimes\Psi_B^0|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\qquad-\hbar\omega(\mathbf{k})\Big\langle(H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big((\sum_{j_A}\mathbf{p}_{j_A})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\Big)\otimes\Psi_B^0|\\
&\qquad\quad|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Bigg\} \qquad (3.0.11)\\
&\qquad+\Bigg\{\Big\langle\Psi_A^0\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big((\sum_{j_B}\mathbf{p}_{j_B})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\qquad-\hbar\omega(\mathbf{k})\Big\langle\Psi_A^0\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big((\sum_{j_B}\mathbf{p}_{j_B})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\Big)|\\
&\qquad\quad|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Bigg\}\Bigg]\Bigg]. \qquad (3.0.12)
\end{aligned}
$$

Theorem 3.0.6. *Assume (A1) and (A2) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Then*

$$
\begin{aligned}
V_1^\sigma(\Lambda, \mathbf{R}) =& V_3^\sigma(\Lambda, \mathbf{R}) = 0, \\
V_2^\sigma(\Lambda, \mathbf{R}) =& \langle \Psi_0 | Q_R | \Psi_0 \rangle, && (3.0.13) \\
V_4^\sigma(\Lambda, \mathbf{R}) =& - \langle Q_R \Psi_0 | T^\sigma | Q_R \Psi_0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}} && (3.0.14) \\
& + F_6(\mathbf{R}, \sigma) && (3.0.15) \\
& + F_7(\mathbf{R}, \sigma) + F_8(\mathbf{R}, \sigma) && (3.0.16) \\
& + M_A(\mathbf{R}, \sigma) + M_B(\mathbf{R}, \sigma) && (3.0.17) \\
& - \langle \Psi_0 | Q_R | \Psi_0 \rangle \Big(\| T_A^\sigma H'_{\sigma,A} (\Psi_A^0 \otimes \Omega) \|^2 + \| T_B^\sigma H'_{\sigma,B} (\Psi_B^0 \otimes \Omega) \|^2 \Big). && (3.0.18)
\end{aligned}
$$

Remarks:
The terms (3.0.13) and (3.0.14) only contain the interatomic Coulomb potential Q_R and correspond exactly to the first- and second-order energy corrections that arise in the context of the perturbative analysis of the Born-Oppenheimer potential energy surface in the case without radiation field, see [Gar07]. The reason they appear as a second- and fourth-order correction (with respect to the perturbation parameter e) in our situation is that if the radiation field is taken into account, the perturbation is a sum of both linear and quadratic terms e.
To further analyze (3.0.13), (3.0.14) and (3.0.18) (the latter owing its $\mathbf{R}$-dependence only to the prefactor $\langle \Psi_0 | Q_R | \Psi_0 \rangle$, which is identical to (3.0.13)), we will employ a multipole expansion of Q_R. This technique, which exploits the exponential decay of the atomic ground states Ψ_A^0 and Ψ_B^0 and involves a spatial cutoff, will be developed in Section 5.1 and applied to (3.0.13), (3.0.14) and (3.0.18) in Sections 5.2 and 5.3. There it will turn out that (3.0.13) and (3.0.18) decay faster than any inverse power of R as $R \to \infty$, while the lowest-order contribution (in $1/R$) to $-\langle Q_R \Psi_0 | T^\sigma | Q_R \Psi_0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}}$ is proportional to $1/R^6$, and is given a by version of the well-known so-called London term (1.0.3) involving the smeared Coulomb potential.
The terms (3.0.15) and (3.0.16) which originate from (4.3.3) below are generated solely by the interaction operators $H'_{\sigma,A}$ and $H'_{\sigma,B}$, while (3.0.17) (which will be further analyzed in Section 5.4) contains both the field interaction and the interatomic Coulomb interaction. As it will turn out, after the removal of the infrared-cutoff σ,

- The term (3.0.15) and parts of (3.0.17) are responsible for the (asymptotic) cancellation of the $1/R^6$-contribution from $-\langle Q_R \Psi_0 | T^\sigma | Q_R \Psi_0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}}$, see Section 6.6.

- The term $F_7(\mathbf{R}, \sigma)$ and parts of (3.0.17) combine to give the asymptotic $1/R^7$-decay of $V_4^\sigma(\Lambda, \mathbf{R})$, the coefficient agreeing with the one predicted by Casimir and Polder, see Section 6.

- The term $F_8(\mathbf{R}, \sigma)$ and the remaining parts of (3.0.17) are of higher order than $1/R^7$, see also Section 6.

Chapter 4

Proof of Theorem 3.0.6

4.1 Derivation of a formula for the fourth-order energy correction

The first step in the proof of Theorem 3.0.6 consists of deriving formulas for the energy corrections up to fourth order (in e), using general features and symmetries of the perturbation problem at hand. In particular, we conclude that the first- and third-order corrections vanish altogether.
The perturbation problems (2.4.1) for the compound system and (2.4.4), (2.4.5) for the separated atoms have the following features in common:

i. They are of the form $H(e) = H_0 + eH' + e^2H''$, with $H(e)$ acting on a Hilbert space of the form $\mathcal{H} \otimes \mathcal{F}$, where $\mathcal{H}$ is a Hilbert space and and $\mathcal{F}$ is a bosonic Fock space.

ii. H_0 is a self-adjoint operator which has the non-interacting structure

$$H_0 = \overline{\tilde{H} \otimes I_{\mathcal{F}} + I_{\mathcal{H}} \otimes H_{f \geq \sigma}},$$

where $\tilde{H}$ is a self-adjoint operator on $\mathcal{H}$ and $H_{f\geq\sigma}$ is the infrared-regularized free field Hamiltonian. H_0 has a simple eigenvalue E_0 at the bottom of its spectrum, and the corresponding eigenvector (ground state) is of the form $\Psi_0 = \psi \otimes \Omega$, where $\psi \in \mathcal{H}$ and $\Omega \in \mathcal{F}$ is the vacuum vector. Furthermore, E_0 is separated from the rest of the spectrum by a spectral gap $\Delta > 0$.

iii. The part of H' which acts on $\mathcal{F}$ maps the n-th sector to the $(n+1)$-th and to the $(n-1)$-th sector.

iv. The part of H'' which acts on $\mathcal{F}$ maps the n-th sector to itself, the $(n-2)$-th and to the $(n+2)$-th sector.

By the assumptions on H_0 we have $\text{spec}\,(H_0 - E_0) \subset \{0\} \dot{\cup} [\Delta, \infty)$, with 0 being an isolated eigenvalue. Restricting this operator to the orthogonal complement of the ground state Ψ_0 yields

$$\text{spec}\left((H_0 - E_0)_{|\{\Psi_0\}^\perp}\right) \subset [\Delta, \infty),$$

which shows that $0 \in \rho\left((H_0 - E_0)_{|\{\Psi_0\}^\perp}\right)$, and thus the reduced resolvent

$$T := ((H_0 - E_0)_{|\{\Psi_0\}^\perp})^{-1}$$

exists and is bounded, see also Proposition 2.6.4 above. The structure of H_0 implies that T leaves individual Fock space levels invariant (see also Lemma 4.2.5). Let P and $P^\perp$ denote the orthogonal projections onto $\{\Psi_0\}$ and $\{\Psi_0\}^\perp$, respectively. For a perturbation problem of this form, assuming that for e in a neighbourhood of 0 we have an analytic representation of the ground state eigenvalue $E(e)$ and the ground state $\psi(e)$ of $H(e)$, we can solve the eigenvalue equation

$$H(e)\psi(e) = \left(H_0 + eH' + e^2H''\right)\psi(e) = E(e)\psi(e) = \Big(\sum_{i=0}^{\infty} e^i\, E_i\Big)\Big(\sum_{j=0}^{\infty} e^j \psi_j\Big)$$

order by order and find

$$\begin{aligned}
E_1 =& \langle\Psi_0|H'|\Psi_0\rangle = 0 \text{ (by ii and iii)} && (4.1.1)\\
E_2 =& -\langle H'\Psi_0|T|H'\Psi_0\rangle + \langle\Psi_0|H''|\Psi_0\rangle && (4.1.2)\\
E_3 =& -\langle H'\Psi_0|T(E_1 - P^\perp H')TP^\perp|H'\Psi_0\rangle \\
&-\langle H'\Psi_0|TP^\perp|H'P\psi_1\rangle \\
&-\langle H'\Psi_0|TP^\perp|H''\Psi_0\rangle \\
&-\langle H''\Psi_0|TP^\perp|H'\Psi_0\rangle \\
&-\langle\Psi_0|\psi_1\rangle\,\left(E_2 - \langle\Psi_0|H''\Psi_0\rangle\right).
\end{aligned}$$

Using that $\{\Psi_0\}$ is one-dimensional (property ii) above), one concludes $P\psi' = \mu\Psi_0$ for a complex number $\mu = \langle\Psi_0|\psi'\rangle$, which, together with the formula for E_2 already established, leads to the simplification

$$\begin{aligned}
E_3 =& -\langle H'\Psi_0|T(E_1 - P^\perp H')TP^\perp|H'\Psi_0\rangle \\
&-\langle H'\Psi_0|TP^\perp|H''\Psi_0\rangle - \langle H''\Psi_0|TP^\perp|H'\Psi_0\rangle \\
=& -\langle H'\Psi_0|T(E_1 - P^\perp H')TP^\perp|H'\Psi_0\rangle \\
&-2\mathrm{Re}\left[\langle H'\Psi_0|T|H''\Psi_0\rangle\right].
\end{aligned}$$

Now using $E_1 = 0$ and properties iii) and iv) above, as well as the Fock level invariance of the reduced resolvent and the mutual orthogonality of different Fock space sectors, we conclude

$$E_3 = 0. \tag{4.1.3}$$

Using this and the same arguments again, one arrives at the simplified expression

$$\begin{aligned}
E_4 =& -\langle H'\Psi_0|TH'TH'T|H'\Psi_0\rangle - E_2\|TH'\Psi_0\|^2 \\
&-\langle H''\Psi_0|T|H''\Psi_0\rangle + 2\mathrm{Re}\left[\langle H'\Psi_0|TH'T|H''\Psi_0\rangle\right] \\
&+\langle H'\Psi_0|TH''T|H'\Psi_0\rangle
\end{aligned} \tag{4.1.4}$$

for the fourth-order energy correction.

4.2 Analysis of the reduced resolvent

Having established explicit formulas for the energy corrections up to fourth order in the preceding section, we will now apply these to the specific perturbation problems at hand to obtain a simplified expression for the interaction potential

$$V(\Lambda, \mathbf{R}) = \sum_{i=0}^{4} e^i \, V_i^\sigma(\Lambda, \mathbf{R}).$$

To this end, we will first set out to conduct a detailed analysis of the reduced resolvent

$$T^\sigma = ((H_0^\sigma - E_0)_{|\{\Psi_0\}^\perp})^{-1}.$$

More precisely, after proving two auxiliary technical results, we first investigate the action of T^σ on a number of invariant subspaces (Lemma 4.2.5 below). Following this, we show in Section 4.2.3 how the action of T^σ on several subspaces of $\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}_\sigma^{(1)}$ and $\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}_\sigma^{(2)}$ can be understood pointwise in the photon coordinates $(\mathbf{k}, \lambda)$ and $(\mathbf{k}_1, \lambda, \mathbf{k}_2, \mu)$, respectively. The corresponding results will be used heavily in the calculations which convert the contributions to $V(\Lambda, \mathbf{R})$ into integrals over photon momenta. Sections 4.2.4 and 4.2.5 are concerned with properties of T^σ inherited from the atomic Hamiltonians H_A and H_B, in particular commutativity with the atomic ground state projections and parity operators, as well as rotation invariance.
In Section 4.2.6 we establish and collect some important operator identities that will be used in the calculations later on.

4.2.1 Some results on tensor products of operators

Lemma 4.2.1. *Let $A : D(A) \to H$ be a densely defined operator which is boundedly invertible, and let $M \subset H$ be a reducing subspace of A. Then M is a reducing subspace of A^{-1}, and the restrictions $A_{|M}$ and $A_{|M^\perp}$ are invertible, with inverses given by*

$$(A_{|M})^{-1} = (A^{-1})_{|M}, \quad (A_{|M^\perp})^{-1} = (A^{-1})_{|M^\perp}.$$

Proof. Since A is boundedly invertible, we have $D(A^{-1}) = H$. For any $u \in H$ we can find an element $v \in D(A)$ such that $u = Av$. Let P_M be the orthogonal projection onto M. Then

$$A^{-1}P_M u = A^{-1}P_M Av = A^{-1}AP_M v = P_M v = P_M A^{-1}u,$$

where we have used the assumption $P_M A \subset AP_M$ in the second identity. This shows that $A^{-1}P_M = P_M A^{-1}$, and thus that M is a reducing subspace for A^{-1}. In particular, the restrictions $(A^{-1})_{|M}$ and $(A^{-1})_{|M^\perp}$ are well-defined, and for any $u \in M$ we have

$$(A^{-1})_{|M}u = A^{-1}u \in M \cap D(A) = D(A_{|M})$$

and thus

$$A_{|M}(A^{-1})_{|M}u = AA^{-1}u = u.$$

On the other hand,

$$(A^{-1})_{|M} A_{|M} u = (A^{-1})_{|M} Au = A^{-1} Au = u$$

for any $u \in D(A) \cap M$, since then $Au \in M$ by assumption. This shows the assertion for the operators involving the subspace M. The corresponding result for those involving $M^\perp$ is shown completely analogously. □

Lemma 4.2.2. *Let H be a Hilbert space and let $A : D(A) \to H$ and $B : D(B) \to H$ be bounded closable operators with a common dense domain D which is left invariant by both of them, i.e. $A\mathcal{D} \subset \mathcal{D}$, $B\mathcal{D} \subset \mathcal{D}$. Then if A and B commute on $\mathcal{D}$, i.e. $ABu = BAu$ for all $u \in \mathcal{D}$, so do their closures, i.e.*

$$(\overline{A})(\overline{B})u = (\overline{B})(\overline{A})u$$

for all $u \in H$.

Proof. Let $u \in H$. Since $\overline{A}$ and $\overline{B}$ are closed by construction, we can choose $\{u_n\} \subset \mathcal{D}$ with $u_n \to u$ and $Bu_n \to \overline{B}u$. By the boundedness of A, the sequence Au_n also converges (it is a Cauchy sequence), and by the closedness of $\overline{A}$ this limit has to equal $\overline{A}u$. Thus we obtain

$$(\overline{A})(\overline{B})u = (\overline{A})(\lim_{n\to\infty} Bu_n) \underbrace{=}_{\overline{A} \text{ cont.}} \lim_{n\to\infty} (\overline{A}) \underbrace{Bu_n}_{\in\mathcal{D}} = \lim_{n\to\infty} ABu_n = \lim_{n\to\infty} B(Au_n),$$

which shows that $\lim_{n\to\infty} B(Au_n)$ exists. Now the closedness of $\overline{B}$ implies that

$$\lim_{n\to\infty} B(Au_n) = \overline{B}(\lim_{n\to\infty} Au_n) = (\overline{B})(\overline{A})u,$$

which proves the assertion. □

Definition 4.2.3. A symmetric operator $A : D(A) \to H$ is called positive if

$$\langle u|Au\rangle \geq 0$$

for all $u \in D(A)$.

Lemma 4.2.4. *Let A and B be positive self-adjoint operators on the Hilbert spaces H_1 and H_2, respectively. Then $A \otimes B$, $I \otimes B$, $A \otimes I$ and $A \otimes I + I \otimes B$ are positive operators on $H_1 \otimes H_2$, and $D(\overline{A+B}) \subset D(\overline{A \otimes I})$, $D(\overline{A+B}) \subset D(\overline{I \otimes B})$.*

Proof. By the positivity of A and B, we have $\mathrm{spec}(A) \subset [0, \infty)$ and $\mathrm{spec}(B) \subset [0, \infty)$, and thus

$$\mathrm{spec}(\overline{A \otimes B}) = \overline{(\mathrm{spec}(A))(\mathrm{spec}(B))} \subset [0, \infty),$$
$$\mathrm{spec}(\overline{A \otimes I + I \otimes B}) = \overline{(\mathrm{spec}(A)) + (\mathrm{spec}(B))} \subset [0, \infty),$$

and $\mathrm{spec}(\overline{A \otimes I}) = \mathrm{spec}(A)$, $\mathrm{spec}(\overline{I \otimes B}) = \mathrm{spec}(B)$ (see [RS80], Theorem VIII.33). This in turn implies that $\overline{A \otimes B}$, $\overline{A \otimes I + I \otimes B}$, $\overline{A \otimes I}$ and $\overline{I \otimes B}$ are positive. Thus the assertion on the positivity of the operators follows by restriction.

Next let $u \in D(\overline{A+B})$ and choose $\{u_n\} \subset D(A)\widehat{\otimes}D(B)$ with $u_n \to u$ and $(A+B)u_n \to (\overline{A+B})u$, which is possible by construction of the closure. Since $\{u_n\} \subset D(A)\widehat{\otimes}D(B) \subset D(A)\widehat{\otimes}H_2$, it suffices to show that $(A\otimes I)u_n \to \overline{A\otimes I}u$ to prove the assertion on the domains. To this end, consider

$$\begin{aligned}\|(A+B)(u_n-u_m)\|^2 =& \|A(u_n-u_m)\|^2 + \|B(u_n-u_m)\|^2 \\ &+ 2\mathrm{Re}\left[\langle (A\otimes I)(u_n-u_m), (I\otimes B)(u_n-u_m)\rangle\right].\end{aligned}$$

Since A and B are self-adjoint and $A\otimes B$ is positive, we have

$$\langle (A\otimes I)(u_n-u_m), (I\otimes B)(u_n-u_m)\rangle = \langle u_n-u_m, (A\otimes B)(u_n-u_m)\rangle \geq 0,$$

which implies

$$\|(A+B)(u_n-u_m)\|^2 \geq \|A(u_n-u_m)\|^2 + \|B(u_n-u_m)\|^2.$$

Now as a convergent sequence $(A+B)u_n$ is a Cauchy sequence, and the inequality shows that Au_n and Bu_n are also Cauchy sequences, which converge since $\mathcal{H}_A\otimes\mathcal{H}_B$ is complete. But now the closability of $A\otimes I$ and $I\otimes B$ implies that $u \in D(\overline{A\otimes I}) \cap D(\overline{I\otimes B})$ and that $(A\otimes I)u_n \to (\overline{A\otimes I})u$ and $(I\otimes B)u_n \to (\overline{I\otimes B})u$.

□

4.2.2 Action of T^σ on reducing subspaces

Lemma 4.2.5 (Properties of the reduced resolvent)**.** *Let the assumptions (A1) and (A2) be satisfied and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Let $H_0^\sigma = \overline{H_A + H_B + H_{f\geq\sigma}}$ and assume without loss of generality that $\inf\mathrm{spec}(H_A) = \mathrm{spec}(H_B) = 0$, which implies $\inf\mathrm{spec}(H_0^\sigma) = 0$. Recall that H_0^σ has a spectral gap $\min\{\Delta_A, \Delta_B, \sigma\}$. Let Ψ_A^0 and Ψ_B^0 denote the non-degenerate ground states of H_A and H_B, respectively. Set $\Psi_0 := \Psi_A^0 \otimes \Psi_B^0 \otimes \Omega$ and let $n \in \mathbb{N}$, $n \geq 1$. Then the reduced resolvent*

$$T^\sigma = \left(H_0^\sigma|_{\{\Psi_0\}^\perp}\right)^{-1}$$

has the following invariant (closed) subspaces, on which it acts as indicated:

i. 'Particle excitations':

$$\{\Psi_A^0\otimes\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}, \quad \left(\overline{H_A+H_B}|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}\otimes I_{\{\Omega\}}, \tag{4.2.1}$$

$$\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}, \quad I_{\{\Psi_A^0\}}\otimes\left(H_B|_{\{\Psi_B^0\}^\perp}\right)^{-1}\otimes I_{\{\Omega\}}, \tag{4.2.2}$$

$$\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}\widehat{\otimes}\{\Omega\}, \quad \left(H_A|_{\{\Psi_A^0\}^\perp}\right)^{-1}\otimes I_{\{\Psi_B^0\}}\otimes I_{\{\Omega\}}, \tag{4.2.3}$$

$$\overline{(\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp)}\widehat{\otimes}\{\Omega\}, \quad \left(\overline{H_A+H_B}|_{\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}}\right)^{-1}\otimes I_{\{\Omega\}}. \tag{4.2.4}$$

ii. 'Field excitations':

$$\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}\widehat{\otimes}\{\Omega\}^\perp, \quad I_{\{\Psi_A^0\}}\otimes I_{\{\Psi_B^0\}}\otimes\left(H_{f\geq\sigma}|_{\{\Omega\}^\perp}\right)^{-1}, \tag{4.2.5}$$

$$\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}\widehat{\otimes}\mathcal{F}_\sigma^{(n)}, \quad I_{\{\Psi_A^0\}}\otimes I_{\{\Psi_B^0\}}\otimes\left(\oplus_{\lambda=1,2}\frac{1}{\hbar(\omega(\mathbf{k}_1)+\cdots+\omega(\mathbf{k}_n))}\right). \tag{4.2.6}$$

iii. 'Mixed excitations':

$$\{\Psi_A^0\}\widehat{\otimes}(\overline{\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}), \quad I_{\{\Psi_A^0\}}\otimes\underbrace{\left(\overline{H_B+H_{f\geq\sigma}}|_{\overline{\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}}\right)^{-1}}_{=T_B^\sigma}, \tag{4.2.7}$$

$$\{\Psi_A^0\}\widehat{\otimes}(\overline{\{\Psi_B^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(n)}}), \quad I_{\{\Psi_A^0\}}\otimes\left(\overline{H_B+\hbar(\omega(\mathbf{k}_1)+\cdots+\omega(\mathbf{k}_n))}|_{\overline{\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}}\right)^{-1}, \tag{4.2.8}$$

$$\{\Psi_B^0\}\widehat{\otimes}(\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}), \quad I_{\{\Psi_B^0\}}\otimes\underbrace{\left(\overline{H_A+H_{f\geq\sigma}}|_{\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}}\right)^{-1}}_{=T_A^\sigma}, \tag{4.2.9}$$

$$\{\Psi_B^0\}\widehat{\otimes}(\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(n)}}), \quad I_{\{\Psi_B^0\}}\otimes\left(\overline{H_A+\hbar(\omega(\mathbf{k}_1)+\cdots+\omega(\mathbf{k}_n))}|_{\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}}\right)^{-1}, \tag{4.2.10}$$

$$\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}, \quad \left(\overline{H_A+H_B+H_{f\geq\sigma}}|_{\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}}\right)^{-1}, \tag{4.2.11}$$

$$\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(n)}}, \quad \left(\overline{H_A+H_B+\hbar(\omega(\mathbf{k}_1)+\cdots+\omega(\mathbf{k}_n))}|_{\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}^\perp}}\right)^{-1}. \tag{4.2.12}$$

Proof. By Proposition 2.6.4, $\{\Psi_0\}$, $\{\Psi_0\}^\perp$ and all the subspaces of $\{\Psi_0\}^\perp$ mentioned in the assertion are reducing subspaces for $\overline{H_A+H_B+H_f^\sigma}$,

$$\mathrm{spec}((\overline{H_A+H_B+H_f^\sigma})_{|\{\Psi_0\}^\perp})=[\min\{\Delta_A,\Delta_B,\hbar\sigma\},\infty),$$

and in particular

$$T^\sigma=((\overline{H_A+H_B+H_f^\sigma})_{|\{\Psi_0\}^\perp})^{-1}$$

exists and is bounded. By Lemma 4.2.1, all the subspaces comprising $\{\Psi_0\}^\perp$ are reducing subspaces of T^σ, and restriction commutes with taking the inverse.
Next we will analyze the action of T^σ on the individual subspaces. To this end, denote $S:=H_A+H_B+H_{f\geq\sigma}$, with domain $D(H_A)\widehat{\otimes}D(H_B)\widehat{\otimes}D(H_{f\geq\sigma})$.
i) First of all consider $S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}$ (no closure of the subspace needs to be taken, since $\{\Psi_A^0\}\widehat{\otimes}\{\Omega\}$ is finite-dimensional, see Lemma 2.6.1). As H_A and $H_{f\geq\sigma}$ act trivially on $\{\Psi_A^0\}$ and $\{\Omega\}$, respectively, we conclude

$$\overline{S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}}=\overline{I_{\{\Psi_A^0\}}\otimes H_{B|\{\Psi_B^0\}^\perp}\otimes I_{\{\Psi_B^0\}}},$$

from which we deduce

$$\mathrm{spec}(\overline{S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}})=\mathrm{spec}(H_{B|\{\Psi_B^0\}^\perp})$$

as operators on the Hilbert space $\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}$, see e.g. [RS80]). In particular, $\overline{S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}}\geq\Delta_B>0$. We claim that

$$\begin{aligned}\left(\overline{S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}}\right)^{-1}&=\overline{I_{\{\Psi_A^0\}}\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\otimes I_{\{\Omega\}}}\\&=I_{\{\Psi_A^0\}}\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\otimes I_{\{\Omega\}},\end{aligned} \tag{4.2.13}$$

which is (4.2.2). Note that $S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}$ is essentially self-adjoint by Corollary 2.6.3, so it is in particular densely defined, symmetric and closable. Furthermore, its closure is positive ($\geq \Delta_B > 0$, see above) and thus in particular one-to-one. So in order to prove the first equality in (4.2.13), by ([Wei76], Satz 5.2) it suffices to verify that $S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}$ is one-to-one and has the (set-theoretic) inverse $I_{\{\Psi_A^0\}}\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\otimes I_{\{\Omega\}}$ on its range. The latter follows from $\mathrm{Ran}(S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}) = \{\Psi_A^0\}\widehat{\otimes}(\mathrm{Ran}(H_{B|\{\Psi_B^0\}^\perp}))\widehat{\otimes}\{\Omega\}$, while the former follows from the relation

$$\mathrm{Ker}(S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}) \subset \mathrm{Ker}(\overline{(S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}})}) = \{0\},$$

where the last equality holds since $\overline{(S_{|\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}})}$ is boundedly invertible. The second equality in (4.2.13) holds since $I_{\{\Psi_A^0\}}\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\otimes I_{\{\Omega\}}$ is bounded and defined on all of $\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}$, which is a closed subspace by Lemma 2.6.1 (note that $\{\Psi_A^0\}$ and $\{\Omega\}$ are one-dimensional) and thus equal to the Hilbert space $\{\Psi_A^0\}\otimes\{\Psi_B^0\}^\perp\otimes\{\Omega\}$.
The identities (4.2.3) and (4.2.5) are proven completely analogous. To show (4.2.6) one uses in addition the decomposition $\{\Omega\}^\perp = \oplus_{n=1}^\infty \mathcal{F}_\sigma^{(n)}$, which consists of reducing subspaces for the operator $H_{f|\{\Omega\}^\perp}$ by construction of the second quantization. Note that the bounded invertibility of $H^\sigma_{f|\{\Omega\}\perp}$ and $H^\sigma_{f|\mathcal{F}_\sigma^{(n)}} = \oplus_{\lambda=1,2}\hbar(\omega(\mathbf{k}_1)+\cdots+\omega(\mathbf{k}_n))$ is due to the infrared regularization.
ii) To see (4.2.4), first set $M := \overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}$ and note that

$$\overline{S_{M\widehat{\otimes}\{\Omega\}}} = \overline{(H_A+H_B)_{|M}\otimes I_{\{\Omega\}}},$$

since the action of $H_{f\geq\sigma}$ on $\{\Omega\}$ is trivial. Furthermore,

$$(H_A+H_B)_{|M}\otimes I_{\{\Omega\}} \subset \overline{((H_A+H_B)_{|M})}\otimes I_{\{\Omega\}},$$

and both operators are essentially self-adjoint (the former by Corollary 2.6.3, the latter by the construction of tensor product operators and the fact that $\overline{((H_A+H_B)_{|M})} = (\overline{H_A+H_B})_{|M}$ is self-adjoint by arguments analogous to those in i)). The uniqueness of the closure now implies

$$\overline{(H_A+H_B)_{|M}\otimes I_{\{\Omega\}}} = \overline{\overline{((H_A+H_B)_{|M})}\otimes I_{\{\Omega\}}},$$

which allows us to conclude $\mathrm{spec}(\overline{S_{M\widehat{\otimes}\{\Omega\}}}) = \mathrm{spec}(\overline{((H_A+H_B)_{|M})})$ To calculate the latter set, we observe that

$$H_{A|\{\Psi_A^0\}^\perp} + H_{B|\{\Psi_B^0\}^\perp} \subset (H_A+H_B)_{|M} \subset \overline{((H_A+H_B)_{|M})},$$

which shows that $\overline{((H_A+H_B)_{|M})}$ is a self-adjoint extension of $H_{A|\{\Psi_A^0\}^\perp}+H_{B|\{\Psi_B^0\}^\perp}$, the latter being essentially self-adjoint by construction of the tensor product of operators. Again by the uniqueness of the self-adjoint closure, we deduce

$$\overline{((H_A+H_B)_{|M})} = \overline{H_{A|\{\Psi_A^0\}^\perp}+H_{B|\{\Psi_B^0\}^\perp}},$$

which implies

$$\mathrm{spec}(\overline{((H_A+H_B)_{|M})}) = \overline{\mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp})+\mathrm{spec}(H_{B|\{\Psi_B^0\}^\perp})} \subset [\Delta_A+\Delta_B,\infty).$$

In particular,

$$(\overline{((H_A + H_B)_{|M})})^{-1} = (\overline{(H_A + H_B)}_{|M})^{-1}$$

exists and is bounded. As in i), we first check that $(H_A+H_B)_{|M} \otimes I_{\{\Omega\}}$ is one-to-one, which holds since its closure $\overline{S_{M\widehat{\otimes}\{\Omega\}}}$ is boundedly invertible. Furthermore, $(H_A + H_B)_{|M} \otimes I_{\{\Omega\}}$ has the set-theoretic inverse $(\overline{((H_A + H_B)_{|M})})^{-1} \otimes I_{\{\Omega\}}$ on its range, which allows us to conclude that

$$\left(\overline{S_{M\widehat{\otimes}\{\Omega\}}}\right)^{-1} = \overline{(\overline{(H_A + H_B)}_{|M})^{-1} \otimes I_{\{\Omega\}}} = (\overline{(H_A + H_B)}_{|M})^{-1} \otimes I_{\{\Omega\}},$$

where the last identity follows since M is closed and $\{\Omega\}$ is finite-dimensional. Thus we have proved (4.2.4). The proof of the remaining asserted identities is completely analogous. □

4.2.3 Fiber decomposition of T^σ with respect to photon momenta

Lemma 4.2.6 (Fiber decomposition)**.** *Let A be a densely defined symmetric positive operator on a Hilbert space H, $A \geq a > 0$, which is essentially self-adjoint on a core $\mathcal{D}$. Let $\Omega \subset \mathbb{R}^n$ and $\omega : \Omega \to \mathbb{R}_{\geq 0}$ be measurable, and let T_ω be the self-adjoint realization of multiplication by ω on $L^2(\Omega)$. Then $A + T_\omega$ is essentially self-adjoint on $\mathcal{D}\widehat{\otimes}D(T_\omega)$,* $\operatorname{spec}(\overline{A + T_\omega}) \subset [a, \infty)$,

$$\inf \operatorname{spec}(\overline{A + T_\omega}) = \inf \operatorname{spec}(\overline{A}) + \inf \operatorname{spec}(T_\omega) \geq a > 0,$$

and under the isomorphism $H \otimes L^2(\Omega) \cong L^2(\Omega; H)$ we have

$$\left(\overline{A + T_\omega}\right)^{-1} = \int_\Omega^\oplus (\overline{A} + \omega(\mathbf{k}))^{-1} d\mathbf{k},$$

i.e.

$$\left(\left(\overline{A + T_\omega}\right)^{-1} \varphi\right)(\mathbf{k}) = (\overline{A} + \omega(\mathbf{k}))^{-1} \varphi(\mathbf{k})$$

for any $\varphi \in H \otimes L^2(\Omega)$ and $\mathbf{k} \in \Omega$. In particular,

$$\langle \varphi | (\overline{A + T_\omega})^{-1} \psi \rangle_{H \otimes L^2(\Omega)} = \int_\Omega \langle \varphi(\mathbf{k}) | (\overline{A} + \omega(\mathbf{k}))^{-1} | \psi(\mathbf{k}) \rangle_H \, d\mathbf{k} \tag{4.2.14}$$

for any $\varphi, \psi \in H \otimes L^2(\Omega)$.

Proof. The assertions on the essential self-adjointness of $A + T_\omega$ and the spectrum of its closure are standard results from operator theory, see e.g. [RS78]. Since $\overline{A} \geq a > 0$ and $\operatorname{ess\,ran} \omega \subset [0, \infty)$, the operator $(A + \omega(\mathbf{k}))^{-1}$ exists and is bounded (as an operator on H) for almost all $\mathbf{k} \in \Omega$. Furthermore, $0 \notin \operatorname{spec}(\overline{A + T_\omega})$, so that $(\overline{A + T_\omega})^{-1}$ exists and is a bounded operator on $H \otimes L^2(\Omega)$.
Under the isomorphism mentioned above a vector $u \otimes v \in H \otimes L^2(\Omega)$ is identified with the map $\mathbf{k} \mapsto v(\mathbf{k})u =: (u \otimes v)(\mathbf{k}) \in H$. Correspondingly, for $u \otimes v \in \mathcal{D}\widehat{\otimes}L^2(\Omega)$, we have

$$(A \otimes I)(u \otimes v) \mapsto v(\mathbf{k})(Au) = A(v(\mathbf{k})u) = A(\mathbf{k})(v(\mathbf{k})u) = A(\mathbf{k})(u \otimes v)(\mathbf{k}),$$

where $\mathbf{k} \mapsto A(\mathbf{k}) = A$ is the constant map. By linear extension, this generalizes to $(A \otimes I)\varphi \mapsto A(\mathbf{k})\varphi(\mathbf{k})$ for any $\varphi \in \mathcal{D}\widehat{\otimes}L^2(\Omega)$. Analogously, $(I \otimes T_\omega)(\varphi) \mapsto \omega(\mathbf{k})\varphi(\mathbf{k})$ for

$\varphi \in H \widehat{\otimes} D(T_\omega)$. In terms of fiber decompositions of operators (see [RS78] for the definition of the concept) this means

$$A \otimes I = \int_\Omega^\oplus A d\mathbf{k}, \quad I \otimes T_\omega = \int_\Omega^\oplus \omega(\mathbf{k}) d\mathbf{k}. \tag{4.2.15}$$

Note that both $\mathbf{k} \mapsto A(\mathbf{k}) = A$ and $\mathbf{k} \mapsto \omega(\mathbf{k})$ are measurable in the sense of [RS78], Chapter XIII, see below.
For $\varphi, \psi \in H \otimes L^2(\Omega)$ the map $\mathbf{k} \mapsto \langle \varphi | (\overline{A} + \omega(\mathbf{k}))^{-1} | \psi \rangle$ is a composition $\mathbf{k} \mapsto \omega(\mathbf{k}) \mapsto (\overline{A} + \omega(\mathbf{k}))^{-1} \mapsto \langle \varphi | (\overline{A} + \omega(\mathbf{k}))^{-1} | \psi \rangle$ of a measurable and two continuous maps, and thus $\mathbf{k} \mapsto (\overline{A} + \omega(\mathbf{k}))^{-1}$ is a measurable map from Ω to $\mathcal{L}(H)$ by definition. Since $\overline{A} + \omega(\mathbf{k})$ (and thus also $(\overline{A} + \omega(\mathbf{k}))^{-1}$) is self-adjoint and positive (on $D(\overline{A}) \subset H$ and H, respectively) for almost all $\mathbf{k} \in \Omega$, we obtain the estimate

$$\begin{aligned}
\|\overline{A} + \omega(\cdot)\|^{-1} :=& \operatorname*{ess\,sup}_{\mathbf{k} \in \Omega} \|(\overline{A} + \omega(\mathbf{k}))^{-1}\|_{\mathcal{L}(H)} \\
=& \operatorname*{ess\,sup}_{\mathbf{k} \in \Omega} \Big(\sup \operatorname{spec}((\overline{A} + \omega(\mathbf{k}))^{-1} \Big) \\
=& \operatorname*{ess\,sup}_{\mathbf{k} \in \Omega} \Big(1/(\inf \operatorname{spec}((\overline{A} + \omega(\mathbf{k}))) \Big) \\
\leq& \Big(1/(\inf \operatorname{spec}(\overline{A}) \Big) \leq 1/a < \infty,
\end{aligned}$$

where in the second to last step we have used that $\overline{A} + \omega(\mathbf{k}) \geq \overline{A}$ since $\operatorname{ess\,ran} \omega \subset [0, \infty)$. This shows that $(\overline{A} + \omega(\cdot))^{-1} \in L^\infty(\Omega; \mathcal{L}(H))$, and thus $\int_\Omega^\oplus (\overline{A} + \omega(\mathbf{k}))^{-1} d\mathbf{k}$ is well-defined. Now fix $\psi \in \mathcal{D} \widehat{\otimes} D(T_\omega) \subset D(\overline{A + T_\omega})$ and write $\psi = (\overline{A + T_\omega})^{-1} (\overline{A + T_\omega}) \psi$. Under the isomorphism, this maps to $\psi(\mathbf{k}) = \big((\overline{A + T_\omega})^{-1} (\overline{A + T_\omega}) \psi \big)(\mathbf{k})$. On the other hand, $\psi(\mathbf{k}) \in \mathcal{D} = D(A + \omega(\mathbf{k})) \subset D(\overline{A} + \omega(\mathbf{k}))$, and by the bounded invertibility of $\overline{A} + \omega(\mathbf{k})$ we have

$$\psi(\mathbf{k}) = (\overline{A} + \omega(\mathbf{k}))^{-1} (\overline{A} + \omega(\mathbf{k})) \psi(\mathbf{k}). \tag{4.2.16}$$

Defining $\varphi := (\overline{A + T_\omega}) \psi \in H \otimes L^2(\Omega)$ and $\chi(\mathbf{k}) := (\overline{A} + \omega(\mathbf{k})) \psi(\mathbf{k}) = (A + \omega(\mathbf{k})) \psi(\mathbf{k}) \in H$, and using that on $\mathcal{D} \widehat{\otimes} D(T_\omega)$ we have $\overline{A + T_\omega} = A + T_\omega = \int_\Omega^\otimes (A + \omega(\mathbf{k})) d\mathbf{k}$ by (4.2.15), this implies

$$\varphi(\mathbf{k}) = \Big((\overline{A + T_\omega}) \psi \Big)(\mathbf{k}) = (A + \omega(\mathbf{k})) \psi(\mathbf{k}) = \chi(\mathbf{k}). \tag{4.2.17}$$

Putting together (4.2.16) and (4.2.17), we obtain

$$\big((\overline{A + T_\omega})^{-1} \varphi \big)(\mathbf{k}) = \psi(\mathbf{k}) = (\overline{A} + \omega(\mathbf{k}))^{-1} \chi(\mathbf{k}) = (\overline{A} + \omega(\mathbf{k}))^{-1} \varphi(\mathbf{k}).$$

This shows

$$\big(\overline{A + T_\omega} \big)^{-1} = \int_{\mathbb{R}^n}^\oplus (\overline{A} + \omega(\mathbf{k}))^{-1} d\mathbf{k}$$

on $\overline{A + T_\omega} \big(\mathcal{D} \widehat{\otimes} D(T_\omega) \big)$, which is dense in $H \otimes L^2(\Omega)$ since $\overline{A + T_\omega}$ is onto and has $\mathcal{D} \widehat{\otimes} D(T_\omega)$ as a core by construction of the operator closure. $\square$

In the next Lemma we apply the previous abstract result to the reduced resolvent T^σ restricted to various of its invariant subspaces.

Lemma 4.2.7 (Fiber decomposition of reduced resolvents)**.** *Assume (A1) and (A2) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Set $\Omega_\sigma := \{\omega(\mathbf{k}) \geq \sigma\} \subset \mathbb{R}^3$. Then*

i.

$$\begin{aligned}
&\left(T^\sigma_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}} \psi\right)(\mathbf{k},\lambda) \\
&= \left(\overline{H_A + H_B} + \hbar\omega(\mathbf{k})_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}}\right)^{-1} \psi(\mathbf{k},\lambda),
\end{aligned}$$

$$\begin{aligned}
&\left\langle \varphi | T^\sigma_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}} | \psi \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}} \\
&= \sum_{\lambda=1,2} \int_{\Omega_\sigma} d\mathbf{k} \left\langle \varphi(\mathbf{k},\lambda) | \left(\overline{H_A + H_B} + \hbar\omega(\mathbf{k})_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}}\right)^{-1} | \psi(\mathbf{k},\lambda) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}
\end{aligned} \tag{4.2.18}$$

for all $\varphi, \psi \in \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}$, and

$$\begin{aligned}
&\left(T^\sigma_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}} \psi\right)(\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu) \\
&= \left(\overline{H_A + H_B} + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}}\right)^{-1} \psi(\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu),
\end{aligned}$$

$$\begin{aligned}
&\left\langle \varphi | T^\sigma_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}} | \psi \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}} \\
&= \sum_{\lambda,\mu=1,2} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \\
&\left\langle \varphi(\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu) | \left(\overline{H_A + H_B} + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}}\right)^{-1} | \psi(\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}
\end{aligned} \tag{4.2.19}$$

for all $\varphi, \psi \in \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}$.

ii.

$$\begin{aligned}
&\left(T^\sigma_{\overline{A|\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}} \psi\right)(\mathbf{k},\lambda) \\
&= \left(H_A + \hbar\omega(\mathbf{k})_{|\{\Psi_A^0\}^\perp}\right)^{-1} \psi(\mathbf{k},\lambda),
\end{aligned}$$

$$\begin{aligned}
&\left\langle \varphi | T^\sigma_{\overline{A|\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}} | \psi \right\rangle_{\mathcal{H}_A \otimes \mathcal{F}} \\
&= \sum_{\lambda=1,2} \int_{\Omega_\sigma} d\mathbf{k} \left\langle \varphi(\mathbf{k},\lambda) | \left(H_A + \hbar\omega(\mathbf{k})_{|\{\Psi_A^0\}^\perp}\right)^{-1} | \psi(\mathbf{k},\lambda) \right\rangle_{\mathcal{H}_A}
\end{aligned}$$

for all $\varphi, \psi \in \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}$, *and*

$$\begin{aligned}
&\left(T^\sigma_{\overline{A|\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}} \psi\right)(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu) \\
&= \left(H_A + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{|\{\Psi_A^0\}^\perp}\right)^{-1} |\psi(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu),
\end{aligned}$$

$$\begin{aligned}
&\left\langle \varphi | T^\sigma_{\overline{A|\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}} | \psi \right\rangle_{\mathcal{H}_A \otimes \mathcal{F}} \\
&= \sum_{\lambda,\mu=1,2} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \\
&\left\langle \varphi(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu) | \left(H_A + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{|\{\Psi_A^0\}^\perp}\right)^{-1} |\psi(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu)\right\rangle_{\mathcal{H}_A}
\end{aligned}$$

for all $\varphi, \psi \in \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}$.

iii.

$$\begin{aligned}
&\left(T^\sigma_{\overline{B|\{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}} \psi\right)(\mathbf{k}, \lambda) \\
&= \left(H_B + \hbar\omega(\mathbf{k})_{|\{\Psi_B^0\}^\perp}\right)^{-1} \psi(\mathbf{k}, \lambda),
\end{aligned}$$

$$\begin{aligned}
&\left\langle \varphi | T^\sigma_{\overline{B|\{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}} | \psi \right\rangle_{\mathcal{H}_B \otimes \mathcal{F}} \\
&= \sum_{\lambda=1,2} \int_{\Omega_\sigma} d\mathbf{k} \left\langle \varphi(\mathbf{k}, \lambda) | \left(H_B + \hbar\omega(\mathbf{k})_{|\{\Psi_B^0\}^\perp}\right)^{-1} |\psi(\mathbf{k}, \lambda)\right\rangle_{\mathcal{H}_B}
\end{aligned}$$

for all $\varphi, \psi \in \overline{\{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}$, *and*

$$\begin{aligned}
&\left(T^\sigma_{\overline{B|\{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}} \psi\right)(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu) \\
&= \left(H_B + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}\right)^{-1} \psi(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu),
\end{aligned}$$

$$\begin{aligned}
&\left\langle \varphi | T^\sigma_{\overline{B|\{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}} | \psi \right\rangle_{\mathcal{H}_B \otimes \mathcal{F}} \\
&= \sum_{\lambda,\mu=1,2} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \\
&\left\langle \varphi(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu) | \left(H_B + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}\right)^{-1} |\psi(\mathbf{k}_1, \mathbf{k}_2, \lambda, \mu)\right\rangle_{\mathcal{H}_B}
\end{aligned}$$

for all $\varphi, \psi \in \overline{\{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}$.

Proof. We prove i) and remark that the assertions in ii) and iii) are shown completely analogously. Since $\mathcal{F}_\sigma^{(1)} = W_\sigma \oplus W_\sigma$, where $W_\sigma = L^2(\Omega_\sigma)$, we have the natural isomorphism

$$\begin{aligned}\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}} &= (\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}) \otimes \mathcal{F}_\sigma^{(1)} \\ \cong &((\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}) \otimes W_\sigma) \oplus ((\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}) \otimes W_\sigma) \\ \cong &L^2(\Omega_\sigma; \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}) \oplus L^2(\Omega_\sigma; \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}),\end{aligned}$$

where the first equality holds by the definition of the tensor product of Hilbert spaces. With respect to this isomorphism a vector $\varphi \in \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}$ is represented by the function $\mathbf{k} \mapsto \begin{pmatrix} \varphi(\mathbf{k}, \lambda = 1) \\ \varphi(\mathbf{k}, \lambda = 2) \end{pmatrix}$, on which T^σ acts as

$$\begin{pmatrix} \int_{\Omega_\sigma}^\oplus \left(\overline{H_A + H_B} + \hbar\omega(\mathbf{k})_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}} \right)^{-1} d\mathbf{k} & 0 \\ 0 & \int_{\Omega_\sigma}^\oplus \left(\overline{H_A + H_B} + \hbar\omega(\mathbf{k})_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}} \right)^{-1} d\mathbf{k} \end{pmatrix},$$

see Lemmas 4.2.5 and 4.2.6. Since $\mathcal{F}_\sigma^{(2)} = S_2(W_\sigma \otimes W_\sigma)$ (S_2 being the symmetrizer, i.e. the orthogonal projection onto the subspace of functions which are invariant under permutation of the variables) and $H_{f \geq \sigma}$ commutes with S_2, we conclude that, as above, T^σ acts on a function

$$(\mathbf{k}_1, \mathbf{k}_2) \mapsto \begin{pmatrix} \varphi(\mathbf{k}_1, \mathbf{k}_2, \lambda = 1, \mu = 1) \\ \varphi(\mathbf{k}_1, \mathbf{k}_2, \lambda = 1, \mu = 2) \\ \varphi(\mathbf{k}_1, \mathbf{k}_2, \lambda = 2, \mu = 1) \\ \varphi(\mathbf{k}_1, \mathbf{k}_2, \lambda = 2, \mu = 2) \end{pmatrix} \in \varphi \in \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(2)}}$$

by componentwise application of

$$\int_{\Omega_\sigma \times \Omega_\sigma}^\oplus \left(\overline{H_A + H_B} + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{\overline{|\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}} \right)^{-1} d\mathbf{k}_1 d\mathbf{k}_2,$$

The identities (4.2.18) and (4.2.19) now follow by (4.2.14) and the definition of the inner product on $\mathcal{F}_\sigma^{(1)}$ and $\mathcal{F}_\sigma^{(2)}$, respectively. □

4.2.4 Ground state and parity invariance

Throughout this section, we assume (A1) and (A2), as well as $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. In particular, Proposition 2.5.2 then guarantees the existence of real, non-degenerate and rotation-invariant ground states $\{\Psi_A^0\}$ and $\{\Psi_B^0\}$ of H_A and H_B, which both possess a definite parity. In the following, we will show that the reduced resolvent commutes with the orthogonal projections onto Ψ_A^0 and Ψ_B^0 and with the parity operators.

Lemma 4.2.8 (Ground state projections commute with T^σ)**.** *Let*

$$T^\sigma = \Big((H_0^\sigma - E_0)|_{\{\Psi_0\}^\perp}\Big)^{-1}$$

be the reduced resolvent of $H_0^\sigma = \overline{H_A + H_B + H_{f\geq\sigma}}$*, where* $\Psi_0 = \Psi_A^0 \otimes \Psi_B^0 \otimes \Omega$*, and* Ψ_A^0*,* Ψ_B^0 *are the (non-degenerate) ground states of* H_A *and* H_B*, respectively. Then the projections*

$$\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}} = P_{\{\Psi_A^0\}\widehat{\otimes}(\mathcal{H}_B \otimes \mathcal{F})}$$

and

$$\overline{P_{\{\Psi_B^0\}} \otimes I_{\mathcal{H}_A \otimes \mathcal{F}}} = P_{\{\Psi_B^0\}\widehat{\otimes}(\mathcal{H}_A \otimes \mathcal{F})}$$

commute with T^σ *on* $\{\Psi_0\}^\perp$*, i.e.*

$$[T, \overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}}]_{\{\Psi_0\}^\perp} = [T, \overline{P_{\{\Psi_B^0\}} \otimes I_{\mathcal{H}_A \otimes \mathcal{F}}}]_{\{\Psi_0\}^\perp} = 0.$$

Proof. Without loss of generality, assume $E_A^0 = E_B^0 = 0$, i.e. $E_0 = E_A^0 + E_B^0 = 0$. The fact that $\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}}$ and $\overline{P_{\{\Psi_B^0\}} \otimes I_{\mathcal{H}_A \otimes \mathcal{F}}}$ are the projections onto $\{\Psi_A^0\}\widehat{\otimes}(\mathcal{H}_B \otimes \mathcal{F})$ and $\{\Psi_B^0\}\widehat{\otimes}(\mathcal{H}_A \otimes \mathcal{F})$ follows from Lemma 2.6.1. Let $u = u_1 \otimes u_2 \otimes u_3 \in (\mathcal{H}_A \widehat{\otimes} \mathcal{H}_B \widehat{\otimes} \mathcal{F}) \cap \{\Psi_0\}^\perp$. Then $(\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})u = (P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}})u = (c\Psi_A^0) \otimes u_2 \otimes u_3 \in \{\Psi_A^0\}\widehat{\otimes}(\{\Psi_B^0\}\widehat{\otimes}\{\Omega\})^\perp$, which is an invariant subspace of T^σ (Lemma 4.2.5). We have $T^\sigma(\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})u = T^\sigma((c\Psi_A^0) \otimes u_2 \otimes u_3) = (c\Psi_A^0) \otimes T_B^\sigma(u_2 \otimes u_3)$, where

$$T_B^\sigma = \Big((\overline{H_B + H_{f\geq\sigma}})|_{\{\Psi_B^0 \otimes \Omega\}^\perp}\Big)^{-1}$$

(this follows from $T^\sigma_{|\{\Psi_A^0\}\widehat{\otimes}(\{\Psi_B^0\}\widehat{\otimes}\{\Omega\})^\perp} = I_{\{\Psi_A^0\}} \otimes T_B^\sigma$, see Lemma 4.2.5). On the other hand,

$$\begin{aligned}
&(\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})T^\sigma u \\
=&(\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})T^\sigma\Big((\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})u + (\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})^\perp u\Big) \\
=&(\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})\Big((c\Psi_A^0) \otimes T_B^\sigma(u_2 \otimes u_3)\Big) \\
&+ (\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}})T^\sigma\Big((I - P_{\{\Psi_A^0\}}u_1) \otimes u_2 \otimes u_3\Big) \\
=&(c\Psi_A^0) \otimes T_B^\sigma(u_2 \otimes u_3) + (P_{\{\Psi_A^0\}\widehat{\otimes}(\mathcal{H}_B \otimes \mathcal{F})})T^\sigma\Big(\underbrace{(I - P_{\{\Psi_A^0\}}u_1) \otimes u_2 \otimes u_3}_{\in \{\Psi_A^0\}^\perp\widehat{\otimes}(\{\Psi_B^0\}\widehat{\otimes}\{\Omega\})^\perp}\Big).
\end{aligned}$$

Since $\{\Psi_A^0\}^\perp\widehat{\otimes}(\{\Psi_B^0\}\widehat{\otimes}\{\Omega\})^\perp$ is also an invariant subspace for T^σ, we conclude

$$(P_{\{\Psi_A^0\}\widehat{\otimes}(\mathcal{H}_B \otimes \mathcal{F})})T^\sigma((I - P_{\{\Psi_A^0\}}u_1) \otimes u_2 \otimes u_3) = 0,$$

which (by linear extension) implies that $T^\sigma\overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}} = \overline{P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}}T^\sigma$ on $(\mathcal{H}_A\widehat{\otimes}\mathcal{H}_B\widehat{\otimes}\mathcal{F}) \cap \{\Psi_0\}^\perp$, the latter being a dense subspace of $(\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}) \cap \{\Psi_0\}^\perp$ by construction, so that the assertion follows. □

Definition 4.2.9 (Parity operators)**.** Let $\psi \in \mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}$, where $\mathcal{H}_A$ and $\mathcal{H}_B$ are (symmetry subspaces of) $L^2(\mathbb{R}^{3Z_A})$ and $L^2(\mathbb{R}^{3Z_B})$ and $\mathcal{F}$ is a bosonic Fock space over a Hilbert space $\mathcal{H}$. The parity operators P_A ('with respect to A'), P_B ('with respect to B') and the 'joint' parity operator P are defined by their action in the representation

$$\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F} \cong L^2(\mathbb{R}^{3Z_A} \times \mathbb{R}^{3Z_B}; \oplus_{n=0}^{\infty} \mathcal{F}^{(n)})$$

as follows:

$$\begin{aligned}
(P_A\psi)_n(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}, \mathbf{y}_1, \ldots, \mathbf{y}_{Z_B}) &:= \psi_n(-\mathbf{x}_1, \ldots, -\mathbf{x}_{Z_A}, \mathbf{y}_1, \ldots, \mathbf{y}_{Z_B}),\\
(P_B\psi)_n(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}, \mathbf{y}_1, \ldots, \mathbf{y}_{Z_B}) &:= \psi_n(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}, -\mathbf{y}_1, \ldots, -\mathbf{y}_{Z_B}),\\
(P\psi)_n(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}, \mathbf{y}_1, \ldots, \mathbf{y}_{Z_B}) &:= \psi_n(-\mathbf{x}_1, \ldots, -\mathbf{x}_{Z_A}, -\mathbf{y}_1, \ldots, -\mathbf{y}_{Z_B}).
\end{aligned}$$

Remark 4.2.10. Note that P_A, P_B and P are isometries satisfying $P_A^2 = P_B^2 = P^2 = I_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}}$ (such operators are sometimes called 'involutions'). Since they differ from $\pm I$, they all have spectrum $\{1, -1\}$. The two eigenvalues 1 and -1 correspond to functions of 'even' and 'odd' parity (with respect to A, B or 'joint'). The orthogonal projections onto the eigenspaces can be constructed explicitly by decomposing a given function into its even and odd part ($f = f_+ + f_-$, $f_\pm(x) := 1/2(f(x) \pm f(-x))$. Furthermore, note that $P_A P_B = P_B P_A = P$.

Lemma 4.2.11 (Parity invariance properties of the reduced resolvent)**.** *Let P, P_A, P_B be the parity operators defined above. Let*

$$T^\sigma = \left((H_0^\sigma - E_0)|_{\{\Psi_0\}^\perp}\right)^{-1}$$

be the reduced resolvent of $H_0^\sigma = \overline{H_A + H_B + H_{f\geq\sigma}}$, where $\Psi_0 = \Psi_A^0 \otimes \Psi_B^0 \otimes \Omega$, and Ψ_A^0, Ψ_B^0 are the (non-degenerate) ground states of H_A and H_B, respectively. Then

$$[T^\sigma, P \otimes I_{\mathcal{F}}] = [T^\sigma, P_A \otimes I_{\mathcal{H}_B} \otimes I_{\mathcal{F}}] = [T^\sigma, I_{\mathcal{H}_A} \otimes P_B \otimes I_{\mathcal{F}}] = 0.$$

In particular, T^σ leaves the (separate and joint) parity eigenspaces invariant.

Proof. Assume without loss of generality that $E_0 = 0$. First note that $H_A \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}$, $I_{\mathcal{H}_A} \otimes H_B \otimes I_{\mathcal{F}}$ and $I_{\mathcal{H}_A \otimes \mathcal{H}_B} \otimes H_{f\geq\sigma}$ all commute with P_A, P_B and P on their respective domains $D(H_A)\widehat{\otimes}(\mathcal{H}_B \otimes \mathcal{F})$, $D(H_B)\widehat{\otimes}(\mathcal{H}_A \otimes \mathcal{F})$ and $(\mathcal{H}_A \otimes \mathcal{H}_B)\widehat{\otimes}D(H_{f\geq\sigma})$ in the sense that $P_A(H_A \otimes I_{\mathcal{H}_B \otimes \mathcal{F}}) \subset (H_A \otimes I_{\mathcal{H}_B \otimes \mathcal{F}})P_A$ etc. (see Proposition 2.5.1). Therefore $H_A + H_B + H_{f\geq\sigma}$ (with domain $D(H_A)\widehat{\otimes}D(H_B)\widehat{\otimes}D(H_{f\geq\sigma})$) commutes with P_A, P_B and P. Since ψ_A and ψ_B are non-degenerate eigenfunctions of H_A and H_B by assumption, they must have a definite parity, i.e. be eigenfunctions of P_A and P_B, respectively. This implies that $\{\Psi_0\}$ is a common eigenspace of P_A, P_B and P. Therefore its orthogonal complement $\{\Psi_0\}^\perp$ is left invariant by P_A, P_B and P, which in turn implies that $H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp}$ (with domain $(D(H_A)\widehat{\otimes}D(H_B)\widehat{\otimes}D(H_{f\geq\sigma})) \cap \{\Psi_0\}^\perp$) commutes with P_A, P_B and P.
By Corollary 2.6.3,

$$H_0^\sigma|_{\{\Psi_0\}^\perp} = \overline{H_A + H_B + H_{f\geq\sigma}}|_{\{\Psi_0\}^\perp} = \overline{H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp}}.$$

Now let $u \in D(\overline{H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp}})$ and choose

$$\{u_n\} \subset D(H_A)\widehat{\otimes}D(H_B)\widehat{\otimes}D(H_{f\geq\sigma})) \cap \{\Psi_0\}^\perp$$

with $u_n \to u$ and $(H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp})u_n \to H^\sigma_0|_{\{\Psi_0\}^\perp}u$. Since P_A, P_B and P are continuous, we have

$$\begin{aligned} PH^\sigma_0|_{\{\Psi_0\}^\perp}u &= \lim_{n\to\infty} P(H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp})u_n \\ &= \lim_{n\to\infty} (H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp})Pu_n, \end{aligned}$$

where we have used the above commutator property. The continuity of P implies $Pu_n \to Pu$, and the construction of the operator closure allows us to conclude $Pu \in D(H^\sigma_0|_{\{\Psi_0\}^\perp})$ and

$$\lim_{n\to\infty} (H_A + H_B + H^\sigma_{f|\{\Psi_0\}^\perp})Pu_n = H^\sigma_0|_{\{\Psi_0\}^\perp}Pu,$$

which establishes $PH^\sigma_0|_{\{\Psi_0\}^\perp} \subset H^\sigma_0|_{\{\Psi_0\}^\perp}P$. Now for any densely defined operator $A : D(A) \to H$ which is boundedly invertible and which commutes with a bounded operator B (in the sense that $BA \subset AB$), we have

$$A^{-1}Bu = A^{-1}BAv = A^{-1}ABv = Bv = BA^{-1}u$$

for every $u \in H$ and $v = A^{-1}u \in D(A)$, which shows $[A^{-1}, B] = 0$. Applying this to $A = H^\sigma_0|_{\{\Psi_0\}^\perp}$ and $B = P$ proves the assertion. The same argument is valid for P_A and P_B.

□

4.2.5 Rotation invariance

In this section we establish results that will allow us to exploit the rotation invariance of the atomic Hamiltonians and their ground states in simplifying certain expressions that will arise in the conversion of the $V^\sigma_i(\Lambda, \mathbf{R})$ into integrals over photon momenta. More precisely, the results in this section will allow us to eliminate the polarization vectors from the calculation, resulting in expressions whose only photon degrees of freedom are the momenta.
We begin by proving a simple fact about expectations of rotation-invariant operators on states involving a rotation-invariant wave function and components of the position and momentum operators.

Lemma 4.2.12 (Rotation invariance I)**.** *Let $a, b \in \mathbb{R}^3$. Suppose that $\Psi, \Phi \in L^2(\mathbb{R}^{3N})$ and the bounded operator $A : L^2(\mathbb{R}^{3N}) \to L^2(\mathbb{R}^{3N})$ are rotation invariant in the sense that for every $R \in SO(3)$, $(U_R\Psi)(x_1, \ldots, x_N) = \Psi(R^{-1}x_1, \ldots, R^{-1}x_N) = \Psi(x_1, \ldots, x_N)$ for almost all $(x_1, \ldots, x_N) \in \mathbb{R}^{3N}$, and $[A, U_R] = 0$. Furthermore, let $p_i = -\mathrm{i}\hbar\nabla_{x_i}$ be the operator of momentum for the i-th particle. Then the following identities hold.*

i. If $x_i\Psi$ and $x_j\Phi$ are in $L^2(\mathbb{R}^{3N})$, then

$$\langle (a \cdot x_i)\Psi | A | (b \cdot x_j)\Phi \rangle = \frac{1}{3}(a \cdot b)\langle x_i\Psi | A | x_j\Phi \rangle.$$

ii. If Ψ and Ψ are in $H^1(\mathbb{R}^{3N})$, then

$$\langle (a \cdot p_i)\Psi | A | (b \cdot p_j)\Phi \rangle = \frac{1}{3}(a \cdot b)\langle p_i\Psi | A | p_j\Phi \rangle.$$

iii. If $x_i\Psi$ and $x_j\Phi$ are in $L^2(\mathbb{R}^{3n})$ and $\Psi, \Phi \in H^1(\mathbb{R}^{3N})$, then

$$\langle (a \cdot x_i)\Psi | A | (b \cdot p_j)\Phi \rangle = \frac{1}{3}(a \cdot b)\langle x_i\Psi | A | p_j\Phi \rangle.$$

Proof. i) Choose $R \in SO(3)$ such that $R\hat{a} = e_1$, the first standard unit vector of $\mathbb{R}^3$. Then

$$\begin{aligned}
&\langle (a \cdot x_i)\Psi | A | (b \cdot x_j)\Phi \rangle \\
=&\langle (U_R((a \cdot x_i)\Psi) | U_R A | (b \cdot x_j)\Phi \rangle \\
=&\langle (a \cdot R^{-1}x_i)\Psi | A[(b \cdot R^{-1}x_j)\Phi] \rangle \\
=&\langle (Ra \cdot x_i)\Psi | A[(Rb \cdot x_j)\Phi] \rangle \\
=&\langle (Ra \cdot x_i)\Psi | Rb \cdot A[x_j\Phi] \rangle \\
=&|a||b| \int dx (e_1 \cdot x_i)\overline{\Psi(x)}(R\hat{b} \cdot A[x_j\Phi](x)) \\
=&|a||b| \int dx\ x_i^1\ \overline{\Psi(x)}\Big[(e_1 \cdot R\hat{b})(e_1 \cdot A[x_j\Phi](x)) + (e_2 \cdot R\hat{b})(e_2 \cdot A[x_j\Phi](x)) \\
&+ (e_3 \cdot R\hat{b})(e_3 \cdot A[x_j\Phi](x))\Big] \\
=&|a||b|(\hat{a} \cdot \hat{b}) \int dx\ x_i^1\ \overline{\Psi(x)}A[x_j^1\Phi](x) \\
&+ (e_2 \cdot R\hat{b}) \int dx\ x_i^1\ \overline{\Psi(x)}A[x_j^2\Phi](x) \\
&+ (e_3 \cdot R\hat{b}) \int dx\ x_i^1\ \overline{\Psi(x)}A[x_j^3\Phi](x).
\end{aligned}$$

In the second term, x_i^1 changes sign under the transformation $T \times \cdots \times T$, $T : (e_1, e_2, e_3) \mapsto (-e_1, e_2, -e_3) \in SO(3)$, while the remaining integrand $\overline{\Psi(x)}A[x_j^2\Phi](x)$ is invariant under this transformation by assumption. Thus the second term vanishes. For the first term, apply the same argument with the transformation $T : (e_1, e_2, e_3) \mapsto (-e_1, -e_2, e_3) \in SO(3)$ Thus only the term

$$\begin{aligned}
|a||b|(\hat{a} \cdot \hat{b}) \int dx\ x_i^1\ \overline{\Psi(x)}A[x_j^1\Phi](x) =&(a \cdot b) \int dx\ \ \overline{\Psi(x)}A[x_j^1\Phi](x) \\
=&(a \cdot b)\langle x_i^1\Psi | A | x_j^1\Phi \rangle
\end{aligned}$$

survives. Repeating the above argument with matrices $R \in SO(3)$ mapping $\hat{a}$ to e_2 and e_3, respectively, and summing up yields

$$3\langle (a \cdot x_i)\Psi | A | (b \cdot x_j)\Phi \rangle = \sum_{\alpha=1}^{3} (a \cdot b)\langle x_i^\alpha\Psi | A | x_j^\alpha\Phi \rangle = (a \cdot b)\langle x_i\Psi | A | x_j\Phi \rangle.$$

ii), iii) Since for an H^1-function Ψ which is rotationally invariant in the above sense, we have

$$U_R[\nabla_{x_i}\Psi](x) = [\nabla_{x_i}\Psi]([R^{-1} \times \cdots \times R^{-1}]x) = R^{-1}[\nabla_{x_i}\Psi(x)]$$

(as is easily seen using the chain rule), the operators p_i exhibit the same transformation behaviour as x_i, so the above reasoning applies without alteration. □

To generalize the preceding result to situations in which not one but two components of the position (or momentum) operators appear on each side of the inner product, we first prove the following two results, which will yield a decomposition of the resolvent of an operator with a 'non-interacting' structure:

Lemma 4.2.13. *Let $A : D(A) \to H$ be a positive self-adjoint operator satisfying $A \geq \Delta > 0$. Then for any $\lambda \geq 0$ and all $x \in H$, the identity*

$$(A + \lambda)^{-1}x = \int_0^\infty ds\, \mathrm{e}^{-\lambda s}\, \mathrm{e}^{-sA}x$$

holds.

Proof. Since $A - \Delta \geq 0$, we have $-A + \Delta \leq 0$ and $(0, \infty) \subset \rho(-A + \Delta)$. Furthermore, $\|\lambda(\lambda - (-A + \Delta))^{-1}\| \leq |\lambda|(\mathrm{dist}(\lambda, \mathrm{spec}(-A + \Delta))^{-1} \leq 1$ for any $\lambda > 0$. By the Hille-Yosida theorem (or by the spectral theorem), $\exp(t(-A + \Delta))$ is a strongly continuous contraction semigroup. For generators of such groups we have the following representation of the resolvent in terms of the semigroup (see e.g. [Eva98], Theorem 7.4.3):

$$-(-A + \Delta - \mu)^{-1}x = (A - \Delta + \mu)^{-1}x = \int_0^\infty ds\, \mathrm{e}^{-\mu s}\, \mathrm{e}^{-s(A-\Delta)}x$$

for every $x \in H$ and $\mu \in (0, \infty)$. Choosing $\mu = \Delta + \lambda > 0$ and using the Trotter product formula (see [RS80], Theorem VIII.31) to conclude

$$\mathrm{e}^{-(\Delta+\lambda)s}\mathrm{e}^{-sA+\Delta s}x = \mathrm{e}^{-\lambda s}\mathrm{e}^{-sA}x$$

yields the assertion. □

Lemma 4.2.14. *Let A and B be self-adjoint operators on the Hilbert spaces H_1 and H_2, respectively. Assume that $A \geq \Delta_A > 0$, $B \geq \Delta_B > 0$, and consider the self-adjoint operator $T = \overline{A \otimes I + I \otimes B}$. Then* $\mathrm{spec}(T) \subset [\Delta_A + \Delta_B, \infty)$, *$T \geq \Delta_A + \Delta_B > 0$, and*

$$\mathrm{e}^{-tT} = \overline{\mathrm{e}^{-tA} \otimes \mathrm{e}^{-tB}}, \quad \forall t \geq 0, \tag{4.2.20}$$

$$\left(\mathrm{e}^{-tT}\right)_{|H_1 \widehat{\otimes} H_2} = \mathrm{e}^{-tA} \otimes \mathrm{e}^{-tB}, \quad \forall t \geq 0. \tag{4.2.21}$$

Proof. The assertion on the self-adjointness, the spectrum and the semi-boundedness of T are standard properties of the tensor product operator construction (see e.g. [RS80], Theorem VIII.33).
Since $A \geq \Delta_A > 0$, $B \geq \Delta_B > 0$, $T \geq \Delta_A + \Delta_B =: \Delta > 0$, we know that e^{-tA}, e^{-tB} and e^{-tT} are strongly continuous contraction semigroups.
Let $u \otimes v \in D(A)\widehat{\otimes}D(B)$ and define $\psi(t) := \mathrm{e}^{-tT}(u \otimes v)$. Since $D(A)\widehat{\otimes}D(B) \subset D(T)$, we have $\psi(t) \in C^1([0, \infty), D(T))$ and $\frac{d}{dt}\psi(t) = -T\psi(t)$, i.e. $\psi(t)$ solves the initial value problem $\frac{d}{dt}\psi(t) = -T\psi(t)$, $\psi(0) = u \otimes v$. Now consider $\tilde{\psi}(t) := \mathrm{e}^{-tA}u \otimes \mathrm{e}^{-tB}v$. We have $\tilde{\psi}(t) \in D(A)\widehat{\otimes}D(B)$ for all $t \geq 0$ since e^{-tA} and e^{-tB} leave the domains of their respective

generators invariant. Furthermore, $\tilde{\psi}(0) = u \otimes v$, and

$$\begin{aligned}
&\frac{d}{dt}\tilde{\psi}(t) = \lim_{h\to 0}\left(\frac{1}{h}(\tilde{\psi}(t+h) - \tilde{\psi}(t))\right)\\
=&\lim_{h\to 0}\Big(\frac{1}{h}\Big[(\mathrm{e}^{-(t+h)A} - \mathrm{e}^{-tA})u \otimes \mathrm{e}^{-tB}v + \mathrm{e}^{-(t+h)A}u \otimes (\mathrm{e}^{-(t+h)B} - \mathrm{e}^{-tB})v\Big]\Big)\\
=&(-A\mathrm{e}^{-tA})u \otimes \mathrm{e}^{-tB}v + \mathrm{e}^{-tA}u \otimes (-B\mathrm{e}^{-tB}v)\\
=&-(A \otimes I + I \otimes B)\left(\mathrm{e}^{-tA}u \otimes \mathrm{e}^{-tB}v\right)\\
=&-(A \otimes I + I \otimes B)\,\tilde{\psi}(t)\\
=&-T\tilde{\psi}(t)
\end{aligned}$$

by the semigroup properties of $\exp(-tA)$ and $\exp(-tB)$ and the fact that $T = A \otimes I + I \otimes B$ on $D(A)\widehat{\otimes}D(B)$ by construction. But this means that $\tilde{\psi}(t)$ also solves the above initial value problem. Since this solution is unique (this is a standard result on operator semigroups, see e.g. [Wer00], Satz VII.4.), we conclude

$$\psi(t) = \mathrm{e}^{-tT}(u \otimes v) = \left(\mathrm{e}^{-tA} \otimes \mathrm{e}^{-tB}\right)(u \otimes v) = \tilde{\psi}(t).$$

Since this identity holds for any $u \otimes v \in D(A)\widehat{\otimes}D(B)$, it extends to all of $D(A)\widehat{\otimes}D(B)$ by linearity and to all of $H_1 \otimes H_2$ by density, proving (4.2.20). This last step requires passing to the closure $\overline{(\mathrm{e}^{-tA} \otimes \mathrm{e}^{-tB})}$ of the operator on the right-hand side. Restricting both sides of (4.2.20) to the algebraic tensor product $D(\exp(-tA)\otimes\exp(-tB)) = H_1\widehat{\otimes}H_2 \subset H_1 \otimes H_2$ yields (4.2.21) □

Next we apply the preceding results to the resolvent of

$$(\overline{H_A \otimes I + I \otimes H_B})_{\overline{|\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}}.$$

Proposition 4.2.15. *Assume (A1) and (A2) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Let H_A and H_B be the atomic Hamiltonians, and assume without loss of generality that* $\inf\mathrm{spec}(H_A) = \inf\mathrm{spec}(H_B) = 0$. *Let $\omega(\mathbf{k}) = c|\mathbf{k}|$ be the photonic dispersion relation. Then*

$$\begin{aligned}
&\left((\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{\overline{|\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}}\right)^{-1}(u \otimes v)\\
=&\int_0^\infty ds\,\left(\mathrm{e}^{-s(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}}u \otimes \mathrm{e}^{-s(H_B+\omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}}v\right)
\end{aligned}$$

for all $\mathbf{k}_1, \mathbf{k}_2 \in \mathbb{R}^3$ and every $u \otimes v \in \mathcal{H}_A \otimes \mathcal{H}_B$. Furthermore, the operators

$$\mathrm{e}^{-s(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}} \text{ and } \mathrm{e}^{-s(H_B+\omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}}$$

are rotation invariant operators on the Hilbert spaces $\{\Psi_A^0\}^\perp \subset \mathcal{H}_A$ and $\{\Psi_B^0\}^\perp \subset \mathcal{H}_B$ (in the sense that they commute with the family U_R of operators, see Proposition 2.5.1).

Proof. Set $M := \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}$. We have $(\overline{H_A + H_B})_{|M} \geq \Delta_A + \Delta_B =: \Delta > 0$. Thus by Lemma 4.2.13, we have (choosing $z = \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2) \geq 0$)

$$\begin{aligned}
&\Big((\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{|\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}}\Big)^{-1} (u \otimes v) \\
&= \int_0^\infty ds\, \mathrm{e}^{-(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))s}\, \mathrm{e}^{-s(\overline{H_A+H_B})_{|M}} (u \otimes v).
\end{aligned}$$

By Corollary 2.6.3, $(\overline{H_A + H_B})_{|M} = \overline{(H_A + H_B)_{|M}}$. Furthermore, as was shown in the proof of Lemma 4.2.5, $\overline{(H_A + H_B)_{|M}} = \overline{H_{A|\{\Psi_A^0\}^\perp} + H_{B|\{\Psi_B^0\}^\perp}}$. We have $H_{A|\{\Psi_A^0\}^\perp} \geq \Delta_A > 0$, $H_{B|\{\Psi_B^0\}^\perp} \geq \Delta_B > 0$, and thus Lemma 4.2.14 is applicable, yielding

$$\begin{aligned}
&\Big((\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))_{|\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}}\Big)^{-1} (u \otimes v) \\
&= \int_0^\infty ds\, \mathrm{e}^{-s(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))} \left(\mathrm{e}^{-sH_{A|\{\Psi_A^0\}^\perp}} u \otimes \mathrm{e}^{-sH_{B|\{\mathbf{A}\}^\perp}} v\right) \\
&= \int_0^\infty ds\, \mathrm{e}^{-s(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}} u \otimes \mathrm{e}^{-s(H_B+\omega(\mathbf{k}_s))_{|\{\Psi_B^0\}^\perp}} v,
\end{aligned}$$

where we have used the Trotter product formula ([RS80], Theorem VIII.31) for the last identity.
To prove the claim about the rotation invariance, recall that by Proposition 2.5.1, $U_R H_{A,B} = H_{A,B} U_R$ (as unbounded operators on $\mathcal{H}_{A,B}$). As noted in the remarks after that proposition, the ground state eigenfunctions Ψ_A^0 and Ψ_B^0, and thus also the one-dimensional subspaces spanned by them, are invariant under the family $\{U_R | R \in SO(3)\}$. Therefore the orthogonal complements $\{\Psi_A^0\}^\perp$ and $\{\Psi_B^0\}^\perp$ are also left invariant by the family $\{U_R | R \in SO(3)\}$, so that we immediately obtain $U_R(H_{A,B|\{\psi_{A,B}^0\}^\perp}) = (H_{A,B|\{\psi_{A,B}^0\}^\perp})U_R$ as operators on $\{\psi_{A,B}^0\}^\perp$. Now the identities

$$\mathrm{e}^{-s(H_{A,B}+\omega(\mathbf{k}))_{|\{\psi_{A,B}^0\}^\perp}} U_R = U_R \mathrm{e}^{-s(H_{A,B}+\omega(\mathbf{k}))_{|\{\psi_{A,B}^0\}^\perp}}$$

(as bounded operators on $\{\psi_{A,B}^0\}^\perp$) follow from the spectral theorem (see e.g. [Con90], Theorem 4.11). □

We now prove the generalization of Lemma 4.2.12 to the situation of operators that allow for a decomposition of the form just considered.

Lemma 4.2.16 (Rotation invariance II)**.** *Let $a, b, c, d \in \mathbb{R}^3$ and let the operators p_i be as in Lemma 4.2.12. Suppose that $N = Z_A + Z_B$ and let A be a bounded operator on $L^2(\mathbb{R}^{3N}) = L^2(\mathbb{R}^{3Z_A}) \otimes L^2(\mathbb{R}^{3Z_B})$ which allows for a decomposition of the form*

$$A = C \int_0^\infty du\, A_1(u) \otimes A_2(u),$$

(or into linear combination of terms of this type), where the $A_1(u)$ and $A_2(u)$ are bounded, rotationally invariant operators on $L^2(\mathbb{R}^{3Z_A})$ and $L^2(\mathbb{R}^{3Z_B})$, respectively, and convergence of the integral holds at least in the weak operator topology. Suppose further that Ψ_A^0 and Ψ_B^0 are rotation invariant in the sense of Lemma 4.2.12. Then for any $i_A, j_A \in \{1 \ldots, Z_A\}$ and any $i_B, j_B \in \{1 \ldots, Z_B\}$,

i. If $x_{i_A,j_A}\Psi_A \in L^2(\mathbb{R}^{3Z_A})$, $x_{i_B,j_B}\Psi_B \in L^2(\mathbb{R}^{3Z_B})$, *then*

$$\begin{aligned}&\langle (x_{i_A}\cdot a)\Psi_A \otimes (x_{i_B}\cdot b)\Psi_B|A|(x_{j_A}\cdot c)\Psi_A \otimes (x_{j_B}\cdot d)\Psi_B\rangle\\ =&\frac{1}{9}(a\cdot c)(b\cdot d)\sum_{\alpha,\beta=1}^{3}\langle x_{i_A}^{\alpha}\Psi_A \otimes x_{i_B}^{\beta}\Psi_B|A|x_{j_A}^{\alpha}\Psi_A \otimes x_{j_B}^{\beta}\Psi_B\rangle.\end{aligned}$$

ii. If $\Psi_{A,B} \in H^1(\mathbb{R}^{3Z_{A,B}})$ *and* $x_{j_A}\Psi_A \in L^2(\mathbb{R}^{3Z_A})$, $x_{j_B}\Psi_B \in L^2(\mathbb{R}^{3Z_B})$, *then*

$$\begin{aligned}&\langle (p_{i_A}\cdot a)\Psi_A \otimes (p_{i_B}\cdot b)\Psi_B|A|(x_{j_A}\cdot c)\Psi_A \otimes (x_{j_B}\cdot d)\Psi_B\rangle\\ =&\frac{1}{9}(a\cdot c)(b\cdot d)\sum_{\alpha,\beta=1}^{3}\langle p_{i_A}^{\alpha}\Psi_A \otimes p_{i_B}^{\beta}\Psi_B|A|x_{j_A}^{\alpha}\Psi_A \otimes x_{j_B}^{\beta}\Psi_B\rangle.\end{aligned}$$

Proof. i) Suppose that A is given in the above integral representation, where we assume $C = 1$ without loss of generality. We calculate

$$\begin{aligned}&\langle (x_{i_A}\cdot a)\Psi_A \otimes (x_{i_B}\cdot b)\Psi_B|A|(x_{j_A}\cdot c)\Psi_A \otimes (x_{j_B}\cdot d)\Psi_B\rangle\\ =&\int_0^\infty du\langle (x_{i_A}\cdot a)\Psi_A|A_1(u)|(x_{j_A}\cdot c)\Psi_A\rangle\langle (x_{i_B}\cdot b)\Psi_B|A_2(u)|(x_{j_B}\cdot d)\Psi_B\rangle\\ =&\frac{1}{3}\frac{1}{3}(a\cdot c)(b\cdot d)\int_0^\infty du\langle x_{i_A}\Psi_A|A_1(u)|x_{j_A}\Psi_A\rangle\langle x_{i_B}\Psi_B|A_2(u)|x_{j_B}\Psi_B\rangle\\ =&\frac{1}{9}(a\cdot c)(b\cdot d)\int_0^\infty du\sum_{\alpha,\beta=1}^{3}\langle x_{i_A}^{\alpha}\Psi_A|A_1(u)|x_{j_A}^{\alpha}\Psi_A\rangle\langle x_{i_B}^{\beta}\Psi_B|A_2(u)|x_{j_B}^{\beta}\Psi_B\rangle\\ =&\frac{1}{9}(a\cdot c)(b\cdot d)\sum_{\alpha,\beta=1}^{3}\int_0^\infty du\langle x_{i_A}^{\alpha}\Psi_A \otimes x_{i_B}^{\beta}\Psi_B|A_1(u)\otimes A_2(u)|x_{j_A}^{\alpha}\Psi_A \otimes x_{j_B}^{\beta}\Psi_B\rangle\\ =&\frac{1}{9}(a\cdot c)(b\cdot d)\sum_{\alpha,\beta=1}^{3}\langle x_{i_A}^{\alpha}\Psi_A \otimes x_{i_B}^{\beta}\Psi_B|A|x_{j_A}^{\alpha}\Psi_A \otimes x_{j_B}^{\beta}\Psi_B\rangle,\end{aligned}$$

where the second equality follows from Lemma 4.2.12. If A is given as a linear combination of terms of the above type, the argument still applies, since the initial and final matrix elements are linear in A.
ii), iii) follow immediately, since under rotations, the momentum operators transform exactly like x_i, see the proof of Lemma 4.2.12. □

By a simple argument involving the diagonalization of symmetric matrices, we can generalize the preceding result even further:

Lemma 4.2.17. *Assume the hypotheses of Lemma 4.2.16. In addition, let* A, B *be real, symmetric* 3×3 *matrices. Then for any* $i_A, j_A \in \{1\ldots, Z_A\}$ *and any* $i_B, j_B \in \{1\ldots, Z_B\}$,

i. If $x_{i_A,j_A}\Psi_A \in L^2(\mathbb{R}^{3Z_A})$, $x_{i_B,j_B}\Psi_B \in L^2(\mathbb{R}^{3Z_B})$, *then*

$$\begin{aligned}&\left\langle \left(\mathbf{x}_{i_A}\Psi_A^0 \cdot A(\mathbf{x}_{i_B}\Psi_B^0)\right)|T|\left(\mathbf{x}_{j_A}\Psi_A^0 \cdot B(\mathbf{x}_{j_B}\Psi_B^0)\right)\right\rangle\\ =&\frac{1}{9}\mathrm{tr}[AB]\sum_{\alpha,\beta=1}^{3}\left\langle \mathbf{x}_{i_A}^{\alpha}\Psi_A^0 \otimes \mathbf{x}_{i_B}^{\beta}\Psi_B^0|T|\mathbf{x}_{j_A}^{\alpha}\Psi_A^0 \otimes \mathbf{x}_{j_B}^{\beta}\Psi_B^0\right\rangle.\end{aligned}$$

ii. If $\Psi_{A,B} \in H^1(\mathbb{R}^{3Z_{A,B}})$ *and* $x_{j_A}\Psi_A \in L^2(\mathbb{R}^{3Z_A})$, $x_{j_B}\Psi_B \in L^2(\mathbb{R}^{3Z_B})$, *then*

$$\begin{aligned}&\Big\langle \big(\mathbf{p}_{i_A}\Psi_A^0 \cdot A(\mathbf{p}_{i_B}\Psi_B^0)\big)\,|T|\,\big(\mathbf{p}_{j_A}\Psi_A^0 \cdot B(\mathbf{p}_{j_B}\Psi_B^0)\big)\Big\rangle\\ =&\frac{1}{9}\mathrm{tr}[AB]\sum_{\alpha,\beta=1}^{3}\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0 \otimes \mathbf{p}_{i_B}^{\beta}\Psi_B^0|T|\mathbf{p}_{j_A}^{\alpha}\Psi_A^0 \otimes \mathbf{p}_{j_B}^{\beta}\Psi_B^0\Big\rangle.\end{aligned}$$

Proof. Since A is symmetric, we can find an orthogonal matrix O_A which transforms A into the diagonal matrix $D = \mathrm{diag}\,(\lambda_1, \lambda_2, \lambda_2)$, the λ_i being the eigenvalues of A. We conclude

$$\begin{aligned}&\big(\mathbf{x}_{i_A}\Psi_A^0 \cdot A(\mathbf{x}_{i_B}\Psi_B^0)\big) = \big(\mathbf{x}_{i_A}\Psi_A^0 \cdot (O_A^T D O_A)(\mathbf{x}_{i_B}\Psi_B^0)\big)\\ =&\big(O_A(\mathbf{x}_{i_A}\Psi_A^0) \cdot (DO_A)(\mathbf{x}_{i_B}\Psi_B^0)\big) = \sum_{i=1}^{3}\lambda_i (O_A(\mathbf{x}_{i_A}\Psi_A^0)\cdot e_i)\,(e_i \cdot O_A(\mathbf{x}_{i_B}\Psi_B^0))\\ =&\sum_{i=1}^{3}\lambda_i(\mathbf{x}_{i_A}\Psi_A^0 \cdot O_A^T e_i)\,(O_A^T e_i \cdot \mathbf{x}_{i_B}\Psi_B^0).\end{aligned}$$

Here the e_i constitute the canonical orthonormal basis of $\mathbb{R}^3$. The corresponding transformation for B yields

$$(\mathbf{x}_{j_A}\Psi_A^0 \cdot B(\mathbf{x}_{j_B}\Psi_B^0)) = \sum_{i=1}^{3}\tilde{\lambda}_i(\mathbf{x}_{j_A}\Psi_A^0 \cdot O_B^T e_i)\,(O_B^T e_i \cdot \mathbf{x}_{j_B}\Psi_B^0),$$

so that we obtain

$$\begin{aligned}&\Big\langle \big(\mathbf{x}_{i_A}\Psi_A^0 \cdot A(\mathbf{x}_{i_B}\Psi_B^0)\big)\,|T|\,\big(\mathbf{x}_{j_A}\Psi_A^0 \cdot B(\mathbf{x}_{j_B}\Psi_B^0)\big)\Big\rangle\\ =&\sum_{i,j}\lambda_i\,\tilde{\lambda}_j\Big\langle (\mathbf{x}_{i_A}\Psi_A^0 \cdot O_A^T e_i)\,(O_A^T e_i \cdot \mathbf{x}_{i_B}\Psi_B^0)|T|(\mathbf{x}_{j_A}\Psi_A^0 \cdot O_B^T e_j)\,(O_B^T e_j \cdot \mathbf{x}_{j_B}\Psi_B^0)\Big\rangle,\end{aligned}$$

to which Lemma 4.2.16 is applicable, yielding

$$\begin{aligned}&\Big\langle \big(\mathbf{x}_{i_A}\Psi_A^0 \cdot A(\mathbf{x}_{i_B}\Psi_B^0)\big)\,|T|\,\big(\mathbf{x}_{j_A}\Psi_A^0 \cdot B(\mathbf{x}_{j_B}\Psi_B^0)\big)\Big\rangle\\ =&\frac{1}{9}\sum_{i,j}\lambda_i\,\tilde{\lambda}_j(O_A^T e_i \cdot O_B^T e_j)\,(O_B^T e_j \cdot O_A^T e_i)\sum_{\alpha,\beta=1}^{3}\Big\langle \mathbf{x}_{i_A}^{\alpha}\Psi_A^0 \otimes \mathbf{x}_{i_B}^{\beta}\Psi_B^0|T|\mathbf{x}_{j_A}^{\alpha}\Psi_A^0 \otimes \mathbf{x}_{j_B}^{\beta}\Psi_B^0\Big\rangle.\end{aligned}$$

An easy calculation shows that

$$\sum_{i,j}\lambda_i\,\tilde{\lambda}_j(O_A^T e_i \cdot O_B^T e_j)\,(O_B^T e_j \cdot O_A^T e_i) = \sum_{i=1}^{3}\lambda_i\,(e_i \cdot (O_A B O_A^T)e_i),$$

and by the definition and the cyclicity of the trace we find

$$\begin{aligned}\mathrm{tr}[AB] &= \mathrm{tr}[O_A^T D O_A B] = \mathrm{tr}[DO_A B O_A^T]\\ &= \sum_{i=1}^{3}(e_i \cdot (DO_A B O_A^T)e_i) = \sum_{i=1}^{3}\lambda_i\,(e_i \cdot (O_A B O_A^T)e_i),\end{aligned}$$

finishing the proof of the first assertion. The second claim is proven completely analogously. □

4.2.6 Resolvent and operator identities

In the following lemma we collect some important identities involving the reduced resolvent T^σ restricted to some of its invariant subspaces. We will need these identities in Chapters 3 and 5, when we convert the matrix elements occurring in $V^\sigma_{VdW}(\mathbf{R})$ to integrals over photon momenta.

Note: In the statement of the following lemma, we will stick to the convention that if an operator $(H_{A,B}+\omega(\mathbf{k}))$ appears on the left of $(\overline{H_A+H_B})^{-1}$, it is understood that it is applied to the left-hand side of the inner product.

Lemma 4.2.18 (Resolvent and operator identities)**.** *Assume (A1) and (A2) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Then the following identities and relations hold:*

i.

$$\begin{aligned}&(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_{A,B}+\omega(\mathbf{k}_2))^{-1}\\ =&(H_{B,A}+\omega(\mathbf{k}_1))^{-1}(H_{A,B}+\omega(\mathbf{k}_2))^{-1}\\ &-(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_{B,A}+\omega(\mathbf{k}_1))^{-1}\end{aligned}$$

as bounded operators on $\{\Psi^0_A\}^\perp\widehat{\otimes}\{\Psi^0_B\}^\perp \subset \mathcal{H}_A\widehat{\otimes}\mathcal{H}_B$ for all $(\mathbf{k}_1,\mathbf{k}_2)\in\mathbb{R}^6$.

ii.

$$\begin{aligned}[(H_A+\omega(\mathbf{k}_1))^{-1}\otimes I_{\mathcal{H}_B}, I_{\mathcal{H}_A}\otimes(H_B+\omega(\mathbf{k}_2))^{-1}] =&0, \qquad (4.2.22)\\ [(H_{A,B}+\omega(\mathbf{k}_1))^{-1}\otimes I_{\mathcal{H}_{B,A}}, \frac{1}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}] =&0,\\ [(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}, \frac{1}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}] =&0,\\ [(\overline{H_A+H_B})^{-1}, \frac{1}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}] =&0\end{aligned}$$

as bounded operators on $\{\Psi^0_A\}^\perp\widehat{\otimes}\{\Psi^0_B\}^\perp \subset \mathcal{H}_A\otimes\mathcal{H}_B$ for all $(\mathbf{k}_1,\mathbf{k}_2)\neq(0,0)\in\mathbb{R}^6$.

$$\begin{aligned}[(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}, \overline{(H_{A,B}+\omega(\mathbf{k}_3))^{-1}\otimes I_{\mathcal{H}_{B,A}}}] =&0, \qquad (4.2.23)\\ [\overline{(H_A+\omega(\mathbf{k}_1))^{-1}\otimes I_{\mathcal{H}_B}}, \overline{I_{\mathcal{H}_A}\otimes(H_A+\omega(\mathbf{k}_2))^{-1}}] =&0 \qquad (4.2.24)\end{aligned}$$

as bounded operators on $\{\Psi^0_A\}^\perp\widehat{\otimes}\{\Psi^0_B\}^\perp \subset \mathcal{H}_A\otimes\mathcal{H}_B$ for all $\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3\in\mathbb{R}^3$. Furthermore,

$$\begin{aligned}&(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}\left(\overline{(H_{A,B}+\omega(\mathbf{k}_3))\otimes I_{\mathcal{H}_{B,A}}}\right)\\ \subset&\left(\overline{(H_{A,B}+\omega(\mathbf{k}_3))\otimes I_{\mathcal{H}_{B,A}}}\right)(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}. \qquad (4.2.25)\end{aligned}$$

iii.

$$\begin{aligned}&\Big\langle(H_A+\omega(\mathbf{k}_1))^{-1}(H_B+\omega(\mathbf{k}_2))^{-2}+(H_B+\omega(\mathbf{k}_1))^{-1}(H_A+\omega(\mathbf{k}_2))^{-2}\\ &-\Big((H_A+\omega(\mathbf{k}_1))^{-1}+(H_B+\omega(\mathbf{k}_2))^{-1}\Big)^2(\overline{H_A+H}_B+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}\Big\rangle_\psi\\ =&\Big\langle(H_A+\omega(\mathbf{k}_2))^{-2}(H_B+\omega(\mathbf{k}_1))^{-1}-(H_A+\omega(\mathbf{k}_1))^{-2}(H_B+\omega(\mathbf{k}_2))^{-1}\Big\rangle_\psi \qquad (4.2.26)\end{aligned}$$

for all $\psi \in \{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \subset \mathcal{H}_A \otimes \mathcal{H}_B$ *and all* $\mathbf{k}_1, \mathbf{k}_2 \in \mathbb{R}^3$.

iv.

$$\begin{aligned}
&\Big\langle \big((H_A + \hbar\omega(\mathbf{k}))^{-1} + (H_B + \hbar\omega(\mathbf{k}))^{-1}\big)\,(\overline{H_A + H_B})^{-1} \\
&\quad - (H_A + \hbar\omega(\mathbf{k}))^{-1}(H_B + \hbar\omega(\mathbf{k}))^{-1} \Big\rangle_\psi \\
=&2\hbar\omega(\mathbf{k}) \Big\langle (H_A + \omega(\mathbf{k}))^{-1}(H_B + \omega(\mathbf{k}))^{-1}(\overline{H_A + H_B})^{-1} \Big\rangle_\psi
\end{aligned}$$

for all $\psi \in \{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp$ *and all* $\mathbf{k} \in \mathbb{R}^3$.

v.

$$\begin{aligned}
&\Big\langle (\overline{H_A + H_B})^{-1} \\
&\Big[(H_A + \hbar\omega(\mathbf{k}_1))^{-1}(H_A + \hbar\omega(\mathbf{k}_2))^{-1} \otimes H_B \\
&+ H_A \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1}(H_B + \hbar\omega(\mathbf{k}_2))^{-1}\Big] \Big\rangle_\psi \\
=&\Big\langle (H_A + \hbar\omega(\mathbf{k}_1))^{-1}(H_A + \hbar\omega(\mathbf{k}_2))^{-1} \otimes I \Big\rangle_\psi \\
&+ \Big\langle I \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1}(H_B + \hbar\omega(\mathbf{k}_2))^{-1} \Big\rangle_\psi \\
&- \Big\langle (\overline{H_A + H_B})^{-1}\Big[(H_A + \hbar\omega(\mathbf{k}_1))^{-1} \otimes I + I \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1}\Big] \Big\rangle_\psi \\
&+ \hbar\omega(\mathbf{k}_2)\Big\langle (\overline{H_A + H_B})^{-1}\Big[(H_A + \hbar\omega(\mathbf{k}_1))^{-1}(H_A + \hbar\omega(\mathbf{k}_2))^{-1} \otimes I \\
&+ I \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1}(H_B + \hbar\omega(\mathbf{k}_2))^{-1}\Big] \Big\rangle_\psi
\end{aligned}$$

for all $\psi \in D(H_{A|\{\Psi_A^0\}^\perp}) \widehat{\otimes} D(H_{B|\{\Psi_B^0\}^\perp})$ *and any* $\mathbf{k}_1, \mathbf{k}_2 \in \mathbb{R}^3$.

Proof. i) Noting that $(H_A + \omega(\mathbf{k}_2))^{-1} \otimes (H_B + \omega(\mathbf{k}_1))^{-1}$ maps $\mathcal{H}_A \widehat{\otimes} \mathcal{H}_B$ to $D(H_A) \widehat{\otimes} D(H_B) = D(H_A + H_B)$ and $(\overline{H_A + H_B})^{-1}$ extends $(H_A + H_B)^{-1}$, we calculate

$$\begin{aligned}
&(\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1}(H_A + \omega(\mathbf{k}_2))^{-1} \otimes I_{\mathcal{H}_B} \\
=&(\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1}(H_A + \omega(\mathbf{k}_2))^{-1} \otimes \big((H_B + \omega(\mathbf{k}_1))(H_B + \omega(\mathbf{k}_1))^{-1}\big) \\
=&(\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1}\Big(H_A + H_B + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2) - (H_A + \omega(\mathbf{k}_2))\Big) \\
&\times \Big((H_A + \omega(\mathbf{k}_2))^{-1} \otimes (H_B + \omega(\mathbf{k}_1))^{-1}\Big) \\
=&\Big((H_A + \omega(\mathbf{k}_2))^{-1} \otimes (H_B + \omega(\mathbf{k}_1))^{-1}\Big) \\
&- (\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1}\Big(I_{\mathcal{H}_A} \otimes (H_B + \omega(\mathbf{k}_1))^{-1}\Big)
\end{aligned}$$

on $\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp$. By exchanging the roles of A and B, the second assertion follows.
ii) The first four identities hold trivially due to the non-interacting structure of the operator $(H_{A,B} + \omega(\mathbf{k}))^{-1} \otimes I_{\mathcal{H}_{B,A}}$ and the fact that $1/(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))$ is multiplication by a constant on $\mathcal{H}_A \otimes \mathcal{H}_B$. To prove (4.2.23), set $M := \overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp}$. As in the proof of Lemma

4.2.5, we see that M is a reducing subspace for $(H_A+\omega(\mathbf{k}_3))^{-1}\otimes I_{\mathcal{H}_B}$, on which it is closable with boundedly invertible, self-adjoint closure

$$\overline{((H_A+\omega(\mathbf{k}_3))^{-1}\otimes I_{\mathcal{H}_B})_{|M}}.$$

Equally, we have that

$$(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M}=\overline{((H_A+\omega(\mathbf{k}_1))\otimes I_{\mathcal{H}_B}+I_{\mathcal{H}_A}\otimes(H_B+\omega(\mathbf{k}_2)))_{|M}}$$

is self-adjoint and boundedly invertible on the Hilbert space M. This implies the relations

$$\begin{aligned}
&\exp\left(\mathrm{i}t(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M}\right)\\
=&\overline{\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}\right)\otimes\exp\left(\mathrm{i}t(H_B+\omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}\right)},\\
&\exp\left(\mathrm{i}t(\overline{(H_A+\omega(\mathbf{k}_3))^{-1}\otimes I_{\mathcal{H}_B})_{|M}}\right)\\
=&\overline{\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_3))_{|\{\Psi_A^0\}^\perp}\right)\otimes I_{\{\Psi_B^0\}^\perp}}
\end{aligned}$$

for the corresponding unitary groups (see [Wei76], Satz 8.35). By definition, two (unbounded) self-adjoint operators commute if and only if all their spectral projections commute. By [RS80], Thm.VIII.13, this equivalent to either their resolvents or their unitary groups commuting (in the usual sense for bounded operators). Now it is clear that

$$\begin{aligned}
&\Big[\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}\right)\otimes\exp\left(\mathrm{i}t(H_B+\omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}\right),\\
&\quad\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_3))_{|\{\Psi_A^0\}^\perp}\right)\otimes I_{\{\Psi_B^0\}^\perp}\Big]\\
=&\Big[\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}\right),\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_3))_{|\{\Psi_A^0\}^\perp}\right)\Big]\\
&\otimes\left[\exp\left(\mathrm{i}t(H_B+\omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}\right),I_{\{\Psi_B^0\}^\perp}\right]=0
\end{aligned}$$

on $\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$, the latter being a dense invariant subspace of the Hilbert space $M=\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}$ for these operators. Applying Lemma 4.2.2, we conclude

$$\begin{aligned}
&\Big[\overline{\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_1))_{|\{\Psi_A^0\}^\perp}\right)\otimes\exp\left(\mathrm{i}t(H_B+\omega(\mathbf{k}_2))_{|\{\Psi_B^0\}^\perp}\right)},\\
&\overline{\exp\left(\mathrm{i}t(H_A+\omega(\mathbf{k}_3))_{|\{\Psi_A^0\}^\perp}\right)\otimes I_{\{\Psi_B^0\}^\perp}}\Big]=0,
\end{aligned}$$

as bounded operators on M, which is equivalent to

$$[(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M},\overline{((H_A+\omega(\mathbf{k}_3))\otimes I_{\mathcal{H}_B})_{|M}}]=0,$$

and also to

$$[\left((\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M}\right)^{-1},\left(\overline{((H_A+\omega(\mathbf{k}_3))\otimes I_{\mathcal{H}_B})_{|M}}\right)^{-1}]=0$$

as bounded operators on M by the above and [RS80], Thm.VIII.13. As in the proof of Lemma 4.2.5,

$$\Big(\overline{((H_A+\omega(\mathbf{k}_3))\otimes I_{\mathcal{H}_B})_{|M}}\Big)^{-1} = \overline{(H_A+\omega(\mathbf{k}_3))^{-1}\otimes I_{\mathcal{H}_B})_{|M}},$$

and restricting to the (dense) subspace $\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$ proves (4.2.23). Identity 4.2.24 is proven completely analogous. To see (4.2.25), note that by the above, all spectral projections of $(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M}$ and $\overline{((H_A+\omega(\mathbf{k}_3))\otimes I_{\mathcal{H}_B})_{|M}}$ commute. Using the spectral representation of $((\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M})^{-1}$ and the fact that $((\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M})^{-1}$ maps M to $D(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))_{|M})$, which is contained in $D(\overline{(H_A\otimes I_{\mathcal{H}_B})_{|M}})$ (see Lemma 4.2.4), the assertion follows.
iii) Since the operator $(H_B+\omega(\mathbf{k}_1))^{-1}(H_A+\omega(\mathbf{k}_2))^{-2}$ appears on both sides of the asserted equality (note that the two factors commute on $\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$), it suffices to show that

$$\begin{aligned}&\Big\langle (H_A+\omega(\mathbf{k}_1))^{-1}(H_B+\omega(\mathbf{k}_2))^{-2}\\ &\quad-\Big((H_A+\omega(\mathbf{k}_1))^{-1}+(H_B+\omega(\mathbf{k}_2))^{-1}\Big)^2(\overline{H_A+H}_B+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}\Big\rangle_\psi\\ =&-\Big\langle (H_A+\omega(\mathbf{k}_1))^{-2}(H_B+\omega(\mathbf{k}_2))^{-1}\Big\rangle_\psi,\end{aligned}$$

which is easily seen by inserting a factor

$$(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))$$

in front of the first summand on the left-hand side and the right-hand side, noting that

$$\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2) = H_A+H_B+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)$$

on $D(H_A)\widehat{\otimes}D(H_B)$, and using the commutator relations from ii), as well as our convention about the ordering of operators stated above.
iv) The operator $(H_A+\hbar\omega(\mathbf{k}))^{-1}(H_B+\hbar\omega(\mathbf{k}))^{-1}$ maps $\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$ to $D(H_A)\widehat{\otimes}D(H_B)$, on which $\overline{H_A+H_B}=H_A+H_B$, so that we can insert an identity and obtain

$$\begin{aligned}&\Big\langle \big((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,(\overline{H_A+H_B})^{-1}\\ &\quad-(H_A+\hbar\omega(\mathbf{k}))^{-1}(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big\rangle_\psi\\ =&\Big\langle \big((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,(\overline{H_A+H_B})^{-1}\\ &\quad-(\overline{H_A+H_B})^{-1}(H_A+H_B+2\omega(\mathbf{k})-2\omega(\mathbf{k}))(H_A+\hbar\omega(\mathbf{k}))^{-1}(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big\rangle_\psi\\ =&\Big\langle \big((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,(\overline{H_A+H_B})^{-1}\\ &\quad-(\overline{H_A+H_B})^{-1}\Big((H_B+\hbar\omega(\mathbf{k}))^{-1}+(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big)\\ &\quad+2\omega(\mathbf{k})(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}))^{-1}(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big\rangle_\psi.\end{aligned}$$

Now by the commutator relation from ii) and the fact that

$$\overline{(H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I} = (H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I$$

on $\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp$, we infer

$$
\begin{aligned}
&\Big\langle \big((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,(\overline{H_A+H_B})^{-1}\\
&\quad-(\overline{H_A+H_B})^{-1}\Big((H_B+\hbar\omega(\mathbf{k}))^{-1}+(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big)\Big\rangle_\psi\\
=&\Big\langle (\overline{H_A+H_B})^{-1}\,\Big(\overline{(H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I}+\overline{I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}}\Big)\\
&\quad-(\overline{H_A+H_B})^{-1}\Big((H_B+\hbar\omega(\mathbf{k}))^{-1}+(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big)\Big\rangle_\psi\\
=&\Big\langle (\overline{H_A+H_B})^{-1}\,\big((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I+I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\\
&\quad-(\overline{H_A+H_B})^{-1}\Big((H_B+\hbar\omega(\mathbf{k}))^{-1}+(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big)\Big\rangle_\psi\\
=&0,
\end{aligned}
$$

so that the assertion follows by noting that due to the commutator relations in ii) and our above convention, we have

$$
\begin{aligned}
&\Big\langle 2\omega(\mathbf{k})(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}))^{-1}(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big\rangle_\psi\\
=&2\omega(\mathbf{k})\Big\langle (H_A+\hbar\omega(\mathbf{k}))^{-1}(H_B+\hbar\omega(\mathbf{k}))^{-1}(\overline{H_A+H_B})^{-1}\Big\rangle_\psi.
\end{aligned}
$$

v) Since $\psi\in D(H_{A|\{\Psi_A^0\}^\perp})\widehat{\otimes}D(H_{B|\{\Psi_B^0\}^\perp})$, we can add and subtract operators H_A and H_B to obtain

$$
\begin{aligned}
&\Big\langle (\overline{H_A+H_B})^{-1}\\
&\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes H_B\\
&+H_A\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big\rangle_\psi\\
=&\Big\langle (\overline{H_A+H_B})^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(H_A+H_B)\\
&+(H_A+H_B)(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\\
&-(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}H_A\otimes I\\
&-I\otimes H_B(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big\rangle_\psi.
\end{aligned}
$$

Using the commutator relation (4.2.25), the fact that $H_A+H_B=\overline{H_A+H_B}$ on $D(H_{A|\{\Psi_A^0\}^\perp})\widehat{\otimes}D(H_{B|\{\Psi_B^0\}^\perp})$ and the commutativity of H_B and $(H_B+\hbar\omega(\mathbf{k}_1))^{-1}$ on

$D(H_B)$, we obtain

$$
\begin{aligned}
&\Big\langle (\overline{H_A+H_B})^{-1}\Big[(\overline{H_A+H_B})\Big((H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes I\Big)\\
&+(\overline{H_A+H_B})\Big(I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big)\\
&-(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}H_A\otimes I\\
&-I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}H_B(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big\rangle_\psi\\
=&\Big\langle (H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes I\\
&+I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\\
&-(\overline{H_A+H_B})^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}H_A\otimes I\\
&+I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}H_B(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big\rangle_\psi.
\end{aligned}
$$

Finally, using the relations $(H_A+\hbar\omega(\mathbf{k}_2))^{-1}H_A = I-\hbar\omega(\mathbf{k}_2)(H_A+\hbar\omega(\mathbf{k}_2))^{-1}$ and $H_B(H_B+\hbar\omega(\mathbf{k}_2))^{-1} = I-\hbar\omega(\mathbf{k}_2)(H_B+\hbar\omega(\mathbf{k}_2))^{-1}$ (which hold on $D(H_A)$ and $\mathcal{H}_B$, respectively), we arrive at the asserted expression. □

4.3 Exploiting invariance properties and cancellations with infinitely separated problems

In the next step of the proof of Theorem 3.0.6 we apply the formulas for the energy corrections derived in Section 4.1, exploit the properties of the reduced resolvent established in Section 4.2, and collect simplifications that arise from cancellations of terms occurring in the energy corrections of both the compound and the infinitely separated systems. Recall the definitions

$$
\begin{aligned}
T^\sigma =&((H_0^\sigma - E_0)_{|\{\Psi_0\}^\perp})^{-1},\quad \Psi_0=\Psi_A^0\otimes\Psi_B^0\otimes\Omega,\\
T_A^\sigma =&((\overline{H_A+H_{f\geq\sigma}}-E_A^0))_{|\{\Psi_A^0\otimes\Omega\}^\perp})^{-1},\\
T_B^\sigma =&((\overline{H_B+H_{f\geq\sigma}}-E_B^0))_{|\{\Psi_B^0\otimes\Omega\}^\perp})^{-1}
\end{aligned}
$$

of the reduced resolvents of H_0, $\overline{H_A+H_{f\geq\sigma}}$ and $\overline{H_B+H_{f\geq\sigma}}$ from Proposition 2.6.5. The above-mentioned simplifications are collected in the following result.

Lemma 4.3.1. *Assume the hypotheses of Theorem 3.0.6. Then*

$$
\begin{aligned}
&V_1^\sigma(\Lambda,\mathbf{R}) = V_3^\sigma(\Lambda,\mathbf{R}) = 0,\\
&V_2^\sigma(\Lambda,\mathbf{R}) = \langle\Psi_0|Q_R|\Psi_0\rangle, && (4.3.1)\\
&V_4^\sigma(\Lambda,\mathbf{R})\\
=&-\big\langle Q_R\Psi_0|T^\sigma|Q_R\Psi_0\big\rangle && (4.3.2)\\
&-\Big\{2\,\mathrm{Re}\Big[\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle + \big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle\Big]\\
&\quad+\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle + \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle\Big\} && (4.3.3)\\
&-2\mathrm{Re}\Big[\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle + \big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle\\
&\quad+\big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle + \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle\Big] && (4.3.4)\\
&-2\mathrm{Re}\Big[\big\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\big\rangle\Big] && (4.3.5)\\
&+2\mathrm{Re}\Big[\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_{\sigma,B}\Psi_0\big\rangle + \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_{\sigma,A}\Psi_0\big\rangle\\
&\quad+\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma|H''_{\sigma,A}\Psi_0\big\rangle + \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma|H''_{\sigma,B}\Psi_0\big\rangle\Big] && (4.3.6)\\
&+\Big\{2\,\mathrm{Re}\Big[\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle\Big] + \big\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle\\
&\quad+\big\langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle\Big\} && (4.3.7)\\
&+2\mathrm{Re}\Big[\big\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|Q_R\Psi_0\big\rangle\Big] + \big\langle H'_\sigma\Psi_0|T^\sigma Q_R T^\sigma|H'_\sigma\Psi_0\big\rangle && (4.3.8)\\
&+\Big\{\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle\,\|T_B^\sigma\,H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\\
&\quad+\big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\,\|T_A^\sigma H'_\sigma(\Psi_A^0\otimes\Omega)\|^2\\
&\quad-\big\langle\Psi_0|Q_R|\Psi_0\big\rangle\Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2+\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big)\\
&\quad-\big\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\big\rangle\,\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\\
&\quad-\big\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\big\rangle\,\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2\Big\} && (4.3.9)
\end{aligned}
$$

Proof. We apply the formulae from Section 4.1 to $V_i^\sigma(\Lambda,\mathbf{R})$, $i=1,2,3,4$. As concerns the first- and third-order contributions, (4.1.1) and (4.1.3) implies

$$V_1^\sigma(\Lambda,\mathbf{R}) = V_3^\sigma(\Lambda,\mathbf{R}) = 0.$$

For the second-order term, (4.1.2) yields

$$
\begin{aligned}
V_2^\sigma(\Lambda,\mathbf{R}) =&E_2^\sigma(R) - (E_{2,A}^\sigma + E_{2,B}^\sigma)\\
=&-\big\langle H'_\sigma\Psi_0|T^\sigma|H'_\sigma\Psi_0\big\rangle + \big\langle\Psi_0|H''_\sigma|\Psi_0\big\rangle\\
&-\Big[-\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle + \big\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\big\rangle\\
&\quad-\big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle + \big\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\big\rangle\Big].
\end{aligned}
$$

Noting that

$$H'_\sigma = H'_{\sigma,A} + H'_{\sigma,B}, \quad H''_\sigma = H''_{\sigma,A} + H''_{\sigma,B} + Q_R,$$

recalling that $\|\Psi_A^0\| = \|\Psi_B^0\| = 1$ and using the fact that T^σ leaves the spaces $\{\Psi_A^0\}\widehat{\otimes}(\{\Psi_B^0\}\widehat{\otimes}\{\Omega\})^\perp$ and $\{\Psi_B^0\}\widehat{\otimes}(\{\Psi_A^0\}\widehat{\otimes}\{\Omega\})^\perp$ invariant (Lemma 4.2.5), we find

$$\begin{aligned}
&- \big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle + \big\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\big\rangle\\
&- \big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle + \big\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\big\rangle\\
=&- \big\langle H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle + \big\langle\Psi_0|H''_{\sigma,A}|\Psi_0\big\rangle - \big\langle H'_{\sigma,B}\Psi_0|T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle + \big\langle\Psi_0|H''_{\sigma,B}|\Psi_0\big\rangle,
\end{aligned}$$

which has the consequence that most terms in $V_2^\sigma(\Lambda,\mathbf{R})$ cancel and that we are left with

$$V_2^\sigma(\Lambda,\mathbf{R}) = - \big\langle H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle - \big\langle H'_{\sigma,B}\Psi_0|T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle + \big\langle\Psi_0|Q_R|\Psi_0\big\rangle.$$

By Lemma A.8.2, $\big\langle H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle = \big\langle H'_{\sigma,B}\Psi_0|T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle = 0$, so that

$$V_2^\sigma(\Lambda,\mathbf{R}) = \big\langle\Psi_0|Q_R|\Psi_0\big\rangle = (4.3.1). \tag{4.3.10}$$

This remaining term, which is independent of the infrafred regularization parameter σ, will turn out to vanish up to arbitrary order in $1/R$ by the exponential localization of Ψ_A^0 and Ψ_B^0 and the rotational invariance of the corresponding one-particle densities, see Section 5.2. As regards the fourth-order term $V_4^\sigma(\mathbf{R})$, we use (4.1.4) to find

$$\begin{aligned}
V_4^\sigma(\Lambda,\mathbf{R}) =& E_4^\sigma(\mathbf{R}) - (E_{4,A}^\sigma + E_{4,B}^\sigma)\\
=&\Big\{-\big\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma H'_\sigma T^\sigma|H'_\sigma\Psi_0\big\rangle\\
&\quad + \big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma H'_{\sigma,A}T_A^\sigma H'_{\sigma,A}T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle\\
&\quad + \big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma H'_{\sigma,B}T_B^\sigma H'_{\sigma,B}T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\Big\} &(4.3.11)\\
&+\Big\{-E_2^\sigma(\mathbf{R})\|T^\sigma H'_\sigma\Psi_0\|^2\\
&\quad + E_{2,A}^\sigma\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 + E_{2,B}^\sigma\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big]\Big\} &(4.3.12)\\
&+\Big\{-\big\langle H''_\sigma\Psi_0|T^\sigma|H''_\sigma\Psi_0\big\rangle + \big\langle H''_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H''_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle\\
&\quad + \big\langle H''_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H''_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\Big\} &(4.3.13)\\
&+2\mathrm{Re}\Bigg[\big\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_\sigma\Psi_0\big\rangle - \Big[\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma H'_{\sigma,A}T_A^\sigma|H''_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle\\
&\qquad + \big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma H'_{\sigma,B}T_B^\sigma|H''_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\Big]\Bigg] &(4.3.14)\\
&+\Big\{\big\langle H'_\sigma\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_\sigma\Psi_0\big\rangle - \Big[\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma H''_{\sigma,A}T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle\\
&\quad + \big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma H''_{\sigma,B}T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\Big]\Big\}. &(4.3.15)
\end{aligned}$$

Again using $H'_\sigma = H'_{\sigma,A} + H'_{\sigma,B}$, $H''_\sigma = H''_{\sigma,A} + H''_{\sigma,B} + Q_R$, $\|\Psi_A^0\| = \|\Psi_B^0\| = 1$ and the invariance of $\{\Psi_A^0\}\widehat{\otimes}(\{\Psi_B^0\}\widehat{\otimes}\{\Omega\})^\perp$ and $\{\Psi_B^0\}\widehat{\otimes}(\{\Psi_A^0\}\widehat{\otimes}\{\Omega\})^\perp$ under T^σ, we find

$$\begin{aligned}
(4.3.11) =& -\big\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma H'_\sigma T^\sigma|H'_\sigma\Psi_0\big\rangle \\
&- \Big[-\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma H'_{\sigma,A}T_A^\sigma H'_{\sigma,A}T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle \\
&\quad - \big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma H'_{\sigma,B}T_B^\sigma H'_{\sigma,B}T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\Big] \\
=& -\big\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma H'_\sigma T^\sigma|H'_\sigma\Psi_0\big\rangle \\
&- \Big[-\big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle \\
&\quad - \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle\Big] \\
=&(4.3.3) + (4.3.4) \\
&- \big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle \\
&- \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle \\
&+ \big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle \\
&+ \big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle \\
=&(4.3.3) + (4.3.4).
\end{aligned}$$

Similarly,

$$\begin{aligned}
(4.3.13) =& -\big\langle H''_\sigma\Psi_0|T^\sigma|H''_\sigma\Psi_0\big\rangle \\
&- \Big[-\big\langle H''_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H''_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle - \big\langle H''_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H''_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\Big] \\
=& -\big\langle H''_\sigma\Psi_0|T^\sigma|H''_\sigma\Psi_0\big\rangle - \Big[-\big\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,A}\Psi_0\big\rangle - \big\langle H''_{\sigma,B}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\big\rangle\Big] \\
=& -\big\langle Q_R\Psi_0|T^\sigma|Q_R\Psi_0\big\rangle - 2\mathrm{Re}\big[\big\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\big\rangle\big] \\
&- 2\mathrm{Re}\big[\big\langle Q_R\Psi_0|T^\sigma|(H''_{\sigma,A}+H''_{\sigma,B})\Psi_0\big\rangle\big].
\end{aligned}$$

Note that $Q_R\Psi_0 \in (\mathcal{H}_A\otimes\mathcal{H}_B)\otimes\{\Omega\}$ and $(H''_{\sigma,A}+H''_{\sigma,B})\Psi_0 \in \{\Psi_0\}\oplus(\mathcal{H}_A\otimes\mathcal{H}_B)\otimes\mathcal{F}^{(2)}$. Since T^σ acts trivially on $\{\Psi_0\}$ and since different Fock space sectors are mutually orthogonal, we can use the invariance of Fock space levels under T^σ (Lemma 4.2.5) to conclude $2\mathrm{Re}[\big\langle Q_R\Psi_0|T^\sigma|(H''_{\sigma,A}+H''_{\sigma,B})\Psi_0\big\rangle] = 0$, which leads to

$$(4.3.13) = -\big\langle Q_R\Psi_0|T^\sigma|Q_R\Psi_0\big\rangle - 2\mathrm{Re}[\big\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\big\rangle] = (4.3.5) + (4.3.2).$$

Using the same arguments, we find

$$
\begin{aligned}
&(4.3.14)+(4.3.15)\\
=&2\mathrm{Re}\Bigg[\left\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_\sigma\Psi_0\right\rangle-\Big[\left\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma H'_{\sigma,A}T_A^\sigma|H''_{\sigma,A}(\Psi_A^0\otimes\Omega)\right\rangle\\
&\qquad+\left\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma H'_{\sigma,B}T_B^\sigma|H''_{\sigma,B}(\Psi_B^0\otimes\Omega)\right\rangle\Big]\Bigg]\\
&+\left\langle H'_\sigma\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_\sigma\Psi_0\right\rangle-\Big[\left\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma H''_{\sigma,A}T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\right\rangle\\
&\qquad+\left\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma H''_{\sigma,B}T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\right\rangle\Big]\\
=&2\mathrm{Re}\Bigg[\left\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_\sigma\Psi_0\right\rangle-\Big[\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma|H''_{\sigma,A}\Psi_0\right\rangle\\
&\qquad+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma|H''_{\sigma,B}\Psi_0\right\rangle\Big]\Bigg]\\
&+\left\langle H'_\sigma\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_\sigma\Psi_0\right\rangle-\Big[\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,A}\Psi_0\right\rangle\\
&\qquad+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\right\rangle\Big]\\
=&2\mathrm{Re}\Bigg[\left\langle (H'_{\sigma,A}+H'_{\sigma,B})\Psi_0|T^\sigma(H'_{\sigma,A}+H'_{\sigma,B})T^\sigma|(H''_{\sigma,A}+H''_{\sigma,B})\Psi_0\right\rangle\\
&\qquad-\Big[\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma|H''_{\sigma,A}\Psi_0\right\rangle+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma|H''_{\sigma,B}\Psi_0\right\rangle\Big]\Bigg]\\
&+\left\langle (H'_{\sigma,A}+H'_{\sigma,B})\Psi_0|T^\sigma(H''_{\sigma,A}+H''_{\sigma,B})T^\sigma|(H'_{\sigma,A}+H'_{\sigma,B})\Psi_0\right\rangle\\
&-\Big[\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,A}\Psi_0\right\rangle+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\right\rangle\Big]\\
&+2\mathrm{Re}\left[\left\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|Q_R\Psi_0\right\rangle\right]+\left\langle H'_\sigma\Psi_0|T^\sigma Q_R T^\sigma|H'_\sigma\Psi_0\right\rangle\\
=&2\mathrm{Re}\Bigg[\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_{\sigma,B}\Psi_0\right\rangle+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_{\sigma,A}\Psi_0\right\rangle\\
&\qquad+\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma|H''_{\sigma,A}\Psi_0\right\rangle+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma|H''_{\sigma,B}\Psi_0\right\rangle\Bigg]\\
&+2\,\mathrm{Re}\left[\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_{\sigma,B}\Psi_0\right\rangle\right]+\left\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\right\rangle\\
&+\left\langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\right\rangle\\
&+2\mathrm{Re}\left[\left\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|Q_R\Psi_0\right\rangle\right]+\left\langle H'_\sigma\Psi_0|T^\sigma Q_R T^\sigma|H'_\sigma\Psi_0\right\rangle\\
=&(4.3.6)+(4.3.7)+(4.3.8),
\end{aligned}
$$

and

$$
\begin{aligned}
&(4.3.12)\\
&= -E_2^\sigma(\mathbf{R})\|T^\sigma H'_\sigma\Psi_0\|^2\\
&\quad + E_{2,A}^\sigma\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 - E_{2,B}^\sigma\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\\
&= \Big[\big\langle H'_\sigma\Psi_0|T^\sigma|H'_\sigma\Psi_0\big\rangle - \big\langle\Psi_0|H''_\sigma|\Psi_0\big\rangle\Big]\|T^\sigma H'_\sigma\Psi_0\|^2 \qquad (4.3.16)\\
&\quad + \Big[-\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle + \big\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\big\rangle\Big]\\
&\qquad \times\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2\\
&\quad + \Big[-\big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle + \big\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\big\rangle\Big]\\
&\qquad \times\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2.
\end{aligned}
$$

By Lemma A.8.2, the cross terms in $\|T^\sigma H'_\sigma\Psi_0\|^2$ and $\big\langle H'_\sigma\Psi_0|T^\sigma|H'_\sigma\Psi_0\big\rangle$ vanish, yielding

$$
\begin{aligned}
&(4.3.16)\\
&= \Big[\big\langle H'_\sigma\Psi_0|T^\sigma|H'_\sigma\Psi_0\big\rangle - \big\langle\Psi_0|H''_\sigma|\Psi_0\big\rangle\Big]\|T^\sigma H'_\sigma\Psi_0\|^2\\
&= \Big[\big\langle H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,A}\Psi_0\big\rangle + \big\langle H'_{\sigma,B}\Psi_0|T^\sigma|H'_{\sigma,B}\Psi_0\big\rangle - \big\langle\Psi_0|H''_{\sigma,A}+H''_{\sigma,B}+Q_R|\Psi_0\big\rangle\Big]\\
&\qquad \times\Big[\|T^\sigma H'_{\sigma,A}\Psi_0\|^2 + \|T^\sigma H'_{\sigma,B}\Psi_0\|^2\Big]\\
&= \Big[\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle + \big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^{\sigma,B}|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\\
&\quad - \big\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\big\rangle - \big\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\big\rangle - \big\langle\Psi_0|Q_R|\Psi_0\big\rangle\Big]\\
&\quad \times\Big[\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big],
\end{aligned}
$$

where the last identity follows from the invariance properties of the reduced resolvent T^σ (see Lemma 4.2.5). Thus most contributions cancel, and we are left with

$$
\begin{aligned}
(4.3.12) = &-\big\langle\Psi_0|Q_R|\Psi_0\big\rangle\Big[\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big]\\
&+\big\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\big\rangle\,\|T_B^\sigma\,H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\\
&+\big\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\big\rangle\,\|T_A^\sigma H'_\sigma(\Psi_A^0\otimes\Omega)\|^2\\
&-\big\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\big\rangle\,\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\\
&-\big\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\big\rangle\,\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2\\
&=(4.3.9),
\end{aligned}
$$

which finishes the proof. □

4.4 Converting matrix elements into integrals over photon momenta

The remainder of the proof of Theorem 3.0.6 consists of analyzing the terms (4.3.3) through (4.3.9) one by one, which is done in the following series of lemmas. The assertions of these lemmas constitute the results of the process of converting the $V_i^\sigma(\Lambda, \mathbf{R})$ into integrals over photon momenta which contain effective atomic quantities like the dynamic polarizabilities $\alpha_E^{A,B}(\mathbf{k})$. As will be discussed below, we will also encounter some important cancellations in the course of the calculations, see (4.4.12), (4.4.13) and Lemma 4.4.5 below.
The proofs of Lemmas 4.4.1 through 4.4.5 will be given in the remaining sections of this chapter. The method employed is common to all of them and will be outlined in Section 4.4.1.
Define

$$S_{A,B} := \left\langle \mathbf{v}_{A,B}\psi^0_{A,B} | H_{A,B} | \mathbf{v}_{A,B}\psi^0_{A,B} \right\rangle \tag{4.4.1}$$

and note that $S_{A,B}$ are well-defined since $\mathbf{x}_i\psi^0_{A,B} \in H^2(\mathbb{R}^{3Z_{A,B}})$ by the remarks in section 2.5.

Lemma 4.4.1. *Assume the hypotheses of Theorem 3.0.6 Then*

$$(4.3.4) = 0, \tag{4.4.2}$$

and

$$\begin{aligned}
&(4.3.3)\\
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 |C(\mathbf{k}_2)|^2 (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times \Bigg[4\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\left(\sum_{\alpha,\beta=1}^{3} \left\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | (\overline{H_A + H_B})^{-1} | \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \right\rangle\right)\\
&\qquad + (\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{2\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)} - 6\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\right)\\
&\qquad + \left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2) + \alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)} - \hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\right)\\
&\qquad + \hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2) + 2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2 T_5(\mathbf{k}_1,\mathbf{k}_2)\\
&\qquad - 8\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_5(\mathbf{k}_1,\mathbf{k}_1) + \hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2 T_6(\mathbf{k}_1,\mathbf{k}_2)\\
&\qquad + 2\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg(\sum_{\alpha,\beta=1}^{3} \langle (H_A + \hbar\omega(\mathbf{k}_1))^{-1}(H_A + \hbar\omega(\mathbf{k}_2))^{-1}\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta} S_B\\
&\qquad\qquad + S_A\langle (H_B + \hbar\omega(\mathbf{k}_1))^{-1}(H_B + \hbar\omega(\mathbf{k}_2))^{-1}\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\Bigg) \qquad (4.4.3)\\
&\qquad + \frac{4S_AS_B}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))} \qquad (4.4.4)\\
&\qquad + \left(S_A\alpha_E^B(\mathbf{k}_1) + \alpha_E^A(\mathbf{k}_1)S_B\right)\left(-4\hbar\omega(\mathbf{k}_1) + \hbar\frac{4\omega(\mathbf{k}_1)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg] \qquad (4.4.5)
\end{aligned}$$

$$
\begin{aligned}
&-\frac{1}{9}\frac{1}{(m_e)^4}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2 \\
&\quad\Big\langle \mathbf{p}^\alpha_{i_A}\Psi^0_A\otimes\mathbf{p}^\beta_{j_B}\Psi^0_B | 8(H_A+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_B+\omega(\mathbf{k}_2))^{-1} \\
&\qquad + 4(H_A+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_A+\omega(\mathbf{k}_1))^{-1} \\
&\qquad + 4(H_B+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_B+\omega(\mathbf{k}_1))^{-1}|\mathbf{p}^\alpha_{k_A}\Psi^0_A\otimes\mathbf{p}^\beta_{l_B}\Psi^0_B\Big\rangle.
\end{aligned}
\tag{4.4.6}
$$

Lemma 4.4.2. *Assume the hypotheses of Theorem 3.0.6. Then*

$$
\begin{aligned}
&(4.3.5) \\
&= -\frac{4}{9\hbar^5}S_A S_B\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2).
\end{aligned}
\tag{4.4.7}
$$

Lemma 4.4.3. *Assume the hypotheses of Theorem 3.0.6. Then*

$$
\begin{aligned}
&(4.3.6)+(4.3.7) \\
&=\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\,(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}} \\
&\quad\times\Bigg[\frac{8}{9\hbar^5(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}S_A S_B && (4.4.8)\\
&\qquad+\frac{2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}{9\hbar^2}\Bigg(\Big\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle S_B \\
&\qquad\qquad+S_A\Big\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle\Bigg) && (4.4.9)\\
&\qquad+\frac{4}{9\hbar^3}(S_A\alpha^B_E(\mathbf{k}_1)+\alpha^A_E(\mathbf{k}_1)S_B)\left(\frac{\omega(\mathbf{k}_1)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-\omega(\mathbf{k}_1)\right)\Bigg] && (4.4.10)\\
&\quad+\frac{1}{2m_e c^2}\Big(\sum_{\alpha=1}^{3}\|\,(\mathbf{G}^{\mathbf{x}_\mathbf{k}}_\sigma)_\alpha\,\|^2_W\Big)\Big(Z_A\|TH'_B\Psi_0\|^2+Z_B\|TH'_A\Psi_0\|^2\Big). && (4.4.11)
\end{aligned}
$$

Lemma 4.4.4. *Assume the hypotheses of Theorem 3.0.6. Then*

$$
\begin{aligned}
(4.3.8) &= 2\mathrm{Re}\left[\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|Q_R\Psi_0\rangle\right]+\langle H'_\sigma\Psi_0|T^\sigma Q_R T^\sigma|H'_\sigma\Psi_0\rangle \\
&= M_A(\mathbf{R},\sigma)+M_B(\mathbf{R},\sigma).
\end{aligned}
$$

Cancellation of lower-homogeneity-terms. Two important cancellations occur at this point. Firstly, as we have pointed out in the introduction, homogeneity of (parts of) the integrands will be a guiding principle in grouping terms and detecting cancellations, as well as in the analysis of their large R-asymptotics. In fact, all of the terms we encounter have the structure of Fourier integrals, and it will turn out that the 'partial' homogeneity is crucial in determining the decay as $R\to\infty$. For instance, consider the terms (4.4.4),

(4.4.7) and (4.4.8). The function

$$\frac{1}{\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)$$

occurring in the integral (recall the definition of $C(\mathbf{k})$ from (3.0.3)) is homogeneous of degree -3, which hints at a $1/R^3$-decay of the integral (the integrals are taken over $\mathbb{R}^6$). However, as is easily seen by inspection,

$$(4.4.4)+(4.4.7)+(4.4.8)=0. \tag{4.4.12}$$

Similarly, the terms (4.4.3), (4.4.5), (4.4.9) and (4.4.10), which contain integrands of homogeneity 0 and -1, respectively, cancel:

$$(4.4.3)+(4.4.5)+(4.4.9)+(4.4.10)=0 \tag{4.4.13}$$

Note that these cancellations do not involve the Coulomb potential. In particular, for them to occur it does not make a difference whether or not a smeared charge distribution is used, or whether or not the Coulomb potential is multipole-expanded.

Cancellation of R-independent terms. Secondly, the terms and (4.4.6) and (4.4.11) are independent of $\mathbf{R}$, and (4.3.9) will turn out also to contain $\mathbf{R}$-independent contributions. Fortunately, the next lemma asserts that their sum reduces to a term which will later be shown to decay faster than any inverse power of R, see Chapter 5.

Lemma 4.4.5. *Assume the hypotheses of Theorem 3.0.6. Then*

$$(4.3.9)+(4.4.6)+(4.4.11)=-\langle\Psi_0|Q_R|\Psi_0\rangle\Big(\|T^\sigma_A H'_{\sigma,A}(\Psi^0_A\otimes\Omega)\|^2+\|T^\sigma_B H'_{\sigma,B}(\Psi^0_B\otimes\Omega)\|^2\Big).$$

4.4.1 Outline of the method for the proofs of Lemmas 4.4.1 through 4.4.5

Before giving the proofs of Lemmas 4.4.1 through 4.4.5, we describe the general scheme according to which they all proceed.

i. Use the structure of the interaction operators $H'_\sigma = H'_{\sigma,A}+H'_{\sigma,B}$, the invariance properties of the reduced resolvent T^σ (see Lemmas 4.2.5, 4.2.11) and orthogonality arguments to reduce the number of terms and to simplify their structure. For instance, many operators T^σ 'collapse' into the operators T^σ_A and T^σ_B.

ii. Use a fiber decomposition of T^σ_A and T^σ_B (see Lemma 4.2.7) to convert the matrix elements into integrals over the photon momenta, the integrands involving matrix elements over the electronic coordinates.

iii. Use rotation invariance properties of the ground states and the unperturbed operators, as well as the transformation behaviour of the position and momentum operators to eliminate sums over the photon polarizations involving the polarization vectors (see Lemmas 4.2.12 through 4.2.17).

iv. This procedure produces matrix elements of resolvents of the atomic Hamiltonians $((H_A+\omega(\mathbf{k}))^{-1}$, $(H_A+H_B+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)^{-1})$ etc.) on states of the form $\mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes \mathbf{p}_{j_B}^{\beta}\Psi_B^0$, which are then further transformed into matrix elements on states of the form $\mathbf{x}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{x}_{j_B}^{\beta}\Psi_B^0$ by using Proposition 2.5.1, the commutator relation from Lemma A.7.1 and a number of operator identities collected in Lemma 4.2.18. This conversion of the matrix elements is crucial in order to be able to compare the contributions to the interaction potential obtained from the quantized radiation field to those containing the (multipole-expanded) interatomic Coulomb potential, which involve the position operators $\mathbf{x}_i$ per se.

v. The resulting integrals can be further simplified by exploiting symmetry with respect to the photon momentum integration variables $\mathbf{k}_1$ and $\mathbf{k}_2$, see Lemma A.10.1 and Remark 4.4.6

4.4.2 Proof of Lemma 4.4.1

We begin by proving (4.4.2). First note that

$$\begin{aligned}(4.3.4) &= -2\mathrm{Re}\Big[\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle\\ &\qquad+\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\rangle\\ &\qquad+\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\rangle\\ &\qquad+\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle\Big]\\ &= -2\mathrm{Re}\Big[\langle H'_{\sigma,A}T^\sigma H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,A}T^\sigma H'_{\sigma,B}\Psi_0\rangle\\ &\qquad+\langle H'_{\sigma,A}T^\sigma H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,B}T^\sigma H'_{\sigma,A}\Psi_0\rangle &(4.4.14)\\ &\qquad+\langle H'_{\sigma,B}T^\sigma H'_{\sigma,B}\Psi_0|T^\sigma|H'_{\sigma,B}T^\sigma H'_{\sigma,A}\Psi_0\rangle &(4.4.15)\\ &\qquad+\langle H'_{\sigma,B}T^\sigma H'_{\sigma,B}\Psi_0|T^\sigma|H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle\Big]. &(4.4.16)\end{aligned}$$

We will show that the four terms in square brackets vanish individually. Using the definition of the perturbation operators $H'_{\sigma,A}$ and $H'_{\sigma,B}$, Lemma A.8.1 and the invariance properties of the reduced resolvent T^σ (Lemma 4.2.5), we find

$$\begin{aligned}H'_{\sigma,A}T^\sigma H'_{\sigma,A}\Psi_0 &\in \{\Psi_B^0\}\widehat{\otimes}\left(\mathcal{H}_A\otimes(\mathcal{F}_\sigma^{(0)}\oplus\mathcal{F}_\sigma^{(2)})\right),\\ H'_{\sigma,A}T^\sigma H'_{\sigma,B}\Psi_0 &\in \overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}(\mathcal{F}_\sigma^{(0)}\oplus\mathcal{F}_\sigma^{(2)})}.\end{aligned}$$

Since these subspaces are conserved by T^σ (Lemma 4.2.5) and are mutually orthogonal due to the occurrence of $\{\Psi_B^0\}$ in one of them and $\{\Psi_B^0\}^\perp$ in the other, we conclude that

$$\langle H'_{\sigma,A}T^\sigma H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,A}T^\sigma H'_{\sigma,B}\Psi_0\rangle = 0.$$

By the same argument, we find that (4.4.14) through (4.4.16) also vanish, establishing (4.4.2).

In the remainder of the proof we establish the identity involving

(4.3.3)

$$
\begin{aligned}
&= -\Big[2\,\mathrm{Re}\Big[\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,B}\Psi_0\rangle + \langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle\Big] \\
&\qquad + \langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\rangle + \langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma H'_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle\Big] \\
&=: S^{AABB} + S^{ABAB} + S^{ABBA} + S^{BAAB}. \qquad (4.4.17)
\end{aligned}
$$

In the following we will demonstrate steps ii) to v) from the procedure outlined above using the term

$$
\begin{aligned}
S^{ABBA} &= -\langle\Psi_0|H'_{\sigma,A}T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,B}T^\sigma H'_{\sigma,A}|\Psi_0\rangle \\
&= -\langle H'_{\sigma,B}T^\sigma H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,B}T^\sigma H'_{\sigma,A}|\Psi_0\rangle.
\end{aligned}
$$

By Lemma A.8.1, we have $H'_{\sigma,A}\Psi_0 \in \{\Psi_B^0\}\widehat{\otimes}\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(1)}}$ (note that $H'_{\sigma,A}$ only acts on the coordinates of atom A), which is left invariant by T^σ and on which it acts as $I_{\{\Psi_B^0\}}\otimes T_A^\sigma$ (see Lemma 4.2.5), yielding

$$
S^{ABBA} = -\langle H'_{\sigma,B}T_A^\sigma H'_{\sigma,A}\Psi_0|T^\sigma|H'_{\sigma,B}T_A^\sigma H'_{\sigma,A}|\Psi_0\rangle.
$$

The vector potential contained in $H'_{\sigma,B}$ maps the subspace $\{\Psi_B^0\}\widehat{\otimes}\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(1)}}$ to $\mathcal{H}_B\otimes\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Omega\}}$ and $\mathcal{H}_B\otimes\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(2)}}$, which are invariant subspaces for T^σ. Thus $H'_{\sigma,B}(T_A^\sigma H'_{\sigma,A}\Psi_0)$ and $T^\sigma H'_{\sigma,B}(T_A^\sigma H'_{\sigma,A}\Psi_0)$ consist of two contributions from these respective subspaces. By the mutual orthogonality of Fock space levels, the cross-terms vanish, so that only the contributions which have the structure

$$
\langle\mathcal{H}_B\otimes\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Omega\}}|\mathcal{H}_B\otimes\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Omega\}}\rangle
$$

and

$$
\langle\mathcal{H}_B\otimes\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(2)}}|\mathcal{H}_B\otimes\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(2)}}\rangle,
$$

respectively, survive. The first of these is

$$
S_1^{ABBA} := -\frac{1}{(m_ec)^4}\sum_{i_A,j_B,k_A,l_B}\Big\langle\mathbf{p}_{j_B}\cdot a(G_\sigma^{\mathbf{R}})u_{i_A}|T^\sigma|\mathbf{p}_{l_B}\cdot a(G_\sigma^{\mathbf{R}})u_{k_A}\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\{\Omega\}},
$$

where $u_{i_A} := \Psi_B^0\otimes T_A^\sigma\left[\mathbf{p}_{i_A}\cdot a^\dagger(G_\sigma^0)(\Psi_A^0\otimes\Omega)\right]$. Again using Lemma A.8.1, we find

$$
\mathbf{p}_{l_B}\cdot a(G_\sigma^{\mathbf{R}})u_{k_A} \in \overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}},
$$

so that we can use Lemma 4.2.5 (T^σ acts as $(\overline{H_A+H_B})^{-1}$ on $\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\{\Omega\}}$) to deduce

$$
S_1^{ABBA} = -\frac{1}{(m_ec)^4}\sum_{i_A,j_B,k_A,l_B}\Big\langle\mathbf{p}_{j_B}\cdot a(G_\sigma^{\mathbf{R}})u_{i_A}|(\overline{H_A+H_B})^{-1}|\mathbf{p}_{l_B}\cdot a(G_\sigma^{\mathbf{R}})u_{k_A}\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\{\Omega\}}.
$$

By the fiber decomposition of T_A^σ (Lemma 4.2.7), u_{i_A} can be represented as

$$u_{i_A}(\mathbf{k},\lambda) = \left(c\,C(\mathbf{k})\mathbf{e}(\mathbf{k},\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]\right)\Psi_B^0,$$

so that by the definition of the annihilation operator,

$$\begin{aligned}
&\mathbf{p}_{j_B}\cdot a(G_\sigma^{\mathbf{R}})u_{i_A}\\
=&\mathbf{p}_{j_B}\Psi_B^0\cdot\left(\sum_{\lambda=1,2}\int_{\Omega_\sigma} d\mathbf{k}_1\, c^2\,\overline{C(\mathbf{k}_1)}\mathbf{e}(\mathbf{k}_1,\lambda)\mathrm{e}^{i\mathbf{k}_1\cdot\mathbf{R}}\left(C(\mathbf{k}_1)\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]\right)\right)\\
=&\int_{\Omega_\sigma} d\mathbf{k}_1\, c^2\,C(\mathbf{k}_1)|^2\mathrm{e}^{i\mathbf{k}_1\cdot\mathbf{R}}\mathbf{p}_{j_B}\Psi_B^0\cdot(I-\hat{\mathbf{k}}_1\otimes\hat{\mathbf{k}}_1)(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{i_A}\Psi_A^0],
\end{aligned}$$

where we have also used Lemma A.6.1. Recall the definition of the set $\Omega_\sigma = \{\mathbf{k}\in\mathbb{R}^3|\omega(\mathbf{k})\geq\sigma\}$. Fubini's theorem yields

$$\begin{aligned}
&S_1^{ABBA}\\
=&-\frac{1}{(m_e)^4}\sum_{i_A,j_B,k_A,l_B}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\mathrm{e}^{-i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Big\langle\mathbf{p}_{j_B}\Psi_B^0\cdot(I-\hat{\mathbf{k}}_1\otimes\hat{\mathbf{k}}_1)(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]|(\overline{H_A+H_B})^{-1}|\\
&\qquad|\mathbf{p}_{l_B}\Psi_B^0\cdot(I-\hat{\mathbf{k}}_2\otimes\hat{\mathbf{k}}_2)(H_A+\hbar\omega(\mathbf{k}_2))^{-1}[\mathbf{p}_{k_A}\Psi_A^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
=&-\frac{1}{(m_e)^4}\sum_{i_A,j_B,k_A,l_B}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\mathrm{e}^{-i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Big\langle\mathbf{p}_{i_A}\Psi_A^0\cdot(I-\hat{\mathbf{k}}_1\otimes\hat{\mathbf{k}}_1)\mathbf{p}_{j_B}\Psi_B^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\\
&\qquad|\mathbf{p}_{k_A}\Psi_A^0\cdot(I-\hat{\mathbf{k}}_2\otimes\hat{\mathbf{k}}_2)\mathbf{p}_{l_B}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}.
\end{aligned}$$

Now $(I-\hat{\mathbf{k}}_1\otimes\hat{\mathbf{k}}_1)$ and $(I-\hat{\mathbf{k}}_2\otimes\hat{\mathbf{k}}_2)$ are real symmetric matrices, and since $(H_A+\hbar\omega(\mathbf{k}))^{-1}$ is rotation-invariant by Proposition 2.5.1 and $(\overline{H_A+H_B})^{-1}$ allows for an integral decomposition into tensor products of rotation-invariant operators by Proposition 4.2.15, the operator $(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}$ satisfies the assumptions of Lemma 4.2.17, so that we obtain

$$\begin{aligned}
&S_1^{ABBA}\\
=&-\frac{1}{(m_e)^4}\frac{1}{9}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\mathrm{e}^{-i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\mathrm{tr}\left[(I-\hat{\mathbf{k}}_1\otimes\hat{\mathbf{k}}_1)(I-\hat{\mathbf{k}}_2\otimes\hat{\mathbf{k}}_2)\right]\\
&\times\Big\langle\mathbf{p}_{j_B}\alpha\Psi_B^0\cdot\otimes\mathbf{p}_{i_A}^\beta\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\\
&\qquad\times(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_{l_B}^\alpha\Psi_B^0\otimes\mathbf{p}_{k_A}^\beta\Psi_A^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}.
\end{aligned}$$

Noting that $\mathrm{tr}[(I-\hat{\mathbf{k}}_1\otimes\hat{\mathbf{k}}_1)(I-\hat{\mathbf{k}}_2\otimes\hat{\mathbf{k}}_2)]=(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)$, we arrive at

$$
\begin{aligned}
&S_1^{ABBA}\\
=&-\frac{1}{(m_e)^4}\frac{1}{9}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\mathrm{e}^{-i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\\
&\Big\langle\mathbf{p}_{j_B}\alpha\Psi_B^0\cdot\otimes\mathbf{p}_{i_A}^{\beta}\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\\
&\qquad\times(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_{l_B}^{\alpha}\Psi_B^0\otimes\mathbf{p}_{k_A}^{\beta}\Psi_A^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}.
\end{aligned}
$$

Note that we are allowed to replace $\mathrm{e}^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}$ by $\mathrm{e}^{i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}$, since the remaining integrand is invariant under the change of variables $\mathbf{k}_2\to-\mathbf{k}_2$. The second contribution to the term S^{ABBA} is

$$
\begin{aligned}
S_2^{ABBA}:=-\frac{1}{(m_ec)^4}\sum_{i_A,j_B,k_A,l_B}&\Big\langle\mathbf{p}_{j_B}\cdot a^\dagger(G_\sigma^{\mathbf{R}})u_{i_A}|(\overline{H_A+H_B+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))})^{-1}|\\
&\qquad\mathbf{p}_{l_B}\cdot a^\dagger(G_\sigma^{\mathbf{R}})u_{k_A}\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}_\sigma},
\end{aligned}
$$

where we have used that $\mathbf{p}_{l_B}\cdot a^\dagger(G_\sigma^{\mathbf{R}})u_{k_A}\in\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp\widehat{\otimes}\mathcal{F}_\sigma^{(2)}}$ (Lemma A.8.1) and that T^σ acts as $(\overline{H_A+H_B+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))})^{-1}$ on this subspace by Lemma 4.2.5. Analogously to the above, $\mathbf{p}_{j_B}\cdot a^\dagger(G_\sigma^{\mathbf{R}})u_{i_A}$ has the representation

$$
\begin{aligned}
&(\mathbf{p}_{j_B}\cdot a^\dagger(G_\sigma^{\mathbf{R}})u_{i_A})(\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu)\\
=&\frac{1}{\sqrt{2}}\oplus_{\lambda,\mu=1,2}\,c^2C(\mathbf{k}_1)C(\mathbf{k}_2)\\
&\times\Big[\mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}}(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_A+\hbar\omega(\mathbf{k}_2))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]\\
&\qquad+\mathrm{e}^{-i\mathbf{k}_2\cdot\mathbf{R}}(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]\Big],
\end{aligned}
$$

where we have used the fiber decomposition of T_A^σ (Lemma 4.2.7) and the definition of the creation operators. Using the definition of the inner product on $\mathcal{F}_\sigma^{(2)}$ and the fiber decomposition of $(\overline{H_A+H_B+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))})^{-1}$ (Lemma 4.2.7), this yields

$$
\begin{aligned}
&S_2^{ABBA} \\
&= -\frac{1}{(m_e)^4}\frac{1}{2}\sum_{i_A,j_B,k_A,l_B}\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2 \\
&\quad\times\Bigg[\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_A+\hbar\omega(\mathbf{k}_2))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]| \\
&\qquad\quad |(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_A+\hbar\omega(\mathbf{k}_2))^{-1}[\mathbf{p}_{k_A}\Psi_A^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\qquad+\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]| \\
&\qquad\quad |(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{k_A}\Psi_A^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\qquad+\mathrm{e}^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_A+\hbar\omega(\mathbf{k}_2))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]| \\
&\qquad\quad |(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{k_A}\Psi_A^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\qquad+\mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]| \\
&\qquad\quad |(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_A+\hbar\omega(\mathbf{k}_2))^{-1}[\mathbf{p}_{k_A}\Psi_A^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg] \\
&= -\frac{1}{(m_e)^4}\frac{1}{2}\sum_{i_A,j_B,k_A,l_B}\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2 \\
&\quad\times\Bigg[\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{i_A}\Psi_A^0| \\
&\qquad\quad |(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{k_A}\Psi_A^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\qquad+\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{i_A}\Psi_A^0| \\
&\qquad\quad |(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{k_A}\Psi_A^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\qquad+\mathrm{e}^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{i_A}\Psi_A^0| \\
&\qquad\quad |(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{k_A}\Psi_A^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\qquad+\mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}\Big\langle(\mathbf{p}_{j_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{i_A}\Psi_A^0| \\
&\qquad\quad |(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}| \\
&\qquad\quad |(\mathbf{p}_{l_B}\Psi_B^0\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{k_A}\Psi_A^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg].
\end{aligned}
$$

Next we exploit the rotation invariance of the operator

$$(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}$$

and apply Lemma 4.2.16 (note that by Proposition 4.2.15, its assumptions are satisfied), yielding

$$\begin{aligned}
&S_2^{ABBA}\\
&=-\frac{1}{(m_e)^4}\frac{1}{2}\frac{1}{9}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\\
&\quad\times\Bigg[(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_1,\lambda))(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\Big\langle\mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0|\\
&\qquad|(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\\
&\qquad|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&\quad+(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{e}(\mathbf{k}_2,\mu))(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_1,\lambda))\Big\langle\mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0|\\
&\qquad|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\\
&\qquad|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&\quad+\mathrm{e}^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))^2\Big\langle\mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0|\\
&\qquad|(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\\
&\qquad|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&\quad+\mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))^2\Big\langle\mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0|\\
&\qquad|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\\
&\qquad|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg],
\end{aligned}$$

a form which allows us to compute the sums over λ and μ using Lemma A.6.1:

$$
\begin{aligned}
&S_2^{ABBA} \\
&= -\frac{1}{(m_e)^4}\frac{1}{2}\frac{1}{9}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2 \\
&\quad\times\Bigg[4\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0| \\
&\qquad |(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1} \qquad (4.4.18)\\
&\qquad +(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}| \qquad (4.4.19)\\
&\qquad |\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\quad +\mathrm{e}^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0| \\
&\qquad |(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}| \\
&\qquad |\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \qquad (4.4.20)\\
&\quad +\mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0| \\
&\qquad |(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}| \\
&\qquad |\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg]. \qquad (4.4.21)
\end{aligned}
$$

For the final step we note that the exchange $\mathbf{k}_1 \leftrightarrow \mathbf{k}_2$ transforms (4.4.19) into (4.4.18) and (4.4.21) into (4.4.20), so that an application of Lemma A.10.1 yields

$$
\begin{aligned}
&S_2^{ABBA} \\
&= -\frac{1}{(m_e)^4}\frac{4}{9}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2 \\
&\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}| \\
&\qquad |\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \qquad (4.4.22)
\end{aligned}
$$

$$
\begin{aligned}
&-\frac{1}{(m_e c)^4}\frac{1}{9}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2) \\
&\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{k_B}^{\beta}\Psi_B^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}| \\
&\qquad |\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}. \qquad (4.4.23)
\end{aligned}
$$

Note that we can replace $\mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}$ by $\mathrm{e}^{i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}$ in (4.4.23), since the remaining integrand is invariant under the change of variables $\mathbf{k}_1 \to -\mathbf{k}_1$. This will be done in the

following. The term (4.4.22) is independent of $\mathbf{R}$ and will be part of the term (4.4.6).

Using commutators and operator identities. Applying the preceding procedure to the contributions in S^{AABB}, S^{ABAB} and S^{BAAB} (see (4.4.17)) which are analogous to S_1^{ABBA} and S_2^{ABBA}, we obtain

$$
\begin{aligned}
&S_1^{AABB} + S_1^{ABAB} + S_1^{ABBA} + S_1^{BAAB} \\
&= -\frac{1}{9m_e^4} \sum_{i_A,j_B,k_A,l_B} \sum_{\alpha,\beta=1}^{3} \int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 |C(\mathbf{k}_2)|^2 (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad \Big\langle \mathbf{p}_{i_A}^{\alpha} \Psi_A^0 \otimes \mathbf{p}_{j_B}^{\beta} \Psi_B^0 \Big| \frac{1}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))} \Big[2(H_A + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_1))^{-1} \\
&\quad + 2(H_A + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} \Big] + (\overline{H_A + H_B})^{-1} \Big[(H_A + \hbar\omega(\mathbf{k}_1))^{-1} (H_A + \hbar\omega(\mathbf{k}_2))^{-1} \\
&\quad + (H_B + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} + 2(H_A + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} \Big] \Big| \\
&\quad |\mathbf{p}_{k_A}^{\alpha} \Psi_A^0 \otimes \mathbf{p}_{l_B}^{\beta} \Psi_B^0 \Big\rangle
\end{aligned}
$$

and

$$
\begin{aligned}
&S_2^{AABB} + S_2^{ABAB} + S_2^{ABBA} + S_2^{BAAB} \\
&= -\frac{1}{(m_e)^4} \frac{1}{9} \sum_{i_A,j_B,k_A,l_B} \sum_{\alpha,\beta=1}^{3} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 \, |C(\mathbf{k}_2)|^2 \mathrm{e}^{i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}} (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \\
&\quad \Big\langle \mathbf{p}_{i_A}^{\alpha} \Psi_A^0 \otimes \mathbf{p}_{k_B}^{\beta} \Psi_B^0 | \\
&\qquad \Big| (H_A + \hbar\omega(\mathbf{k}_1))^{-1} (\overline{H_A + H_B} + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)))^{-1} (H_A + \hbar\omega(\mathbf{k}_2))^{-1} \\
&\qquad + (H_B + \hbar\omega(\mathbf{k}_1))^{-1} (\overline{H_A + H_B} + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} \\
&\qquad + 2(H_A + \hbar\omega(\mathbf{k}_1))^{-1} (\overline{H_A + H_B} + \hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)))^{-1} (H_B + \hbar\omega(\mathbf{k}_1))^{-1} \Big| \\
&\qquad |\mathbf{p}_{k_A}^{\alpha} \Psi_A^0 \otimes \mathbf{p}_{l_B}^{\beta} \Psi_B^0 \Big\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \qquad (4.4.24) \\
&\quad -\frac{1}{9} \frac{1}{(m_e)^4} \sum_{i_A,j_B,k_A,l_B} \sum_{\alpha,\beta=1}^{3} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 |C(\mathbf{k}_2)|^2 \\
&\quad \Big\langle \mathbf{p}_{i_A}^{\alpha} \Psi_A^0 \otimes \mathbf{p}_{j_B}^{\beta} \Psi_B^0 | 8(H_A + \omega(\mathbf{k}_1))^{-1} (\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1} (H_B + \omega(\mathbf{k}_2))^{-1} \\
&\quad + 4(H_A + \omega(\mathbf{k}_1))^{-1} (\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1} (H_A + \omega(\mathbf{k}_1))^{-1} \\
&\quad + 4(H_B + \omega(\mathbf{k}_1))^{-1} (\overline{H_A + H_B} + \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))^{-1} (H_B + \omega(\mathbf{k}_1))^{-1} | \mathbf{p}_{k_A}^{\alpha} \Psi_A^0 \otimes \mathbf{p}_{l_B}^{\beta} \Psi_B^0 \Big\rangle.
\end{aligned}
$$

To further simplify (4.4.24), we employ Lemma 4.2.18 i), which allows us to calculate

$$\begin{aligned}
&(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\\
&+(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\\
&+2(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
=&(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&-(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big]\\
&+(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\\
&-(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\Big]\\
&+2(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}
\end{aligned}$$

as bounded operators on $\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$. By the commutator relation Lemma 4.2.18 ii) (4.2.22), this equals

$$\begin{aligned}
&(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\Big[2(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}-(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}\\
&\quad+(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big](H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&-(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}.
\end{aligned}$$

Using Lemma 4.2.18 ii) (4.2.23), (4.2.24), we can rewrite this as

$$\begin{aligned}
&(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\\
&+(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&-(\overline{(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I_{\mathcal{H}_B}})(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}(\overline{I_{\mathcal{H}_A}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}}).
\end{aligned}$$

Sandwiching this with two vectors $\varphi,\psi\in\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$ yields

$$\begin{aligned}
&\Big\langle\varphi|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]|\psi\Big\rangle\\
&+\Big\langle((H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I)\varphi|(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}|(I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1})\psi\Big\rangle\\
&-\Big\langle\overline{((H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I)}\varphi|(\overline{H_A+H_B}+\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)))^{-1}|(\overline{I_{\mathcal{H}_A}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}})\psi\Big\rangle\\
=&\Big\langle\varphi|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]|\psi\Big\rangle,
\end{aligned}$$

since

$$\overline{((H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}\otimes I_{\mathcal{H}_{B,A}})}=((H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}\otimes I_{\mathcal{H}_{B,A}})$$

on $\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp$. The choice $\varphi=\mathbf{p}_{i_A}^\alpha\Psi_A^0\otimes\mathbf{p}_{j_B}^\beta\Psi_B^0$, $\psi=\mathbf{p}_{k_A}^\alpha\Psi_A^0\otimes\mathbf{p}_{l_B}^\beta\Psi_B^0$ now establishes

(4.4.24)

$$
\begin{aligned}
&= -\frac{1}{9m_e^4}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{j_B}^{\beta}\Psi_B^0\Big|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&\qquad\times\Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle,
\end{aligned}
$$

and thus, upon adding all contributions,

(4.3.3)

$$
\begin{aligned}
&= S_1^{AABB}+S_1^{ABAB}+S_1^{ABBA}+S_1^{BAAB}\\
&\quad+S_2^{AABB}+S_2^{ABAB}+S_2^{ABBA}+S_2^{BAAB}\\
&= -\frac{1}{9m_e^4}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{j_B}^{\beta}\Psi_B^0\Big|\\
&\qquad\Big|\frac{1}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}\Big[2(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&\qquad+2(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big] \qquad (4.4.25)\\
&\qquad+\Big\{(\overline{H_A+H_B})^{-1}\Big[(\overline{H}_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\\
&\qquad\qquad+(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\\
&\qquad+2(\overline{H_A+H_B})^{-1}(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\} \qquad (4.4.26)\\
&\qquad+(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big| \qquad (4.4.27)\\
&\qquad\Big|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle\\
&-\frac{1}{9}\frac{1}{(m_e)^4}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\\
&\quad\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{j_B}^{\beta}\Psi_B^0\Big|\\
&\qquad\Big|8(H_A+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_B+\omega(\mathbf{k}_2))^{-1}\\
&\qquad+4(H_A+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_A+\omega(\mathbf{k}_1))^{-1}\\
&\qquad+4(H_B+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_B+\omega(\mathbf{k}_1))^{-1}\Big|\\
&\qquad\Big|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle. \qquad (4.4.28)
\end{aligned}
$$

Note that (4.4.28) equals (4.4.6), so it remains to investigate the terms (4.4.25), (4.4.26) and (4.4.27).

Converting momentum into position operators. To this end, we implement step v) from the procedure outlined above, i.e. we will convert the atomic matrix elements between states of the form $\mathbf{p}_{i_A}^{\alpha}\Psi_A^0 \otimes \mathbf{p}_{j_B}^{\beta}\Psi_B^0$ to expressions involving states of the form $\mathbf{x}_{i_A}^{\alpha}\Psi_A^0 \otimes \mathbf{x}_{j_B}^{\beta}\Psi_B^0$. This is done using the commutator relations

$$\mathbf{p}_{i_{A,B}}\Psi_{A,B}^0 = \frac{\mathrm{i}m_e}{\hbar}H_{A,B}(\mathbf{x}_{i_{A,B}}\Psi_{A,B}^0)$$

from Lemma A.7.1 (recall that $H_{A,B}$ denote $H_{A,B} - E_{A,B}^0$), and after that the operator identities from Lemma 4.2.18.
First we consider the term (4.4.25). By Proposition 2.5.1,

$$\mathbf{x}_{i_A}^{\alpha}\Psi_A^0 \otimes \mathbf{x}_{j_B}^{\beta}\Psi_B^0 \in H^2(\mathbb{R}^{3Z_A})\widehat{\otimes}H^2(\mathbb{R}^{3Z_B}),$$

so that we can apply Lemma A.7.1, the definition of $\mathbf{v}_{A,B}$ and the identity

$$H_{A,B}(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1} = I - \hbar\omega(\mathbf{k})(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1},$$

to obtain, after rearranging terms,

$$\begin{aligned}
&\sum_{i_A,j_B,k_A,l_B}\Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{j_B}^{\beta}\Psi_B^0\Big|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&\qquad + (H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes\mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle\\
&=\Big(\frac{m_e}{\hbar}\Big)^4\Big\langle(H_A\otimes H_B)(\mathbf{v}_A^{\alpha}\otimes\mathbf{v}_B^{\beta})\Big|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\\
&\qquad + (H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big|(H_A\otimes H_B)(\mathbf{v}_A^{\alpha}\otimes\mathbf{v}_B^{\beta})\Big\rangle\\
&=\Big(\frac{m_e}{\hbar}\Big)^4\Big\langle 2(H_A\otimes H_B)-\hbar\omega(\mathbf{k}_1)\left(2(I\otimes H_B)+H_A\otimes I\right)-\hbar\omega(\mathbf{k}_2)(H_A\otimes I)\\
&\qquad+\hbar^2\omega(\mathbf{k}_1)^2\Big(I\otimes I+2(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes H_B+H_A\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big)\\
&\qquad+\hbar^2\omega(\mathbf{k}_2)^2\left(H_A\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\right)+\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)(I\otimes I)\\
&\qquad-\hbar^3\omega(\mathbf{k}_1)^3\left((H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I+I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\right)\\
&\qquad-\hbar^3\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)^2\left(I\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\right)\\
&\qquad-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\left((H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I\right)\\
&\qquad+\hbar^4\omega(\mathbf{k}_1)^4\left((H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\right)\\
&\qquad+\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\left((H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\right)\Big\rangle_{\mathbf{v}_A^{\alpha}\otimes\mathbf{v}_B^{\beta}}.
\end{aligned}$$

Recalling the definitions of $S_{A,B}$, $\alpha_E^{A,B}(\mathbf{k})$ (see (4.4.1) and Theorem 2.8.4, respectively) and defining the magnetic polarizabilities

$$\alpha_M^{A,B} := \langle \mathbf{v}_{A,B}\psi_{A,B}^0 | \mathbf{v}_{A,B}\psi_{A,B}^0\rangle, \tag{4.4.29}$$

(these are well-defined, since $\mathbf{x}_i\psi^0_{A,B} \in H^2(\mathbb{R}^{3Z_{A,B}})$ by the remarks in section 2.5), we find

$$
\begin{aligned}
&(4.4.25)\\
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Big[4S_AS_B - 2\hbar\omega(\mathbf{k}_1)\left(2\alpha^A_MS_B + S_A\alpha^B_M\right) - 2\hbar\omega(\mathbf{k}_2)S_A\alpha^B_M\\
&\qquad + 2\hbar^2\omega(\mathbf{k}_1)^2\Big(\alpha^A_M\alpha^B_M + 2\alpha^A_E(\mathbf{k}_1)S_B + S_A\alpha^B_E(\mathbf{k}_1)\Big)\\
&\qquad + 2\hbar^2\omega(\mathbf{k}_2)^2\left(S_A\alpha^B_E(\mathbf{k}_2)\right) + \hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)(I\otimes I)\\
&\qquad - 2\hbar^3\omega(\mathbf{k}_1)^3\ \left(\alpha^A_E(\mathbf{k}_1)\alpha^B_M + \alpha^A_M\alpha^B_E(\mathbf{k}_1)\right)\\
&\qquad - 2\hbar^3\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)^2\alpha^A_M\alpha^B_E(\mathbf{k}_2) - 2\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\alpha^A_E(\mathbf{k}_1)\alpha^B_M\\
&\qquad + 2\hbar^4\omega(\mathbf{k}_1)^4\alpha^A_E(\mathbf{k}_1)\alpha^B_E(\mathbf{k}_1) + 2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\alpha^A_E(\mathbf{k}_1)\alpha^B_E(\mathbf{k}_2)\Big].
\end{aligned}
$$

According to Lemma A.10.1, we can replace

$$
\begin{aligned}
-2\hbar\omega(\mathbf{k}_1)2\alpha^A_MS_B \text{ by } &-2(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)\hbar\alpha^A_MS_B,\\
2\hbar^2\omega(\mathbf{k}_1)^2\alpha^A_M\alpha^B_M \text{ by } &\left(\omega(\mathbf{k}_1)^2+\omega(\mathbf{k}_2)^2\right)\hbar^2\alpha^A_M\alpha^B_M,\\
2\hbar^2\omega(\mathbf{k}_2)^2\left(S_A\alpha^B_E(\mathbf{k}_2)\right) \text{ by } &2\hbar^2\omega(\mathbf{k}_1)^2\left(S_A\alpha^B_E(\mathbf{k}_1)\right),\\
-2\hbar^3\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)^2\alpha^A_M\alpha^B_E(\mathbf{k}_2) \text{ by } &-2\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\alpha^A_M\alpha^B_E(\mathbf{k}_1), \text{ and}\\
2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\alpha^A_E(\mathbf{k}_1)\alpha^B_E(\mathbf{k}_2) \text{ by } &\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\left(\alpha^A_E(\mathbf{k}_1)\alpha^B_E(\mathbf{k}_2)+\alpha^A_E(\mathbf{k}_2)\alpha^B_E(\mathbf{k}_1)\right),
\end{aligned}
$$

obtaining

$$
\begin{aligned}
&(4.4.25)\\
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Big[4S_AS_B - 2\hbar\left(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)\right)\left(\alpha^A_MS_B + S_A\alpha^B_M\right)\\
&\qquad + \hbar^2\alpha^A_M\alpha^B_M\Big(\underbrace{\omega(\mathbf{k}_1)^2+2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)+\omega(\mathbf{k}_2)^2}_{=(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^2}\Big)\\
&\qquad + 4\hbar^2\omega(\mathbf{k}_1)^2\left(S_A\alpha^B_E(\mathbf{k}_1) + \alpha^A_E(\mathbf{k}_1)S_B\right)\\
&\qquad - 2\hbar^3\left(\omega(\mathbf{k}_1)^3+\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\right)\left(\alpha^A_E(\mathbf{k}_1)\alpha^B_M + \alpha^A_M\alpha^B_E(\mathbf{k}_1)\right)\\
&\qquad + 2\hbar^4\omega(\mathbf{k}_1)^4\alpha^A_E(\mathbf{k}_1)\alpha^B_E(\mathbf{k}_1)\\
&\qquad + \hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\left(\alpha^A_E(\mathbf{k}_1)\alpha^B_E(\mathbf{k}_2)+\alpha^A_E(\mathbf{k}_2)\alpha^B_E(\mathbf{k}_1)\right)\Big].
\end{aligned}
$$

Now consider (4.4.27). Using the commutator relation from Lemma A.7.1, the definition of $\mathbf{v}_{A,B}$, the identity $(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}H_{A,B} = I - \hbar\omega(\mathbf{k})(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}$ (which holds

on $D(H_{A,B})$) and the commutator relation from Lemma 4.2.18ii)(4.2.22), we find

$$\begin{aligned}
&\sum_{i_A,j_B,k_A,l_B} \Big\langle \mathbf{p}^{\alpha}_{i_A}\Psi^0_A \otimes \mathbf{p}^{\beta}_{j_B}\Psi^0_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_1))^{-1} \\
&\qquad\qquad \times \Big[(H_A+\hbar\omega(\mathbf{k}_2))^{-1}+(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big] \Big| \mathbf{p}^{\alpha}_{k_A}\Psi^0_A \otimes \mathbf{p}^{\beta}_{l_B}\Psi^0_B \Big\rangle \\
&= \Big(\frac{m_e}{\hbar}\Big)^4 \Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| \big((I-\hbar\omega(\mathbf{k}_1)(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\big)\big(I-\hbar\omega(\mathbf{k}_2)(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\big) \\
&\qquad\qquad \otimes (H_B-\hbar\omega(\mathbf{k}_1)+(\hbar\omega(\mathbf{k}_1))^2(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big] \\
&\qquad + \big(H_A-\hbar\omega(\mathbf{k}_1)+(\hbar\omega(\mathbf{k}_1))^2(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\big) \\
&\qquad\qquad \otimes \Big[\big((I-\hbar\omega(\mathbf{k}_1)(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\big) \\
&\qquad\qquad\qquad \times \big(I-\hbar\omega(\mathbf{k}_2)(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\big)\Big]\Big| \\
&\qquad \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle.
\end{aligned}$$

Rearranging terms yields

$$\begin{aligned}
&\Big(\frac{m_e}{\hbar}\Big)^4 \Big[\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| H_A\otimes I + I\otimes H_B \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad - \hbar\omega(\mathbf{k}_1)\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes H_B \\
&\qquad\qquad + H_A\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1} + 2I\otimes I \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad - \hbar\omega(\mathbf{k}_2)\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes H_B + H_A\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1} \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad + 2(\hbar\omega(\mathbf{k}_1))^2\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1} + (H_B+\hbar\omega(\mathbf{k}_1))^{-1} \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad + \hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes H_B \\
&\qquad\qquad + H_A\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1} \\
&\qquad\qquad + (H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes I + I\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1} \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad - \hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes I \\
&\qquad\qquad + I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1} \\
&\qquad\qquad + (H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1} \\
&\qquad\qquad + (H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1} \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad - 2\hbar^3\omega(\mathbf{k}_1)^3\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1} \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \\
&\quad + \hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)\Big\langle \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big| (H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1} \\
&\qquad\qquad \otimes (H_B+\hbar\omega(\mathbf{k}_1))^{-1} \\
&\qquad\qquad + (H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1} \\
&\qquad\qquad \times (H_B+\hbar\omega(\mathbf{k}_2))^{-1} \Big| \mathbf{v}^{\alpha}_A \otimes \mathbf{v}^{\beta}_B \Big\rangle \Big].
\end{aligned}$$

Setting

$$T_3(\mathbf{k}_1,\mathbf{k}_2) := \sum_{\alpha=1}^{3} \Big\langle (H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1} \Big\rangle_{\mathbf{v}_A^\alpha} \alpha_M^B + \alpha_M^A \Big\langle (H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1} \Big\rangle_{\mathbf{v}_B^\alpha}$$

and using the definitions of $S_{A,B}$, $\alpha_M^{A,B}$, $\alpha_E^{A,B}(\mathbf{k})$, $T_4(\mathbf{k}_1,\mathbf{k}_2)$ (for the latter, see (3.0.6)) and Lemma A.10.1 (exchange $\mathbf{k}_2$ and $\mathbf{k}_1$ for otherwise symmetric integrands), we obtain

(4.4.27)

$$\begin{aligned}
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad\times\Bigg[S_A\alpha_M^B + \alpha_M^A S_B - 2\hbar\omega(\mathbf{k}_1)\left(\alpha_E^A(\mathbf{k}_1)S_B + S_A\alpha_E^B(\mathbf{k}_1) + \alpha_M^A\alpha_M^B\right) \\
&\qquad + \left(2\hbar^2\omega(\mathbf{k}_1)^2 + \hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\right)\left(\alpha_E^A(\mathbf{k}_1)\alpha_M^B + \alpha_M^A\alpha_E^B(\mathbf{k}_1)\right) \\
&\qquad + \hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg[\sum_{\alpha=1}^{3}\Big\langle \mathbf{v}_A^\alpha\Big|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{v}_A^\alpha\Big\rangle S_B \\
&\qquad + S_A\sum_{\beta=1}^{3}\Big\langle \mathbf{v}_B^\beta\Big|(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{v}_B^\beta\Big\rangle\Bigg] \\
&\qquad - \hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\Big[T_3(\mathbf{k}_1,\mathbf{k}_2) + \alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1) + \alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)\Big] \\
&\qquad - 2\hbar^3\omega(\mathbf{k}_1)^3\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1) + \hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2)\Bigg].
\end{aligned}$$

Next we consider (4.4.26). Performing the same steps as for (4.4.27), we end up with

(4.4.26)

$$\begin{aligned}
&= -\frac{1}{9m_e^4}\Big(\frac{m_e}{\hbar}\Big)^4\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad\times\Bigg[\Bigg\{\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|(H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\qquad + \Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|(I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\qquad + 2\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}(H_A\otimes H_B)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg\} \qquad (4.4.30)
\end{aligned}$$

$$
\begin{aligned}
&- 2\hbar\omega(\mathbf{k}_1)\Bigg[\Bigg\{\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(H_A\otimes(H_B+\omega(\mathbf{k}_1))^{-1}\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+\Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big((H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg\} \qquad (4.4.31)\\
&+\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(H_A\otimes I+I\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg] \qquad (4.4.32)\\
&+\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg[\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1} \\
&\qquad\times\Big(H_A\otimes(H_B+\omega(\mathbf{k}_1))^{-1}(H_B+\omega(\mathbf{k}_2))^{-1}\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\quad+\Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1} \\
&\qquad\times\Big((H_A+\omega(\mathbf{k}_1))^{-1}(H_A+\omega(\mathbf{k}_2))^{-1}\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\quad+2\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(I\otimes I\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg] \qquad (4.4.33)\\
&+2(\hbar\omega(\mathbf{k}_1))^2\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes H_B \\
&\qquad+H_A\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big]\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \qquad (4.4.34)\\
&-2\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I \\
&\qquad+I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big]\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1} \\
&\qquad\otimes(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg].
\end{aligned}
$$

Using Lemma 4.2.18ii)(4.2.22) and the fact that on $D(H_A)\widehat{\otimes}D(H_B)$ we have $\overline{H_A\otimes I}=H_A\otimes I$ and $\overline{I\otimes H_B}=I\otimes H_B$, we find

$$
\begin{aligned}
&\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|(H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+\Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|(I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+2\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}(H_A\otimes H_B)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
=&\Big\langle (\overline{H_A\otimes I})(\overline{H_A+H_B})^{-1}(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+\Big\langle (\overline{I\otimes H_B})(\overline{H_A+H_B})^{-1}(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+\Big\langle (\overline{H_A\otimes I})(\overline{H_A+H_B})^{-1}(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&+\Big\langle (\overline{I\otimes H_B})(\overline{H_A+H_B})^{-1}(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle
\end{aligned}
$$

$$
\begin{aligned}
&=\Big\langle \Big[(\overline{H_A\otimes I})+(\overline{I\otimes H_B})\Big](\overline{H_A+H_B})^{-1}(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(H_A\otimes I+I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&=\Big\langle (\overline{H_A+H_B})^{-1}(H_A+H_B)(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(H_A+H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle.
\end{aligned}
$$

On $D(H_A)\widehat{\otimes}D(H_B)$ we have $\overline{H_A+H_B}=H_A+H_B$, and thus, again recalling the definitions of $\alpha_M^{A,B}$ and $S_{A,B}$ ((4.4.29) and (4.4.1)), we obtain

$$
(4.4.30)=-\frac{S_A\alpha_M^B+\alpha_M^AS_B}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}.
$$

For the same reason,

$$
(4.4.32)=\frac{2\alpha_M^A\alpha_M^B}{\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\hbar\omega(\mathbf{k}_1)(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}.
$$

Furthermore, by Proposition 2.5.1, $\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\in D(H_A)\widehat{\otimes}D(H_B)$, so that we can write

$$
\begin{aligned}
&\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(H_A\otimes(H_B+\omega(\mathbf{k}_1))^{-1}\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\quad+\Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big((H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&=\Big\langle (H_A+H_B-H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(H_A\otimes(H_B+\omega(\mathbf{k}_1))^{-1}\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\quad+\Big\langle (H_A-H_A+H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big((H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&=\Big\langle \mathbf{v}_A^\alpha|H_A|\mathbf{v}_A^\alpha\Big\rangle\Big\langle \mathbf{v}_B^\beta|(H_B+\omega(\mathbf{k}_1))^{-1}|\mathbf{v}_B^\beta\Big\rangle+\Big\langle \mathbf{v}_A^\alpha|(H_A+\omega(\mathbf{k}_1))^{-1}|\mathbf{v}_A^\alpha\Big\rangle\Big\langle \mathbf{v}_B^\beta|H_B|\mathbf{v}_B^\beta\Big\rangle\\
&\quad-\Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(H_A\otimes(H_B+\omega(\mathbf{k}_1))^{-1}\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\quad-\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big((H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle.
\end{aligned}
$$

Again using Lemma 4.2.18ii)(4.2.22), the fact that on $D(H_A)\widehat{\otimes}D(H_B)$ we have $\overline{H_A\otimes I}=H_A\otimes I$ and $\overline{I\otimes H_B}=I\otimes H_B$, and the identities

$$
H_{A,B}(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}=I-\hbar\omega(\mathbf{k})(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1},
$$

we obtain

$$
\begin{aligned}
&-\Big\langle (I\otimes H_B)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big(H_A\otimes(H_B+\omega(\mathbf{k}_1))^{-1}\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\quad-\Big\langle (H_A\otimes I)\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big((H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\Big)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&=-\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big[\big(I-\hbar\omega(\mathbf{k}_1)(H_A+\omega(\mathbf{k}_1))^{-1}\big)\otimes H_B\\
&\quad+H_A\otimes\big(I-\hbar\omega(\mathbf{k}_1)(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\big)\Big]\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&=-\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}(H_A+H_B)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\quad+\hbar\omega(\mathbf{k}_1)\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big[(H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\\
&\quad+H_A\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big]\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle
\end{aligned}
$$

$$
\begin{aligned}
&= -\Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big| (I\otimes I) \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \\
&\quad + \hbar\omega(\mathbf{k}_1) \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big| (\overline{H_A + H_B})^{-1} \Big[(H_A + \omega(\mathbf{k}_1))^{-1} \otimes H_B \\
&\quad + H_A \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1} \Big] \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle .
\end{aligned}
$$

Now using the definitions of $\alpha_E^{A,B}(\mathbf{k})$, $S_{A,B}$ and $\alpha_M^{A,B}$, we conclude

$$
\begin{aligned}
&(4.4.31) \\
&= -\frac{1}{9\hbar^4} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 |C(\mathbf{k}_2)|^2 (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad \times \Bigg[2\hbar\omega(\mathbf{k}_1) \left[\alpha_M^A \alpha_M^B - S_A \alpha_E^B(\mathbf{k}_1) - \alpha_E^A(\mathbf{k}_1) S_B \right] \\
&\qquad - 2(\hbar\omega(\mathbf{k}_1))^2 \sum_{\alpha,\beta=1}^{3} \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big| (\overline{H_A + H_B})^{-1} \Big[(H_A + \omega(\mathbf{k}_1))^{-1} \otimes H_B \\
&\qquad\qquad + H_A \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1} \Big] \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \Bigg] .
\end{aligned}
$$

Note that last term cancels (4.4.34). Repeating the same arguments for the term (4.4.33), we find

$$
\begin{aligned}
&(4.4.33) \\
&= -\frac{1}{9\hbar^4} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 |C(\mathbf{k}_2)|^2 \omega(\mathbf{k}_1) (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad \times \Bigg[\hbar^2 \omega(\mathbf{k}_1)\omega(\mathbf{k}_2) \Bigg[2 \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big| (\overline{H_A + H_B})^{-1} \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \\
&\qquad + \sum_{\alpha=1}^{3} \Big\langle \mathbf{v}_A^\alpha \Big| (H_A + \hbar\omega(\mathbf{k}_1))^{-1} (H_A + \hbar\omega(\mathbf{k}_2))^{-1} \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle S_B \\
&\qquad + S_A \sum_{\beta=1}^{3} \Big\langle \mathbf{v}_B^\beta \Big| (H_B + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} \Big| \mathbf{v}_B^\beta \Big\rangle \\
&\qquad - \sum_{\alpha,\beta=1}^{3} \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big| (\overline{H_A + H_B})^{-1} \Big[(H_A + \omega(\mathbf{k}_1))^{-1} \otimes H_B \\
&\qquad\qquad + H_A \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1} \Big] \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \Bigg] \qquad (4.4.35) \\
&\quad + \hbar^3 \omega(\mathbf{k}_1)\omega(\mathbf{k}_2)^2 \\
&\qquad \times \Bigg[\sum_{\alpha,\beta=1}^{3} \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big| (\overline{H_A + H_B})^{-1} \Big[(H_A + \omega(\mathbf{k}_1))^{-1} (H_A + \omega(\mathbf{k}_2))^{-1} \otimes H_B \\
&\qquad\qquad + H_A \otimes (H_B + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} \Big] \Big| \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \Bigg] \Bigg] .
\end{aligned}
$$

To simplify (4.4.35), we use the fact that $\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \in D(H_A)\widehat{\otimes}D(H_B)$, the relation $(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}H_{A,B} = I - \hbar\omega(\mathbf{k})(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}$ (valid on $D(H_{A,B})$), the commutator relation (4.2.22) from Lemma 4.2.18ii) and the fact that $\overline{H_A+H_B} = H_A+H_B$, $\overline{(H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I} = (H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I$ and $\overline{I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}} = I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}$ on $D(H_A)\widehat{\otimes}D(H_B)$, to conclude

$$
\begin{aligned}
&\sum_{\alpha,\beta=1}^{3}\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\left((H_A+\omega(\mathbf{k}_1))^{-1}\otimes H_B\right)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
=&\sum_{\alpha,\beta=1}^{3}\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\left((H_A+\omega(\mathbf{k}_1))^{-1}(H_A+H_B-H_B)\right)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
=&\sum_{\alpha,\beta=1}^{3}\Bigg[\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{(H_A+\omega(\mathbf{k}_1))^{-1}\otimes I})(\overline{H_A+H_B})^{-1}(H_A+H_B)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\qquad-\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\left(I-\hbar\omega(\mathbf{k}_1)(H_A+\omega(\mathbf{k}_1))^{-1}\right)\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg]\\
=&\alpha_E^A(\mathbf{k}_1)\alpha_M^B\\
&-\sum_{\alpha,\beta=1}^{3}\Bigg[\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\qquad+\hbar\omega(\mathbf{k}_1)\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}(H_A+\omega(\mathbf{k}_1))^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg],
\end{aligned}
$$

and accordingly for the second term in (4.4.35), yielding

$$
\begin{aligned}
&(4.4.35)\\
=&\alpha_E^A(\mathbf{k}_1)\alpha_M^B+\alpha_M^A\alpha_E^B(\mathbf{k}_1)\\
&-2\sum_{\alpha,\beta=1}^{3}\Bigg[\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\\
&\qquad+\hbar\omega(\mathbf{k}_1)\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big[(H_A+\omega(\mathbf{k}_1))^{-1}\\
&\qquad\qquad+(H_B+\omega(\mathbf{k}_1))^{-1}\Big]\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg].
\end{aligned}
$$

Putting everything together, setting

$$
\begin{aligned}
T_1(\mathbf{k}) :=& \sum_{\alpha=1}^{3}\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta|(\overline{H}_A+\overline{H}_B)^{-1}\Big[(\overline{H}_A+\hbar\omega(\mathbf{k}))^{-1}+(\overline{H}_B+\hbar\omega(\mathbf{k}))^{-1}\Big]|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\rangle,\\
T_2(\mathbf{k}_1,\mathbf{k}_2) :=& \sum_{\alpha,\beta=1}^{3}\Big\langle(\overline{H_A+H_B})^{-1}\Big((H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}H_B\\
&+H_A(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big)\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta},
\end{aligned}
$$

and recalling the definition of $T_5(\mathbf{k}_1, \mathbf{k}_2)$ from (3.0.7), we have arrived at

(4.4.26)

$$
\begin{aligned}
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\omega(\mathbf{k}_1)(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad\times\Bigg[S_A\alpha_M^B+\alpha_M^A S_B+2\hbar\omega(\mathbf{k}_1)\left[\alpha_M^A\alpha_M^B-S_A\alpha_E^B(\mathbf{k}_1)-\alpha_E^A(\mathbf{k}_1)S_B-\alpha_M^A\alpha_M^B\right] \\
&\qquad+\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg[2\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\qquad\quad+\sum_{\alpha=1}^3\Big\langle \mathbf{v}_A^\alpha\Big|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle S_B \\
&\qquad\quad+S_A\sum_{\beta=1}^3\Big\langle \mathbf{v}_B^\beta\Big|(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{v}_B^\beta\Big\rangle \\
&\qquad\quad-\Bigg[\alpha_E^A(\mathbf{k}_1)\alpha_M^B+\alpha_M^A\alpha_E^B(\mathbf{k}_1)-2\sum_{\alpha,\beta=1}^3\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\qquad\qquad+\hbar\omega(\mathbf{k}_1)T_1(\mathbf{k}_1)\Bigg]\Bigg] \\
&\qquad+\hbar^3\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)^2T_2(\mathbf{k}_1,\mathbf{k}_2)-2\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)T_1(\mathbf{k}_1) \\
&\qquad+2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)\Bigg] \\
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\omega(\mathbf{k}_1)(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\quad\times\Bigg[S_A\alpha_M^B+\alpha_M^A S_B+2\hbar\omega(\mathbf{k}_1)\left[-S_A\alpha_E^B(\mathbf{k}_1)-\alpha_E^A(\mathbf{k}_1)S_B\right] \\
&\qquad+4\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|(\overline{H_A+H_B})^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle \\
&\qquad+\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg[\sum_{\alpha=1}^3\Big\langle \mathbf{v}_A^\alpha\Big|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle S_B \\
&\qquad\quad+S_A\sum_{\beta=1}^3\Big\langle \mathbf{v}_B^\beta\Big|(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big|\mathbf{v}_B^\beta\Big\rangle\Bigg] \\
&\qquad-\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\left(\alpha_E^A(\mathbf{k}_1)\alpha_M^B+\alpha_M^A\alpha_E^B(\mathbf{k}_1)\right) \\
&\qquad+\hbar^3\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)^2T_2(\mathbf{k}_1,\mathbf{k}_2)-3\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)T_1(\mathbf{k}_1) \\
&\qquad+2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)\Bigg].
\end{aligned}
$$

Adding all contributions, noting that $T_2(\mathbf{k}_2,\mathbf{k}_1)=T_2(\mathbf{k}_1,\mathbf{k}_2)$ (this follows from commutativity of resolvents of a fixed operator) and using Lemma A.10.1 now yields

$$
\begin{aligned}
&(4.4.25)+(4.4.26)+(4.4.27)\\
=&-\frac{1}{9\hbar^4}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Bigg[\frac{4S_AS_B}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}+(S_A\alpha_M^B+\alpha_M^AS_B)\underbrace{(2-2\frac{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)})}_{=0}\\
&\quad+\big(S_A\alpha_E^B(\mathbf{k}_1)+\alpha_E^A(\mathbf{k}_1)S_B\big)\left(-4\hbar\omega(\mathbf{k}_1)+\hbar\frac{4\omega(\mathbf{k}_1)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\quad+4\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\left(\sum_{\alpha,\beta=1}^3\Big\langle\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta|(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\right)\\
&\quad+(\alpha_E^A(\mathbf{k}_1)\alpha_M^A+\alpha_M^B\alpha_E^B(\mathbf{k}_1)\underbrace{\left(2\hbar^2\omega(\mathbf{k}_1)^2-\frac{2\hbar^2\omega(\mathbf{k}_1)^3}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-\frac{2\hbar^2\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)}_{=0}\\
&\quad+(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(-2\hbar^3\omega(\mathbf{k}_1)^3+\frac{2\hbar^3\omega(\mathbf{k}_1)^4}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\quad+\big(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\big)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)+\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\quad+\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)\Big[-3T_1(\omega(\mathbf{k}_1))+T_2(\mathbf{k}_1,\mathbf{k}_2)-T_3(\mathbf{k}_1,\mathbf{k}_2)\Big]\\
&\quad+\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2)+2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)\\
&\quad+2\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Big(\sum_{\alpha,\beta=1}^3\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}S_B\\
&\quad+S_A\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\Big).
\end{aligned}
$$

Next we claim that

$$
\begin{aligned}
&-3T_1(\mathbf{k}_1)+T_2(\mathbf{k}_1,\mathbf{k}_2)-T_3(\mathbf{k}_1,\mathbf{k}_2)\\
=&-4\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1)-8\hbar\omega(\mathbf{k}_1)T_5(\mathbf{k}_1,\mathbf{k}_1)+\hbar\omega(\mathbf{k}_2)T_6(\mathbf{k}_1,\mathbf{k}_2).
\end{aligned}
$$

To see this, observe that by Lemma 4.2.18 v) (recall the definition of the magnetic polarizabilities $\alpha_M^{A,B}$ from (4.4.29)),

$$
\begin{aligned}
&T_2(\mathbf{k}_1,\mathbf{k}_2)\\
=&\sum_{\alpha,\beta=1}^3\Bigg[\Big\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes I\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\\
&\qquad+\Big\langle I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\\
&\qquad-\Big\langle(\overline{H_A+H_B}^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}\otimes I+I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}\Big]\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\Bigg]
\end{aligned}
$$

$$
\begin{aligned}
&+\hbar\omega(\mathbf{k}_2)\sum_{\alpha,\beta=1}^{3}\Big\langle(\overline{H_A+H_B}^{-1}\Big[(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\otimes I\\
&\qquad\qquad+I\otimes(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big]\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\\
=&T_3(\mathbf{k}_1,\mathbf{k}_2)-T(\mathbf{k}_1)+\hbar\omega(\mathbf{k}_2)T_6(\mathbf{k}_1,\mathbf{k}_2).
\end{aligned}
$$

Furthermore, Lemma 4.2.18 iv) yields

$$
\begin{aligned}
&T_1(\mathbf{k})\\
=&\alpha_E^A(\mathbf{k})\alpha_E^B(\mathbf{k})+2\hbar\omega(\mathbf{k})\sum_{\alpha,\beta=1}^{3}\Big\langle(\overline{H_A+H_B}^{-1}\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\\
=&\alpha_E^A(\mathbf{k})\alpha_E^B(\mathbf{k})+2\hbar\omega(\mathbf{k})T_5(\mathbf{k},\mathbf{k}).
\end{aligned}
$$

Using this result, we can now rewrite

$$
\begin{aligned}
&(4.4.25)+(4.4.26)+(4.4.27)\\
=&-\frac{1}{9\hbar^4}\int_{\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Bigg[4\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg(\sum_{\alpha,\beta=1}^{3}\Big\langle\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta|(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle\Bigg)\\
&\quad+(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(-4\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)-2\hbar^3\omega(\mathbf{k}_1)^3+\frac{2\hbar^3\omega(\mathbf{k}_1)^4}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\quad+\left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)+\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\quad+\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2)+2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)\\
&\quad-8\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_5(\mathbf{k}_1,\mathbf{k}_1)+\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_6(\mathbf{k}_1,\mathbf{k}_2)\\
&\quad+2\hbar^2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Big[\sum_{\alpha,\beta=1}^{3}\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}S_B\\
&\quad+S_A\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\Big]\\
&\quad+\frac{4S_AS_B}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}+\left(S_A\alpha_E^B(\mathbf{k}_1)+\alpha_E^A(\mathbf{k}_1)S_B\right)\left(-4\hbar\omega(\mathbf{k}_1)+\hbar\frac{4\omega(\mathbf{k}_1)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg].
\end{aligned}
$$

Finally, noting that on $\mathbb{R}^6\setminus\{0\}$ we have the identity

$$
\begin{aligned}
&-4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)-2\omega(\mathbf{k}_1)^3+\frac{2\omega(\mathbf{k}_1)^4}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\\
=&\frac{-4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2-6\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\\
=&\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\left(\frac{-2\omega(\mathbf{k}_1)^2-4\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)-4((\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^2-\omega(\mathbf{k}_2)^2-2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2))}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
=&\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\left(2(\omega(\mathbf{k}_2)-\omega(\mathbf{k}_1))+4\frac{\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}+2\frac{\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-4\omega(\mathbf{k}_1)-4\omega(\mathbf{k}_2)\right)
\end{aligned}
$$

$$=\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\left(-2\frac{\omega(\mathbf{k}_2)(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-6\omega(\mathbf{k}_1)+4\frac{\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}+2\frac{\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)$$
$$=\frac{2\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-6\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)$$

finishes the proof of Lemma 4.4.1.

4.4.3 Proof of Lemma 4.4.2

By the definition of the perturbation operators $H''_{\sigma,A}$ and $H''_{\sigma,B}$ (see (2.4.6) and (2.4.7)),

$$\begin{aligned}(4.3.5) &= -2\mathrm{Re}[\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\rangle]\\ &= -\left(\frac{1}{2m_e}\right)^2 2\mathrm{Re}\Big\langle(\frac{Z_A}{c^2}\mathbf{A}_\sigma(0)^2)\Psi_0|T^\sigma|(\frac{Z_B}{c^2}\mathbf{A}_\sigma(\mathbf{R})^2)\Psi_0\Big\rangle_{\mathcal{H}}.\end{aligned}$$

Recall the definitions of the magnetic vector potential $\mathbf{A}_\sigma(\mathbf{x}) = a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma) + a(\mathbf{G}^{\mathbf{x}}_\sigma)$, with the coupling functions $\mathbf{G}^{\mathbf{x}}_\sigma(\mathbf{k},\lambda) = \chi_\sigma(\mathbf{k})\mathbf{G}^{\mathbf{x}}(\mathbf{k},\lambda) = \chi_\sigma(\mathbf{k})\, cC(\mathbf{k})\mathbf{e}(\mathbf{k},\lambda)\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}}$. Note that since $\Psi_0 = \Psi^0_A\otimes\Psi^0_B\otimes\Omega$ and $a(f)\Omega = 0$ for any $f\in W_\sigma = L^2(\Omega_\sigma)\oplus L^2(\Omega_\sigma)$, we have, using the commutator relation $[a(f),a^\dagger(g)] = \langle f,g\rangle_{W_\sigma}$,

$$\begin{aligned}&\mathbf{A}_\sigma(\mathbf{x}_k)^2\Psi_0\\ =&\Big[a^\dagger(\mathbf{G}^{\mathbf{x}_k}_\sigma)\cdot a^\dagger(\mathbf{G}^{\mathbf{x}_k}_\sigma) + a(\mathbf{G}^{\mathbf{x}_k}_\sigma)\cdot a(\mathbf{G}^{\mathbf{x}_k}_\sigma) + 2a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma)\cdot a(\mathbf{G}^{\mathbf{x}}_\sigma) + \langle\mathbf{G}^{\mathbf{x}}_\sigma,\mathbf{G}^{\mathbf{x}}_\sigma\rangle_{W_\sigma}\Big]\Psi_0\\ =&a^\dagger(\mathbf{G}^{\mathbf{x}_k}_\sigma)\cdot a^\dagger(\mathbf{G}^{\mathbf{x}_k}_\sigma)\Psi_0 + (\sum_{\alpha=1}^3\|(\mathbf{G}^{\mathbf{x}}_\sigma)_\alpha\|^2_{W_\sigma})\Psi_0,\end{aligned}$$

so that $\mathbf{A}_\sigma(\mathbf{0})^2\Psi_0$ and $\mathbf{A}_\sigma(\mathbf{R})^2\Psi_0$ are elements of $\Psi_0\oplus\{\Psi^0_A\}\widehat{\otimes}\{\Psi^0_B\})\widehat{\otimes}\mathcal{F}^{(2)}$. On $\{\Psi_0\}$, the reduced resolvent T^σ acts trivially (this subspace is projected out in its construction), and for the remaining terms we can use the invariance properties of T^σ (Lemma 4.2.5) and the mutual orthogonality of Fock space levels to conclude

$$\begin{aligned}&-2\mathrm{Re}[\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\rangle]\\ =&-\frac{Z_A\,Z_B}{2m_e^2c^4}\,\mathrm{Re}\Big\langle a^\dagger(\mathbf{G}^0_\sigma)\cdot a^\dagger(\mathbf{G}^0_\sigma)\Psi_0|T^\sigma|a^\dagger(\mathbf{G}^{\mathbf{R}}_\sigma)\cdot a^\dagger(\mathbf{G}^{\mathbf{R}}_\sigma)\Psi_0\Big\rangle.\end{aligned}$$

By the definition of the creation operators,

$$\begin{aligned}&\Big(a^\dagger(\mathbf{G}^{\mathbf{x}_k}_\sigma)\cdot a^\dagger(\mathbf{G}^{\mathbf{x}_k}_\sigma)\Psi_0\Big)(\mathbf{x}_1,\ldots,\mathbf{x}_N,\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu)\\ =&\frac{2}{\sqrt{2}}(\mathbf{G}^{\mathbf{x}_k}_\sigma(\mathbf{k}_1,\lambda)\cdot\mathbf{G}^{\mathbf{x}_k}_\sigma(\mathbf{k}_2,\mu)(\Psi^0_A\otimes\Psi^0_B)(\mathbf{x}_1,\ldots,\mathbf{x}_N).\end{aligned}$$

Furthermore, T^σ acts as $I_{\mathcal{H}_A} \otimes I_{\mathcal{H}_B} \otimes \frac{1}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}$ on $\{\Psi_A^0\}\widehat{\otimes}\{\Psi_B^0\}\widehat{\otimes}\mathcal{F}_\sigma^{(2)}$ (see Lemma 4.2.5), so that we find

$$\begin{aligned}
&-2\mathrm{Re}[\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\rangle] \\
&= -\frac{Z_A\,Z_B}{m_e^2}\underbrace{\|\Psi_A^0\|^2\,\|\Psi_B^0\|^2}_{=1} \\
&\quad\times \mathrm{Re}\left[\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2}{(\hbar\omega(\mathbf{k}_1)+\hbar\omega(\mathbf{k}_2))}\,(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))^2\,\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\right].
\end{aligned}$$

Lemma A.6.1 asserts that $\sum_{\lambda,\mu=1,2}(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))^2 = 1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2$, which allows us to carry out the summation over the photon polarizations, and by Lemma A.9.1, $Z_{A,B} = (2m_e)/(3\hbar^2)S_{A,B}$, which finally yields

$$\begin{aligned}
&-2\mathrm{Re}[\langle H''_{\sigma,A}\Psi_0|T^\sigma|H''_{\sigma,B}\Psi_0\rangle] \\
&= -\frac{4}{9\hbar^5}S_A S_B\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2),
\end{aligned}$$

finishing the proof.

4.4.4 Proof of Lemma 4.4.3

i) We first consider the term

$$\begin{aligned}
&(4.3.7) \\
=&\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_{\sigma,B}\Psi_0\rangle + \langle H'_{\sigma,B}\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_{\sigma,A}\Psi_0\rangle \\
&+\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\rangle + \langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle \\
=&2\,\mathrm{Re}\left[\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_{\sigma,B}\Psi_0\rangle\right] + \langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\rangle \\
&+\langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle. \qquad (4.4.36)
\end{aligned}$$

To analyze these expressions further, we first consider a general term of the form

$$\frac{1}{2m_e^3c^4}\sum_{i\in I_i,j\in I_j,k\in I_k}\left\langle T^\sigma(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i))\Psi_0|\mathbf{A}_\sigma(\mathbf{x}_k)^2|T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0\right\rangle_{\mathcal{H}}, \qquad (4.4.37)$$

where the sets I_i, I_j, I_k over which the indices are ranging will be specified below. Note that so far, no dipole approximation has been employed. By the invariance properties of the reduced resolvent T^σ (see Lemma 4.2.5), we have

$$T^\sigma(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i))\Psi_0 \in \mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}_\sigma^{(1)}$$

for any i. Therefore only the contributions to $\mathbf{A}(\mathbf{x}_k)^2T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0$ which lie in $\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}_\sigma^{(1)}$ contribute to the inner product. Recall the definitions of the magnetic vector potential $\mathbf{A}_\sigma(\mathbf{x}) = a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}}) + a(\mathbf{G}_\sigma^{\mathbf{x}})$, the one-photon space $W_\sigma = L^2(\Omega_\sigma)\oplus L^2(\Omega_\sigma)$ and the coupling functions $\mathbf{G}_\sigma^{\mathbf{x}}(\mathbf{k},\lambda) = \chi_\sigma(\mathbf{k})\mathbf{G}^{\mathbf{x}}(\mathbf{k},\lambda) = \chi_\sigma(\mathbf{k})\,c\,C(\mathbf{k})\mathbf{e}(\mathbf{k},\lambda)\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}}$. Using

the commutator relation $[a(f), a^\dagger(g)] = \langle f, g\rangle_{W_\sigma}$, we find that for fixed $\mathbf{x} \in \mathbb{R}^3$, the part of $\mathbf{A}_\sigma(\mathbf{x})^2$ which leaves each Fock space level invariant is given by

$$2\, a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma) \cdot a(\mathbf{G}^{\mathbf{x}}_\sigma) + \langle \mathbf{G}^{\mathbf{x}}_\sigma, \mathbf{G}^{\mathbf{x}}_\sigma\rangle_{W_\sigma}.$$

On a function $v \in W_\sigma = \mathcal{F}^{(1)}_\sigma$, this operator acts as

$$\begin{aligned}&\left[\left(2\, a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma) \cdot a(\mathbf{G}^{\mathbf{x}}_\sigma) + \langle \mathbf{G}^{\mathbf{x}}_\sigma, \mathbf{G}^{\mathbf{x}}_\sigma\rangle_{W_\sigma}\right) v\right](\mathbf{k}, \lambda)\\ =&\sum_{\alpha=1}^{3} 2(\mathbf{G}^{\mathbf{x}}_\sigma)_\alpha(\mathbf{k}, \lambda)\langle (\mathbf{G}^{\mathbf{x}}_\sigma)_\alpha, v\rangle_{W_\sigma} + \|(\mathbf{G}^{\mathbf{x}}_\sigma)_\alpha\|^2_{W_\sigma} v(\mathbf{k}, \lambda).\end{aligned}$$

Note that $\|(\mathbf{G}^{\mathbf{x}}_\sigma)_\alpha\|^2_{W_\sigma}$ is independent of $\mathbf{x}$ by construction of $\mathbf{G}^{\mathbf{x}}_\sigma$. Thus

(4.4.37)

$$\begin{aligned}=&\frac{1}{2m_e^3c^4}\Bigg[2\sum_{i\in I_i, j\in I_j, k\in I_k}\Big\langle T^\sigma(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i))\Psi_0\Big|\\ &\qquad\qquad\Big|\sum_{\alpha=1}^{3}(\mathbf{G}^{\mathbf{x}_k}_\sigma)_\alpha(\mathbf{k},\lambda)\langle(\mathbf{G}^{\mathbf{x}_k}_\sigma)_\alpha, T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0\rangle_{W_\sigma}\Big\rangle_{\mathcal{H}}\\ &+(\sharp I_k)(\sum_{\alpha=1}^{3}\|(\mathbf{G}^{\mathbf{x}}_\sigma)_\alpha\|^2_{W_\sigma})\Big\langle\sum_{i\in I_i}(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i))\Psi_0|(T^\sigma)^2|(\sum_{j\in I_J}\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0\Big\rangle_{\mathcal{H}}\Bigg].\end{aligned}$$

Using the fiber decomposition of T^σ with respect to photon momenta (Lemma 4.2.7) and the definition of the inner product on W_σ, we calculate

$$\begin{aligned}&\langle \mathbf{G}^{\mathbf{x}_k}_\sigma, T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0\rangle_{W_\sigma}\\ =&\sum_{\lambda=1,2}\int_{\Omega_\sigma} d\mathbf{k}\,\overline{\mathbf{G}^{\mathbf{x}_k}(\mathbf{k},\lambda)}\,[T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0](\mathbf{k},\lambda)\\ =&\sum_{\lambda=1,2}\int_{\Omega_\sigma} d\mathbf{k}\,\overline{\mathbf{G}^{\mathbf{x}_k}(\mathbf{k},\lambda)}\,\left(T^\sigma(\mathbf{k})[(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))(\Psi^0_A\otimes\Psi^0_B)](\mathbf{k},\lambda)\right),\end{aligned}$$

where $T^\sigma(\mathbf{k})$ denotes the resulting $\mathbf{k}$-dependent operator (this will depend on the subspace on which T^σ is applied). Note that this expression is still a function of the electronic variables. Since $\Psi_0 \in \mathcal{H}_A \otimes \mathcal{H}_B \otimes \{\Omega\}$, we have $a(\mathbf{G}^{\mathbf{x}}_\sigma)\Psi_0 = 0$, and thus

$$\begin{aligned}&(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))(\Psi^0_A\otimes\Psi^0_B) = (\mathbf{p}_{\mathbf{x}_j}\cdot a^\dagger(\mathbf{G}^{\mathbf{x}_j}_\sigma))(\Psi^0_A\otimes\Psi^0_B)\\ =&c\,C(\mathbf{k})\left(\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_{x_j}(\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_j}(\Psi^0_A\otimes\Psi^0_B))\right).\end{aligned} \tag{4.4.38}$$

Using this, we conclude

$$\begin{aligned}
&\langle \mathbf{G}_\sigma^{\mathbf{x}_k}, T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0\rangle_{W_\sigma}\\
=&c^2\sum_{\lambda=1,2}\int_{\Omega_\sigma} d\mathbf{k}\,|C(\mathbf{k})|^2\mathbf{e}(\mathbf{k},\lambda)\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_k}\left(T^\sigma(\mathbf{k})\left(\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_j(\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_j}(\Psi_A^0\otimes\Psi_B^0))\right)\right),
\end{aligned}$$

and, again using 4.4.38, the fiber decomposition of T^σ and Fubini's theorem, arrive at

$$\begin{aligned}
&\Big\langle T^\sigma(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i))\Psi_0|\sum_{\alpha=1}^3(\mathbf{G}_\sigma^{\mathbf{x}_k})_\alpha(\mathbf{k},\lambda)\langle(\mathbf{G}_\sigma^{\mathbf{x}_k})_\alpha, T^\sigma(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))\Psi_0\rangle_{W_\sigma}\Big\rangle_{\mathcal{H}}\\
=&c^2\Big\langle T^\sigma(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i))\Psi_0\Big|\mathbf{G}_\sigma^{\mathbf{x}_k}\cdot\Big[\sum_{\mu=1,2}\int_{\Omega_\sigma}d\mathbf{k}_2\,|C(\mathbf{k}_2)|^2\mathbf{e}(\mathbf{k}_2,\mu)\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\\
&\qquad\times\left(T^\sigma(\mathbf{k}_2)\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_j(\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}(\Psi_A^0\otimes\Psi_B^0))\right)\right)\Big]\Big\rangle_{\mathcal{H}}\\
=&c^4\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2\,|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\\
&\qquad\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot T^\sigma(\mathbf{k}_1)\left[\mathbf{p}_{\mathbf{x}_i}(\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}(\Psi_A^0\otimes\Psi_B^0))\right]\Big|\\
&\qquad\qquad\Big|\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot T^\sigma(\mathbf{k}_2)\left[\mathbf{p}_{\mathbf{x}_j}(\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}(\Psi_A^0\otimes\Psi_B^0))\right]\right)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
=&c^4\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2\,|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\\
&\qquad\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot T^\sigma(\mathbf{k}_1)\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}((\Psi_A^0\otimes\Psi_B^0))\right]\Big|\\
&\qquad\qquad\Big|\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot T^\sigma(\mathbf{k}_2)\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}((\Psi_A^0\otimes\Psi_B^0))\right]\right)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B},
\end{aligned}$$

where in the last step we have used the Coulomb gauge condition $\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{k}=0$.
To proceed, we need to specify the index sets I_i, I_j and I_k.
Case 1a): $i\in\{1,\dots,Z_A\}$, $k\in\{Z_{A+1},\dots,N\}$, $j\in\{Z_{A+1},\dots,N\}$. This generates the term

$$\begin{aligned}
M_1:=&\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot T^\sigma(\mathbf{k}_1)\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}((\Psi_A^0\otimes\Psi_B^0))\right]|\\
&\qquad\times\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot T^\sigma(\mathbf{k}_2)\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}((\Psi_A^0\otimes\Psi_B^0))\right]\right)\Big\rangle_{\mathcal{H}_A\otimes h_B}\\
=&\Big\langle\Big[\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}\Psi_A^0\right]\Big]\otimes\Psi_B^0))\,|\\
&\qquad\times\Psi_A^0\otimes\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_B+\hbar\omega(\mathbf{k}_2)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}\Psi_B^0\right]\right)\right]\Big\rangle_{\mathcal{H}_A\otimes h_B}\\
=&\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}\Psi_A^0\right]|\Psi_A^0\Big\rangle_{\mathcal{H}_A}\\
&\qquad\times\Big\langle\Psi_B^0|\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_B+\hbar\omega(\mathbf{k}_2)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}\Psi_B^0\right]\right)\Big\rangle_{\mathcal{H}_B}
\end{aligned}$$

$$
\begin{aligned}
=&\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\underbrace{\left\langle (H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}\Psi_A^0\right]|\Psi_A^0\right\rangle_{\mathcal{H}_A}}_{=0}\\
&\times\left\langle\Psi_B^0|\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\left(\mathbf{e}(\mathbf{k}_2,\mu)\cdot(H_B+\hbar\omega(\mathbf{k}_2)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}\Psi_B^0\right]\right)\right\rangle_{\mathcal{H}_B}\\
=&\,0.
\end{aligned}
$$

Case 1b): $i\in\{1,\dots,Z_A\}$, $k\in\{1,\dots,Z_A\}$, $j\in\{Z_{A+1},\dots,N\}$. Analogously to the above, we obtain

$$
\begin{aligned}
M_2:=&\left\langle \mathrm{e}^{\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\mathbf{e}(\mathbf{k}_1,\lambda)\cdot(H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}\Psi_A^0\right]|\Psi_A^0\right\rangle_{\mathcal{H}_A}\\
&\times\underbrace{\left\langle\Psi_B^0|\left((H_B+\hbar\omega(\mathbf{k}_2)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}\Psi_B^0\right]\right)\right\rangle_{\mathcal{H}_B}}_{=0}\cdot\mathbf{e}(\mathbf{k}_2,\mu)\\
=&0.
\end{aligned}
$$

Case 2): $i,j\in\{1,\dots,Z_A\}$, $k\in\{Z_{A+1},\dots,N\}$. In this case the term

$$
\begin{aligned}
M_3:=&\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\left((H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}\Psi_A^0\right]\right)\Big|\\
&\Big|\mathbf{e}(\mathbf{k}_2,\mu)\cdot\left((H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}\Psi_A^0\right]\right)\Big\rangle_{\mathcal{H}_A}\\
&\times\left\langle\Psi_B^0|\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\Psi_B^0\right\rangle_{\mathcal{H}_B}.
\end{aligned}
$$

is generated. Note that the last term is proportional to $\hat{\rho}_B(\mathbf{k}_1-\mathbf{k}_2)$, the Fourier transform of the ground state density of atom B.

Case 3): $i,j\in\{Z_{A+1},\dots,N\}$, $k\in\{1,\dots,Z_A\}$. Analogously to the previous case, we obtain

$$
\begin{aligned}
M_4:=&\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\left((H_B+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_i}\mathbf{p}_{\mathbf{x}_i}\Psi_B^0\right]\right)\Big|\\
&\Big|\mathbf{e}(\mathbf{k}_2,\mu)\cdot\left((H_B+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathrm{e}^{-\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_j}\mathbf{p}_{\mathbf{x}_j}\Psi_B^0\right]\right)\Big\rangle_{\mathcal{H}_B}\\
&\times\left\langle\Psi_A^0|\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\cdot\mathbf{x}_k}\mathrm{e}^{\mathrm{i}\mathbf{k}_2\cdot\mathbf{x}_k}\Psi_A^0\right\rangle_{\mathcal{H}_A}.
\end{aligned}
$$

Now we return to the analysis of (4.4.36). The first term corresponds exactly to case 1) upon setting $\mathbf{x}_i=0$, $\mathbf{x}_j=\mathbf{R}$, $\mathbf{x}_k=0$ (case a)), $\mathbf{x}_k=\mathbf{R}$ (case b)) in the exponentials, so we conclude

$$
2\,\mathrm{Re}\left[\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_\sigma T^\sigma|H'_{\sigma,B}\Psi_0\right]\rangle_{\mathcal{H}}\rangle=0.
$$

The second term corresponds to case 2). Upon setting $\mathbf{x}_i = 0 = \mathbf{x}_j$, $\mathbf{x}_k = \mathbf{R}$ in the exponentials, it becomes

$$\begin{aligned}
&\langle H'_{\sigma,A}\Psi_0|T^\sigma H''_{\sigma,B}T^\sigma|H'_{\sigma,A}\Psi_0\rangle \\
=&\frac{Z_B}{2m_e^3c^4}\Bigg[2c^4\sum_{i,j\in\{1,\dots,Z_A\}}\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2\,|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2 \\
&\qquad\times(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}} \\
&\qquad\times\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\Big((H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathbf{p}_{\mathbf{x}_i}\Psi_A^0\right]\Big)\Big| \\
&\qquad\qquad\Big|\mathbf{e}(\mathbf{k}_2,\mu)\cdot\Big((H_A+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_A^0\}^\perp})^{-1}\left[\mathbf{p}_{\mathbf{x}_j}\Psi_A^0\right]\Big)\Big\rangle_{\mathcal{H}_A} \\
&\quad+(\sum_{\alpha=1}^3\|(\mathbf{G}_\sigma^{\mathbf{x}})_\alpha\|_{W_\sigma}^2)\underbrace{\sum_{i_A,j_A}\Big\langle(\mathbf{p}_{\mathbf{x}_{i_A}}\cdot\mathbf{A}_\sigma(0))\Psi_0|(T^\sigma)^2|(\mathbf{p}_{\mathbf{x}_{j_B}}\cdot\mathbf{A}_\sigma(0))\Psi_0\Big\rangle_{\mathcal{H}}}_{=(m_ec^2)\|T^\sigma H'_{\sigma,A}\Psi_0\|^2}\Bigg]. \qquad (4.4.39)
\end{aligned}$$

For the third term, which corresponds to case 3), we obtain (this time setting $\mathbf{x}_i = \mathbf{x}_j = \mathbf{R}$, $\mathbf{k}_k = 0$)

$$\begin{aligned}
&\langle H'_{\sigma,B}\Psi_0|T^\sigma H''_{\sigma,A}T^\sigma|H'_{\sigma,B}\Psi_0\rangle \\
=&\frac{Z_A}{2m_e^3c^4}\Bigg[2c^4\sum_{i_B,j_B}\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2\,|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2 \\
&\qquad\times(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\mathrm{e}^{-\mathrm{i}(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}} \\
&\qquad\Big\langle\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\Big((H_B+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathbf{p}_{\mathbf{x}_i}\Psi_B^0\right]\Big)\Big| \\
&\qquad\qquad\Big|\mathbf{e}(\mathbf{k}_2,\mu)\cdot\Big((H_B+\hbar\omega(\mathbf{k}_1)_{|\{\Psi_B^0\}^\perp})^{-1}\left[\mathbf{p}_{\mathbf{x}_j}\Psi_B^0\right]\Big)\Big\rangle_{\mathcal{H}_B} \\
&\quad+(\sum_{\alpha=1}^3\|(\mathbf{G}_\sigma^{\mathbf{x}})_\alpha\|_{W_\sigma}^2)\underbrace{\sum_{i_B,j_B}\Big\langle(\mathbf{p}_{\mathbf{x}_{i_B}}\cdot\mathbf{A}_\sigma(\mathbf{R}))\Psi_0|(T^\sigma)^2|(\mathbf{p}_{\mathbf{x}_{j_B}}\cdot\mathbf{A}_\sigma(\mathbf{R}))\Psi_0\Big\rangle_{\mathcal{H}}}_{=(m_ec)^2\|T^\sigma H'_{\sigma,B}\Psi_0\|^2}\Bigg]. \\
&\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad (4.4.40)
\end{aligned}$$

Since the remaining integrands are invariant under the change of variables $(\mathbf{k}_1,\mathbf{k}_2)\mapsto(\pm\mathbf{k}_1,\pm\mathbf{k}_2)$, we can replace both $\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}}$ and $\mathrm{e}^{-\mathrm{i}(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{R}}$ by $\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}$, add (4.4.39) to (4.4.40) and exploit rotation invariance of the ground states Ψ_A^0,Ψ_B^0 and the operators $H_{A,B}$ by applying Lemma 4.2.12, obtaining

(4.3.7)

$$
\begin{aligned}
=&\frac{1}{2m_ec^2}\Big(\sum_{\alpha=1}^{3}\|\,(\mathbf{G}_\sigma^{\mathbf{x_k}})_\alpha\,\|_{W_\sigma}^2\Big)\Big(Z_A\|T^\sigma H'_{\sigma,B}\Psi_0\|^2+Z_B\|T^\sigma H'_{\sigma,A}\Psi_0\|^2\Big)\\
&+\frac{1}{3m_e^3}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\,\Big(\sum_{\lambda,\mu=1,2}(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))^2\Big)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\\
&\qquad\times\Bigg[Z_B\sum_{i,j=1}^{Z_A}\langle\mathbf{p}_i\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1)^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_j\Psi_A^0\rangle_{\mathcal{H}_A}\\
&\qquad\quad+Z_A\sum_{i,j=Z_A+1}^{N}\langle\mathbf{p}_i\Psi_B^0|(H_B+\hbar\omega(\mathbf{k}_1)^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_j\Psi_B^0\rangle_{\mathcal{H}_B}\Bigg].
\end{aligned}
$$

By Lemma A.6.1, $\sum_{\lambda,\mu=1,2}(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))^2=1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2$.

ii) Next we analyze

$$
\begin{aligned}
&(4.3.6)\\
=&2\mathrm{Re}\Bigg[\Big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_{\sigma,B}\Psi_0\Big\rangle+\Big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_\sigma T^\sigma|H''_{\sigma,A}\Psi_0\Big\rangle\\
&+\Big\langle H'_{\sigma,A}\Psi_0|T^\sigma H'_{\sigma,B}T^\sigma|H''_{\sigma,A}\Psi_0\Big\rangle+\Big\langle H'_{\sigma,B}\Psi_0|T^\sigma H'_{\sigma,A}T^\sigma|H''_{\sigma,B}\Psi_0\Big\rangle\Bigg].
\end{aligned}
\tag{4.4.41}
$$

First consider a general term of the form

$$
\frac{2}{2m_e^3c^4}\sum_{i\in I_i,j\in I_j,k\in I_k}\mathrm{Re}\Bigg[\Big\langle T^\sigma[(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i)\Psi_0]|(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))T^\sigma[\mathbf{A}_\sigma(\mathbf{x}_k)^2\Psi_0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}}\Bigg],
$$

with the index sets I_i,I_j,I_k to be specified below. Since $\Psi_0=(\Psi_A^0\otimes\Psi_B^0\otimes\Omega)$ and $a(\mathbf{G}_\sigma^{\mathbf{x}})\Omega=0$ for any $\mathbf{x}\in\mathbb{R}^3$, we have

$$
\begin{aligned}
\mathbf{A}_\sigma(\mathbf{x}_k)^2\Psi_0=&\Big[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})+a(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a(\mathbf{G}_\sigma^{\mathbf{x}_k})+2a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}})\cdot a(\mathbf{G}_\sigma^{\mathbf{x}})+\langle\mathbf{G}_\sigma^{\mathbf{x}},\mathbf{G}_\sigma^{\mathbf{x}}\rangle_{W_\sigma}\Big]\Psi_0\\
=&a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0+(\sum_{\alpha=1}^{3}\|(\mathbf{G}_\sigma^{\mathbf{x}})_\alpha\|_{W_\sigma}^2)\Psi_0,
\end{aligned}
$$

which is a vector in $\mathcal{H}_A\otimes\mathcal{H}_B\otimes(\mathcal{F}_\sigma^{(2)}\oplus\{\Omega\})$. Consequently, only the contribution from the second Fock space level survives under application of the reduced resolvent, i.e. $T^\sigma[\mathbf{A}_\sigma(\mathbf{x}_k)^2\Psi_0]=T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]$. Furthermore, by the invariance properties of T^σ (Lemma 4.2.5) and the structure of the operator $(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))$, we have

$$
T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}_\sigma^{(2)}
$$

and

$$\begin{aligned}
&(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]\\
=&(\mathbf{p}_{\mathbf{x}_j}\cdot a(\mathbf{G}_\sigma^{\mathbf{x}_j}))T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]+(\mathbf{p}_{\mathbf{x}_j}\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_j}))T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]\\
&\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes(\mathcal{F}_\sigma^{(1)}\oplus\mathcal{F}_\sigma^{(3)}).
\end{aligned}$$

On the other hand, by the same arguments,

$$T^\sigma[(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i)\Psi_0]=T^\sigma[(\mathbf{p}_{\mathbf{x}_i}\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_i})\Psi_0]\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}_\sigma^{(1)},$$

so that the mutual orthogonality of Fock space levels implies

$$\begin{aligned}
&\Big\langle T^\sigma[(\mathbf{p}_{\mathbf{x}_i}\cdot\mathbf{A}_\sigma(\mathbf{x}_i)\Psi_0]|(\mathbf{p}_{\mathbf{x}_j}\cdot\mathbf{A}_\sigma(\mathbf{x}_j))T^\sigma[\mathbf{A}_\sigma(\mathbf{x}_k)^2\Psi_0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}}\\
=&\Big\langle T^\sigma[(\mathbf{p}_{\mathbf{x}_i}\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_i})\Psi_0]|(\mathbf{p}_{\mathbf{x}_j}\cdot a(\mathbf{G}_\sigma^{\mathbf{x}_j}))T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}}. \qquad (4.4.42)
\end{aligned}$$

By the definition of the creation operators,

$$\begin{aligned}
&\Big(a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0\Big)(\mathbf{x}_1,\ldots,\mathbf{x}_N,\mathbf{k}_1,\mathbf{k}_2,\lambda,\mu)\\
=&\frac{2}{\sqrt{2}}(\mathbf{G}_\sigma^{\mathbf{x}_k}(\mathbf{k}_1,\lambda)\cdot\mathbf{G}_\sigma^{\mathbf{x}_k}(\mathbf{k}_2,\mu)(\Psi_A^0\otimes\Psi_B^0)(\mathbf{x}_1,\ldots,\mathbf{x}_N).
\end{aligned}$$

On $\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}_\sigma^{(2)}$, T^σ acts as $I\otimes I\otimes\frac{1}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}$ (see Lemma 4.2.5), so that we can use the definition of the annihilation operators to calculate

$$\begin{aligned}
&(\mathbf{p}_{\mathbf{x}_j}\cdot a(\mathbf{G}_\sigma^{\mathbf{x}_j}))T^\sigma[a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_k})\Psi_0]\\
=&\sqrt{2}\sqrt{2}\mathbf{p}_{\mathbf{x}_j}(\Psi_A^0\otimes\Psi_B^0)\\
&\cdot\Big[\sum_{\mu=1,2}\int_{\Omega_\sigma}d\mathbf{k}_2\frac{1}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}\overline{\mathbf{G}^{\mathbf{x}_j}(\mathbf{k}_2,\mu)}\Big(\mathbf{G}^{\mathbf{x}_k}(\mathbf{k}_1,\lambda)\cdot\mathbf{G}^{\mathbf{x}_k}(\mathbf{k}_2,\mu)\Big)\Big].
\end{aligned}$$

Now using the fiber decomposition of T^σ (with respect to photon momenta) on $\mathcal{F}_\sigma^{(1)}$ (Lemma 4.2.7), the definition of the coupling functions $\mathbf{G}^{\mathbf{x}}$ and Fubini's theorem, we obtain

$$\begin{aligned}
&(4.4.42)\\
=&2c^4\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2\,\frac{|C(\mathbf{k}_1)|^2\,C(\mathbf{k}_2)|^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\,(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\\
&\Big\langle T^\sigma(\mathbf{k}_1)[\mathrm{e}^{-\mathrm{i}\mathbf{k}_1\mathbf{x}_i}\,\big(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{\mathbf{x}_i}(\Psi_A^0\otimes\Psi_B^0)\big)\Big|\\
&\qquad\Big|\mathrm{e}^{\mathrm{i}\mathbf{k}_2\mathbf{x}_j}\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{x}_k}\,\big(\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{\mathbf{x}_j}(\Psi_A^0\otimes\Psi_B^0)\big)]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}, \qquad (4.4.43)
\end{aligned}$$

where $T^\sigma(\mathbf{k}_1)$ denotes the resulting $\mathbf{k}$-dependent operator (which depends on the subspace of $\mathcal{H}_A\otimes\mathcal{H}_B$ to which T^σ is applied). Note that we have also used the Coulomb gauge condition $\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{k}=0$.

A case analysis similar to the above shows that the only cases which occur in (4.3.6) and in which (4.4.43) does not vanish are
1) $i,j \in \{1,\dots,Z_A\}$, $k \in \{Z_{A+1},\dots,N\}$, which corresponds to the third term in (4.4.41) upon setting $\mathbf{x}_i = \mathbf{x}_j = 0$, $\mathbf{x}_k = \mathbf{R}$ in the exponentials, and
2) $i,j \in \{Z_{A+1},\dots,N\}$, $k \in \{1,\dots,Z_A\}$, corresponding to the fourth term in (4.4.41) upon setting $\mathbf{x}_i = \mathbf{x}_j = \mathbf{R}$, $\mathbf{x}_k = 0$ in the exponentials. We conclude

$$
\begin{aligned}
&(4.3.6)\\
=&\frac{2}{m_e^3}\mathrm{Re}\Bigg[\sum_{\lambda,\mu=1,2}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2\, \frac{|C(\mathbf{k}_1)|^2\, C(\mathbf{k}_2)|^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\,(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu))\\
&\qquad\times\Bigg[Z_B \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\sum_{i_A,j_A}\Big\langle \mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{\mathbf{x}_{i_A}}\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{\mathbf{x}_{j_A}}\Psi_A^0\Big\rangle_{\mathcal{H}_A}\\
&\qquad+Z_A\mathrm{e}^{\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\sum_{i_B,j_B}\Big\langle \mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{p}_{\mathbf{x}_{i_B}}\Psi_B^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\mathbf{e}(\mathbf{k}_2,\mu)\cdot\mathbf{p}_{\mathbf{x}_{j_B}}\Psi_B^0\Big\rangle_{\mathcal{H}_B}\Bigg]\Bigg].
\end{aligned}
$$

Using rotation invariance of the ground states Ψ_A^0, Ψ_B^0 and the operators $H_{A,B}$ (Lemma 4.2.12), the identity $\sum_{\lambda,\mu=1,2}(\mathbf{e}(\mathbf{k}_1,\lambda)\cdot\mathbf{e}(\mathbf{k}_2,\mu)) = 1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2$ from Lemma A.6.1 and the fact that as above, we can replace $\mathbf{k}_i$ by $-\mathbf{k}_i$ in the exponentials, yields

$$
\begin{aligned}
&(4.3.6)\\
=&\frac{2}{3m_e^3}\mathrm{Re}\Bigg[\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2\,\frac{|C(\mathbf{k}_1)|^2\, C(\mathbf{k}_2)|^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\\
&\qquad\times\Bigg[Z_B\sum_{i_A,j_A}\Big\langle \mathbf{p}_{\mathbf{x}_{i_A}}\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\mathbf{p}_{\mathbf{x}_{j_A}}\Psi_A^0\Big\rangle_{\mathcal{H}_A}\\
&\qquad\quad+Z_A\sum_{i_B,j_B}\Big\langle \mathbf{p}_{\mathbf{x}_{i_B}}\Psi_B^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\mathbf{p}_{\mathbf{x}_{j_B}}\Psi_B^0\Big\rangle_{\mathcal{H}_B}\Bigg]\Bigg].
\end{aligned}
$$

So far we have shown that

$$
\begin{aligned}
&(4.3.6)+(4.3.7)\\
=&\frac{2}{3m_e^3}\mathrm{Re}\Bigg[\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2\frac{|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\qquad\times\Bigg[Z_B\sum_{i,j=1}^{Z_A}\langle\mathbf{p}_i\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}|\mathbf{p}_j\Psi_A^0\rangle\\
&\qquad\quad+Z_A\sum_{i,j=Z_A+1}^{N}\langle\mathbf{p}_i\Psi_B^0)|H_B+\hbar\omega(\mathbf{k}_1))^{-1}|\mathbf{p}_j\Psi_B^0\rangle\Bigg]\Bigg] \qquad (4.4.44)\\
&+\frac{1}{2m_ec^2}\Big(\sum_{\alpha=1}^{3}\|\,(\mathbf{G}_\sigma^{\mathbf{x_k}})_\alpha\,\|_{W_\sigma}^2\Big)\Big(Z_A\|T^\sigma H'_{\sigma,B}\Psi_0\|^2+Z_B\|T^\sigma H'_{\sigma,A}\Psi_0\|^2\Big) \qquad (4.4.45)
\end{aligned}
$$

$$+\frac{1}{3m_e^3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\,(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}$$
$$\times\Bigg[Z_B\sum_{i,j=1}^{Z_A}\langle\mathbf{p}_i\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1)^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_j\Psi_A^0\rangle_{\mathcal{H}_A}$$
$$+Z_A\sum_{i,j=Z_A+1}^{N}\langle\mathbf{p}_i\Psi_B^0|(H_B+\hbar\omega(\mathbf{k}_1)^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_j\Psi_B^0\rangle_{\mathcal{H}_B}\Bigg]. \tag{4.4.46}$$

Remark 4.4.6. Since the atomic ground states Ψ_A^0 and Ψ_B^0 are chosen to be real functions, the states $\mathbf{x}_i^\alpha\Psi_{A,B}^0$ are also real, and so are any states the form $A(x_i^\alpha\Psi_{A,B}^0)$ with a real operator A. On the other hand, states of the form $A(\mathbf{p}_i^\alpha\Psi_{A,B}^0)$ are purely imaginary. (We call a vector real if it is an eigenfunction of the involution $\psi\mapsto\overline{\psi}$ corresponding to the eigenvalue 1. Analogously, a vector is called purely imaginary if it is an eigenfunction corresponding to the eigenvalue -1. An operator is called real if it commutes with this involution.)
Since all the resolvents occurring in the matrix elements are real operators, all occurring matrix elements are real. (For the matrix elements involving $\mathbf{p}_i^\alpha\Psi_{A,B}^0$ this follows since the inner product of two purely imaginary states is real.) Furthermore, they are invariant under $(\mathbf{k}_1,\mathbf{k}_2)\to(-\mathbf{k}_1,-\mathbf{k}_2)$. The remaining integrands in all integrals under consideration are of the form $f(\mathbf{k}_1,\mathbf{k}_2)\exp(-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R})$, with f real and satisfying $f(-\mathbf{k}_1,-\mathbf{k}_2)=f(\mathbf{k}_1,\mathbf{k}_2)$. It follows that all integrals are real, so that any 'real parts' in front of them can be dropped. (Note that the domain of integration, $\{\omega(\mathbf{k}_1)\geq\sigma\}\times\{\omega(\mathbf{k}_2)\geq\sigma\}$, is invariant under the change of coordinates $(\mathbf{k}_1,\mathbf{k}_2)\to(-\mathbf{k}_1,-\mathbf{k}_2)$)

Combining terms (4.4.46) and (4.4.44). According to remark 4.4.6, the real part in front of (4.4.44) can be dropped, so that applying the commutator relation (recall the convention $H_{A,B}=H_{A,B}-E_{A,B}^0$)

$$\mathbf{p}_i\psi_{A,B}^0=\frac{\mathrm{i}m_e}{\hbar}[H_{A,B},x_i]\psi_{A,B}^0=\frac{\mathrm{i}m_e}{\hbar}H_{A,B}(\mathbf{x}_i\psi_{A,B}^0)$$

from Lemma A.7.1 to the terms in (4.4.46)+(4.4.44), one obtains

$$\begin{aligned}&(4.4.46)+(4.4.44)\\ =&\frac{1}{3m_e^3}\left(\frac{m_e}{\hbar}\right)^2\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\,(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\\ &\times\Bigg[\frac{2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}\Bigg(Z_B\sum_{i,j=1}^{Z_A}\langle\mathbf{x}_i\Psi_A^0|H_A(H_A+\hbar\omega(\mathbf{k}_1))^{-1}H_A|\mathbf{x}_j\Psi_A^0\rangle\\ &\quad+Z_A\sum_{i,j=1}^{Z_B}\langle\mathbf{x}_i\Psi_B^0|H_B(H_B+\hbar\omega(\mathbf{k}_1))^{-1}H_B|\mathbf{x}_j\Psi_B^0\rangle\Bigg)\\ &\quad+Z_B\sum_{i,j=1}^{Z_A}\langle\mathbf{x}_i\Psi_A^0|H_A(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\omega(\mathbf{k}_2))^{-1}H_A|\mathbf{x}_j\Psi_A^0\rangle\end{aligned}$$

$$
\begin{aligned}
&+ Z_A \sum_{i,j=1}^{Z_B} \langle \mathbf{x}_i \Psi_B^0 | H_B (H_B + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \omega(\mathbf{k}_2))^{-1} H_B | \mathbf{x}_j \Psi_B^0 \rangle \Bigg] \\
=&\frac{1}{3m_e\hbar^2} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 \, |C(\mathbf{k}_2)|^2 \, (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\mathbf{R}} \\
&\times \Bigg[Z_B \sum_{i,j=1}^{Z_A} \Big\langle \mathbf{x}_i \Psi_A^0 | \frac{2}{\hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))} H_A \Big(I - \hbar\omega(\mathbf{k}_1)(H_A + \hbar\omega(\mathbf{k}_1))^{-1} \Big) \\
&\quad + \Big(I - \hbar\omega(\mathbf{k}_1)(H_A + \hbar\omega(\mathbf{k}_1))^{-1} \Big) \Big(I - \hbar\omega(\mathbf{k}_2)(H_A + \hbar\omega(\mathbf{k}_2))^{-1} \Big) | \mathbf{x}_j \Psi_A^0 \Big\rangle \\
&\quad + Z_A \sum_{i,j=1}^{Z_B} \Big\langle \mathbf{x}_i \Psi_B^0 | \frac{2}{\hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))} H_B \Big(I - \hbar\omega(\mathbf{k}_1)(H_B + \hbar\omega(\mathbf{k}_1))^{-1} \Big) \\
&\quad + I - \hbar\omega(\mathbf{k}_1)(H_B + \hbar\omega(\mathbf{k}_1))^{-1} - \hbar\omega(\mathbf{k}_2)(H_B + \hbar\omega(\mathbf{k}_2))^{-1} \\
&\quad + (\hbar\omega(\mathbf{k}_1))(\hbar\omega(\mathbf{k}_2))(H_B + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} | \mathbf{x}_j \Psi_B^0 \Big\rangle \Bigg] \\
=&\frac{1}{3m_e\hbar^2} \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 \, |C(\mathbf{k}_2)|^2 \, (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\mathbf{R}} \\
&\times \Bigg[(Z_A \alpha_M^B + Z_B \alpha_M^A) - \frac{2\omega(\mathbf{k}_1)}{\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)} (Z_A \alpha_M^B + Z_B \alpha_M^A) \\
&\quad + \frac{2}{\hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))} (Z_A S_B + S_A Z_B) \\
&\quad + \frac{2(\hbar\omega(\mathbf{k}_1))^2}{\hbar(\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2))} \Big(Z_A \alpha_E^B(\mathbf{k}_1) + \alpha_E^A(\mathbf{k}_1) Z_B \Big) \\
&\quad - 2\hbar\omega(\mathbf{k}_1) \Big(Z_A \alpha_E^B(\mathbf{k}_1) + \alpha_E^A(\mathbf{k}_1) Z_B \Big) \\
&\quad + (\hbar\omega(\mathbf{k}_1))(\hbar\omega(\mathbf{k}_2)) \Bigg[Z_B \sum_{i,j=1}^{Z_A} \langle \mathbf{x}_i \Psi_A^0 | (H_A + \hbar\omega(\mathbf{k}_1))^{-1} (H_A + \hbar\omega(\mathbf{k}_2))^{-1} | \mathbf{x}_j \Psi_A^0 \rangle \\
&\quad + Z_A \sum_{i,j=1}^{Z_B} \langle \mathbf{x}_i \Psi_B^0 | (H_B + \hbar\omega(\mathbf{k}_1))^{-1} (H_B + \hbar\omega(\mathbf{k}_2))^{-1} | \mathbf{x}_j \Psi_B^0 \rangle \Bigg] \Bigg],
\end{aligned}
$$

where we have used Lemma A.10.1. Since the remaining integrand is invariant under the exchange $\mathbf{k}_1 \leftrightarrow \mathbf{k}_2$, and since $\Omega_\sigma \times \Omega_\sigma$ is composed of twice the same subset of $\mathbb{R}^3$, we can replace

$$
\int d\mathbf{k}_1 d\mathbf{k}_2 \ldots \frac{2\omega(\mathbf{k}_1)}{\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)} = \int d\mathbf{k}_1 d\mathbf{k}_2 \ldots \frac{\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)} = \int d\mathbf{k}_1 d\mathbf{k}_2 \ldots 1,
$$

so that the first two terms of $[\ldots]$ (which contain integrands of homogeneity -2) add up to zero, and we are left with

$$
\begin{aligned}
&(4.4.46) + (4.4.44)\\
=&\frac{1}{3m_e\hbar^2}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\,(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\\
&\quad\times\Bigg[\frac{2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}(Z_A S_B + S_A Z_B)\\
&\qquad+\frac{2(\hbar\omega(\mathbf{k}_1))^2}{\hbar(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}\Big(Z_A\alpha_E^B(\mathbf{k}_1)+\alpha_E^A(\mathbf{k}_1)Z_B\Big)\\
&\qquad-2\hbar\omega(\mathbf{k}_1)\Big(Z_A\alpha_E^B(\mathbf{k}_1)+\alpha_E^A(\mathbf{k}_1)Z_B\Big)\\
&\qquad+(\hbar\omega(\mathbf{k}_1))(\hbar\omega(\mathbf{k}_2))\Bigg[Z_B\sum_{i,j=1}^{Z_A}\langle\mathbf{x}_i\Psi_A^0|(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{x}_j\Psi_A^0\rangle\\
&\qquad+Z_A\sum_{i,j=1}^{Z_B}\langle\mathbf{x}_i\Psi_B^0|(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}|\mathbf{x}_j\Psi_B^0\rangle\Bigg]\Bigg].
\end{aligned}
$$

Now by Lemma A.9.1, $Z_{A,B} = (2m_e)/(3\hbar^2)S_{A,B}$, so that - recalling the definition of the dynamic polarizabilities $\alpha_E^{A,B}(\mathbf{k})$ from Theorem 2.8.4 - we obtain

$$
\begin{aligned}
&(4.4.46) + (4.4.44)\\
=&\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\,(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\mathbf{R}}\\
&\quad\times\Bigg[\frac{8}{9\hbar^5(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}S_A S_B\\
&\qquad+\frac{2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}{9\hbar^2}\Bigg[\Big\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle S_B\\
&\qquad+S_A\Big\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle\Bigg]\\
&\qquad+\frac{4}{9\hbar^3}(S_A\alpha_E^B(\mathbf{k}_1)+\alpha_E^A(\mathbf{k}_1)S_B)\left(\frac{\omega(\mathbf{k}_1)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-\omega(\mathbf{k}_1)\right)\Bigg],
\end{aligned}
$$

finishing the proof.

4.4.5 Proof of Lemma 4.4.4

By the definition (2.4.2) of the perturbation operator H'_σ, we have

$$
\begin{aligned}
(4.3.8) =&2\mathrm{Re}\left[\langle H'_\sigma\Psi_0|T^\sigma H'_\sigma T^\sigma|Q_R\Psi_0\rangle\right]+\langle H'_\sigma\Psi_0|T^\sigma Q_R T^\sigma|H'_\sigma\Psi_0\rangle\\
=&\left(-\frac{1}{m_e c}\right)^2\sum_{i,j=1}^{N}\Bigg[2\mathrm{Re}\left[\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma(\mathbf{p}_j\cdot\mathbf{A}_\sigma)T^\sigma|Q_R\Psi_0\rangle\right]\\
&\qquad+\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma Q_R T^\sigma|(\mathbf{p}_j\cdot\mathbf{A}_\sigma)\Psi_0\rangle\Bigg].
\end{aligned}
$$

The sum over the electron coordinates i and j splits into four contributions:

$$\sum_{i,j} = \sum_{\substack{i \in \{1,\dots,Z_A\},\\ j \in \{1,\dots,Z_A\}}} + \sum_{\substack{i \in \{Z_A+1,\dots,N\},\\ j \in \{Z_A+1,\dots,N\}}} + \sum_{\substack{i \in \{1,\dots,Z_A\},\\ j \in \{Z_A+1,\dots,N\}}} + \sum_{\substack{i \in \{Z_A+1,\dots,N\},\\ j \in \{1,\dots,Z_A\}}} .$$

To simplify notation, we will denote these cases by using indices (i_A, j_A), (i_B, j_B) et cetera. The proof consists of two parts. In the first part, we will investigate the third and fourth sum and show that

$$T_1 := \frac{1}{m_e^2 c^2} \left(\sum_{i_A, j_B} + \sum_{i_B, j_A} \right) \Big[2\mathrm{Re}\left[\langle (\mathbf{p}_i \cdot \mathbf{A}_\sigma)\Psi_0 | T^\sigma (\mathbf{p}_j \cdot \mathbf{A}_\sigma) T^\sigma | Q_R \Psi_0 \rangle \right] + \langle (\mathbf{p}_i \cdot \mathbf{A}_\sigma)\Psi_0 | T^\sigma Q_R T^\sigma | (\mathbf{p}_j \cdot \mathbf{A}_\sigma)\Psi_0 \rangle \Big] = M_B(\mathbf{R}, \sigma).$$

In the second part we will establish the corresponding identity

$$T_2 := \frac{1}{m_e^2 c^2} \left(\sum_{i_A, j_A} + \sum_{i_B, j_B} \right) \Big[2\mathrm{Re}\left[\langle (\mathbf{p}_i \cdot \mathbf{A}_\sigma)\Psi_0 | T^\sigma (\mathbf{p}_j \cdot \mathbf{A}_\sigma) T^\sigma | Q_R \Psi_0 \rangle \right] + \langle (\mathbf{p}_i \cdot \mathbf{A}_\sigma)\Psi_0 | T^\sigma Q_R T^\sigma | (\mathbf{p}_j \cdot \mathbf{A}_\sigma)\Psi_0 \rangle \Big] = M_A(\mathbf{R}, \sigma).$$

Part i) First consider

$$\sum_{i_A, j_B} 2\mathrm{Re}\Big[\Big\langle (\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))\Psi_0 | T^\sigma (\mathbf{p}_{j_B} \cdot \mathbf{A}_\sigma(\mathbf{R})) T^\sigma | Q_R \Psi_0 \Big\rangle \Big] = \sum_{i_A, j_B} 2\mathrm{Re}\Big[\Big\langle T^\sigma (\mathbf{p}_{j_B} \cdot \mathbf{A}_\sigma(\mathbf{R})) T^\sigma (\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))\Psi_0 | Q_R \Psi_0 \Big\rangle \Big].$$

By the structure of Ψ_0, the form of the vector potential $\mathbf{A}_\sigma(\mathbf{x}) = a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma) + a(\mathbf{G}^{\mathbf{x}}_\sigma)$ and Lemma A.8.1, we have

$$(\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))\Psi_0 = (\mathbf{p}_{i_A} \cdot a^\dagger(\mathbf{G}^0_\sigma))\Psi_0 = \mathbf{p}_{i_A} \Psi^0_A \otimes \Psi^0_B \otimes a^\dagger(\mathbf{G}^0_\sigma)\Omega \in \{\Psi^0_A\}^\perp \widehat{\otimes} \{\Psi^0_B\} \widehat{\otimes} \mathcal{F}^{(1)}_\sigma,$$

and thus the invariance properties of T^σ (Lemma 4.2.5) imply that

$$T^\sigma (\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))\Psi_0 = T^\sigma_A \Big(\mathbf{p}_{i_A} \Psi^0_A \otimes a^\dagger(\mathbf{G}^0_\sigma)\Omega \Big) \otimes \Psi^0_B \in \{\Psi^0_A\}^\perp \widehat{\otimes} \{\Psi^0_B\} \widehat{\otimes} \mathcal{F}^{(1)}_\sigma.$$

By the same arguments,

$$\begin{aligned}
&T^\sigma(\mathbf{p}_{j_B}\cdot\mathbf{A}_\sigma(\mathbf{R}))T^\sigma(\mathbf{p}_{i_A}\cdot\mathbf{A}_\sigma(0))\Psi_0\\
=&T^\sigma\Big[\mathbf{p}_{j_B}\cdot\Big(a^\dagger(\mathbf{G}^{\mathbf{R}}_\sigma)+a(\mathbf{G}^{\mathbf{R}}_\sigma)\Big)\Big[T^\sigma_A\Big(\mathbf{p}_{i_A}\Psi^0_A\otimes a^\dagger(\mathbf{G}^0_\sigma)\Omega\Big)\otimes\Psi^0_B\Big]\Big]\\
&\in\{\Psi^0_A\}^\perp\widehat{\otimes}\{\Psi^0_B\}^\perp\widehat{\otimes}\Big(\mathcal{F}^{(0)}_\sigma\oplus\mathcal{F}^{(2)}_\sigma\Big).
\end{aligned}$$

On the other hand, $Q_R\Psi_0\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes\{\Omega\}$, so that the mutual orthogonality of Fock space levels implies that

$$\Big\langle T^\sigma\Big[\mathbf{p}_{j_B}\cdot a^\dagger(\mathbf{G}^{\mathbf{R}}_\sigma)\Big[T^\sigma_A\Big(\mathbf{p}_{i_A}\Psi^0_A\otimes a^\dagger(\mathbf{G}^0_\sigma)\Omega\Big)\otimes\Psi^0_B\Big]\Big]|Q_R\Psi_0\Big\rangle=0.$$

For the remaining contribution we use the invariance properties of T^σ, the fiber decomposition of T^σ_A (Lemma 4.2.7) and the definition of the annihilation operator $a(\mathbf{G}^{\mathbf{R}}_\sigma)$ to obtain

$$\begin{aligned}
&\Big\langle T^\sigma\Big[\mathbf{p}_{j_B}\cdot a(\mathbf{G}^{\mathbf{R}}_\sigma)\Big[T^\sigma_A\Big(\mathbf{p}_{i_A}\Psi^0_A\otimes a^\dagger(\mathbf{G}^0_\sigma)\Omega\Big)\otimes\Psi^0_B\Big]\Big]|Q_R\Psi_0\Big\rangle_{\mathcal{H}}\\
=&\Big\langle(\overline{H_A+H_B})^{-1}\Big[\sum_{\lambda=1,2}\int_{\Omega_\sigma}d\mathbf{k}\Big(\mathbf{p}_{j_B}\Psi^0_B\cdot\overline{\mathbf{G}^{\mathbf{R}}_\sigma(\mathbf{k},\lambda)}\Big)\Big((H_A+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{i_A}\Psi^0_A\cdot\mathbf{G}^0_\sigma(\mathbf{k},\lambda)]\Big)\Big]\Big|\\
&\Big|Q_R(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B},
\end{aligned}$$

which by Fubini's theorem and the definition of the coupling functions $\mathbf{G}^{\mathbf{x}}_\sigma(\mathbf{k},\lambda)$ equals

$$\begin{aligned}
&c^2\sum_{\lambda=1,2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\Big\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}[\big(\mathbf{p}_{i_A}\Psi^0_A\cdot\mathbf{e}(\mathbf{k},\lambda)\big)]\otimes\big(\mathbf{p}_{j_B}\Psi^0_B\cdot\mathbf{e}(\mathbf{k},\lambda)\big)\Big|(\overline{H_A+H_B})^{-1}\Big|\\
&\qquad\qquad\Big|Q_R(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}.
\end{aligned}$$

Interchanging the roles of the indices, one finds

$$\begin{aligned}
&2\mathrm{Re}\Big[\Big\langle(\mathbf{p}_{i_B}\cdot\mathbf{A}_\sigma(\mathbf{R}))\Psi_0|T^\sigma(\mathbf{p}_{j_A}\cdot\mathbf{A}_\sigma(0)T^\sigma|Q_R\Psi_0\Big\rangle\Big]\\
=&c^2\sum_{\lambda=1,2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\Big\langle\big(\mathbf{p}_{j_A}\Psi^0_A\cdot\mathbf{e}(\mathbf{k},\lambda)\big)\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}[\big(\mathbf{p}_{i_B}\Psi^0_B\cdot\mathbf{e}(\mathbf{k},\lambda)\big)]\Big|(\overline{H_A+H_B})^{-1}\Big|\\
&\qquad\qquad\Big|Q_R(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
=&c^2\sum_{\lambda=1,2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\Big\langle\big(\mathbf{p}_{j_A}\Psi^0_A\cdot\mathbf{e}(\mathbf{k},\lambda)\big)\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}[\big(\mathbf{p}_{i_B}\Psi^0_B\cdot\mathbf{e}(\mathbf{k},\lambda)\big)]\Big|(\overline{H_A+H_B})^{-1}\Big|\\
&\qquad\qquad\Big|Q_R(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B},
\end{aligned}$$

where we have used the fact that $C(\mathbf{k})$, $\mathbf{e}(\mathbf{k},\lambda)$ and $\omega(\mathbf{k})$ are invariant under the change of variables $\mathbf{k}\mapsto-\mathbf{k}$. By arguments completely analogous to the ones just given (and the fact that $T^\sigma Q_R T^\sigma$ is symmetric), one establishes

$$
\begin{aligned}
&\frac{1}{m_e^2c^2}\left(\sum_{i_A,j_B}+\sum_{i_B,j_A}\right)\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma Q_R T^\sigma|(\mathbf{p}_j\cdot\mathbf{A}_\sigma)\Psi_0\rangle\\
=&\frac{2}{m_e^2}\mathrm{Re}\Bigg[\sum_{i_A,j_B}\sum_{\lambda=1,2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\times\Bigg[\Big\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{i_A}\cdot\mathbf{e}(\mathbf{k},\lambda)\Psi_A^0]\otimes\Psi_B^0\Big|Q_R\Big|\\
&\qquad\qquad\Big|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_{j_B}\Psi_B^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg].
\end{aligned}
$$

Summarizing, we have found

$$
\begin{aligned}
T_1=&\frac{1}{m_e^2c^2}\left(\sum_{i_A,j_B}+\sum_{i_B,j_A}\right)\Bigg[2\mathrm{Re}\left[\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma(\mathbf{p}_j\cdot\mathbf{A}_\sigma)T^\sigma|Q_R\Psi_0\rangle\right]\\
&\qquad+\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma Q_R T^\sigma|(\mathbf{p}_j\cdot\mathbf{A}_\sigma)\Psi_0\rangle\Bigg]\\
=&\frac{2}{m_e^2}\mathrm{Re}\Bigg[\sum_{i_A,j_B}\sum_{\lambda=1,2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\times\Bigg[\Big\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{i_A}\cdot\mathbf{e}(\mathbf{k},\lambda)\Psi_A^0]\otimes\Psi_B^0|Q_R|\\
&\qquad\qquad\Big|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_{j_B}\Psi_B^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&\qquad\quad+\Big\langle\left((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)\\
&\qquad\qquad\times\Big[(\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_{i_A}\Psi_A^0)\otimes(\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_{j_B}\Psi_B^0)\Big]\Big|\\
&\qquad\qquad\Big|(\overline{H_A+H_B})^{-1}|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg]\Bigg].
\end{aligned}
$$

Applying Lemma A.6.1, we can carry out the summation over the polarizabilities:

$$
\begin{aligned}
T_1=&\frac{2}{m_e^2}\mathrm{Re}\Bigg[\sum_{i_A,j_B}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\times\Bigg[\Big\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{i_A}\Psi_A^0]\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\Big|\\
&\qquad\qquad\Big|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{j_B}\Psi_B^0]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Big]\\
&\qquad+\Big\langle\left((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)\Big[\mathbf{p}_{i_A}\Psi_A^0(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{p}_{j_B}\Psi_B^0\Big]\Big|\\
&\qquad\Big|(\overline{H_A+H_B})^{-1}|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg]\Bigg].
\end{aligned}
$$

Next we note that by Proposition 2.5.1, $\mathbf{v}_A = \sum_{i_A} \mathbf{x}_{i_A} \Psi_A^0 \in H^2(\mathbb{R}^{3Z_A})$ and $\mathbf{v}_B = \sum_{j_B} \mathbf{x}_{j_B} \Psi_B^0 \in H^2(\mathbb{R}^{3Z_B})$, so we can apply the commutator relation

$$\mathbf{p}_{i_{A,B}} \Psi_{A,B}^0 = \frac{\mathrm{i} m_e}{\hbar} H_{A,B} \left(\mathbf{x}_{i_{A,B}} \Psi_{A,B}^0 \right)$$

from Lemma A.7.1, as well as the relations

$$(H_{A,B} + \hbar\omega(\mathbf{k}))^{-1} H_{A,B} = I - \hbar\omega(\mathbf{k})(H_{A,B} + \hbar\omega(\mathbf{k}))^{-1},$$

which hold on $D(H_{A,B})$, yielding

$$\begin{aligned}
T_1 = \frac{2}{\hbar^2} \mathrm{Re} \Bigg[\int_{\Omega_\sigma} d\mathbf{k} |C(\mathbf{k})|^2 \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} \\
\times \Bigg[(\mathrm{i}(-\mathrm{i}) \Big\langle (H_A + \hbar\omega(\mathbf{k}))^{-1} [H_A \mathbf{v}_A] \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R \Big| \\
\Big| \Psi_A^0 \otimes (H_B + \hbar\omega(\mathbf{k}))^{-1} [H_B \mathbf{v}_B] \Big\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \\
+ (-\mathrm{i})^2 \Big\langle \left((H_A + \hbar\omega(\mathbf{k}))^{-1} + (H_B + \hbar\omega(\mathbf{k}))^{-1} \right) \\
\times \Big[(H_A \mathbf{v}_A)(1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}})(H_B \mathbf{v}_B) \Big] \Big| \\
\Big| (\overline{H_A + H_B})^{-1} | Q_R (\Psi_A^0 \otimes \Psi_B^0) \Big\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \Bigg] \Bigg] \\
= \frac{2}{\hbar^2} \mathrm{Re} \Bigg[\int_{\Omega_\sigma} d\mathbf{k} |C(\mathbf{k})|^2 \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} \\
\Bigg[\Big\langle (I - \hbar\omega(\mathbf{k})(H_A + \hbar\omega(\mathbf{k}))^{-1}) \mathbf{v}_A] \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R \Big| \\
\Big| \Psi_A^0 \otimes (I - \hbar\omega(\mathbf{k})(H_B + \hbar\omega(\mathbf{k}))^{-1}) \mathbf{v}_B \Big\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \\
- \Big\langle \left((I - (H_A + \hbar\omega(\mathbf{k}))^{-1}) \mathbf{v}_A \right) (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}})(H_B \mathbf{v}_B) | (\overline{H_A + H_B})^{-1} \Big| \\
\Big| Q_R (\Psi_A^0 \otimes \Psi_B^0) \Big\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \\
- \Big\langle (H_A \mathbf{v}_A)(1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) \left((I - (H_B + \hbar\omega(\mathbf{k}))^{-1}) \mathbf{B}_A \right) | (\overline{H_A + H_B})^{-1} \Big| \\
\Big| Q_R (\Psi_A^0 \otimes \Psi_B^0) \Big\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \Bigg] \Bigg].
\end{aligned}$$

To simplify notation, set $v := \mathbf{v}_A (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) \mathbf{v}_B$ and $u := Q_R (\Psi_A^0 \otimes \Psi_B^0)$. Then

$$
\begin{aligned}
T_1 =& \frac{2}{\hbar^2}\mathrm{Re}\Bigg[\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2 \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} \\
&\times \Bigg[\Big\langle \mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} && (4.4.47)\\
&\quad -\hbar\omega(\mathbf{k})\Big\langle[(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} && (4.4.48)\\
&\quad -\hbar\omega(\mathbf{k})\Big\langle\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} && (4.4.49)\\
&\quad +(\hbar\omega(\mathbf{k}))^2\Big\langle[(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\big| \\
&\qquad\qquad \big|\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
&\quad -\Big\langle(H_A\otimes I+I\otimes H_B)v|(\overline{H_A+H_B})^{-1}|u\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} && (4.4.50)\\
&\quad +\hbar\omega(\mathbf{k})\Big\langle\big((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes H_B+H_A\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,v\big| \\
&\qquad\qquad \big|(\overline{H_A+H_B})^{-1}|u\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg]\Bigg]. && (4.4.51)
\end{aligned}
$$

Note that since $v\in D(H_A)\widehat{\otimes}D(H_B)$ and $\overline{H_A+H_B}=H_A\otimes I+I\otimes H_B$ on $D(H_A)\widehat{\otimes}D(H_B)$, we have

$$(4.4.50) = -\langle \mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B|Q_R(\Psi_A^0\otimes\Psi_B^0)\rangle = -(4.4.47),$$

where for the last identity we have used that $\sum_{j_B}\mathbf{x}_{j_B}$ and Q_R are real multiplication operators.

To further analyze (4.4.51), note that $((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I)v$ and $(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1})v$ both lie in $D(H_A)\widehat{\otimes}D(H_B)$, which allows us to calculate

$$
\begin{aligned}
(4.4.51) =& \hbar\omega(\mathbf{k})\Big\langle(\overline{H_A+H_B})^{-1}\Big[(H_A+H_B-H_A)((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I) \\
&\qquad +(H_A+H_B-H_B)(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1})\Big]v|u\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B} \\
=& \hbar\omega(\mathbf{k})\Big\langle\big((H_A+\omega(\mathbf{k}))^{-1}\otimes I+I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,v|u\Big\rangle \\
&-\hbar\omega(\mathbf{k})\Big\langle(\overline{H_A+H_B})^{-1}\Big[(\underbrace{H_A(H_A+\hbar\omega(\mathbf{k}))^{-1}}_{=I-\hbar\omega(\mathbf{k})(H_A+\hbar\omega(\mathbf{k}))^{-1}}\otimes I) \\
&\qquad +(I\otimes\underbrace{H_B(H_B+\hbar\omega(\mathbf{k}))^{-1}}_{=I-\hbar\omega(\mathbf{k})(H_B+\hbar\omega(\mathbf{k}))^{-1}})\Big]v|u\Big\rangle \\
=& \hbar\omega(\mathbf{k})\Big\langle\big((H_A+\omega(\mathbf{k}))^{-1}\otimes I+I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,v|u\Big\rangle && (4.4.52)\\
&-2\hbar\omega(\mathbf{k})\Big\langle v|(\overline{H_A+H_B})^{-1}|u\Big\rangle \\
&+(\hbar\omega(\mathbf{k}))^2\Big\langle\big((H_A+\hbar\omega(\mathbf{k}))^{-1}+(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\,u|(\overline{H_A+H_B})^{-1}|v\Big\rangle.
\end{aligned}
$$

Again using that $\sum_{i_A} \mathbf{x}_{i_A}$, $\sum_{j_B} \mathbf{x}_{j_B}$ and Q_R are real multiplication operators, we find

$$
\begin{aligned}
&(4.4.52)\\
=&\hbar\omega(\mathbf{k})\Big\langle[(H_A+\omega(\mathbf{k}))^{-1}\mathbf{v}_A](1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&+\hbar\omega(\mathbf{k})\Big\langle\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})[(H_B+\omega(\mathbf{k}))^{-1}\mathbf{v}_B]|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
=&\hbar\omega(\mathbf{k})\Big\langle[(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&+\hbar\omega(\mathbf{k})\Big\langle\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R|\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
=&-((4.4.48)+(4.4.49))
\end{aligned}
$$

Adding up all terms, we arrive at

$$
\begin{aligned}
T_1=\frac{2}{\hbar^2}\mathrm{Re}\Bigg[&\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\times\Bigg[-2\hbar\omega(\mathbf{k})\Big\langle\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B|(\overline{H_A+H_B})^{-1}|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0|Q_R|\\
&\qquad\qquad\Big|\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\Big\rangle\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\right)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)|(\overline{H_A+H_B})^{-1}|\\
&\qquad\qquad\Big|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle\left(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)|(\overline{H_A+H_B})^{-1}|\\
&\qquad\qquad\Big|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big]\Bigg]\\
=&M_B(\mathbf{R},\sigma),
\end{aligned}
$$

finishing the first part of the proof.

Part ii) In this part of the proof we will show that

$$
\begin{aligned}
&\frac{1}{m_e^2c^2}\left(\sum_{i_A,j_A}+\sum_{i_B,j_B}\right)\Big[2\mathrm{Re}\left[\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma(\mathbf{p}_j\cdot\mathbf{A}_\sigma)T^\sigma|Q_R\Psi_0\rangle\right]\\
&+\langle(\mathbf{p}_i\cdot\mathbf{A}_\sigma)\Psi_0|T^\sigma Q_R T^\sigma|(\mathbf{p}_j\cdot\mathbf{A}_\sigma)\Psi_0\rangle\Big]=M_A(\mathbf{R},\sigma).
\end{aligned}
$$

First of all, consider the contribution

$$
T_{2a}:=\frac{1}{m_e^2c^2}\sum_{i_A,j_A}\langle(\mathbf{p}_{i_A}\cdot\mathbf{A}_\sigma(0))\Psi_0|T^\sigma Q_R T^\sigma|(\mathbf{p}_{j_A}\cdot\mathbf{A}_\sigma(0))\Psi_0\rangle
$$

and note that by Lemma A.8.1, we have

$$
(\mathbf{p}_{i_A}\cdot\mathbf{A}_\sigma(0))\Psi_0=[(\mathbf{p}_{i_A}\cdot\mathbf{A}_\sigma(0))(\Psi_A^0\otimes\Omega)]\otimes\Psi_B^0\in\{\Psi_A^0\}^\perp\otimes\mathcal{F}_\sigma^{(1)}\widehat{\otimes}\{\Psi_B^0\},
$$

on which T^σ acts as $I_{\{\Psi_B^0\}} \otimes T_A^\sigma = I_{\{\Psi_B^0\}} \otimes \left(\overline{H_A + H_{f\geq\sigma}}|_{\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}_\sigma^{(1)}}}\right)^{-1}$ according to Lemma 4.2.5, (4.2.9). Thus we obtain

$$
\begin{aligned}
&T_{2a}\\
=&\frac{1}{m_e^2 c^2} \sum_{i_A, j_A} \Big\langle T_A^\sigma[(\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))(\Psi_A^0 \otimes \Omega)] \otimes \Psi_B^0 | Q_R | T_A^\sigma[(\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(0))(\Psi_A^0 \otimes \Omega)] \otimes \Psi_B^0 \Big\rangle.
\end{aligned}
$$

Using the definition of $\mathbf{A}_\sigma$ and applying the fiber decomposition of T_A^σ with respect to photon momenta (Lemma 4.2.7) now yields

$$
\begin{aligned}
T_{2a} = \frac{1}{m_e^2} \sum_{i_A, j_A} \sum_{\lambda=1,2} \int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \\
\Big\langle (H_A + \hbar\omega(\mathbf{k}))^{-1}[(\mathbf{p}_{i_A} \cdot \mathbf{e}(\mathbf{k}, \lambda))\Psi_A^0] \otimes \Psi_B^0 | Q_R \big| \\
\big| (H_A + \hbar\omega(\mathbf{k}))^{-1}[(\mathbf{p}_{j_A} \cdot \mathbf{e}(\mathbf{k}, \lambda))\Psi_A^0] \otimes \Psi_B^0 \Big\rangle,
\end{aligned}
$$

and an application of Lemma A.6.1 allows us to carry out the λ-summation, resulting in

$$
\begin{aligned}
T_{2a} = \frac{1}{m_e^2} \sum_{i_A, j_A} \sum_{\lambda=1,2} \int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \\
\Big\langle (H_A + \hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{i_A} \Psi_A^0] \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R \big| \\
\big| (H_A + \hbar\omega(\mathbf{k}))^{-1}[\mathbf{p}_{j_A} \Psi_A^0] \otimes \Psi_B^0 \Big\rangle \Big].
\end{aligned}
$$

As above, by Proposition 2.5.1, we have $\mathbf{v}_A := \sum_{i_A} \mathbf{x}_{i_A} \Psi_A^0 \in H^2(\mathbb{R}^{3Z_A})$ and $\mathbf{v}_B := \sum_{j_B} \mathbf{x}_{j_B} \Psi_B^0 \in H^2(\mathbb{R}^{3Z_B})$, so we can apply the commutator relation

$$
\mathbf{p}_{i_{A,B}} \Psi_{A,B}^0 = \frac{\mathrm{i} m_e}{\hbar} (H_{A,B} \mathbf{x}_{i_{A,B}} \Psi_{A,B}^0)
$$

from Lemma A.7.1 and the relations $(H_{A,B} + \hbar\omega(\mathbf{k}))^{-1} H_{A,B} = I - \hbar\omega(\mathbf{k})(H_{A,B} + \hbar\omega(\mathbf{k}))^{-1}$, which hold on $D(H_{A,B})$, yielding

$$
\begin{aligned}
T_{2a} =& \frac{1}{\hbar^2} \int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \\
&\times \Big[\Big\langle \mathbf{v}_A \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | \mathbf{v}_A \otimes \Psi_B^0 \Big\rangle \\
&\quad - \hbar\omega(\mathbf{k}) \Big\langle (H_A + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_A \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | \mathbf{v}_A \otimes \Psi_B^0 \Big\rangle \\
&\quad - \hbar\omega(\mathbf{k}) \Big\langle \mathbf{v}_A \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | (H_A + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_A \otimes \Psi_B^0 \Big\rangle \\
&\quad + (\hbar\omega(\mathbf{k}))^2 \Big\langle (H_A + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_A \otimes \Psi_B^0 | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | (H_A + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_A \otimes \Psi_B^0 \Big\rangle \Big] \\
=&(3.0.9).
\end{aligned}
$$

By exchanging the roles of A and B, one proves that

$$\begin{aligned}
T_{2b} :=& \frac{1}{m_e^2 c^2} \sum_{i_B, j_B} \langle (\mathbf{p}_{i_B} \cdot \mathbf{A}_\sigma(\mathbf{R}))\Psi_0 | T^\sigma Q_R T^\sigma | (\mathbf{p}_{j_B} \cdot \mathbf{A}_\sigma(\mathbf{R}))\Psi_0 \rangle \\
=& \frac{1}{\hbar^2} \int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \\
& \times \Bigg[\Big\langle \Psi_A^0 \otimes \mathbf{v}_B | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | \Psi_A^0 \otimes \mathbf{v}_B \Big\rangle \\
& \quad - \hbar\omega(\mathbf{k}) \Big\langle \Psi_A^0 \otimes (H_B + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_B | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | \Psi_A^0 \otimes \mathbf{v}_B \Big\rangle \\
& \quad - \hbar\omega(\mathbf{k}) \Big\langle \Psi_A^0 \otimes \mathbf{v}_B | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | \Psi_A^0 \otimes (H_B + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_B \Big\rangle \\
& \quad + (\hbar\omega(\mathbf{k}))^2 \Big\langle \Psi_A^0 \otimes (H_B + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_B | (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) Q_R | \Psi_A^0 \otimes (H_B + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_B \Big\rangle \Bigg] \\
=& (3.0.10).
\end{aligned}$$

It remains to investigate

$$\frac{1}{m_e^2 c^2} \left(\sum_{i_A, j_A} + \sum_{i_B, j_B} \right) 2\mathrm{Re} \left[\langle (\mathbf{p}_i \cdot \mathbf{A}_\sigma)\Psi_0 | T^\sigma (\mathbf{p}_j \cdot \mathbf{A}_\sigma) T^\sigma | Q_R \Psi_0 \rangle \right].$$

In the first contribution

$$T_{2c} := \frac{2}{m_e^2 c^2} \sum_{i_A, j_A} \mathrm{Re} \left[\langle (\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0)\Psi_0 | T^\sigma (\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(0)) T^\sigma | Q_R \Psi_0 \rangle \right],$$

we move all resolvents to the left and obtain

$$T_{2c} = \frac{2}{m_e^2 c^2} \sum_{i_A, j_A} \mathrm{Re} \left[\langle T^\sigma \underbrace{(\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(0)) T^\sigma ([(\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))(\Psi_A^0 \otimes \Omega)] \otimes \Psi_B^0)}_{=:u} | Q_R \Psi_0 \rangle \right].$$

Next we use the invariance properties of T^σ (see Lemma 4.2.5) to simplify this expression: as already shown above,

$$T^\sigma([(\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))(\Psi_A^0 \otimes \Omega)] \otimes \Psi_B^0) = [T_A^\sigma((\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(0))(\Psi_A^0 \otimes \Omega))] \otimes \Psi_B^0,$$

and this vector lies in $\{\Psi_B^0\} \widehat{\otimes} (\{\Psi_A^0\}^\perp \otimes \mathcal{F}_\sigma^{(1)})$. Therefore, since $(\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(0))$ does not act on the coordinates of B,

$$u \in \{\Psi_B^0\} \widehat{\otimes} (\mathcal{H}_A \otimes (\mathcal{F}_\sigma^{(0)} \oplus \mathcal{F}_\sigma^{(2)})) = \left(\{\Psi_B^0\} \widehat{\otimes} (\mathcal{H}_A \otimes \{\Omega\}) \right) \oplus \left(\{\Psi_B^0\} \widehat{\otimes} (\mathcal{H}_A \otimes \mathcal{F}_\sigma^{(2)}) \right).$$

Let u_1 and u_2 denote the projections of u onto the two subspaces on the right-hand side. From the invariance properties of T^σ we conclude that also $T^\sigma u \in \left(\{\Psi_B^0\} \widehat{\otimes} (\mathcal{H}_A \otimes \{\Omega\}) \right) \oplus \left(\{\Psi_B^0\} \widehat{\otimes} (\mathcal{H}_A \otimes \mathcal{F}_\sigma^{(2)}) \right)$. Since the Coulomb potential Q_R acts only on the particle coordinates, it follows for the right-hand side in the above inner product that $Q_R \Psi_0 \in (\mathcal{H}_A \otimes \mathcal{H}_B) \widehat{\otimes} \{\Omega\}$. But now the mutual orthogonality of Fock space levels implies that

the only non-vanishing contributions to T_{2c} are the terms containing $\langle T^\sigma u_1 | Q_R \Psi_0 \rangle$. We calculate u_1 explicitly. Recalling

$$\mathbf{A}_\sigma(\mathbf{x}) := a^\dagger(\mathbf{G}^{\mathbf{x}}_\sigma) + a(\mathbf{G}^{\mathbf{x}}_\sigma),$$

where $\mathbf{G}^{\mathbf{x}}_\sigma = \chi_\sigma(\mathbf{k})\, c\, C(\mathbf{k}) \mathbf{e}(\mathbf{k},\lambda) \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}}$, and using that $(\mathbf{p}_{i_A} \cdot a(\mathbf{G}^0_\sigma))(\Psi^0_A \otimes \Omega) = 0$, we conclude

$$u_1 = [(\mathbf{p}_{j_A} \cdot a(\mathbf{G}^0_\sigma)) T^\sigma_A ((\mathbf{p}_{i_A} \cdot a^\dagger(\mathbf{G}^0_\sigma))(\Psi^0_A \otimes \Omega))] \otimes \Psi^0_B.$$

By the definition of the action of the creation operators on $\{\Omega\}$, that of the annihilation operators on $\mathcal{F}^{(1)}_\sigma$ and the fiber decomposition of T^σ_A with respect to photon momenta (Lemma 4.2.7), this equals

$$\begin{aligned}
&u_1 \\
=&\left[(\mathbf{p}_{j_A} \cdot \left(\sum_{\lambda=1,2} \int_{\Omega_\sigma} d\mathbf{k}\, (\overline{\mathbf{G}^0_\sigma(\mathbf{k},\lambda)}) (H_A + \hbar\omega(\mathbf{k}))^{-1} [(\mathbf{p}_{i_A} \Psi^0_A \cdot \mathbf{G}^0_\sigma)(\mathbf{k},\lambda)] \right) \right] \otimes \Psi^0_B \\
=&c^2 \left[(\mathbf{p}_{j_A} \cdot \left(\int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \left(\sum_{\lambda=1,2} \mathbf{e}(\mathbf{k},\lambda) \cdot \mathbf{e}(\mathbf{k},\lambda) \right) (H_A + \hbar\omega(\mathbf{k}))^{-1} [\mathbf{p}_{i_A} \Psi^0_A] \right) \right] \otimes \Psi^0_B \\
=&c^2 \left[(\mathbf{p}_{j_A} \cdot \left(\int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \left(1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}} \right) (H_A + \hbar\omega(\mathbf{k}))^{-1} [\mathbf{p}_{i_A} \Psi^0_A] \right) \right] \otimes \Psi^0_B,
\end{aligned}$$

where we have used Lemma A.6.1 for the last identity. Now since $u_1 \in \{\Psi^0_B\} \widehat{\otimes} (\mathcal{H}_A \otimes \{\Omega\})$, we conclude

$$\begin{aligned}
&T^\sigma u_1 \\
=&c^2 (H_{A|\{\Psi^0_A\}^\perp})^{-1} \left[(\mathbf{p}_{j_A} \cdot \left(\int_{\Omega_\sigma} d\mathbf{k}\, |C(\mathbf{k})|^2 \left(1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}} \right) (H_A + \hbar\omega(\mathbf{k}))^{-1} [\mathbf{p}_{i_A} \Psi^0_A] \right) \right] \otimes \Psi^0_B,
\end{aligned}$$

(see Lemma 4.2.5), and using the above commutator relation and the relation $(H_A + \hbar\omega(\mathbf{k}))^{-1} H_A = I - \hbar\omega(\mathbf{k})(H_A + \hbar\omega(\mathbf{k}))^{-1}$ finally yields

$$\begin{aligned}
T_{2c} =& 2\mathrm{Re} \Bigg[\int_{\Omega_\sigma} d\mathbf{k} \left(\frac{-\mathrm{i}}{m_e \hbar} \right) |C(\mathbf{k})|^2 \\
&\times \Bigg[\Big\langle (H_{A|\{\Psi^0_A\}^\perp})^{-1} \Big((\sum_{j_A} \mathbf{p}_{j_A}) \cdot (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) \mathbf{v}_A \Big) \otimes \Psi^0_B | Q_R (\Psi^0_A \otimes \Psi^0_B) \Big\rangle \\
&- \hbar\omega(\mathbf{k}) \Big\langle (H_{A|\{\Psi^0_A\}^\perp})^{-1} \Big((\sum_{j_A} \mathbf{p}_{j_A}) \cdot (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) (H_A + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_A \Big) \otimes \Psi^0_B \Big| \\
&\qquad \Big| Q_R (\Psi^0_A \otimes \Psi^0_B) \Big\rangle \Bigg] \Bigg] = (3.0.11).
\end{aligned}$$

The last identity to be established, namely

$$\begin{aligned}
T_{2d} :=& \frac{2}{m_e^2 c^2} \sum_{i_B, j_B} \mathrm{Re} \left[\langle (\mathbf{p}_{i_A} \cdot \mathbf{A}_\sigma(\mathbf{R}) \Psi_0 | T^\sigma (\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(\mathbf{R})) T^\sigma | Q_R \Psi_0 \rangle \right] \\
=& (3.0.12),
\end{aligned}$$

is proven completely analogous by exchanging the roles of A and B.

4.4.6 Proof of Lemma 4.4.5

First recall

$$
\begin{aligned}
&(4.3.9)\\
&= -\langle\Psi_0|Q_R|\Psi_0\rangle\Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2+\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big) && (4.4.53)\\
&\quad+\langle H'_{\sigma,A}(\Psi_A^0\otimes\Omega)|T_A^\sigma|H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\rangle\ \|T_B^\sigma\, H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2 && (4.4.54)\\
&\quad+\langle H'_{\sigma,B}(\Psi_B^0\otimes\Omega)|T_B^\sigma|H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\rangle\ \|T_A^\sigma H'_\sigma(\Psi_A^0\otimes\Omega)\|^2 && (4.4.55)\\
&\quad-\langle(\Psi_A^0\otimes\Omega)|H''_{\sigma,A}|(\Psi_A^0\otimes\Omega)\rangle\ \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2 && (4.4.56)\\
&\quad-\langle(\Psi_B^0\otimes\Omega)|H''_{\sigma,B}|(\Psi_B^0\otimes\Omega)\rangle\ \|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2. && (4.4.57)
\end{aligned}
$$

Note that apart from $\langle\Psi_0|Q_R|\Psi_0\rangle\|T^\sigma H'_\sigma\Psi_0\|^2$, all contributions are R-independent. We will start by investigating the second and third contribution. Applying the general scheme outlined in Section 4.4.1 above (i.e. using the definition of the perturbation operators $H'_{\sigma,A}$ and $H'_{\sigma,B}$, the invariance properties of T^σ (Lemma 4.2.5), the fiber decomposition of the reduced resolvent on $\mathcal{F}_\sigma^{(1)}$ (Lemma 4.2.7), rotation invariance of Ψ_A^0 and Ψ_B^0 and the polarization vector identities from Lemma A.6.1), we arrive at

$$
\begin{aligned}
&(4.4.54)+(4.4.55)\\
&=\frac{4}{9}\frac{1}{(m_e)^4}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2\,|C(\mathbf{k}_2)|^2\sum_{i_A,j_A,i_B,j_B}\sum_{\alpha,\beta=1}^{3}\\
&\qquad\Big\langle\mathbf{p}_{i_A}^\alpha\Psi_A^0\otimes\mathbf{p}_{i_B}^\beta\Psi_B^0|(H_A+\omega(\mathbf{k}_1))^{-1}(H_B+\omega(\mathbf{k}_2))^{-2}\\
&\qquad\qquad+(H_B+\omega(\mathbf{k}_1))^{-1}(H_A+\omega(\mathbf{k}_2))^{-2}|\mathbf{p}_{j_A}^\alpha\Psi_A^0\otimes\mathbf{p}_{j_B}^\beta\Psi_B^0\Big\rangle. && (4.4.58)
\end{aligned}
$$

Now recall the term (4.4.6). It can be rewritten as

$$
\begin{aligned}
&(4.4.6)\\
&=-\frac{1}{9}\frac{1}{(m_e)^4}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2\\
&\quad\Big\langle\mathbf{p}_{i_A}^\alpha\Psi_A^0\otimes\mathbf{p}_{j_B}^\beta\Psi_B^0\big|\\
&\qquad\big|8(H_A+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_B+\omega(\mathbf{k}_2))^{-1}\\
&\qquad+4(H_A+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_A+\omega(\mathbf{k}_1))^{-1}\\
&\qquad+4(H_B+\omega(\mathbf{k}_1))^{-1}(\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}(H_B+\omega(\mathbf{k}_1))^{-1}\big|\\
&\qquad\qquad\big|\mathbf{p}_{k_A}^\alpha\Psi_A^0\otimes\mathbf{p}_{l_B}^\beta\Psi_B^0\Big\rangle
\end{aligned}
$$

$$
\begin{aligned}
= & -\frac{4}{9}\frac{1}{(m_e)^4}\sum_{i_A,j_B,k_A,l_B}\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2 \\
& \Big\langle \mathbf{p}_{i_A}^{\alpha}\Psi_A^0\otimes \mathbf{p}_{j_B}^{\beta}\Psi_B^0 | \Big((H_A+\omega(\mathbf{k}_1))^{-1}+(H_B+\omega(\mathbf{k}_2))^{-1}\Big)^2 \\
& \qquad \times (\overline{H_A+H_B}+\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^{-1}|\mathbf{p}_{k_A}^{\alpha}\Psi_A^0\otimes \mathbf{p}_{l_B}^{\beta}\Psi_B^0\Big\rangle, \qquad (4.4.59)
\end{aligned}
$$

where we have used the commutator relations from Lemma 4.2.18 ii) and the fact that by Lemma A.10.1), we can change variables from $\mathbf{k}_1$ to $\mathbf{k}_2$ in the last term without changing the value of the integral. Now using the operator identity Lemma 4.2.18 iii) and the fact that under the exchange $\mathbf{k}_1 \leftrightarrow \mathbf{k}_2$ (note that $\Omega_\sigma \times \Omega_\sigma$ is also invariant with respect to this), the second term in (4.2.26) becomes minus the first term, we conclude that by the symmetry of the remaining integrand,

$$(4.4.54) + (4.4.55) + (4.4.6) = 0.$$

For the last two contributions to (4.3.9), we find

$$
\begin{aligned}
& (4.4.56) + (4.4.57) \\
= & -\langle\Psi_0|H''_{\sigma,A}|\Psi_0\rangle\|T^\sigma H'_{\sigma,B}\Psi_0\|^2 - \langle\Psi_0|H''_{\sigma,B}|\Psi_0\rangle\|T^\sigma H'_{\sigma,A}\Psi_0\|^2 \\
= & -Z_A\frac{1}{2m_e c^2}\sum_{\alpha=1}^{3}\|\,(\mathbf{G}_\sigma^{\mathbf{x}})_\alpha\,\|_{W_\sigma}^2\|T^\sigma H'_{\sigma,B}\Psi_0\|^2 \\
& -Z_B\frac{1}{2m_e c^2}\sum_{\alpha=1}^{3}\|\,(\mathbf{G}_\sigma^{\mathbf{x}})_\alpha\,\|_{W_\sigma}^2\|T^\sigma H'_{\sigma,A}\Psi_0\|^2 \\
= & -Z_A\frac{2}{2m_e}\left(\int_{\{\omega(\mathbf{k})\geq\sigma\}}|C(\mathbf{k})|^2 d\mathbf{k}\right)\|T^\sigma H'_{\sigma,B}\Psi_0\|^2 \\
& -Z_B\frac{2}{2m_e}\left(\int_{\{\omega(\mathbf{k})\geq\sigma\}}|C(\mathbf{k})|^2 d\mathbf{k}\right)\|T^\sigma H'_{\sigma,A}\Psi_0\|^2,
\end{aligned}
$$

which is cancelled by the term (4.4.11), finishing the proof of Lemma 4.4.5.

Chapter 5

Analysis of terms containing the Coulomb interaction

In this chapter we carry out the analysis of the terms in $V_i^\sigma(\Lambda, \mathbf{R})$, $i = 1, 2, 3, 4$, which contain the smeared interatomic Coulomb potential Q_R. See the introduction for an overview. The main tool is the so-called multipole expansion, which is a series expansion of Q_R on a restrained configuration space, and which we introduce in the next section.

5.1 Multipole expansion of the (smeared) interatomic Coulomb potential Q_R

Following Gardner ([Gar07]), we derive a tail estimate for the quadratic form of this series. If the series is truncated at order L, the right-hand side of this estimate involves the quotient $\left(\frac{4(d+1)}{R}\right)^{L+1}$, where R is the interatomic distance and d characterizes the size of the area of $\mathbb{R}^{3N}$ to which the electron coordinates are restricted. Furthermore, we establish that the quadratic form of Q_R, if evaluated on exponentially decaying wave functions, decays exponentially outside this area as $d \to \infty$. We will make the assumptions (A1) on the form factor ψ throughout this section. Writing Q_R in position space (see A.5) yields

$$\begin{aligned} &Q_R \\ =&\frac{1}{4\pi} \sum_{i_A, j_B} \int_{\mathbb{R}^6} dy dy' \psi(y)\psi(y') \\ &\times \Big(\frac{1}{|\mathbf{R} - y + y'|} + \frac{1}{|\mathbf{x}_{i_A} - \mathbf{x}_{j_B} - \mathbf{R} - y + y'|} - \frac{1}{|\mathbf{x}_{i_A} - \mathbf{R} - y + y'|} - \frac{1}{|\mathbf{x}_{j_B} + \mathbf{R} - y + y'|} \Big). \end{aligned} \tag{5.1.1}$$

For two vectors $\mathbf{r}, \mathbf{R} \in \mathbb{R}^3$ satisfying $|\mathbf{r}| < |\mathbf{R}|$, the series expansion

$$\frac{1}{|\mathbf{r} - \mathbf{R}|} = \sum_{l=0}^{\infty} \frac{|\mathbf{r}|^l}{|\mathbf{R}|^{l+1}} P_l(\cos\theta)$$

converges (absolutely). Here $P_l(x)$ is the l-th Legendre polynomial and θ is the angle enclosing the vectors $\mathbf{r}$ and $\mathbf{R}$, defined via the inner product by $\mathbf{r}\cdot\mathbf{R} = |\mathbf{r}|\,|\mathbf{R}|\cos\theta$. Let $R := |\mathbf{R}|$ and choose $d \le R/4$, $R_0 := 6\,\mathrm{diam}\,\mathrm{supp}\,\psi$. Then for $y, y' \in \mathrm{supp}\,\psi$, $|\mathbf{x}_{i_A}|$, $|\mathbf{x}_{j_B}| \le d$ and $R > R_0$, we have

$$\begin{aligned}
|\mathbf{R}+y-y'| \ge& |R-|y-y'|| \ge +R-2\,\mathrm{diam}\,\mathrm{supp}\,\psi\\
\ge& (2/3)R \ge (8/3)d \ge 2d\\
\ge& |\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|,\\
|\mathbf{R}+y-y'| \ge& |\mathbf{x}_{i_A}|,\\
|-\mathbf{R}+y-y'| \ge& |\mathbf{x}_{j_B}|,
\end{aligned}$$

and thus the multipole expansion is applicable to the last three terms in (5.1.1), yielding

$$\begin{aligned}
Q_R =& \frac{1}{4\pi}\sum_{i_A,j_B}\int_{\mathbb{R}^6} dydy'\psi(y)\,\psi(y')\\
&\times\Bigg[\frac{1}{|\mathbf{R}-y+y'|}+\sum_{l=0}^{\infty}\frac{|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|^l}{|\mathbf{R}+y-y'|^{l+1}}P_l\left(\cos\theta_{\mathbf{x}_{i_A}-\mathbf{x}_{j_B},\,\mathbf{R}+y-y'}\right)\\
&\quad-\sum_{l=0}^{\infty}\frac{|\mathbf{x}_{i_A}|^l}{|\mathbf{R}+y-y'|^{l+1}}P_l\left(\cos\theta_{\mathbf{x}_{i_A},\,\mathbf{R}+y-y'}\right)\\
&\quad-\sum_{l=0}^{\infty}\frac{|\mathbf{x}_{j_B}|^l}{|-\mathbf{R}+y-y'|^{l+1}}P_l\left(\cos\theta_{\mathbf{x}_{j_B},\,-\mathbf{R}+y-y'}\right)\Bigg].
\end{aligned}$$

As in the non-smeared case (see [Gar07]), one easily checks - using the definition of the Legendre polynomials - that the $(l=0)$- and $(l=1)$-terms vanish. For the latter, one uses in addition that the change of variables $y \leftrightarrow y'$ does not change the value of the integral and one can thus replace $|-\mathbf{R}+y-y'| = |\mathbf{R}+y'-y|$ by $|\mathbf{R}+y-y'|$. Evaluation of the $(l=2)$-term, which again uses the property just mentioned, yields

$$\begin{aligned}
Q_2 :=& \frac{1}{4\pi}\sum_{i_A,j_B}\int_{\mathbb{R}^6} dydy'\psi(y)\,\psi(y')\\
&\times\frac{1}{|\mathbf{R}+y-y'|^3}\Big[\mathbf{x}_{i_A}\cdot\Big(I_{\mathbb{R}^3}-3(\widehat{\mathbf{R}+y-}\,y')\otimes(\widehat{\mathbf{R}+y-}\,y')\Big)\,\mathbf{x}_{j_B}\Big],
\end{aligned}$$

which corresponds exactly to the non-smeared dipole operator, as can be seen by formally replacing ψ by δ_0. Now noting that in the sense of the distributional Fourier transform on $\mathbb{R}^3$, we have

$$\mathfrak{F}\left(\frac{1}{|\mathbf{k}|^3}\Big[I_{\mathbb{R}^3}-3(\widehat{\mathbf{k}}\otimes\widehat{\mathbf{k}})\Big]\right)=\frac{2}{(2\pi)^{1/2}}\,\hat{\mathbf{k}}\otimes\hat{\mathbf{k}},$$

Q_2 can be expressed as

$$\begin{aligned}
Q_2 &= \frac{(2\pi)^{3/2}}{4\pi}\frac{2}{(2\pi)^{1/2}}\sum_{i_A,j_B}\int d\mathbf{k}\frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2}\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}}(\mathbf{x}_{i_A}\cdot\mathbf{k})\,(\mathbf{k}\cdot\mathbf{x}_{j_B})\\
&= \sum_{i_A,j_B}\int d\mathbf{k}\frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2}\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}}(\mathbf{x}_{i_A}\cdot\mathbf{k})\,(\mathbf{k}\cdot\mathbf{x}_{j_B})\\
&=: \tilde{Q}. \qquad (5.1.2)
\end{aligned}$$

To exhibit the mathematical structure of the higher terms contributing to Q_R more clearly, we will need two results from the spectral theory of the Laplacian. Firstly, we use the addition theorem for Legendre polynomials (see e.g. [WW96]), which furnishes a connection between the l-th Legendre polynomial and the spherical harmonics Y_{lm} of degree l: for two unit vectors $\hat{x}, \hat{y} \in \mathbb{R}^3$, we have

$$P_l(\hat{x} \cdot \hat{y}) = \frac{4\pi}{2l+1} \sum_{m=-l}^{l} Y_{lm}^*[(\theta,\varphi)_{\hat{x}}]\, Y_{lm}[(\theta,\varphi)_{\hat{y}}],$$

where the tuple $(\theta,\varphi)_{\hat{x}}$ denotes the angular variables of a unit vector $\hat{x} \in \mathbb{R}^3$. The second result tells us how to expand a translated regular solid harmonic into spherical harmonics depending on the individual vectors comprising the translate, see e.g. [ST77]. More precisely, we have

$$\begin{aligned}
&\sqrt{\frac{4\pi}{2l+1}}|a-b|^l Y_{lm}[(\theta,\varphi)_{a-b}] \\
=&\sum_{l_1=0}^{l} \binom{2l}{2l_1}^{1/2} \sum_{m_1=-l_1}^{l_1} \sqrt{\frac{4\pi}{2l_1+1}} \sqrt{\frac{4\pi}{2(l-l_1)+1}} C_{l,m,l_1,m_1} \\
&\qquad \times |a|^{l_1}\, |b|^{l-l_1} Y_{l_1 m_1}[(\theta,\varphi)_a]\, Y_{(l-l_1)(m-m_1)}[(\theta,\varphi)_b],
\end{aligned}$$

for any $a, b \in \mathbb{R}^3$, where

$$C_{l,m,l_1,m_1} = \binom{l+m}{l_1+m_1}^{1/2} \binom{l-m}{l_1-m_1}^{1/2} \binom{2l}{2l_1}^{-1/2}$$

is a Clebsch-Gordan coefficient. Applying these results to Q_R yields

$$\begin{aligned}
Q_R =& Q_2 + \frac{1}{4\pi} \sum_{i_A, j_B} \int dy dy' \psi(y), \psi(y') \\
&\times \Bigg[\sum_{l=3}^{\infty} \Bigg[\sqrt{\frac{4\pi}{2l+1}} \sum_{m=-l}^{l} \sum_{l_1=0}^{l} \sum_{m_1=-l_1}^{l_1} \binom{2l}{2l_1}^{1/2} \sqrt{\frac{4\pi}{2l_1+1}} \sqrt{\frac{4\pi}{2(l-l_1)+1}} C_{l,m,l_1,m_1} \\
&\quad \times \frac{|\mathbf{x}_{i_A}|^{l_1}\, |\mathbf{x}_{j_B}|^{l-l_1}}{|\mathbf{R}+y-y'|^{l+1}} Y_{l_1 m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\, Y_{(l-l_1)(m-m_1)}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}] Y_{lm}[(\theta,\varphi)_{\mathbf{R}+y-y'}] \\
&\quad - \frac{4\pi}{2l+1} \sum_{m=-l}^{l} \frac{|\mathbf{x}_{i_A}|^l}{|\mathbf{R}+y-y'|^{l+1}} Y_{lm}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\, Y_{lm}[(\theta,\varphi)_{\mathbf{R}+y-y'}] \\
&\quad - \frac{4\pi}{2l+1} \sum_{m=-l}^{l} \frac{|\mathbf{x}_{j_B}|^l}{|-\mathbf{R}+y-y'|^{l+1}} Y_{lm}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\, Y_{lm}[(\theta,\varphi)_{-\mathbf{R}+y-y'}] \Bigg] \Bigg].
\end{aligned}$$

Since the multipole expansion converges absolutely and uniformly with respret to y and y' (note that $\mathrm{supp}(\psi(\cdot)\,\psi(\cdot'))$ is compact), we can exchange the summation and the $dydy'$-

integration. Next define

$$
\begin{aligned}
u_{l,m,l_1,m_1}(\mathbf{R}) :=& \underbrace{\frac{1}{4\pi}\sqrt{\frac{4\pi}{2l+1}}\binom{2l}{2l_1}^{1/2}\sqrt{\frac{4\pi}{2l_1+1}}\sqrt{\frac{4\pi}{2(l-l_1)+1}}C_{l,m,l_1,m_1}}_{=:\tilde{C}_{l,m,l_1,m_1}} \\
&\times\left(\int dydy'\psi(y),\psi(y')\frac{Y_{lm}[(\theta,\varphi)_{\mathbf{R}+y-y'}]}{|\mathbf{R}+y-y'|^{l+1}}\right), \\
v_{l,m}(\mathbf{R}) :=& \frac{1}{2l+1}\int dydy'\psi(y),\psi(y')\frac{Y_{lm}[(\theta,\varphi)_{\mathbf{R}+y-y'}]}{|\mathbf{R}+y-y'|^{l+1}}, \\
w_{l,m}(\mathbf{R}) :=& \frac{1}{2l+1}\int dydy'\psi(y),\psi(y')\frac{Y_{lm}[(\theta,\varphi)_{-\mathbf{R}+y-y'}]}{|-\mathbf{R}+y-y'|^{l+1}}.
\end{aligned}
$$

Note that these quantities are well-defined for all l since we have chosen $R > R_0 = 4\,\mathrm{diam}\,\mathrm{supp}\,\psi$, which implies $|\mathbf{R}+y| > 3\,\mathrm{diam}\,\mathrm{supp}\,\psi$ for all $y \in \mathrm{supp}\,\psi$, and thus ensures that $1/|\pm\mathbf{R}+y-y'|^{l+1}$ is continuous on $\mathrm{supp}\,\psi \times \mathrm{supp}\,\psi \subset \mathbb{R}^6$.
Summarizing these results, we obtain

Lemma 5.1.1. *Let ψ satisfy the assumptions (A1). Choose $0 < d \leq R/4$ and $R_0 = 6\,\mathrm{diam}\,\mathrm{supp}\,\psi$ as above. Then for $R > R_0$ and $|\mathbf{x}_{i_A}|$, $|\mathbf{x}_{j_B}| \leq d$,*

$$
\begin{aligned}
Q_R =& Q_2 \\
&+ \sum_{i_A,j_B}\sum_{l=3}^{\infty}\Bigg[\sum_{m=-l}^{l}\sum_{l_1=0}^{l}\sum_{m_1=-l_1}^{l_1}|\mathbf{x}_{i_A}|^{l_1}\,|\mathbf{x}_{j_B}|^{l-l_1}\,Y^*_{l_1m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}] \\
&\qquad\times Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\,u_{l,m,l_1,m_1}(\mathbf{R}) \\
&\quad - \sum_{m=-l}^{l}|\mathbf{x}_{i_A}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\,v_{l,m}(\mathbf{R}) \\
&\quad - \sum_{m=-l}^{l}|\mathbf{x}_{j_B}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\,w_{l,m}(\mathbf{R})\Bigg] =: \sum_{l=2}^{\infty}Q_l. \qquad (5.1.3)
\end{aligned}
$$

Furthermore, by the parity of the spherical harmonics of degree l and the invariance of $\psi(y)\psi(y')$ under $y \leftrightarrow y'$, it holds that $w_{l,m}(\mathbf{R}) = (-1)^l\,v_{l,m}(\mathbf{R})$.

Large R-asymptotics of the interatomic Coulomb potential Q_R

Lemma 5.1.2. *Suppose that ψ satisfies the assumptions (A1). Let $\Lambda > 0$, $l \geq 2$ and let $\hat{\mathbf{R}} \in \mathbb{R}^3$, $|\hat{\mathbf{R}}| = 1$ be fixed. Then*

$$
\begin{aligned}
&\lim_{R\to\infty}\int dydy'\frac{R^3\psi_0(Ry)\,R^3\psi_0(Ry')}{|\Lambda\hat{\mathbf{R}}+y-y'|^{l+1}}Y_{lm}[(\theta,\varphi)_{\Lambda\hat{\mathbf{R}}+y-y'}] \qquad (5.1.4)\\
=&\frac{Y_{lm}[(\theta,\varphi)_{\Lambda\hat{\mathbf{R}}}]}{|\Lambda\hat{\mathbf{R}}|^{l+1}} = \frac{1}{\Lambda^{l+1}}Y_{lm}[(\theta,\varphi)_{\hat{\mathbf{R}}}].
\end{aligned}
$$

Proof. Consider the function $g : \mathbb{R}^6 \to \mathbb{C}$,

$$g((y', y)) = \frac{1}{|\Lambda\hat{\mathbf{R}} + (y' - y)|^{l+1}} Y_{lm}[(\theta, \varphi)_{\Lambda\hat{\mathbf{R}}+(y'-y)}],$$

which is continuous and bounded away from the set $S := \{(y', y) | \, y = y' + \Lambda\hat{\mathbf{R}}\} = \{(y', y' + \Lambda\hat{\mathbf{R}}) | \, y' \in \mathbb{R}^3\}$ of its singularities. Define $\Psi_R(y', y) := R^3\psi_0(Ry)\, R^3\psi_0(Ry')$. Since $\operatorname{supp} \Psi_R \subset B_{1/R}(0) \times B_{1/R}(0)$, we have $|\Lambda\hat{\mathbf{R}} + y'| > 1/R$ for $R > R_0 = 4 \operatorname{diam} \operatorname{supp} \psi = 4/\Lambda$ and $|y'| \leq 1/R$. This shows that $\big(B_{1/R}(0) \times B_{1/R}(0)\big) \cap S = \emptyset$, and in particular $\operatorname{supp} \Psi_R \cap S = \emptyset$, which in turn implies that $g|_{\operatorname{supp} \Psi_R}$ is continuous and bounded. This allows us to interpret the integral in (5.1.4) as $(\Psi_R * g)((0, 0))$. By the assumptions on ψ_0, Ψ_R is a Dirac sequence (with respect to the parameter R), so that the assertion follows from the standard result that the convolution of a continuous function f with a Dirac sequence converges pointwise to the function f, see e.g. [Eva98]. □

Rescaling variables by $(y, y') \mapsto (\Lambda/Ry, \Lambda/Ry')$ and using the properties of ψ (see assumption (A1)) yields the identity

$$\begin{aligned} &\int dy dy' \frac{\psi(y), \psi(y')}{|\mathbf{R} + y - y'|^{l+1}} Y_{lm}[(\theta, \varphi)_{\mathbf{R}+y-y'}] \\ =&\frac{\Lambda^{l+1}}{R^{l+1}} \int dy dy' \frac{R^3\psi_0(Ry)\, R^3\psi_0(Ry')}{|\Lambda\hat{\mathbf{R}} + y - y'|^{l+1}} Y_{lm}[(\theta, \varphi)_{\Lambda\hat{\mathbf{R}}+y-y'}]. \end{aligned} \tag{5.1.5}$$

In view of (5.1.5), applying Lemma 5.1.2 to (5.1.3) yields

Lemma 5.1.3. *Suppose that ψ satisfies the assumptions (A1). Choose $0 < d \leq R/4$ and $R_0 = 4 \operatorname{diam} \operatorname{supp} \psi$ as above and let $\hat{\mathbf{R}} \in \mathbb{R}^3$, $|\hat{\mathbf{R}}| = 1$ be fixed. Then for $l \in \mathbb{N}$, $l \geq 2$ and $|\mathbf{x}_{i_A}|$, $|\mathbf{x}_{j_B}| \leq d$,*

$$\begin{aligned} Q_2 &\sim \frac{1}{4\pi} \sum_{i_A, j_B} \frac{1}{R^3} \Big[\mathbf{x}_{i_A} \cdot \Big(I_{\mathbb{R}^3} - 3\widehat{\mathbf{R}} \otimes \widehat{\mathbf{R}}\Big)\, \mathbf{x}_{j_B}\Big], \\ u_{l,m,l_1,m_1}(\mathbf{R}) &\sim \tilde{C}_{l,m,l_1,m_1} \frac{1}{R^{l+1}} Y_{lm}[(\theta, \varphi)_{\hat{\mathbf{R}}}] =: \frac{1}{R^{l+1}} \tilde{u}_{l,m,l_1,m_1}(\hat{\mathbf{R}}), \\ v_{l,m}(\mathbf{R}) &\sim \frac{1}{2l+1} \frac{1}{R^{l+1}} Y_{lm}[(\theta, \varphi)_{\hat{\mathbf{R}}}] =: \frac{1}{R^{l+1}} \tilde{v}_{l,m}(\hat{\mathbf{R}}), \\ w_{l,m}(\mathbf{R}) &\sim \frac{1}{2l+1} \frac{1}{R^{l+1}} Y_{lm}[(\theta, \varphi)_{-\hat{\mathbf{R}}}] = (-1)^l \frac{1}{R^{l+1}} \tilde{v}_{l,m}(\hat{\mathbf{R}}) \end{aligned}$$

as $R \to \infty$, and consequently, for $l \geq 3$,

$$\begin{aligned} Q_l \sim &\sum_{i_A, j_B} \frac{1}{R^{l+1}} \\ &\times \Bigg[\sum_{m=-l}^{l} \sum_{l_1=0}^{l} \sum_{m_1=-l_1}^{l_1} |\mathbf{x}_{i_A}|^{l_1} |\mathbf{x}_{j_B}|^{l-l_1} Y^*_{l_1 m_1}[(\theta, \varphi)_{\mathbf{x}_{i_A}}] Y^*_{(l-l_1)(m-m_1)}[(\theta, \varphi)_{\mathbf{x}_{j_B}}] \tilde{u}_{l,m,l_1,m_1}(\hat{\mathbf{R}}) \\ &\quad - \sum_{m=-l}^{l} \Big(|\mathbf{x}_{i_A}|^l Y^*_{lm}[(\theta, \varphi)_{\mathbf{x}_{i_A}}] + (-1)^l |\mathbf{x}_{j_B}|^l Y^*_{lm}[(\theta, \varphi)_{\mathbf{x}_{j_B}}]\Big) \tilde{v}_{l,m}(\hat{\mathbf{R}}) \Bigg] \end{aligned}$$

as $R \to \infty$.

5.1.1 Error estimates for the multipole expansion

Lemma 5.1.4 (Tail estimate)**.** *Suppose that ψ satisfies the assumptions (A1). Let $\Psi, \Phi \in L^2(\mathbb{R}^{3N})$ and let $0 < (d+1) \leq R/4$. Furthermore, assume $R > R_0 := 6 \operatorname{diam} \operatorname{supp} \psi$ and let $\Omega_d := \{|\mathbf{x}_i| \leq d| \leq d\} \subset \mathbb{R}^{3N}$. Let $\chi_{\Omega_d}(\mathbf{x}_1, \ldots, \mathbf{x}_N) = \Pi_{i=1}^N \chi_d(\mathbf{x}_i)$, where $\chi_d \in C_0^\infty(\mathbb{R}^3)$, $0 \leq \chi_d \leq 1$, $\chi_d(\mathbf{x}) = 1$ if $|\mathbf{x}| \leq d$, $\chi_d(\mathbf{x}) = 0$ if $|\mathbf{x}| > d+1$ and $\chi_d(R\mathbf{x}) = \chi_d(\mathbf{x})$ for all $R \in SO(3)$, be a smooth characteristic function of Ω_d. Then for any $L \in \mathbb{N}$, $L \geq 2$,*

$$\begin{aligned}
&\left| \left\langle \Psi | Q_R \chi_{\Omega_d} | \Phi \right\rangle_{L^2(\mathbb{R}^{3N})} - \left\langle \Psi | \Big(\sum_{l=2}^{L} Q_l \Big) \chi_{\Omega_d} | \Phi \right\rangle_{L^2(\mathbb{R}^{3N})} \right| \\
\leq & Z_A \, Z_B \frac{1}{R} \left(\frac{4(d+1)}{R} \right)^{L+1} \|\Phi\|_{L^2(\mathbb{R}^{3N}} \|\Psi\|_{L^2(\mathbb{R}^{3N})}.
\end{aligned}$$

Proof. The argument is a modification of the one given in [Gar07]. By the choice of d and R_0 and Lemma 5.1.1, the multipole expansion converges on $\operatorname{supp} \chi_{\Omega_d} \subset \Omega_{d+1}$. Therefore, using Fubini's theorem, we find

$$\begin{aligned}
I := & \left| \left\langle \Psi | Q_R \chi_{\Omega_d} | \Phi \right\rangle_{L^2(\mathbb{R}^{3N})} - \left\langle \Psi | \Big(\sum_{l=2}^{L} Q_l \Big) \chi_{\Omega_d} | \Phi \right\rangle_{L^2(\mathbb{R}^{3N})} \right| \\
\leq & \frac{1}{4\pi} \sum_{l=L+1}^{\infty} \sum_{i_A, j_B} \Bigg| \Big\langle \Psi | \chi_{\Omega_d} \int_{\mathbb{R}^6} dy dy' \psi(y) \psi(y') \\
& \times \Bigg[\frac{|\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^l}{|\mathbf{R} + y - y'|^{l+1}} P_l \left(\cos \theta_{\mathbf{x}_{i_A} - \mathbf{x}_{j_B}, \mathbf{R} + y - y'} \right) \\
& \quad - \frac{|\mathbf{x}_{i_A}|^l}{|\mathbf{R} + y - y'|^{l+1}} P_l \left(\cos \theta_{\mathbf{x}_{i_A}, \mathbf{R} + y - y'} \right) \\
& \quad - \frac{|\mathbf{x}_{j_B}|^l}{|-\mathbf{R} + y - y'|^{l+1}} P_l \left(\cos \theta_{\mathbf{x}_{j_B}, -\mathbf{R} + y - y'} \right) \Bigg] | \Phi \Big\rangle_{L^2(\mathbb{R}^{3N})} \Bigg|.
\end{aligned}$$

Note that $\cos \theta \in [-1, 1]$ and $|P_n(x)| \leq 1$ for $x \in [-1, 1]$. The term $1/|\mathbf{R} + y - y'|$ is independent of $\mathbf{x}_{i_A}$ and $\mathbf{x}_{j_B}$, and in the last term we can replace $|-\mathbf{R} + y - y'|$ with $|\mathbf{R} + y - y'|$ by virtue of the $y \leftrightarrow y'$-symmetry of the remaining integrand. This yields

$$\begin{aligned}
I \leq & \frac{1}{4\pi} \sum_{l=L+1}^{\infty} \left| \int_{\mathbb{R}^6} dy dy' \frac{\psi(y) \, \psi(y')}{|\mathbf{R} + y - y'|^{l+1}} \right| \\
& \times \left(\sum_{i_A, j_B} \left| \left\langle \Psi | \Big(|\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^l - |\mathbf{x}_{i_A}|^l - |\mathbf{x}_{j_B}|^l \Big) \chi_{\Omega_d} | \Phi \right\rangle_{L^2(\mathbb{R}^{3N})} \right| \right) \\
\leq & \frac{1}{4\pi} \sum_{l=L+1}^{\infty} \left| \int_{\mathbb{R}^6} dy dy' \frac{\psi(y) \, \psi(y')}{|\mathbf{R} + y - y'|^{l+1}} \right| \\
& \times \left(\sum_{i_A, j_B} \Big(2(d+1)^l + (2(d+1))^l \Big) \, \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\chi_{\Omega_d} \Phi\|_{L^2(\mathbb{R}^{3N})} \right)
\end{aligned}$$

$$\leq \frac{1}{4\pi} \sum_{l=L+1}^{\infty} \left| \int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\,\psi(y')}{|\mathbf{R}+y-y'|^{l+1}} \right| \times \left(\sum_{i_A, j_B} \left(2(d+1)^l + (2(d+1))^l \right) \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})} \right).$$

On $\operatorname{supp}\psi \times \operatorname{supp}\psi$, we have $|\mathbf{R}+y-y'| \geq R - 2\operatorname{diam}\operatorname{supp}\psi \geq 2/3R$, and thus

$$\int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\,\psi(y')}{|\mathbf{R}+y-y'|^{l+1}} \leq \left(\frac{3}{2R}\right)^{l+1} \|\psi\|_{L^1}^2 = \left(\frac{3}{2R}\right)^{l+1}. \tag{5.1.6}$$

This implies

$$\begin{aligned} I \leq & \frac{1}{4\pi} \sum_{l=L+1}^{\infty} \left(\frac{3}{2R}\right)^{l+1} Z_A\, Z_B \left(2(d+1)^l + (2(d+1))^l \right) \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})} \\ = & \frac{1}{4\pi} Z_A\, Z_B \sum_{l=L+1}^{\infty} \frac{1}{R} \left(\frac{4(d+1)}{R}\right)^l \left(3\left(\frac{3}{8}\right)^l + \frac{3}{2}\left(\frac{3}{4}\right)^l \right) \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})}. \end{aligned}$$

Furthermore, $(4(d+1)/R)^l \leq (4(d+1)/R)^{L+1}$ for all $l \geq L+1$, since $4(d+1)/R \leq 1$ by assumption, yielding

$$\begin{aligned} I \leq & \frac{1}{4\pi} Z_A\, Z_B \frac{1}{R} \left(\frac{4(d+1}{R}\right)^{L+1} \sum_{l=L+1}^{\infty} \left(3\left(\frac{3}{8}\right)^l + \frac{3}{2}\left(\frac{3}{4}\right)^l \right) \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})} \\ \leq & \frac{1}{4\pi} Z_A\, Z_B \frac{1}{R} \left(\frac{4(d+1)}{R}\right)^{L+1} \left(\frac{3\left(\frac{3}{8}\right)^{L+1}}{5/8} + \frac{\frac{3}{2}\left(\frac{3}{4}\right)^{L+1}}{1/4} \right) \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})} \\ \leq & \frac{3}{4\pi} Z_A\, Z_B \frac{1}{R} \left(\frac{4(d+1)}{R}\right)^{L+1} \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})} \\ \leq & Z_A\, Z_B \frac{1}{R} \left(\frac{4(d+1)}{R}\right)^{L+1} \|\Psi\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})}. \end{aligned}$$

□

Lemma 5.1.5 (Exponential decay of the quadratic form)**.** *Let ψ satisfy the assumptions (A1). Suppose that $f \in H^1(\mathbb{R}^{3N})$, $g \in L^2(\mathbb{R}^{3N})$ and that these functions satisfy the pointwise bounds*

$$\begin{aligned} |f(\mathbf{x}_1, \ldots, \mathbf{x}_N)| &\leq C_1 \mathrm{e}^{-\gamma_1(|\mathbf{x}_1| + \ldots |\mathbf{x}_N|)}, \\ |g(\mathbf{x}_1, \ldots, \mathbf{x}_N)| &\leq C_2 \mathrm{e}^{-\gamma_2(|\mathbf{x}_1| + \ldots |\mathbf{x}_N|)} \end{aligned}$$

for some positive constants C_1, C_2, γ_1, γ_2.
Let $\Omega_d := \{|\mathbf{x}_i| \leq d | i = 1, \ldots, N\} \subset \mathbb{R}^{3N}$. Let $\chi_{\Omega_d}(\mathbf{x}_1, \ldots, \mathbf{x}_N) = \Pi_{i=1}^N \chi_d(\mathbf{x}_i)$, where $\chi_d \in C_0^\infty(\mathbb{R}^3)$, $0 \leq \chi_d \leq 1$, $\chi_d(\mathbf{x}) = 1$ if $|\mathbf{x}| \leq d$, $\chi_d(\mathbf{x}) = 0$ if $|\mathbf{x}| > d+1$ and $\chi_d(R\mathbf{x}) = \chi_d(\mathbf{x})$ for all $R \in SO(3)$, be a smooth characteristic function of Ω_d.
Then there exist positive constants C and γ, independent of $\mathbf{R}$, d and the ultraviolet-cutoff Λ, such that

$$\left| \langle f | Q_R (1 - \chi_{\Omega_d}) | g \rangle_{L^2(\mathbb{R}^{3N})} \right| \leq C(1 + \frac{1}{R})\, \mathrm{e}^{-\gamma d}.$$

Proof. By Lemma A.1.1, Q_R is a bounded operator from $H^1(\mathbb{R}^{3N})$ to $L^2(\mathbb{R}^{3N})$, and its operator norm can be bounded by $C_Q(1+1/R)$, where $C_Q > 0$ is independent of R and the ultraviolet-cutoff Λ. Thus we obtain

$$\begin{aligned}&\left|\langle f|Q_R(1-\chi_{\Omega_d})|g\rangle_{L^2(\mathbb{R}^{3N})}\right| \\ \leq&\|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|f\|_{H^1(\mathbb{R}^{3N})}\|(1-\chi_{\Omega_d})g\|_{L^2(\mathbb{R}^{3N})} \\ \leq&C_Q(1+1/R)\|f\|_{H^1(\mathbb{R}^{3N})}\|g\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)},\end{aligned}$$

where for the second inequality we have used that $1-\chi_{\Omega_d} \leq 1$ and $\operatorname{supp}(1-\chi_{\Omega_d}) \subset L^2(\mathbb{R}^{3N}\setminus\Omega_d)$. The claim now follows from Lemma A.3.1 □

5.2 Analysis of $V_2^\sigma(R)$ and $-\langle\Psi_0|Q_R|\Psi_0\rangle\|T^\sigma H'_\sigma\Psi_0\|^2$

In this section we combine the results of the previous section with the spherical symmetry of the ground states Ψ_A^0 and Ψ_B^0 to prove that the terms

$$\langle\Psi_0|Q_R|\Psi_0\rangle$$

and

$$-\langle\Psi_0|Q_R|\Psi_0\rangle\Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2+\|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big)$$

occurring in $\sum_{i=1}^4 e^i V_i^\sigma(\mathbf{R})$ decay faster than any inverse power of R as $R\to\infty$.

Lemma 5.2.1. *Let ψ satisfy the assumptions (A1). Choose $d < |\mathbf{R}|/2$, set $\Omega_d := \{|\mathbf{x}_i| \leq d, i = 1,\dots,N\} \subset \mathbb{R}^{3N}$ and let χ_{Ω_d} be a smooth characteristic function of Ω_d as in Lemma 5.1.4. Suppose that $\Psi_A^0 \in L^2(\mathbb{R}^{3Z_A})$, $\Psi_B^0 \in L^2(\mathbb{R}^{3Z_B})$ are normalized and spherically symmetric in the sense that $\Psi_A^0(R\mathbf{x}_1,\dots,R\mathbf{x}_{Z_A}) = \Psi_A^0(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})$ and $\Psi_B^0(R\mathbf{x}_{Z_A+1},\dots,R\mathbf{x}_{Z_A+Z_B}) = \Psi_B^0(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_{Z_A+Z_B})$ for all rotations $R\in SO(3)$. Then for any $l \geq 2$,*

$$\langle\Psi_A^0\otimes\Psi_B^0|Q_l\chi_{\Omega_d}|\Psi_A^0\otimes\Psi_B^0\rangle_{L^2(\mathbb{R}^{3N})} = 0.$$

Proof. In the case $l = 2$, the result is most easily seen by the representation (5.1.2) for Q_2, which is odd under the change of variables $(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A},\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_{Z_A+Z_B}) \mapsto (-\mathbf{x}_1,\dots,-\mathbf{x}_{Z_A},\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_{Z_A+Z_B})$, while the remaining integrand $\chi_{\Omega_d}|\Psi_A^0|^2\,|\Psi_B^0|^2$ is invariant under this transformation by the assumptions on χ_{Ω_d} and the fact that Ψ_A^0 and Ψ_B^0 possess a definite parity (see the remarks after Proposition 2.5.1). For the case $l \geq 3$,

recall the definition (5.1.3) of the terms

$$Q_l = \sum_{i_A, j_B} \Bigg[\sum_{m=-l}^{l} \sum_{l_1=0}^{l} \sum_{m_1=-l_1}^{l_1} |\mathbf{x}_{i_A}|^{l_1} \, |\mathbf{x}_{j_B}|^{l-l_1} \, Y^*_{l_1 m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}] \, Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]$$
$$\times u_{l,m,l_1,m_1}(\mathbf{R}) \tag{5.2.1}$$
$$- \sum_{m=-l}^{l} |\mathbf{x}_{i_A}|^{l} \, Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{i_A}}] \, v_{l,m}(\mathbf{R}) \tag{5.2.2}$$
$$- \sum_{m=-l}^{l} |\mathbf{x}_{j_B}|^{l} \, Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \, w_{l,m}(\mathbf{R}) \Bigg]. \tag{5.2.3}$$

Each summand in (5.2.1) only depends on one variable from $\{\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}\}$ and one from $\{\mathbf{x}_{Z_A+1}, \ldots, \mathbf{x}_{Z_A+Z_B}\}$, so that after renaming variables and using the antisymmetry of the wave functions (χ_{Ω_d} is invariant under permutation of the coordinates), the corresponding term in the integral $\langle \Psi_A^0 \otimes \Psi_B^0 | Q_l \chi_{\Omega_d} | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3N})}$ reduces to an integral of the form

$$\int_{\operatorname{supp}\chi_d \times \operatorname{supp}\chi_d} d\mathbf{x}_A d\mathbf{x}_B \rho_{A,\Omega_d}(\mathbf{x}_A) \, \rho_{B,\Omega_d}(\mathbf{x}_B) |\mathbf{x}_A|^{l_1} \, |\mathbf{x}_B|^{l-l_1}$$
$$\times Y^*_{l_1 m_1}[(\theta,\varphi)_{\mathbf{x}_A}] \, Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_B}]$$
$$= \Big(\int_{\operatorname{supp}\chi_d} d\mathbf{x}_A \, \rho_{A,\Omega_d}(\mathbf{x}_A) \, |\mathbf{x}_A|^{l_1} \, Y^*_{l_1 m_1}[(\theta,\varphi)_{\mathbf{x}_A}] \Big)$$
$$\times \Big(\int_{\operatorname{supp}\chi_d} d\mathbf{x}_B \, \rho_{B,\Omega_d}(\mathbf{x}_B) \, |\mathbf{x}_B|^{l-l_1} \, Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_B}] \Big),$$

where ρ_{A,Ω_d} and ρ_{B,Ω_d} are the one-particle density matrices of $\Psi_A^0|_{\operatorname{supp}\chi_{\Omega_d}}$ and $\Psi_B^0|_{\operatorname{supp}\chi_{\Omega_d}}$, whose spherical symmetry (in the above sense) is inherited by the former: $\rho_{A,B,\Omega_d}(R\mathbf{x}) = \rho_{A,B,\Omega_d}(\mathbf{x})$ for all $\mathbf{x} \in \mathbb{R}^3$ and any $R \in SO(3)$, as is easily seen by a change of variables in the definition of ρ_{A,B,Ω_d} (note that the region $\operatorname{supp}\chi_{\Omega_d}$ is left invariant by rotations $(R, \ldots, R)$, $R \in SO(3)$). Thus $\rho_{A,\Omega_d}(\mathbf{x}_A)|\mathbf{x}_A|^{l_1}$ and $\rho_{B,\Omega_d}(\mathbf{x}_B)|\mathbf{x}_B|^{l-l_1}$ depend only on the radial variables $|\mathbf{x}_A|$ and $|\mathbf{x}_B|$, respectively. Spherical harmonics $Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}}]$ average to zero upon angular integration with respect to $\mathbf{x}$ if $l \geq 1$, which follows from their orthogonality properties, the $(l = 0)$-spherical harmonic being a constant function. But since $l \geq 3$ in our case, at least one of the numbers l_1 and $l - l_1$ is greater or equal to one, so that the corresponding integral vanishes. An analogous argument shows that the terms in (5.2.2) and (5.2.3) integrate to zero. □

Combining Lemmas 5.1.4, 5.1.5 and 5.2.1, we obtain the following result on

$$V_2^{\sigma}(R) = \langle \Psi_0 | Q_R | \Psi_0 \rangle$$

from (4.3.10), which says that this contribution to the interaction potential can be made smaller than any given power of $1/|\mathbf{R}|$.

Lemma 5.2.2. *Assume the hypotheses of Theorem 3.0.6.*
Then for any $L \geq 3$, there exist constants $C_1, C_2' \geq 0$, independent of $\mathbf{R}$ and the ultraviolet-cutoff Λ, such that for $R > R_0 := 6 \operatorname{diam} \operatorname{supp} \psi$,

$$\left|\langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3(Z_A+Z_B)})}\right| \leq Z_A\, Z_B \left(\frac{1}{|\mathbf{R}|}\right)^L + C_1(1+\frac{1}{|\mathbf{R}|})\mathrm{e}^{-C_2\sqrt{|\mathbf{R}|}}.$$

In particular,

$$\lim_{R\to\infty}\left(R^k \langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3(Z_A+Z_B)})}\right) = 0$$

for any $k \geq 0$.

Proof. By Proposition 2.5.1 iv) a), Ψ_A^0 and Ψ_B^0 satisfy pointwise exponential bounds, so that we can use Lemmas 5.1.4 and 5.1.5 to conclude that for any $0 < (d+1) \leq R/4$ and any $\tilde{L} \geq 2$,

$$\begin{aligned}
&|\langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3(Z_A+Z_B)})}| \\
=&|\langle \Psi_A^0 \otimes \Psi_B^0 | Q_R \chi_{\Omega_d} | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3N})} + \langle \Psi_A^0 \otimes \Psi_B^0 | Q_R(1-\chi_{\Omega_d}) | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3N})}| \\
\leq&|\langle \Psi_A^0 \otimes \Psi_B^0 | \Big(\sum_{l=2}^{\tilde{L}} Q_l\Big) \chi_{\Omega_d} | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3N})}| + Z_A\, Z_B \frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{\tilde{L}+1} \\
&+ C_1(1+\frac{1}{|\mathbf{R}|})\mathrm{e}^{-\tilde{C}'d}
\end{aligned}$$

for suitable constants $C_1, \tilde{C}' \geq 0$. As before, $\Omega_d := \{|\mathbf{x}_{i_A}| \leq d,\, |\mathbf{x}_{j_B}| \leq d\} \subset \mathbb{R}^{3N}$ and χ_{Ω_d} is a smooth characteristic function of Ω_d as in Lemma 5.1.4. By Proposition 2.5.2, Ψ_A^0 and Ψ_B^0 are spherically symmetric in the sense that $\Psi_A^0(R\mathbf{x}_1, \ldots, R\mathbf{x}_{Z_A}) = \Psi_A^0(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A})$ and $\Psi_B^0(R\mathbf{x}_{Z_A+1}, \ldots, R\mathbf{x}_{Z_A+Z_B}) = \Psi_B^0(\mathbf{x}_{Z_A+1}, \ldots, \mathbf{x}_{Z_A+Z_B})$ for all rotations $R \in SO(3)$. Thus the hypotheses of Lemma 5.2.1 are satisfied, which allows us to conclude that

$$|\langle \Psi_A^0 \otimes \Psi_B^0 | \Big(\sum_{l=2}^{\tilde{L}} Q_l\Big) \chi_{\Omega_d} | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3N})}| = 0,$$

and by choosing $(d+1) = 1/4|\mathbf{R}|^{1/2} < 1/4|\mathbf{R}|$, we obtain

$$\begin{aligned}
&\langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3(Z_A+Z_B)})} \\
\leq& Z_A\, Z_B \frac{1}{|\mathbf{R}|}\left(\frac{1}{|\mathbf{R}|}\right)^{\frac{\tilde{L}+1}{2}} + C_1(1+\frac{1}{|\mathbf{R}|})\mathrm{e}^{-\tilde{C}'1/4\sqrt{|\mathbf{R}|}},
\end{aligned}$$

so that the assertion follows by choosing $\tilde{L} = 2L-3$ and $C_2 = 1/4\tilde{C}'$. □

Lemma 5.2.3. *Assume the hypotheses of Theorem 3.0.6. Then*

$$\lim_{R\to\infty}\left(R^k \langle \Psi_0 | Q_R | \Psi_0 \rangle \Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0 \otimes \Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0 \otimes \Omega)\|^2\Big)\right) = 0$$

for any $k \geq 0$ and any $\sigma \geq 0$, and

$$\lim_{R\to\infty}\lim_{\sigma\to 0}\left(R^k \langle \Psi_0 | Q_R | \Psi_0 \rangle \Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0 \otimes \Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0 \otimes \Omega)\|^2\Big)\right) = 0$$

for any $k \geq 0$.

Proof. Using the fiber decomposition of the reduced resolvents $T^{\sigma}_{A,B}$ (see Lemma 4.2.7) and rotation invariance of the operators $H_{A,B}$ (Lemma 4.2.12), one finds

$$\begin{aligned}
&\Big(\|T^{\sigma}_{A}H'_{\sigma,A}(\Psi^0_A\otimes\Omega)\|^2+\|T^{\sigma}_{B}H'_{\sigma,B}(\Psi^0_B\otimes\Omega)\|^2\Big)\\
=&\frac{\hbar}{3}\int_{\Omega_\sigma}\frac{|\rho(\mathbf{k})|^2}{\omega(\mathbf{k})}\Bigg[\Big\langle\sum_{i_A}\mathbf{p}_{i_A}\Psi^0_A\mid|(H_A+\hbar\omega(\mathbf{k}))^{-2}||\sum_{j_A}\mathbf{p}_{j_A}\Psi^0_A\Big\rangle_{\mathcal{H}_A}\\
&+\Big\langle\sum_{i_B}\mathbf{p}_{i_B}\Psi^0_B\mid|(H_B+\hbar\omega(\mathbf{k}))^{-2}||\sum_{j_B}\mathbf{p}_{j_B}\Psi^0_A\Big\rangle_{\mathcal{H}_B}\Bigg],
\end{aligned}$$

which is independent of $\mathbf{R}$. Note that the $\mathbf{k}$-integrals converge, since $\rho\in\mathcal{S}(\mathbb{R}^3)$ and the inner products in the integrand are uniformly bounded with respect to $\mathbf{k}$, which follows from the resolvent estimates

$$\|(H_{A,B}+\hbar\omega(\mathbf{k}))^{-2}\|\leq\frac{1}{\Delta_{A,B}}.$$

By dominated convergence, the $(\sigma\to 0)$-limit exists, and the assertions now follow from Lemma 5.2.2. □

5.3 Analysis of the London term

5.3.1 Error estimate for the London term

The preceding results of this chapter allow us to analyze the large R-asymptotics of the term

$$-\langle Q_R\Psi_0|T^{\sigma}|Q_R\Psi_0\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}},$$

which arises as the second-order energy correction with respect to the interatomic Coulomb potential in the non-QED context of Friesecke and Gardner ([Fri], [Gar07]). First note that since $Q_R\Psi_0\in(\mathcal{H}_A\otimes\mathcal{H}_B)\widehat{\otimes}\{\Omega\}$, the invariance properties of T^{σ} (see Lemma 4.2.5) imply

$$\begin{aligned}
&\big\langle Q_R\Psi_0|T^{\sigma}|Q_R\Psi_0\big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}}\\
=&\big\langle Q_R(\Psi^0_A\otimes\Psi^0_B)|\left((\overline{H_A+H_B})_{|\{\Psi^0_A\otimes\Psi^0_B\}^{\perp}}\right)^{-1}|Q_R(\Psi^0_A\otimes\Psi^0_B\big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B},
\end{aligned}\tag{5.3.1}$$

which shows that this term is independent of the infrared regularization parameter σ. Our goal in this section is to estimate the error made by replacing (5.3.1) with the lowest-order contribution of its multipole expanded version, the so-called London term, which is formally given by

$$\big\langle Q_2(\Psi^0_A\otimes\Psi^0_B)|\left((\overline{H_A+H_B})|_{\{\Psi^0_A\otimes\Psi^0_B\}^{\perp}}\right)^{-1}|Q_2(\Psi^0_A\otimes\Psi^0_B)\big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B},\tag{5.3.2}$$

where

$$\begin{aligned}
Q_2=&\frac{1}{4\pi}\sum_{i_A,j_B}\int_{\mathbb{R}^6}dydy'\psi(y)\,\psi(y')\\
&\times\frac{1}{|\mathbf{R}+y-y'|^3}\Big[\mathbf{x}_{i_A}\cdot\Big(I_{\mathbb{R}^3}-3(\widehat{\mathbf{R}+y-}y')\otimes(\widehat{\mathbf{R}+y-}y')\Big)\mathbf{x}_{j_B}\Big]
\end{aligned}$$

is the (smeared) dipole operator, see Section 5.1. However, to obtain a convergent expression for the multipole expansion, we introduce a spatially cutoff version of (5.3.2), which is then compared to (5.3.1).

Lemma 5.3.1. *Assume the hypotheses of Theorem 3.0.6. Let* $R_0 = 6\,\mathrm{diam}\,\mathrm{supp}\,\psi$ *and* $R > R_0$. *Choose* $0 < d \leq R/4$, *define the set* $\Omega_d := \{|\mathbf{x}_i| \leq d, i = 1, \ldots, N\} \subset \mathbb{R}^{3N}$ *and let* χ_{Ω_d} *be a smooth characteristic function of* Ω_d *as in Lemma 5.1.5.*
Then for any $L \in \mathbb{N}$, $L \geq 2$, *there exist positive constants* C_1, C_2, γ *(independent of* R *and* d *but depending on the ultraviolet-cutoff* Λ *via properties of the atomic Hamiltonians* $H_{A,B}$*), such that*

$$\begin{aligned}
&\Big|\Big\langle Q_R(\Psi_A^0 \otimes \Psi_B^0)| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} |Q_R(\Psi_A^0 \otimes \Psi_B^0)\Big\rangle \\
&- \Big\langle Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} |Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)\Big\rangle\Big| \qquad (5.3.3)\\
\leq& C_1\Big[\frac{1}{R^2}\left(\frac{4(d+1)}{R}\right)^{2L+2} + \sum_{l=2}^{L} \frac{1}{R^2}\left(\frac{4(d+1)}{R}\right)^{L+1+l}\Big] \\
&+ C_2 \mathrm{e}^{-\gamma d} + \mathcal{O}(1/R^8),
\end{aligned}$$

where the coefficients of the last contribution are independent of d.

Proof. First write

$$\Psi_A^0 \otimes \Psi_B^0 = \chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) + (1 - \chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0),$$

which leads to

$$\begin{aligned}
&\Big\langle Q_R(\Psi_A^0 \otimes \Psi_B^0)| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} |Q_R(\Psi_A^0 \otimes \Psi_B^0)\Big\rangle \\
=&\Big\langle Q_R\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} |Q_R\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)\Big\rangle \qquad (5.3.4)\\
&+ \Big\langle Q_R(1 - \chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0)| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} |Q_R(1 - \chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0)\Big\rangle \qquad (5.3.5)\\
&+ 2\mathrm{Re}\Big[\Big\langle Q_R(1 - \chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0)| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} |Q_R\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)\Big\rangle\Big]. \qquad (5.3.6)
\end{aligned}$$

By Proposition 2.5.1 and the assumptions on χ_{Ω_d}, the functions $\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)$ and $(1 - \chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0)$ are elements of $H^1(\mathbb{R}^{3N})$. Furthermore, Proposition 2.5.1 iv) a) guarantees the pointwise exponential decay of Ψ_A^0 and Ψ_B^0. Thus we can apply A.3.1 b) (note that that $\Psi_A^0 \otimes \Psi_B^0$ is an eigenfunction of $H_A + H_B$ corresponding to the eigenvalue 0 and that the smeared Coulomb potentials occurring in H_A and H_B satisfy the relevant assumptions therein), to infer the existence of positive constants C and γ', independent of d, such that

$$\|(1 - \chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \leq C\mathrm{e}^{-\gamma' d}.$$

By Lemma A.1.1, Q_R is a bounded operator from $H^1(\mathbb{R}^{3N})$ to $L^2(\mathbb{R}^{3N})$, and its operator norm $\|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}$ can be bounded from above by

$$C_Q(1+1/R) < C_Q(1+1/R_0) =: C',$$

where $C_Q > 0$ (and thus also C') is independent of R and Λ. This allows us to estimate (5.3.5) and (5.3.6) as follows:

$$\begin{aligned}
&\Big|\Big\langle Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big|\\
\leq&\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|^2_{H^1(\mathbb{R}^{3N})}\|Q_R\|^2_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|(\overline{H_A+H_B})^{-1}\|_{L^2(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\\
\leq&C^2\,\mathrm{e}^{-2\gamma' d}(C')^2\,\|(\overline{H_A+H_B})^{-1}\|_{L^2(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})},\\
&\Big|\Big\langle Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big|\\
\leq&C\,\mathrm{e}^{-\gamma' d}(C')^2\|(\overline{H_A+H_B})^{-1}\|_{L^2(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})}\\
\leq&C\,\mathrm{e}^{-\gamma' d}(C')^2\|(\overline{H_A+H_B})^{-1}\|_{L^2(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|\Psi_A^0\otimes\Psi_B^0\|_{H^1(\mathbb{R}^{3N})},
\end{aligned}$$

where we have used $\|\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})}\leq\|\Psi_A^0\otimes\Psi_B^0\|_{H^1(\mathbb{R}^{3N})}$ in the last step.
It remains to investigate the term 5.3.4. Lemma 5.1.1 yields the convergence of the multipole expansion on $\operatorname{supp}\chi_{\Omega_d}$, which means that we have

$$\begin{aligned}
&\Big\langle Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|(\overline{H_A+H_B})^{-1}|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
=&\Big\langle\Big(\sum_{l=2}^{L}Q_l+\sum_{l=L+1}^{\infty}Q_l\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big|(\overline{H_A+H_B})^{-1}\Big|\\
&\qquad\Big|\Big(\sum_{m=2}^{L}Q_m+\sum_{m=L+1}^{\infty}Q_m\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
=&\sum_{l,m=2}^{L}\Big\langle Q_l\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|(\overline{H_A+H_B})^{-1}|Q_m\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle \qquad (5.3.7)\\
&+\Big\langle\Big(\sum_{l=L+1}^{\infty}Q_l\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|(\overline{H_A+H_B})^{-1}|\Big(\sum_{m=L+1}^{\infty}Q_m\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle \qquad (5.3.8)\\
&+2\mathrm{Re}\Big[\Big\langle\Big(\sum_{l=2}^{L}Q_l\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|(\overline{H_A+H_B})^{-1}|\Big(\sum_{m=L+1}^{\infty}Q_m\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big]. \qquad (5.3.9)
\end{aligned}$$

The tail estimate established in the proof of Lemma 5.1.4 leads to

$$
\begin{aligned}
&|(5.3.8)+(5.3.9)| \\
\leq&\left[\left(Z_AZ_B\frac{1}{R}\left(\frac{4(d+1)}{R}\right)^{L+1}\right)^2+Z_AZ_B\frac{1}{R}\left(\frac{4(d+1)}{R}\right)^{L+1}\left|\sum_{l=2}^{L}Q_l(\mathbf{x}_1,\dots,\mathbf{x}_N)\right|\right] \\
&\times\|\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\|_{L^2(\mathbb{R}^{3N})}\|(\overline{H_A+H_B})^{-1}\|_{L^2(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})} \\
\leq&\left[\left(Z_AZ_B\frac{1}{R}\left(\frac{4(d+1)}{R}\right)^{L+1}\right)^2+\frac{1}{4\pi}(Z_AZ_B)^2\sum_{l=2}^{L}C_l\frac{1}{R^2}\left(\frac{4(d+1)}{R}\right)^{l}\right] \\
&\times\|\Psi_A^0\otimes\Psi_B^0\|_{L^2(\mathbb{R}^{3N})}\|(\overline{H_A+H_B})^{-1}\|_{L^2(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})},
\end{aligned}
$$

where we have defined the constants $C_l := 3\left(\frac{3}{8}\right)^l+\frac{3}{2}\left(\frac{3}{4}\right)^l$ and used the fact that

$$
\|\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\|_{L^2(\mathbb{R}^{3N})}\leq\|\Psi_A^0\otimes\Psi_B^0\|_{L^2(\mathbb{R}^{3N})}
$$

by dominated convergence. Finally, we show that

$$
(5.3.7)=\left\langle Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|(\overline{H_A+H_B})^{-1}|Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\right\rangle+\mathcal{O}(1/R^8),
$$

where the coefficients of the $\mathcal{O}(1/R^8)$ can be chosen to be independent of d. To see this, note that in view of Lemma 5.1.3 (or again the estimates established in the proof of Lemma 5.1.4), we have

$$
\left\langle Q_l\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|(\overline{H_A+H_B})^{-1}|Q_m\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\right\rangle=\mathcal{O}(1/R^{l+m+2}),
$$

and the coefficients on the right-hand side can be bounded independently of d by enlarging the domains of integration from $\operatorname{supp}\chi_{\Omega_d}$ to $\mathbb{R}^{3N}$, yielding convergent expressions thanks to the exponential decay of Ψ_A^0 and Ψ_B^0. Thus what is left to show is that the term

$$
2\mathrm{Re}\Big[\langle Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}|Q_3\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\rangle\Big],
$$

with

$$
\begin{aligned}
Q_2=&\frac{1}{4\pi}\sum_{i_A,j_B}\int_{\mathbb{R}^6}dydy'\psi(y)\,\psi(y') \\
&\times\frac{1}{|\mathbf{R}+y-y'|^3}\Big[\mathbf{x}_{i_A}\cdot\Big(I_{\mathbb{R}^3}-3(\widehat{\mathbf{R}+y-y'})\otimes(\widehat{\mathbf{R}+y-y'})\Big)\mathbf{x}_{j_B}\Big]
\end{aligned}
$$

and

$$
\begin{aligned}
Q_3=\sum_{i_A,j_B}\Bigg[&\sum_{m=-3}^{3}\sum_{l_1=0}^{3}\sum_{m_1=-l_1}^{l_1}|\mathbf{x}_{i_A}|^{l_1}\,|\mathbf{x}_{j_B}|^{3-l_1}\,Y^*_{l_1m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\,Y^*_{(3-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \\
&\times u_{3,m,l_1,m_1}(\mathbf{R}) \qquad (5.3.10) \\
&-\sum_{m=-3}^{3}\Big(|\mathbf{x}_{i_A}|^3\,Y^*_{3m}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]+(-1)^3|\mathbf{x}_{j_B}|^3\,Y^*_{3m}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\Big)\,v_{3,m}(\mathbf{R})\Bigg], \qquad (5.3.11)
\end{aligned}
$$

vanishes, since it is the only one that could yield contributions asymptotically proportional to $1/R^7$. Since Ψ_A^0 and Ψ_B^0 are non-degenerate, they are eigenfunctions of the parity operators P_A and P_B (acting on the variables $\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A}$ and $\mathbf{x}_{Z_A+1}, \ldots, \mathbf{x}_N$) with eigenvalues ε_A and ε_B, respectively. By construction, χ_{Ω_d} has parity 1 with respect to both P_A and P_B. Now $P_A[Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)] = -\varepsilon_A\, Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)$ and $P_B[Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)] = -\varepsilon_B\, Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)$, as is easily seen from the structure of Q_2. In particular, $Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) \in \{\Psi_A^0\}^\perp \otimes \{\Psi_B^0\}^\perp$. On the other hand, for $l_1 = 1$, $3 - l_1 = 2$, the terms $Y^*_{1m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\, Y^*_{2(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}](\Psi_A^0 \otimes \Psi_B^0)$ have parity ε_B with respect to P_B, since $Y^*_{2(m-m_1)}$ has parity $(-1)^2 = 1$. The same argument holds for the case $l_1 = 2$, $l - l_1 = 1$, with ε_A and P_A instead. Since the operator $\left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1}$ leaves the eigenspaces of P_A and P_B invariant individually (see Lemma 4.2.11), the contributions from (5.3.10) vanish.
The two terms in (5.3.11) both only depend on either of the variables $\mathbf{x}_{i_A}$ and $\mathbf{x}_{j_B}$, which implies that the product of (5.3.11) and $\Psi_A^0 \otimes \Psi_B^0$ is a sum consisting of one term from $\mathcal{H}_A \otimes \{\Psi_B^0\}$ and one term from $\{\Psi_A^0\} \otimes \mathcal{H}_B$. Since $\{\Psi_A^0\} \otimes \mathcal{H}_B$ and $\mathcal{H}_A \otimes \{\Psi_B^0\}$ are invariant subspaces of $\left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1}$ (see Lemma 4.2.5) and $Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) \in \{\Psi_A^0\}^\perp \otimes \{\Psi_B^0\}^\perp$ by the above, the contributions from (5.3.11) also vanish. □

5.3.2 Integral representation of the London term

Define

$$\tilde{L}(d) := \sum_{\alpha,\beta=1}^{3} \left\langle \chi_{\Omega_d}\left(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta\right) | \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} | \chi_{\Omega_d}\left(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta\right) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}$$

and recall the definition of the operator

$$\begin{aligned} Q_2 =& \frac{1}{4\pi} \sum_{i_A, j_B} \int_{\mathbb{R}^6} dy dy' \psi(y)\, \psi(y') \\ & \times \frac{1}{|\mathbf{R} + y - y'|^3} \left[\mathbf{x}_{i_A} \cdot \left(I_{\mathbb{R}^3} - 3(\widehat{\mathbf{R} + y - y'}) \otimes (\widehat{\mathbf{R} + y - y'}) \right) \mathbf{x}_{j_B} \right] \end{aligned}$$

from the multipole expansion of Q_R. As the next result shows, we can convert the London term (5.3.3) into an integral over photon momenta, thereby putting it on equal footing with the perturbation terms generated by the radiation field. For this step, it is essential that Q_R contains the smeared Coulomb potential, since only then the operator Q_2 has a momentum space representation via the distributional Fourier transform, see (5.1.2). As noted in the introduction, this will also play a role in the (asymptotic) cancellation at order $1/R^6$, which will be discussed in detail in Section 6.6.

Lemma 5.3.2. *Assume the hypotheses of Theorem 3.0.6 and let χ_{Ω_d} be as in Lemma 5.3.1. Then*

$$\begin{aligned} & \left\langle Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) | \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp}\right)^{-1} | Q_2\chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \\ =& \frac{1}{9}\tilde{L}(d) \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\, |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot\mathbf{R}} (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2. \end{aligned}$$

Proof. By (5.1.2), $Q_2 = \tilde{Q}$, where

$$\tilde{Q} = \sum_{i_A, j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\rho(\mathbf{k})|^2}{|\mathbf{k}|^2} (\mathbf{x}_{i_A} \cdot \mathbf{k})(\mathbf{x}_{j_B} \cdot \mathbf{k}) \mathrm{e}^{-\mathrm{i}\mathbf{k}\mathbf{R}}.$$

Exploiting rotation invariance (Lemma 4.2.16) and recalling the definition of the dipole moments $\mathbf{v}_{A,B}$ from (3.0.4), we find

$$\begin{aligned}
&\left\langle \tilde{Q}\, \chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) \middle| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \middle| \tilde{Q}\, \chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0 \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \\
&= \sum_{i_A, j_B, k_A, l_B} \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2\, |\rho(\mathbf{k}_2)|^2}{|\mathbf{k}_1|^2\, |\mathbf{k}_2|^2} \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}} \\
&\qquad \left\langle \chi_{\Omega_d}\left((\mathbf{x}_{i_A} \cdot \mathbf{k}_1) \Psi_A^0 \otimes (\mathbf{x}_{j_B} \cdot \mathbf{k}_1) \Psi_B^0 \right) \middle| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \middle| \right. \\
&\qquad\quad \left. \chi_{\Omega_d}\left((\mathbf{x}_{k_A} \cdot \mathbf{k}_2) \Psi_A^0 \otimes (\mathbf{x}_{l_B} \cdot \mathbf{k}_2) \Psi_B^0 \right) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}
\end{aligned}$$

$$\begin{aligned}
&= \frac{1}{9} \sum_{\alpha, \beta = 1}^{3} \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2\, |\rho(\mathbf{k}_2)|^2}{|\mathbf{k}_1|^2\, |\mathbf{k}_2|^2} \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}} (\mathbf{k}_1 \cdot \mathbf{k}_2)^2 \\
&\qquad \left\langle \chi_{\Omega_d}\left(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \right) \middle| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \middle| \chi_{\Omega_d}\left(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \right) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B} \\
&= \frac{1}{9} \sum_{\alpha, \beta = 1}^{3} \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\, |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}} (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2 \\
&\qquad \left\langle \chi_{\Omega_d}\left(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \right) \middle| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \middle| \chi_{\Omega_d}\left(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \right) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}.
\end{aligned} \tag{5.3.12}$$

□

5.4 Decomposition and analysis of the mixed terms $M_A(\mathbf{R}, \sigma)$ and $M_B(\mathbf{R}, \sigma)$

The remainder of Chapter 5 is devoted to the analysis of the terms $M_A(\mathbf{R}, \sigma)$ and $M_B(\mathbf{R}, \sigma)$ from $V_4^\sigma(\Lambda, \mathbf{R})$ (see (3.0.17)), which contain both the quantized radiation field and the (smeared) interatomic Coulomb potential.
In the present section we state a result providing a representation of these terms which will later be used to identify their contributions at orders $1/R^6$ and $1/R^7$ and to provide error estimates for the remaining ones. Its proof, which is split into a series of lemmas, will be given in the remaining sections of this chapter.

Theorem 5.4.1. *Assume the hypotheses of Theorem 3.0.6. Let* $R_0 = 6\,\mathrm{diam}\,\mathrm{supp}\,\psi$ *and* $R > R_0$. *Choose* $0 < d \leq R/4$. *Then there exist positive constants* $C_1(l)$, $C_2(l)$, C, γ, *independent of* $\mathbf{R}$, σ *and* d *(but depending on* Λ *via* ρ *and properties of* $H_{A,B}$*), such that for any* $L \in \mathbb{N}$, $L \geq 2$, *we have the representation*

$$\begin{aligned} &M_A(\mathbf{R},\sigma) + M_B(\mathbf{R},\sigma) \\ =&M_6(\mathbf{R},\sigma,d) + M_7(\mathbf{R},d) + M_{89}^L(\mathbf{R},d) \\ &+ M_{IN,ERR}^L(\mathbf{R},d) + M_{OUT}(\mathbf{R},\sigma,d) + M_{IR}^L(\mathbf{R},\sigma,d), \end{aligned}$$

and its contributions (which will be defined below) have the following properties:

- $\lim_{\sigma\to 0} M_6(\mathbf{R},\sigma,d)$ *exists. Furthermore,*

$$\lim_{R\to\infty}\left(R^k \lim_{\sigma\to 0} M_6(\mathbf{R},\sigma,R^{1/2})\right) = 0 \tag{5.4.1}$$

for $k < 6$, *and*

$$\lim_{R\to\infty}\left(R^6 \lim_{\sigma\to 0} M_6(\mathbf{R},\sigma,R^{1/2})\right) = \frac{1}{3(2\pi)^2} L(\infty) \tag{5.4.2}$$

(see (3.0.5) for the definition of $L(\infty)$*).*

- $$\lim_{R\to\infty}\left(R^k M_7(\mathbf{R},d)\right) = 0 \quad \textit{uniformly in } d > 0 \textit{ for any } k < 7,$$
$$\lim_{R\to\infty}\left(R^7 M_7(\mathbf{R},R^{1/2})\right) = -\frac{32}{9}\frac{\hbar c}{(2\pi)^3}\alpha_E^A(0)\,\alpha_E^B(0).$$

- $$\lim_{R\to\infty}(R^k\, M_{89}^L(\mathbf{R},d) = 0, \quad \textit{for any } k < 8, \textit{ uniformly in } d > 0. \tag{5.4.3}$$

- $$\begin{aligned} \left|M_{IR}^L(\mathbf{R},\sigma,d)\right| \leq &\sum_{l\geq 4, l \textit{ even}}^{L} \left(\frac{3}{2R}\right)^{l+1} \sup_{s\in[0,\sigma/(\Lambda c)]} |\rho_0(s)|^2 C_1(l)\left(\frac{\sigma}{c}\right)^3 \\ &+ \sum_{l\geq 2, l \textit{ even}}^{L} \sup_{s\in[0,\sigma/(\Lambda c)]} |\rho_0(s)|^2 \left(\frac{3}{2R}\right)^{l+1} C_2(l)\left(\frac{\sigma}{c}\right)^4. \end{aligned} \tag{5.4.4}$$

  In particular,

$$\lim_{\sigma\to 0} M_{IR}^L(\mathbf{R},\sigma,d) = 0.$$

- $$M_{IN,ERR}^L(\mathbf{R},d) = \mathcal{O}\left(\frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{L+1}\right),$$

  with coefficients that depend on the ultraviolet-cutoff scale Λ *via* $\rho_0(\mathbf{k}/\Lambda)$ *and properties of* $H_{A,B}$, *but which are independent of* σ.

- $$|M_{OUT}(\mathbf{R},\sigma,d)| \leq C(1+1/R)\mathrm{e}^{-\gamma d}.$$

  Furthermore,

  $$\lim_{\sigma\to 0}\left(M_{OUT}(\mathbf{R},\sigma,d)\right)$$

  exists and satisfies the same estimate.

5.5 Proof of Theorem 5.4.1

5.5.1 Some definitions

To prepare for the following, we split the terms $M_A(\mathbf{R},\sigma)$ and $M_B(\mathbf{R},\sigma)$ into a spatially cutoff 'inner part' and a corresponding 'outer part'. Set $\Omega_d := \{|\mathbf{x}_1| \leq d, \ldots |\mathbf{x}_N| \leq d\} \subset \mathbb{R}^{3N}$ and let $\chi_{\Omega_d} = \Pi_{i=1}^N \chi d(\cdot_i)$ be a smooth, rotation-invariant characteristic function of Ω_d as in Section 5.1 (respectively Section 5.3).

Definition 5.5.1. Using the notational conventions introduced in Section 3, we define

$$\begin{aligned}
&A_{IN}(\mathbf{R},\sigma,d)\\
&:= \frac{1}{\hbar^2}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\\
&\times\Bigg[\Big\langle \mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\quad-\hbar\omega(\mathbf{k})\Big\langle (H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\quad-\hbar\omega(\mathbf{k})\Big\langle \mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle (H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\otimes\Psi_B^0\Big\rangle\\
&\quad+\Big\langle \Psi_A^0\otimes\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle\\
&\quad-\hbar\omega(\mathbf{k})\Big\langle \Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|\Psi_A^0\otimes\mathbf{v}_B\Big\rangle\\
&\quad-\hbar\omega(\mathbf{k})\Big\langle \Psi_A^0\otimes\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\Big\rangle\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle \Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B|(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})Q_R\chi_{\Omega_d}|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\Big\rangle\Bigg]\\
&+2\mathrm{Re}\Bigg[\int_{\Omega_\sigma} d\mathbf{k}\left(\frac{-\mathrm{i}}{m_e\hbar}\right)|C(\mathbf{k})|^2\\
&\times\Bigg[\Big\langle (H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big((\sum_{j_A}\mathbf{p}_{j_A})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_A\Big)\otimes\Psi_B^0|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&\quad-\hbar\omega(\mathbf{k})\Big\langle (H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big((\sum_{j_A}\mathbf{p}_{j_A})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A\Big)\otimes\Psi_B^0\Big|\\
&\qquad\qquad\Big|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle
\end{aligned}$$

$$+\Big\langle \Psi_A^0 \otimes (H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big((\sum_{j_B}\mathbf{p}_{j_B})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle$$
$$-\hbar\omega(\mathbf{k})\Big\langle \Psi_A^0 \otimes (H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big((\sum_{j_B}\mathbf{p}_{j_B})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\Big)\Big|$$
$$\Big|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big]\Big]$$

and

$$B_{IN}(\mathbf{R},\sigma,d)$$
$$:=\frac{2}{\hbar^2}\mathrm{Re}\Big[\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}$$
$$\times\Big[-2\hbar\omega(\mathbf{k})\Big\langle\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B|(\overline{H_A+H_B})^{-1}|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle$$
$$+(\hbar\omega(\mathbf{k}))^2\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0|Q_R\chi_{\Omega_d}\Big|$$
$$\Big|\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\Big\rangle$$
$$+(\hbar\omega(\mathbf{k}))^2\Big\langle\big((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\big)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)|(\overline{H_A+H_B})^{-1}\Big|$$
$$\Big|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle$$
$$+(\hbar\omega(\mathbf{k}))^2\Big\langle\big(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\big)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)|(\overline{H_A+H_B})^{-1}\Big|$$
$$\Big|Q_R\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big]\Big].$$

The terms $A_{OUT}(\mathbf{R},\sigma,\mathbf{d})$ and $B_{OUT}(\mathbf{R},\sigma,d)$ are defined analogously by replacing χ_{Ω_d} with $1-\chi_{\Omega_d}$. Set

$$M_{OUT}(\mathbf{R},\sigma,d):=A_{OUT}(\mathbf{R},\sigma,\mathbf{d})+B_{OUT}(\mathbf{R},\sigma,d).$$

Obviously,

$$M_A(\mathbf{R},\sigma)+M_B(\mathbf{R},\sigma)=A_{IN}(\mathbf{R},\sigma,\mathbf{d})+B_{IN}(\mathbf{R},\sigma,d)+M_{OUT}(\mathbf{R},\sigma,d). \tag{5.5.1}$$

The proof of theorem 5.4.1 is divided into the following series of lemmas. For an overview of its strategy and the methods employed, see Section 1.3 of the introduction.

5.5.2 Multipole expansion of the Coulomb part

The first step in the proof of Theorem 5.4.1 is to employ the multipole expansion of the smeared interatomic Coulomb potential. Recalling the definition of the terms Q_l from Section 5.1 and letting χ_{Ω_d} be as in the preceding section, we make the following

Definition 5.5.2. Set

$$
\begin{aligned}
&\tilde{B}_l(\mathbf{R},\sigma,d)\\
&:=\frac{2}{\hbar^2}\mathrm{Re}\Bigg[\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\quad\times\Bigg[-2\hbar\omega(\mathbf{k})\Big\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\Big|\\
&\qquad\qquad\Big|Q_l\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\Omega_d)}\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0\Big|\\
&\qquad\qquad\Big|Q_l\chi_{\Omega_d}\left(\Psi_A^0\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle(\overline{H_A+H_B})^{-1}\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\right)[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\Big|\\
&\qquad\qquad\Big|Q_l\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\qquad+(\hbar\omega(\mathbf{k}))^2\Big\langle(\overline{H_A+H_B})^{-1}\left(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\Big|\\
&\qquad\qquad\Big|Q_l\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Bigg]\Bigg],
\end{aligned}
\tag{5.5.2}
$$

and define $\tilde{A}_l(\mathbf{R},\sigma,d)$ analogously, replacing Q_R by Q_l in $A_{IN}(\mathbf{R},\sigma,d)$. Furthermore, for $L\in\mathbb{N}$, $L\geq 2$, define

$$
M^L_{IN,ERR}(\mathbf{R},d):=A_{IN}(\mathbf{R},\sigma,d)+B_{IN}(\mathbf{R},\sigma,d)-\sum_{l=2}^{L}\left(\tilde{A}_l(\mathbf{R},\sigma,d)+\tilde{B}_l(\mathbf{R},\sigma,d)\right). \tag{5.5.3}
$$

Lemma 5.5.3 (Multipole error estimate for mixed terms, inside)**.** *Assume the hypotheses of Theorem 5.4.1. Then for any $L\in N$, $L\geq 2$,*

$$
M^L_{IN,ERR}(\mathbf{R},d)=\mathcal{O}\left(\frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{L+1}\right),
$$

where the higher-order coefficients depend on the ultraviolet-cutoff scale Λ via $\rho_0(\mathbf{k}/\Lambda)$ and properties of $H_{A,B}$, but are independent of σ.

Proof. By Lemma 5.1.1, the multipole expansion of Q_R converges on $\operatorname{supp}\chi_{\Omega_d}$, yielding

$$
\begin{aligned}
&\left|B_{IN}(\mathbf{R},\sigma,d)-\sum_{l=2}^{L}\tilde{B}_l(\mathbf{R},\sigma,d)\right| \\
\leq&\frac{2}{\hbar^2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2 \\
&\times\Bigg[2\hbar\omega(\mathbf{k})\Big|\Big\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\Big|\Big(\sum_{l=L+1}^{\infty}Q_l\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Big| \\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big|\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0\Big| \\
&\qquad\qquad\Big|\Big(\sum_{l=L+1}^{\infty}Q_l\Big)\chi_{\Omega_d}\left(\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Big| \\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big|\Big\langle(\overline{H_A+H_B})^{-1}[\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\right)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)]\Big| \\
&\qquad\qquad\Big|\Big(\sum_{l=L+1}^{\infty}Q_l\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Big| \\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big|\Big\langle(\overline{H_A+H_B})^{-1}[\left(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)\Big(\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B\Big)]\Big| \\
&\qquad\qquad\Big|\Big(\sum_{l=L+1}^{\infty}Q_l\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Big|\Bigg].
\end{aligned}
$$

Note that the $\mathbf{k}$-integrals converge due to the ultraviolet-cutoff ρ contained in $C(\mathbf{k})$. Carrying out the vector operations inside the matrix elements, noting that $\operatorname{supp}\chi_{\Omega_d}\subset\Omega_{d+1}$ and applying Lemma 5.1.4, we obtain the estimate

$$
\begin{aligned}
&\left|B_{IN}(\mathbf{R},\sigma,d)-\sum_{l=2}^{L}\tilde{B}_l(\mathbf{R},\sigma,d)\right| \\
\leq&\frac{2}{\hbar^2}\int_{\Omega_\sigma}|C(\mathbf{k})|^2\,Z_AZ_B\frac{3}{4\pi}\frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{L+1}\Bigg[\sum_{\alpha,\beta=1}^{3}\left|\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\right| \\
&\quad\times\Bigg[(\hbar\omega(\mathbf{k}))^2\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\|_{L^2(\Omega_{A,d+1})}\|\Psi_B^0\|_{L^2(\Omega_{B,(d+1)})}\|\Psi_A^0\|_{L^2(\Omega_{A,(d+1)})} \\
&\qquad\times\|(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\|_{L^2(\Omega_{B,d+1})} \\
&\qquad+(\hbar\omega(\mathbf{k}))^2\|(\overline{H_A+H_B})^{-1}((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I)\mathbf{v}_A^\alpha\mathbf{v}_B^\beta\|_{L^2(\Omega_{d+1})} \\
&\qquad\times\|\Psi_A^0\otimes\Psi_B^0\|_{L^2(\Omega_{d+1})} \\
&\qquad+(\hbar\omega(\mathbf{k}))^2\|(\overline{H_A+H_B})^{-1}(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1})\mathbf{v}_A^\alpha\mathbf{v}_B^\beta\|_{L^2(\Omega_{d+1})} \\
&\qquad\times\|\Psi_A^0\otimes\Psi_B^0\|_{L^2(\Omega_{d+1})} \\
&\qquad+2\hbar\omega(\mathbf{k})\|(\overline{H_A+H_B})^{-1}\mathbf{v}_A^\alpha\mathbf{v}_B^\beta\|_{L^2(\Omega_{d+1})}\|\Psi_A^0\otimes\Psi_B^0\|_{L^2(\Omega_{d+1})}\Bigg]\Bigg],
\end{aligned}
$$

where $\Omega_{A,d}=\{|\mathbf{x}_1|\leq,\ldots,|\mathbf{x}_{Z_A}|\leq d\}$, $\Omega_{B,d}=\{|\mathbf{x}_{Z_A+1}|\leq,\ldots,|\mathbf{x}_N|\leq d\}$.

We have $|\delta_{\alpha,\beta} - \frac{k_\alpha k_\beta}{|\mathbf{k}|^2}| \leq 2$ for all $\mathbf{k} \in \mathbb{R}^3 \setminus \{0\}$. Since $\Psi_A^0, \mathbf{v}_A^\alpha \in L^2(\mathbb{R}^{3Z_A})$ and $\Psi_B^0, \mathbf{v}_B^\beta \in L^2(\mathbb{R}^{3Z_B})$ (for $\mathbf{v}_A^\alpha$ and $\mathbf{v}_B^\beta$ this follows from the exponential decay of the atomic ground states, see Proposition 2.5.1), all L^2-norms on the bounded subsets $\Omega_{A,d+1}$, $\Omega_{B,d+1}$ and Ω_{d+1} can be estimated from above by the corresponding L^2-norms on the whole space. Using the fact that Ψ_A^0 and Ψ_B^0 are normalized, this leads to

$$
\begin{aligned}
&\left| B_{IN}(\mathbf{R},\sigma,d) - \sum_{l=2}^{L} \tilde{B}_l(\mathbf{R},\sigma,d) \right| \\
\leq &\frac{4}{\hbar^2} \int_{\Omega_\sigma} |C(\mathbf{k})|^2 \, Z_A Z_B \frac{3}{4\pi} \frac{1}{|\mathbf{R}|} \left(\frac{4(d+1)}{|\mathbf{R}|} \right)^{L+1} \\
&\times \Bigg[\sum_{\alpha,\beta=1}^{3} \Bigg[(\hbar\omega(\mathbf{k}))^2 \|(H_A + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})} \|(H_B + \hbar\omega(\mathbf{k}))^{-1} \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})} \\
&\quad + (\hbar\omega(\mathbf{k}))^2 \|(\overline{H_A + H_B})^{-1} ((H_A + \hbar\omega(\mathbf{k}))^{-1} \otimes I) \mathbf{v}_A^\alpha \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^N)} \\
&\quad + (\hbar\omega(\mathbf{k}))^2 \|(\overline{H_A + H_B})^{-1} (I \otimes (H_B + \hbar\omega(\mathbf{k}))^{-1}) \mathbf{v}_A^\alpha \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^N)} \\
&\quad + 2\hbar\omega(\mathbf{k}) \|(\overline{H_A + H_B})^{-1} \mathbf{v}_A^\alpha \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^N)} \Bigg] \Bigg].
\end{aligned}
$$

In the next step we use the resolvent norm estimates

$$
\begin{aligned}
\|(\overline{H_A + H_B})^{-1}\| &\leq 1/(\Delta_A + \Delta_B), \\
\|(H_A + \hbar\omega(\mathbf{k}))^{-1}\| &\leq 1/(\hbar\omega(\mathbf{k}) + \Delta_A) \leq 1/\Delta_A, \\
\|(H_B + \hbar\omega(\mathbf{k}))^{-1}\| &\leq 1/(\hbar\omega(\mathbf{k}) + \Delta_B) \leq 1/\Delta_B,
\end{aligned}
$$

which hold on the subspace $\{\Psi_A^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp$, $\{\Psi_A^0\}^\perp$ and $\{\Psi_B^0\}^\perp$, respectively (note that $\mathbf{v}_A^\alpha \in \{\Psi_A^0\}^\perp$ and $\mathbf{v}_B^\beta \in \{\Psi_B^0\}^\perp$ by parity). Recall that Δ_A and Δ_B are the spectral gaps of the atomic Hamiltonians H_A and H_B. Recalling the definition $|C(\mathbf{k})|^2 = (\hbar|\rho(\mathbf{k})|^2)/(2\omega(\mathbf{k}))$, we conclude

$$
\begin{aligned}
&\left| B_{IN}(\mathbf{R},\sigma,d) - \sum_{l=2}^{L} \tilde{B}_l(\mathbf{R},\sigma,d) \right| \\
\leq &Z_A Z_B \frac{3}{2\pi} \frac{1}{|\mathbf{R}|} \left(\frac{4(d+1)}{|\mathbf{R}|} \right)^{L+1} \left(\sum_{\alpha=1}^{3} \|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})} \right) \left(\sum_{\beta=1}^{3} \|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})} \right) \\
&\times \left(\int_{\Omega_\sigma} d\mathbf{k} |\rho(\mathbf{k})|^2 \left[\frac{4}{\Delta_A + \Delta_B} + 2\hbar\omega(\mathbf{k}) \left(\frac{1}{\Delta_A \, \Delta_B} + \frac{1}{\Delta_A + \Delta_B} \left(\frac{1}{\Delta_A} + \frac{1}{\Delta_B} \right) \right) \right] \right).
\end{aligned}
$$

Noting that the integrand in the $\mathbf{k}$-integral is non-negative, we can pass to the integral over all of $\mathbb{R}^3$ (which is finite since $\rho \in \mathcal{S}(\mathbb{R}^3)$), obtaining the final result

$$
\begin{aligned}
&\left|\tilde{B}(\mathbf{R},\sigma,d)-\sum_{l=2}^{L}\tilde{B}_l(\mathbf{R},\sigma,d)\right| \\
\leq & Z_AZ_B\frac{3}{2\pi}\frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{L+1}\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)\left(\sum_{\beta=1}^{3}\|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\right) \\
&\times\left(\int_{\mathbb{R}^3}d\mathbf{k}|\rho_0(\mathbf{k}/\Lambda)|^2\left[\frac{4}{\Delta_A+\Delta_B}+2\hbar\omega(\mathbf{k})\left(\frac{1}{\Delta_A\,\Delta_B}+\frac{1}{\Delta_A+\Delta_B}\left(\frac{1}{\Delta_A}+\frac{1}{\Delta_B}\right)\right)\right]\right),
\end{aligned}
$$

where we have highlighted the dependence of the coefficient on the scale Λ of the ultraviolet-cutoff.

Performing the same steps of the proof for the term $\tilde{A}_l(\mathbf{R},\sigma,d)$, we obtain

$$
\begin{aligned}
&\left|A_{IN}(\mathbf{R},\sigma,d)-\tilde{A}_l(\mathbf{R},\sigma,d)\right| \\
\leq&\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2Z_AZ_B\frac{3}{4\pi}\frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{L+1}\left[\sum_{\alpha,\beta=1}^{3}\left|\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|}\right|\right. \\
&\times\left[\frac{1}{\hbar^2}\left(\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\|\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}+\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\right)\right. \\
&\quad+\frac{2}{\hbar}\omega(\mathbf{k})\Big[\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\|\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})} \\
&\qquad\qquad+\|(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\Big] \\
&\quad+\omega(\mathbf{k})^2\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})} \\
&\quad+\omega(\mathbf{k})^2\|(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\|(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})} \\
&\quad+\frac{2}{m_e\hbar}\Big[\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}(\sum_{j_A}\mathbf{p}_{j_A}^\alpha)\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})} \\
&\qquad\qquad+\|(H_{B|\{\Psi_B^0\}^\perp})^{-1}(\sum_{j_B}\mathbf{p}_{j_B}^\alpha)\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\Big] \\
&\quad+\frac{2}{m_e}\omega(\mathbf{k})\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}(\sum_{j_A}\mathbf{p}_{j_A}^\alpha)(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})} \\
&\quad\left.+\frac{2}{m_e}\omega(\mathbf{k})\|(H_{B|\{\Psi_B^0\}^\perp})^{-1}(\sum_{j_B}\mathbf{p}_{j_B}^\alpha)(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\right]\Bigg] \\
\leq&2\int_{\Omega_\sigma}d\mathbf{k}\,|C(\mathbf{k})|^2Z_AZ_B\frac{3}{4\pi}\frac{1}{|\mathbf{R}|}\left(\frac{4d}{|\mathbf{R}|}\right)^{L+1} \\
&\times\left[\frac{1}{\hbar^2}\left(\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)^2+\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right)^2\right)\right. \\
&\quad\left.+\frac{2\omega(\mathbf{k})}{\hbar}\left(\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)^2\frac{1}{\Delta_A}+\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right)^2\frac{1}{\Delta_B}\right)\right.
\end{aligned}
$$

$$
\begin{aligned}
&+\omega(\mathbf{k})^2\left(\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)^2\frac{1}{\Delta_A^2}+\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right)^2\frac{1}{\Delta_B^2}\right)\\
&+\sum_{\alpha,\beta=1}^{3}\Bigg[\frac{2}{m_e\hbar}\Big[\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}(\sum_{j_A}\mathbf{p}_{j_A}^\alpha)\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}\\
&\qquad+\|(H_{B|\{\Psi_B^0\}^\perp})^{-1}(\sum_{j_B}\mathbf{p}_{j_B}^\alpha)\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\Big]\\
&+\frac{2}{m_e}\omega(\mathbf{k})\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}(\sum_{j_A}\mathbf{p}_{j_A}^\alpha)(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}\\
&+\frac{2}{m_e}\omega(\mathbf{k})\|(H_{B|\{\Psi_B^0\}^\perp})^{-1}(\sum_{j_B}\mathbf{p}_{j_B}^\alpha)(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\Bigg]\Bigg].
\end{aligned}
$$

By Proposition 2.5.1, $\mathbf{v}_A^\alpha \in H^2(\mathbb{R}^{3Z_A})$ and $\mathbf{v}_B^\alpha \in H^2(\mathbb{R}^{3Z_B})$. Since the $\mathbf{p}_{i_A,j_B}^\alpha$ are relatively bounded with respect to $H_{A,B}$ and $D(H_{A,B}) = H^2(\mathbb{R}^{3Z_{A,B}})$, we have

$$
\begin{aligned}
&\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}(\sum_{j_A}\mathbf{p}_{j_A}^\alpha)\psi\|_{L^2(\mathbb{R}^{3Z_A})}\\
\le& C\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}\|\left(\|\psi\|_{L^2(\mathbb{R}^{3Z_A})}+\|H_A\psi\|_{L^2(\mathbb{R}^{3Z_A})}\right)\\
\le& \tilde{C}\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}\|\,\|\psi\|_{H^2(\mathbb{R}^{3Z_A})}
\end{aligned}
$$

for any $\psi \in H^2(\mathbb{R}^{3Z_A})$, and accordingly for H_B. Note that by taking maxima if necessary, we can find constants C and $\tilde{C}$ which work for any j_A, j_B, α and both H_A and H_B. Furthermore,

$$
\begin{aligned}
&\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}+\|H_A(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}\\
\le&\frac{1}{\Delta_A}\|\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}+\|\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}+\hbar\omega(\mathbf{k})\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}\\
\le&\left(\frac{1}{\Delta_A}+1+\frac{\hbar\omega(\mathbf{k})}{\Delta_A}\right)\|\mathbf{v}_A^\beta\|_{L^2(\mathbb{R}^{3Z_A})}.
\end{aligned}
$$

Recalling $\|(H_{A|\{\Psi_A^0\}^\perp})^{-1}\| \le 1/\Delta_A$ and the definition of $C(\mathbf{k})$, we conclude

$$
\begin{aligned}
&\left|A_{IN}(\mathbf{R},\sigma,d)-\tilde{A}_l(\mathbf{R},\sigma,d)\right|\\
\le& Z_AZ_B\frac{3}{4\pi}\frac{1}{|\mathbf{R}|}\left(\frac{4d}{|\mathbf{R}|}\right)^{L+1}\int_{\Omega_\sigma}d\mathbf{k}\,|\rho_0(\mathbf{k}/\Lambda)|^2\\
&\times\Bigg[\frac{1}{\hbar\omega(\mathbf{k})}\left(\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)^2+\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right)^2\right)\\
&\quad+2\left(\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)^2\frac{1}{\Delta_A}+\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right)^2\frac{1}{\Delta_B}\right)\\
&\quad+\hbar\omega(\mathbf{k})\left(\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)^2\frac{1}{\Delta_A^2}+\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right)^2\frac{1}{\Delta_B^2}\right)
\end{aligned}
$$

$$
\begin{aligned}
&+\frac{2\tilde{C}}{m_e\omega(\mathbf{k})}\left(\frac{Z_A}{\Delta_A}\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{H^2}\right)+\frac{Z_B}{\Delta_B}\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{H^2}\right)\right)\\
&+\frac{2\hbar C}{m_e}\left(\frac{Z_A}{\Delta_A}\left(\frac{1}{\Delta_A}+1+\frac{\hbar\omega(\mathbf{k})}{\Delta_A}\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2}\right)\right.\\
&\left.+\frac{Z_B}{\Delta_B}\left(\frac{1}{\Delta_B}+1+\frac{\hbar\omega(\mathbf{k})}{\Delta_B}\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2}\right)\right)\Bigg],
\end{aligned}
$$

which is finite since $\rho\in\mathcal{S}(\mathbb{R}^3)$. □

Lemma 5.5.4 (Multipole error estimate for mixed terms, outside)**.** *Assume the hypotheses of theorem 5.4.1. Then there exist positive constants C and γ, independent of σ, $\mathbf{R}$, d (but depending on Λ via $\rho(\mathbf{k}/\Lambda)$ and the properties of $H_{A,B}$), such that*

$$
|M_{OUT}(\mathbf{R},\sigma,d)|\leq C(1+1/|\mathbf{R}|)\,\mathrm{e}^{-\gamma d}.
$$

Furthermore,

$$
\lim_{\sigma\to 0}\left(M_{OUT}(\mathbf{R},\sigma,d)\right)
$$

exists and satisfies the same estimate.

Proof. To simplify notation, we first define

$$
\begin{aligned}
&f_1^{\alpha\beta}(\mathbf{R},d)\\
&:=\left\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta]|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\right\rangle_{L^2(\mathbb{R}^{3N})},\\
&f_2^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\\
&:=\left\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\otimes\Psi_B^0|Q_R(1-\chi_{\Omega_d})\left(\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\right)\right\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\left\langle(\overline{H_A+H_B})^{-1}[(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta]|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\right\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\left\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A^\alpha\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta]|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\right\rangle_{L^2(\mathbb{R}^{3N})},\\
&f_3^{\alpha\beta}(\mathbf{R},d)\\
&:=\left\langle\mathbf{v}_A^\alpha\otimes\Psi_B^0|Q_R(1-\chi_{\Omega_d})\left(\mathbf{v}_A^\beta\otimes\Psi_B^0\right)\right\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\left\langle\Psi_A^0\otimes\mathbf{v}_B^\alpha|Q_R(1-\chi_{\Omega_d})\left(\Psi_A^0\otimes\mathbf{v}_B^\beta\right)\right\rangle_{L^2(\mathbb{R}^{3N})},\\
&f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\\
&:=2\,\mathrm{Re}\Big[\left\langle(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\otimes\Psi_B^0|Q_R(1-\chi_{\Omega_d})\left(\mathbf{v}_A^\beta\otimes\Psi_B^0\right)\right\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\left\langle\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha|Q_R(1-\chi_{\Omega_d})\left(\Psi_A^0\otimes\mathbf{v}_B^\beta\right)\right\rangle_{L^2(\mathbb{R}^{3N})}\Big],
\end{aligned}
$$

$$
\begin{aligned}
&f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\\
&:=\Big\langle (H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\otimes\Psi_B^0|Q_R(1-\chi_{\Omega_d})\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\Big\langle \Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha|Q_R(1-\chi_{\Omega_d})\left(\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\right)\Big\rangle_{L^2(\mathbb{R}^{3N})},\\
&f_6^{\alpha\beta}(\mathbf{R},d)\\
&:=\Big\langle (H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big[(\sum_{j_A}\mathbf{p}_{j_A}^\alpha)\mathbf{v}_A^\beta\Big]\otimes\Psi_B^0\Big|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\Big\langle \Psi_A^0\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big[(\sum_{j_B}\mathbf{p}_{j_B}^\alpha)\mathbf{v}_B^\beta\Big]\Big|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})},\\
&f_7^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\\
&:=\Big\langle (H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big[(\sum_{j_A}\mathbf{p}_{j_A})^\alpha(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\Big]\otimes\Psi_B^0\Big|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\Big\langle \Psi_A^0\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big[(\sum_{j_B}\mathbf{p}_{j_B}^\alpha)(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\Big]\Big|Q_R(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}
\end{aligned}
$$

and note that

$$
\begin{aligned}
&M_{OUT}(\mathbf{R},\sigma,d)=A_{OUT}(\mathbf{R},\sigma,d)+B_{OUT}(\mathbf{R},\sigma,d)\\
&=\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2(\delta_{\alpha,\beta}-\hat{\mathbf{k}}_\alpha\hat{\mathbf{k}}_\beta)\\
&\quad\times\Bigg[\frac{1}{\hbar^2}\Big[f_3^{\alpha\beta}(\mathbf{R},d)-\hbar\omega(\mathbf{k})f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})+(\hbar\omega(\mathbf{k}))^2f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\Big]\\
&\qquad+2\mathrm{Re}\Bigg[\left(\frac{-\mathrm{i}}{m_e\hbar}\right)\Big[f_6^{\alpha\beta}(\mathbf{R},d)-\hbar\omega(\mathbf{k})f_7^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\Big]\Bigg]\\
&\qquad+\frac{1}{\hbar^2}\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\Big[-2\hbar\omega(\mathbf{k})f_1^{\alpha\beta}(\mathbf{R},d)+(\hbar\omega(\mathbf{k}))^2f_2^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\Big]\Bigg]\Bigg].
\end{aligned}
$$

Using $|(\delta_{\alpha,\beta}-\hat{\mathbf{k}}_\alpha\hat{\mathbf{k}}_\beta)|\leq 2$, recalling $|C(\mathbf{k})|^2=(\hbar|\rho(\mathbf{k})|^2)/(2\omega(\mathbf{k}))$ and noting that the integrands are regular at $\mathbf{k}=0$, we immediately conclude

$$
\begin{aligned}
&|M_{OUT}(\mathbf{R},\sigma,d)|\\
&\leq\sum_{\alpha,\beta=1}^{3}\int_{\Omega_\sigma}d\mathbf{k}|\rho(\mathbf{k})|^2\\
&\quad\times\Bigg[4|f_1^{\alpha\beta}(\mathbf{R},d)|+2\hbar\omega(\mathbf{k})|f_2^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|+\frac{1}{\hbar\omega(\mathbf{k})}|f_3^{\alpha\beta}(\mathbf{R},d)|\\
&\qquad+|f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|+\hbar\omega(\mathbf{k})|f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|\\
&\qquad+\frac{2}{m_e\omega(\mathbf{k})}|f_6^{\alpha\beta}(\mathbf{R},d)|+\frac{2\hbar}{m_e}|f_7^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|\Bigg],
\end{aligned}
\tag{5.5.4}
$$

and positivity of the integrands yields

$$
\begin{aligned}
&|M_{OUT}(\mathbf{R},\sigma,d)| \\
\leq& 4\,\|\rho\|^2_{L^2(\mathbb{R}^3)}(\Lambda)\left(\sum_{\alpha,\beta=1}^{3}|f_1^{\alpha\beta}(\mathbf{R},d)|\right)+\frac{1}{\hbar}\|\rho/\sqrt{\omega}\|^2_{L^2(\mathbb{R}^3)}(\Lambda)\left(\sum_{\alpha,\beta=1}^{3}|f_3^{\alpha\beta}(\mathbf{R},d)|\right) \\
&+\frac{2}{m_e}\|\rho/\sqrt{\omega}\|^2_{L^2(\mathbb{R}^3)}(\Lambda)\left(\sum_{\alpha,\beta=1}^{3}|f_6^{\alpha\beta}(\mathbf{R},d)|\right) \\
&+\int_{\mathbb{R}^3}d\mathbf{k}\,\hbar\omega(\mathbf{k})|\rho_0(\mathbf{k}/\Lambda)|^2\left(\sum_{\alpha,\beta=1}^{3}2|f_2^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|+|f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|\right) \\
&+\int_{\mathbb{R}^3}d\mathbf{k}\,|\rho_0(\mathbf{k}/\Lambda)|^2\left(\sum_{\alpha,\beta=1}^{3}|f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|+\frac{2\hbar}{m_e}|f_7^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|\right),
\end{aligned}
$$

where we have highlighted the dependence of ρ on the ultraviolet-cutoff Λ. By the fact that $\rho_0 \in \mathcal{S}(\mathbb{R}^3)$ by assumption and the bounds to be proven below, all $\mathbf{k}$-integrals are finite.

In the remainder of the proof we will establish ($\mathbf{k}$-independent) exponential bounds on the $f_i^{\alpha\beta}$. As we will show below, for $i = 1, 2, 3, 6, 7$, this can be accomplished by a method analogous to the one used in the proof of Lemma 5.3.1.

In the case of $f_4^{\alpha\beta}$ and $f_5^{\alpha\beta}$, however, this method will have to be modified slightly, using an argument based on the maximum principle for elliptic PDEs to extract information about the decay of the one-particle density of the functions $(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha$ and $(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha$.

Let us first turn to the cases $i = 1, 2, 3, 6, 7$. The arguments given in the proof of Lemma 5.3.1, together with the resolvent and relative boundedness estimates already used in the proof of Lemma 5.5.3, yield

$$
\begin{aligned}
&\sum_{\alpha,\beta=1}^{3}|f_1^{\alpha\beta}(\mathbf{R},d)| \\
\leq& \|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \\
&\times\frac{1}{\Delta_A+\Delta_B}\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right), \\
&\sum_{\alpha,\beta=1}^{3}|f_2^{\alpha\beta}(\mathbf{R},d,\mathbf{k})| \\
\leq& \|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \\
&\times\left(\frac{1}{\Delta_A\,\Delta_B}+\frac{1}{\Delta_A+\Delta_B}\left(\frac{1}{\Delta_A}+\frac{1}{\Delta_B}\right)\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^\alpha\|_{L^2(\mathbb{R}^{3Z_B})}\right),
\end{aligned}
$$

$$
\begin{aligned}
&\sum_{\alpha,\beta=1}^{3} |f_3^{\alpha\beta}(\mathbf{R},d)| \\
&\leq \|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})} \|\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \\
&\quad\times\left[\sum_{\alpha,\beta=1}^{3} \|\sum_{i_A,j_A} \mathbf{x}_{i_A}^{\alpha}\mathbf{x}_{j_A}^{\beta}\Psi_A^0\|_{L^2(\mathbb{R}^{3Z_A})} + \|\sum_{i_B,j_B} \mathbf{x}_{i_B}^{\alpha}\mathbf{x}_{j_B}^{\beta}\Psi_B^0\|_{L^2(\mathbb{R}^{3Z_B})}\right], \qquad (5.5.5)\\
&\sum_{\alpha,\beta=1}^{3} |f_6^{\alpha\beta}(\mathbf{R},d)| \\
&\leq \|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})} \|\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \\
&\quad\times\left[\frac{Z_A\tilde{C}}{\Delta_A}\left(\sum_{\beta=1}^{3}\|\mathbf{v}_A^{\beta}\|_{H^2(\mathbb{R}^{3Z_A})}\right) + \frac{Z_B\tilde{C}}{\Delta_B}\left(\sum_{\beta=1}^{3}\|\mathbf{v}_B^{\beta}\|_{H^2(\mathbb{R}^{3Z_B})}\right)\right], \\
&\sum_{\alpha,\beta=1}^{3} |f_7^{\alpha\beta}(\mathbf{R},d,\mathbf{k})| \\
&\leq \|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})} \|\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \\
&\quad\times\left[\frac{Z_AC}{\Delta_A}\left(\frac{1}{\Delta_A}+1+\frac{\hbar\omega(\mathbf{k})}{\Delta_A}\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^{\alpha}\|_{L^2(\mathbb{R}^{3Z_A})}\right)\right. \\
&\qquad\left.+\frac{Z_BC}{\Delta_B}\left(\frac{1}{\Delta_B}+1+\frac{\hbar\omega(\mathbf{k})}{\Delta_B}\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^{\alpha}\|_{L^2(\mathbb{R}^{3Z_B})}\right)\right].
\end{aligned}
$$

Note that the norms $\|\sum_{i_A,j_A}\mathbf{x}_{i_A}^{\alpha}\mathbf{x}_{j_A}^{\beta}\Psi_A^0\|_{L^2(\mathbb{R}^{3Z_A})}$ and $\|\sum_{i_B,j_B}\mathbf{x}_{i_B}^{\alpha}\mathbf{x}_{j_B}^{\beta}\Psi_B^0\|_{L^2(\mathbb{R}^{3Z_B})}$ occurring in (5.5.5) are finite due to the exponential decay of Ψ_A^0 and Ψ_B^0. These estimates show that we can find a positive constant C', independent of $\mathbf{R}$, σ and d (but depending on Λ via properties of $H_{A,B}$ and via $\rho_0(\mathbf{k}/\Lambda)$), such that

$$
\begin{aligned}
&|M_{OUT}(\mathbf{R},\sigma,d)| \\
&\leq C'\|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\|\|(1-\chi_{\Omega_d})(\Psi_A^0\otimes\Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \\
&\quad+\int_{\mathbb{R}^3} d\mathbf{k}|\rho_0(\mathbf{k}/\Lambda)|^2\Big[\sum_{\alpha,\beta=1}^{3}|f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})| + \hbar\omega(\mathbf{k})|f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|\Big]. \qquad (5.5.6)
\end{aligned}
$$

By Lemma A.1.1,

$$\|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})} \leq C_Q(1+1/R),$$

with a positive constant C_Q that is independent of R and Λ. The exponential decay of Ψ_A^0 and Ψ_B^0 (see Proposition 2.5.1) implies the existence of positive constants C_A, C_B, γ_A and γ_B, such that

$$
\begin{aligned}
\big|\Psi_A^0(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})\big| &\leq C_A\,\mathrm{e}^{-\gamma_A(|\mathbf{x}_1|+\cdots+|\mathbf{x}_{Z_A}|)},\\
\big|\Psi_B^0(\mathbf{x}_{Z_A+1},\ldots,\mathbf{x}_N)\big| &\leq C_B\,\mathrm{e}^{-\gamma_B(|\mathbf{x}_{Z_A+1}|+\cdots+|\mathbf{x}_N|)}.
\end{aligned}
$$

From this we conclude that $\mathbf{v}_A^{\alpha}$ and $\mathbf{v}_B^{\beta}$ are also exponentially decaying, and we can find

positive constants C_A^α, C_B^β, γ_A^α and γ_B^β, such that

$$|\mathbf{v}_A^\alpha(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})| = \Big|\sum_{i_A} \mathbf{x}_{i_A}^\alpha \Psi_A^0(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\Big| \le C_A^\alpha \,\mathrm{e}^{-\gamma_A^\alpha(|\mathbf{x}_1|+\cdots+|\mathbf{x}_{Z_A}|)},$$
$$\Big|\mathbf{v}_B^\beta(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Big| = \Big|\sum_{j_B} \mathbf{x}_{i_B}^\beta \Psi_B^0(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Big| \le C_B^\beta \,\mathrm{e}^{-\gamma_B^\beta(|\mathbf{x}_{Z_A+1}|+\cdots+|\mathbf{x}_N|)},$$

and by taking maxima and minima, respectively, we can find positive constants C_A', C_B', γ_A' and γ_B' such that

$$|\mathbf{v}_A^\alpha(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})| \le C_A' \,\mathrm{e}^{-\gamma_A'(|\mathbf{x}_1|+\cdots+|\mathbf{x}_{Z_A}|)},$$
$$\Big|\mathbf{v}_B^\beta(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Big| \le C_B' \,\mathrm{e}^{-\gamma_B'(|\mathbf{x}_{Z_A+1}|+\cdots+|\mathbf{x}_N|)},$$

the right-hand sides now being independent of α and β. By Lemma A.3.1 ii) (recall that $\Psi_A^0 \otimes \Psi_B^0$ is an eigenfunction of $H_A + H_B$ corresponding to the eigenvalue 0, and the smeared Coulomb potentials occurring in H_A and H_B satisfy the assumptions of Lemma A.3.1), there exist positive constants C_1 and γ_1, independent of d, such that

$$\|(1-\chi_{\Omega_d})(\Psi_A^0 \otimes \Psi_B^0)\|_{H^1(\mathbb{R}^{3N})} \le C_1 \mathrm{e}^{-\gamma_1 d}. \tag{5.5.7}$$

It remains to estimate $|f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|$ and $|f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|$. By Lemma A.1.1,

$$\begin{aligned}
&\Big|f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})\Big| \\
\le& 2\|Q_R\|_{H^1(\mathbb{R}^{3N}),L^2(\mathbb{R}^{3N})}\Big[\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha \otimes \Psi_B^0\|_{H^1(\mathbb{R}^{3N})}\|(1-\chi_{\Omega_d})(\mathbf{v}_A^\beta \otimes \Psi_B^0)\|_{L^2(\mathbb{R}^{3N})} \\
&+ \|\Psi_A^0 \otimes (H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha\|_{H^1(\mathbb{R}^{3N})}\|(1-\chi_{\Omega_d})(\Psi_A^0 \otimes \mathbf{v}_B^\beta)\|_{L^2(\mathbb{R}^{3N})}\Big] \\
\le& 2C_Q(1+1/R)\Big[\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha \otimes \Psi_B^0\|_{H^1(\mathbb{R}^{3N})}\|\mathbf{v}_A^\beta \otimes \Psi_B^0\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)} \\
&+ \|\Psi_A^0 \otimes (H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha\|_{H^1(\mathbb{R}^{3N})}\|\Psi_A^0 \otimes \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\Big],
\end{aligned}$$

where for the second inequality we have used that $1-\chi_{\Omega_d} \le 1$ and $\mathrm{supp}\,(1-\chi_{\Omega_d}) \subset L^2(\mathbb{R}^{3N}\setminus\Omega_d)$. Now Lemma A.3.1 i) yields the existence of positive constants C_2, C_3, γ_2 and γ_3 such that

$$\|\mathbf{v}_A^\beta \otimes \Psi_B^0\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)} \le C_2 \,\mathrm{e}^{-\gamma_2 d},$$
$$\|\Psi_A^0 \otimes \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)} \le C_3 \,\mathrm{e}^{-\gamma_3 d}.$$

Noting that $(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha \otimes \Psi_B^0$ and $\Psi_A^0 \otimes (H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha$ are independent of σ, $\mathbf{R}$ and d, we can find positive constants C_4 and γ_4 such that

$$|f_4^{\alpha\beta}(\mathbf{R},d,\mathbf{k})| \le C_4(1+1/R)\mathrm{e}^{-\gamma_4 d}. \tag{5.5.8}$$

In the last part of the proof we investigate $f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})$. Applying the Cauchy-Schwarz inequality, Lemma A.1.1 and using $1-\chi_{\Omega_d} \le 1$, $\mathrm{supp}\,(1-\chi_{\Omega_d}) \subset L^2(\mathbb{R}^{3N}\setminus\Omega_d)$, yields

$$
\begin{aligned}
&|f_5^{\alpha\beta}(\mathbf{R},d,\mathbf{k})|\\
=&\Big|\Big\langle (H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\otimes\Psi_B^0|Q_R(1-\chi_{\Omega_d})\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&+\Big\langle \Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha|Q_R(1-\chi_{\Omega_d})\left(\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Big|\\
\leq& C_Q(1+1/R)\Big[\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha\otimes\Psi_B^0\|_{H^1(\mathbb{R}^{3N})}\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\\
&+\|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\alpha\|_{H^1(\mathbb{R}^{3N})}\|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\Big].
\end{aligned}
$$

The reason we have to modify our previous method at this point is the appearance of the terms $\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}$ and $\|\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}$. Since they involve the reduced resolvents $(H_{A,B}+\hbar\omega(\mathbf{k}))^{-1}$, we cannot establish their exponential decay (in d) directly as above.
The first step is to estimate these terms by expressions only involving one-particle densities. Put $\varphi_{\beta,\mathbf{k}}:=(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta$ and consider the decomposition

$$
\mathbb{R}^{3N}\setminus\Omega_d=\cup_J\Omega_J
$$

into 2^N-1 disjoint subsets, where every Ω_J is of the form $(\cdots\times\{|\mathbf{x}_{i_J}|>d\}\times\dots)$ for some $i_J\in\{1,\dots,N\}$ (see the proof of Lemma A.3.1 i)). Then

$$
\begin{aligned}
&\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\|^2_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\\
=&\sum_J\int_{\Omega_J}|\varphi_{\beta,\mathbf{k}}|^2(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\,|\Psi_B^0|^2(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)d\mathbf{x}_1\dots d\mathbf{x}_N.
\end{aligned}
$$

Since the integrand is positive, we can replace each Ω_J by the larger set

$$
\mathbb{R}^3\times\cdots\times\{|\mathbf{x}_{i_J}|>d\}\times\cdots\times\mathbb{R}^3,
$$

obtaining

$$
\begin{aligned}
&\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\|^2_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\\
\leq&\sum_J\int_{\mathbb{R}^3\times\cdots\times\{|\mathbf{x}_{i_J}|>d\}\times\cdots\times\mathbb{R}^3}|\varphi_{\beta,\mathbf{k}}|^2(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\,|\Psi_B^0|^2(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)d\mathbf{x}_1\dots d\mathbf{x}_N.
\end{aligned}
$$

Collecting the cases in which $i_J\in\{1,\dots,Z_A\}$ and $i_J\in\{Z_A+1,\dots,N\}$, respectively, using the partial antisymmetry of $\varphi_{\beta,\mathbf{k}}$ and Ψ_B^0 and employing the definition of the one-particle densities $\rho_{\varphi_{\beta,\mathbf{k}}}$ and $\rho_{\Psi_B^0}$, we find

$$
\begin{aligned}
&\|(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\beta\otimes\Psi_B^0\|^2_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\\
\leq&C_{Z_A,Z_B}\left(\int_{\{|\mathbf{x}_1|>d\}\times\mathbb{R}^3\times\cdots\times\mathbb{R}^3}|\varphi_{\beta,\mathbf{k}}|^2(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})d\mathbf{x}_1\dots d\mathbf{x}_{Z_A}\right)\underbrace{\|\Psi_B^0\|^2_{L^2(\mathbb{R}^{3Z_B})}}_{=1}\\
&+C'_{Z_A,Z_B}\|\varphi_{\beta,\mathbf{k}}\|^2_{L^2(\mathbb{R}^{3Z_A})}\int_{\{|\mathbf{x}_{Z_A+1}|>d\}\times\mathbb{R}^3\times\cdots\times\mathbb{R}^3}|\Psi_B^0|^2(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)d\mathbf{x}_{Z_A+1}\dots d\mathbf{x}_N
\end{aligned}
$$

$$=C_{Z_A,Z_B}\frac{1}{Z_B}\int_{\{|\mathbf{x}|>d\}}\rho_{\varphi_{\beta,\mathbf{k}}}(\mathbf{x})d\mathbf{x}+C'_{Z_A,Z_B}\|\varphi_{\beta,\mathbf{k}}\|^2_{L^2(\mathbb{R}^{3Z_A})}\frac{1}{Z_A}\int_{\{|\mathbf{x}|>d\}}\rho_{\Psi^0_B}(\mathbf{x})d\mathbf{x},$$

where C_{Z_A,Z_B} and C'_{Z_A,Z_B} are combinatorial constants. Analogously,

$$\begin{aligned}&\|\Psi^0_A\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}^\beta_B\|^2_{L^2(\mathbb{R}^{3N}\setminus\Omega_d)}\\ \leq& C_{Z_A,Z_B}\frac{1}{Z_A}\left(\int_{\{|\mathbf{x}|>d\}}\rho_{\Psi^0_A}(\mathbf{x})d\mathbf{x}\right)\|\tilde{\varphi}_{\beta,\mathbf{k}}\|^2_{L^2(\mathbb{R}^{3Z_A})}+C'_{Z_A,Z_B}\frac{1}{Z_B}\int_{\{|\mathbf{x}|>d\}}\rho_{\tilde{\varphi}_{\beta,\mathbf{k}}}(\mathbf{x})d\mathbf{x},\end{aligned}$$

where we have set $\tilde{\varphi}_{\beta,\mathbf{k}}:=(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}^\beta_B$. Now the exponential decay of Ψ^0_A and Ψ^0_B implies the existence of positive constants C_5, C_6, γ_5, γ_6 such that

$$\begin{aligned}\int_{\{|\mathbf{x}|>d\}}\rho_{\Psi^0_A}(\mathbf{x})d\mathbf{x}&\leq C_5\mathrm{e}^{-\gamma_5 d},\\ \int_{\{|\mathbf{x}|>d\}}\rho_{\Psi^0_B}(\mathbf{x})d\mathbf{x}&\leq C_6\mathrm{e}^{-\gamma_6 d},\end{aligned}$$

and by Lemma A.2.2,

$$\begin{aligned}\rho_{\varphi_{\beta,\mathbf{k}}}(\mathbf{x})&\leq C_7\mathrm{e}^{-\gamma_7|\mathbf{x}|},\\ \rho_{\tilde{\varphi}_{\beta,\mathbf{k}}}(\mathbf{x})&\leq C_8\mathrm{e}^{-\gamma_8|\mathbf{x}|}\end{aligned}$$

for almost all $\mathbf{x}\in\mathbb{R}^3$ and suitable positive constants C_7, C_8, γ_7 and γ_8 which are independent of $\mathbf{k}$. Thus we can find postive constants $\tilde{C}_7$, $\tilde{C}_8$, $\tilde{\gamma}_7$ and $\tilde{\gamma}_8$, independent of $\mathbf{k}$ and d, such that

$$\begin{aligned}\int_{\{|\mathbf{x}|>d\}}\rho_{\varphi_{\beta,\mathbf{k}}}(\mathbf{x})d\mathbf{x}&\leq\tilde{C}_7\mathrm{e}^{-\tilde{\gamma}_7 d},\\ \int_{\{|\mathbf{x}|>d\}}\rho_{\tilde{\varphi}_{\beta,\mathbf{k}}}(\mathbf{x})d\mathbf{x}&\leq\tilde{C}_8\mathrm{e}^{-\tilde{\gamma}_8 d}.\end{aligned}$$

Collecting all estimates yields

$$|f^{\alpha\beta}_5(\mathbf{R},d,\mathbf{k})|\leq C_9(1+1/R)\mathrm{e}^{-\gamma_9 d}\tag{5.5.9}$$

for positive constants C_9 and γ_9 which are independent of σ, $\mathbf{R}$, $\mathbf{k}$ and d. Finally, plugging (5.5.7), (5.5.8) and (5.5.9) into (5.5.6) finishes the proof of the first assertion of the lemma. The second assertion follows by a dominated convergence argument applied to (5.5.4). □

5.5.3 Simplification of lower-order terms

Lemma 5.5.5. *Assume the hypotheses of Theorem 5.4.1 and recall the terms $\tilde{A}_l(\mathbf{R},\sigma,d)$ and $\tilde{B}_l(\mathbf{R},\sigma,d)$ from Definition 5.5.2. Then for all $l\in\mathbb{N}$, $l\geq 2$,*

$$\tilde{A}_l(\mathbf{R},\sigma,d)=0.$$

Furthermore,

$$\tilde{B}_l(\mathbf{R},\sigma,d)=0$$

for odd l, and for even $l=2s$ we have

$$\begin{aligned}
&\tilde{B}_{2s}(\mathbf{R},\sigma,d)\\
=&\frac{2}{\hbar^2}\mathrm{Re}\Bigg[\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\
&\times\Bigg[-2\hbar\omega(\mathbf{k})\Big\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]|\tilde{Q}_{2s}\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi_B^0\Big|\\
&\qquad\qquad\Big|\tilde{Q}_{2s}\chi_{\Omega_d}\left(\Psi_A^0\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle(\overline{H_A+H_B})^{-1}\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\right)[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\Big|\\
&\qquad\qquad\Big|\tilde{Q}_{2s}\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+(\hbar\omega(\mathbf{k}))^2\Big\langle(\overline{H_A+H_B})^{-1}\left(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\Big|\\
&\qquad\qquad\Big|\tilde{Q}_{2s}\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Bigg]\Bigg],
\end{aligned}$$

where

$$\begin{aligned}
\tilde{Q}_{2s}=\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}&|\mathbf{x}_{i_A}|^{l_1}\,|\mathbf{x}_{j_B}|^{2s-l_1}\,Y^*_{l_1m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\\
&\times Y^*_{(2s-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\,u_{2s,m,l_1,m_1}(\mathbf{R}).
\end{aligned}$$

Proof. We begin by proving the assertion about $\tilde{A}_l(\mathbf{R},\sigma,d)$. The first step is to perform the $\mathbf{k}$-integration first to generate one $\mathbf{k}$-independent 3×3-Matrix in each contribution. This is done by defining the following functions:

$$\begin{aligned}
&\tilde{g}_1(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})\\
:=&\frac{1}{\hbar^2}\int_{\Omega_\sigma}d\mathbf{k}|C(\mathbf{k})|^2\overline{(\sum_{i_A}\mathbf{x}_{i_A}\Psi_A^0)}(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(\sum_{j_A}\mathbf{x}_{j_A}\Psi_A^0),
\end{aligned}$$

$$\tilde{g}_2(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})$$
$$:= -\frac{1}{\hbar}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})\overline{(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big[\sum_{i_A}\mathbf{x}_{i_A}\Psi_A^0\Big]}(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(\sum_{j_A}\mathbf{x}_{j_A}\Psi_A^0),$$
$$\tilde{g}_3(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})$$
$$:= \int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2\overline{(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big[\sum_{i_A}\mathbf{x}_{i_A}\Psi_A^0\Big]}(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\Big[\sum_{j_A}\mathbf{x}_{j_A}\Psi_A^0\Big],$$
$$\tilde{g}_4(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})$$
$$:=\frac{-\mathrm{i}}{m_e\hbar}\int_{\Omega_\sigma} d\mathbf{k}\,|C(\mathbf{k})|^2\,\Psi_A^0\,\overline{(H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big[(\sum_{j_A}\mathbf{p}_{j_A})(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(\sum_{i_A}\mathbf{x}_{i_A}\Psi_A^0)\Big]},$$
$$\tilde{g}_5(\mathbf{x}_1,\ldots,\mathbf{x}_{Z_A})$$
$$:=\frac{\mathrm{i}}{m_e}\int_{\Omega_\sigma} d\mathbf{k}\,|C(\mathbf{k})|^2\omega(\mathbf{k})\,\Psi_A^0$$
$$\times\overline{(H_{A|\{\Psi_A^0\}^\perp})^{-1}\Big[(\sum_{j_A}\mathbf{p}_{j_A})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}(\sum_{i_A}\mathbf{x}_{i_A}\Psi_A^0)\Big]},$$
$$\tilde{g}_6(\mathbf{x}_{Z_A+1},\ldots,\mathbf{x}_N)$$
$$:= \int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\overline{(\sum_{i_B}\mathbf{x}_{i_B}\Psi_B^0)}(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(\sum_{j_B}\mathbf{x}_{j_B}\Psi_B^0),$$
$$\tilde{g}_7(\mathbf{x}_{Z_A+1},\ldots,\mathbf{x}_N)$$
$$:= -\frac{1}{\hbar}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})\overline{(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big[\sum_{i_B}\mathbf{x}_{i_B}\Psi_B^0\Big]}(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(\sum_{j_B}\mathbf{x}_{j_B}\Psi_B^0),$$
$$\tilde{g}_8(\mathbf{x}_{Z_A+1},\ldots,\mathbf{x}_N)$$
$$:= \int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2\overline{(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big[\sum_{i_B}\mathbf{x}_{i_B}\Psi_B^0\Big]}(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_B+\hbar\omega(\mathbf{k}))^{-1}\Big[\sum_{j_B}\mathbf{x}_{j_B}\Psi_B^0\Big],$$
$$\tilde{g}_9(\mathbf{x}_{Z_A+1},\ldots,\mathbf{x}_N)$$
$$:=\frac{-\mathrm{i}}{m_e\hbar}\int_{\Omega_\sigma} d\mathbf{k}\,|C(\mathbf{k})|^2\,\Psi_B^0\overline{(H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big[(\sum_{j_B}\mathbf{p}_{j_B})(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(\sum_{i_B}\mathbf{x}_{i_B}\Psi_B^0)\Big]},$$
$$\tilde{g}_{10}(\mathbf{x}_{Z_A+1},\ldots,\mathbf{x}_N)$$
$$:=\frac{\mathrm{i}}{m_e}\int_{\Omega_\sigma} d\mathbf{k}\,|C(\mathbf{k})|^2\omega(\mathbf{k})\,\Psi_B^0$$
$$\times\overline{(H_{B|\{\Psi_B^0\}^\perp})^{-1}\Big[(\sum_{j_B}\mathbf{p}_{j_B})\cdot(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_B+\hbar\omega(\mathbf{k}))^{-1}(\sum_{i_B}\mathbf{x}_{i_B}\Psi_B^0)\Big]}.$$

Note that $\tilde{g}_1$ through $\tilde{g}_5$ are in $L^1(\mathbb{R}^{3Z_A})$ and $\tilde{g}_6$ through $\tilde{g}_{10}$ are in $L^1(\mathbb{R}^{3Z_B})$. Furthermore, all the $\tilde{g}_i$ are invariant under permutation of the variables.
The key in the following argument is the insight that all functions $\tilde{g}_i$ are invariant under the families $\{U_R|R\in SO(3)\}$, which act on $L^1(\mathbb{R}^{3Z_A})$ and $L^1(\mathbb{R}^{3Z_B})$, respectively. This can be seen as follows. The dispersion relation $\omega(\mathbf{k})$ and the ultraviolet-cutoff function ρ (occurring in $C(\mathbf{k})$) are invariant under rotations in $\mathbb{R}^3$ by assumption. The ground states Ψ_A^0 and Ψ_A^0 are left invariant by the families $\{U_R\}$ by the conclusions after Propo-

sition 2.5.1. Finally, the operators $(H_{A|\{\Psi_A^0\}^\perp})^{-1}$, $(H_{B|\{\Psi_B^0\}^\perp})^{-1}$, $(H_A+\hbar\omega(\mathbf{k}))^{-1}$ and $(H_B+\hbar\omega(\mathbf{k}))^{-1}$ commute with the families $\{U_R\}$ by Proposition 2.5.1 and the fact that commutativity with these families is inherited by the resolvents of H_A and H_B (see the proof of Lemma 4.2.11). Using these ingredients, we calculate, e.g. for the function $\tilde{g}_8$ which originates from the term (3.0.10):

$$
\begin{aligned}
&U_R[\tilde{g}_8(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)]\\
=&\tilde{g}_8(R^{-1}\mathbf{x}_{Z_A+1},\dots,R^{-1}\mathbf{x}_N)\\
=&\sum_{i_B,j_B}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2 U_R\Big[\overline{(H_B+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{x}_{i_B}\Psi_B^0)}\Big]\\
&\qquad\times(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})U_R\Big[(H_B+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{x}_{j_B}\Psi_B^0)\Big]\\
=&\sum_{i_B,j_B}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2\overline{(H_B+\hbar\omega(\mathbf{k}))^{-1}(U_R[\mathbf{x}_{i_B}\Psi_B^0])}\\
&\qquad\times(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_B+\hbar\omega(\mathbf{k}))^{-1}(U_R[\mathbf{x}_{j_B}\Psi_B^0])\Big]\\
=&\sum_{i_B,j_B}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2\left(\overline{R^{-1}[(H_B+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{x}_{i_B}\Psi_B^0)]}\right)\\
&\qquad\times(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\left(R^{-1}[(H_B+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{x}_{j_B}\Psi_B^0)]\right)\\
=&\sum_{i_B,j_B}\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2\left(\overline{(H_B+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{x}_{i_B}\Psi_B^0)}\right)\\
&\qquad\times(R^{-1}R-(R\hat{\mathbf{k}})\otimes(R\hat{\mathbf{k}}))\left([(H_B+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{x}_{j_B}\Psi_B^0)]\right)\\
=&\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\omega(\mathbf{k})^2\overline{(H_B+\hbar\omega(\mathbf{k}))^{-1}(\sum_{i_B}\mathbf{x}_{i_B}\Psi_B^0)}\\
&\qquad\times(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_B+\hbar\omega(\mathbf{k}))^{-1}(\sum_{j_B}\mathbf{x}_{j_B}\Psi_B^0)\\
=&\tilde{g}_8(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N).
\end{aligned}
$$

In the last step we have used the change of variables $\mathbf{k}':=R\mathbf{k}$ (note that $R\hat{\mathbf{k}}=\widehat{R\mathbf{k}}$, since $R\in SO(3)$, and that the domain Ω_σ is invariant under rotations). The argument for the remaining $\tilde{g}_i$ is completely analogous. Next we define the functions

$$
g_i(\mathbf{x}):=\begin{cases}\chi_d(\mathbf{x})\int\Pi_{j=2}^{Z_A}\chi_d(\mathbf{x}_j)\tilde{g}_i(\mathbf{x},\mathbf{x}_2,\dots,\mathbf{x}_{Z_A})d\mathbf{x}_2\dots d\mathbf{x}_{Z_A}, & i=1,\dots,5,\\ \chi_d(\mathbf{x})\int\Pi_{j=Z_A+1}^{N}\chi_d(\mathbf{x}_j)\tilde{g}_i(\mathbf{x},\mathbf{x}_{Z_A+2},\dots,\mathbf{x}_N)d\mathbf{x}_{Z_A+2}\dots d\mathbf{x}_N), & i=6,\dots,10,\end{cases}
$$

and note that since the $\tilde{g}_i$ are invariant under permutation of the variables, this definition does not depend on which $3Z_A-3$ (resp. $3Z_B-3$) variables we trace out. Now a simple change of variables shows that the g_i inherit the rotation invariance, i.e. $g_i(R^{-1}\mathbf{x})=g_i(\mathbf{x})$ for all $\mathbf{x}\in\mathbb{R}^3$, $R\in SO(3)$ and $i=1,\dots,10$ (recall that the functions χ_d where chosen to be invariant under $SO(3)$). Using the definitions of the functions $\tilde{g}_i$ and Fubini's theorem, we conclude

$$
\begin{aligned}
&\tilde{A}_l(\mathbf{R},\sigma,d)\\
&=\int_{\mathbb{R}^{3N}}\Bigg[|\Psi_B^0|^2(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Big(\tilde{g}_1+2\mathrm{Re}[\tilde{g}_2]+\tilde{g}_3+2\mathrm{Re}[\tilde{g}_4+\tilde{g}_5]\Big)(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\\
&\qquad\qquad+|\Psi_A^0|^2(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\Big(\tilde{g}_6+2\mathrm{Re}[\tilde{g}_7]+\tilde{g}_8+2\mathrm{Re}[\tilde{g}_9+\tilde{g}_{10}]\Big)(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Bigg]\\
&\qquad\times Q_l(\mathbf{x}_1,\dots,\mathbf{x}_N)\chi_{\Omega_d}(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_1\dots d\mathbf{x}_N\\
&=\sum_{i_A,j_B}\int_{\mathbb{R}^{3N}}d\mathbf{x}_1\dots d\mathbf{x}_N\,\chi_{\Omega_d}(\mathbf{x}_1,\dots,\mathbf{x}_N)\\
&\quad\times\Bigg[|\Psi_B^0|^2(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Big(\tilde{g}_1+2\mathrm{Re}[\tilde{g}_2]+\tilde{g}_3+2\mathrm{Re}[\tilde{g}_4+\tilde{g}_5]\Big)(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\\
&\qquad+|\Psi_A^0|^2(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})\Big(\tilde{g}_6+2\mathrm{Re}[\tilde{g}_7]+\tilde{g}_8+2\mathrm{Re}[\tilde{g}_9+\tilde{g}_{10}]\Big)(\mathbf{x}_{Z_A+1},\dots,\mathbf{x}_N)\Bigg]\\
&\quad\times\Bigg[\sum_{m=-l}^{l}\sum_{l_1=0}^{l}\sum_{m_1=-l_1}^{l_1}|\mathbf{x}_{i_A}|^{l_1}\,|\mathbf{x}_{j_B}|^{l-l_1}\,Y^*_{l_1m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\,Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\,u_{l,m,l_1,m_1}(\mathbf{R})\\
&\qquad-\sum_{m=-l}^{l}|\mathbf{x}_{i_A}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\,v_{l,m}(\mathbf{R})-\sum_{m=-l}^{l}|\mathbf{x}_{j_B}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\,w_{l,m}(\mathbf{R})\Bigg].
\end{aligned}
$$

Using the invariance of $|\Psi_A^0|^2$, $|\Psi_B^0|^2$, the $\tilde{g}_i$ and χ_{Ω_d} under permutation of the electron coordinates, we can rename variables and obtain

$$
\begin{aligned}
&\tilde{A}_l(\mathbf{R},\sigma,d)\\
&=Z_A\,Z_B\\
&\quad\times\Bigg[-\sum_{m=-l}^{l}v_{l,m}(\mathbf{R})\,\|\Pi_{i=Z_A+1}^{N}\chi_d(\cdot_i)\Psi_B^0\|^2_{L^2(\mathbb{R}^{3Z_B})}\\
&\qquad\qquad\times\left(\int_{\mathbb{R}^3}|\mathbf{x}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}}]\,\Big(g_1+2\mathrm{Re}[g_2]+g_3+2\mathrm{Re}[g_4+g_5]\Big)(\mathbf{x})\,d\mathbf{x}\right) \qquad (5.5.10)\\
&\qquad-\sum_{m=-l}^{l}v_{l,m}(\mathbf{R})\,\|\Pi_{i=1}^{Z_A}\chi_d(\cdot_i)(\tilde{g}_6+2\mathrm{Re}[\tilde{g}_7]+\tilde{g}_8+2\mathrm{Re}[\tilde{g}_9+\tilde{g}_{10}])\|_{L^1(\mathbb{R}^{3Z_A})}\\
&\qquad\qquad\times\left(\int_{\mathbb{R}^3}|\mathbf{x}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}}]\,(1/Z_A)\rho_{A,d}(\mathbf{x})\,d\mathbf{x}\right) \qquad (5.5.11)\\
&\qquad-\sum_{m=-l}^{l}w_{l,m}(\mathbf{R})\,\|\Pi_{i=1}^{Z_A}\chi_d(\cdot_i)\Psi_A^0\|^2_{L^2(\mathbb{R}^{3Z_A})}\\
&\qquad\qquad\times\left(\int_{\mathbb{R}^3}|\mathbf{x}|^{l}\,Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}}]\,\Big(g_6+2\mathrm{Re}[g_7]+g_8+2\mathrm{Re}[g_9+g_{10}]\Big)(\mathbf{x})\,d\mathbf{x}\right) \qquad (5.5.12)
\end{aligned}
$$

$$
\begin{aligned}
&- \sum_{m=-l}^{l} w_{l,m}(\mathbf{R}) \, \|\Pi_{i=Z_A+1}^{N} \chi_d(\cdot_i)(\tilde{g}_1 + 2\mathrm{Re}[\tilde{g}_2] + \tilde{g}_3 + 2\mathrm{Re}[\tilde{g}_4 + \tilde{g}_5])\|_{L^1(\mathbb{R}^{3Z_B})} \\
&\qquad \times \left(\int_{\mathbb{R}^3} |\mathbf{x}|^l \, Y_{lm}^*[(\theta,\varphi)_{\mathbf{x}}] \, (1/Z_B) \rho_{B,d}(\mathbf{x}) \, d\mathbf{x} \right) \qquad (5.5.13) \\
&+ \sum_{m=-l}^{l} \sum_{l_1=0}^{l} \sum_{m_1=-l_1}^{l_1} u_{l,m,l_1,m_1}(\mathbf{R}) \\
&\qquad \times \Bigg[\left(\int_{\mathbb{R}^3} |\mathbf{x}|^{l_1} Y_{l_1 m_1}^*[(\theta,\varphi)_{\mathbf{x}}] \Big(g_1 + 2\mathrm{Re}[g_2] + g_3 + 2\mathrm{Re}[g_4 + g_5] \Big)(\mathbf{x}) \, d\mathbf{x} \right) \\
&\qquad\qquad \times \left(\int_{\mathbb{R}^3} |\mathbf{x}|^{l-l_1} \, Y_{(l-l_1)(m-m_1)}^*[(\theta,\varphi)_{\mathbf{x}}] \, (1/Z_B) \rho_{B,d}(\mathbf{x}) \, d\mathbf{x} \right) \qquad (5.5.14) \\
&\qquad\quad + \left(\int_{\mathbb{R}^3} |\mathbf{x}|^{l_1} Y_{l_1 m_1}^*[(\theta,\varphi)_{\mathbf{x}}] \, (1/Z_A) \rho_{A,d}(\mathbf{x}) \, d\mathbf{x} \right) \\
&\qquad\qquad \times \Bigg(\int_{\mathbb{R}^3} |\mathbf{x}|^{l-l_1} \, Y_{(l-l_1)(m-m_1)}^*[(\theta,\varphi)_{\mathbf{x}}] \\
&\qquad\qquad\qquad \times \Big(g_6 + 2\mathrm{Re}[g_7] + g_8 + 2\mathrm{Re}[g_9 + g_{10}] \Big)(\mathbf{x}) \, d\mathbf{x} \Bigg) \Bigg] \Bigg], \qquad (5.5.15)
\end{aligned}
$$

where $\rho_{A,d}$, $\rho_{B,d}$ are the one-particle densities of $\Pi_{i=1}^{Z_A} \chi_d(\cdot_i)\Psi_A^0$ and $\Pi_{i=Z_A+1}^{N} \chi_d(\cdot_i)\Psi_B^0$, respectively. But now all the integrands in the terms (5.5.10) through (5.5.13) have the structure of a product of a function on $\mathbb{R}^3$ which is invariant under $SO(3)$, and a spherical harmonic of degree $l \geq 2$. The latter integrates to zero against a spherically symmetric function due to the mutual orthogonality of spherical harmonics of different degrees and the $(l = 0)$- spherical harmonic being a constant function. In the two terms (5.5.14) and (5.5.15) at least one of the two spherical harmonics $Y_{l_1 m_1}^*$ and $Y_{(l-l_1)(m-m_1)}^*$ has degree ≥ 1, so that the corresponding integral against the spherically symmetric functions involved vanishes, which causes the two products (5.5.14) and (5.5.15) to vanish. Summarizing, all the contributions to $\tilde{A}_l(\mathbf{R}, \sigma, d)$ vanish, and the assertion on $\tilde{A}_l(\mathbf{R}, \sigma, d)$ is proved.
Next we turn to the investigation of $\tilde{B}_l(\mathbf{R}, \sigma, d)$. To this end, consider the term (5.5.2). Set $f := (\overline{H_A + H_B})^{-1} \Big(\mathbf{v}_A (1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) \mathbf{v}_B \Big)$. Since Ψ_A^0 and Ψ_B^0 are eigenfunctions of the parity operators P_A and P_B with eigenvalues ε_A and ε_B, respectively (see the remarks after Proposition 2.5.1), $\mathbf{v}_A$ and $\mathbf{v}_A$ have parity $-\varepsilon_A$ and $-\varepsilon_B$. By Lemma 4.2.11, these parities are left invariant by the operator $(\overline{H_A + H_B})^{-1}$, so that the function f has parity $-\varepsilon_A$ with respect to $P_A \otimes I$ and $-\varepsilon_B$ with respect to $I \otimes P_B$. By definition, for $l \geq 3$,

$$Q_l = \sum_{i_A, j_B} \Bigg[\sum_{m=-l}^{l} \sum_{l_1=0}^{l} \sum_{m_1=-l_1}^{l_1} |\mathbf{x}_{i_A}|^{l_1} \, |\mathbf{x}_{j_B}|^{l-l_1} \, Y^*_{l_1 m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}]$$
$$\times \, Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \, u_{l,m,l_1,m_1}(\mathbf{R}) \tag{5.5.16}$$
$$- \sum_{m=-l}^{l} |\mathbf{x}_{i_A}|^{l} \, Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{i_A}}] \, v_{l,m}(\mathbf{R}) \tag{5.5.17}$$
$$- \sum_{m=-l}^{l} |\mathbf{x}_{j_B}|^{l} \, Y^*_{lm}[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \, w_{l,m}(\mathbf{R}) \Bigg]. \tag{5.5.18}$$

The term (5.5.17) in Q_l only depends on the variable $\mathbf{x}_{i_A} \in \{\mathbf{x}_1, \dots \mathbf{x}_{Z_A}\}$, so that the corresponding contribution to the inner product in (5.5.2) vanishes upon integrating over the variables $\mathbf{x}_{Z_A+1}, \dots, \mathbf{x}_N$, since $\Pi_{i=Z_A+1}^{N} \chi_d(\cdot_i) \Psi_B^0$ and f have opposite parity with respect to P_B. Analogously, the contribution from (5.5.18) vanishes.
If l is odd, then at least one of the numbers l_1 and $l - l_1$ has to be even. Assume without loss of generality that l_1 is even. Then the fact that the spherical harmonics Y^*_{lm} have parity $(-1)^l$ and the assumption that the functions χ_d comprising χ_{Ω_d} are invariant under $SO(3)$ imply that the function

$$|\mathbf{x}_{i_A}|^{l_1} \, |\mathbf{x}_{j_B}|^{l-l_1} \, Y^*_{l_1 m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}] \, Y^*_{(l-l_1)(m-m_1)}[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \, u_{l,m,l_1,m_1}(\mathbf{R}) \chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0)$$

occurring in (5.5.16) has parity ε_A with respect to $P_A \otimes I$. Thus its integral against f with respect to the variables $\mathbf{x}_1, \dots, \mathbf{x}_{Z_A}$ vanishes, proving the assertion for the term (5.5.2). The claim for the remaining terms is proven analogously, noting that all resolvents involved conserve parities with respect to P_A and P_B by Lemma 4.2.11. □

5.5.4 Infrared regularization errors for mixed terms

After having established some simplifications in the lowest-order terms of the multipole expansion, we further analyze the remaining contributions $\sum_s \tilde{B}_{2s}(\mathbf{R}, \sigma, d)$ by splitting them into σ-dependent and σ-independent terms.

Definition 5.5.6. Recalling the notation and the definitions of Section 5.1 and letting χ_{Ω_d} be as in the preceding sections, we define

$$T_\Lambda^{l,m}(\mathbf{R})$$
$$:= \int_{\mathbb{R}^6} dy dy' \frac{R^3 \psi_0(Ry) \, R^3 \psi_0(Ry')}{|\Lambda \hat{\mathbf{R}} + y - y'|^{l+1}} Y_{l,m}[(\theta,\varphi)_{\Lambda\hat{\mathbf{R}}+y-y'}],$$
$$S_{\alpha,\beta}^{2s,l_1,m,m_1}(d)$$
$$:= \Big\langle (\overline{H_A + H_B})^{-1} [\mathbf{v}_A^\alpha \mathbf{v}_B^\beta] \Big| (\sum_{i_A} |\mathbf{x}_{i_A}|^{l_1} Y^*_{l_1,m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}])$$
$$\times (\sum_{j_B} |\mathbf{x}_{j_B}|^{l-l_1} Y^*_{l-l_1,m-m_1}[(\theta,\varphi)_{\mathbf{x}_{j_B}}]) \chi_{\Omega_d}(\Psi_A^0 \otimes \Psi_B^0) \Big\rangle_{L^2(\mathbb{R}^{3N})},$$

$$
\begin{aligned}
&T^{2s,l_1,m,m_1}_{\alpha,\beta}(d,|\mathbf{k}|)\\
&:=\Big\langle((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}^\alpha_A)\otimes\Psi^0_B\Big|(\sum_{i_A}|\mathbf{x}_{i_A}|^{l_1}Y^*_{l_1,m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}])\\
&\qquad\times(\sum_{j_B}|\mathbf{x}_{j_B}|^{l-l_1}Y^*_{l-l_1,m-m_1}[(\theta,\varphi)_{\mathbf{x}_{j_B}}])\,\chi_{\Omega_d}\left(\Psi^0_A\otimes((H_B+\hbar\omega\mathbf{k})^{-1}\mathbf{v}^\beta_B)\right)\Big\rangle_{L^2(\mathbb{R}^{3N})} \qquad (5.5.19)\\
&+\Big\langle(\overline{H_A+H_B})^{-1}\Big[((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}^\alpha_A)\otimes\mathbf{v}^\beta_B\Big]\Big|(\sum_{i_A}|\mathbf{x}_{i_A}|^{l_1}Y^*_{l_1,m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}])\\
&\qquad\times(\sum_{j_B}|\mathbf{x}_{j_B}|^{l-l_1}Y^*_{l-l_1,m-m_1}[(\theta,\varphi)_{\mathbf{x}_{j_B}}])\chi_{\Omega_d}(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{L^2(\mathbb{R}^{3N})} \qquad (5.5.20)\\
&+\Big\langle(\overline{H_A+H_B})^{-1}\Big[\mathbf{v}^\alpha_A\otimes((H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}^\beta_B)\Big]\Big|(\sum_{i_A}|\mathbf{x}_{i_A}|^{l_1}Y^*_{l_1,m_1}[(\theta,\varphi)_{\mathbf{x}_{i_A}}])\\
&\qquad\times(\sum_{j_B}|\mathbf{x}_{j_B}|^{l-l_1}Y^*_{l-l_1,m-m_1}[(\theta,\varphi)_{\mathbf{x}_{j_B}}])\chi_{\Omega_d}(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{L^2(\mathbb{R}^{3N})}, \qquad (5.5.21)
\end{aligned}
$$

$$
\begin{aligned}
&B_1(s,d,\mathbf{R})\\
&:=\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde{C}_{2s,m,l_1,m_1}(-2)S^{2s,l_1,m,m_1}_{\alpha,\beta}(d)T^{2s,m}_{\Lambda}(\mathbf{R})\left(\frac{\Lambda}{R}\right)^{2s+1}\\
&\qquad\times\left(\int_{\mathbb{R}^3}d\mathbf{k}|\rho(\mathbf{k})|^2\left(\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\right)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\right)\Bigg], \qquad (5.5.22)
\end{aligned}
$$

$$
\begin{aligned}
&B_2(s,d,\mathbf{R})\\
&:=\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde{C}_{2s,m,l_1,m_1}\hbar\,T^{2s,m}_{\Lambda}(\mathbf{R})\left(\frac{\Lambda}{R}\right)^{2s+1}\\
&\qquad\times\left(\int_{\mathbb{R}^3}d\mathbf{k}\,\omega(\mathbf{k})|\rho(\mathbf{k})|^2\left(\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\right)T^{2s,l_1,m,m_1}_{\alpha,\beta}(d,|\mathbf{k}|)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\right)\Bigg], \qquad (5.5.23)
\end{aligned}
$$

$$
\begin{aligned}
&B_3(\sigma,s,d,\mathbf{R})\\
&:=\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde{C}_{2s,m,l_1,m_1}2S^{2s,l_1,m,m_1}_{\alpha,\beta}(d)T^{2s,m}_{\Lambda}(\mathbf{R})\left(\frac{\Lambda}{R}\right)^{2s+1}\\
&\qquad\times\left(\int_{B_{\sigma/c}(0)}d\mathbf{k}|\rho_0(\mathbf{k}/\Lambda)|^2\left(\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\right)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\right)\Bigg],
\end{aligned}
$$

$$
\begin{aligned}
&B_4(\sigma,s,d,\mathbf{R})\\
&:=\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde{C}_{2s,m,l_1,m_1}(-\hbar)T^{2s,m}_{\Lambda}(\mathbf{R})\left(\frac{\Lambda}{R}\right)^{2s+1}\\
&\qquad\times\left(\int_{B_{\sigma/c}(0)}d\mathbf{k}\,\omega(\mathbf{k})|\rho_0(\mathbf{k}/\Lambda)|^2\left(\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\right)T^{2s,l_1,m,m_1}_{\alpha,\beta}(d,|\mathbf{k}|)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\right)\Bigg].
\end{aligned}
$$

Note that the $\mathbf{k}$-integrals over $\mathbb{R}^3$ exist, since $|\rho|^2 \in \mathcal{S}(\mathbb{R}^3)$ and the remaining integrands are in $L^1_{loc}(\mathbb{R}^3)$. For (5.5.23) the latter fact will become evident from estimates on $T^{2s,l_1,m,m_1}_{\alpha,\beta}(d,|\mathbf{k}|)$ derived below. Recalling Definition 5.5.2, the definition of the Q_l (see section 5.1) and using $\int_{\Omega_\sigma} = \int_{\mathbb{R}^3} - \int_{B_{\sigma/c}(0)}$ immediately leads to

$$\begin{aligned}&\tilde{B}_{2s}(\mathbf{R},\sigma,d)\\ =&B_1(s,d,\mathbf{R}) + B_2(s,d,\mathbf{R}) + B_3(\sigma,s,d,\mathbf{R}) + B_4(\sigma,s,d,\mathbf{R}).\end{aligned} \tag{5.5.24}$$

The analysis of the last two terms is the subject of the present section, and the terms $B_1(s,d,\mathbf{R})$ and $B_2(s,d,\mathbf{R})$ will be investigated in Section 5.5.5.
The following result concerns the σ-dependence of $B_3(\sigma,s,d,\mathbf{R})$ and $B_4(\sigma,s,d,\mathbf{R})$ and will imply the claim (5.4.4) of Theorem 5.4.1 upon defining

$$M^L_{IR}(\mathbf{R},\sigma,d) := \sum_{l\geq 4, l \text{ even}}^{L} B_3(\sigma,l/2,d,\mathbf{R}) + \sum_{l\geq 2, l \text{ even}}^{L} B_4(\sigma,l/2,d,\mathbf{R}), \tag{5.5.25}$$

where $L \in \mathbb{N}$, $L \geq 2$.

Lemma 5.5.7 (Infrared regularization errors for mixed terms)**.** *Assume the hypotheses of Theorem 5.4.1 and let $s = l/2 \geq 1$. Then there exist positive constants $C_1(l), C_2(l)$, independent of σ, d and R (but depending on Λ via properties of $H_{A,B}$), such that*

$$\begin{aligned}&\Big|B_3(\sigma,s,d,R) + B_4(\sigma,s,d,R)\Big|\\ \leq&\left(\frac{3}{2R}\right)^{l+1}\left(\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\right)\left(C_1(l)\left(\frac{\sigma}{c}\right)^3 + C_2(l)\left(\frac{\sigma}{c}\right)^4\right).\end{aligned}$$

In particular,

$$\lim_{\sigma\to 0}\left(B_3(\sigma,s,d,R) + B_4(\sigma,s,d,R)\right) = 0.$$

Proof. Recall from Section 5.1 that

$$\left(\frac{\Lambda}{R}\right)^{l+1} T^{l,m}_{\Lambda}(\mathbf{R}) = \int_{\mathbb{R}^6} dydy' \frac{\psi(y)\,\psi(y')}{|\mathbf{R}+y-y'|^{l+1}} Y_{l,m}[(\theta,\varphi)_{\mathbf{R}+y-y'}].$$

Using Fubini's theorem and reversing the steps taken in Section 5.1 (i.e. addition theorem for Legendre polynomials and results about spherical harmonics of translates), we obtain

$$\begin{aligned}B_4(\sigma,l/2,d,\mathbf{R}) = -\frac{\hbar}{4\pi}\,\mathrm{Re}\Bigg[\sum_{\alpha,\beta=1}^{3}\int_{B_{\sigma/c}(0)} d\mathbf{k}\omega(\mathbf{k})|\rho_0(\mathbf{k}/\Lambda)|^2(\delta_{\alpha,\beta} - \frac{\mathbf{k}_\alpha\mathbf{k}_\beta}{|\mathbf{k}|^2})\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\ \times\left(\int_{\mathbb{R}^6} dydy' \frac{\psi(y)\,\psi(y')}{|\mathbf{R}+y-y'|^{l+1}} W_{\alpha,\beta}(\mathbf{k},d,\mathbf{R},y,y')\right)\Bigg],\end{aligned}$$

where

$$
\begin{aligned}
&W_{\alpha,\beta}(\mathbf{k},d,\mathbf{R},y,y')\\
:=&\Big\langle((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha)\otimes\Psi_B^0\big|\big(\sum_{i_A,j_B}|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|^l P_l(\cos\theta_{\mathbf{x}_{i_A}-\mathbf{x}_{j_B},\mathbf{R}+y-y'})\big)\\
&\qquad\times\chi_{\Omega_d}\big(\Psi_A^0\otimes((H_B+\hbar\omega\mathbf{k})^{-1}\mathbf{v}_B^\beta)\big)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&+\Big\langle(\overline{H_A+H_B})^{-1}[((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha)\otimes\mathbf{v}_B^\beta]\big|\\
&\qquad\big|\big(\sum_{i_A,j_B}|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|^l P_l(\cos\theta_{\mathbf{x}_{i_A}-\mathbf{x}_{j_B},\mathbf{R}+y-y'})\big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&+\Big\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A^\alpha\otimes((H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta)]\big|\\
&\qquad\big|\big(\sum_{i_A,j_B}|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|^l P_l(\cos\theta_{\mathbf{x}_{i_A}-\mathbf{x}_{j_B},\mathbf{R}+y-y'})\big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}.
\end{aligned}
$$

To estimate $W_{\alpha,\beta}(\mathbf{k},d,\mathbf{R},y,y')$, define

$$
u_d(\mathbf{x},\mathbf{R},y,y'):=\sum_{i_A,j_B}|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|^l P_l(\cos\theta_{\mathbf{x}_{i_A}-\mathbf{x}_{j_B},\mathbf{R}+y-y'})\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)
$$

and note that we can rewrite

$$
\begin{aligned}
W_{\alpha,\beta}(\mathbf{k},d,\mathbf{R},y,y')=&\Big\langle((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha)\otimes\overline{((H_B+\hbar\omega\mathbf{k})^{-1}\mathbf{v}_B^\beta)}|\tilde{u}_d\Big\rangle_{L^2(\mathbb{R}^6)}\\
&+\Big\langle(\overline{H_A+H_B})^{-1}[((H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A^\alpha)\otimes\mathbf{v}_B^\beta]|u_d\Big\rangle_{L^2(\mathbb{R}^6)}\\
&+\Big\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A^\alpha\otimes((H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B^\beta)]|u_d\Big\rangle_{L^2(\mathbb{R}^6)},
\end{aligned}
$$

where $\tilde{u}_d$ is defined by replacing Ψ_B^0 with $\overline{\Psi_B^0}$ in u_d. Next we use the resolvent estimates

$$
\begin{aligned}
\|(\overline{H_A+H_{B|W}})^{-1}\|&\le\frac{1}{\Delta_A+\Delta_B},\\
\|((H_A+\hbar\omega(\mathbf{k}))_{|\{\Psi_A^0\}^\perp})^{-1}\|&\le\frac{1}{\Delta_A},\\
\|((H_B+\hbar\omega(\mathbf{k}))_{|\{\Psi_B^0\}^\perp})^{-1}\|&\le\frac{1}{\Delta_B}
\end{aligned}
$$

(recall the definition of the subspace $W=\overline{\{\Psi_A^0\}^\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}$) to obtain

$$
\Big|W_{\alpha,\beta}(\mathbf{k},d,\mathbf{R},y,y')\Big|\le\|u_d\|_{L^2(\mathbb{R}^{3N})}\|\mathbf{v}_A^\alpha\|\|\mathbf{v}_B^\beta\|\left(\frac{1}{\Delta_A\Delta_B}+\frac{1}{\Delta_A+\Delta_B}\left(\frac{1}{\Delta_A}+\frac{1}{\Delta_B}\right)\right).
$$

As far as the norm of u_d is concerned, we use the fact that $|P_l(x)|\le 1$ for $x\in[-1,1]$ (see

[Gar07]) to estimate

$$\begin{aligned}\|u_d\|_{L^2(\mathbb{R}^{3N})} =& \sqrt{\int_{\mathbb{R}^{3N}} \left| \sum_{i_A,j_B} |\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^2 P_l(\cos\theta_{\mathbf{x}_{i_A}-\mathbf{x}_{j_B},\mathbf{R}+y-y'}) \chi_{\Omega_d} \Psi_A^0 \Psi_B^0 \right|^2} \\ \leq& \sqrt{\int_{\mathbb{R}^{3N}} \chi_{\Omega_d}^2 \left(\sum_{i_A,j_B} |\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^2 \right)^2 |\Psi_A^0|^2 \, |\Psi_B^0|^2} \\ \leq& \left\| \sum_{i_A,j_B} |\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^2 (\Psi_A^0 \otimes \Psi_B^0) \right\|_{L^2(\mathbb{R}^{3N})} < \infty,\end{aligned}$$

the last expression being finite due to the exponential decay of the ground states Ψ_A^0 and Ψ_B^0 (see Proposition 2.5.1). As seen in the proof of Lemma 5.1.4, for $R > R_0$ we have

$$\left| \int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\,\psi(y')}{|\mathbf{R}+y-y'|^{l+1}} \right| \leq \left(\frac{3}{2R}\right)^{l+1}.$$

Finally, using

$$\left| \int_{B_{\sigma/c}(0)} d\mathbf{k}\, \omega(\mathbf{k}) |\rho_0(\mathbf{k}/\Lambda)|^2 \Big(\delta_{\alpha,\beta} - \frac{\mathbf{k}_\alpha \mathbf{k}_\beta}{|\mathbf{k}|^2} \Big) \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} \right| \leq 8\pi c \left(\frac{\sigma}{c}\right)^4 \sup_{s\in[0,\sigma/(\Lambda c)]} |\rho_0(s)|^2$$

(see also the proof of Lemma 6.1.1), we arrive at

$$\begin{aligned}&|B_4(\sigma, l/2, d, \mathbf{R})| \\ \leq& \left(\frac{3}{2R}\right)^{l+1} \left(\frac{\sigma}{c}\right)^4 \left(\sup_{s\in[0,\sigma/(\Lambda c)]} |\rho_0(s)|^2 \right) (2\hbar c) \|\mathbf{v}_A^\alpha\| \|\mathbf{v}_B^\beta\| \\ &\times \left(\frac{1}{\Delta_A \Delta_B} + \frac{1}{\Delta_A + \Delta_B} \left(\frac{1}{\Delta_A} + \frac{1}{\Delta_B} \right) \right) \Big\| \sum_{i_A,j_B} |\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^2 (\Psi_A^0 \otimes \Psi_B^0) \Big\|_{L^2(\mathbb{R}^{3N})},\end{aligned}$$

which proves the first part of the claim. Following the same steps as above, we obtain

$$\begin{aligned}&B_3(\sigma, l/2, d, \mathbf{R}) \\ =& \frac{2}{4\pi} \mathrm{Re} \Bigg[\sum_{\alpha,\beta=1}^3 \left(\int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\,\psi(y')}{|\mathbf{R}+y-y'|^{l+1}} \Big\langle (\overline{H_A + H_B})^{-1} [\mathbf{v}_A^\alpha \mathbf{v}_B^\beta] | u_d \Big\rangle_{L^2(\mathbb{R}^{3N})} \right) \\ &\times \left(\int_{B_{\sigma/c}(0)} d\mathbf{k}\, \omega(\mathbf{k}) |\rho_0(\mathbf{k}/\Lambda)|^2 \Big(\delta_{\alpha,\beta} - \frac{\mathbf{k}_\alpha \mathbf{k}_\beta}{|\mathbf{k}|^2} \Big) \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} \right) \Bigg],\end{aligned}$$

with u_d as above. The estimates just established, together with

$$\left| \int_{B_{\sigma/c}(0)} d\mathbf{k}\, \omega(\mathbf{k}) |\rho_0(\mathbf{k}/\Lambda)|^2 \Big(\delta_{\alpha,\beta} - \frac{\mathbf{k}_\alpha \mathbf{k}_\beta}{|\mathbf{k}|^2} \Big) \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} \right| \leq 8\pi (\sigma/c)^3 \sup_{s\in[0,\sigma/(\Lambda c)]} |\rho_0(s)|^2,$$

lead to

$$|B_3(\sigma, l/2, d, \mathbf{R})| \leq 4 \left(\frac{3}{2R}\right)^{l+1} \left(\frac{\sigma}{c}\right)^3 \left(\sup_{s\in[0,\sigma/(\Lambda c)]} |\rho_0(s)|^2\right) \times \frac{1}{\Delta_A + \Delta_B} \|\mathbf{v}_A^\alpha\| \|\mathbf{v}_B^\beta\| \Big\| \sum_{i_A, j_B} |\mathbf{x}_{i_A} - \mathbf{x}_{j_B}|^2 (\Psi_A^0 \otimes \Psi_B^0) \Big\|_{L^2(\mathbb{R}^{3N})},$$

proving the second part of the claim and finishing the proof. □

5.5.5 $1/R^6$- and $1/R^7$- contributions to $M_A(\mathbf{R}, \sigma)$ and $M_B(\mathbf{R}, \sigma)$

The final step in the proof of Theorem 5.4.1 is to identify the contributions at the orders $1/R^6$ and $1/R^7$. To this end, we first establish some results on the asymptotic behaviour of certain Fourier integrals in the next section.

Asymptotics of distributional Fourier transforms

In the following two sections we will have to understand the large $|\mathbf{R}|$-asymptotics of a class of Fourier integrals of the form

$$I(\mathbf{R}) = \int_{\mathbb{R}^n} d\mathbf{k}\, T(\mathbf{k}) g(\mathbf{k}) \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}},$$

where $\mathbf{R} \in \mathbb{R}^n$, $g \in \mathcal{S}(\mathbb{R}^n)$, $g(-\mathbf{k}) = g(\mathbf{k})$ for all $\mathbf{k} \in \mathbb{R}^n$, and T is an element of $L^1_{loc}(\mathbb{R}^n)$ which is homogeneous of degree $\alpha > -n$.
An approach which we have found to be quite useful in this context is to investigate $I(\mathbf{R})$ by methods involving the Fourier transform of distributions. Note that the homogeneity of T ensures that T satisfies an integral growth estimate which implies $T \in \mathcal{S}'(\mathbb{R}^n)$ (see e.g. [Str94]). Thus $I(\mathbf{R})$ can be written in 'dual' notation as follows:

$$I(\mathbf{R}) = \langle T \mathrm{e}^{-\mathrm{i}(\cdot\mathbf{R})}, g\rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)} = \langle T, \mathrm{e}^{-\mathrm{i}(\cdot\mathbf{R})} g\rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)}$$

Since g is even, we have

$$\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}} g(\mathbf{k}) = \left(\widehat{\mathrm{e}^{\mathrm{i}(\cdot\mathbf{R})} \widehat{g(\cdot)}}\right)(\mathbf{k}),$$

so that, using the definition and the properties of the distributional Fourier transform, we find

$$\begin{aligned} I(\mathbf{R}) =& \langle T, \widehat{\mathrm{e}^{\mathrm{i}(\cdot\mathbf{R})} \widehat{g}}\rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)} = \langle \hat{T}, \widehat{\mathrm{e}^{\mathrm{i}(\cdot\mathbf{R})} g}\rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)} \\ =& \langle \hat{T}, \tau_{\mathbf{R}}[\widehat{g}]\rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)}, \end{aligned}$$

with $\tau_{\mathbf{R}}$ denoting the operator of translation by $\mathbf{R} \in \mathbb{R}^n$. Since T is homogeneous of degree α, $\hat{T}$ is homogeneous of degree $-n-\alpha$ (see [Str94]). Recall that a distribution T is called homogeneous of degree α if it satisfies $\langle T, S_\lambda \varphi\rangle = 1/\lambda^\alpha \langle T, \varphi\rangle$ for all $\varphi \in \mathcal{S}(\mathbb{R}^n)$ and all $0 \neq \lambda \in \mathbb{R}$. Here S_λ denotes the scaling operator, i.e. $(S_\lambda \varphi)(\cdot) = \lambda^n \varphi(\lambda \cdot)$. The relation

between the scaling operator and the operator of translation is given by $S_\lambda \tau_a = \tau_{a/\lambda} S_\lambda$, $a \in \mathbb{R}^n$. Using these facts, we obtain that for any $0 \neq \lambda \in \mathbb{R}$,

$$\begin{aligned} I(\mathbf{R}) =& \frac{\lambda^{-n-\alpha}}{\lambda^{-n-\alpha}} \langle \hat{T}, \tau_{\mathbf{R}}[\widehat{g}] \rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)} = \lambda^{-n-\alpha} \langle \hat{T}, S_\lambda \tau_{\mathbf{R}}[\,\widehat{g}\,] \rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)} \\ =& \lambda^{-n-\alpha} \langle \hat{T}, \tau_{(1/\lambda)\mathbf{R}} S_\lambda[\,\widehat{g}\,] \rangle_{\mathcal{S}'(\mathbb{R}^n),\mathcal{S}(\mathbb{R}^n)}. \end{aligned}$$

In the applications below, we will have $n = 3$ or $n = 6$, and the test function g will be given by $|\rho(\mathbf{k})|^2$ and $|\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2$, respectively.
The assumptions (A1) on the form factor ψ imply that $|\rho|^2 \in \mathcal{S}(\mathbb{R}^3)$, and that the Fourier transform of $|\rho|^2$ is given by

$$\begin{aligned} & \widehat{|\rho|^2} = \widehat{\rho}\,\widehat{\overline{\rho}} = \widehat{\rho}\,\widehat{\rho} \\ =& (2\pi)^{-3/2} (\hat{\rho} * \hat{\rho}) = (2\pi)^{-3/2} \Big((\Lambda^3 \tilde{\psi}_0(\Lambda \cdot)) * (\Lambda^3 \tilde{\psi}_0(\Lambda \cdot)) \Big) \\ =& (2\pi)^{-3/2} \Lambda^3 (\psi_0 * \psi_0)(\Lambda \cdot), \end{aligned}$$

where $\tilde{f}(x) = f(-x)$ denotes reflection and we have used that ρ is real and ψ is even. The last equality follows from properties of the convolution, with the Fourier convention (2.1.6) used in this work. Note that the assumed properties of ψ_0 imply that $\Lambda^3 \psi_0(\Lambda \cdot)$ and $\Lambda^3 (\psi_0 * \psi_0)(\Lambda \cdot)$ are Dirac sequences. For the latter fact, note that $\int (\psi_0 * \psi_0)(x) dx = (\int \psi_0(x) dx)^2 = 1$ and $\psi_0 * \psi_0 \in C_0^\infty(\mathbb{R}^3)$.
Applying the preceding arguments to the two cases $g(\mathbf{k}) = |\rho(\mathbf{k})|^2$ and $g(\mathbf{k}_1, \mathbf{k}_2) = |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2$ and choosing $\lambda = R/\Lambda$, we find

$$\int_{\mathbb{R}^3} d\mathbf{k}\, T(\mathbf{k}) |\rho(\mathbf{k})|^2 \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}} = (2\pi)^{-3/2} \left(\frac{\Lambda}{R} \right)^{3+\alpha} \Big\langle \hat{T}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3 (\psi_0 * \psi_0)(R\cdot)] \Big\rangle_{\mathcal{S}'(\mathbb{R}^3),\mathcal{S}(\mathbb{R}^3)}$$

and

$$\begin{aligned} & \int_{\mathbb{R}^6} d\mathbf{k}_1 d\mathbf{k}_2\, T(\mathbf{k}_1, \mathbf{k}_2) |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\ =& (2\pi)^{-3} \left(\frac{\Lambda}{R} \right)^{6+\alpha} \Big\langle \hat{T}, \tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}[R^3 (\psi_0 * \psi_0)(R\cdot_1) R^3 (\psi_0 * \psi_0)(R\cdot_2)] \Big\rangle_{\mathcal{S}'(\mathbb{R}^6),\mathcal{S}(\mathbb{R}^6)}. \end{aligned}$$

Two things are important to note at this point. Firstly, the rescaling has generated inverse powers of $R = |\mathbf{R}|$, which is connected to the homogeneity of T. Secondly, it has produced Dirac sequences of test functions which are parametrized by the interatomic distance R and which are shifted away from the origin by an amount of the order of the ultraviolet-cutoff Λ. These observations motivate the next results, which will allow us to estimate the large-R decay of certain terms from the interaction potential which involve both the radiation field and the Coulomb potential, see below.
The following two lemmas state that under the additional assumption that the distributional Fourier transform $\hat{T}$ is represented by a smooth function outside the set of its singularities, we can calculate the $(R \to \infty)$-limits of the terms in pointed brackets explicitly.

Lemma 5.5.8. *Let $\Lambda > 0$ and $\mathbf{R} = R\hat{\mathbf{R}} \in \mathbb{R}^3$. Let $T \in \mathcal{S}'(\mathbb{R}^3)$ be such that away from $0 \in \mathbb{R}^3$, the distribution $\hat{T} \in \mathcal{S}'(\mathbb{R}^3)$ is represented by a function $\hat{T}(\cdot) \in C^\infty(\mathbb{R}^3 \setminus \{0\})$ when restricted to test functions of compact support, i.e.*

$$\langle \hat{T}, \varphi \rangle = \int \hat{T}(\mathbf{k})\varphi(\mathbf{k})d\mathbf{k}$$

for all $\varphi \in \mathcal{C}_0^\infty(\mathbb{R}^3)$ with $0 \notin \operatorname{supp}\varphi$. Furthermore, assume that ψ_0 is even, $\psi_0 \in C_0^\infty(\mathbb{R}^3)$, $\operatorname{supp}\psi_0 \subset B_1(0)$ and $\int \psi_0 = 1$. Then

$$\lim_{R\to\infty} \Big\langle \hat{T}, \tau_{\Lambda\hat{\mathbf{R}}}\Big[R^3(\psi_0 * \psi_0)(R\cdot)\Big]\Big\rangle = \hat{T}(\Lambda\hat{\mathbf{R}}). \tag{5.5.26}$$

Proof. Set $\Psi_R := \Big(R^3(\psi_0 * \psi_0)(R\cdot_1)\Big)$. Since $\operatorname{supp}(\psi_0 * \psi_0) \subset 2\operatorname{supp}\psi_0 \subset B_2(0)$, it follows that $\operatorname{supp}(\psi_0 * \psi_0)(R\cdot) \subset B_{2/R}(0)$ and

$$\operatorname{supp}\Big(\tau_{\Lambda\hat{\mathbf{R}}}\Psi_R(\cdot)\Big) \subset B_{2/R}(\Lambda\hat{\mathbf{R}}),$$

so that

$$0 \notin \operatorname{supp}\Big(\tau_{\Lambda\hat{\mathbf{R}}}\Psi_R(\cdot)\Big) \Leftrightarrow |\Lambda\hat{\mathbf{R}}| > 2/R \Leftrightarrow R > 2/\Lambda.$$

Choose $R_0 \geq 2/\Lambda$ and a bounded open set $\Omega \supset \operatorname{supp}\Big(\tau_{\Lambda\hat{\mathbf{R}}}\Psi_R(\cdot)\Big)$ with $0 \notin \Omega$, for instance, $\Omega := B_{2/R_0}(\Lambda\hat{\mathbf{R}})$. By the hypothesis on the distribution $\hat{T}$, we have $\hat{T}|_\Omega \in C_b^\infty(\Omega)$, and in particular $\hat{T}|_\Omega \in L^p(\Omega)$ for any $1 \leq p \leq \infty$. Furthermore, $\Psi_R|_\Omega \in C_0^\infty(\Omega)$ for all $R > R_0$, and Ψ_R is a Dirac sequence, which implies that $\hat{T} * \Psi_R \to \hat{T}$ in $L^p(\Omega)$ for $1 \leq p < \infty$. Since $\hat{T}|_\Omega$ is continuous, we conclude that also $\hat{T} * \Psi_R \to \hat{T}$ pointwise in Ω. Now for $R \geq R_0$ and by the choice of Ω,

$$\Big\langle \hat{T}, \tau_{\Lambda\hat{\mathbf{R}}}\Big[R^3(\psi_0 * \psi_0)(R\cdot)\Big]\Big\rangle = \int_\Omega \hat{T}(\mathbf{k})\Psi_R(\mathbf{k} - \Lambda\hat{\mathbf{R}})d\mathbf{k} = \Big(\hat{T} * \Psi_R\Big)(\Lambda\hat{\mathbf{R}}),$$

where we have used the definition of the convolution and the fact that Ψ_R is even. Since $\Lambda\hat{\mathbf{R}} \in \Omega$ by construction, this proves the assertion. □

The next result is a modification of the preceding one for the case $n = 6$, under the additional assumptions that the occurring test functions have a special structure.

Lemma 5.5.9. *Let $\Lambda > 0$ and $\mathbf{R} = R\hat{\mathbf{R}} \in \mathbb{R}^3$. Let $T \in \mathcal{S}'(\mathbb{R}^6)$ be such that away from the set $S = (\{0\} \times \mathbb{R}^3) \cup (\mathbb{R}^3 \times \{0\}) \subset \mathbb{R}^6$, the distribution $\hat{T} \in \mathcal{S}'(\mathbb{R}^6)$ is represented by a function $\hat{T}(\cdot_1, \cdot_2) \in C^\infty(\mathbb{R}^6 \setminus S)$ when restricted to test functions of compact support, i.e.*

$$\langle \hat{T}, \varphi \rangle = \int \hat{T}(\mathbf{k}_1, \mathbf{k}_2)\varphi(\mathbf{k}_1, \mathbf{k}_2)d\mathbf{k}_1 d\mathbf{k}_2$$

for all $\varphi \in \mathcal{C}_0^\infty(\mathbb{R}^6)$ with $S \cap \operatorname{supp}\varphi = \emptyset$. Furthermore, assume that ψ_0 is even, $\psi_0 \in C_0^\infty(\mathbb{R}^3)$, $\operatorname{supp}\psi_0 \subset B_1(0)$ and $\int \psi_0 = 1$. Then

$$\lim_{R\to\infty} \Big\langle \hat{T}, \tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Big[R^3(\psi_0 * \psi_0)(R\cdot_1)\, R^3(\psi_0 * \psi_0)(R\cdot_2)\Big]\Big\rangle = \hat{T}(\Lambda\hat{\mathbf{R}}, \Lambda\hat{\mathbf{R}}). \tag{5.5.27}$$

Proof. Set $\Psi_R := \Big(R^3(\psi_0 * \psi_0)(R\cdot_1)\Big)\Big(R^3(\psi_0 * \psi_0)(R\cdot_2)\Big)$. Since

$$\operatorname{supp}(\psi_0 * \psi_0) \subset 2\operatorname{supp}\psi_0 \subset B_2(0),$$

it follows that $\operatorname{supp}(\psi_0 * \psi_0)(R\cdot) \subset B_{2/R)}(0)$ and

$$\begin{aligned}\operatorname{supp}\Big(\tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Psi_R(\cdot_1,\cdot_2)\Big) =& \operatorname{supp}\Big(\tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Big[R^3(\psi_0 * \psi_0)(R\cdot_1)\, R^3(\psi_0 * \psi_0)(R\cdot_2)\Big]\Big)\\ &\subset B_{2/R}(\Lambda\hat{\mathbf{R}}) \times B_{2/R}(\Lambda\hat{\mathbf{R}}),\end{aligned}$$

so that

$$S \cap \operatorname{supp}\Big(\tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Psi_R(\cdot_1,\cdot_2)\Big) = \emptyset \Leftrightarrow |\Lambda\hat{\mathbf{R}}| > 2/R \Leftrightarrow R > 2/\Lambda.$$

Choose $R_0 \geq 2/\Lambda$ and a bounded open set $\Omega \supset \operatorname{supp}\Big(\tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Psi_R(\cdot_1,\cdot_2)\Big)$ with $0 \notin \Omega$, for instance $\Omega := B_{2/R_0}(\Lambda\hat{\mathbf{R}}) \times B_{2/R_0}(\Lambda\hat{\mathbf{R}})$. By the hypothesis on the distribution $\hat{T}$, we have $\hat{T}|_\Omega \in C_b^\infty(\Omega)$, and in particular $\hat{T}|_\Omega \in L^p(\Omega)$ for any $1 \leq p \leq \infty$. Furthermore, $\Psi_R|_\Omega \in C_0^\infty(\Omega)$ for all $R > R_0$, and Ψ_R is a Dirac sequence, which implies that $\hat{T} * \Psi_R \to \hat{T}$ in $L^p(\Omega)$ for $1 \leq p < \infty$. Since $\hat{T}|_\Omega$ is continuous, we conclude that also $\hat{T} * \Psi_R \to \hat{T}$ pointwise in Ω. Now for $R \geq R_0$ and by the choice of Ω,

$$\begin{aligned}&\Big\langle \hat{T}, \tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Big[R^3(\psi_0 * \psi_0)(R\cdot_1)\, R^3(\psi_0 * \psi_0)(R\cdot_2)\Big]\Big\rangle\\ =&\int_\Omega \hat{T}(\mathbf{k}_1,\mathbf{k}_2)\Psi_R(\mathbf{k}_1 - \Lambda\hat{\mathbf{R}}, \mathbf{k}_2 - \Lambda\hat{\mathbf{R}})d\mathbf{k}_1 d\mathbf{k}_2\\ =&\Big(\hat{T} * \Psi_R\Big)(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}}),\end{aligned}$$

where we have used the definition of the convolution and the fact that Ψ_R is even. Since $(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}}) \in \Omega$ by construction, this proves the assertion. □

The next two lemmas constitute a generalization of the well-known fact that the Fourier transform maps multiplication by polynomials to derivatives to the case of tempered distributions which are represented by smooth functions outside their sets of singularities. We will use these results in the following sections to calculate

$$\lim_{R\to\infty}\Big\langle \hat{T}, \tau_{\Lambda\hat{\mathbf{R}}}\Big[R^3(\psi_0 * \psi_0)(R\cdot)\Big]\Big\rangle$$

and

$$\lim_{R\to\infty}\Big\langle \hat{T}, \tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}\Big[R^3(\psi_0 * \psi_0)(R\cdot_1)\, R^3(\psi_0 * \psi_0)(R\cdot_2)\Big]\Big\rangle$$

in situations where $\hat{T}$ is not known explicitly (and might not even be globally representable by an L^1_{loc}-function), but where T has the structure of a product of a polynomial with a function whose distributional Fourier transform is known explicitly.

Lemma 5.5.10. *Suppose that $f \in L^1_{loc}(\mathbb{R}^n)$ is of the form $f(\mathbf{k}) = P(-\mathrm{i}\mathbf{k})g(\mathbf{k})$, where $P(x) = \sum_{|\alpha|\leq m} c_\alpha x^\alpha$ is a polynomial of degree m (multi-index notation is used here) and $g \in L^1_{loc}(\mathbb{R}^n) \cap C^\infty(\mathbb{R}^n \setminus \{0\})$ grows at most polynomially at infinity (for instance because it is homogeneous). Then f also grows at most polynomially at infinity, and thus f and g define tempered distributions $T_f, T_g \in \mathcal{S}'(\mathbb{R}^n)$. Furthermore, suppose that $\hat{T}_g = T_{\hat{g}}$ for a function $\hat{g} \in L^1_{loc}(\mathbb{R}^n) \cap C^\infty(\mathbb{R}^n \setminus \{0\})$ (necessarily also fulfilling a growth estimate at infinity).*
Then on test functions $\varphi \in C_0^\infty(\mathbb{R}^n)$ with $0 \notin \operatorname{supp}\varphi$, $\hat{T}_f$ is given by $P(D)\hat{g}$, where $D = \nabla_{\mathbf{k}}$ denotes the classical derivative)

Proof. Let $\tilde{T}$ denote the operator of reflection, i.e. $\langle \tilde{T}, \varphi\rangle := \langle T, \tilde{\varphi}\rangle$, where $\tilde{\varphi}(x) := \varphi(-x)$. Let $\varphi \in C_0^\infty(\mathbb{R}^n)$ and assume $0 \notin \operatorname{supp}\varphi$. By the definition of the distributional Fourier transform, the multiplication of tempered distributions with C^∞-functions growing sufficiently slow at infinity, the behaviour of the Fourier transformation under multiplication with polynomials in the coordinates, and the partial derivative of distributions, we obtain

$$\begin{aligned}
\langle \hat{T}_f, \varphi\rangle &= \langle T_f, \hat{\varphi}\rangle = \langle T_{P(-\mathrm{i}\mathbf{k})g}, \hat{\varphi}\rangle \\
&= \langle T_g, P(-\mathrm{i}\mathbf{k})\hat{\varphi}\rangle = \langle T_g, \widehat{P(-\mathrm{i}\mathbf{k})}\hat{\varphi}\rangle = \langle T_g, \widehat{P(\mathrm{i}\mathbf{k})}\hat{\hat{\varphi}}\rangle \\
&= \langle T_g, \widehat{P(-D)}\hat{\hat{\varphi}}\rangle = \langle \widehat{T_g}, P(-D)\varphi\rangle = \langle P(D)\widehat{T_g}, \varphi\rangle \\
&= \langle T_{\hat{g}}, P(-D)\varphi\rangle.
\end{aligned}$$

Now choose functions $\chi_1 \in C^\infty(\mathbb{R}^n)$ and $\chi_2 \in C_0^\infty(\mathbb{R}^n)$ with $\chi_1 + \chi_2 = 1$ and $\chi_2 = 1$ on $B_L(0)$, where $L > 0$ is a parameter to be chosen below. This yields

$$\begin{aligned}
\langle \hat{T}_f, \varphi\rangle &= \langle T_{\chi_1\hat{g}+\chi_2\hat{g}}, P(-D)\varphi\rangle \\
&= \langle \chi_1 T_{\hat{g}}, P(-D)\varphi\rangle + \langle \chi_2 T_{\hat{g}}, P(-D)\varphi\rangle
\end{aligned}$$

(note that χ_1 is bounded and thus $\chi_1 T_{\hat{g}}$ is a tempered distribution.) Since $0 \notin \operatorname{supp}\chi_1$ and $\hat{g} \in C^\infty(\mathbb{R}^n \setminus \{0\})$, we have $\chi_1\hat{g} \in C^\infty(\mathbb{R}^n)$. Therefore, the partial derivative $P(D)T_{\chi_1\hat{g}}$ exists (as a tempered distribution) and is given by $T_{P(D)(\chi_1\hat{g})}$. Furthermore, $\chi_2\hat{g} \in C_0^\infty(\mathbb{R}^n \setminus \{0\}) \cap L^1_{loc}(\mathbb{R}^3)$ since χ_2 has compact support. Thus we have

$$\langle \hat{T}_f, \varphi\rangle = \langle T_{P(D)(\chi_1\hat{g})}, \varphi\rangle + \langle T_{\chi_2\hat{g}}, P(-D)\varphi\rangle.$$

Now since $0 \notin \operatorname{supp}\varphi$ by assumption, we also have $0 \notin \operatorname{supp}(P(-D)\varphi)$, and thus we can find an L such that $\chi_1 \equiv 1$ on $\operatorname{supp}\varphi$ and $\operatorname{supp}(\chi_2\hat{g}) \cap \operatorname{supp}(P(-D)\varphi) = \emptyset$. But then $P(D)\chi_1\hat{g} = P(D)\hat{g}$ (as $C^\infty(\mathbb{R}^n)$-functions on $\operatorname{supp}\varphi$), and

$$\langle T_{\chi_2\hat{g}}, P(-D)\varphi\rangle = 0,$$

i.e. we have shown that

$$\langle \widehat{T_{P(-i\mathbf{k})g}}, \varphi\rangle = \langle T_{P(D)\hat{g}}, \varphi\rangle$$

for all $\varphi \in C_0^\infty(\mathbb{R}^n)$ with $0 \notin \operatorname{supp}\varphi$. □

Remark: In the above Lemma, we could also assume the weaker condition that $\langle \hat{T}_g, \varphi\rangle = \langle \hat{g}, \varphi\rangle$ for all $\varphi \in C_0^\infty(\mathbb{R}^n)$ such that $0 \notin \operatorname{supp}\varphi$.

Lemma 5.5.11. *Suppose that $f \in L^1_{loc}(\mathbb{R}^{2n})$ is of the form*

$$f(\mathbf{k}_1, \mathbf{k}_2) = P(-\mathrm{i}\mathbf{k}_1, -\mathrm{i}\mathbf{k}_2) g(\mathbf{k}_1, \mathbf{k}_2),$$

where P is a polynomial and $g \in L^1_{loc}(\mathbb{R}^{2n}) \cap C^\infty(\mathbb{R}^{2n} \setminus S)$, with $S = (\mathbb{R}^n \times \{0\}) \cup (\{0\} \times \mathbb{R}^n)$, grows at most polynomially at infinity. Then f also grows at most polynomially at infinity, and thus f and g define tempered distributions $T_f, T_g \in \mathcal{S}'(\mathbb{R}^n)$.
Suppose that the distributional Fourier transform of g satisfies $\hat{T}_g = T_{\hat{g}}$ for a function $\hat{g} \in L^1_{loc}(\mathbb{R}^{2n}) \cap C^\infty(\mathbb{R}^n \setminus S)$.
Then on test functions of the form $\varphi(\cdot_1)\varphi(\cdot_2) \in C_0^\infty(\mathbb{R}^{2n})$, where $\varphi \in C_0^\infty(\mathbb{R}^n)$ and $0 \notin \operatorname{supp}\varphi$, $\hat{T}_f$ is given by $P(D)\hat{g}$, where $D = \nabla_{\mathbf{k}}$ is the classical derivative.

Proof. As in the proof of Lemma 5.5.10, we begin by noting that

$$\langle \hat{T}_f, (\varphi(\cdot_1)\varphi(\cdot_2)\rangle = \langle T_{\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2)\rangle.$$

Now choose functions $\chi_1 \in C^\infty(\mathbb{R}^n)$, $\chi_2 \in C_0^\infty(\mathbb{R}^n)$ with $\chi_1 + \chi_2 = 1$ and $\chi_2 = 1$ on $B_L(0)$, where $L > 0$ is a parameter to be chosen below. Using $(\chi_1 + \chi_2)(\cdot_1)(\chi_1 + \chi_2)(\cdot_2) \equiv 1$ (as a multiplier on $\mathbb{R}^{2n}$) yields

$$\begin{aligned}
&\langle \hat{T}_f, (\varphi(\cdot_1)\varphi(\cdot_2))\rangle = \langle T_{\chi_1\chi_1\hat{g} + \chi_2\chi_2\hat{g} + \chi_1\chi_2\hat{g} + \chi_2\chi_1\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\rangle \\
=&\Big\langle \chi_1(\cdot_1)\chi_1(\cdot_2)T_{\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle + \Big\langle \chi_2(\cdot_1)\chi_2(\cdot_2)T_{\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle \\
&+ \Big\langle \chi_1(\cdot_1)\chi_2(\cdot_2)T_{\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle + \Big\langle \chi_2(\cdot_1)\chi_1(\cdot_2)T_{\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle.
\end{aligned}$$

Note that since χ_1 and χ_2 are smooth and bounded, all the $\chi_i\chi_j T_{\hat{g}}$ are tempered distributions. By the structure of the singular set S, we have $S \cap \operatorname{supp}(\chi_1(\cdot_1)\chi_1(\cdot_1)) = \emptyset$, and the assumption $\hat{g} \in C^\infty(\mathbb{R}^{2n} \setminus S)$ thus implies $\chi_1(\cdot_1)\chi_1(\cdot_2)\hat{g} \in C^\infty(\mathbb{R}^{2n})$. Therefore, the partial derivative $P(D)T_{\chi_1\chi_1\hat{g}}$ exists (as a tempered distribution) and is given by $T_{P(D)(\chi_1\chi_1\hat{g})}$. Thus we have

$$\begin{aligned}
\Big\langle \chi_1(\cdot_1)\chi_1(\cdot_2)T_{\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle =& \Big\langle P(D)T_{\chi_1\chi_1\hat{g}}, (\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle \\
=& \Big\langle T_{P(D)(\chi_1\chi_1\hat{g})}, (\varphi(\cdot_1)\varphi(\cdot_2))\Big\rangle.
\end{aligned}$$

Now since $0 \notin \operatorname{supp}\varphi$ by assumption, we can find an L such that $\chi_1 \equiv 1$ on $\operatorname{supp}\varphi$ and $\operatorname{supp}\chi_2 \cap \operatorname{supp}\varphi = \emptyset$, which implies that $P(D)\chi_1\chi_1\hat{g} = P(D)\hat{g}$ as $C^\infty(\mathbb{R}^{2n})$-functions on $\operatorname{supp}(\varphi(\cdot_1)\varphi(\cdot_2))$. Furthermore,

$$\operatorname{supp}(P(-D)(\varphi(\cdot_1)\varphi(\cdot_2)) \subset \operatorname{supp}((\varphi(\cdot_1)\varphi(\cdot_2)),$$

with the consequence that that for such an L,

$$\begin{aligned}
&\operatorname{supp}(\chi_2(\cdot_1)\chi_2(\cdot_2)\tilde{g}) \cap \operatorname{supp}(P(-D)(\varphi(\cdot_1)\varphi(\cdot_2)) \\
=&\operatorname{supp}(\chi_1(\cdot_1)\chi_2(\cdot_2)\tilde{g}) \cap \operatorname{supp}(P(-D)(\varphi(\cdot_1)\varphi(\cdot_2)) \\
=&\operatorname{supp}(\chi_2(\cdot_1)\chi_1(\cdot_2)\tilde{g}) \cap \operatorname{supp}(P(-D)(\varphi(\cdot_1)\varphi(\cdot_2)) \\
=&\emptyset,
\end{aligned}$$

which in turn implies

$$\begin{aligned}\langle T_{\chi_2\chi_2\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\rangle =&\langle T_{\chi_1\chi_2\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\rangle\\ =&\langle T_{\chi_2\chi_1\hat{g}}, P(-D)(\varphi(\cdot_1)\varphi(\cdot_2))\rangle\\ =&0,\end{aligned}$$

finishing the proof. □

No $1/R^7$ (or less)-contributions from higher terms in multipole expansion

In this section we will show that the contributions $B_1(s,d,\mathbf{R})$ and $B_2(s,d,\mathbf{R})$ to (5.5.24) (see (5.5.22) and (5.5.23) above for their definition) in the lower-order terms of the multipole expansion decay strictly faster than $1/R^7$ if $s \geq 2$.
Upon setting

$$M_{89}^L(\mathbf{R},d) := \sum_{l\geq 4, l \text{ even}}^{L} B_1(l/2,d,\mathbf{R}) + B_2(l/2,d,\mathbf{R}),$$

for $L \in N$, $L \geq 2$, the corresponding claim in Theorem 5.4.1 (see (5.4.3)) will follow from Lemmas 5.5.12 and 5.5.13 below.

Lemma 5.5.12. *Assume the hypotheses of Theorem 5.4.1 and let $s \in \mathbb{N}$, $s \geq 2$ and $k < 8$. Then*

$$\lim_{R\to\infty} (R^k\, B_1(s,d,\mathbf{R})) = 0$$

uniformly in d.

Proof. We will first estimate $S_{\alpha,\beta}^{2s,l_1,m,m_1}(d)$. Introduce the function

$$\begin{aligned}v_d(\mathbf{x}_1,\dots,\mathbf{x}_N) :=&\Big(\sum_{i_A} |\mathbf{x}_{i_A}|^{l_1} Y_{l_1,m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\Big)\\ &\times\Big(\sum_{j_B} |\mathbf{x}_{j_B}|^{l-l_1} Y_{l-l_1,m-m_1}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\Big)\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0),\end{aligned}$$

where χ_{Ω_d} is the smooth characteristic function of the set Ω_d introduced in the assumptions of Theorem 5.4.1. Then we can write

$$S_{\alpha,\beta}^{2s,l_1,m,m_1}(d) = \Big\langle (\overline{H_A+H_B})^{-1}[\mathbf{v}_A^\alpha \mathbf{v}_B^\beta] | v_d\Big\rangle_{L^2(\mathbb{R}^{3N})},$$

which yields the estimate

$$|S_{\alpha,\beta}^{2s,l_1,m,m_1}(d)| \leq \|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\|(\overline{H_A+H_B})^{-1}\|\|v_d\|_{L^2(\mathbb{R}^{3N})}.$$

By the exponential decay of Ψ_A^0 and Ψ_B^0 (see Proposition 2.5.1),

$$v_\infty := \Big(\sum_{i_A} |\mathbf{x}_{i_A}|^{l_1} Y_{l_1,m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\Big)\Big(\sum_{j_B} |\mathbf{x}_{j_B}|^{l-l_1} Y_{l-l_1,m-m_1}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}]\Big)(\Psi_A^0\otimes\Psi_B^0)$$

is in $L^2(\mathbb{R}^{3N})$. Furthermore, $\|v_d\| \leq \|v_\infty\|$ by construction of χ_{Ω_d}, so that we conclude

$$|S^{2s,l_1,m,m_1}_{\alpha,\beta}(d)| \leq \|\mathbf{v}^\alpha_A\|_{L^2(\mathbb{R}^{3Z_A})}\|\mathbf{v}^\beta_B\|_{L^2(\mathbb{R}^{3Z_B})}\|(\overline{H_A+H_B})^{-1}\|\|v_\infty\|_{L^2(\mathbb{R}^{3N})},$$

this estimate now being independent of d. By Lemma 5.1.2, $\lim\limits_{R\to\infty} T^{2s,m}_\Lambda(\mathbf{R})$ exists, so $T^{2s,m}_\Lambda(\mathbf{R})$ is bounded with respect to R, but depends on Λ. Next we investigate the integral

$$\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\left(\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2})\right)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}.$$

The contribution

$$\delta_{\alpha,\beta}\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}$$

decays faster than any inverse power of R, since it is the Fourier transform of the Schwartz function $|\rho|^2$ (up to a constant). Note, however, that by the scaling $\rho(\mathbf{k}) = \rho_0(\mathbf{k}/\Lambda)$, the rate of decay depends on Λ. Writing the second contribution in distributional form yields

$$-\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}(x) = -\frac{1}{(2\pi)^{3/2}}\left(\frac{\Lambda}{R}\right)^3\left\langle\widehat{\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}},\tau_{(\Lambda\hat{\mathbf{R}})}[R^3(\psi_0*\psi_0)(R\cdot)]\right\rangle,$$

see the previous section. Since for $R > R_0 = 2/\Lambda$ we have $0 \notin \mathrm{supp}(\tau_{(\Lambda\hat{\mathbf{R}})}[R^3(\psi_0*\psi_0)(R\cdot)])$, Lemma 5.5.10 tells us that the action of $\widehat{\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}}$ on this test function is given by integration against

$$-\partial_\alpha\partial_\beta(\widehat{\frac{1}{|\mathbf{k}|^2}}) = \frac{(2\pi)^{1/2}}{|x|^3}(\delta_{\alpha,\beta}-3\hat{x}_\alpha\hat{x}_\beta).$$

Lemma 5.5.8 allows us to conclude that

$$\begin{aligned}\lim_{R\to\infty}\left\langle\widehat{\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}},\tau_{(\Lambda\hat{\mathbf{R}})}[R^3(\psi_0*\psi_0)(R\cdot)]\right\rangle =& \frac{(2\pi)^{1/2}}{|\Lambda\hat{\mathbf{R}}|^3}(\delta_{\alpha,\beta}-3\widehat{\Lambda\hat{\mathbf{R}}}_\alpha\widehat{\Lambda\hat{\mathbf{R}}}_\beta)\\ =& \frac{(2\pi)^{1/2}}{\Lambda^3}(\delta_{\alpha,\beta}-3\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta).\end{aligned}$$

In particular, $\langle\widehat{\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}},\tau_{(\Lambda\hat{\mathbf{R}})}[R^3(\psi_0*\psi_0)(R\cdot)]\rangle$ is bounded uniformly in R. We conclude

$$\begin{aligned}&R^k\,B_1(s,d,\mathbf{R})\\ =&-2\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde{C}_{2s,m,l_1,m_1}S^{2s,l_1,m,m_1}_{\alpha,\beta}(d)T^{2s,m}_\Lambda(\mathbf{R})\\ &\times\Bigg[(2\pi)^{3/2}\delta_{\alpha,\beta}\,\Lambda^{2s+1}R^{k-(2s+1)}\widehat{|\rho|^2}(\mathbf{R})\\ &-\frac{1}{(2\pi)^{3/2}}\Lambda^{2s+4}R^{k-(2s+4)}\left\langle\widehat{\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}},\tau_{(\Lambda\hat{\mathbf{R}})}[R^3(\psi_0*\psi_0)(R\cdot)]\right\rangle\Bigg]\Bigg],\end{aligned}$$

and applying the above estimates yields

$$|R^k\,B_1(s,d,\mathbf{R})| \leq C(\Lambda)(R^{k-(2s+1)}|\widehat{|\rho|^2}(\mathbf{R})| + R^{k-(2s+4)}),$$

where the constant $C(\Lambda)$ depends on the ultraviolet-cutoff scale Λ, but is independent of R and the parameter d used in the multipole expansion. The first term on the right-hand side tends to zero as $R \to \infty$ for any choice of k and s by the rapid decay of $\widehat{|\rho|^2}$, while the second term does so if $k < 8$ and $s \geq 2$, proving the assertion. □

Lemma 5.5.13. *Assume the hypotheses of Theorem 5.4.1 and let $s \in \mathbb{N}$, $s \geq 2$. Then*

$$\lim_{R\to\infty} (R^k\, B_2(s, d, \mathbf{R})) = 0$$

uniformly in d for all $k < 9$.

Proof. In contrast to the proof of the preceding Lemma, where the matrix elements occurring in the integrals (collected in the functions $S^{2s,l_1,m,m_1}_{\alpha,\beta}(d)$) were independent of the photon momenta, now we have to deal with the functions $T^{2s,l_1,m,m_1}_{\alpha,\beta}(d, |\mathbf{k}|)$, which do depend on $\mathbf{k}$ and whose Fourier transforms we do not know explicitly. Therefore, we will employ a classic technique to extract the asymptotic behaviour of oscillatory integrals by successive partial integration.
The first step is to note that since $\omega(\mathbf{k})$ and

$$\varphi_{\alpha,\beta}(|\mathbf{k}|, d, \Lambda, 2s, m, m_1, l_1) := |\rho(\mathbf{k})|^2\, T^{2s,l_1,m,m_1}_{\alpha,\beta}(d, |\mathbf{k}|)$$

only depend radially on $\mathbf{k}$, we can carry out the angular integration in (5.5.23) explicitly by using the identity

$$\begin{aligned}&\int \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}(1 - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}})d\Omega_k \\ =&4\pi\Big[(1 - \hat{\mathbf{R}} \otimes \hat{\mathbf{R}})\frac{\sin(kR)}{kR} + (1 - 3\hat{\mathbf{R}} \otimes \hat{\mathbf{R}})\left(\frac{\cos(kR)}{k^2R^2} - \frac{\sin(kR)}{k^3R^3}\right)\Big],\end{aligned}$$

which leads to the integral

$$\begin{aligned}&\int_{\mathbb{R}^3} (\delta_{\alpha,\beta} - \hat{k}_\alpha\hat{k}_\beta)\omega(\mathbf{k})\varphi_{\alpha,\beta}(|\mathbf{k}|, d, \Lambda, 2s, m, m_1, l_1)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}d\mathbf{k} \\ =&4\pi c\int_0^\infty d\xi\Big[(\delta_{\alpha,\beta} - \hat{R}_\alpha\hat{R}_\beta)\big(\frac{1}{R}\xi^2\sin(\xi R)\big) + (\delta_{\alpha,\beta} - 3\hat{R}_\alpha\hat{R}_\beta)\big(\frac{1}{R^2}\xi\cos(\xi R) - \frac{1}{R^3}\sin(\xi R)\big)\Big] \\ &\times \varphi_{\alpha,\beta}(\xi, d, \Lambda, 2s, m, m_1, l_1).\end{aligned}$$

We collect some properties of the functions $\xi^m\varphi_{\alpha,\beta}$ in the following

Lemma 5.5.14. *Under the hypotheses of Lemma 5.5.13, the map*

$$\xi \mapsto \xi^m\varphi_{\alpha,\beta}(\xi, d, \Lambda, 2s, m, m_1, l_1)$$

is in $C^\infty((0,\infty))$. Furthermore, $\xi^m\varphi_{\alpha,\beta}$ and all its derivatives $\frac{d^n}{d\xi^n}(\xi^m\varphi_{\alpha,\beta})$ are continuous at $\xi = 0$ and decay rapidly. In particular, $\frac{d^n}{d\xi^n}(\xi^m\varphi_{\alpha,\beta}) \in L^1((0,\infty)) \cap L^\infty((0,\infty))$, $\lim_{\xi\to\infty}\frac{d^n}{d\xi^n}(\xi^m\varphi_{\alpha,\beta}) = 0$ and $\lim_{\xi\to 0}\frac{d^n}{d\xi^n}(\xi^m\varphi_{\alpha,\beta}) = 0$ if $n < m$. Integrability and boundedness hold uniformly in d.

Proof. First note that

$$\begin{aligned}
&\frac{d^n}{d\xi^n}(\xi^m\varphi_{\alpha,\beta})\\
=&\sum_{k=0}^{n}\binom{n}{k}\frac{d^{n-k}}{d\xi^{n-k}}(\xi^m)\frac{d^k}{d\xi^k}\varphi_{\alpha,\beta}(\xi,\dots)\\
=&\sum_{k=0}^{n}\binom{n}{k}\frac{d^{n-k}}{d\xi^{n-k}}(\xi^m)\left(\sum_{t=0}^{k}\binom{k}{t}\frac{d^{k-t}}{d\xi^{k-t}}(T_{\alpha,\beta}^{2s,l_1,m,m_1}(\xi,d))\frac{d^t}{d\xi^t}(|\rho(\xi)|^2)\right)
\end{aligned}$$

by Leibniz' rule. Since $\rho(\xi)$ is a Schwartz function, the same is true for $\frac{d^t}{d\xi^t}(|\rho(\xi)|^2)$, so it suffices to show that $\frac{d^n}{d\xi^n}(T_{\alpha,\beta}^{2s,l_1,m,m_1}(\xi,d))$ exists, is bounded (uniformly in d) and continuous at zero for all $n\geq 0$. To this end, first consider the term (5.5.19) and define the functions

$$\begin{aligned}
u_{A,d}&:=\Pi_{i=1}^{Z_A}\chi_d(\cdot_i)\,(\sum_{i_A}|\mathbf{x}_{i_A}|^{l_1}Y_{l_1,m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}])\Psi_A^0,\\
u_{B,d}&:=\Pi_{i=Z_A+1}^{N}\chi_d(\cdot_i)\overline{(\sum_{j_B}|\mathbf{x}_{j_B}|^{l-l_1}Y_{l-l_1,m-m_1}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}])}\Psi_B^0.
\end{aligned}$$

Then (5.5.19) becomes

$$\langle\mathbf{v}_A^\alpha|(H_A+\hbar\omega(\xi))^{-1}|u_{A,d}\rangle_{L^2(\mathbb{R}^{3Z_A})}\langle u_{B,d}|(H_B+\hbar\omega(\xi))^{-1}|\mathbf{v}_B^\beta\rangle_{L^2(\mathbb{R}^{3Z_B})}.$$

Using the spectral resolutions $E_A(\lambda)$ and $E_B(\lambda)$ of the self-adjoint operators $H_{A|\{\Psi_A^0\}^\perp}$ and $H_{B|\{\Psi_B^0\}^\perp}$, this can be expressed as

$$\begin{aligned}
&\left(\int_{\mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp})}\frac{1}{\lambda+\hbar\omega(\xi)}d\langle\mathbf{v}_A^\alpha|E_A(\lambda)|u_{A,d}\rangle_{L^2(\mathbb{R}^{3Z_A})}\right)\\
&\times\left(\int_{\mathrm{spec}(H_{B|\{\Psi_B^0\}^\perp})}\frac{1}{\lambda+\hbar\omega(\xi)}d\langle u_{B,d}|E_B(\lambda)|\mathbf{v}_B^\beta\rangle_{L^2(\mathbb{R}^{3Z_B})}\right).
\end{aligned}$$

Note that since $\mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp})\subset[\Delta_A,\infty)$ and $\mathrm{spec}(H_{B|\{\Psi_B^0\}^\perp})\subset[\Delta_B,\infty)$, the functions $\lambda\mapsto\frac{1}{(\lambda+\hbar\omega(\xi))^{n+1}}$ (for $\lambda\in\mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp})$ and $\lambda\in\mathrm{spec}(H_{B|\{\Psi_B^0\}^\perp})$, respectively) are bounded uniformly in ξ for any $n\geq 0$. Thus by a standard result on parameter-dependent integrals, both factors are differentiable with respect to ξ, with derivatives given by

$$\begin{aligned}
&\frac{d^n}{d\xi^n}\left(\int_{\mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp})}\frac{1}{\lambda+\hbar\omega(\xi)}d\langle\mathbf{v}_A^\alpha|E_A(\lambda)|u_{A,d}\rangle_{L^2(\mathbb{R}^{3Z_A})}\right)\\
=&(-1)^n n!(\hbar c)^n\int_{\mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp})}\frac{1}{(\lambda+\hbar\omega(\xi))^{n+1}}d\langle\mathbf{v}_A^\alpha|E_A(\lambda)|u_{A,d}\rangle_{L^2(\mathbb{R}^{3Z_A})}\\
=&(-1)^n n!(\hbar c)^n\langle\mathbf{v}_A^\alpha|(H_A+\hbar\omega(\xi))^{-(n+1)}|u_{A,d}\rangle_{L^2(\mathbb{R}^{3Z_A})},
\end{aligned}$$

and accordingly for the second factor. Continuity at $\xi = 0$ is easily read off from this expression. Using the Cauchy-Schwarz inequality and the resolvent estimate

$$\|(H_A + \hbar\omega(\xi))^{-(n+1)}\| \leq (1/\mathrm{dist}(-\hbar\omega(\xi), \mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp}))^{n+1} \leq (1/\Delta_A)^{n+1},$$

we conclude

$$\begin{aligned}
&\Big|\frac{d^n}{d\xi^n}\Big(\langle \mathbf{v}_A^\alpha|(H_A + \hbar\omega(\xi))^{-1}|u_{A,d}\rangle_{L^2(\mathbb{R}^{3Z_A})}\langle u_{B,d}|(H_B + \hbar\omega(\xi))^{-1}|\mathbf{v}_B^\beta\rangle_{L^2(\mathbb{R}^{3Z_B})}\Big)\Big| \\
\leq &\sum_{t=0}^{n}\binom{n}{t}(n-t)!\,t!\,(\hbar c)^n(1/\Delta_A)^{n-t+1}(1/\Delta_B)^{t+1}\|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})}\|u_{A,d}\|_{L^2(\mathbb{R}^{3Z_A})} \\
&\quad\times\|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})}\|u_{B,d}\|_{L^2(\mathbb{R}^{3Z_B})}.
\end{aligned}$$

The exponential decay of Ψ_A^0 and Ψ_B^0 (see Proposition 2.5.1) implies that

$$\begin{aligned}
u_{A,\infty} &:= \sum_{i_A}|\mathbf{x}_{i_A}|^{l_1}Y_{l_1,m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}]\Psi_A^0 \in L^2(\mathbb{R}^{3Z_A}), \\
u_{B,\infty} &:= \sum_{j_B}|\mathbf{x}_{j_B}|^{l-l_1}Y_{l-l_1,m-m_1}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \in L^2(\mathbb{R}^{3Z_B}),
\end{aligned}$$

and the fact that $0 \leq \chi_d \leq 1$ yields the estimates $\|u_{A,d}\|_{L^2(\mathbb{R}^{3Z_A})} \leq \|u_{A,\infty}\|_{L^2(\mathbb{R}^{3Z_A})}$, $\|u_{B,d}\|_{L^2(\mathbb{R}^{3Z_B})} \leq \|u_{B,\infty}\|_{L^2(\mathbb{R}^{3Z_B})}$, whose right-hand sides are now independent of the parameter d used in the multipole-expansion.
To estimate (5.5.20) and its derivatives, define

$$\begin{aligned}
w_d(\mathbf{x}_1,\ldots,\mathbf{x}_N) := \Bigg(&(\overline{H_A + H_B})^{-1}\Big[(\sum_{i_A}|\mathbf{x}_{i_A}|^{l_1}Y_{l_1,m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}]) \\
&\times(\sum_{j_B}|\mathbf{x}_{j_B}|^{l-l_1}Y_{l-l_1,m-m_1}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}])\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big]\Bigg)(\mathbf{x}_1,\ldots,\mathbf{x}_N)
\end{aligned}$$

and rewrite

$$(5.5.20) = \Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta|\overline{((H_A + \hbar\omega(\xi))^{-1}\otimes I)_{|W}}|w_d\Big\rangle_{L^2(\mathbb{R}^{3N})},$$

where $W = \overline{\{\Psi_A^0\}\perp\widehat{\otimes}\{\Psi_B^0\}^\perp}$. As in the proof of Lemma 4.2.5 (see also the proof of Lemma 4.2.18), one notes that

$$\begin{aligned}
\overline{((H_A + \hbar\omega(\xi))^{-1}\otimes I)_{|W}} &= \Big(\overline{((H_A + \hbar\omega(\xi))\otimes I)_{|W}}\Big)^{-1} \\
&= \Big(\overline{(H_A\otimes I + \hbar\omega(\xi)(I\otimes I))_{|W}}\Big)^{-1}.
\end{aligned}$$

The operator $(H_A\otimes I + \hbar\omega(\xi)(I\otimes I))_{|W} = (H_A\otimes I)_{|W} + \hbar\omega(\xi)I_{|W}$ is essentially self-adjoint and satisfies the inclusion

$$(H_A\otimes I)_{|W} + \hbar\omega(\xi)I_{|W} \subset \overline{(H_A\otimes I)_{|W}} + \hbar\omega(\xi)I_{|W},$$

the latter operator being self-adjoint (as sum of a self-adjoint and a bounded self-adjoint operator). The uniqueness of the self-adjoint extension for essentially self-adjoint operators now implies

$$\overline{(H_A \otimes I + \hbar\omega(\xi)(I \otimes I))_{|W}} = \overline{(H_A \otimes I)_{|W}} + \hbar\omega(\xi) I_{|W},$$

establishing

$$\overline{((H_A + \hbar\omega(\xi))^{-1} \otimes I)_{|W}} = \Big(\overline{(H_A \otimes I)_{|W}} + \hbar\omega(\xi) I_{|W}\Big)^{-1}.$$

This means that we can use the spectral resolution $\tilde{E}(\lambda)$ of the self-adjoint operator $\overline{(H_A \otimes I)_{|W}}$ to conclude

$$\begin{aligned}
&\Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | \overline{((H_A + \hbar\omega(\xi))^{-1} \otimes I)_{|W}} | w_d \Big\rangle_{L^2(\mathbb{R}^{3N})} \\
&= \int_{\mathrm{spec}(\overline{(H_A \otimes I)_{|W}})} \frac{1}{\lambda + \hbar\omega(\xi)} d\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | \tilde{E}(\lambda) | w_d \rangle_{L^2(\mathbb{R}^{3N})}.
\end{aligned}$$

By the proof of Lemma 4.2.5,

$$\mathrm{spec}(\overline{(H_A \otimes I)_{|W}}) = \mathrm{spec}(\overline{H_{A|\{\Psi_A^0\}^\perp} \otimes I_{\{\Psi_B^0\}^\perp}}) = \mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp}) \subset [\Delta_A, \infty),$$

so the function $\lambda \mapsto \frac{1}{\lambda + \hbar\omega(\xi)}$ is uniformly bounded in ξ on $\mathrm{spec}(\overline{(H_A \otimes I)_{|W}})$. The argument already used above directly leads to the estimate

$$\begin{aligned}
&\Big| \frac{d^n}{d\xi^n} \left(\Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | \overline{((H_A + \hbar\omega(\xi))^{-1} \otimes I)_{|W}} | w_d \Big\rangle_{L^2(\mathbb{R}^{3N})} \right) \Big| \\
\leq & n! (\hbar c)^n \left(\frac{1}{\Delta_A} \right)^{n+1} \|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})} \|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})} \|w_d\|_{L^2(\mathbb{R}^{3N})} \\
\leq & n! (\hbar c)^n \left(\frac{1}{\Delta_A} \right)^{n+1} \|\mathbf{v}_A^\alpha\|_{L^2(\mathbb{R}^{3Z_A})} \|\mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3Z_B})} \|(\overline{H_A + H_B})^{-1}\| \|w_\infty\|_{L^2(\mathbb{R}^{3N})},
\end{aligned}$$

where we have set

$$\begin{aligned}
w_\infty := & \Big[\Big(\sum_{i_A} |\mathbf{x}_{i_A}|^{l_1} Y_{l_1,m_1}^*[(\theta,\varphi)_{\mathbf{x}_{i_A}}] \Big) \\
& \times \Big(\sum_{j_B} |\mathbf{x}_{j_B}|^{l-l_1} Y_{l-l_1,m-m_1}^*[(\theta,\varphi)_{\mathbf{x}_{j_B}}] \Big) (\Psi_A^0 \otimes \Psi_B^0) \Big] (\mathbf{x}_1, \ldots, \mathbf{x}_N),
\end{aligned}$$

which is in $L^2(\mathbb{R}^{3N})$ due to the exponential decay of Ψ_A^0 and Ψ_B^0. The corresponding estimate for the term (5.5.21) and its derivatives (with Δ_A replaced by Δ_B) is proven completely analogous, finishing the proof of Lemma 5.5.14. □

Proof of Lemma 5.5.13, continued. Using Lemma A.4.1 and Lemma 5.5.14 (in particular the vanishing of the functions $\frac{d^n}{d\xi^n}(\xi^m\varphi_{\alpha,\beta})$ at infinity for any n,m and at zero if $n<m$), we conclude

$$
\begin{aligned}
&4\pi c\int_0^\infty d\xi\Big[(\delta_{\alpha,\beta}-\hat R_\alpha\hat R_\beta)\big(\frac{1}{R}\xi^2\sin(\xi R)\big)+(\delta_{\alpha,\beta}-3\hat R_\alpha\hat R_\beta)\big(\frac{1}{R^2}\xi\cos(\xi R)-\frac{1}{R^3}\sin(\xi R)\big)\Big]\\
&\qquad\times\varphi_{\alpha,\beta}(\xi,d,\Lambda,2s,m,m_1,l_1)\\
=&4\pi c\Bigg[(\delta_{\alpha,\beta}-\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\Big(-\frac{1}{R^4}\lim_{\xi\to0}\frac{d^2}{d\xi^2}(\xi^2\varphi_{\alpha,\beta})+\frac{1}{R^5}\int_0^\infty\sin(\xi R)\frac{d^4}{d\xi^4}(\xi^2\varphi_{\alpha,\beta})d\xi\Big)\\
&\quad+(\delta_{\alpha,\beta}-3\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\Bigg(-\frac{1}{R^4}(\lim_{\xi\to0}\frac{d}{d\xi}(\xi\varphi_{\alpha,\beta}))+\frac{1}{R^5}\int_0^\infty\sin(\xi R)\frac{d^3}{d\xi^3}(\xi\varphi_{\alpha,\beta})d\xi\\
&\qquad\qquad-\frac{1}{R^4}(\lim_{\xi\to0}\varphi_{\alpha,\beta})+\frac{1}{R^5}\int_0^\infty\sin(\xi R)\frac{d^2}{d\xi^2}\varphi_{\alpha,\beta}d\xi\Bigg)\Bigg]\\
=&4\pi c\Bigg[-2\frac{1}{R^4}(2\delta_{\alpha,\beta}-4\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\,\varphi_{\alpha,\beta}(0,d,\Lambda,2s,m,m_1,l_1)\\
&\quad+\frac{1}{R^5}\Bigg((\delta_{\alpha,\beta}-\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\int_0^\infty\sin(\xi R)\frac{d^4}{d\xi^4}(\xi^2\varphi_{\alpha,\beta})d\xi\\
&\qquad\qquad+(\delta_{\alpha,\beta}-3\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\int_0^\infty\sin(\xi R)\left(\frac{d^3}{d\xi^3}(\xi\varphi_{\alpha,\beta})+\frac{d^2}{d\xi^2}\varphi_{\alpha,\beta}\right)d\xi\Bigg)\Bigg].
\end{aligned}
$$

This strategy has now produced enough powers of $1/R$ to carry out the limit occurring in the assertion. Consider

$$
\begin{aligned}
&R^kB_2(s,d,\mathbf{R})\\
=&\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde C_{2s,m,l_1,m_1}\hbar\,T_\Lambda^{2s,m}(\mathbf{R})\Lambda^{2s+1}\,R^{k-(2s+1)}\\
&\quad\times\int_{\mathbb{R}^3}d\mathbf{k}\,\omega(\mathbf{k})|\rho(\mathbf{k})|^2\left(\delta_{\alpha,\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2})\right)T_{\alpha,\beta}^{2s,l_1,m,m_1}(d,|\mathbf{k}|)\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\Bigg]\\
=&4\pi c\hbar\mathrm{Re}\Bigg[\sum_{m=-2s}^{2s}\sum_{l_1=0}^{2s}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde C_{2s,m,l_1,m_1}\,T_\Lambda^{2s,m}(\mathbf{R})\Lambda^{2s+1}\\
&\quad\times\Bigg[-2R^{(k-4)-(2s+1)}(2\delta_{\alpha,\beta}-4\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\,\varphi_{\alpha,\beta}(0,d,\Lambda,2s,m,m_1,l_1)\\
&\qquad+R^{(k-5)-(2s+1)}\Bigg((\delta_{\alpha,\beta}-\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\int_0^\infty\sin(\xi R)\frac{d^4}{d\xi^4}(\xi^2\varphi_{\alpha,\beta})d\xi\\
&\qquad\qquad+(\delta_{\alpha,\beta}-3\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\int_0^\infty\sin(\xi R)\Big(\frac{d^3}{d\xi^3}(\xi\varphi_{\alpha,\beta})+\frac{d^2}{d\xi^2}\varphi_{\alpha,\beta}\Big)d\xi\Bigg)\Bigg]\Bigg].
\end{aligned}
$$

By Lemma 5.1.2, $\lim_{R\to\infty}T_\Lambda^{2s,m}(\mathbf{R})$ exists, so $T_\Lambda^{2s,m}(\mathbf{R})$ is bounded with respect to R. Now using that $\frac{d^2}{d\xi^2}\varphi_{\alpha,\beta}$, $\frac{d^3}{d\xi^3}(\xi\varphi_{\alpha,\beta})$ and $\frac{d^4}{d\xi^4}(\xi^2\varphi_{\alpha,\beta})$ are in $L^1((0,\infty))$ uniformly in d by Lemma

5.5.14 and employing the (d-independent) bounds on $\varphi_{\alpha,\beta}(0,d,\Lambda,2s,m,m_1,l_1)$ derived in its proof, we arrive at

$$|R^k B_2(s,d,\mathbf{R})| \leq C(\Lambda)\Big(R^{(k-5)-(2s+1)} + R^{(k-4)-(2s+1)}\Big),$$

with a constant $C(\Lambda)$ which is independent of R and d but depends on the ultraviolet-cutoff scale Λ. Now if $k<9$ and $s\geq 2$, the right-hand side contains negative powers of R, proving the assertion. □

$1/R^6$- and $1/R^7$- contributions from mixed terms

Recall that by the decomposition (5.5.1) and the definition of $M^L_{IN,ERR}(\mathbf{R},d)$ see (5.5.3)), we have

$$\begin{aligned}&M_A(\mathbf{R},\sigma,d)+M_B(\mathbf{R},\sigma,d) = A_{IN}(\mathbf{R},\sigma,\mathbf{d})+B_{IN}(\mathbf{R},\sigma,d)+M_{OUT}(\mathbf{R},\sigma,d)\\ =&\sum_{l=2}^{N}\Big(\tilde{A}_l(\mathbf{R},\sigma,d)+\tilde{B}_l(\mathbf{R},\sigma,d)\Big)+M^L_{IN,ERR}(\mathbf{R},d)+M_{OUT}(\mathbf{R},\sigma,d).\end{aligned}$$

In view of the preceding results, the only of these terms which are left to discuss are contained in $\tilde{B}_2(\mathbf{R},\sigma,d)$ (see (5.5.2) for its definition). By Lemma 5.5.5,

$$\begin{aligned}&\tilde{B}_2(\mathbf{R},\sigma,d)\\ =&\frac{2}{\hbar^2}\mathrm{Re}\Bigg[\int_{\Omega_\sigma} d\mathbf{k}|C(\mathbf{k})|^2\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\\ &\times\Bigg[-2\hbar\omega(\mathbf{k})\Big\langle(\overline{H_A+H_B})^{-1}[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]|\tilde{Q}_2\chi_{\Omega_d}(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\ &+(\hbar\omega(\mathbf{k}))^2\Big\langle[(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})(H_A+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_A]\otimes\Psi^0_B\big|\\ &\qquad\big|\tilde{Q}_2\chi_{\Omega_d}\left(\Psi^0_A\otimes[(H_B+\hbar\omega(\mathbf{k}))^{-1}\mathbf{v}_B]\right)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\ &+(\hbar\omega(\mathbf{k}))^2\Big\langle(\overline{H_A+H_B})^{-1}\left((H_A+\hbar\omega(\mathbf{k}))^{-1}\otimes I\right)[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\big|\\ &\qquad\big|\tilde{Q}_2\chi_{\Omega_d}(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\ &+(\hbar\omega(\mathbf{k}))^2\Big\langle(\overline{H_A+H_B})^{-1}\left(I\otimes(H_B+\hbar\omega(\mathbf{k}))^{-1}\right)[\mathbf{v}_A(1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B]\big|\\ &\qquad\big|\tilde{Q}_2\chi_{\Omega_d}(\Psi^0_A\otimes\Psi^0_B)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Bigg]\Bigg],\end{aligned}\tag{5.5.28}$$

with $\tilde{Q}_2$ as defined there. By (5.5.24),

$$\tilde{B}_2(\mathbf{R},\sigma,d) = B_1(1,d,\mathbf{R})+B_2(1,d,\mathbf{R})+B_3(\sigma,1,d,\mathbf{R})+B_4(\sigma,1,d,\mathbf{R}),$$

where the terms on the right-hand side are defined in 5.5.6. Recalling that $B_4(\sigma,1,d,\mathbf{R})$ was already absorbed into the definition of $M^N_{IR}(\mathbf{R},\sigma,d)$ (see (5.5.25)), we are left with discussing

$$M_6(\mathbf{R},\sigma,d) := B_1(1,d,\mathbf{R})+B_3(\sigma,1,d,\mathbf{R})$$

and

$$M_7(\mathbf{R}, d) := B_2(1, d, \mathbf{R}). \tag{5.5.29}$$

The claims (5.4.1) and (5.4.2) about $M_6(\mathbf{R}, \sigma, d)$ in Theorem 5.4.1 follow from the next lemma.

Lemma 5.5.15. *Assume the hypotheses of Theorem 5.4.1. Then for any* $\mathbf{R} \in \mathbb{R}^3$ *and* $0 < d \leq R/4$,

$$\lim_{\sigma \to 0} M_6(\mathbf{R}, \sigma, d)$$

exists. Furthermore,

$$\lim_{R \to \infty} \left(R^k \lim_{\sigma \to 0} M_6(\mathbf{R}, \sigma, R^{1/2}) \right) = 0$$

for $k < 6$, *and*

$$\lim_{R \to \infty} \left(R^6 \lim_{\sigma \to 0} M_6(\mathbf{R}, \sigma, R^{1/2}) \right) = \frac{1}{3(2\pi)^2} L(\infty),$$

with $L(\infty)$ *as defined in 5.5.1.*

Proof. Using $\mathbb{R}^3 \setminus B_{\sigma/c}(0) = \Omega_\sigma$, we find

$$M_6(\mathbf{R}, \sigma, d) = (5.5.28).$$

Recalling the proof of Lemma 5.5.5, we can first replace $\tilde{Q}_2$ by Q_2 (by re-adding the vanishing terms from the multipole expansion) and the latter by its Fourier representation

$$\tilde{Q} = \sum_{i_A, j_B} \int d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}} (\mathbf{x}_{i_A} \cdot \mathbf{k}) \, (\mathbf{k} \cdot \mathbf{x}_{j_B})$$

(see Section 5.1), yielding

$$\begin{aligned} &M_6(\mathbf{R}, \sigma, d) \\ =& -2\mathrm{Re} \Bigg[\int_{\Omega_\sigma \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-i(\mathbf{k}_1 + \mathbf{k}_2)\cdot\mathbf{R}} \\ &\qquad \left\langle \mathbf{v}_A (1 - \hat{\mathbf{k}_1} \otimes \hat{\mathbf{k}_1}) \mathbf{v}_B \middle| (\overline{H_A + H_B})^{-1} \chi_{\Omega_d} \middle| (\mathbf{v}_A \cdot \mathbf{k}_2)(\mathbf{v}_B \cdot \mathbf{k}_2) \right\rangle_{L^2(\mathbb{R}^{3N})} \Bigg]. \end{aligned}$$

Noting that the operator $(\overline{H_A + H_B})^{-1} \chi_{\Omega_d}$ is rotation-invariant in the sense of Lemma 4.2.17, we obtain

$$\begin{aligned} &M_6(\mathbf{R}, \sigma, d) \\ =& -\frac{2}{9} \mathrm{Re} \int_{\Omega_\sigma \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-i(\mathbf{k}_1 + \mathbf{k}_2)\cdot\mathbf{R}} \\ &\qquad \times \underbrace{\mathrm{tr}[(1 - \hat{\mathbf{k}_1} \otimes \hat{\mathbf{k}_1})(\hat{\mathbf{k}_2} \cdot \hat{\mathbf{k}_2})]}_{=1-(\hat{\mathbf{k}_1}\cdot\hat{\mathbf{k}_2})^2} L(d), \end{aligned}$$

where we have defined

$$L(d) := \sum_{\alpha,\beta=1}^{3} \left\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \middle| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \middle| \chi_{\Omega_d} (\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \right\rangle_{L^2(\mathbb{R}^{3N})}. \tag{5.5.30}$$

Since the integrand is of the form $f(\mathbf{k}_1, \mathbf{k}_2)\exp(-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2) \cdot \mathbf{R})$, with f real and satisfying $f(-\mathbf{k}_1, -\mathbf{k}_2) = f(\mathbf{k}_1, \mathbf{k}_2)$, the real part can be dropped. (Note that the domain of integration $\Omega_\sigma \times \mathbb{R}^3$ is invariant under the change of coordinates $(\mathbf{k}_1, \mathbf{k}_2) \to (-\mathbf{k}_1, -\mathbf{k}_2)$). Thus

$$\begin{aligned} & M_6(\mathbf{R}, \sigma, d) \\ =& -\frac{2}{9} L(d) \int_{\Omega_\sigma \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 (1 - (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-i(\mathbf{k}_1 + \mathbf{k}_2) \cdot \mathbf{R}}. \end{aligned}$$

Noting that $|1 - (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2| \leq 2$, recalling $\rho \in \mathcal{S}(\mathbb{R}^3)$ and using dominated convergence, we find

$$\begin{aligned} & \lim_{\sigma \to 0} M_6(\mathbf{R}, \sigma, d) \\ =& -\frac{2}{9} L(d) \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 (1 - (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-i(\mathbf{k}_1 + \mathbf{k}_2) \cdot \mathbf{R}} \\ =& -\frac{2}{9} L(d) \left(\int_{\mathbb{R}^3} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \mathrm{e}^{-i\mathbf{k}_1 \cdot \mathbf{R}} \right) \left(\int_{\mathbb{R}^3} d\mathbf{k}_2 |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-i\mathbf{k}_2 \cdot \mathbf{R}} \right) \\ & + \frac{2}{9} L(d) \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 \frac{\mathbf{k}_1 \cdot \mathbf{k}_2}{|\mathbf{k}_1|^2 |\mathbf{k}_2|^2} \mathrm{e}^{-i(\mathbf{k}_1 + \mathbf{k}_2) \cdot \mathbf{R}} \\ =:& T_1(\mathbf{R}, d) + T_2(\mathbf{R}, d). \end{aligned}$$

Again by dominated convergence,

$$\begin{aligned} & \lim_{d \to \infty} L(d) \\ =& \lim_{d \to \infty} \left(\sum_{\alpha,\beta=1}^{3} \langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \middle| (\overline{H_A + H_B})^{-1} (\chi_{\Omega_d} \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \rangle_{L^2(\mathbb{R}^{3N})} \right) \\ =& \lim_{d \to \infty} \left(\sum_{\alpha,\beta=1}^{3} \langle (\overline{H_A + H_B})^{-1} (\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \middle| (\chi_{\Omega_d} \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \rangle_{L^2(\mathbb{R}^{3N})} \right) \\ =& \sum_{\alpha,\beta=1}^{3} \langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | (\overline{H_A + H_B})^{-1} | \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \rangle_{L^2(\mathbb{R}^{3N})} \\ =& L(\infty). \end{aligned}$$

In particular, $L(d)$ is uniformly bounded with respect to d. Furthermore,

$$T_1(\mathbf{R}, d) = -\frac{2}{9} L(d) (2\pi)^3 \left(\widehat{|\rho|^2}(\mathbf{R}) \right)^2,$$

which has rapid decay with respect to R since ρ is a Schwartz function. Thus

$$\lim_{R \to \infty} (R^k \, T_1(\mathbf{R}, R^{1/2})) = 0$$

for any $k \geq 0$. Next we investigate the integral

$$\begin{aligned} I(\mathbf{R}) :=& \int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 \frac{\mathbf{k}_1\cdot\mathbf{k}_2}{|\mathbf{k}_1|^2|\mathbf{k}_2|^2} \mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\ =& \int_{\mathbb{R}^3} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}} \frac{1}{|\mathbf{k}_1|^2} \int_{\mathbb{R}^3} d\mathbf{k}_2 |\rho(\mathbf{k}_2)|^2 \frac{1}{|\mathbf{k}_2|^2} (\mathbf{k}_1\cdot\mathbf{k}_2)^2 \mathrm{e}^{-i\mathbf{k}_2\cdot\mathbf{R}} \end{aligned}$$

occurring in $T_2(\mathbf{R}, d)$. By Lemma 5.5.10, if $R > 2/\Lambda$,

$$\begin{aligned} &\int_{\mathbb{R}^3} d\mathbf{k}_2 |\rho(\mathbf{k}_2)|^2 \frac{1}{|\mathbf{k}_2|^2} (\mathbf{k}_1\cdot\mathbf{k}_2)^2 \mathrm{e}^{-i\mathbf{k}_2\cdot\mathbf{R}} \\ =& \frac{1}{(2\pi)^{3/2}} \frac{\Lambda^3}{R^3} \Big\langle \widehat{\frac{1}{|\mathbf{k}_2|^2}(\mathbf{k}_1\cdot\mathbf{k}_2)^2}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \Big\rangle \\ =& \frac{1}{(2\pi)^{3/2}} \frac{\Lambda^3}{R^3} \Big\langle -(\mathbf{k}_1\cdot\nabla_{\mathbf{x}_2})^2 \widehat{(\frac{1}{|\mathbf{k}_2|^2})}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \Big\rangle \\ =& \frac{1}{(2\pi)^{3/2}} \frac{\Lambda^3}{R^3} \frac{(2\pi)^{1/2}}{2} \Big\langle \left(\frac{|\mathbf{k}_1|^2}{|\mathbf{x}_2|^3} - 3\frac{(\mathbf{k}_1\cdot\mathbf{x}_2)^2}{|\mathbf{x}_2|^6} \right), \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \Big\rangle. \end{aligned}$$

Thus $I(\mathbf{R})$ splits into the two terms

$$I(\mathbf{R}) = \frac{3}{4\pi}\frac{\Lambda^3}{R^3} \underbrace{\left(\int_{\mathbb{R}^3} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \frac{|\mathbf{k}_1|^2}{|\mathbf{k}_1|^2} \mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}} \right)}_{=(2\pi)^{2/1}\widehat{|\rho|^2}(\mathbf{R})} \Big\langle \frac{1}{|\mathbf{x}_2|^3}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \Big\rangle \tag{5.5.31}$$

$$- \frac{3}{4\pi}\frac{\Lambda^3}{R^3} \sum_{\alpha,\beta=1}^{3} \left(\int_{\mathbb{R}^3} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \frac{\mathbf{k}_1^\alpha \mathbf{k}_1^\beta}{|\mathbf{k}_1|^2} \mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}} \right) \Big\langle \frac{\mathbf{x}_2^\alpha \mathbf{x}_2^\beta}{|\mathbf{x}_2|^5}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \Big\rangle. \tag{5.5.32}$$

By Lemma 5.5.8, $\langle \frac{1}{|\mathbf{x}_2|^3}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \rangle$ converges to $1/\Lambda^3$ as $R\to\infty$. In particular, this expression is bounded in R, so the rapid decay of $\widehat{|\rho|^2}$ implies that $R^k(5.5.31) \to 0$ as $R\to\infty$ for any $k \geq 0$. As above, by Lemma 5.5.10, we have

$$\begin{aligned} &\int_{\mathbb{R}^3} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \frac{\mathbf{k}_1^\alpha \mathbf{k}_1^\beta}{|\mathbf{k}_1|^2} \mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}} \\ =& \frac{\Lambda^3}{R^3}\frac{1}{4\pi} \Big\langle \frac{1}{|\mathbf{x}_1|^3}(\delta_{\alpha,\beta} - 3\hat{\mathbf{x}}_1^\alpha\cdot\hat{\mathbf{x}}_1^\beta), \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)] \Big\rangle \end{aligned}$$

if $R > 2/\Lambda$, and Lemma 5.5.8 implies

$$\begin{aligned} R^6(5.5.32) \to& -\frac{3}{(4\pi)^2}\frac{\Lambda^6}{\Lambda^6} \left(\sum_{\alpha,\beta=1}^{3} (\delta_{\alpha,\beta} - 3\hat{\mathbf{R}}_\alpha\cdot\hat{\mathbf{R}}_\beta)(\hat{\mathbf{R}}_\alpha\cdot\hat{\mathbf{R}}_\beta) \right) \\ =& -\frac{3}{(4\pi)^2}\frac{\Lambda^6}{\Lambda^6} \mathrm{Tr}[(1 - 3\hat{\mathbf{R}}\otimes\hat{\mathbf{R}})(\hat{\mathbf{R}}\otimes\hat{\mathbf{R}})] \\ =& \frac{6}{(4\pi)^2} \end{aligned}$$

as $R \to \infty$, and $R^k(5.5.32) \to 0$ for $k < 6$. This immediately leads to

$$\lim_{R\to\infty} R^k(T_2(\mathbf{R}, R^{1/2})) = \lim_{R\to\infty} \left(\frac{2}{9} L(R^{1/2}) R^k I(\mathbf{R})\right) = 0$$

for any $k < 6$, and

$$\lim_{R\to\infty} \left(R^6(T_2(\mathbf{R}, R^{1/2}))\right) = \lim_{R\to\infty} \left(\frac{2}{9} L(R^{1/2}) R^6 I(\mathbf{R})\right) = \frac{2}{9} L(\infty) \frac{6}{(4\pi)^2},$$

finishing the proof. □

The claim about $M_7(\mathbf{R}, d)$ in Theorem 5.4.1 follows from the next lemma.

Lemma 5.5.16. *Assume the hypotheses of Theorem 5.4.1. Then*

$$\lim_{R\to\infty} \left(R^k M_7(\mathbf{R}, d)\right) = 0$$

uniformly in $d > 0$ for any $k < 7$, and

$$\lim_{R\to\infty} \left(R^7 M_7(\mathbf{R}, R^{1/2})\right) = -\frac{32}{9} \frac{\hbar c}{(2\pi)^3} \alpha_E^A(0)\, \alpha_E^B(0).$$

Proof. Recall the definition (5.5.29). In the proof of Lemma 5.5.13, it was shown that

$$\begin{aligned}
&R^k B_2(1, d, \mathbf{R}) \\
=&4\pi c\hbar \,\mathrm{Re}\Bigg[\sum_{m=-2}^{2} \sum_{l_1=0}^{2} \sum_{m_1=-l_1}^{l_1} \sum_{\alpha,\beta=1}^{3} \tilde{C}_{2,m,l_1,m_1}\, T_\Lambda^{2,m}(\mathbf{R}) \Lambda^{2+1} \\
&\quad \times \Bigg[-4R^{(k-7)}(\delta_{\alpha,\beta} - 2\hat{\mathbf{R}}_\alpha \hat{\mathbf{R}}_\beta)\, \varphi_{\alpha,\beta}(0, d, \Lambda, 2, m, m_1, l_1) \\
&\qquad + R^{(k-8)} \Bigg((\delta_{\alpha,\beta} - \hat{\mathbf{R}}_\alpha \hat{\mathbf{R}}_\beta) \int_0^\infty \sin(\xi R) \frac{d^4}{d\xi^4} (\xi^2 \varphi_{\alpha,\beta}) d\xi \\
&\qquad\quad + (\delta_{\alpha,\beta} - 3\hat{\mathbf{R}}_\alpha \hat{\mathbf{R}}_\beta) \int_0^\infty \sin(\xi R) \left(\frac{d^3}{d\xi^3} (\xi \varphi_{\alpha,\beta}) + \frac{d^2}{d\xi^2} \varphi_{\alpha,\beta} \right) d\xi \Bigg) \Bigg] \Bigg].
\end{aligned}$$

Furthermore, it was established that the term

$$\begin{aligned}
&(\delta_{\alpha,\beta} - \hat{\mathbf{R}}_\alpha \hat{\mathbf{R}}_\beta) \int_0^\infty \sin(\xi R) \frac{d^4}{d\xi^4} (\xi^2 \varphi_{\alpha,\beta}) d\xi \\
+&(\delta_{\alpha,\beta} - 3\hat{\mathbf{R}}_\alpha \hat{\mathbf{R}}_\beta) \int_0^\infty \sin(\xi R) \left(\frac{d^3}{d\xi^3} (\xi \varphi_{\alpha,\beta}) + \frac{d^2}{d\xi^2} \varphi_{\alpha,\beta} \right) d\xi
\end{aligned}$$

can be bounded independently of $\mathbf{R}$ and uniformly in d by a constant which depends on the ultraviolet-cutoff Λ, and that

$$\varphi_{\alpha,\beta}(0, d, \Lambda, 2, m, m_1, l_1) = |\rho(0)|^2 T_{\alpha,\beta}^{2,l_1,m,m_1}(d, 0),$$

which is independent of both Λ and $\mathbf{R}$ and which is bounded uniformly in d. Since $T_\Lambda^{2,m}(\mathbf{R})$ converges to $1/(\Lambda)^3 Y_{2,m}([\theta,\varphi]_{\hat{\mathbf{R}}})$ as $R\to\infty$ by Lemma 5.1.2, it is in particular bounded in R. Combining these facts, we conclude that

$$\lim_{R\to\infty}\Big(R^k B_2(1,R^{1/2},\mathbf{R})\Big)=0$$

for $k<7$, proving the first assertion of the lemma.
By dominated convergence, $T_{\alpha,\beta}^{2,l_1,m,m_1}(d,0)$ has a limit as $d\to\infty$, which we denote by $T_{\alpha,\beta}^{2,l_1,m,m_1}(\infty,0)$ and identify as being obtained from $T_{\alpha,\beta}^{2,l_1,m,m_1}(d,0)$ by omitting the characteristic function χ_{Ω_d}. Recalling $|\rho(0)|^2=1/(2\pi)^3$, we thus conclude that

$$\begin{aligned}
&\lim_{R\to\infty}\Big(R^7 B_2(1,R^{1/2},\mathbf{R})\Big)\\
&=-\frac{16\pi c\hbar}{(2\pi)^3}\mathrm{Re}\Bigg[\sum_{m=-2}^{2}\sum_{l_1=0}^{2}\sum_{m_1=-l_1}^{l_1}\sum_{\alpha,\beta=1}^{3}\tilde{C}_{2,m,l_1,m_1}\frac{\Lambda^3}{\Lambda^3}Y_{2,m}([\theta,\varphi]_{\hat{\mathbf{R}}})\\
&\qquad\qquad\times(\delta_{\alpha,\beta}-2\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\,T_{\alpha,\beta}^{2,l_1,m,m_1}(\infty,0)\Bigg]\\
&=-\frac{16\pi c\hbar}{(2\pi)^3}\mathrm{Re}\Bigg[\sum_{\alpha,\beta=1}^{3}(\delta_{\alpha,\beta}-2\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\\
&\quad\times\Bigg[\Big\langle((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A^\alpha)\otimes\Psi_B^0|W|\Psi_A^0\otimes((H_{B|\{\Psi_B^0\}^\perp})^{-1}\mathbf{v}_B^\beta)\Big\rangle_{L^2(\mathbb{R}^{3N})} &(5.5.33)\\
&\qquad+\Big\langle((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A^\alpha)\otimes\mathbf{v}_B^\beta|(\overline{H_A+H_B})^{-1}|W(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})} &(5.5.34)\\
&\qquad+\Big\langle\mathbf{v}_A^\alpha\otimes((H_{B|\{\Psi_B^0\}^\perp})^{-1}\mathbf{v}_B^\beta)|(\overline{H_A+H_B})^{-1}|W(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{L^2(\mathbb{R}^{3N})}\Bigg]\Bigg], &(5.5.35)
\end{aligned}$$

where we have set

$$\begin{aligned}
W:=\sum_{i_A,j_B}\sum_{m=-2}^{2}\sum_{l_1=0}^{2}\sum_{m_1=-l_1}^{l_1}&\tilde{C}_{2,m,l_1,m_1}\,Y_{2,m}([\theta,\varphi]_{\hat{\mathbf{R}}})\,|\mathbf{x}_{i_A}|^{l_1}\,|\mathbf{x}_{j_B}|^{2-l_1}\\
&\times Y^*_{l_1,m_1}[(\theta,\varphi)_{\hat{\mathbf{x}}_{i_A}}]\,Y^*_{2-l_1,m-m_1}[(\theta,\varphi)_{\hat{\mathbf{x}}_{i_A}}].
\end{aligned}$$

Reversing the steps taken in Section 5.1 (i.e. addition theorem for Legendre polynomials and results about spherical harmonics of translates), we obtain

$$\begin{aligned}
W=&\frac{1}{4\pi}\sum_{i_A,j_B}|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}|^2\,P_2(\hat{\mathbf{R}}\cdot(\widehat{\mathbf{x}_{i_A}-\mathbf{x}}_{j_B}))\\
=&\frac{1}{4\pi}\sum_{i_A,j_B}\mathbf{x}_{i_A}(1-3\hat{\mathbf{R}}\otimes\hat{\mathbf{R}})\mathbf{x}_{j_B}-\frac{1}{2}(|\mathbf{x}_{i_A}|^2+|\mathbf{x}_{j_B}|^2)+\frac{3}{2}((\mathbf{x}_{i_A}\cdot\hat{\mathbf{R}})^2+(\mathbf{x}_{j_B}\cdot\hat{\mathbf{R}})^2),
\end{aligned}$$

where we have used $P_2(x)=1/2(3x^2-1)$. The multiplication operators

$$-\frac{1}{2}(|\mathbf{x}_{i_A}|^2+|\mathbf{x}_{j_B}|^2)+\frac{3}{2}((\mathbf{x}_{i_A}\cdot\hat{\mathbf{R}})^2+(\mathbf{x}_{j_B}\cdot\hat{\mathbf{R}})^2)$$

conserve parity with respect to atom A and B separately, so the corresponding terms in (5.5.33) through (5.5.35) vanish upon integration, and we are left with

$$
\begin{aligned}
&\lim_{R\to\infty}\Big(R^7 B_2(1,R^{1/2},\mathbf{R})\Big)\\
=&-\frac{16\pi c\hbar}{(2\pi)^3(4\pi)}\mathrm{Re}\Bigg[\sum_{\alpha,\beta,\gamma,\delta}(\delta_{\alpha,\beta}-2\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\,(\delta_{\gamma,\delta}-3\hat{\mathbf{R}}_\gamma\hat{\mathbf{R}}_\delta)\\
&\times\Bigg[\Big\langle((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A^\alpha)\otimes\mathbf{v}_B^\delta|\mathbf{v}_A^\gamma\otimes((H_{B|\{\Psi_B^0\}^\perp})^{-1}\mathbf{v}_B^\beta)\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\Big\langle((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A^\alpha)\otimes\mathbf{v}_B^\beta|(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\gamma\otimes\mathbf{v}_B^\delta\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&\quad+\Big\langle\mathbf{v}_A^\alpha\otimes((H_{B|\{\Psi_B^0\}^\perp})^{-1}\mathbf{v}_B^\beta)|(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\gamma\otimes\mathbf{v}_B^\delta\Big\rangle_{L^2(\mathbb{R}^{3N})}\Bigg]\Bigg]\\
=&-\frac{16\pi c\hbar}{(2\pi)^3 4\pi}\mathrm{Re}\Bigg[\sum_{\alpha,\beta,\gamma,\delta}(\delta_{\alpha,\beta}-2\hat{\mathbf{R}}_\alpha\hat{\mathbf{R}}_\beta)\,(\delta_{\gamma,\delta}-3\hat{\mathbf{R}}_\gamma\hat{\mathbf{R}}_\delta)\\
&\times\Bigg[\Big\langle\mathbf{v}_A^\alpha)|((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A^\gamma\Big\rangle\Big\langle\mathbf{v}_B^\delta|((H_{B|\{\Psi_B^0\}^\perp})^{-1}|\mathbf{v}_B^\beta)\Big\rangle\\
&\quad+\Big\langle\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta|(\overline{(H_{A|\{\Psi_A^0\}^\perp})^{-1}\otimes I})(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\gamma\otimes\mathbf{v}_B^\delta\Big\rangle\\
&\quad+\Big\langle\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(\overline{I\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}})(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\gamma\otimes\mathbf{v}_B^\delta\Big\rangle\Bigg]\Bigg].
\end{aligned}
$$

Exploiting rotational invariance (Lemma 4.2.17), applying Lemma 4.2.18 iv) (with $\mathbf{k}=0$) and using the definition of the dynamic polarizabilities $\alpha_E^{A,B}(\mathbf{k})$ yields

$$
\begin{aligned}
&\Big\langle\mathbf{v}_A^\alpha|((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A^\gamma\Big\rangle\Big\langle\mathbf{v}_B^\delta|((H_{B|\{\Psi_B^0\}^\perp})^{-1}|\mathbf{v}_B^\beta)\Big\rangle\\
&+\Big\langle\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta|(\overline{(H_{A|\{\Psi_A^0\}^\perp})^{-1}\otimes I})(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\gamma\otimes\mathbf{v}_B^\delta\Big\rangle\\
&+\Big\langle\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)|(\overline{I\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}})(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^\gamma\otimes\mathbf{v}_B^\delta\Big\rangle\\
=&\frac{1}{9}\delta_{\alpha,\gamma}\,\delta_{\beta,\delta}\Bigg[\underbrace{\Big\langle\mathbf{v}_A|((H_{A|\{\Psi_A^0\}^\perp})^{-1}\mathbf{v}_A\Big\rangle}_{=\alpha_E^A(0)}\underbrace{\Big\langle\mathbf{v}_B|((H_{B|\{\Psi_B^0\}^\perp})^{-1}|\mathbf{v}_B\Big\rangle}_{=\alpha_E^B(0)}\\
&\qquad+\sum_{i,j=1}^3\Big\langle\mathbf{v}_A^i\otimes\mathbf{v}_B^j|(\overline{(H_{A|\{\Psi_A^0\}^\perp})^{-1}\otimes I})(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^i\otimes\mathbf{v}_B^j\Big\rangle\\
&\qquad+\sum_{i,j=1}^3\Big\langle\mathbf{v}_A^i\otimes\mathbf{v}_B^j)|(\overline{I\otimes(H_{B|\{\Psi_B^0\}^\perp})^{-1}})(\overline{H_A+H_B})^{-1}|\mathbf{v}_A^i\otimes\mathbf{v}_B^j\Big\rangle\Bigg]\\
=&\frac{2}{9}\delta_{\alpha,\gamma}\,\delta_{\beta,\delta}\,\alpha_E^A(0)\,\alpha_E^B(0),
\end{aligned}
$$

which finally leads to

$$\begin{aligned}
&\lim_{R\to\infty}\left(R^7 B_2(1,R^{1/2},\mathbf{R})\right)\\
&=-\frac{32\pi c\hbar}{9(2\pi)^3 4\pi}\alpha_E^A(0)\,\alpha_E^B(0)\,\underbrace{\mathrm{Tr}[(1-2\hat{\mathbf{R}}\otimes\hat{\mathbf{R}})\,(1-3\hat{\mathbf{R}}\otimes\hat{\mathbf{R}})]}_{=4},
\end{aligned}$$

proving the second claim. □

Chapter 6

Analysis of pure field terms

In this chapter we we analyze the terms $F_7(\mathbf{R},\sigma)$ and $F_8(\mathbf{R},\sigma)$ in $V_4^\sigma(\Lambda,\mathbf{R})$, which originate purely from the radiation field (see (3.0.16)), and establish the (asymptotic) cancellation of the $1/|\mathbf{R}|^6$-contributions to the interaction potential $V(\Lambda,\mathbf{R})$.
In Section 6.1 we derive error estimates which compare $F_7(\mathbf{R},\sigma)$ and $F_8(\mathbf{R},\sigma)$ to their respective ($\sigma\to 0$)-limits $F_7(\mathbf{R})$ and $F_8(\mathbf{R})$. The latter turn out to exist since the integrands are sufficiently regular at the origin.
In Sections 6.2 and 6.3, which are joint work with Gero Friesecke, we present a method for the asymptotic analysis of a class of singular, formally divergent Fourier integrals, and verify that the integrals encountered in $F_7(\mathbf{R})$ belong to this class. The latter boils down to a careful investigation of regularity properties of the dynamic polarizabilities $\alpha_E^A(\mathbf{k})$ and $\alpha_E^B(\mathbf{k})$. The relevant distributional Fourier transforms are calculated in Section 6.4.
Subsequently, in Section 6.5, we apply this method to show that the lowest power of $1/|\mathbf{R}|$ that enters in $F_7(\mathbf{R})$ is $1/|\mathbf{R}|^7$, and calculate the corresponding (asymptotic) coefficient explicitly. Furthermore, using standard decay estimates for oscillatory integrals involving smooth functions, we show that

$$\lim_{R\to\infty}\Big(R^k F_8(\mathbf{R})\Big)=0$$

for any $k<8$, i.e. that $F_8(\mathbf{R})$ can only contribute to $V(\Lambda,\mathbf{R})$ at orders $1/R^k$ for $k\geq 8$.
In Section 6.6 we show that the $1/|\mathbf{R}|^6$-contributions to $V(\Lambda,\mathbf{R})$ vanish asymptotically, in the sense that they decay faster than any inverse power of $|\mathbf{R}|$. As mentioned in the introduction, this crucially exploits the relation (1.0.10), which is a consequence of using a smeared Coulomb potential for the interaction of the electrons. As an aside, we discuss in Section 6.6.1 why we expect the mechanism of cancellation to break down if a proper Coulomb potential is used.

6.1 Subtraction of infrared-regularized terms and derivation of error estimates

Recall the definition

$$
\begin{aligned}
&F_7(\mathbf{R},\sigma)\\
&= -\frac{1}{9\hbar^4}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |C(\mathbf{k}_1)|^2 |C(\mathbf{k}_2)|^2 (1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Bigg[(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{-4\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2-6\hbar^3\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\qquad+\left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)+\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg]\\
&= -\frac{1}{36\hbar^2}\int_{\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2}{\omega(\mathbf{k}_1)}\frac{|\rho(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_2)} (1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Bigg[(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{-4\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2-6\hbar^3\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\qquad+\left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)+\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg]
\end{aligned}
$$

from (3.0.1). The following result compares this expression, which still depends on the infrared-cutoff σ, to the term which results from taking the limit $\sigma\to 0$, namely to

$$
\begin{aligned}
&F_7(\mathbf{R})\\
&:= -\frac{1}{36\hbar^2}\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2}{\omega(\mathbf{k}_1)}\frac{|\rho(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_2)} (1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Bigg[(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{-4\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2-6\hbar^3\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
&\qquad+\left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)+\frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg].
\end{aligned}
\tag{6.1.1}
$$

Note that this integral converges since $|\rho(\cdot_1)|^2\,|\rho(\cdot_2)|^2\in\mathcal{S}(\mathbb{R}^6)$ and the remaining integrand is in $L^1_{loc}(\mathbb{R}^6)$. The latter fact will become evident from the estimates on $\alpha_E^{A,B}(\mathbf{k})$ derived in the proof of the following lemma.

Lemma 6.1.1. *Assume the hypotheses of Theorem 3.0.6. Then there exist constants C_7 and $C_i>0$, $i=2,\dots,4$, independent of σ and $\mathbf{R}$ (but depending on Λ via properties of $H_{A,B}$), such that*

$$
\begin{aligned}
&\Big|F_7(\mathbf{R},\sigma)-F_7(\mathbf{R})\Big|\\
&\leq\left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)\left(\sum_{i=2}^4\sigma^i\Lambda^{7-i}C_i\right)+\left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)^2\sigma^7 C_7.
\end{aligned}
$$

Proof. First note that

$$
\begin{aligned}
&F_7(\mathbf{R},\sigma) - F_7(\mathbf{R}) \\
=&\frac{\hbar^2}{36\hbar^4}\left(\int_{\Omega_\sigma\times B_{\sigma/c}(0)} + \int_{B_{\sigma/c}(0)\times\Omega_\sigma} + \int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)}\right) d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2}{\omega(\mathbf{k}_1)}\frac{|\rho(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_2)} \\
&\times (1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} \\
&\times \Bigg[(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{-4\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2 - 6\hbar^3\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right) \\
&\quad + \left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2) + \alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(-\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2) + \frac{\hbar^3\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\Bigg].
\end{aligned}
$$

The standard identity $\|(A-\lambda)^{-1}\| = 1/(\mathrm{dist}(\lambda,\mathrm{spec}(A)))$ for self-adjoint operators yields the estimates

$$
|\alpha_E^A(\mathbf{k}_i)\alpha_E^B(\mathbf{k}_j)| \leq \underbrace{\frac{1}{\Delta_A}\frac{1}{\Delta_B}(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|^2)(\sum_{\beta=1}^{3}\|\mathbf{v}_B^\beta\|^2)}_{=:C_{AB}},
$$

i.e. these quantities are bounded uniformly in $\mathbf{k}_1$ and $\mathbf{k}_2$. Note that $|1+(\hat{\mathbf{k}_1}\cdot\hat{\mathbf{k}}_2))^2| \leq 2$, so the only remaining terms in the integral which couple $\mathbf{k}_1$ and $\mathbf{k}_2$ are those containing $1/(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))$. We will use an individual estimate for these terms in each of the three integrals. On $\Omega_\sigma \times B_{\sigma/c}(0)$ we have the estimate

$$
\frac{1}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)} \leq \frac{1}{\sigma/c+\omega(\mathbf{k}_2)} \leq \frac{1}{\omega(\mathbf{k}_2)},
$$

and analogously $1/(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)) \leq 1/\omega(\mathbf{k}_1)$ on $B_{\sigma/c}(0)\times\Omega_\sigma$. For the integral over $B_{\sigma/c}(0)\times B_{\sigma/c}(0)$ we use the estimate $1/(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)) \leq 1/(2\sqrt{\omega(\mathbf{k}_1)}\sqrt{\omega(\mathbf{k}_2)})$, which follows from the basic inequality $ab \leq (1/2)(a^2+b^2)$. Next define

$$
S_i(\sigma,\Lambda) := \int_{B_{\sigma/c}(0)} |\rho(\mathbf{k})|^2\omega(\mathbf{k})^i, \quad M_i(\Lambda) := \int_{\mathbb{R}^3} |\rho(\mathbf{k})|^2\omega(\mathbf{k})^i.
$$

Note that S_i and M_i are finite for $i \geq -1$ since ρ is a Schwartz function, and obviously

$\int_{\Omega_\sigma} |\rho(\mathbf{k})|^2 \omega(\mathbf{k})^i \leq M_i(\Lambda)$. Putting the above estimates together, we obtain

$$
\begin{aligned}
&\Big|F_7(\mathbf{R},\sigma) - F_7(\mathbf{R})\Big| \\
\leq &\frac{\hbar\, C_{AB}}{18}\Bigg[\int_{\Omega_\sigma \times B_{\sigma/c}(0)} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 \frac{1}{\omega(\mathbf{k}_2)} \left(6\omega(\mathbf{k}_1)\omega(\mathbf{k}_2) + 6\omega(\mathbf{k}_1)^2\right) \\
&\quad + \int_{B_{\sigma/c}(0) \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 \frac{1}{\omega(\mathbf{k}_1)} \left(6\omega(\mathbf{k}_1)\omega(\mathbf{k}_2) + 6\omega(\mathbf{k}_1)^2\right) \\
&\quad + \int_{B_{\sigma/c}(0) \times B_{\sigma/c}(0)} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 \frac{1}{2\sqrt{\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}} \left(6\omega(\mathbf{k}_1)\omega(\mathbf{k}_2) + 6\omega(\mathbf{k}_1)^2\right) \\
&\quad + 2\Bigg[\left(\int_{\Omega_\sigma} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \omega(\mathbf{k}_1)\right)\left(\int_{B_{\sigma/c}(0)} d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\right) \\
&\qquad + \left(\int_{B_{\sigma/c}(0)} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \omega(\mathbf{k}_1)\right)\left(\int_{\Omega_\sigma} d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\right) \\
&\qquad + \left(\int_{B_{\sigma/c}(0)} d\mathbf{k}_1 |\rho(\mathbf{k}_1)|^2 \omega(\mathbf{k}_1)\right)\left(\int_{B_{\sigma/c}(0)} d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\right)\Bigg]\Bigg] \\
\leq &\frac{\hbar\, C_{AB}}{18}\Big[8M_0(\Lambda)S_1(\sigma,\Lambda) + 14M_1(\Lambda)S_0(\sigma,\Lambda) + 6M_2(\Lambda)S_{-1}(\sigma,\Lambda) \\
&\qquad + 3S_{1/2}(\sigma,\Lambda)S_{1/2}(\sigma,\Lambda) + 3S_{3/2}(\sigma,\Lambda)S_{-1/2}(\sigma,\Lambda) + 2S_1(\sigma,\Lambda)S_0(\sigma,\Lambda)\Big].
\end{aligned}
$$

Next we use $\rho(\mathbf{k}) = \rho_0(\mathbf{k}/\Lambda) = \rho_0(|\mathbf{k}|/\Lambda)$ and $\omega(\mathbf{k}) = \omega(|\mathbf{k}|)$ to calculate

$$
\begin{aligned}
S_i(\sigma,\Lambda) =& 4\pi c^i \int_0^{\sigma/c} |\rho(s)|^2 s^{i+2} ds \leq 4\pi c^i \frac{\sigma}{c}\left(\sup_{s\in[0,\sigma/c]} |\rho(s)|^2\right)\left(\sup_{s\in[0,\sigma/c]} s^{i+2}\right) \\
=& \underbrace{4\pi c^i \left(\frac{1}{c}\right)^{i+3}}_{=:\tilde{S}_i} \sigma^{i+3} \left(\sup_{s\in[0,\sigma/(c\Lambda)]} |\rho_0(s)|^2\right).
\end{aligned}
$$

By rescaling variables ($\mathbf{k}' = \mathbf{k}/\Lambda$), we find

$$
M_i(\Lambda) = \Lambda^{3+i} \underbrace{\int_{\mathbb{R}^3} |\rho_0(\mathbf{k})|^2 \omega(\mathbf{k})^i}_{=:\tilde{M}_i},
$$

the integral on the right-hand side now being independent of the ultraviolet-cutoff Λ. The assertion follows upon defining

$$
\begin{aligned}
C_2 :=& \frac{\hbar\, C_{AB}}{18} 6\tilde{M}_2\tilde{S}_{-1},\\
C_3 :=& \frac{\hbar\, C_{AB}}{18} 14\tilde{M}_1\tilde{S}_0,\\
C_4 :=& \frac{\hbar\, C_{AB}}{18} 8\tilde{M}_0\tilde{S}_1,\\
C_7 :=& \frac{\hbar\, C_{AB}}{18}(3\tilde{S}_{1/2}^2 + 3\tilde{S}_{3/2}\tilde{S}_{-1/2} + 2\tilde{S}_1\tilde{S}_0).
\end{aligned}
$$

□

Analogous to above, recall the definition

$$
\begin{aligned}
&F_8(\mathbf{R},\sigma)\\
=& -\frac{1}{36\hbar^2}\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2}{\omega(\mathbf{k}_1)}\frac{|\rho(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_2)}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\quad\times\Big[2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)+\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2)\\
&\qquad -8\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_5(\mathbf{k}_1,\mathbf{k}_1)+\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_6(\mathbf{k}_1,\mathbf{k}_2)\Big]
\end{aligned}
$$

from (3.0.2) and set

$$
\begin{aligned}
&F_8(\mathbf{R})\\
:=& -\frac{1}{36\hbar^2}\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 \frac{|\rho(\mathbf{k}_1)|^2}{\omega(\mathbf{k}_1)}\frac{|\rho(\mathbf{k}_2)|^2}{\omega(\mathbf{k}_2)}(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Big[2\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_5(\mathbf{k}_1,\mathbf{k}_2)+\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_4(\mathbf{k}_1,\mathbf{k}_2)\\
&\qquad -8\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)T_5(\mathbf{k}_1,\mathbf{k}_1)+\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2T_6(\mathbf{k}_1,\mathbf{k}_2)\Big].
\end{aligned}
\tag{6.1.2}
$$

Existence of this integral follows from $|\rho(\cdot_1)|^2\,|\rho(\cdot_2)|^2 \in \mathcal{S}(\mathbb{R}^6)$ and the estimates on T_4 through T_6 derived in the proof of the following Lemma.

Lemma 6.1.2. *There exist positive constants $C_1,\ldots,C_4$ (independent of $\mathbf{R}$ and σ, but depending on Λ via properties of $H_{A,B}$, such that*

$$
\begin{aligned}
\Big|F_8(\mathbf{R},\sigma)-F_8(\mathbf{R})\Big| \leq& \big(C_1\sigma^3\Lambda^5+C_2\sigma^4\Lambda^4+C_3\sigma^5\Lambda^3\big)\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\\
&+C_4\sigma^8\left(\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\right)^2.
\end{aligned}
$$

Proof. First we observe that

$$\int_{\Omega_\sigma\times\Omega_\sigma} = \int_{\mathbb{R}^3\times\mathbb{R}^3} - \int_{B_{\sigma/c}(0)\times\Omega_\sigma} - \int_{\Omega_\sigma\times B_{\sigma/c}(0)} - \int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)}.$$

The standard identity $\|(A-\lambda)^{-1}\| = 1/(\mathrm{dist}(\lambda,\mathrm{spec}(A)))$ for self-adjoint operators yields the estimates

$$\begin{aligned}
&\Big|T_5(\mathbf{k}_i,\mathbf{k}_j)\Big| \\
=&\Big|\sum_{\alpha,\beta=1}^{3}\Big\langle(\overline{H_A+H_B})^{-1}(H_A+\hbar(\omega(\mathbf{k}_i))^{-1}(H_B+\hbar(\omega(\mathbf{k}_j))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta}\Big| \\
\leq&\frac{1}{\Delta_A\Delta_B}\|(\overline{H_A+H_B})^{-1}\|\big(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|^2\big)\big(\sum_{\beta=1}^{3}\|\mathbf{v}_B^\beta\|^2\big) =: \tilde{C}_1, \\
&\Big|T_4(\mathbf{k}_1,\mathbf{k}_2)\Big| \\
=&\Big|\sum_{\alpha=1}^{3}\Big\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha}\alpha_E^B(\mathbf{k}_1) \\
&+\alpha_E^A(\mathbf{k}_1)\Big\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_B^\alpha}\Big| \\
\leq&\left(\frac{1}{\Delta_A^2\Delta_B}+\frac{1}{\Delta_A\Delta_B^2}\right)\big(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|^2\big)\big(\sum_{\beta=1}^{3}\|\mathbf{v}_B^\beta\|^2\big) =: \tilde{C}_2, \\
&\Big|T_6(\mathbf{k}_1,\mathbf{k}_2)\Big| \\
\leq&\|(\overline{H_A+H_B})^{-1}\|\left(\frac{1}{\Delta_A^2}+\frac{1}{\Delta_B^2}\right)\big(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^\alpha\|^2\big)\big(\sum_{\beta=1}^{3}\|\mathbf{v}_B^\beta\|^2\big) =: \tilde{C}_3.
\end{aligned}$$

Furthermore, $|(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)|\leq 2$, so that we obtain

$$\begin{aligned}
&\Big|F_8(\mathbf{R},\sigma)-F_8(\mathbf{R})\Big| \\
\leq&\frac{\hbar^2}{18}\Bigg[\tilde{C}_1\left(\int_{B_{\sigma/c}(0)\times\Omega_\sigma}+\int_{\Omega_\sigma\times B_{\sigma/c}(0)}+\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)}\right)d\mathbf{k}_1d\mathbf{k}_2\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2 \\
&\qquad\times\left(2\omega(\mathbf{k}_1)\,\omega(\mathbf{k}_2)+8\omega(\mathbf{k}_1)^2\right) \\
&+\tilde{C}_2\left(\int_{B_{\sigma/c}(0)\times\Omega_\sigma}+\int_{\Omega_\sigma\times B_{\sigma/c}(0)}+\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)}\right)d\mathbf{k}_1d\mathbf{k}_2\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2\,\omega(\mathbf{k}_1)^2 \\
&+\tilde{C}_3\left(\int_{B_{\sigma/c}(0)\times\Omega_\sigma}+\int_{\Omega_\sigma\times B_{\sigma/c}(0)}+\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)}\right)d\mathbf{k}_1d\mathbf{k}_2\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2 \\
&\qquad\times\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\Bigg].
\end{aligned}$$

Recalling the estimate

$$\int_{B_{\sigma/c}(0)} d\mathbf{k}|\rho(\mathbf{k})|^2\omega(\mathbf{k})^i \leq 4\pi c^i \left(\frac{\sigma}{c}\right)^{i+3} \left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)$$

from the proof of Lemma 6.1.1, we arrive at

$$\begin{aligned}
&\left|F_8(\mathbf{R},\sigma) - F_8(\mathbf{R})\right| \\
\leq &\frac{\hbar^2}{18}\Bigg[(2\tilde{C}_1+\tilde{C}_3)\Bigg[8\pi c\left(\frac{\sigma}{c}\right)^4 \sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2 \left(\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\,\omega(\mathbf{k})\right) \\
&\qquad + (4\pi c)^2\left(\frac{\sigma}{c}\right)^8 \left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)^2\Bigg] \\
&+ (8\tilde{C}_1+\tilde{C}_2)\Bigg[4\pi c^2\left(\frac{\sigma}{c}\right)^5 \left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)\left(\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\right) \\
&\qquad + 4\pi\left(\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\,\omega(\mathbf{k})^2\right)\left(\frac{\sigma}{c}\right)^3 \left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right) \\
&\qquad + (4\pi c)^2\left(\frac{\sigma}{c}\right)^8 \left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)^2\Bigg]\Bigg].
\end{aligned}$$

Finally, using $\rho(\mathbf{k}) = \rho_0(\mathbf{k}/\Lambda)$ and rescaling, we find

$$\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\,\omega(\mathbf{k})^i = \Lambda^{3+i}\int_{\mathbb{R}^3} d\mathbf{k}|\rho_0(\mathbf{k})|^2\,\omega(\mathbf{k})^i,$$

the integral on the right-hand side now being independent of the ultraviolet-cutoff Λ. Defining suitable constants $C_1,\ldots,C_4$ completes the proof. □

6.2 Asymptotic analysis of a class of singular and formally divergent Fourier integrals

6.2.1 Single-particle integrals

The first step in the analysis of the integrals occurring in (6.1.1) is to consider simpler integrals of the form

$$I(R) = \int_{\mathbb{R}^n} f\left(\frac{k_1}{R}\right)\alpha\left(\frac{k_1}{R}\right) m(k_1)P_l(k_1)\mathrm{e}^{-\mathrm{i}k_1\cdot a}dk_1, \tag{6.2.1}$$

where $R>0$, $a\in\mathbb{R}^n$ is a non-zero vector, P_l is a homogeneous polynomial of degree l, f is the Fourier transform of a $C_0^\infty(\mathbb{R}^n)$ function χ with integral 1 (so that, in particular, $f(0)=\widehat{\chi}(0)=1/(2\pi)^{n/2}\int_{\mathbb{R}^n}\chi(x)dx = 1/(2\pi)^{n/2}$), m is a singular and slowly-decaying Fourier multiplier such as

$$\frac{1}{|k_1|},\quad \frac{1}{|k_2|^2} \text{ or } \frac{1}{|k_1|(|k_1|+c)}\quad (c>0),$$

and α is a bounded continuous function such as

$$\alpha(k) = \frac{1}{\lambda + |k|} \quad (\lambda > 0).$$

The difficulty is that m and $\widehat{m}$ only make sense in an L^p sense, so standard Fourier calculus is not applicable. Prototypical is the integral

$$I(R) = \int_{\mathbb{R}^3} f\left(\frac{k_1}{R}\right) \frac{1}{1 + \frac{|k_1|}{R}} \frac{1}{|k_1|(|k_1| + c)} |k_1|^2 \mathrm{e}^{-\mathrm{i}k_1 \cdot a} dk_1. \tag{6.2.2}$$

While this integral is perfectly well-defined for finite $R > 0$ (since f is a Schwartz function and $m(k_1) = \frac{1}{|k_1|(|k_1|+c)}$ is in $L^1(|k_1| \leq 1) \cap L^\infty(|k_1| \geq 1)$), as $R \to \infty$ the integral diverges in the sense of Lebesgue, e.g. in polar coordinates and with $a = (0, 0, |a|)$, one formally obtains the limit

$$2\pi \int_{\theta=0}^{\pi} \int_{r=0}^{\infty} \frac{1}{(2\pi)^{3/2}} \frac{r^4}{r(r+c)} \mathrm{e}^{-\mathrm{i}\, r \cos\theta |a|} \sin\theta d\theta dr.$$

Nevertheless, as we shall see, it in fact has a well-defined limit as $R \to \infty$ provided $|a| > 0$, i.e. provided one has an oscillatory factor. (Of course, for $a = 0$ we have $\lim_{R\to\infty} I(R) = \infty$). Recall the Sobolev spaces

$$W^{l,\infty}(\Omega) = \{u \in L^1_{loc}(\Omega) | u, Du, D^2, \ldots, D^l u \in L^\infty(\Omega)\},$$

where $\Omega \subset \mathbb{R}^n$ is an open subset of $\mathbb{R}^n$ and the derivatives appearing above are distributional ones.

Theorem 6.2.1. *Let l be a non-negative integer, f be the Fourier transform of a $C_0^\infty(\mathbb{R}^n)$-function with integral 1, $m \in (L^1 + L^\infty)(\mathbb{R}^n)$, $a \in \mathbb{R}^n \setminus \{0\}$, $\alpha \in C_b(\mathbb{R}^n)$, and P_l be a homogeneous polynomial of degree l. Assume that α and m are real-valued and define $I(R)$ by (6.2.1). Furthermore, suppose*

$$\widehat{m} \in C^l(\mathbb{R}^n \setminus \{0\}) \cap L^1(B_d(0)) \cap L^\infty(\mathbb{R}^n \setminus B_d(0)) \text{ for all } d > 0, \tag{H1}$$

$$\widehat{\alpha} \in \mathcal{M}(\mathbb{R}^n) \cap W^{l,\infty}(\mathbb{R}^n \setminus B_d(0)) \text{ for all } d > 0, \text{ with}$$

$$\|D^i \widehat{\alpha}\|_{L^\infty(\mathbb{R}^n \setminus B_d(0))} \leq \frac{C(i)}{d^{n+i+\delta}} \text{ for some}$$

$$\text{constants } C(i), \delta > 0, \text{ and all } i = 0, \ldots, l. \tag{H2}$$

Then

$$I(R) \to \alpha(0) \left(\overline{P_l(\mathrm{i}\nabla)\widehat{m}}\right)(-a) \text{ as } R \to \infty.$$

Proof. By Plancherel's formula (denoting the Fourier transform both by $\mathfrak{F}[\cdot]$ and by $\widehat{\cdot}$),

$$I(R) = \int_{\mathbb{R}^n} \mathfrak{F}\big[P_l(\cdot) f\left(\frac{\cdot}{R}\right) \mathrm{e}^{-\mathrm{i}a\cdot}\big](x_1) \overline{\mathfrak{F}\big[\alpha\left(\frac{\cdot}{R}\right) m(\cdot)\big](x_1)} dx_1,$$

and by Fourier calculus

$$I(R) = \frac{1}{(2\pi)^{n/2}} \int_{\mathbb{R}^n} P_l(\mathrm{i}\nabla_{x_1}) \left(\widehat{f}(\cdot + a)\right)_R (x_1) \overline{\left(\int_{\mathbb{R}^n} (\widehat{\alpha})_R (x_1 - x_1') \widehat{m}(x_1') dx_1'\right)} dx_1, \tag{6.2.3}$$

where here and below, for any function g on $\mathbb{R}^n$, we use the shorthand $g_R(\cdot) := (S_R g)(\cdot) = R^n g(R\cdot)$ for the action of the scaling operator S_R. For future use we note that by the Fourier inversion theorem,

$$\int_{\mathbb{R}^n} \widehat{f} = (2\pi)^{n/2} f(0) = 1, \tag{6.2.4}$$

$$\int_{\mathbb{R}^n} \widehat{\alpha} = (2\pi)^{n/2} \alpha(0). \tag{6.2.5}$$

To gain intuition, it is useful to consider the special case when α is constant, say $\alpha = 1$. Then $\widehat{\alpha} = (\widehat{\alpha})_R = (2\pi)^{n/2}\delta$, so

$$\int_{\mathbb{R}^n} (\widehat{\alpha})_R(x_1 - x_1')\widehat{m}(x_1')dx_1' = (2\pi)^{n/2}\widehat{m}(x_1).$$

But since $a \neq 0$, for sufficiently large R the support of $(\widehat{f}(\cdot + a))_R$ is bounded away from zero, so we can integrate by parts to obtain

$$I(R) = \frac{(2\pi)^{n/2}}{(2\pi)^{n/2}} \int_{\mathbb{R}^n} \left(\widehat{f}(\cdot + a)\right)_R \overline{P_l(\mathrm{i}\nabla)\widehat{m}},$$

which tends to

$$\left(\int_{\mathbb{R}^n} \widehat{f}(\cdot + a)\right) \overline{P_l(\mathrm{i}\nabla)\widehat{m}}(-a) = \frac{1}{(2\pi)^{n/2}} \overline{P_l(\mathrm{i}\nabla)\widehat{m}}(-a)$$

by the continuity of $P_l(\mathrm{i}\nabla)\widehat{m}$ and the fact that $(\widehat{f}(\cdot + a))_R$ is a Dirac sequence centered at $-a$.

For non-constant α, much more work is needed. First of all, we pick $d > 0$ so small that whenever $|x_1' - x_1| \leq d$ and $|x_1 - (-a)| \leq d$, then $|x_1'| \geq d$. For instance, the choice $d = |a|/3$ will do. Next we introduce a partition of unity $1 = \chi_< + \chi_>$ with $\chi_<, \chi_> \in C^\infty(\mathbb{R}^n)$, $\operatorname{supp}\chi_< \subset B_d(0)$, $\operatorname{supp}\chi_> \subset \mathbb{R}^n \setminus B_{d/2}(0)$, $\chi_<, \chi_> \geq 0$. We now split $I(R)$ into two parts:

$$I(R) = I_<(R) + I_>(R),$$

with

$$I_\sharp(R) := \frac{1}{(2\pi)^{n/2}} \int_{\mathbb{R}^n} P_l(\mathrm{i}\nabla_{x_1}) \left(\widehat{f}(\cdot + a)\right)_R (x_1) \times \overline{\left(\int_{\mathbb{R}^n} \chi_\sharp(x_1 - x_1')(\widehat{\alpha})_R(x_1 - x_1')\widehat{m}(x_1')dx_1'\right)} dx_1, \tag{6.2.6}$$

where $\sharp \in \{<, >\}$ First we analyze $I_<(R)$. For sufficiently large R, $\operatorname{supp}(\widehat{f}(\cdot + a))_R \subset B_d(-a)$, and hence $|x_1'| \geq d$ for $x_1 \in \operatorname{supp}(\widehat{f}(\cdot + a))_R$, $x_1 - x_1' \in \operatorname{supp}\chi_<$. Integrating by parts, using $\nabla_{x_1}(\chi_<(\widehat{\alpha})_R)(x_1 - x_1') = -\nabla_{x_1'}(\chi_<(\widehat{\alpha})_R)(x_1 - x_1')$, and integrating by parts

again gives

$$\begin{aligned} I_<(R) =& \frac{1}{(2\pi)^{n/2}}(-1)^l \int_{\mathbb{R}^n} \left(\widehat{f}(\cdot + a)\right)_R (x_1) \\ & \qquad \times \overline{\left(\int_{\mathbb{R}^n} P_l(\mathrm{i}\nabla_{x_1'})(\chi_<(\widehat{\alpha})_R)(x_1 - x_1')\widehat{m}(x_1')dx_1'\right)} dx_1 \\ =& \frac{1}{(2\pi)^{n/2}}(-1)^{2l} \int_{\mathbb{R}^n} \left(\widehat{f}(\cdot + a)\right)_R (x_1) \\ & \qquad \times \overline{\left(\int_{\mathbb{R}^n} (\chi_<(\widehat{\alpha})_R)(x_1 - x_1')P_l(\mathrm{i}\nabla_{x_1'})\widehat{m}(x_1')dx_1'\right)} dx_1. \end{aligned} \tag{6.2.7}$$

Note that the first integration by parts does not produce a sign $(-1)^l$ since $\mathrm{i}\nabla_{x_1}$ moves underneath a conjugation sign, but the second integration by parts does since $\mathrm{i}\nabla_{x_1'}$ stays underneath the conjugation sign.
Strictly speaking, in our derivation of (6.2.7), the intermediate expressions including $\nabla_{x_1}(\chi_<(\widehat{\alpha})_R)$ and $\nabla_{x_1'}(\chi_<(\widehat{\alpha})_R)$ are only well-defined when $(\widehat{\alpha})_R$ is smooth, but (6.2.7) can be justified for arbitrary Radon measures $(\widehat{\alpha})_R$ by using that $C_0^\infty(\mathbb{R}^n)$ is weak$*$ dense in $\mathcal{M}(\mathbb{R}^n)$ and noting that the expressions (6.2.6) and (6.2.7) are continuous with respect to weak$*$-convergence of $(\widehat{\alpha})_R$.
We now pass to the limit in (6.2.7). We abbreviate $P_l(\mathrm{i}\nabla_{x_1'})\widehat{m}(x_1') =: g(x_1')$, note that if $x_1 \in \operatorname{supp}(\widehat{f}(\cdot + a))_R$ then $x_1 \in B_d(-a)$ for all sufficiently large R, and claim that the inner integral in (6.2.7) satisfies

$$\sup_{x_1 \in B_d(-a)} \left| \int_{\mathbb{R}^n} (\chi_<(\widehat{\alpha})_R)\,(x_1 - x_1')g(x_1')dx_1' - \left(\int_{\mathbb{R}^n} \widehat{\alpha}(-\cdot)\right) g(x_1) \right| \to 0 \tag{6.2.8}$$

as $R \to \infty$. Indeed, by the changes of variables $z = x_1' - x_1$, $z' = Rz$, we have

$$\begin{aligned} & \int_{\mathbb{R}^n} (\chi_<(\widehat{\alpha})_R)\,(x_1 - x_1')g(x_1')dx_1' - \left(\int_{\mathbb{R}^n} \widehat{\alpha}(-\cdot)\right) g(x_1) \\ =& \int_{\mathbb{R}^n} (\chi_<(\widehat{\alpha})_R)\,(-z)\,(g(z + x_1) - g(x_1))\,dz + g(x_1) \int_{\mathbb{R}^n} \underbrace{(\chi_< - 1)}_{=\chi_>}(\widehat{\alpha})_R(-z)dz \\ =& \int_{B_d(0)} \left(\chi_<\left(\frac{\cdot}{R}\right)\widehat{\alpha}\right)_R (-z)\,(g(z + x_1) - g(x_1))\,dz \\ & + g(x_1) \int_{\mathbb{R}^n} \left(\chi_>\left(\frac{\cdot}{R}\right)\widehat{\alpha}\right)_R (-z)dz \\ =& \int_{B_{Rd}(0)} \left(\chi_<\left(\frac{\cdot}{R}\right)\widehat{\alpha}\right)(-z')\left(g\left(\frac{z'}{R} + x_1\right) - g(x_1)\right)dz' \\ & + g(x_1) \int_{\mathbb{R}^n} \chi_>\left(\frac{-z'}{R}\right)\widehat{\alpha}(-z')dz'. \end{aligned}$$

For $x_1 \in B_d(-a)$, the absolute value of the second term is bounded by

$$\|g\|_{L^\infty(B_d(-a))}\|\widehat{\alpha}\|_{\mathcal{M}(\mathbb{R}^n \setminus B_{Rd/2}(0))} \to 0 \quad (R \to \infty).$$

To estimate the first term, we split the integral into $\int_{B_{R_0 d}(0)} + \int_{B_{Rd}(0)\setminus B_{R_0 d}(0)}$, where $R_0 < R$ is fixed. Hence

$$\begin{aligned}
&\int_{B_{Rd}(0)} \left(\chi_< \left(\frac{\cdot}{R}\right) \widehat{\alpha}\right)(-z') \left(g\left(\frac{z'}{R} + x_1\right) - g(x_1)\right) dz' \\
\leq &\left(\int_{B_{R_0 d}(0)} |\widehat{\alpha}|\right) \sup_{z' \in B_{R_0 d}(0)} \left|g\left(\frac{z'}{R} + x_1\right) - g(x_1)\right| \\
&+ \left(\int_{B_{Rd}(0)\setminus B_{R_0 d}(0)} |\widehat{\alpha}|\right) 2 \sup_{x_1' \in B_{2d}(-a)} |g(x_1')|.
\end{aligned}$$

It follows that

$$\begin{aligned}
&\limsup_{R\to\infty} \sup_{x_1 \in B_d(-a)} \left|\int_{\mathbb{R}^n} (\chi_<(\widehat{\alpha})_R)(x_1 - x_1') g(x_1') dx_1' - \left(\int_{\mathbb{R}^n} \widehat{\alpha}(-\cdot)\right) g(x_1)\right| \\
\leq &\|\widehat{\alpha}\|_{\mathcal{M}(\mathbb{R}^n \setminus B_{R_0 d}(0))} 2 \|g\|_{L^\infty(B_{2d}(-a))}.
\end{aligned}$$

Since R_0 is arbitrary and $\|\widehat{\alpha}\|_{\mathcal{M}(\mathbb{R}^n\setminus B_{R_0 d}(0))} \to 0$ as $R_0 \to \infty$, this establishes (6.2.8). Since the convergence of the inner integral in (6.2.7) is, by (6.2.8), uniform on $B_d(-a) \supset \operatorname{supp}(\widehat{f}(\cdot + a))_R$, it follows that expression (6.2.7) and the expression obtained by replacing the inner integral by its limit, which we denote by $\tilde{I}_<(R)$, have the same limit:

$$\begin{aligned}
&\left|I_<(R) - \tilde{I}_<(R)\right| \\
\leq &\frac{1}{(2\pi)^{n/2}} \left\|\left(\widehat{f}(\cdot + a)\right)_R\right\|_{L^1(\mathbb{R}^n)} \\
&\quad \times \sup_{x_1 \in B_d(-a)} \left|\int_{\mathbb{R}^n} (\chi_<(\widehat{\alpha})_R)(x_1 - x_1') g(x_1') dx_1' - \left(\int_{\mathbb{R}^n} \widehat{\alpha}(-\cdot)\right) g(x_1)\right| \\
&\to 0 \quad (R \to \infty).
\end{aligned} \tag{6.2.9}$$

Finally we pass to the limit in $\tilde{I}_<(R)$. Since g is continuous on $B_d(-a) \supset \operatorname{supp}(\widehat{f}(\cdot + a))_R$,

$$\tilde{I}_<(R) \to \frac{1}{(2\pi)^{n/2}} \left(\int_{\mathbb{R}^n} \widehat{f}\right) \overline{\left(\int_{\mathbb{R}^n} \widehat{\alpha}(-\cdot)\right)} \overline{P_l(\mathrm{i}\nabla_{x_1})\widehat{m}(-a)} \quad (R \to \infty). \tag{6.2.10}$$

Next we observe that the right-hand side of (6.2.10) equals the asserted limit of $I(R)$ in the theorem. This is because $\int_{\mathbb{R}^n} \widehat{f} = 1$ by (6.2.4), and because the relation $\widehat{\alpha}(-\cdot) = \overline{\widehat{\alpha}}$ (which holds since α is real-valued) and (6.2.5) imply

$$\overline{\int_{\mathbb{R}^n} \widehat{\alpha}(-\cdot)} = \overline{\int_{\mathbb{R}^n} \overline{\widehat{\alpha}}} = \int_{\mathbb{R}^n} \widehat{\alpha} = (2\pi)^{n/2} \alpha(0).$$

Altogether we have shown that

$$I_<(R) \to \alpha(0) \left(\overline{P_l(\mathrm{i}\nabla)\widehat{m}}\right)(-a) \tag{6.2.11}$$

as $R \to \infty$.
It remains to investigate $I_>(R)$. We integrate by parts to obtain

$$\begin{aligned}
&\left|(2\pi)^{n/2} I_>(R)\right| \\
=&\Big| \int_{\mathbb{R}^n} \Big(\widehat{f}(\cdot + a)\Big)_R (x_1) \overline{\int_{\mathbb{R}^n} P_l(\mathrm{i}\nabla_{x_1}) \left(\chi_>(\widehat{\alpha})_R\right)(x_1 - x_1') \widehat{m}(x_1') dx_1' dx_1} \Big| \\
\leq& \big\| \big(\widehat{f}(\cdot + a)\big)_R \big\|_{L^1(\mathbb{R}^n)} \sup_{x_1 \in \mathbb{R}^n} \Big| \int_{\mathbb{R}^n} \underbrace{P_l(\mathrm{i}\nabla_{x_1}) \left(\chi_>(\widehat{\alpha})_R\right)(x_1 - x_1')}_{=:h(x_1 - x_1')} \widehat{m}(x_1') dx_1' \Big|. \qquad (6.2.12)
\end{aligned}$$

Note that this time, the derivatives cannot be moved onto $\widehat{m}$ as in (6.2.7) since the region of integration with respect to x_1' contains the singular point $x_1' = 0$, at which $\widehat{m}$ may not be differentiable. But on the other hand, this time the derivatives on $\chi_>(\widehat{\alpha})_R$ are well-defined, since the singular point $x_1 - x_1' = 0$ at which $(\widehat{\alpha})_R$ may not be differentiable is now excluded by the presence of the cutoff function $\chi_>$ which vanishes near $x_1 - x_1' = 0$. By Leibniz' rule,

$$h = P_l(\mathrm{i}\nabla) \left(\chi_>(\widehat{\alpha})_R\right) = \sum_{\nu=0}^{M} \sum_{i=0}^{l} \Big(\tilde{P}_{l-i}^{(\nu)}(\mathrm{i}\nabla)\chi_>\Big) \Big(P_i^{(\nu)}(\mathrm{i}\nabla)(\widehat{\alpha})_R\Big)$$

for some homogeneous polynomials $P_i^{(\nu)}$ and $\tilde{P}_{l-i}^{(\nu)}$ of degree i and $l-i$, respectively. Here M is the number of monomials in P_l. Since $\operatorname{supp} \chi_> \subset \mathbb{R}^n \setminus B_{d/2}(0)$,

$$\begin{aligned}
&\sup_{x_1 \in \mathbb{R}^n} \Big| \int_{\mathbb{R}^n} h(x_1 - x_1') \widehat{m}(x_1') dx_1' \Big| \\
=& \sup_{x_1 \in \mathbb{R}^n} \Big| \int_{|x_1'| < d} h(x_1 - x_1') \widehat{m}(x_1') dx_1' + \int_{|x_1'| \geq d} h(x_1 - x_1') \widehat{m}(x_1') dx_1' \Big| \\
\leq& \|h\|_{L^\infty(\mathbb{R}^n \setminus B_{d/2}(0))} \|\widehat{m}\|_{L^1(B_d(0))} + \|h\|_{L^1(\mathbb{R}^n \setminus B_{d/2}(0))} \|\widehat{m}\|_{L^\infty(\mathbb{R}^n \setminus B_d(0))}. \qquad (6.2.13)
\end{aligned}$$

The norms of $\widehat{m}$ appearing above are finite due to the hypotheses (H1), and the norms of h can be estimated with the help of (H2): For some constants $C, \tilde{C}$,

$$\begin{aligned}
\|h\|_{L^\infty(\mathbb{R}^n \setminus B_{d/2}(0))} \leq& C \sum_{i=0}^{l} \|D^{l-i} \chi_>\|_{L^\infty(\mathbb{R}^n)} \|D^i (\widehat{\alpha})_R\|_{L^\infty(\mathbb{R}^n \setminus B_{d/2}(0))} \\
=& C \sum_{i=0}^{l} \|D^{l-i} \chi_>\|_{L^\infty(\mathbb{R}^n)} R^{n+i} \|D^i \widehat{\alpha}\|_{L^\infty(\mathbb{R}^n \setminus B_{Rd/2}(0))} \\
\underset{(\widehat{H}2)}{\leq}& C \sum_{i=0}^{l} \|D^{l-i} \chi_>\|_{L^\infty(\mathbb{R}^n)} R^{n+i} \frac{C(i)}{\left(\frac{Rd}{2}\right)^{n+i+\delta}} \\
\leq& \tilde{C} \|\chi_>\|_{W^{l,\infty}(\mathbb{R}^n)} \frac{1}{R^\delta} \qquad (6.2.14)
\end{aligned}$$

and

$$\begin{aligned}
\|h\|_{L^1(\mathbb{R}^n \setminus B_{d/2}(0))} \leq& C \sum_{i=0}^{l} \|D^{l-i} \chi_>\|_{L^\infty(\mathbb{R}^n)} \|D^i (\widehat{\alpha})_R\|_{L^1(\mathbb{R}^n \setminus B_{d/2}(0))} \\
\leq& C \sum_{i=0}^{l} \|D^{l-i} \chi_>\|_{L^\infty(\mathbb{R}^n)} R^i \|D^i \widehat{\alpha}\|_{L^1(\mathbb{R}^n \setminus B_{Rd/2}(0))}.
\end{aligned}$$

But (H2) implies

$$\|D^i\widehat{\alpha}\|_{L^1(\mathbb{R}^n\setminus B_d(0))} \leq \int_{\mathbb{R}^n\setminus B_d(0)} \frac{C(i)}{|z|^{n+i+\delta}}dz = \frac{\tilde{C}(i)}{d^{i+\delta}}$$

with $\tilde{C}(i) = |S^{n-1}|\frac{C(i)}{i+\delta}$, whence

$$\|h\|_{L^1(\mathbb{R}^n\setminus B_{d/2}(0))} \leq C'\|\chi_>\|_{W^{l,\infty}(\mathbb{R}^n)}\frac{1}{R^\delta} \tag{6.2.15}$$

for a suitable constant $C' > 0$. Combining (6.2.12), (6.2.13), (6.2.14) and (6.2.15) yields

$$|I_>(R)| \leq \text{const} \cdot \frac{1}{R^\delta}. \tag{6.2.16}$$

Together with (6.2.11) this establishes the theorem. □

Remark 6.2.2. In the application we have in mind, where α is a dynamic polarizability, as we will show (H2) is valid with $\delta = 1$. Hence (6.2.16) shows that the contribution $I_>(R)$ from the long-range part of the polarizability is $\mathcal{O}(1/R)$.

6.2.2 Two-particle integrals

Having established results concerning the asymptotic behaviour of integrals of the form (6.2.1), we continue to generalize these to integrals of the form encountered in (6.1.1), namely

$$I(R) = \int_{\mathbb{R}^n\times\mathbb{R}^n} f\left(\frac{k_1}{R}\right) g\left(\frac{k_2}{R}\right) \alpha\left(\frac{k_1}{R}\right) \beta\left(\frac{k_2}{R}\right) P(k_1,k_2)m(k_1,k_2)\mathrm{e}^{-\mathrm{i}k_1\cdot a}\mathrm{e}^{-\mathrm{i}k_1\cdot b}. \tag{6.2.17}$$

We will use the following function spaces. Here l_1, l_2 are non-negative integers, and d is a positive real number.

$$\begin{aligned}
X^{0,0,d}(\mathbb{R}^{2n}) := \{u \in L^1_{loc}(\mathbb{R}^{2n}) | u \in & L^1(|x_1| \leq d, |x_2| \leq d) \\
& \cap L^1(|x_2| \leq d; L^\infty(|x_1| > d)) \\
& \cap L^1(|x_1| \leq d; L^\infty(|x_2| > d)) \\
& \cap L^\infty(|x_1| > d, |x_2| > d)\}, \\
X^{l_1,l_2,d}(\mathbb{R}^{2n}) := \{u \in L^1_{loc}(\mathbb{R}^{2n}) | & u \in X^{0,0,d}(\mathbb{R}^{2n}), \\
& D^{l_1}_{x_1} u \in L^1(|x_2| \leq d; L^\infty(|x_1| > d)) \\
& \qquad \cap L^\infty(|x_1| > d, |x_2| > d), \\
& D^{l_2}_{x_2} u \in L^1(|x_1| \leq d; L^\infty(|x_2| > d)) \\
& \qquad \cap L^\infty(|x_1| > d, |x_2| > d)\}.
\end{aligned}$$

The spaces $X^{l_1,l_2,d}(\mathbb{R}^{2n})$ are Banach spaces with the following norms (where sup denotes essential supremum):

$$\begin{aligned}
\|u\|_{X^{0,0,d}} :=& \int_{B_d(0)\times B_d(0)} |u| \\
&+ \int_{|x_2|\le d} \sup_{|x_1|>d} |u(x_1,x_2)|dx_2 \\
&+ \int_{|x_1|\le d} \sup_{|x_2|>d} |u(x_1,x_2)|dx_1 \\
&+ \sup_{|x_1|>d,|x_2|>d} |u(x_1,x_2)|, \\
\|u\|_{X^{l_1,l_2,d}} :=& \|u\|_{X^{0,0,d}} \\
&+ \int_{|x_2|\le d} \sup_{|x_1|>d} |D_{x_1}^{l_1} u(x_1,x_2)|dx_2 \\
&+ \sup_{|x_1|>d,|x_2|>d} |D_{x_1}^{l_1} u(x_1,x_2)| \\
&+ \int_{|x_1|\le d} \sup_{|x_2|>d} |D_{x_2}^{l_2} u(x_1,x_2)|dx_1 \\
&+ \sup_{|x_1|>d,|x_2|>d} |D_{x_2}^{l_2} u(x_1,x_2)|.
\end{aligned}$$

Note that the derivatives $D_{x_1}^{l_1}u$, $D_{x_2}^{l_2}u$ are not required to be in $X^{0,0,d}$, because the coordinates with respect to which u is being differentiated only enter into the $X^{l_1,l_2,d}$ norm in the 'tail' region $\mathbb{R}^n \setminus B_d(0)$, but not in the 'head' region B_d. This is essential for the applicability of the theorem below to the multipliers arising in the study of the interaction potential $V(\Lambda, \mathbf{R})$.

Theorem 6.2.3. *Suppose that f, g are Fourier transforms of non-negative $C_0^\infty(\mathbb{R}^n)$ functions with integral 1, $m \in (L^1 + L^\infty)(\mathbb{R}^{2n})$ and that $P(k_1,k_2)$ is a polynomial which is homogeneous of degree l_1 in k_1 and homogeneous of degree l_2 in k_2. Furthermore, let α, β be bounded continuous functions on $\mathbb{R}^n$ and let $a, b \in \mathbb{R}^n \setminus \{0\}$. Assume that α, β and m are real-valued, and define $I(R)$ by (6.2.17). Suppose that*

$$\begin{aligned}
&\widehat{m} \in C^{l_1,l_2}((\mathbb{R}^n \setminus \{0\})^2) \cap X^{l_1,l_2,d} \text{ for all } d > 0, && \text{(H1)}\\
&\widehat{\alpha}, \widehat{\beta} \in \mathcal{M}(\mathbb{R}^n), \widehat{\alpha} \in W^{l_1,\infty}(\mathbb{R}^n \setminus B_d(0)), \\
&\widehat{\beta} \in W^{l_2,\infty}(\mathbb{R}^n \setminus B_d(0)) \text{ for all } d > 0, \text{ with} \\
&\|D^i \widehat{\alpha}\|_{L^\infty(\mathbb{R}^n \setminus B_d(0))} \le \frac{C(i)}{d^{n+i+\delta}},\ i = 0, \dots, l_1, \\
&\|D^j \widehat{\beta}\|_{L^\infty(\mathbb{R}^n \setminus B_d(0))} \le \frac{\tilde{C}(j)}{d^{n+j+\delta}},\ j = 0, \dots, l_2 \\
&\text{for some constants } C(i), \tilde{C}(j), \delta > 0. && \text{(H2)}
\end{aligned}$$

Then

$$I(R) \to \alpha(0)\beta(0)\left(\overline{P(\mathrm{i}\nabla_{x_1}, \mathrm{i}\nabla_{x_2})\widehat{m}}\right)(-a,-b) \text{ as } R \to \infty.$$

Proof. Because the assertion is linear in P, we can assume without loss of generality that $P(k_1,k_2)=P_1(k_1)P_2(k_2)$, with P_i homogeneous of degree l_i, $i=1,2$. By the real-valuedness of α,β and m, as in the proof of Theorem 6.2.1, we have

$$I(R)=\frac{1}{(2\pi)^n}\int_{\mathbb{R}^{2n}} P_1(\mathrm{i}\nabla_{x_1})P_2(\mathrm{i}\nabla_{x_2})(\widehat{f}(\cdot+a))_R(x_1)(\widehat{g}(\cdot+b))_R(x_2) \times\overline{\left(\int_{\mathbb{R}^{2n}}(\widehat{\alpha})_R(x_1-x_1')(\widehat{\beta})_R(x_2-x_2')\widehat{m}(x_1',x_2')dx_1'dx_2'\right)}dx_1dx_2. \quad (6.2.18)$$

Let $d:=\min\{|a|,|b|\}/3$, and let $\chi_<,\chi_>\in C^\infty(\mathbb{R}^n)$ be the partition of unity introduced in the proof of Theorem 6.2.1. We have

$$I(R)=I_{<<}(R)+I_{<>}(R)+I_{><}(R)+I_{>>}(R),$$

where, for $u,v\in\{<,>\}$,

$$I_{uv}(R)=\frac{1}{(2\pi)^n}\int_{\mathbb{R}^{2n}} P_1(\mathrm{i}\nabla_{x_1})P_2(\mathrm{i}\nabla_{x_2})(\widehat{f}(\cdot+a))_R(x_1)(\widehat{g}(\cdot+b))_R(x_2) \times\overline{\Big(\int_{\mathbb{R}^{2n}}\chi_u(x_1-x_1')(\widehat{\alpha})_R(x_1-x_1')\chi_v(x_2-x_2')(\widehat{\beta})_R(x_2-x_2') \cdot\overline{\widehat{m}(x_1',x_2')dx_1'dx_2'}\Big)}dx_1dx_2. \quad (6.2.19)$$

The first integral, $I_{<<}(R)$, has exactly the same structure as the integral $I_<(R)$ in the proof of Theorem 6.2.1, with $\mathbb{R}^{2n}$ instead of $\mathbb{R}^n$, and $\widehat{f}(\cdot+a)\otimes\widehat{g}(\cdot+b)$, $(\chi_<\otimes\chi_<)\cdot((\widehat{\alpha})_R\otimes(\widehat{\beta})_R)$ instead of $\widehat{f}(\cdot+a)$, $\chi_<\cdot(\widehat{\alpha})_R$, respectively. Hence arguing as in the derivation of (6.2.11), we obtain

$$I_{<<}(R)\to\alpha(0)\beta(0)\left(\overline{P_1(\mathrm{i}\nabla_{x_1})P_2(\mathrm{i}\nabla_{x_2})\widehat{m}}\right)(-a,-b)\text{ as }R\to\infty. \quad (6.2.20)$$

Next we consider $I_{<>}(R)$. By (6.2.7),

$$I_{<>}(R)=\frac{1}{(2\pi)^n}\int_{\mathbb{R}^{2n}}(\widehat{f}(\cdot+a))_R(x_1)(\widehat{g}(\cdot+b))_R(x_2) \times\overline{\Big(\int_{\mathbb{R}^{2n}}\left(\chi_<(\widehat{\alpha})_R\right)(x_1-x_1')P_1(\mathrm{i}\nabla_{x_1'})\widehat{m}(x_1',x_2') \cdot\overline{P_2(\mathrm{i}\nabla_{x_2})\left(\chi_>(\widehat{\beta})_R\right)(x_2-x_2')dx_1'dx_2'}\Big)}dx_1dx_2.$$

Instead of analyzing the integral over x_1 and x_1' in the finer manner of equation (6.2.8), we use the following estimate: assuming R is so large that $\operatorname{supp}(\widehat{f}(\cdot+a))_R\subset B_d(-a)$,

$$\begin{aligned}&\left|\int\mathbb{R}^{2n}(\widehat{f}(\cdot+a))_R(x_1)\left(\chi_<(\widehat{\alpha})_R\right)(x_1-x_1')P_1(\mathrm{i}\nabla_{x_1'})\widehat{m}(x_1',x_2')dx_1dx_1'\right|\\ \le&\|\widehat{f}\|_{L^1(\mathbb{R}^n)}\|\widehat{\alpha}\|_{\mathcal{M}(\mathbb{R}^n)}\sup_{|x_1'|>d}\left|P_1(\mathrm{i}\nabla_{x_1'})\widehat{m}(x_1',x_2')\right|. \end{aligned}\quad (6.2.21)$$

Here we have used that if $x_1\in\operatorname{supp}(\widehat{f}(\cdot+a))_R\subset B_d(-a)$ and $x_1-x_1'\in\operatorname{supp}\chi_<\subset B_d(0)$, then $|x_1'|\ge|a|-2d\ge d$ (since $d\le|a|/3$). Abbreviating $h:=P_2(\mathrm{i}\nabla)(\chi_>(\widehat{\beta})_R)$, it follows

that

$$
\begin{aligned}
|I_{<>}(R)| \leq & \frac{1}{(2\pi)^n} \|\widehat{f}\|_{L^1(\mathbb{R}^n)} \|\widehat{\alpha}\|_{\mathcal{M}(\mathbb{R}^n)} \\
& \times \Bigg[\Big(\int_{|x_2'| \leq d} \sup_{|x_1'|>d} \big| P_1(\mathrm{i}\nabla_{x_1'}) \widehat{m}(x_1', x_2') \big| dx_2' \Big) \|\widehat{g}\|_{L^1(\mathbb{R}^n)} \|h\|_{L^\infty(\mathbb{R}^n)} \\
& \quad + \sup_{|x_2'|>d} \sup_{|x_1'|>d} \big| P_1(\mathrm{i}\nabla_{x_1'}) \widehat{m}(x_1', x_2') \big| \|\widehat{g}\|_{L^1(\mathbb{R}^n)} \|h\|_{L^1(\mathbb{R}^n)} \Bigg].
\end{aligned}
$$

Since $\operatorname{supp} h \subset \mathbb{R}^n \setminus B_{d/2}(0)$, $\|h\|_{L^p(\mathbb{R}^n)} = \|h\|_{L^p(\mathbb{R}^n \setminus B_{d/2}(0))}$ $(1 \leq p \leq \infty)$, and by (6.2.14), (6.2.15) and (H2),

$$
\|h\|_{L^\infty(\mathbb{R}^n \setminus B_{d/2}(0))} \leq \tilde{C} \|\chi_>\|_{W^{l_2,\infty}(\mathbb{R}^n)} \frac{1}{R^\delta}, \tag{6.2.22}
$$

$$
\|h\|_{L^1(\mathbb{R}^n \setminus B_{d/2}(0))} \leq C' \|\chi_>\|_{W^{l_2,\infty}(\mathbb{R}^n)} \frac{1}{R^\delta}. \tag{6.2.23}
$$

Summarizing and noting that the expressions involving $\widehat{m}$ are controlled by $\|\widehat{m}\|_{X^{l_1,0,d}}$, this yields

$$
|I_{<>}(R)| \leq \text{const} \cdot \|\chi_>\|_{W^{l_2,\infty}(\mathbb{R}^n)} \|\widehat{m}\|_{X^{l_1,0,d}} \frac{1}{R^\delta} \to 0 \text{ as } R \to \infty. \tag{6.2.24}
$$

The integral $I_{><}(R)$ can be treated analogously by moving $P_1(\mathrm{i}\nabla_{x_1})$ onto $\chi_>(\widehat{\alpha})_R$ and $P_2(\mathrm{i}\nabla_{x_2'})$ onto $\widehat{m}(x_1', x_2')$, yielding

$$
\begin{aligned}
|I_{><}(R)| = & \Bigg| \frac{1}{(2\pi)^n} \int_{\mathbb{R}^{2n}} (\widehat{f}(\cdot + a))_R(x_1) (\widehat{g}(\cdot + b))_R(x_2) \\
& \quad \times \overline{\Big(\int_{\mathbb{R}^{2n}} (\chi_<(\widehat{\beta})_R)(x_2 - x_2') P_2(\mathrm{i}\nabla_{x_2'}) \widehat{m}(x_1', x_2') } \\
& \qquad \overline{\cdot P_1(\mathrm{i}\nabla_{x_1}) \, (\chi_>(\widehat{\alpha})_R) \, (x_1 - x_1') dx_1' dx_2' \Big)} dx_1 dx_2 \Bigg| \\
\leq & \text{const} \cdot \|\chi_>\|_{W^{l_1,\infty}(\mathbb{R}^n)} \|\widehat{m}\|_{X^{0,l_2,d}} \frac{1}{R^\delta} \to 0 \text{ as } R \to \infty.
\end{aligned} \tag{6.2.25}
$$

It remains to study the long-range part $I_{>>}(R)$. We move $P_1(\mathrm{i}\nabla_{x_1})$ and $P_2(\mathrm{i}\nabla_{x_2})$ onto $\chi_>(\widehat{\alpha})_R$ and $\chi_>(\widehat{\beta})_R$, respectively, and estimate the integrals over x_1 and x_2 by taking L^1

norms of $\widehat{f}$ and $\widehat{g}$ and the supremum over x_1, x_2 of the the inner integral:

$$|I_{>>}(R)| = \Bigg| \frac{1}{(2\pi)^n} \int_{\mathbb{R}^{2n}} (\widehat{f}(\cdot + a))_R(x_1)(\widehat{g}(\cdot + b))_R(x_2) \times \overline{\Big(\int_{\mathbb{R}^{2n}} P_1(\mathrm{i}\nabla_{x_1})(\chi_>(\widehat{\alpha})_R)(x_1 - x_1') \cdot \overline{P_2(\mathrm{i}\nabla_{x_2}) \left(\chi_>(\widehat{\beta})_R \right) (x_2 - x_2') \widehat{m}(x_1', x_2') dx_1' dx_2'} \Big) dx_1 dx_2} \Bigg| \tag{6.2.26}$$

$$\leq \frac{1}{(2\pi)^n} \|\widehat{f}\|_{L^1(\mathbb{R}^n)} \|\widehat{g}\|_{L^1(\mathbb{R}^n)} \times \sup_{x_1, x_2 \in \mathbb{R}^n} \Bigg| \int_{\mathbb{R}^{2n}} P_1(\mathrm{i}\nabla_{x_1})(\chi_>(\widehat{\alpha})_R)(x_1 - x_1') \cdot P_2(\mathrm{i}\nabla_{x_2}) \left(\chi_>(\widehat{\beta})_R \right) (x_2 - x_2') \widehat{m}(x_1', x_2') dx_1' dx_2' \Bigg| \tag{6.2.27}$$

To estimate the integral on the right, we split it into four terms:

$$\int_{\mathbb{R}^{2n}} = \int_{|x_1'|, |x_2'| \leq d} + \int_{|x_1'| \leq d, |x_2'| > d} + \int_{|x_1'| > d, |x_2'| \leq d} + \int_{|x_1'|, |x_2'| > d}.$$

It follows that, for any $x_1, x_2 \in \mathbb{R}^n$,

$$\begin{aligned}
&\Bigg| \int_{\mathbb{R}^{2n}} P_1(\mathrm{i}\nabla_{x_1})(\chi_>(\widehat{\alpha})_R)(x_1 - x_1') \cdot P_2(\mathrm{i}\nabla_{x_2}) \left(\chi_>(\widehat{\beta})_R \right) (x_2 - x_2') \widehat{m}(x_1', x_2') dx_1' dx_2' \Bigg| \\
\leq &\Big(\int_{B_d(0) \times B_d(0)} |\widehat{m}| \Big) \|P_1(\mathrm{i}\nabla)(\chi_>(\widehat{\alpha})_R)\|_{L^\infty(\mathbb{R}^n)} \|P_2(\mathrm{i}\nabla)(\chi_>(\widehat{\beta})_R)\|_{L^\infty(\mathbb{R}^n)} \\
&+ \Big(\int_{|x_1'| \leq d} \sup_{|x_2'| > d} |\widehat{m}(x_1', x_2')| dx_1' \Big) \|P_1(\mathrm{i}\nabla)(\chi_>(\widehat{\alpha})_R)\|_{L^\infty(\mathbb{R}^n)} \|P_2(\mathrm{i}\nabla)(\chi_>(\widehat{\beta})_R)\|_{L^1(\mathbb{R}^n)} \\
&+ \Big(\int_{|x_2'| \leq d} \sup_{|x_1'| > d} |\widehat{m}(x_1', x_2')| dx_2' \Big) \|P_1(\mathrm{i}\nabla)(\chi_>(\widehat{\alpha})_R)\|_{L^1(\mathbb{R}^n)} \|P_2(\mathrm{i}\nabla)(\chi_>(\widehat{\beta})_R)\|_{L^\infty(\mathbb{R}^n)} \\
&+ \Big(\sup_{|x_1'| > d, |x_2'| > d} |\widehat{m}(x_1', x_2')| \Big) \|P_1(\mathrm{i}\nabla)(\chi_>(\widehat{\alpha})_R)\|_{L^1(\mathbb{R}^n)} \|P_2(\mathrm{i}\nabla)(\chi_>(\widehat{\beta})_R)\|_{L^1(\mathbb{R}^n)}.
\end{aligned} \tag{6.2.28}$$

But, as already used repeatedly, $\operatorname{supp} \chi_> \subset \mathbb{R}^n \setminus B_{d/2}(0)$, so the $L^p(\mathbb{R}^n)$ norms appearing in (6.2.28) equal the corresponding $L^p(\mathbb{R}^n \setminus B_{d/2}(0))$ norms. Estimating the latter by (6.2.14), (6.2.15) and (H2) gives

$$\begin{aligned}
\|P_1(\mathrm{i}\nabla)(\chi_>(\widehat{\alpha})_R)\|_{L^p(\mathbb{R}^n \setminus B_{d/2}(0))} &\leq \tilde{C}_1 \|\chi_>\|_{W^{l_1,\infty}(\mathbb{R}^n)} \frac{1}{R^\delta} \quad (p = 1, \infty), \\
\|P_2(\mathrm{i}\nabla)(\chi_>(\widehat{\beta})_R)\|_{L^p(\mathbb{R}^n \setminus B_{d/2}(0))} &\leq \tilde{C}_2 \|\chi_>\|_{W^{l_2,\infty}(\mathbb{R}^n)} \frac{1}{R^\delta} \quad (p = 1, \infty)
\end{aligned}$$

for suitable positive constants $\tilde{C}_i$, $i = 1, 2$. Since the norms of $\widehat{m}$ appearing in (6.2.28) are estimated by the norm $\|\widehat{m}\|_{X^{0,0,d}}$, it follows that

$$\begin{aligned} |I_{>>}(R)| \leq & \text{const} \cdot \|\chi_>\|_{W^{l_1,\infty}(\mathbb{R}^n)} \|\chi_>\|_{W^{l_2,\infty}(\mathbb{R}^n)} \|\widehat{m}\|_{X^{0,0,d}} \frac{1}{R^\delta} \\ & \to 0 \quad (R \to \infty). \end{aligned} \tag{6.2.29}$$

Combining (6.2.20), (6.2.24), (6.2.25) and (6.2.29) completes the proof. □

6.3 Regularity of dynamic polarizabilities

We prove here the subtle fact that the dynamic polarizabilities $\alpha_E^{A,B}(\mathbf{k})$ (for which we will use the shorthand α) contained in the term $F_7(\mathbf{R})$ (see(6.1.1)) satisy the regularity conditions

$$\widehat{\alpha} \in \mathcal{M}(\mathbb{R}^3), \tag{6.3.1}$$

$$\|D^i \widehat{\alpha}\|_{L^\infty(\mathbb{R}^3 \setminus B_d(0))} \leq \frac{C(i)}{d^{3+i+\delta}}, \; i = 0, 1, 2, \ldots, \; \delta > 0 \tag{6.3.2}$$

required for the rigorous asymptotic method introduced in Section 6.2. These properties of $\widehat{\alpha}$ are subtle because α is far from being a Schwartz function: it has a $|\mathbf{k}|$-singularity at $\mathbf{k} = 0$ and only decays like $1/|\mathbf{k}|$ at infinity. In fact, the analysis below shows that (6.3.2) holds for each i with sharp optimal incremental exponent $\delta = 1$, but fails for every $i = 0, 1, 2, \ldots$ when $\delta > 1$. In the remainder of this and the next section, we will refrain from using bold face notation for vectors in $\mathbb{R}^3$ in order to simplify notation.
As can be read off from their definition (see Theorem 2.8.4), the dynamic polarizabilities are of the form

$$\alpha(k) = \int_{\mathbb{R}} dm(\lambda) \frac{1}{\lambda + \gamma|k|} \quad (k \in \mathbb{R}^3), \tag{6.3.3}$$

with $m \in \mathcal{P}(\mathbb{R})$ (the space of propability measures), $\operatorname{supp} m \subset [\lambda_0, \infty)$ for some $\lambda_0 > 0$, and $\gamma > 0$. Physically, m is a spectral measure and the term $\gamma|k|$ is a dispersion relation, with $\gamma|k| = \hbar\omega(k) = \hbar c|k|$, whence $\gamma = \hbar c$.

Theorem 6.3.1. *Let α be given by (6.3.3). Then*

i. $\widehat{\alpha} \in L^1(\mathbb{R}^3)$, and in particular α satisfies (6.3.1).

ii. α satisfies (6.3.2) with $\delta = 1$.

Proof. First of all, note that one can assume without loss of generality that $\gamma = 1$, because otherwise

$$\alpha(k) = \frac{1}{\gamma} \int dm(\lambda) \frac{1}{\frac{\lambda}{\gamma} + |k|} = \frac{1}{\gamma} \int dm'(\lambda') \frac{1}{\lambda' + |k|},$$

where $m'(A) := m(\gamma \cdot A)$ for any Borel set $A \subset \mathbb{R}$, i.e. α is of the required form up to a constant prefactor.
Next we will need a very explicit expression for the Fourier transform $\widehat{\alpha}$. To this end

we use what one may call a 'Slater transform', i.e. we write $1/(\lambda + |k|)$ as an infinite superposition of Slater-type orbitals,

$$\frac{1}{\lambda + |k|} = \int_0^\infty \mathrm{e}^{-(\lambda+|k|)s} ds = \int_0^\infty \mathrm{e}^{-\lambda s} \mathrm{e}^{-|k|s} ds \tag{6.3.4}$$

and use the well-known formula (see e.g. [Str94])

$$\widehat{\mathrm{e}^{-s|\cdot|}}(x) = \frac{4}{(2\pi)^{1/2}} \frac{s}{(s^2 + |x|^2)^2}.$$

Consequently

$$\mathfrak{F}\Big[\frac{1}{\lambda + |\cdot|}\Big](x) = \frac{4}{(2\pi)^{1/2}} \int_0^\infty \mathrm{e}^{-\lambda s} \frac{s}{(s^2 + |x|^2)^2} ds, \tag{6.3.5}$$

see also (6.4.4).

Estimate 1 For all $\lambda \in \operatorname{supp} m$, by the assumptions on m and by (6.3.4), (6.3.5), we have

$$\begin{aligned}
0 \leq & \mathfrak{F}\Big[\frac{1}{\lambda + |\cdot|}\Big](x) \leq \frac{4}{(2\pi)^{1/2}} \int_0^\infty \mathrm{e}^{-\lambda_0 s} \frac{s}{(s^2 + |x|^2)^2} ds \\
= & \frac{4}{(2\pi)^{1/2}} \int_0^\infty \mathrm{e}^{-\lambda_0 s} \left(-\frac{1}{2} \frac{d}{ds} \frac{1}{s^2 + |x|^2} \right) ds \\
= & \underbrace{\Big[-\frac{2}{(2\pi)^{1/2}} \frac{\mathrm{e}^{-\lambda_0 s}}{s^2 + |x|^2} \Big]_{s=0}^\infty}_{= \frac{2}{(2\pi)^{1/2}|x|^2}} \underbrace{- \frac{2}{(2\pi)^{1/2}} \int_0^\infty \lambda_0 \, \mathrm{e}^{-\lambda_0 s} \frac{1}{s^2 + |x|^2} ds}_{\leq 0} \\
\leq & \frac{2}{(2\pi)^{1/2}|x|^2}.
\end{aligned}$$

Multiplying by the measure $m(\lambda)$ and integrating over λ yields

$$0 \leq \mathfrak{F}\Big[\int_{\mathbb{R}} dm(\lambda) \frac{1}{\lambda + |\cdot|}\Big](x) = \widehat{\alpha}(x) \leq \frac{2}{(2\pi)^{1/2}|x|^2}. \tag{6.3.6}$$

Estimate 2 By the assumptions on m and by (6.3.4), (6.3.5),

$$\begin{aligned}
0 \leq & \mathfrak{F}\Big[\frac{1}{\lambda + |\cdot|}\Big](x) \leq \frac{4}{(2\pi)^{1/2}} \int_0^\infty \mathrm{e}^{-\lambda_0 s} \frac{s}{(s^2 + |x|^2)^2} ds \\
\leq & \frac{4}{(2\pi)^{1/2}} \frac{1}{|x|^4} \int_0^\infty \mathrm{e}^{-\lambda_0 s} s \, ds.
\end{aligned} \tag{6.3.7}$$

An elementary calculation shows that the integral appearing on the right-hand side equals $1/\lambda_0^2$. Multiplying (6.3.7) by the measure $m(\lambda)$ and integrating over λ yields

$$0 \leq \widehat{\alpha}(x) \leq \frac{4}{\lambda_0^2 (2\pi)^{1/2}} \frac{1}{|x|^4}. \tag{6.3.8}$$

Since $1/|x|^2 \in L^1(|x| \leq 1)$ and $1/|x|^4 \in L^1(|x| > 1)$, (6.3.6) and (6.3.8) show in particular that $\widehat{\alpha} \in L^1(\mathbb{R}^3)$, establishing assertion i).

Derivative Estimates Let D_x^l be a partial differential operator l which is of the form

$$D_x^l = \prod_{i=1}^{3} \frac{\partial^{l_i}}{\partial x_i^{l_i}}, \quad l_i \in \mathbb{N} \cup \{0\}, \quad \sum_{i=1}^{3} l_i = l.$$

By (6.3.5),

$$D_x^l \widehat{\alpha}(x) = \frac{4}{(2\pi)^{1/2}} \int_{\mathbb{R}} dm(\lambda) \int_0^\infty \mathrm{e}^{-\lambda s}\, s\, D_x^l \frac{1}{(s^2+|x|^2)^2}. \tag{6.3.9}$$

We claim that

$$D_x^l \frac{1}{(s^2+|x|^2)^2} = \begin{cases} \sum_{j=0, j\,\mathrm{even}}^{l} P_j(x) \frac{1}{(s^2+|x|^2)^{(l+j)/2+2}}, & l \,\mathrm{even} \\ \sum_{j=1, j\,\mathrm{odd}}^{l} P_j(x) \frac{1}{(s^2+|x|^2)^{(l+j)/2+2}}, & l \,\mathrm{odd} \end{cases}, \tag{6.3.10}$$

where the P_j are (l-dependent) homogeneous polynomials of degree j. I.e.,

$$\begin{aligned} D_x^0 \frac{1}{(s^2+|x|^2)^2} =& P_0(x) \frac{1}{(s^2+|x|^2)^2}, \\ D_x^1 \frac{1}{(s^2+|x|^2)^2} =& P_1(x) \frac{1}{(s^2+|x|^2)^3}, \\ D_x^2 \frac{1}{(s^2+|x|^2)^2} =& P_2(x) \frac{1}{(s^2+|x|^2)^4} + P_0(x) \frac{1}{(s^2+|x|^2)^3}, \end{aligned}$$

et cetera. This is easily proved by induction, noting that (for any $m > 0$ and with P_j denoting any homogeneous polynomial of degree j),

$$\begin{aligned} D_x^1 P_0(x) \frac{1}{(s^2+|x|^2)^m} =& P_1(x) \frac{1}{(s^2+|x|^2)^{m+1}}, \\ D_x^1 P_j(x) \frac{1}{(s^2+|x|^2)^m} =& P_{j+1}(x) \frac{1}{(s^2+|x|^2)^{m+1}} + P_{j-1}(x) \frac{1}{(s^2+|x|^2)^m}. \end{aligned}$$

Using the fact that for any P_j there exists a constant C such that

$$|P_j(x)| \le C|x|^j \le C(s^2+|x|^2)^{j/2},$$

it now follows from (6.3.10) that, for suitable constants C_l,

$$\left| D_x^l \frac{1}{(s^2+|x|^2)^2} \right| \le C_l \frac{1}{(s^2+|x|^2)^{l/2+2}}. \tag{6.3.11}$$

Using this estimate for the right-hand side of (6.3.9) gives

$$\begin{aligned} \left| D_x^l \widehat{\alpha}(x) \right| \le& \frac{4}{(2\pi)^{1/2}} \int_{\mathbb{R}} dm(\lambda) \int_0^\infty \mathrm{e}^{-\lambda_0 s}\, s\, \frac{C_l}{|x|^{l+4}} ds, \\ =& \frac{4C_l}{\lambda_0^2 (2\pi)^{1/2}} \frac{1}{|x|^{l+4}} \quad (l = 0, 1, 2, \dots), \end{aligned} \tag{6.3.12}$$

establishing assertion ii) and completing the proof. □

Next we verify the regularity conditions (6.3.1) and (6.3.2) for pointwise products of dynamic polarizabilities, e.g. $(\alpha_E^A(\mathbf{k}_1))^2$. The latter are of the form

$$\alpha(k) = \int_{\mathbb{R}} \int_{\mathbb{R}} dm(\lambda) dm'(\lambda') \frac{1}{\lambda + \gamma|k|} \frac{1}{\lambda' + \gamma|k|} \quad (k \in \mathbb{R}^3), \tag{6.3.13}$$

with $m, m' \in \mathcal{P}(\mathbb{R})$, $\operatorname{supp} m, \operatorname{supp} m' \subset [\lambda_0, \infty)$ for some $\lambda_0 > 0$, and $\gamma > 0$.

Theorem 6.3.2. *Let α be given by (6.3.13). Then*

i. $\widehat{\alpha} \in L^1(\mathbb{R}^3)$, in particular α satisfies (6.3.1).

ii. α satisfies (6.3.2) with $\delta = 1$.

Proof. As in the proof of Theorem 6.3.1, we may assume $\gamma = 1$. Applying the 'Slater transform' trick introduced there to both factors $1/(\lambda + |k|)$ and $1/(\lambda' + |k|)$ yields the explicit representation

$$\alpha(k) = \int_{\mathbb{R}} \int_{\mathbb{R}} dm(\lambda) dm'(\lambda') \int_{s=0}^{\infty} \int_{s'=0}^{\infty} \mathrm{e}^{-\lambda s} \mathrm{e}^{-\lambda' s'} \mathrm{e}^{-(s+s')|k|} ds ds',$$

and consequently

$$\widehat{\alpha}(k) = \int_{\mathbb{R}} \int_{\mathbb{R}} dm(\lambda) dm'(\lambda') \underbrace{\frac{4}{(2\pi)^{1/2}} \int_{s=0}^{\infty} \int_{s'=0}^{\infty} \mathrm{e}^{-\lambda s} \mathrm{e}^{-\lambda' s'} \frac{s + s'}{((s+s')^2 + |x|^2)^2} ds ds'}_{=: T_{\lambda, \lambda'}(x)}. \tag{6.3.14}$$

To estimate $\widehat{\alpha}$ at short range, we calculate

$$\begin{aligned} 0 \leq T_{\lambda,\lambda'}(x) =& \frac{4}{(2\pi)^{1/2}} \int_{s=0}^{\infty} \int_{s'=0}^{\infty} \mathrm{e}^{-\lambda s} \mathrm{e}^{-\lambda' s'} \Big(-\frac{1}{2} \frac{d}{ds} \frac{1}{|x|^2 + (s+s')^2} \Big) ds ds' \\ =& -\frac{2}{(2\pi)^{1/2}} \int_0^{\infty} \mathrm{e}^{-\lambda' s'} \Big[\frac{\mathrm{e}^{-\lambda s}}{|x|^2 + (s+s')^2} \Big]_{s=0}^{\infty} ds' \\ & - \frac{2}{(2\pi)^{1/2}} \int_0^{\infty} \int_0^{\infty} \lambda \, \mathrm{e}^{-\lambda s} \mathrm{e}^{-\lambda' s'} \frac{1}{|x|^2 + (s+s')^2} ds ds'. \end{aligned}$$

The first term equals

$$\frac{2}{(2\pi)^{1/2}} \int_0^{\infty} \mathrm{e}^{-\lambda' s'} \frac{1}{|x|^2 + (s')^2} ds'$$

and can be bounded from above for any $\lambda' \in \operatorname{supp} m'$ by

$$\frac{2}{(2\pi)^{1/2}} \int_0^{\infty} \mathrm{e}^{-\lambda_0 s'} \frac{1}{|x|^2} ds' = \frac{2}{\lambda_0 (2\pi)^{1/2}} \frac{1}{|x|^2}.$$

The second term is less or equal to zero. It follows that

$$0 \leq \widehat{\alpha}(x) \leq \frac{2}{\lambda_0 (2\pi)^{1/2}} \frac{1}{|x|^2}. \tag{6.3.15}$$

At long range, the representation (6.3.14) immediately gives

$$0 \leq \widehat{\alpha}(x) \leq \frac{4}{\lambda_0 (2\pi)^{1/2}} \underbrace{\Big(\int_0^\infty \int_0^\infty \mathrm{e}^{-\lambda_0 (s+s')} \, (s+s') ds ds' \Big)}_{=:C_*} \frac{1}{|x|^4}. \tag{6.3.16}$$

As before, (6.3.15) and (6.3.16) imply assertion i). Finally, combining (6.3.14) with the derivative estimate (6.3.11) (with $s+s'$ in place of s) yields

$$\big| D_x^l \widehat{\alpha}(x) \big| \leq C_* \frac{C_l}{|x|^{l+4}},$$

establishing assertion ii) and completing the proof. □

6.4 Calculation of distributional Fourier transforms

In order to apply Theorem 6.2.3 to analyze the large-R-asymptotics of $F_7(\mathbf{R})$, we need to calculate the distributional Fourier transform of the functions

$$W_1(\mathbf{k}_1, \mathbf{k}_2) := \frac{1}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1| + |\mathbf{k}_2|)}, \quad W_2(\mathbf{k}_1, \mathbf{k}_2) := \frac{1}{|\mathbf{k}_1||\mathbf{k}_2|^2}.$$

Lemma 6.4.1. *W_1 and W_2 define tempered distributions $S \in \mathcal{S}'(\mathbb{R}^6)$, and their distributional Fourier transforms are given by*

$$\widehat{W_1}(\mathbf{k}_1, \mathbf{k}_2) = \frac{1}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1| + |\mathbf{k}_2|)} = W_1(\mathbf{k}_1, \mathbf{k}_2),$$
$$\widehat{W_2}(\mathbf{k}_1, \mathbf{k}_2) = \frac{1}{|\mathbf{k}_1|^2 |\mathbf{k}_2|} = W_2(\mathbf{k}_2, \mathbf{k}_1).$$

Proof. First note that both W_1 and W_2 are in $L^1_{loc}(\mathbb{R}^6)$ and decay like inverse polynomials at infinity. For W_2 this is immediate, and for W_1 it follows from the elementary estimate $|\mathbf{k}_1| + |\mathbf{k}_2| \geq \sqrt{2}\sqrt{|\mathbf{k}_1|}\sqrt{|\mathbf{k}_2|}$. Thus we conclude $W_1, W_2 \in \mathcal{S}'(\mathbb{R}^6)$, and thus their distributional Fourier transforms are well-defined. Furthermore, since both functions are homogeneous of degree -3, we know that both $\widehat{W_1}$ and $\widehat{W_2}$ are also homogeneous of degree $-n - \alpha = -6 - (-3) = -3$ (as distributions).
Since W_2 is of product structure, the second assertion of the lemma follows by separation of variables and the well known formulas

$$\widehat{\frac{1}{|\cdot|}} = \frac{2}{(2\pi)^{1/2}} \frac{1}{|\cdot|^2}, \quad \widehat{\frac{1}{|\cdot|^2}} = \frac{(2\pi)^{1/2}}{2} \frac{1}{|\cdot|}$$

on $\mathcal{S}'(\mathbb{R}^3)$, see e.g. [Str94] (note that the Fourier convention used there differs from ours). To prove the assertion about W_1, note that by Fubini's theorem, the $\mathbb{R}^6$-Fourier transform of a test function $\varphi \in \mathcal{S}(\mathbb{R}^6)$ can be written as the the consecutive application to φ of the partial Fourier transform with respect to $\mathbf{k}_1$, $\mathbf{k}_2$, respectively. That is to say,

$$\begin{aligned} \hat{\varphi}(\mathbf{k}_1, \mathbf{k}_2) =& \frac{1}{(2\pi)^{6/2}} \int_{\mathbb{R}^6} \varphi(\tilde{\mathbf{k}}_1, \tilde{\mathbf{k}}_2) \mathrm{e}^{-\mathrm{i}(\tilde{\mathbf{k}}_1 \cdot \mathbf{k}_1 + \tilde{\mathbf{k}}_2 \cdot \mathbf{k}_2)} d\tilde{\mathbf{k}}_1 d\tilde{\mathbf{k}}_2 \\ =& \Big[(\mathfrak{F}_1 \otimes I)(I \otimes \mathfrak{F}_2) \varphi \Big] (\mathbf{k}_1, \mathbf{k}_2), \end{aligned}$$

where

$$[(\mathfrak{F}_1 \otimes I)\varphi]\,(\mathbf{k}_1, \mathbf{k}_2) = \frac{1}{(2\pi)^{3/2}} \int_{\mathbb{R}^3} \varphi(\tilde{\mathbf{k}}_1, \mathbf{k}_2) \mathrm{e}^{-\mathrm{i}(\tilde{\mathbf{k}}_1 \cdot \mathbf{k}_1)} d\tilde{\mathbf{k}}_1.$$

Using this and the fact that for fixed $\mathbf{k}_2 \in \mathbb{R}^3$, $[(\mathfrak{F}_1 \otimes \mathfrak{F}_2)\varphi](\cdot, \mathbf{k}_2) \in \mathcal{S}(\mathbb{R}^3)$, we find

$$\begin{aligned}
&\langle \widehat{W_1}, \varphi \rangle = \langle W_1, \hat{\varphi} \rangle \\
=&\langle W_1, (\mathfrak{F}_1 \otimes \mathfrak{F}_2)\varphi \rangle \\
=&\int_{\mathbb{R}^3} \frac{1}{|\mathbf{k}_2|} \Big[\int_{\mathbb{R}^3} \frac{1}{|\mathbf{k}_1|\,(|\mathbf{k}_1| + |\mathbf{k}_2|)} [(\mathfrak{F}_1 \otimes \mathfrak{F}_2)\varphi](\mathbf{k}_1, \mathbf{k}_2) d\mathbf{k}_1 \Big] d\mathbf{k}_2 \\
=&\int_{\mathbb{R}^3} \frac{1}{|\mathbf{k}_2|} \Big\langle \frac{1}{|\mathbf{k}_1|\,(|\mathbf{k}_1| + |\mathbf{k}_2|)}, \Big[(\mathfrak{F}_1 \otimes \mathfrak{F}_2)\varphi \Big](\cdot, \mathbf{k}_2) \Big\rangle_{\mathbf{k}_1} d\mathbf{k}_2 \\
=&\int_{\mathbb{R}^3} \frac{1}{|\mathbf{k}_2|} \Big\langle \mathfrak{F}_1 \Big[\frac{1}{|\mathbf{k}_1|\,(|\mathbf{k}_1| + |\mathbf{k}_2|)} \Big], \Big[(I \otimes \mathfrak{F}_2)\varphi \Big](\cdot, \mathbf{k}_2) \Big\rangle_{\mathbf{k}_1} d\mathbf{k}_2. \qquad (6.4.1)
\end{aligned}$$

Now for $\mathbf{k}_1 \neq 0$, $\mathbf{k}_2 \neq 0$,

$$\frac{1}{(|\mathbf{k}_1|(|\mathbf{k}_1| + |\mathbf{k}_2|)} = \frac{1}{|\mathbf{k}_2|} \left(\frac{1}{|\mathbf{k}_1|} - \frac{1}{(|\mathbf{k}_1| + |\mathbf{k}_2|)} \right). \qquad (6.4.2)$$

For $0 \neq a \in \mathbb{R}$, consider the function $f_a(\mathbf{k}) = 1/(|\mathbf{k}| + a)$ on $\mathbb{R}^3$. We have $f_a \in L^1_{loc}(\mathbb{R}^3)$, and f_a decays like $1/|\mathbf{k}|$ for $|\mathbf{k}| \to \infty$, so $f_a \in \mathcal{S}'(\mathbb{R}^3)$, and thus its distributional Fourier transform $\hat{f}_a$ is well-defined. Using the representation

$$\frac{1}{|\mathbf{k}| + a} = \int_0^\infty \mathrm{e}^{-(|\mathbf{k}|+a)s} ds,$$

we will calculate $\hat{f}_a$ explicitly. To this end, let $\varphi \in \mathcal{S}(\mathbb{R}^3)$. Then

$$\begin{aligned}
\langle \hat{f}_a, \varphi \rangle =& \langle \frac{1}{|\cdot| + a}, \hat{\varphi} \rangle = \int_{\mathbb{R}^3} \frac{1}{|\mathbf{k}| + a} \hat{\varphi}(\mathbf{k}) d\mathbf{k} \\
=& \int_{\mathbb{R}^3} \Big[\int_0^\infty \mathrm{e}^{-|\mathbf{k}|s} \mathrm{e}^{-as} ds \Big] \hat{\varphi}(\mathbf{k}) d\mathbf{k}. \qquad (6.4.3)
\end{aligned}$$

Note that by using $|\mathrm{e}^{-|\mathbf{k}|\,|s|} \mathrm{e}^{-as} \hat{\varphi}(\mathbf{k})| \leq \mathrm{e}^{-as} |\hat{\varphi}(\mathbf{k})|$, we can estimate

$$\int_{\mathbb{R}^3 \times (0,\infty)} \Big| \mathrm{e}^{-|\mathbf{k}|\,|s|} \mathrm{e}^{-a|s|} \hat{\varphi}(\mathbf{k}) \Big| \, d\mathbf{k} \otimes ds \leq \Big(\int_0^\infty \mathrm{e}^{-a|s|} ds \Big) \Big(\|\hat{\varphi}\|_{L^1(\mathbb{R}^3)} \Big),$$

the right-hand side being finite by the assumption $a \neq 0$, so that we can apply Fubini's theorem to (6.4.3) and obtain

$$\langle \hat{f}_a, \varphi \rangle = \int_0^\infty \mathrm{e}^{-a|s|} \Big[\int_{\mathbb{R}^3} \mathrm{e}^{-|\mathbf{k}|\,|s|} \hat{\varphi}(\mathbf{k}) d\mathbf{k} \Big] ds.$$

For $s \neq 0$, an explicit expression for the distributional Fourier transform of $\mathrm{e}^{-|\cdot|s}$ (see for instance [Str94]; note that a different Fourier convention is used by the author) yields

$$\langle \mathrm{e}^{-|\cdot|s}, \widehat{\varphi} \rangle = \langle \widehat{\mathrm{e}^{-|\cdot|s}}, \varphi \rangle = \frac{4}{(2\pi)^{1/2}} \int_{\mathbb{R}^3} \frac{s}{(s^2 + |\mathbf{k}|^2)^2} \varphi(\mathbf{k}) d\mathbf{k}.$$

Furthermore, the integral

$$\frac{4}{(2\pi)^{1/2}}\int_0^\infty \frac{s}{(s^2+|\mathbf{k}|^2)^2}\mathrm{e}^{-as}ds \tag{6.4.4}$$

can be calculated explicitly and equals

$$g_a(\mathbf{k}) := \frac{2}{(2\pi)^{1/2}}\frac{1}{|\mathbf{k}|^2} - \frac{2}{(2\pi)^{1/2}}\frac{a}{|\mathbf{k}|}\Big[\sin(a|\mathbf{k}|)\mathrm{Ci}(a|\mathbf{k}|) - \cos(a|\mathbf{k}|)(\mathrm{Si}(a|\mathbf{k}|) - \frac{\pi}{2})\Big].$$

It is easily checked that $g_a(\mathbf{k}) \in L^1(\mathbb{R}^3)$ (for instance by noting that $\int_{\mathbb{R}^3}\frac{s}{(s^2+|\mathbf{k}|^2)^2}d\mathbf{k} = \widehat{\widehat{\mathrm{e}^{-|\cdot|s}}}(0) = 1$), so that

$$\frac{4}{(2\pi)^{1/2}}\int_{\mathbb{R}^3}\Big[\int_0^\infty \frac{s}{(s^2+|\mathbf{k}|^2)^2}\mathrm{e}^{-as}ds\Big]|\varphi(\mathbf{k})|d\mathbf{k} = \int_{\mathbb{R}^3} g_a(\mathbf{k})|\varphi(\mathbf{k})|d\mathbf{k} < \infty.$$

Since the integrand is non-negative (for g_a this follows from its explicit representation (6.4.4)), Tonelli's theorem (in conjunction with Fubini's theorem) implies

$$\langle \hat{f}_a, \varphi\rangle = \int_0^\infty \mathrm{e}^{-a|s|}\Big[\int_{\mathbb{R}^3}\mathrm{e}^{-|\mathbf{k}|\,|s|}\hat{\varphi}(\mathbf{k})d\mathbf{k}\Big]ds = \int_{\mathbb{R}^3} g_a(\mathbf{k})\varphi(\mathbf{k})d\mathbf{k}.$$

Thus we have shown that $\hat{f}_a = g_a$ as distributions. In view of (6.4.2), this yields

$$\begin{aligned}
&\mathfrak{F}_1\left[\frac{1}{|\mathbf{k}_1|\,(|\mathbf{k}_1|+|\mathbf{k}_2|)}\right]\\
=&\frac{1}{|\mathbf{k}_2|}\left(\mathfrak{F}_1\left[\frac{1}{|\mathbf{k}_1|}\right] - \mathfrak{F}_1\left[\frac{1}{|\mathbf{k}_1|+|\mathbf{k}_2|}\right]\right)\\
=&\frac{1}{|\mathbf{k}_2|}\Big(\frac{2}{(2\pi)^{1/2}}\frac{1}{|\mathbf{k}_1|^2} - g_{|\mathbf{k}_2|}(\mathbf{k}_1)\Big)\\
=&\frac{1}{|\mathbf{k}_2|}\left(\frac{2}{(2\pi)^{1/2}}\frac{|\mathbf{k}_2|}{|\mathbf{k}_1|}\Big[\sin(|\mathbf{k}_2|\,|\mathbf{k}_1|)\mathrm{Ci}(|\mathbf{k}_2|\,\mathbf{k}_1|) - \cos(|\mathbf{k}_2|\,|\mathbf{k}_1|)(\mathrm{Si}(|\mathbf{k}_2|\,|\mathbf{k}_1|) - \frac{\pi}{2})\Big]\right)\\
=&\frac{2}{(2\pi)^{1/2}}\frac{1}{|\mathbf{k}_1|}\Big[\sin(|\mathbf{k}_2|\,|\mathbf{k}_1|)\mathrm{Ci}(|\mathbf{k}_2|\,\mathbf{k}_1|) - \cos(|\mathbf{k}_2|\,|\mathbf{k}_1|)(\mathrm{Si}(|\mathbf{k}_2|\,|\mathbf{k}_1|) - \frac{\pi}{2})\Big].
\end{aligned}$$

Plugging this into (6.4.1), we obtain

$$\begin{aligned}
\langle\widehat{W_1},\varphi\rangle = &\frac{2}{(2\pi)^{1/2}}\int_{\mathbb{R}^6}\frac{1}{|\mathbf{k}_1|\,|\mathbf{k}_2|}\left[(I\otimes\mathfrak{F}_2)\varphi\right](\mathbf{k}_1,\mathbf{k}_2)\\
&\Big[\sin(|\mathbf{k}_2|\,|\mathbf{k}_1|)\mathrm{Ci}(|\mathbf{k}_2|\,\mathbf{k}_1|) - \cos(|\mathbf{k}_2|\,|\mathbf{k}_1|)(\mathrm{Si}(|\mathbf{k}_2|\,|\mathbf{k}_1|) - \frac{\pi}{2})\Big]d\mathbf{k}_1 d\mathbf{k}_2.
\end{aligned}$$

By Fubini's theorem and the definition of the dual pairing of $\mathcal{S}'(\mathbb{R}^3)$ and $\mathcal{S}(\mathbb{R}^3)$,

$$\begin{aligned}
&\langle\widehat{W_1},\varphi\rangle\\
=&\int_{\mathbb{R}^3}\frac{1}{|\mathbf{k}_1|^2}\Big\langle\frac{|\mathbf{k}_1|}{|\mathbf{k}_2|}\frac{2}{(2\pi)^{1/2}}\Big[\sin(|\mathbf{k}_2|\,|\mathbf{k}_1|)\mathrm{Ci}(|\mathbf{k}_2|\,\mathbf{k}_1|)\\
&\qquad - \cos(|\mathbf{k}_2|\,|\mathbf{k}_1|)(\mathrm{Si}(|\mathbf{k}_2|\,|\mathbf{k}_1|) - \frac{\pi}{2})\Big], \Big[(I\otimes\mathfrak{F}_2)\varphi\Big](\mathbf{k}_1,\cdot)\Big\rangle_{\mathbf{k}_2} d\mathbf{k}_1\\
=&\int_{\mathbb{R}^3}\frac{1}{|\mathbf{k}_1|^2}\Big\langle\mathfrak{F}_2\Big[\frac{|\mathbf{k}_1|}{|\mathbf{k}_2|}\frac{2}{(2\pi)^{1/2}}\Big[\sin(|\mathbf{k}_2|\,|\mathbf{k}_1|)\mathrm{Ci}(|\mathbf{k}_2|\,\mathbf{k}_1|)\\
&\qquad - \cos(|\mathbf{k}_2|\,|\mathbf{k}_1|)(\mathrm{Si}(|\mathbf{k}_2|\,|\mathbf{k}_1|) - \frac{\pi}{2})\Big]\Big], \varphi(\mathbf{k}_1,\cdot)\Big\rangle_{\mathbf{k}_2} d\mathbf{k}_1.
\end{aligned}$$

By the Fourier inversion theorem on $\mathcal{S}'(\mathbb{R}^3)$,

$$\begin{aligned}&\mathfrak{F}_2\Big[\frac{|\mathbf{k}_1|}{|\mathbf{k}_2|}\frac{2}{(2\pi)^{1/2}}\Big[\sin(|\mathbf{k}_2|\,|\mathbf{k}_1|)\mathrm{Ci}(|\mathbf{k}_2|\,\mathbf{k}_1|)-\cos(|\mathbf{k}_2|\,|\mathbf{k}_1|)(\mathrm{Si}(|\mathbf{k}_2|\,|\mathbf{k}_1|)-\frac{\pi}{2})\Big]\Big]\\ =&\frac{1}{|\mathbf{k}_2|}-\frac{1}{|\mathbf{k}_1|+|\mathbf{k}_2|}\end{aligned}$$

(note that this latter function is invariant under the reflection $\mathbf{k}_2 \to -\mathbf{k}_2$), so we end up with

$$\begin{aligned}&\langle\widehat{W_1},\varphi\rangle\\ =&\int_{\mathbb{R}^3}\frac{1}{|\mathbf{k}_1|^2}\left\langle\frac{1}{|\cdot|}-\frac{1}{|\mathbf{k}_1|+|\cdot|},\varphi(\mathbf{k}_1,\cdot)\right\rangle_{\mathbf{k}_2}d\mathbf{k}_1\\ =&\int_{\mathbb{R}^3}\frac{1}{|\mathbf{k}_1|^2}\left\langle\frac{|\mathbf{k}_1|}{|\cdot|\,(|\mathbf{k}_1|+|\cdot|)},\varphi(\mathbf{k}_1,\cdot)\right\rangle_{\mathbf{k}_2}d\mathbf{k}_1\\ =&\int_{\mathbb{R}^6}\frac{1}{|\mathbf{k}_1|\,|\mathbf{k}_2|\,(|\mathbf{k}_1|+|\mathbf{k}_2|)}\varphi(\mathbf{k}_1,\mathbf{k}_2)d\mathbf{k}_1d\mathbf{k}_2,\end{aligned}$$

finishing the proof. □

6.5 Asymptotics of $F_7(\mathbf{R})$ and $F_8(\mathbf{R})$

In this section we first use the results of the previous three sections to analyze the large-$|\mathbf{R}|$-asymptotics of the term $F_7(\mathbf{R})$ from (6.1.1), see the following theorem. Subsequently, we shall analyze the asymptotic behaviour of the term $F_8(\mathbf{R})$ from (6.1.2).

Theorem 6.5.1. *Assume the hypotheses of Theorem 3.0.6. Then*

$$\lim_{R\to\infty}\Big(R^kF_7(\mathbf{R})\Big)=0$$

for any $0\le k<7$, *and*

$$\lim_{R\to\infty}\big(R^7F_7(\mathbf{R})\big)=\frac{41}{2}\frac{\hbar c}{(2\pi)^3}\frac{1}{9}\alpha_E^A(0)\,\alpha_E^B(0).$$

Proof. Recalling the definition of $C(\mathbf{k})$ and $\omega(\mathbf{k})$ (for the former, see (3.0.3)) and using the identity

$$\begin{aligned}&\frac{-4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2-6\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\\ =&\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\\ &\times\left(\frac{-2\omega(\mathbf{k}_1)^2-4\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)-4((\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))^2-\omega(\mathbf{k}_2)^2-2\omega(\mathbf{k}_1)\omega(\mathbf{k}_2))}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\ =&\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\\ &\times\left(2(\omega(\mathbf{k}_2)-\omega(\mathbf{k}_1))+4\frac{\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}+2\frac{\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-4\omega(\mathbf{k}_1)-4\omega(\mathbf{k}_2)\right)\end{aligned}$$

$$
\begin{aligned}
=&\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\\
&\times\left(-2\frac{\omega(\mathbf{k}_2)(\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2))}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-6\omega(\mathbf{k}_1)+4\frac{\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}+2\frac{\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}\right)\\
=&\frac{2\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2}{\omega(\mathbf{k}_1)+\omega(\mathbf{k}_2)}-6\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)
\end{aligned}
$$

which holds on $\mathbb{R}^6\setminus(0,0)$, we find

$$
\begin{aligned}
F_7(\mathbf{R})=&-\frac{\hbar c}{36}\int_{\mathbb{R}^6}d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Bigg[(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_1))\left(\frac{2|\mathbf{k}_1||\mathbf{k}_2|}{|\mathbf{k}_1|+|\mathbf{k}_2|}-6|\mathbf{k}_1|\right)\\
&\quad+\left(\alpha_E^A(\mathbf{k}_1)\alpha_E^B(\mathbf{k}_2)+\alpha_E^A(\mathbf{k}_2)\alpha_E^B(\mathbf{k}_1)\right)\left(\frac{|\mathbf{k}_1||\mathbf{k}_2|}{|\mathbf{k}_1|+|\mathbf{k}_2|}-|\mathbf{k}_1|\right)\Bigg].
\end{aligned}
$$

Rescaling variables $\mathbf{k}_i\mapsto(R/\Lambda)\mathbf{k}_i$ and recalling $\rho(\mathbf{k})=\rho_0(\mathbf{k}/\Lambda)$ leads to

$$
\begin{aligned}
F_7(\mathbf{R})=&-\frac{\hbar c}{36}\left(\frac{\Lambda}{R}\right)^7\int_{\mathbb{R}^6}d\mathbf{k}_1d\mathbf{k}_2|\rho_0(\mathbf{k}_1/R)|^2|\rho_0(\mathbf{k}_2/R)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\Lambda\hat{\mathbf{R}}}\\
&\times\Bigg[(\alpha_E^A((\Lambda/R)\mathbf{k}_1)\alpha_E^B((\Lambda/R)\mathbf{k}_1))\left(\frac{2|\mathbf{k}_1||\mathbf{k}_2|}{|\mathbf{k}_1|+|\mathbf{k}_2|}-6|\mathbf{k}_1|\right)\\
&\quad+\Big(\alpha_E^A((\Lambda/R)\mathbf{k}_1)\alpha_E^B((\Lambda/R)\mathbf{k}_2)+\alpha_E^A((\Lambda/R)\mathbf{k}_2)\alpha_E^B((\Lambda/R)\mathbf{k}_1)\Big)\\
&\qquad\times\left(\frac{|\mathbf{k}_1||\mathbf{k}_2|}{|\mathbf{k}_1|+|\mathbf{k}_2|}-|\mathbf{k}_1|\right)\Bigg],
\end{aligned}
$$

Using assumption (A1), the ensuing relation (2.1.10) and the behaviour of the Fourier transform under convolutions, we conclude

$$
\begin{aligned}
&\widehat{|\rho_0|^2}=\widehat{\rho_0\,\overline{\rho_0}}=\widehat{\rho_0\,\rho_0}\\
=&(2\pi)^{-3/2}(\widehat{\rho_0}*\widehat{\rho_0})=(2\pi)^{-3/2}(\tilde{\psi_0}*\tilde{\psi_0})\\
=&(2\pi)^{-3/2}(\psi_0*\psi_0)
\end{aligned}
$$

where $\tilde{f}(x)=f(-x)$ denotes reflection, and we have used that ρ_0 is real and ψ_0 is even. Thus, using that ρ_0 is even by (A1), we find

$$
|\rho_0(\frac{\cdot}{R})|^2=\widehat{\widehat{|\rho_0|^2}}(\frac{\cdot}{R})=(2\pi)^{-3/2}\underbrace{\mathfrak{F}\Big[\psi_0*\psi_0\Big]}_{=:f}(\frac{\cdot}{R})
$$

It follows from the assumptions (A1) that $\psi_0*\psi_0$ is a non-negative $C_0^\infty(\mathbb{R}^3)$ function with integral 1 (note that $\int(\psi_0*\psi_0)(x)dx=(\int\psi_0(x)dx)^2=1$ and $\psi_0*\psi_0\in C_0^\infty(\mathbb{R}^3)$). Using this, we rewrite $F_7(\mathbf{R})$ as

$$
F_7(\mathbf{R})=-\frac{\hbar c}{36}\left(\frac{\Lambda}{R}\right)^7\frac{1}{(2\pi)^3}(I_1(\mathbf{R}),+I_2(\mathbf{R})),\tag{6.5.1}
$$

where

$$I_1(\mathbf{R}) := \int_{\mathbb{R}^6} d\mathbf{k}_1 d\mathbf{k}_2 f(\mathbf{k}_1/R) f(\mathbf{k}_2/R)(|\mathbf{k}_1|^2|\mathbf{k}_2|^2 + (\mathbf{k}_1\cdot\mathbf{k}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\Lambda\hat{\mathbf{R}}} \times (\alpha_E^A((\Lambda/R)\mathbf{k}_1)\alpha_E^B((\Lambda/R)\mathbf{k}_1)) \underbrace{\left(\frac{2}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1|+|\mathbf{k}_2|)} - \frac{6}{|\mathbf{k}_1||\mathbf{k}_2|^2}\right)}_{=:m_1(\mathbf{k}_1,\mathbf{k}_2)}, \tag{6.5.2}$$

$$I_2(\mathbf{R}) := \int_{\mathbb{R}^6} d\mathbf{k}_1 d\mathbf{k}_2 f(\mathbf{k}_1/R) f(\mathbf{k}_2/R)(|\mathbf{k}_1|^2|\mathbf{k}_2|^2 + (\mathbf{k}_1\cdot\mathbf{k}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\Lambda\hat{\mathbf{R}}} \times \left(\alpha_E^A((\Lambda/R)\mathbf{k}_1)\alpha_E^B((\Lambda/R)\mathbf{k}_2) + \alpha_E^A((\Lambda/R)\mathbf{k}_2)\alpha_E^B((\Lambda/R)\mathbf{k}_1)\right) \times \underbrace{\left(\frac{1}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1|+|\mathbf{k}_2|)} - \frac{1}{|\mathbf{k}_1||\mathbf{k}_2|^2}\right)}_{=:m_2(\mathbf{k}_1,\mathbf{k}_2)}. \tag{6.5.3}$$

Note that both $I_1(\mathbf{R})$ and $I_2(\mathbf{R})$ are of the form (6.2.17), with $f = g$, $a = b = \Lambda\hat{\mathbf{R}}$ and

$$P(\mathbf{k}_1,\mathbf{k}_2) = (|\mathbf{k}_1|^2|\mathbf{k}_2|^2 + (\mathbf{k}_1\cdot\mathbf{k}_2)^2)$$

As we have just shown, f is the Fourier transform of a non-negative $C_0^\infty(\mathbb{R}^3)$ function with integral 1. Furthermore, the multipliers m_1 and m_2 are elements of $(L^1+L^\infty)(\mathbb{R}^6)$, and P is a polynomial which is homogeneous of degree 2 in both $\mathbf{k}_1$ and $\mathbf{k}_2$. By Lemma 6.4.1,

$$\widehat{m_1}(\mathbf{k}_1,\mathbf{k}_2) = \frac{2}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1|+|\mathbf{k}_2|)} - \frac{6}{|\mathbf{k}_1|^2|\mathbf{k}_2|},$$
$$\widehat{m_2}(\mathbf{k}_1,\mathbf{k}_2) = \frac{1}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1|+|\mathbf{k}_2|)} - \frac{1}{|\mathbf{k}_1|^2|\mathbf{k}_2|},$$

and these functions are elements of $C^{2,2}((\mathbb{R}^n\setminus\{0\})^2)\cap X^{2,2,d}$ for all $d>0$. Furthermore, Theorems 6.3.2 and 6.3.1 state that $\alpha_E^A((\Lambda/R)\mathbf{k}_1)\alpha_E^B((\Lambda/R)\mathbf{k}_1)$, $\alpha_E^A((\Lambda/R)\mathbf{k}_1)\alpha_E^B((\Lambda/R)\mathbf{k}_2)$ and $\alpha_E^A((\Lambda/R)\mathbf{k}_2)\alpha_E^B((\Lambda/R)\mathbf{k}_1)$ satisfy the assumptions (H2) of Theorem 6.2.3. Applying said theorem now yields

$$\begin{aligned}
&\lim_{R\to\infty}(I_1(\mathbf{R})+I_2(\mathbf{R}))\\
=&\alpha_E^A(0)\alpha_E^B(0)\left[\left(\left(\Delta_{\mathbf{k}_1}\otimes\Delta_{\mathbf{k}_2} + (\nabla_{\mathbf{k}_1}\cdot\nabla_{\mathbf{k}_2})^2\right)\widehat{m_1}\right)(-\Lambda\hat{\mathbf{R}},-\Lambda\hat{\mathbf{R}})\right.\\
&\left.+2\left(\left(\Delta_{\mathbf{k}_1}\otimes\Delta_{\mathbf{k}_2} + (\nabla_{\mathbf{k}_1}\cdot\nabla_{\mathbf{k}_2})^2\right)\widehat{m_2}\right)(-\Lambda\hat{\mathbf{R}},-\Lambda\hat{\mathbf{R}})\right]\\
=&\alpha_E^A(0)\alpha_E^B(0)\left[4\left(\left(\Delta_{\mathbf{k}_1}\otimes\Delta_{\mathbf{k}_2} + (\nabla_{\mathbf{k}_1}\cdot\nabla_{\mathbf{k}_2})^2\right)\frac{1}{|\mathbf{k}_1||\mathbf{k}_2|(|\mathbf{k}_1|+|\mathbf{k}_2|)}\right)(-\Lambda\hat{\mathbf{R}},-\Lambda\hat{\mathbf{R}})\right.\\
&\left.-8\left(\left(\Delta_{\mathbf{k}_1}\otimes\Delta_{\mathbf{k}_2} + (\nabla_{\mathbf{k}_1}\cdot\nabla_{\mathbf{k}_2})^2\right)\frac{1}{|\mathbf{k}_1|^2|\mathbf{k}_2|}\right)(-\Lambda\hat{\mathbf{R}},-\Lambda\hat{\mathbf{R}})\right].
\end{aligned}$$

An elementary calculation shows

$$\begin{aligned}
&\Big[\Delta_{\mathbf{k}_1}\otimes\Delta_{\mathbf{k}_2}+(\nabla_{\mathbf{k}_1}\cdot\nabla_{\mathbf{k}_2})^2\Big]\Big(\frac{1}{|\mathbf{k}_1|\,|\mathbf{k}_2|\,(|\mathbf{k}_1|+|\mathbf{k}_2|)}\Big)\\
=&\frac{24\,(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2+24}{|\mathbf{k}_1|\,|\mathbf{k}_2|\,(|\mathbf{k}_1|+|\mathbf{k}_2|)^5}\\
&+\left[\frac{1}{|\mathbf{k}_1|^3|\mathbf{k}_2|^3(|\mathbf{k}_1|+|\mathbf{k}_2|)}+\frac{1}{|\mathbf{k}_1|^2|\mathbf{k}_2|^2(|\mathbf{k}_1|+|\mathbf{k}_2|)^3}\right]\Big(24\,(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2-8\Big)
\end{aligned}$$

and

$$\begin{aligned}
&\Big[\Delta_{\mathbf{k}_1}\otimes\Delta_{\mathbf{k}_2}+(\nabla_{\mathbf{k}_1}\cdot\nabla_{\mathbf{k}_2})^2\Big]\Big(\frac{1}{|\mathbf{k}_1|^2|\mathbf{k}_2|}\Big)\\
=&-4\left(\frac{1}{|\mathbf{k}_1|^3|\mathbf{k}_2|^4}+\frac{1}{|\mathbf{k}_1|^4|\mathbf{k}_2|^3}\right)+12\,(\mathbf{k}_1\cdot\mathbf{k}_2)^2\left(\frac{1}{|\mathbf{k}_1|^5|\mathbf{k}_2|^6}+\frac{1}{|\mathbf{k}_1|^6|\mathbf{k}_2|^5}\right).
\end{aligned}$$

Evaluating these two expressions at the point $(-\Lambda\hat{\mathbf{R}},-\Lambda\hat{\mathbf{R}})$ thus yields

$$\lim_{R\to\infty}(I_1(\mathbf{R})+I_2(\mathbf{R}))=\alpha_E^A(0)\alpha_E^B(0)\Big[4\frac{23}{2}\frac{1}{\Lambda^7}-8\frac{16}{\Lambda^7}\Big]=\frac{-82}{\Lambda^7}\alpha_E^A(0)\alpha_E^B(0),$$

and the assertion of the theorem follows by inspection of (6.5.1). □

Next we investigate the large $|\mathbf{R}|$-asymptotics of the term $F_8(\mathbf{R})$ (see 6.1.2). As we will show below using standard estimates for oscillatory integrals involving smooth functions, this term can only contribute to $V(\Lambda,\mathbf{R})$ at orders $1/R^k$ for $k\geq 8$.
From (6.1.2), recall the definition

$$\begin{aligned}
&F_8(\mathbf{R})\\
=&-\frac{1}{9\hbar^4}\int_{\mathbb{R}^3\times\mathbb{R}^3}d\mathbf{k}_1d\mathbf{k}_2|C(\mathbf{k}_1)|^2|C(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\times\Big[\hbar^4\omega(\mathbf{k}_1)^2\omega(\mathbf{k}_2)^2\,(2T_5(\mathbf{k}_1,\mathbf{k}_2)+T_6(\mathbf{k}_1,\mathbf{k}_2)) &(6.5.4)\\
&\quad+\hbar^4\omega(\mathbf{k}_1)^3\omega(\mathbf{k}_2)\,(T_4(\mathbf{k}_1,\mathbf{k}_2)-8T_5(\mathbf{k}_1,\mathbf{k}_1))\Big], &(6.5.5)
\end{aligned}$$

where

$$\begin{aligned}
T_4(\mathbf{k}_1,\mathbf{k}_2)=&\sum_{\alpha=1}^3\Big[\Big\langle(H_A+\hbar\omega(\mathbf{k}_1))^{-1}(H_A+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha}\alpha_E^B(\mathbf{k}_1)\\
&+\alpha_E^A(\mathbf{k}_1)\Big\langle(H_B+\hbar\omega(\mathbf{k}_1))^{-1}(H_B+\hbar\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_B^\alpha}\Big],\\
T_5(\mathbf{k}_1,\mathbf{k}_2)=&\sum_{\alpha,\beta=1}^3\Big\langle(\overline{H_A+H_B})^{-1}(H_A+\hbar(\omega(\mathbf{k}_1))^{-1}\otimes(H_B+\hbar(\omega(\mathbf{k}_2))^{-1}\Big\rangle_{\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta},
\end{aligned}$$

$$T_6(\mathbf{k}_1,\mathbf{k}_2) = \sum_{\alpha,\beta=1}^{3} \Big[\Big\langle (\overline{H_A + H_B})^{-1} \Big[(H_A + \hbar(\omega(\mathbf{k}_1))^{-1} (H_A + \hbar(\omega(\mathbf{k}_2))^{-1} + (H_B + \hbar(\omega(\mathbf{k}_1))^{-1} (H_B + \hbar(\omega(\mathbf{k}_2))^{-1} \Big] \Big\rangle_{\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta} \Big]$$

were defined in (3.0.6), (3.0.7) and (3.0.8), respectively. Define the functions

$$\begin{aligned}
\tilde{g}_1(\mathbf{k}_1,\mathbf{k}_2) &:= 2T_5(\mathbf{k}_1,\mathbf{k}_2) + T_6(\mathbf{k}_1,\mathbf{k}_2), \\
\tilde{g}_2(\mathbf{k}_1,\mathbf{k}_2) &:= T_4(\mathbf{k}_1,\mathbf{k}_2) - 8T_5(\mathbf{k}_1,\mathbf{k}_1), \\
g_1(\mathbf{k}_1,\mathbf{k}_2) &:= |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 \tilde{g}_1(\mathbf{k}_1,\mathbf{k}_2), \\
g_2(\mathbf{k}_1,\mathbf{k}_2) &:= |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 \tilde{g}_2(\mathbf{k}_1,\mathbf{k}_2).
\end{aligned}$$

Lemma 6.5.2 (Properties of $g_i(\mathbf{k}_1,\mathbf{k}_2)$)**.** *Let $k,l \in \mathbb{N}_0$. The functions*

$$(\xi_1,\xi_2) \mapsto \xi_1^k\, \xi_2^l\, g_i(\xi_1,\xi_2), \quad i = 1,2$$

are in $C^\infty((0,\infty)\times(0,\infty)) \cap C([0,\infty)\times[0,\infty))$. Furthermore, $\xi_1^k\, \xi_2^l\, g_i(\xi_1,\xi_2)$ and all their partial derivatives $\frac{\partial^{n+m}}{\partial \xi_1^n \xi_2^m}(\xi_1^k \xi_2^l\, g_i(\xi_1,\xi_2))$ are rapidly decaying. In particular,

$$\frac{\partial^{n+m}}{\partial \xi_1^n \xi_2^m}(\xi_1^k \xi_2^l\, g_i(\xi_1,\xi_2)) \in L^1((0,\infty)\times(0,\infty)) \cap L^\infty((0,\infty)\times(0,\infty),$$

$\lim_{\xi\to\infty} \frac{d^n}{d\xi^n}(\xi^m \varphi_{\alpha,\beta}) = 0$ *and* $\lim_{\xi\to 0} \frac{d^n}{d\xi^n}(\xi^m \varphi_{\alpha,\beta}) = 0$ *if* $n < m$.

Proof. The proof proceeds along the lines of the one of Lemma 5.5.14. The properties of the ultraviolet-cutoff ρ and the photonic dispersion relation $\omega(\mathbf{k})$ imply $g_i(\mathbf{k}_1,\mathbf{k}_2) = g_i(|\mathbf{k}_1|,|\mathbf{k}_2|)$, and the reduced resolvents $((H_{A,B} + \hbar\omega(\mathbf{k}))_{|\{\Psi^0_{A,B}\}^\perp})^{-1}$ exist for all $\mathbf{k} \in \mathbb{R}^3$, so the two functions g_i are well-defined on $[0,\infty)\times[0,\infty)$. Since $\rho \in \mathcal{S}(\mathbb{R}^3)$, the function $(\xi_1,\xi_2) \mapsto \xi_1^k\, \xi_2^l |\rho(\xi_1)|^2 |\rho(\xi_2)|^2$ is in $C^\infty((0,\infty)\times(0,\infty)) \cap C([0,\infty)\times[0,\infty))$, and all its partial derivatives are continuous on $[0,\infty)\times[0,\infty)$ and decay rapidly. As a consequence, it suffices to show that $\frac{\partial^{n+m}}{\partial \xi_1^n \xi_2^m} T_i(\xi_1,\xi_2))$, $i = 4,5,6$, and $\frac{\partial^n}{\partial \xi_1^n} T_5(\xi_1,\xi_1))$ exist and are bounded and continuous on $[0,\infty)\times[0,\infty)$ for all $n,m \geq 0$.
We first consider T_5. Recall the subspace $W = \overline{\{\Psi^0_A\}^\perp \widehat{\otimes} \{\Psi^0_B\}^\perp}$ and the proof of Lemma 5.5.14, where it was shown that

$$\overline{((H_A + \hbar\omega(\xi_1))^{-1} \otimes I)_{|W}} = \Big(\overline{(H_A \otimes I)_{|W}} + \hbar\omega(\xi) I_{|W} \Big)^{-1}$$

and

$$\overline{(I \otimes (H_B + \hbar\omega(\xi))^{-1})_{|W}} = \Big(\overline{(I \otimes H_B)_{|W}} + \hbar\omega(\xi) I_{|W} \Big)^{-1}.$$

Using the spectral resolution $E_B(\lambda)$ of the self-adjoint operator $\overline{(I \otimes H_B)_{|W}}$ and noting that by the proof of Lemma 4.2.5,

$$\operatorname{spec}(\overline{(I \otimes H_B)_{|W}}) = \operatorname{spec}(\overline{I_{|\{\Psi^0_A\}^\perp} \otimes H_{B|\{\Psi^0_B\}^\perp}}) = \operatorname{spec}(H_{B|\{\Psi^0_B\}^\perp}) \subset [\Delta_B, \infty),$$

and thus that the function $\lambda \mapsto \frac{1}{(\lambda+\hbar\omega(\xi))^{m+1}}$ on $\mathrm{spec}(\overline{(H_A \otimes I)_{|W}})$ is uniformly bounded in ξ for any $m \geq 0$, a standard result on the differentiation of parameter-dependent integrals yields

$$
\begin{aligned}
&\frac{\partial^m}{\partial \xi_2^m} T_5(\xi_1, \xi_2) \\
=&(-1)^m m! (\hbar c)^m \sum_{\alpha,\beta=1}^{3} \int_{\mathrm{spec}(\overline{(I\otimes H_B)_{|W}})} \frac{1}{(\lambda + \hbar c \xi_2)^{m+1}} d\Big\langle \overline{((H_A + \hbar(\omega(\xi_1))_{|\{\Psi_A^0\}^\perp})^{-1} \otimes I_{|\{\Psi_B^0\}^\perp})} \\
&\qquad \times (\overline{(H_A + H_B)_{|W}})^{-1} (\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) | E_B(\lambda) | \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \\
=&(-1)^m m! (\hbar c)^m \sum_{\alpha,\beta=1}^{3} \Big\langle \overline{((H_A + \hbar(\omega(\xi_1))_{\{\Psi_A^0\}^\perp})^{-1} \otimes I_{|\{\Psi_B^0\}^\perp})} (\overline{(H_A + H_B)_{|W}})^{-1} (\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \Big| \\
&\qquad \Big| \Big(\overline{(I \otimes H_B)_{|W}} + \hbar\omega(\xi_2) I_{|W} \Big)^{-(m+1)} | \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle \\
=&(-1)^m m! (\hbar c)^m \sum_{\alpha,\beta=1}^{3} \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | (\overline{(H_A + H_B)_{|W}})^{-1} ((H_A + \hbar\omega(\xi_1))_{|\{\Psi_A^0\}^\perp})^{-1} \\
&\qquad \times ((H_B + \hbar\omega(\xi_2))_{|\{\Psi_B^0\}^\perp})^{-(m+1)} | \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle.
\end{aligned}
$$

This is a (jointly) continuous function on $[0,\infty) \times [0,\infty)$ since the reduced resolvents $\xi \mapsto ((H_{A,B} + \hbar\omega(\xi))_{|\{\Psi_{A,B}^0\}^\perp})^{-1}$ are continuous on $[0,\infty)$. Applying the same argument again, this time using the spectral resolution of $\overline{(H_A \otimes I)_{|W}}$, yields

$$
\begin{aligned}
&\frac{\partial^{m+n}}{\partial \xi_1^n \partial \xi_2^m} T_5(\xi_1, \xi_2) \\
=&(-1)^{m+n} m! n! (\hbar c)^{m+n} \sum_{\alpha,\beta=1}^{3} \Big\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | (\overline{(H_A + H_B)_{|W}}))^{-1} ((H_A + \hbar\omega(\xi_1))_{|\{\Psi_A^0\}^\perp})^{-(n+1)} \\
&\qquad \times ((H_B + \hbar\omega(\xi_2))_{\{\Psi_B^0\}^\perp})^{-(m+1)} | \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta \Big\rangle,
\end{aligned}
$$

which, again by the continuity of the resolvents of $H_{A|\{\Psi_A^0\}^\perp}$ and $H_{B|\{\Psi_B^0\}^\perp}$, is a (jointly) continuous function on $[0,\infty) \times [0,\infty)$. To prove boundedness, we use the Cauchy-Schwarz inequality and the resolvent estimates

$$
\begin{aligned}
&\|((H_A + \hbar\omega(\xi))_{|\{\Psi_A^0\}^\perp})^{-(n+1)}\| \leq (1/\mathrm{dist}(-\hbar\omega(\xi), \mathrm{spec}(H_{A|\{\Psi_A^0\}^\perp}))^{n+1} \leq (1/\Delta_A)^{n+1}, \\
&\|((H_B + \hbar\omega(\xi))_{|\{\Psi_B^0\}^\perp})^{-(n+1)}\| \leq (1/\mathrm{dist}(-\hbar\omega(\xi), \mathrm{spec}(H_{B|\{\Psi_B^0\}^\perp}))^{n+1} \leq (1/\Delta_B)^{n+1}
\end{aligned}
$$

to conclude

$$
\begin{aligned}
&\Big| \frac{\partial^{m+n}}{\partial \xi_1^n \partial \xi_2^m} T_5(\xi_1, \xi) \Big| \\
\leq& m! n! (\hbar c)^{m+n} \frac{1}{\Delta_A^{n+1} \Delta_B^{m+1}} \Big(\sum_{\alpha=1}^{3} \|\mathbf{v}_A^\alpha\|^2 \Big) \Big(\sum_{\beta=1}^{3} \|\mathbf{v}_B^\beta\|^2 \Big) (\|\overline{(H_A + H_B)_{|W}})^{-1}\|.
\end{aligned}
$$

Since $T_5(\xi_1,\xi_1)$ can be viewed as the composition of $T_5(\xi_1,\xi_2)$ with the C^∞-map $\xi_1 \mapsto (\xi_1,\xi_1)$, the assertion on $\frac{\partial^n}{\partial\xi_1^n}T_5(\xi_1,\xi_1))$ also follows.
Next we turn to T_4. Using the same arguments as above, we find

$$\begin{aligned}
&\frac{d^n}{d\xi^n}\alpha_E^{A,B}(\xi)\\
=&(-1)^n n!(\hbar c)^n \sum_{\alpha=1}^{3}\Big\langle \mathbf{v}_{A,B}^{\alpha}|((H_{A,B}+\hbar c\xi)_{|\{\Psi_{A,B}^0\}^\perp})^{-(n+1)}|\mathbf{v}_{A,B}^{\alpha}\Big\rangle,\\
&\frac{\partial^{m+n}}{\partial\xi_1^n\partial\xi_2^m}\left[\sum_{\alpha=1}^{3}\Big\langle((H_{A,B}+\hbar c\xi_1)_{|\{\Psi_{A,B}^0\}^\perp})^{-1}((H_{A,B}+\hbar c\xi_2)_{|\{\Psi_{A,B}^0\}^\perp})^{-1}\Big\rangle_{\mathbf{v}_{A,B}^{\alpha}}\right]\\
=&(-1)^{m+n} m!n!(\hbar c)^{m+n}\sum_{\alpha=1}^{3}\Big\langle((H_{A,B}+\hbar c\xi_1)_{|\{\Psi_{A,B}^0\}^\perp})^{-(n+1)}\\
&\qquad\qquad\times((H_{A,B}+\hbar c\xi_2)_{|\{\Psi_{A,B}^0\}^\perp})^{-(m+1)}\Big\rangle_{\mathbf{v}_{A,B}^{\alpha}}
\end{aligned}$$

and the corresponding estimates

$$\begin{aligned}
&\Big|\frac{d^n}{d\xi^n}\alpha_E^{A,B}(\xi)\Big|\\
\leq& n!(\hbar c)^n\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_{A,B}^{\alpha}\|^2\right)\left(\frac{1}{\Delta_{A,B}}\right)^{n+1},\\
&\left|\frac{\partial^{m+n}}{\partial\xi_1^n\partial\xi_2^m}\left[\sum_{\alpha=1}^{3}\Big\langle((H_{A,B}+\hbar c\xi_1)_{|\{\Psi_{A,B}^0\}^\perp})^{-1}((H_{A,B}+\hbar c\xi_2)_{|\{\Psi_{A,B}^0\}^\perp})^{-1}\Big\rangle_{\mathbf{v}_{A,B}^{\alpha}}\right]\right|\\
\leq& m!n!(\hbar c)^{m+n}\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_{A,B}^{\alpha}\|^2\right)\left(\frac{1}{\Delta_{A,B}}\right)^{m+n+2}.
\end{aligned}$$

Now the Leibniz rule yields the existence and continuity of $\frac{\partial^{m+n}}{\partial\xi_1^n\partial\xi_2^m}T_4(\xi_1,\xi_2)$ for any $n,m\in\mathbb{N}_0$, together with the uniform estimate

$$\begin{aligned}
&\Big|\frac{\partial^{m+n}}{\partial\xi_1^n\partial\xi_2^m}T_4(\xi_1,\xi_2)\Big|\\
\leq&\sum_{s=0}^{n}\binom{n}{s}m!(n-s)!s!(\hbar c)^{m+n}\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_A^{\alpha}\|^2\right)\left(\sum_{\alpha=1}^{3}\|\mathbf{v}_B^{\alpha}\|^2\right)\\
&\times\left[\left(\frac{1}{\Delta_A}\right)^{m+n-s+2}\left(\frac{1}{\Delta_B}\right)^{s+1}+\left(\frac{1}{\Delta_A}\right)^{s+1}\left(\frac{1}{\Delta_B}\right)^{m+n-s+2}\right].
\end{aligned}$$

The assertion about $\frac{\partial^{n+m}}{\partial\xi_1^n\xi_2^m}T_6(\xi_1,\xi_2))$ is proven completely analogous. □

Having established the regularity properties of the functions $g_i(\mathbf{k}_1,\mathbf{k}_2)$ in the preceding lemma, we are in a position to analyze the large-$|\mathbf{R}|$-asymptotics of $F_8(\mathbf{R})$.

Lemma 6.5.3. *Assume the hypotheses of Theorem 3.0.6. Then*

$$\lim_{R\to\infty}\Big(R^k F_8(\mathbf{R})\Big)=0$$

for any $k<8$.

Proof. We first consider (6.5.4). Since $g_1(\mathbf{k}_1,\mathbf{k}_2)=g_1(|\mathbf{k}_1|,|\mathbf{k}_2|)$, we switch to 'double' polar coordinates on $\mathbb{R}^3\times\mathbb{R}^3$ and obtain

$$(6.5.4)=-\frac{(\hbar c)^2(4\pi)^2}{18}\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\,\xi_1^3\,\xi_2^3\,g_1(\xi_1,\xi_2)\,W(\xi_1,\xi_2),$$

where

$$\begin{aligned}
&W(\xi_1,\xi_2)\\
=&\frac{2}{R^2}\frac{\sin(\xi_1R)\sin(\xi_2R)}{\xi_1\xi_2}+\frac{2}{R^3}\left(\frac{\sin(\xi_1R)\cos(\xi_2R)}{\xi_1\xi_2^2}+\frac{\sin(\xi_2R)\cos(\xi_1R)}{\xi_1^2\xi_2}\right)\\
&-\frac{1}{R^4}\left(\frac{2\sin(\xi_1R)\sin(\xi_2R)}{\xi_1\xi_2^3}+\frac{2\sin(\xi_2R)\sin(\xi_1R)}{\xi_1^3\xi_2}-\frac{6\cos(\xi_1R)\cos(\xi_2R)}{\xi_1^2\xi_2^2}\right) &(6.5.6)\\
&-\frac{6}{R^5}\left(\frac{\cos(\xi_1R)\sin(\xi_2R)}{\xi_1^2\xi_2^3}+\frac{\cos(\xi_2R)\sin(\xi_1R)}{\xi_1^3\xi_2^2}\right) &(6.5.7)\\
&+\frac{6}{R^6}\frac{\sin(\xi_1R)\sin(\xi_2R)}{\xi_1^3\xi_2^3}. &(6.5.8)
\end{aligned}$$

Note that every term in (6.5.4) is of the form

$$C\frac{1}{R^s}\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\,\xi_1^k\,\xi_2^l\,f_1(\xi_1R)\,f_2(\xi_2R)\,g_1(\xi_1,\xi_2),$$

where $s,k,l\geq 0$, $s+k+l=6$ and either $f_i=\sin$ or $f_i=\cos$. But since by Lemma 6.5.2, g_1 satisfies the assumptions of Lemma A.4.2, we can estimate

$$\Big|\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\,\xi_1^k\,\xi_2^l\,f_1(\xi_1R)\,f_2(\xi_2R)\,g_1(\xi_1,\xi_2)\Big|\leq C\frac{1}{R^{k+l+2}},$$

which upon adding all terms yields

$$\big|(6.5.4)\big|\leq C\frac{1}{R^{s+k+l+2}}=C\frac{1}{R^8},$$

proving the assertion for the term (6.5.4).
Next we turn to the analysis of

$$(6.5.5)=\frac{(\hbar c)^2(4\pi)^2}{32}\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\,\xi_1^4\,\xi_2^2\,g_2(\xi_1,\xi_2)\,W(\xi_1,\xi_2),$$

which again consists of terms of the form

$$C\frac{1}{R^s}\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\,\xi_1^k\,\xi_2^l\,f_1(\xi_1R)\,f_2(\xi_2R)\,g_2(\xi_1,\xi_2),$$

with $s+k+l=6$, the only difference being that this time the case $l=-1$ occurs, namely in the first terms in (6.5.6) and (6.5.7) and in (6.5.8). In all these three contributions $f_2(\xi_2 R)=\sin(\xi_2 R)$, so that, using that g_2 satisfies the necessary assumptions according to Lemma 6.5.2, we can use Lemma A.4.2 again to find

$$\big|(6.5.5)\big| \leq C\frac{1}{R^{s+k+l+2}} = C\frac{1}{R^8},$$

finishing the proof. □

6.6 Asymptotic cancellation at order $1/R^6$

In this section we will investigate a crucial feature of the Van der Waals interaction in the context of matter coupled to the radiation field which concerns the lowest-order contribution (in $1/R$) from terms in the interaction potential $V(\Lambda,\mathbf{R})$ due solely to the static Coulomb potential Q_R. More precisely, we will show how this contribution, which decays like $1/R^6$, is counter-balanced by contributions originating from the quantized radiation field, resulting in the net $1/R^7$-decay asserted in Theorem 1.2.2.
As we will show below, there is no exact cancellation at any finite value of R, but an asymptotic one in the sense that the sum of the relevant contributions decays faster than any power of $1/R$. We will see that this lack of an exact cancellation is due to the presence of the spatial cutoff d we had to introduce to rigorously handle the multipole expansion of Q_R. To extract the large R-asymptotics, the scale d has to be coupled to R, and it is only in the limit $d\to\infty$ that the terms containing Q_R converge to cancel those originating purely from the field. However, this convergence will turn out to be superalgebraic, resulting in the fast $1/R$-decay mentioned above.
As mentioned in the introduction, for this to occur it is essential that Q_R is a smeared Coulomb potential. As will be discussed at the end of this section, if a non-smeared potential is used instead, the superalgebraic convergence cannot be expected to hold.
Recall the definitions

$$L(d)=\sum_{\alpha,\beta=1}^{3}\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|\Big((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\Big)^{-1}\Big|\chi\Omega_d(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)\Big\rangle_{L^2(\mathbb{R}^{3N})},$$

$$L(\infty)=\sum_{\alpha,\beta=1}^{3}\Big\langle \mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big|\Big((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\Big)^{-1}\Big|\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta\Big\rangle_{L^2(\mathbb{R}^{3N})}$$

and

$$\tilde{L}(d)=\sum_{\alpha,\beta=1}^{3}\Big\langle \chi\Omega_d(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)\Big|\Big((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\Big)^{-1}\Big|\chi\Omega_d(\mathbf{v}_A^\alpha\otimes\mathbf{v}_B^\beta)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}$$

from (5.5.30), (3.0.5) and Section 5.3.2, respectively. Combining Theorem 3.0.6, the results of Chapter 5, Theorem 6.5.1 and Lemma 6.5.3, we conclude that the only contributions to $V(\mathbf{R})$ which could decay slower than $1/R^7$ are

$$- \left\langle Q_2 \chi_{\Omega_d} (\Psi_A^0 \otimes \Psi_B^0) \right| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \left| Q_2 \chi_{\Omega_d} (\Psi_A^0 \otimes \Psi_B^0) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}, \quad (6.6.1)$$

$$M_6(\mathbf{R}, \sigma, d) = -\frac{2}{9} L(d) \int_{\Omega_\sigma \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-i(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}} \left(1 - (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2 \right)$$

(see the proof of Lemma 5.5.15), and

$$F_6(\mathbf{R}, \sigma) = (3.0.15) = -\frac{1}{9} L(\infty) \int_{\Omega_\sigma \times \Omega_\sigma} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 |\rho(\mathbf{k}_2)|^2 (1 + (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2) \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}},$$

where χ_{Ω_d} is the smooth characteristic function of the set

$$\Omega_d = \{ |\mathbf{x}_i| \leq d, \, i = 1, \ldots, N \} \subset \mathbb{R}^{3N}$$

introduced in Section 5.1. As was shown in Section 5.3, the London term (6.6.1) is the lowest-order contribution (in $1/R$) to the term

$$-\langle Q_R \Psi_0 | T^\sigma | Q_R \Psi_0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}},$$

obtained from a multipole expansion of Q_R on the restrained coordinate set Ω_d. It has an asymptotic decay like $1/R^6$ (see Lemma 5.1.3), and would arise as the second-order energy correction (in e) in the situation without radiation field, in which it determines the overall asymptotic $1/R^6$-decay of the Born-Oppenheimer potential energy surface, see the introduction and the literature cited there. By Lemma 5.3.2,

$$(6.6.1) = \frac{1}{9} \tilde{L}(d) \int_{\mathbb{R}^3 \times \mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2 \, |\rho(\mathbf{k}_2)|^2 \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1 + \mathbf{k}_2)\cdot \mathbf{R}} (\hat{\mathbf{k}}_1 \cdot \hat{\mathbf{k}}_2)^2, \quad (6.6.2)$$

and this representation, which puts (6.6.1), $M_6(\mathbf{R}\sigma, d)$ and $F_6(\mathbf{R}, \sigma)$ on an equal footing and exhibits their similar structure, is possible since Q_R is a smeared Coulomb potential. $F_6(\mathbf{R}, \sigma)$ is a term generated solely by the radiation field, and as such contains the infrared-cutoff σ, but is independent of the spatial cutoff d used in the multipole expansion. $M_6(\mathbf{R}\sigma, d)$ is a mixed term containing both Q_R and contributions from the quantized radiation field, and thus contains both d and σ. The quantity we will investigate in this section is

$$\begin{aligned}
&\tilde{I}_6(\mathbf{R}, \sigma, d) \\
&:= M_6(\mathbf{R}, \sigma, d) + F_6(\mathbf{R}, \sigma) \\
&\quad - \left\langle Q_2 \chi_{\Omega_d} (\Psi_A^0 \otimes \Psi_B^0) \right| \left((\overline{H_A + H_B})|_{\{\Psi_A^0 \otimes \Psi_B^0\}^\perp} \right)^{-1} \left| Q_2 \chi_{\Omega_d} (\Psi_A^0 \otimes \Psi_B^0) \right\rangle_{\mathcal{H}_A \otimes \mathcal{H}_B},
\end{aligned}$$

and by the above, we find

$$
\begin{aligned}
&\tilde{I}_6(\mathbf{R},\sigma,d)\\
=&\frac{2}{9}L(d)\Bigg[\bigg(-\int_{\Omega_\sigma} d\mathbf{k}_1|\rho(\mathbf{k}_1)|^2\mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}}\bigg)\bigg(\int_{\mathbb{R}^3} d\mathbf{k}_2|\rho(\mathbf{k}_2)|^2\mathrm{e}^{-i\mathbf{k}_2\cdot\mathbf{R}}\bigg)\\
&\qquad+\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2\mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&\qquad-\int_{B_{\sigma/c}(0)\times\mathbb{R}^3} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2\mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\Bigg]\\
&-\frac{1}{9}\tilde{L}(d)\int_{\mathbb{R}^6} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2\\
&-\frac{1}{9}L(\infty)\int_{\Omega_\sigma\times\Omega_\sigma} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}. \qquad (6.6.3)
\end{aligned}
$$

To exhibit the mechanism of cancellation more clearly, we rewrite (6.6.3) as

$$
\begin{aligned}
&-\frac{1}{9}L(\infty)\left(\int_{\mathbb{R}^3\times\mathbb{R}^3}-2\int_{B_{\sigma/c}(0)\times\mathbb{R}^3}-\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)}\right)\Big(d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2\\
&\qquad\qquad\times(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\Big)\\
&-\frac{1}{9}L(\infty)\left(\int_{\Omega_\sigma}|\rho(\mathbf{k})|^2\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\right)^2,
\end{aligned}
$$

where we have used the $(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)$-symmetry of the integrand. Note that all integrals exist due to the presence of the ultraviolet-cutoff and the remaining integrands being $L^1_{loc}(\mathbb{R}^6)$-functions. Plugging in this identity, we obtain

$$
\begin{aligned}
&\tilde{I}_6(\mathbf{R},\sigma,d)\\
=&\frac{1}{9}\Big(2L(d)-\tilde{L}(d)-L(\infty)\Big)\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2\mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&+\frac{2}{9}(L(\infty)-L(d))\int_{B_{\sigma/c}(0)\times\mathbb{R}^3} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2\mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\
&-\frac{2}{9}L(d)\left(\int_{\Omega_\sigma} d\mathbf{k}_1|\rho(\mathbf{k}_1)|^2\mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}}\right)\left(\int_{\mathbb{R}^3} d\mathbf{k}_2|\rho(\mathbf{k}_2)|^2\mathrm{e}^{-i\mathbf{k}_2\cdot\mathbf{R}}\right)\\
&-\frac{1}{9}L(\infty)\left(\int_{\Omega_\sigma} d\mathbf{k}|\rho(\mathbf{k})|^2\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\right)^2\\
&+\frac{1}{9}L(\infty)\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)} d\mathbf{k}_1d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}.
\end{aligned}
$$

Next we use the identity

$$
\begin{aligned}
\int_{\Omega_\sigma} d\mathbf{k}|\rho(\mathbf{k})|^2\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}&=\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}-\int_{\mathbb{R}^3\setminus\Omega_\sigma} d\mathbf{k}|\rho(\mathbf{k})|^2\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\\
&=(2\pi)^{3/2}\widehat{|\rho|^2}(\mathbf{R})-\int_{\mathbb{R}^3\setminus\Omega_\sigma} d\mathbf{k}|\rho(\mathbf{k})|^2\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}
\end{aligned}
$$

and obtain

$$\tilde{I}_6(\mathbf{R},\sigma,d) = I_6(\mathbf{R},\sigma,d) + I_6^{IR}(\mathbf{R},\sigma),$$

where we have defined

$$\begin{aligned}
&I_6(\mathbf{R},\sigma,d)\\
&:=\frac{1}{9}\left(2L(d)-\tilde{L}(d)-L(\infty)\right)\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2 \mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} && (6.6.4)\\
&\quad+\frac{2}{9}\big(L(\infty)-L(d)\big)\int_{B_{\sigma/c}(0)\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2 \mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} && (6.6.5)\\
&\quad-\frac{1}{9}\big(2L(d)+L(\infty)\big)\left((2\pi)^{3/2}\,\widehat{|\rho|^2}(\mathbf{R})\right)^2 && (6.6.6)\\
&\quad+\frac{2}{9}\big(L(d)+L(\infty)\big)(2\pi)^{3/2}\,\widehat{|\rho|^2}(\mathbf{R})\int_{B_{\sigma/c}(0)} d\mathbf{k}|\rho(\mathbf{k})|^2\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}, && (6.6.7)\\
&I_6^{IR}(\mathbf{R},\sigma)\\
&:=-\frac{1}{9}L(\infty)\left(\int_{B_{\sigma/c}(0)} d\mathbf{k}|\rho(\mathbf{k})|^2\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\right)^2 && (6.6.8)\\
&\quad+\frac{1}{9}L(\infty)\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2 \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}. && (6.6.9)
\end{aligned}$$

From this representation, the mechanism of the asymptotic cancellation, which will be made precise in the following proposition, can already be read off. The term $I_6^{IR}(\mathbf{R},\sigma)$, on whose $1/R$-decay we do not have information, vanishes as $\sigma\to 0$, which is typical for all the error terms of the infrared regularization we have encountered so far.
The terms (6.6.6) and (6.6.7) contain the Fourier transform of the Schwartz function $|\rho|^2$, and thus decay faster than any inverse power of R. Actually even more is true: by our assumptions on the form factor ψ_0 entering the smeared Coulomb potential and its relation to the ultraviolet-cutoff in the quantized radiation field, which is given by (2.1.10), we have

$$\widehat{|\rho|^2}(\cdot) = (2\pi)^{-3/2}\Lambda^3(\psi_0*\psi_0)(\Lambda\cdot).$$

Furthermore, $\operatorname{supp}\widehat{|\rho|^2}\subset B_{2/\Lambda}(0)$, so that (6.6.6) and (6.6.7) vanish for finite values of R, as soon as $R>2/\Lambda$.
The terms (6.6.4) and (6.6.5) contain the prefactors $2L(d)-\tilde{L}(d)-L(\infty)$ and $L(\infty)-L(d)$, respectively, which will turn out to be responsible for their superalgebraic $1/R$-decay. As we will see,

$$\lim_{d\to\infty} L(d) = \lim_{d\to\infty}\tilde{L}(d) = L(\infty),$$

and this convergence is exponentially fast, so that the $1/R$-decay ensues after coupling the scales via $d=R^{1/2}$.
However, in contrast to the vanishing of (6.6.6) and (6.6.7) at finite values of R, there will in general be no finite d_0 such that

$$L(d) = \tilde{L}(d) = L(\infty)$$

for any $d > d_0$, which is related to the fact that atomic ground state eigenfunctions do not have compact support (see e.g. [RS78]Theorem. XIII.57)
It may be of interest to remark that if all calculations had been performed on a purely formal level, that is

- using the non-smeared dipole operator H' (see (1.0.1)) instead of the interatomic Coulomb potential Q_R in the Hamiltonian,
- using no ultraviolet-cutoff, which corresponds to $\rho \equiv 1/(2\pi)^{3/2}$ and thus $\widehat{|\rho|^2} = 1/(2\pi)^{3/2}\delta_0$,
- using no infrared-cutoff,

then all the terms (6.6.4) through (6.6.7) would vanish for any non-zero value of R, which might explain the claim of an exact cancellation in some of the physics literature (see e.g. [Pow65]).

Proposition 6.6.1. *Assume the hypotheses of Theorem 3.0.6. Then there exist positive constants γ, C_1, C_2, C_3, C_4, independent of R, σ and d, (but depending on Λ via $L(d)$, $L(\infty)$ and the properties of $H_{A,B}$), such that*

$$|I_6(\mathbf{R},\sigma,d)| \leq C_1 \mathrm{e}^{-\gamma d}\Big[\frac{1}{R^3}\Big|\widehat{|\rho|^2}(\mathbf{R})\Big| + \frac{1}{R^6} + \Big(\frac{\sigma}{R}\Big)^3 \sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\Big] \tag{6.6.10}$$

$$+ C_2\left(\Big|\widehat{|\rho|^2}(\mathbf{R})\Big|\right)^2 + C_3\Big|\widehat{|\rho|^2}(\mathbf{R})\Big|\,\sigma^3, \tag{6.6.11}$$

$$\big|I_6^{IR}(\mathbf{R},\sigma)\big| \leq C_4\,\sigma^6\left(\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\right)^2. \tag{6.6.12}$$

In particular,

$$\lim_{\sigma\to 0}\Big(R^k I_6^{IR}(\mathbf{R},\sigma)\Big) = 0$$

for any $\mathbf{R}\in\mathbb{R}^3$, and

$$\lim_{R\to\infty}\Big(R^k I_6(\mathbf{R},\sigma,R^{1/2})\Big) = 0$$

for any $k \geq 0$ and any $\sigma > 0$. Furthermore,

$$\lim_{\sigma\to 0}\left(I_6(\mathbf{R},\sigma,d)\right)$$

exists for any $d \geq 0$ and satisfies the estimate

$$\Big|\lim_{\sigma\to 0}\left(I_6(\mathbf{R},\sigma,d)\right)\Big| \leq C_1\mathrm{e}^{-\gamma d}\Big(\frac{1}{R^3}\Big|\widehat{|\rho|^2}(\mathbf{R})\Big| + \frac{1}{R^6}\Big) + C_2\left(\Big|\widehat{|\rho|^2}(\mathbf{R})\Big|\right)^2. \tag{6.6.13}$$

In particular,

$$\lim_{R\to\infty}\Big(R^k\lim_{\sigma\to 0}\Big(I_6(\mathbf{R},\sigma,R^{1/2})\Big)\Big) = 0$$

for any $k \geq 0$.

Proof. We begin by establishing the exponential decay (in d) of $|L(d) - L(\infty)|$ and $|L(d) - \tilde{L}(d)|$. To this end, note that

$$\begin{aligned}
&|L(d) - L(\infty)| \\
&= \left| \sum_{\alpha,\beta=1}^{3} \left\langle \mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | (\overline{H_A + H_B})^{-1} | (\chi_{\Omega_d} - 1)(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \right\rangle_{L^2(\mathbb{R}^{3N})} \right| \\
&\leq \sum_{\alpha,\beta=1}^{3} \|\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N})} \|(\overline{H_A + H_B})^{-1}\| \|(1 - \chi_{\Omega_d})(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta)\|_{L^2(\mathbb{R}^{3N})}
\end{aligned}$$

and

$$\begin{aligned}
&|L(d) - \tilde{L}(d)| \\
&= \left| \sum_{\alpha,\beta=1}^{3} \left\langle (1 - \chi_{\Omega_d})\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta | (\overline{H_A + H_B})^{-1} | \chi_{\Omega_d}(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta) \right\rangle_{L^2(\mathbb{R}^{3N})} \right| \\
&\leq \sum_{\alpha,\beta=1}^{3} \|(1 - \chi_{\Omega_d})\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N})} \|(\overline{H_A + H_B})^{-1}\| \|\chi_{\Omega_d}(\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta)\|_{L^2(\mathbb{R}^{3N})} \\
&\leq \sum_{\alpha,\beta=1}^{3} \|(1 - \chi_{\Omega_d})\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N})} \|(\overline{H_A + H_B})^{-1}\| \|\mathbf{v}_A^\alpha \otimes \mathbf{v}_B^\beta\|_{L^2(\mathbb{R}^{3N})}.
\end{aligned}$$

The exponential decay of the atomic ground states Ψ_A^0 and Ψ_B^0 implies the existence of positive constants C'_A, C'_B, γ'_A and γ'_B such that

$$\begin{aligned}
|\mathbf{v}_A^\alpha(\mathbf{x}_1, \ldots, \mathbf{x}_{Z_A})| &\leq C'_A \, \mathrm{e}^{-\gamma'_A(|\mathbf{x}_1| + \cdots + |\mathbf{x}_{Z_A}|)}, \\
\left|\mathbf{v}_B^\beta(\mathbf{x}_{Z_A+1}, \ldots, \mathbf{x}_N)\right| &\leq C'_B \, \mathrm{e}^{-\gamma'_B(|\mathbf{x}_{Z_A+1}| + \cdots + |\mathbf{x}_N|)},
\end{aligned}$$

the right-hand sides being independent of α and β, which can be achieved by taking maxima and minima, see also the proof of Lemma 5.5.4. Noting that $\operatorname{supp}(1 - \chi_{\Omega_d}) \subset \mathbb{R}^{3N} \setminus \Omega_d$, we can employ Lemma A.3.1 to obtain positive constants C and γ (independent of $\mathbf{R}$, σ and d, but depending on Λ via smearing in H_A and H_B) such that

$$|L(\infty) - L(d)| \leq C \, \mathrm{e}^{-\gamma d} \tag{6.6.14}$$

and

$$\left|2L(d) - \tilde{L}(d)\right| \leq 2C \, \mathrm{e}^{-\gamma d}. \tag{6.6.15}$$

We proceed by deriving estimates for each of the integrals in (6.6.4) through (6.6.9). As far as the integral in (6.6.4) is concerned, recall the proof of Lemma 5.5.15, where it was

shown that

$$\begin{aligned}
&\Big|\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2 (\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2 \mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\Big| \\
\leq &\frac{3(2\pi)^{3/2}}{4\pi}\frac{\Lambda^3}{R^3}\widehat{|\rho|^2}(\mathbf{R})\Big|\Big\langle \frac{1}{|\mathbf{x}_2|^3}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\mathbf{x}_2)]\Big\rangle\Big| \\
&+\frac{3}{(4\pi)^2}\frac{\Lambda^6}{R^6}\sum_{\alpha,\beta=1}^{3}\Bigg[\Big|\Big\langle \frac{1}{|\mathbf{x}_1|^3}(\delta_{\alpha,\beta}-3\hat{\mathbf{x}}_1^\alpha\hat{\mathbf{x}}_1^\beta), \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\mathbf{x}_2)]\Big\rangle\Big| \\
&\qquad\times\Big|\Big\langle \frac{\mathbf{x}_2^\alpha\mathbf{x}_2^\beta}{|\mathbf{x}_2|^5}, \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\mathbf{x}_2)]\Big\rangle\Big|\Bigg],
\end{aligned} \tag{6.6.16}$$

where the terms in pointed brackets, which do not depend on d or σ, are convergent as $R\to\infty$. In particular, they can be bounded by constants which are independent of R, σ and d. By a similar argument and the estimate

$$\int_{B_{\sigma/c}(0)} d\mathbf{k}|\rho(\mathbf{k})|^2 \leq 4\pi(\sigma/c)^3 \sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2, \tag{6.6.17}$$

we conclude

$$\begin{aligned}
&\Big|\int_{B_{\sigma/c}(0)\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2\,|\rho(\mathbf{k}_2)|^2 (\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}})^2 \mathrm{e}^{-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\Big| \\
\leq &\left(\frac{\sigma}{c}\right)^3\left(\frac{\Lambda}{R}\right)^3\left(\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\right) \\
&\times\left(\sum_{\alpha,\beta=1}^{3}\Big|\Big\langle \frac{1}{|\mathbf{x}|^3}(\delta_{\alpha,\beta}-3\hat{\mathbf{x}}_\alpha\hat{\mathbf{x}}_\beta), \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\mathbf{x}_2)]\Big\rangle\Big|\right)
\end{aligned} \tag{6.6.18}$$

for the integral in (6.6.5), where again the terms $\langle \frac{1}{|\mathbf{x}|^3}(\delta_{\alpha,\beta}-3\hat{\mathbf{x}}_\alpha\hat{\mathbf{x}}_\beta), \tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\mathbf{x}_2)]\rangle$ are convergent as $R\to\infty$. Combining the inequalities (6.6.16), (6.6.18) with (6.6.14), (6.6.15) and defining a suitable constant C_1 establishes (6.6.10).
As regards (6.6.6) and (6.6.7), note that by dominated convergence, $L(d)\to L(\infty)$ as $d\to\infty$, so $L(d)$ can be bounded by a constant independent of d, $\mathbf{R}$ and σ. Note however that $L(d)$ and $L(\infty)$ depend on Λ via the smeared Coulomb potential in the atomic Hamiltonians $H_{A,B}$. Using (6.6.17) and defining suitable constants C_2 and C_3 proves (6.6.11).
Again using (6.6.17), we estimate the integral in (6.6.9) as follows.

$$\begin{aligned}
&\Big|\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2 (\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2 \mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\Big| \\
\leq &\int_{B_{\sigma/c}(0)\times B_{\sigma/c}(0)} d\mathbf{k}_1 d\mathbf{k}_2 |\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2 \underbrace{\Big|(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2\Big|}_{\leq 1} \\
\leq &\left(\int_{B_{\sigma/c}(0)} d\mathbf{k}|\rho(\mathbf{k})|^2\right)^2 \leq (4\pi)^2\left(\frac{\sigma}{c}\right)^6\left(\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\right)^2.
\end{aligned}$$

Finally, using estimate (6.6.17) also for the term (6.6.8) and choosing a suitable constant C_4 yields (6.6.12).
The existence of

$$\lim_{\sigma\to 0}\left(I_6(\mathbf{R},\sigma,d)\right)$$

follows from the fact the only σ-dependent terms in $I_6(\mathbf{R},\sigma,d)$, namely (6.6.5) and (6.6.7), converge to zero as $\sigma\to 0$ by dominated convergence. Finally, the estimate (6.6.13) follows from the estimates for the terms (6.6.4) and (6.6.6) already established above. □

6.6.1 Modification of the cancellation mechanism in the Pauli-Fierz model with proper Coulomb potential

We close this section by discussing how the mechanism of asymptotic cancellation of the $1/R^6$-contributions is modified if instead of the Hamiltonians $H(\mathbf{R})$, H^A and H^B (see Section 2.2), dipole-approximated versions of the Pauli-Fierz Hamiltonian (1.0.8) comprising an ultraviolet-cutoff magnetic vector potential, but a non-smeared Coulomb potential $\tilde{Q}$, are used. For the proof of Proposition 6.6.1, it was crucial to use the representation (6.6.2) of the London term as an integral over photon momenta involving the ultraviolet-cutoff ρ. This representation originates in the relation (2.1.10) between the form factor ψ and ρ, which is a characteristic of the quantized Abraham model. As we will see now, if such a representation is lacking, the sum of the $1/R^6$-contributions is still equal to zero asymptotically, but the superalgebraic decay asserted in Proposition 6.6.1 cannot be expected to hold any longer.
If the smeared interatomic Coulomb potential Q_R is replaced by its non-smeared version $\tilde{Q}_R$ (see (1.0.2) for the case of two electrons) and the corresponding contributions to the interaction potential $V(\Lambda,\mathbf{R})$ are subjected to the multipole-expansion by methods analogous to those used in Chapter 5, the London term (6.6.1) is to be replaced by

$$-\frac{1}{(4\pi)^2}\frac{1}{R^6}\Big\langle \chi_{\Omega_d}(\mathbf{v}_A\cdot(D\mathbf{v}_B))\big|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}|Q_2\chi_{\Omega_d}(\mathbf{v}_A\cdot(D\mathbf{v}_B))\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}, \tag{6.6.19}$$

where $D=1-3\hat{\mathbf{R}}\otimes\hat{\mathbf{R}}$. Using rotation invariance (Lemma 4.2.17), this is easily shown to equal

$$-\frac{2}{3}\frac{1}{(4\pi)^2}\frac{1}{R^6}\tilde{L}(d), \tag{6.6.20}$$

with $\tilde{L}(d)$ as above. Analogously, the term $M_6(\mathbf{R},\sigma,d)$ is to be replaced by

$$\begin{aligned}
&\tilde{M}_6(\mathbf{R},\sigma,d)\\
&:=-\frac{2}{R^3}\frac{1}{4\pi}\int_{\Omega_\sigma}d\mathbf{k}|\rho(\mathbf{k})|^2\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\\
&\qquad\qquad\Big\langle \mathbf{v}_A\cdot((1-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\mathbf{v}_B)\big|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}\big|\mathbf{v}_A\cdot(D\mathbf{v}_B)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&=\frac{2}{9}\frac{1}{4\pi}\frac{1}{R^3}L(d)\int_{\Omega_\sigma}d\mathbf{k}|\rho(\mathbf{k})|^2(1-3(\hat{\mathbf{k}}\cdot\hat{\mathbf{R}})^2)\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}.
\end{aligned}$$

On the other hand, the term $F_6(\mathbf{R},\sigma)$, which is generated purely by the radiation field, is left unaltered. By the same arguments as above ($\rho\in\mathcal{S}(\mathbb{R}^3)$, remaining integrand in

$L^1_{loc}(\mathbb{R}^3)$), the limits

$$\begin{aligned}\lim_{\sigma\to 0}\tilde{M}_6(\mathbf{R},\sigma,d) &= \frac{2}{9}\frac{1}{4\pi}\frac{1}{R^3}L(d)\int_{\mathbb{R}^3} d\mathbf{k}|\rho(\mathbf{k})|^2(1-3(\hat{\mathbf{k}}\cdot\hat{\mathbf{R}})^2)\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\\ &=: \tilde{M}_6(\mathbf{R},0,d),\\ \lim_{\sigma\to 0}F_6(\mathbf{R},\sigma) &= -\frac{1}{9}L(\infty)\int_{\mathbb{R}^3\times\mathbb{R}^3} d\mathbf{k}_1 d\mathbf{k}_2|\rho(\mathbf{k}_1)|^2|\rho(\mathbf{k}_2)|^2(1+(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)^2)\mathrm{e}^{-\mathrm{i}(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}}\\ &=: F_6(\mathbf{R},0)\end{aligned}$$

exist. Rewriting these expressions in distributional form (see Section 5.5.5) yields

$$\tilde{M}_6(\mathbf{R},0,d) = \frac{2}{9}\frac{1}{4\pi}\frac{1}{R^3}L(d)\left((2\pi)^{3/2}\widehat{|\rho|^2}(\mathbf{R})\right) \tag{6.6.21}$$

$$-\frac{2}{3}\frac{1}{4\pi}\frac{1}{R^6}L(d)\frac{\Lambda^3}{(2\pi)^{3/2}}\left\langle\widehat{(\mathbf{k}\cdot\mathbf{R})^2\frac{1}{|\mathbf{k}|^2}},\tau_{\Lambda\hat{\mathbf{R}}}[R^3(\psi_0*\psi_0)(R\cdot)]\right\rangle_{\mathcal{S}'(\mathbb{R}^3),\mathcal{S}(\mathbb{R}^3)}, \tag{6.6.22}$$

$$F_6(\mathbf{R},0) = -\frac{1}{9}L(\infty)\left((2\pi)^{3/2}\widehat{|\rho|^2}(\mathbf{R})\right)^2 \tag{6.6.23}$$

$$-\frac{1}{9}\frac{1}{(2\pi)^3}L(\infty)\frac{\Lambda^6}{R^6}\left\langle\widehat{(\mathbf{k}_1\cdot\mathbf{k}_2)^2\frac{1}{|\mathbf{k}_1|^2|\mathbf{k}_2|^2}},\tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}[\Psi_R]\right\rangle_{\mathcal{S}'(\mathbb{R}^6),\mathcal{S}(\mathbb{R}^6)}, \tag{6.6.24}$$

where $\Psi_R(\mathbf{k}_1,\mathbf{k}_2) := \left(R^3(\psi_0*\psi_0)(R\mathbf{k}_1)\right)\left(R^3(\psi_0*\psi_0)(R\mathbf{k}_2)\right)$. As was discussed in Section 5.5.5, rescaling the photon variables renders R the parameter of the Dirac sequences $R^3(\psi_0*\psi_0)(R\cdot)$ and Ψ_R. As mentioned above, we have $\operatorname{supp}\widehat{|\rho|^2}\subset B_{2/\Lambda}(0)$, and thus (6.6.21) and (6.6.23) vanish as soon as $R>2\Lambda$. Using the methods of Section 5.5.5, one readily shows convergence of the terms in pointed brackets, which leads to

$$\lim_{R\to\infty}\left(R^6\tilde{M}_6(\mathbf{R},0,R^{1/2})\right) = \frac{4}{3}\frac{1}{(4\pi)^2}L(\infty), \tag{6.6.25}$$

$$\lim_{R\to\infty}\left(R^6F_6(\mathbf{R},0)\right) = -\frac{2}{3}\frac{1}{(4\pi)^2}L(\infty). \tag{6.6.26}$$

Combining (6.6.20), (6.6.25) and (6.6.26), we find

$$\lim_{R\to\infty}\left(R^6\left((6.6.19)+\tilde{M}_6(\mathbf{R},0,R^{1/2})+F_6(\mathbf{R},0)\right)\right)=0, \tag{6.6.27}$$

so that the $1/R^6$-contributions indeed cancel asymptotically after the infrared-cutoff is removed. However, the crucial difference to our situation is that the convergence rate cannot be quantified in the same fashion. Recall that Proposition 6.6.1 asserts that in our model, this rate is superalgebraic. The reason we cannot draw the same conclusion in the non-smeared case is that for finite values of R, (6.6.20) is not directly comparable to (6.6.22) and (6.6.24) in the sense that it cannot be written as a (multiple of) a tempered distribution applied to a member of a Dirac sequence parametrized by R. Applying the formal identity

$$-\frac{1}{(2\pi)^3}\int d\mathbf{k}\frac{1}{|k|^2}(\mathbf{k}\otimes\mathbf{k})\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}} = -\frac{1}{4\pi R^3}(1-3|\hat{\mathbf{R}}\otimes\hat{\mathbf{R}}|) \tag{6.6.28}$$

to (6.6.19), we find that formally,

$$
\begin{aligned}
&\text{(6.6.19)}\\
&= -\frac{1}{(2\pi)^6}\frac{1}{9}\tilde{L}(d)\Big\langle (\hat{\mathbf{k}_1}\cdot\hat{\mathbf{k}_2})^2, (\mathrm{e}^{-i\mathbf{k}_1\cdot\mathbf{R}}1)\otimes(\mathrm{e}^{-i\mathbf{k}_2\cdot\mathbf{R}}1)\Big\rangle\\
&= -\frac{1}{(2\pi)^6}\frac{1}{9}\tilde{L}(d)\frac{\Lambda^6}{R^6}\Big\langle \widehat{\frac{(\mathbf{k}_1\cdot\mathbf{k}_2)^2}{|\mathbf{k}_1|^2|\mathbf{k}_2|^2}}, \tau_{(\Lambda\hat{\mathbf{R}},\Lambda\hat{\mathbf{R}})}[\hat{1}\otimes\hat{1}]\Big\rangle\\
&= -\frac{1}{(2\pi)^3}\frac{1}{9}\tilde{L}(d)\frac{\Lambda^6}{R^6}\Big\langle (\nabla_1\cdot\nabla_2)^2\widehat{\frac{1}{|\mathbf{k}_1|^2|\mathbf{k}_2|^2}}, \delta_{-\Lambda\hat{\mathbf{R}}}\otimes\delta_{-\Lambda\hat{\mathbf{R}}}\Big\rangle\\
&= -\frac{1}{(4\pi)^2}\frac{1}{9}\tilde{L}(d)\frac{\Lambda^6}{R^6}\Big\langle \frac{-3}{|\mathbf{k}_1|^3|\mathbf{k}_2|^3}+\frac{9(\mathbf{k}_1\cdot\mathbf{k}_2)^2}{|\mathbf{k}_1|^5|\mathbf{x}_2|^5}, \delta_{-\Lambda\hat{\mathbf{R}}}\otimes\delta_{-\Lambda\hat{\mathbf{R}}}\Big\rangle. \qquad (6.6.29)
\end{aligned}
$$

Although (6.6.28) can be made rigorous if it is understood to hold in the sense of distributions applied to test functions which are supported away from 0 (in that sense, $1/(2\pi)^{3/2}\,\widehat{(\mathbf{k}_\alpha\mathbf{k}_\beta/|\mathbf{k}|^2)} = 1/(4\pi|\mathbf{x}|^3)(\delta_{\alpha,\beta}-3\mathbf{x}_\alpha\mathbf{x}_\beta)$, see Lemma 5.5.10), the delta distribution $\delta_{-\Lambda\hat{\mathbf{R}}}\otimes\delta_{-\Lambda\hat{\mathbf{R}}}$ does not belong to this class. Nevertheless, (6.6.29) indicates that in order to prove that the convergence in (6.6.27) occurs faster than any inverse power of R, one would have to verify the analogous result for the convergence of the Dirac sequences $R^3(\psi_0*\psi_0)(R\cdot)$ and Ψ_R to the Delta distributions δ_0 and $\delta_0\otimes\delta_0$, respectively. To our knowledge, such a result cannot be expected to hold in general.

Chapter 7

Proof of Theorems 1.2.1, 1.2.2 and 1.2.3

In this final chapter we complete the proofs of the main results by combining all the results on the contributions to the interaction potential $V(\Lambda, \mathbf{R})$ that have been established so far.

Proof. We first prove Theorems 2.8.3 and 2.8.4. By Theorem 3.0.6 and Theorem 5.4.1 (with R_0 and d chosen appropriately),

$$\begin{aligned} V_1^\sigma(\Lambda, \mathbf{R}) =& V_3^\sigma(\Lambda, \mathbf{R}) = 0, \\ V_2^\sigma(\Lambda, \mathbf{R}) =& \langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}, \end{aligned}$$

and for any $L \in \mathbb{N}$, $L \geq 2$ (to be chosen later) there is a decomposition

$$\begin{aligned} & V_4^\sigma(\Lambda, \mathbf{R}) \\ =& - \langle Q_R \Psi_0 | T^\sigma | Q_R \Psi_0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B \otimes \mathcal{F}} \\ & + M_6(\mathbf{R}, \sigma, d) + M_7(\mathbf{R}, d) + M_{89}^L(\mathbf{R}, d) \\ & + M_{IN,ERR}^L(\mathbf{R}, d) + M_{OUT}(\mathbf{R}, \sigma, d) + M_{IR}^L(\mathbf{R}, \sigma, d) \\ & - \langle \Psi_0 | Q_R | \Psi_0 \rangle \Big(\| T_A^\sigma H'_{\sigma,A}(\Psi_A^0 \otimes \Omega) \|^2_{\mathcal{H}_A \otimes \mathcal{F}} + \| T_B^\sigma H'_{\sigma,B}(\Psi_B^0 \otimes \Omega) \|^2_{\mathcal{H}_B \otimes \mathcal{F}} \Big) \\ & + F_6(\mathbf{R}, \sigma) + F_7(\mathbf{R}, \sigma) + F_8(\mathbf{R}, \sigma). \end{aligned}$$

Recall that $V_2^\sigma(\Lambda, \mathbf{R})$ is independent of σ, so trivially

$$V_2(\Lambda, \mathbf{R}) := \lim_{\sigma \to 0} V_2^\sigma(\Lambda, \mathbf{R}) = \langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{\mathcal{H}_A \otimes \mathcal{H}_B}.$$

By Lemma 5.2.2,

$$\lim_{R \to \infty} \Big(R^k V_2(\Lambda, \mathbf{R}) \Big) = \lim_{R \to \infty} \Big(R^k \langle \Psi_A^0 \otimes \Psi_B^0 | Q_R | \Psi_A^0 \otimes \Psi_B^0 \rangle_{L^2(\mathbb{R}^{3(N)})} \Big) = 0$$

for any $k \geq 0$. Recalling the definitions of $I_6(\mathbf{R}, \sigma, d)$, $I_6^{IR}(\mathbf{R}, \sigma)$ from Proposition 6.6.1 and those of $F_7(\mathbf{R})$, $F_8(\mathbf{R})$ from Section 6.1 and defining

$$F_{IR}(\mathbf{R}, \sigma) := \left(F_7(\mathbf{R}, \sigma) - F_7(\mathbf{R}) \right) + \left(F_8(\mathbf{R}, \sigma) - F_8(\mathbf{R}) \right),$$

$V_4^\sigma(\Lambda, \mathbf{R})$ can be rewritten as

$$
\begin{aligned}
&V_4^\sigma(\Lambda, \mathbf{R}) \\
=&M_7(\mathbf{R}, d) + F_7(\mathbf{R}) && (7.0.1)\\
&+ F_8(\mathbf{R}) + M_{89}^L(\mathbf{R}, d) + M_{IN,ERR}^L(\mathbf{R}, d) && (7.0.2)\\
&+\Bigg[\Big\langle Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}\Big|Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\\
&\qquad-\Big\langle Q_R(\Psi_A^0\otimes\Psi_B^0)\Big|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^\perp}\right)^{-1}\Big|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle_{\mathcal{H}_A\otimes\mathcal{H}_B}\Bigg] && (7.0.3)\\
&+ M_{OUT}(\mathbf{R}, \sigma, d) + I_6(\mathbf{R}, \sigma, d) && (7.0.4)\\
&- \langle\Psi_0|Q_R|\Psi_0\rangle\Big[\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2_{\mathcal{H}_A\otimes\mathcal{F}} + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2_{\mathcal{H}_B\otimes\mathcal{F}}\Big] && (7.0.5)\\
&+ I_6^{IR}(\mathbf{R}, \sigma) + M_{IR}^L(\mathbf{R}, \sigma, d) + F_{IR}(\mathbf{R}, \sigma). && (7.0.6)
\end{aligned}
$$

The σ-dependent contributions are (7.0.4), (7.0.5) and (7.0.6). Combining Proposition 6.6.1, Theorem 5.4.1 and Lemmas 6.1.1, 6.1.2 yields

$$
\begin{aligned}
&\left|I_6^{IR}(\mathbf{R}, \sigma) + M_{IR}^L(\mathbf{R}, \sigma, d) + F_{IR}(\mathbf{R}, \sigma)\right|\\
\leq&\Bigg[\sum_{l\geq 4, l \text{ even}}^{L}\left(\frac{3}{2R}\right)^{l+1} C_1(l)\left(\frac{\sigma}{c}\right)^3 + \sum_{l\geq 2, l \text{ even}}^{L}\left(\frac{3}{2R}\right)^{l+1} C_2(l)\left(\frac{\sigma}{c}\right)^4\\
&+\sum_{i=2}^{4} C_{i,7-i}\,\sigma^i\Lambda^{7-i} + C_{3,5}\,\sigma^3\Lambda^5 + C_{4,4}\,\sigma^4\Lambda^4 + C_{5,3}\,\sigma^5\Lambda^3\Bigg]\left(\sup_{s\in[0,\sigma/(c\Lambda)]}|\rho_0(s)|^2\right)\\
&+\left(C_6\sigma^6 + C_7\sigma^7 + C_8\sigma^8\right)\left(\sup_{s\in[0,\sigma/(\Lambda c)]}|\rho_0(s)|^2\right)^2
\end{aligned}
$$

for positive constants $C_1(l)$, $C_2(l)$, $C_{i,7-i}$, $C_{3,5}$, $C_{4,4}$ $C_{5,3}$, C_6, C_7, C_8, which are independent of $\mathbf{R}$, σ and d (but depend on the ultraviolet-cutoff Λ via properties of the atomic Hamiltonians H_A and H_B). This estimate immediately implies

$$\lim_{\sigma\to 0}\left(I_6^{IR}(\mathbf{R}, \sigma) + M_{IR}^L(\mathbf{R}, \sigma, d) + F_{IR}(\mathbf{R}, \sigma)\right) = 0.$$

By Lemma 5.2.3,

$$
\begin{aligned}
&\lim_{\sigma\to 0}\Big(\langle\Psi_0|Q_R|\Psi_0\rangle\Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big)\Big)\\
=&\langle\Psi_0|Q_R|\Psi_0\rangle\lim_{\sigma\to 0}\Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big)
\end{aligned}
$$

exists, and

$$\lim_{R\to\infty}\left(\lim_{\sigma\to 0}\left(R^k\langle\Psi_0|Q_R|\Psi_0\rangle\Big(\|T_A^\sigma H'_{\sigma,A}(\Psi_A^0\otimes\Omega)\|^2 + \|T_B^\sigma H'_{\sigma,B}(\Psi_B^0\otimes\Omega)\|^2\Big)\right)\right) = 0.$$

for any $k \geq 0$. The existence of

$$\lim_{\sigma\to 0}\left(I_6(\mathbf{R}, \sigma, d)\right)$$

and

$$\lim_{\sigma\to 0}\left(M_{OUT}(\mathbf{R},\sigma,d)\right)$$

is implied by Proposition 6.6.1 and 5.4.1, thus finishing the proof of Theorem 2.8.3.
To analyze the large-R asymptotics of

$$V_4(\Lambda,\mathbf{R})=\lim_{\sigma\to 0}(V_4^{\sigma}(\Lambda,\mathbf{R}))=\lim_{\sigma\to 0}\Big((7.0.1)+(7.0.2)+(7.0.3)+(7.0.4)\Big),$$

we couple the parameter d, which describes the spatial cutoff scale of the multipole expansion of the interatomic Coulomb potential, to the interatomic distance R by choosing $d:=R^{1/2}$. Note that this is consistent with the assumption $d\leq R/4$ as soon as $R\geq 16$. Proposition 6.6.1 establishes

$$\lim_{R\to\infty}\left(\lim_{\sigma\to 0}R^k\left(I_6(\mathbf{R},\sigma,R^{1/2})\right)\right)=0$$

for any $k\geq 0$, and by Theorem 5.4.1, there exist positive constants and C and γ, independent of σ, $\mathbf{R}$, d (but depending on Λ via $\rho(\mathbf{k}/\Lambda)$ and the properties of $H_{A,B}$), such that

$$\lim_{\sigma\to 0}\left(M_{OUT}(\mathbf{R},\sigma,d)\right)\leq C(1+1/R)\mathrm{e}^{-\gamma d}.$$

Our choice $d=R^{1/2}$ now implies

$$\lim_{R\to\infty}\left(\lim_{\sigma\to 0}\left(M_{OUT}(\mathbf{R},\sigma,R^{1/2})\right)\right)=0.$$

The terms left to be discussed ((7.0.1) through (7.0.3)) are all independent of σ, so that their $(\sigma\to 0)$-limits exist trivially.
As concerns $M^L_{IN,ERR}(\mathbf{R},d)$ and (7.0.3), we employ Theorem 5.4.1 and Lemma 5.3.1 to obtain

$$\begin{aligned}
&\Big|M^L_{IN,ERR}(\mathbf{R},d)+\Big\langle Q_R(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^{\perp}}\right)^{-1}|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&-\Big\langle Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^{\perp}}\right)^{-1}|Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Big|\\
\leq& C_1\Big[\frac{1}{R^2}\left(\frac{4(d+1)}{R}\right)^{2L+2}+\sum_{l=2}^{L}\frac{1}{R^2}\left(\frac{4(d+1)}{R}\right)^{L+1+l}\Big]+C_2\mathrm{e}^{-\gamma d}\\
&+\mathcal{O}(1/R^8)+\mathcal{O}\left(\frac{1}{|\mathbf{R}|}\left(\frac{4(d+1)}{|\mathbf{R}|}\right)^{L+1}\right),
\end{aligned}$$

where C_1, C_2 and γ are positive constants independent of R and d (but depending on Λ via properties of $H_{A,B}$), and the coefficients of the higher-order terms are independent of d, R and σ (but depend on Λ via $\rho_0(\mathbf{k}/\Lambda)$ and properties of $H_{A,B}$). Recalling the choice $d=R^{1/2}$ and choosing L large enough yields

$$\begin{aligned}
\lim_{R\to\infty}\Bigg[R^k\Bigg(&M^L_{IN,ERR}(\mathbf{R},d)+\Big\langle Q_R(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^{\perp}}\right)^{-1}|Q_R(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\\
&-\Big\langle Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)|\left((\overline{H_A+H_B})|_{\{\Psi_A^0\otimes\Psi_B^0\}^{\perp}}\right)^{-1}|Q_2\chi_{\Omega_d}(\Psi_A^0\otimes\Psi_B^0)\Big\rangle\Bigg)\Bigg]=0
\end{aligned}$$

for any $0 \geq k < 8$.
Theorem 5.4.1 and Lemma 6.5.3 imply that

$$\lim_{R\to\infty} (R^k\, F_8(\mathbf{R}) + M^L_{89}(\mathbf{R}, R^{1/2}) = 0$$

for any $k < 8$. Finally, combining Theorem 5.4.1 and Theorem 6.5.1, we find

$$\begin{aligned}
&\lim_{R\to\infty} \Big(R^7 \Big(M_7(\mathbf{R}, R^{1/2}) + F_7(\mathbf{R})\Big)\Big) \\
&= -\frac{32}{9}\frac{\hbar c}{(2\pi)^3}\alpha^A_E(0)\,\alpha^B_E(0) + \frac{41}{2}\frac{1}{9}\frac{\hbar c}{(2\pi)^3}\alpha^A_E(0)\,\alpha^B_E(0) \\
&= -\frac{23}{2}\frac{1}{9}\frac{\hbar c}{(2\pi)^3}\alpha^A_E(0)\,\alpha^B_E(0),
\end{aligned}$$

finishing the proof of Theorem 2.8.4.
We prove the assertion of Theorem 2.8.5 only for $\alpha^A_E(\mathbf{k})$, the other case being completely analogous. To simplify notation, put $\mathbf{q} := \sum_{i=1}^{Z_A} \mathbf{x}_i$, $\Psi(\Lambda) := \Psi^0_A$, $N := Z_A$, $H_N := \tilde{H}_A$ $H_N(\Lambda) := H_A$, and denote the ground state of H_N by Ψ. Using this, we estimate

$$\begin{aligned}
&\big|\alpha^A_E(\mathbf{k}) - \tilde{\alpha}^A_E(\mathbf{k})\big| \\
&= \left|\sum_{\alpha=1}^{3}\Big[\Big\langle \mathbf{q}^\alpha\Psi(\Lambda)|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1}|\mathbf{q}^\alpha\Psi(\Lambda)\Big\rangle - \Big\langle \mathbf{q}^\alpha\Psi|(H_N+\hbar\omega(\mathbf{k}))^{-1}|\mathbf{q}^\alpha\Psi\Big\rangle\Big]\right| \\
&= \Big|\sum_{\alpha=1}^{3}\Big[\Big\langle \mathbf{q}^\alpha\Psi(\Lambda)|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1}\mathbf{q}^\alpha\Psi(\Lambda) - (H_N+\hbar\omega(\mathbf{k}))^{-1}\mathbf{q}^\alpha\Psi\Big\rangle \\
&\qquad + \Big\langle \mathbf{q}^\alpha\Psi(\Lambda) - \mathbf{q}^\alpha\Psi|(H_N+\hbar\omega(\mathbf{k}))^{-1}\mathbf{q}^\alpha\Psi\Big\rangle\Big]\Big| \\
&\leq \sum_{\alpha=1}^{3}\Big[\Big|\Big\langle \mathbf{q}^\alpha\Psi(\Lambda)|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1}(\mathbf{q}^\alpha\Psi(\Lambda) - \mathbf{q}^\alpha\Psi)\Big\rangle\Big| \\
&\qquad + \Big|\Big\langle \mathbf{q}^\alpha\Psi(\Lambda)|\left((H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1} - (H_N+\hbar\omega(\mathbf{k}))^{-1}\right)\mathbf{q}^\alpha\Psi\Big\rangle\Big| \\
&\qquad + \|\mathbf{q}^\alpha\Psi(\Lambda) - \mathbf{q}^\alpha\Psi\|\,\|(H_N+\hbar\omega(\mathbf{k}))^{-1}\mathbf{q}^\alpha\Psi\|\Big] \\
&\leq \sum_{\alpha=1}^{3}\Big[\|\mathbf{q}^\alpha\Psi(\Lambda)\|\,\|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1}\|\,\|\mathbf{q}^\alpha(\Psi(\Lambda)-\Psi)\| \\
&\qquad + \|\mathbf{q}^\alpha\Psi(\Lambda)\|\,\|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1} - (H_N+\hbar\omega(\mathbf{k}))^{-1}\|\,\|\mathbf{q}^\alpha\Psi\| \\
&\qquad + \|\mathbf{q}^\alpha(\Psi(\Lambda)-\Psi)\|\,\|(H_N+\hbar\omega(\mathbf{k}))^{-1}\mathbf{q}^\alpha\Psi\|\Big]. \qquad (7.0.7)
\end{aligned}$$

As was established in the proof of Proposition 2.5.2, $H_N(\Lambda)$ converges to H_N in norm resolvent sense as $\Lambda \to \infty$. This implies that $\|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1}\|$ is uniformly bounded in Λ, and that

$$\|(H_N(\Lambda)+\hbar\omega(\mathbf{k}))^{-1} - (H_N+\hbar\omega(\mathbf{k}))^{-1}\| \to 0$$

as $\Lambda \to \infty$. Of course, the norm resolvent convergence also implies

$$\|\Psi(\Lambda) - \Psi\| \underset{\Lambda\to\infty}{\longrightarrow} 0, \qquad (7.0.8)$$

but this is not sufficient in order to handle the terms $\|\mathbf{q}^\alpha \Psi(\Lambda)\|$ and $\|\mathbf{q}^\alpha(\Psi(\Lambda)-\Psi)\|$: these norms are only finite due to the exponential decay of Ψ and $\Psi(\Lambda)$, and the decay of the latter a priori depends on Λ. However, as we will show, the norm resolvent convergence implies a certain uniformity of the decay of the corresponding one-particle densities, which will turn out to be sufficient to prove the convergence of the above terms. First note that due to $\mathbf{q}^\alpha$ being a sum of one-body operators, we have

$$\|\mathbf{q}^\alpha(\Psi(\Lambda)-\Psi)\| \leq \sum_{i=1}^{N} \|\mathbf{x}_i^\alpha(\Psi(\Lambda)-\Psi)\|. \tag{7.0.9}$$

Putting $\varphi(\Lambda) := \Psi(\Lambda) - \Psi$ and using the antisymmetry of φ and the definition of the one-body density matrix yields

$$\begin{aligned}\|\mathbf{x}_i^\alpha \varphi(\Lambda)\|^2 =& \int_{\mathbb{R}^3} d\mathbf{x}_i |\mathbf{x}_i^\alpha|^2 \int_{\mathbb{R}^{3N-3}} |\varphi(\Lambda)|^2(\mathbf{x}_1,\ldots,\mathbf{x}_N)\widehat{d\mathbf{x}_i} \\ =& \frac{1}{N}\int_{\mathbb{R}^3} |\mathbf{x}^\alpha|^2 \rho_{\varphi(\Lambda)}(\mathbf{x})d\mathbf{x}\end{aligned} \tag{7.0.10}$$

for any $i \in \{1,\ldots,N\}$, where we have used the notation

$$\widehat{d\mathbf{x}_i} = d\mathbf{x}_1 \ldots d\mathbf{x}_{i-1} d\mathbf{x}_{i+1} \ldots d\mathbf{x}_N.$$

The convergence (7.0.8) implies

$$\rho_{\varphi(\Lambda)} \underset{\Lambda\to\infty}{\to} 0$$

in $L^1(\mathbb{R}^3)$, and we have the pointwise estimate

$$\begin{aligned}\rho_{\varphi(\Lambda)}(\mathbf{x}) =& N\int_{\mathbb{R}^{3N-3}} |\Psi(\Lambda)-\Psi|^2(\mathbf{x},\mathbf{x}_2,\ldots,\mathbf{x}_N)\widehat{d\mathbf{x}} \\ \leq& N\int_{\mathbb{R}^{3N-3}} \left(|\Psi(\Lambda)|+|\Psi|\right)^2(\mathbf{x},\mathbf{x}_2,\ldots,\mathbf{x}_N)\widehat{d\mathbf{x}} \\ \leq& 2N\int_{\mathbb{R}^{3N-3}} \left(|\Psi(\Lambda)|^2+|\Psi|^2\right)(\mathbf{x},\mathbf{x}_2,\ldots,\mathbf{x}_N)\widehat{d\mathbf{x}} \\ =& 2N\left(\rho_{\Psi(\Lambda)}(\mathbf{x})+\rho_\Psi(\mathbf{x})\right)\end{aligned}$$

for almost all $\mathbf{x} \in \mathbb{R}^3$. After a slight modification to allow for smeared Coulomb potentials, a result from [Fri] shows that for a given $\varepsilon > 0$ and any $I > 0$, there exist positive constants $C_1(\Lambda)$ (depending on Λ, ε, I and N and γ'), C_2 (depending on ε, I, N) such that

$$\rho_{\Psi(\Lambda)}(\mathbf{x}) \leq C_1(\Lambda)\,\mathrm{e}^{-\gamma'\sqrt{\Sigma_N(\Lambda)-E_N(\Lambda)-\varepsilon}|\mathbf{x}|}, \qquad \rho_\Psi(\mathbf{x}) \leq C_2\,\mathrm{e}^{-\gamma'\sqrt{\Sigma_N-E_N-\varepsilon}|\mathbf{x}|}. \tag{7.0.11}$$

Here $E_N(\Lambda)$ and E_N denote the smallest eigenvalues of $H_N(\Lambda)$ and H_N, respectively, and $\Sigma_N(\Lambda)$ and Σ_N are the respective edges of the essential spectra. By Zhislin's theorem (in a version which allows for smeared Coulomb potentials, as can be obtained by a slight modification of the methods in [Fri03], for instance), $\Sigma_N(\Lambda) = E_{N-1}(\Lambda)$, $\Sigma_N = E_{N-1}$, where $E_{N-1}(\Lambda)$ and $\Sigma_N = E_{N-1}$ are the lowest eigenvalues of $H_{N-1}(\Lambda)$ and H_{N-1}. The norm resolvent convergence of $H_N(\Lambda)$ to H_N (which was established in the proof of Proposition 2.5.2 and which holds for any $N \geq 1$) now implies that for $\Lambda \geq \Lambda_0 > 0$, the quantity

$$\Sigma_N(\Lambda) - E_N(\Lambda) = E_{N-1}(\Lambda) - E_N(\Lambda)$$

stays in a bounded interval $(a, b) \subset \mathbb{R}$ with $0 < a \leq b$. In particular,

$$0 < \inf_{\Lambda \geq \Lambda_0} (\Sigma_N(\Lambda) - E_N(\Lambda)) < \infty.$$

Furthermore, inspection of the proof of (7.0.11) in [Fri] shows that

$$0 \leq \sup_{\Lambda \geq \Lambda_0} C_1(\Lambda) < \infty.$$

Using this, we conclude

$$\rho_{\varphi(\Lambda)}(\mathbf{x}) \leq 2N \left(\rho_{\Psi(\Lambda)}(\mathbf{x}) + \rho_{\Psi}(\mathbf{x}) \right) \leq C \mathrm{e}^{-\gamma |\mathbf{x}|},$$

where we have set $C := 4N \max\{ \sup_{\Lambda \geq \Lambda_0} C_1(\Lambda), C_2 \}$ and

$$\gamma := \gamma' \sqrt{\min\{ \inf_{\Lambda \geq \Lambda_0} (\Sigma_N(\Lambda) - E_N(\Lambda)), \Sigma_N - E_N \} - \varepsilon}.$$

In particular, C and γ are independent of Λ. Using the last estimate, we find

$$|\mathbf{x}^{\alpha}|^2 \rho_{\varphi(\Lambda)}(\mathbf{x}) \leq C |\mathbf{x}^{\alpha}|^2 \mathrm{e}^{-\gamma |\mathbf{x}|},$$

which shows that the sequence $|\cdot^{\alpha}|^2 \rho_{\varphi(\Lambda)}(\cdot)$ is uniformly dominated by the $L^1(\mathbb{R}^3)$-function $|\cdot^{\alpha}|^2 \mathrm{e}^{-\gamma |\cdot|}$. Since also $\rho_{\varphi(\Lambda)} \to 0$ in $L^1(\mathbb{R}^3)$, we can extract a subsequence ρ_{Λ_k} which converges to zero pointwise. The dominated convergence theorem then yields

$$|\cdot^{\alpha}|^2 \rho_{\varphi(\Lambda_k)}(\cdot) \underset{k \to \infty}{\to} 0$$

in $L^1(\mathbb{R}^3)$, and the subsequence principle implies

$$\| |\cdot^{\alpha}|^2 \rho_{\varphi(\Lambda)}(\cdot) \|_{L^1(\mathbb{R}^3)} \underset{\Lambda \to \infty}{\to} 0.$$

Now (7.0.9) and (7.0.10) yield

$$\| \mathbf{q}^{\alpha} (\Psi(\Lambda) - \Psi) \|_{L^2(\mathbb{R}^{3N})} \underset{\Lambda \to \infty}{\to} 0.$$

In particular the sequence $\mathbf{q}^{\alpha} \Psi(\Lambda)$ is uniformly bounded with respect to Λ, so that in light of (7.0.7), we finally obtain

$$\lim_{\Lambda \to \infty} \alpha_E^A(\mathbf{k}) = \tilde{\alpha}_E^A(\mathbf{k}),$$

finishing the proof of Theorem 2.8.5. □

Chapter 8

Remarks and outlook

As we have seen, the mathematical investigation of retarded Van der Waals interactions using perturbation theory required different mathematical methods, even in the context of the simplified model $V(\Lambda, \mathbf{R})$ used by us in the present work. These came into play at different levels of the analysis. The derivation and simplification of the coefficients in the perturbation expansions mainly used concepts from operator theory and relied on symmetries of the operators and ground states involved. On the other hand, analyzing the terms containing the interatomic Coulomb potential and establishing the cancellation of the $1/R^6$-contributions by terms originating from the radiation field exploited exponential localization of eigenfunctions, results from the spectral theory of the Laplacian and PDE estimates. Finally, the asymptotic analysis of the terms that were responsibly for the $1/R^7$-contribution required methods from harmonic analysis. Being singular oscillatory integrals that formally diverge as $R \to \infty$, these terms are fundamentally different mathematically from their counterpart, the London term (1.0.3), which also arises in the perturbative analysis of long-range interactions not involving the radiation field. This fundamental difference is due to the different nature of Schrödinger quantum mechanics and quantum electrodynamics, and the formal divergence of the integrals can be seen as touching on the general issue of divergence phenomena in quantum electrodynamics. Against this backdrop, the asymptotic interaction coefficient c_7 can be seen as an example of an (at least in principle) observable quantity which is stable under the removal of the ultraviolet-cutoff from the theory.

Further research on the problem investigated in the present work could branch out into different directions. One possibility would be to try and investigate the $(\sigma \to 0)$-behaviour of terms in the perturbation expansion which are of higher order than four, i.e. the ones we have not included into the approximate model $V(\Lambda, \mathbf{R})$ for the interaction potential. The derivation of the energy corrections that we have executed up to fourth order (see Section 4.1) could in principle be carried out up to arbitrary order, and there may be a chance of identifying recursive relationships or closed formulas that could allow one to prove $(\sigma \to 0)$-convergence for contributions at all orders. As far as the works by Bach, Fröhlich and Pizzo ([BFP09]) and by Griesemer and Hasler ([GH09]) mentioned in Section 1.2 are concerned, one could try to explicitly evaluate the coefficients of the expansions occurring in their results and compare them to the contributions to $V(\Lambda, \mathbf{R})$ found by us. It would also be highly desirable to go beyond the dipole approximation and carry out the analysis presented here for a model incorporating an $\mathbf{x}$-dependent vector potential $\mathbf{A}$. The

main difficulties this would cause would occur in the procedure converting perturbation matrix elements into integrals over photon momenta, see Section 4.4: the presence of terms of the form $\exp(-\mathrm{i}\mathbf{k}\cdot\mathbf{x})$ in the integrands severely limits the extent to which the resolvent and operator identities from Section 4.2.6 and the symmetry properties of the polarization vectors can be employed. This would probably require the introduction and careful analysis of objects similar to but more complicated than the dynamic polarizabilities $\alpha_E^{A,B}(\mathbf{k})$ of the atoms. Finally, another approach to the problem at hand would be to try and derive expressions for the Born-Oppenheimer potential energy surface that do not rely on perturbation theory at all, for instance by using contour integral representations for the ground state projections of infrared-regularized Pauli-Fierz Hamiltonians directly.

Part III

Appendix

Appendix A

A.1 Smeared Coulomb potential as bounded operator on H^1

Lemma A.1.1. *Let $\Lambda > 0$ and let $\psi = \Lambda^3 \psi_0(\Lambda \cdot)$, where $\psi_0 \in C_0^\infty(\mathbb{R}^3)$ is invariant under $O(3)$ and satisfies $\int \psi_0 = 1$. Consider the smeared interatomic Coulomb potential*

$$\begin{aligned} &Q_R(\mathbf{x}_1, \ldots, \mathbf{x}_N) \\ =&\frac{1}{4\pi} \sum_{i_A, j_B} \int_{\mathbb{R}^6} dy dy' \psi(y)\psi(y') \\ &\times \Big(\frac{1}{|\mathbf{R} - y + y'|} + \frac{1}{|\mathbf{x}_{i_A} - \mathbf{x}_{j_B} - \mathbf{R} - y + y'|} - \frac{1}{|\mathbf{x}_{i_A} - \mathbf{R} - y + y'|} - \frac{1}{|\mathbf{x}_{j_B} + \mathbf{R} - y + y'|} \Big), \end{aligned}$$

where $i_A \in \{1, \ldots, Z_A\}$, $j_B \in \{Z_A + 1, \ldots, Z_A + Z_B = N\}$. There exists a positive constant C_Q, independent of R and the ultraviolet-cutoff Λ, such that

$$\|Q_R \Psi\|_{L^2(\mathbb{R}^{3N})} \leq C_Q \big(1 + \frac{1}{R}\big) \|\Psi\|_{H^1(\mathbb{R}^{3N})},$$

i.e. Q_R is a bounded operator from $H^1(\mathbb{R}^{3N})$ to $L^2(\mathbb{R}^{3N})$, and its operator norm can be bounded by a constant C_Q which is independent of R as long as $R > R_0$.

Proof. First note that all four terms occurring in Q_R are of the form

$$\frac{1}{4\pi} \left((\psi * \psi) * \frac{1}{|\cdot|} \right) (\mathbf{x}),$$

where $\mathbf{x} = \mathbf{R}$, $\mathbf{x} = \mathbf{x}_{i_A} - \mathbf{x}_{j_B}$, $\mathbf{x} = \mathbf{x}_{i_A} - \mathbf{R}$ and $\mathbf{x} = \mathbf{x}_{j_B} + \mathbf{R}$, respectively, as can be seen by a simple calculation involving Fubini's theorem and a change of variables. Furthermore, $\psi * \psi$ defines a (possible signed) Borel measure with total mass 1 on $\mathbb{R}^3$, which satisfies

$$\int_{\mathbb{R}^3} \frac{|(\psi * \psi)|(\mathbf{x})}{1 + |\mathbf{x}|} d\mathbf{x} < \infty$$

and is invariant under rotations. Therefore, we can apply [LL97], Theorem 9.7, which asserts that the potential generated by the smeared charge distribution ψ can be estimated from above by a Coulomb potential with the same total mass as $|\psi * \psi|$:

$$\left| \frac{1}{4\pi} \left((\psi * \psi) * \frac{1}{|\cdot|} \right) (\mathbf{x}) \right| \leq \frac{1}{4\pi} \frac{1}{|\mathbf{x}|} \underbrace{\int_{\mathbb{R}^3} |\psi * \psi|(y) dy}_{=:C}.$$

Note that we have not assumed positivity of ψ_0, otherwise C would be equal to one. Nevertheless, C is independent of the ultraviolet-cutoff Λ, as follows from the scaling behaviour of $\psi(x) = \Lambda^3\psi_0(\Lambda\cdot)$. Using the above estimate, we find

$$\begin{aligned}&|Q_R(\mathbf{x}_1,\dots,\mathbf{x}_N)|\\ \leq&\frac{C}{4\pi}\Big(\frac{Z_AZ_B}{|\mathbf{R}|}+\sum_{i_A,j_B}\frac{1}{|\mathbf{x}_{i_A}-\mathbf{x}_{j_B}-\mathbf{R}|}-\sum_{i_A}\frac{Z_B}{|\mathbf{x}_{i_A}-\mathbf{R}|}-\sum_{j_B}\frac{Z_A}{|\mathbf{x}_{j_B}+\mathbf{R}|}\Big).\end{aligned}$$

The first term is just an (R-dependent) multiple of the identity from $H^1(\mathbb{R}^{3N})$ to $L^2(\mathbb{R}^{3N})$, so it remains to prove the assertion for the remaining terms. We will demonstrate the argument for the third term

$$Q_3 := -\frac{Z_B}{4\pi}\sum_{i_A}\int_{\mathbb{R}^6}\frac{\psi(y)\psi(y')}{|\mathbf{x}_{i_A}-\mathbf{R}-y+y'|}dydy',$$

the other two cases being similar. Assume without loss of generality that $Z_A = 1$. Then for any $\Psi \in H^1(\mathbb{R}^{3N})$, we find

$$\begin{aligned}\|Q_3\Psi\|^2_{L^2(\mathbb{R}^{3N})} &\leq \left(\frac{CZ_B}{4\pi}\right)^2\int_{\mathbb{R}^{3N}}\frac{1}{|\mathbf{x}_1-\mathbf{R}|^2}|\Psi|^2(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_1\dots d\mathbf{x}_N\\ &= \left(\frac{CZ_B}{4\pi}\right)^2\int_{\mathbb{R}^{3N-3}}\left(\int_{\mathbb{R}^3}\frac{1}{|\mathbf{x}_1|^2}|\Psi|^2(\mathbf{x}_1+\mathbf{R},\dots,\mathbf{x}_N)d\mathbf{x}_1\right)d\mathbf{x}_2\dots d\mathbf{x}_N,\end{aligned}$$

and Hardy's inequality on $\mathbb{R}^3$, together with the translation invariance of the gradient, finally yields

$$\begin{aligned}\|Q_3\Psi\|^2_{L^2(\mathbb{R}^{3N})} &\leq 4\left(\frac{CZ_B}{4\pi}\right)^2\int_{\mathbb{R}^{3N-3}}\left(\int_{\mathbb{R}^3}|\nabla_{\mathbf{x}_1}\Psi|^2(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_1\right)d\mathbf{x}_2\dots d\mathbf{x}_N\\ &\leq 4\left(\frac{CZ_B}{4\pi}\right)^2\|\nabla\Psi\|^2_{L^2(\mathbb{R}^{3N})},\end{aligned}$$

where the last inequality can for instance be shown using Plancherel's formula and the positivity of the symbols of the $-\Delta_{\mathbf{x}_i}$. □

A.2 Schrödinger resolvent conserves exponential decay

Lemma A.2.1. *Suppose that $f, g \in L^1(\mathbb{R}^n)$ satisfy the pointwise estimates*

$$\begin{aligned}|f(x)| &\leq C_1\mathrm{e}^{-\gamma_1|x|}, \quad \text{for a.e. } x \in \mathbb{R}^n,\\ |g(x)| &\leq C_2\mathrm{e}^{-\gamma_2|x|}, \quad \text{for a.e. } x \in \mathbb{R}^n \setminus B_{R_0}(0),\end{aligned}$$

with positive constants $C_1, C_2, \gamma_1, \gamma_2$ and $R_0 > 0$. Then for any $0 < \gamma < \min\{\gamma_1, \gamma_2\}$ there exists a positive constant C such that

$$|(f * g)(x)| \leq C\mathrm{e}^{-\gamma|x|}$$

for almost every $x \in \mathbb{R}^n$.

Proof. Using the assumed decay of f and g, we find

$$\begin{aligned}
&|(f*g)(x)| \\
=&\left|\int_{B_{R_0}(0)} f(y)g(x-y)dy + \int_{\mathbb{R}^n\setminus B_{R_0}(0)} f(y)g(x-y)dy\right| \\
\leq& C_2\int_{B_{R_0}(0)} |f(y)|\mathrm{e}^{-\gamma_2|x-y|}dy + C_1C_2\int_{\mathbb{R}^n\setminus B_{R_0}(0)} \mathrm{e}^{-\gamma_1|y|-\gamma_2|x-y|}dy \\
\leq& C_2\left(\sup_{y\in B_{R_0}(0)} \mathrm{e}^{-\gamma_2|x-y|}\right)\int_{B_{R_0}(0)} |f(y)|dy + C_1C_2\int_{\mathbb{R}^n\setminus B_{R_0}(0)} \mathrm{e}^{-\min\{\gamma_1,\gamma_2\}(|y|+|x-y|)}dy \\
\leq& C_2\left(\sup_{y\in B_{R_0}(0)} \mathrm{e}^{-\gamma_2|x-y|}\right)\|f\|_{L^1(\mathbb{R}^n)} + C_1C_2\int_{\mathbb{R}^n} \mathrm{e}^{-\min\{\gamma_1,\gamma_2\}(|y|+|x-y|)}dy. \qquad \text{(A.2.1)}
\end{aligned}$$

For $y\in B_{R_0}(0)$, we have

$$|x-y| \geq ||x|-|y|| \geq |x|-|y| \geq |x|-R_0,$$

which yields

$$\begin{aligned}
C_2\left(\sup_{y\in B_{R_0}(0)} \mathrm{e}^{-\gamma_2|x-y|}\right)\|f\|_{L^1(\mathbb{R}^n)} &\leq \left(C_2\|f\|_{L^1(\mathbb{R}^n)}\mathrm{e}^{\gamma_2R_0}\right)\mathrm{e}^{-\gamma_2|x|} \\
&\leq \left(C_2\|f\|_{L^1(\mathbb{R}^n)}\mathrm{e}^{\gamma_2R_0}\right)\mathrm{e}^{-\min\{\gamma_1,\gamma_2\}|x|}.
\end{aligned}$$

To estimate the second term in (A.2.1), set $\overline{\gamma} := \min\{\gamma_1,\gamma_2\}$, choose $0<\gamma<\overline{\gamma}$ and consider

$$\mathrm{e}^{\gamma|x|}\left(C_1C_2\int_{\mathbb{R}^n} \mathrm{e}^{-\overline{\gamma}(|y|+|x-y|)}dy\right) = C_1C_2\int_{\mathbb{R}^n} \mathrm{e}^{-\overline{\gamma}(|x-y|+|y|-c|x|)}dy, \qquad \text{(A.2.2)}$$

where we have set $c := \gamma/\overline{\gamma} < 1$. Choosing a constant $\tilde{c}$ such that $2\tilde{c}+c\leq 1$, the triangle inequality implies

$$\begin{aligned}
\tilde{c}|x+y|+c|x| \leq& \tilde{c}|x-y|+2\tilde{c}|y|+c|x-y|+c|y| \\
=&(\tilde{c}+c)|x-y|+(2\tilde{c}+c)|y| \\
\leq&|x-y|+|y|.
\end{aligned}$$

Plugging this into (A.2.2) yields

$$\mathrm{e}^{\gamma|x|}\left(C_1C_2\int_{\mathbb{R}^n} \mathrm{e}^{-\overline{\gamma}(|y|+|x-y|)}dy\right) \leq C_1C_2\int_{\mathbb{R}^n} \mathrm{e}^{-\overline{\gamma}\tilde{c}(|x+y|)}dy = C',$$

the right-hand side being a constant which is independent of x due to translation invariance. Thus we have shown that

$$C_1C_2\int_{\mathbb{R}^n} \mathrm{e}^{-\overline{\gamma}(|y|+|x-y|)}dy \leq C'\mathrm{e}^{-\gamma|x|},$$

and the proof is finished upon defining

$$C := \max\{C_2\|f\|_{L^1(\mathbb{R}^n)}\mathrm{e}^{\gamma_2R_0}, C'\}.$$

□

Lemma A.2.2. *Suppose that $f \in L^2_{anti}(\mathbb{R}^{3N})$ is real and that its one-particle density ρ_f is (pointwise) exponentially decaying, i.e. there exist positive constants C' and γ' such that*

$$|\rho_f(\mathbf{x})| \leq C' \mathrm{e}^{-\gamma'|\mathbf{x}|}$$

for almost all $\mathbf{x} \in \mathbb{R}^3$. Suppose that $u \in H^2(\mathbb{R}^{3N}) \cap L^2_{anti}(\mathbb{R}^{3N})$ is a solution of

$$(H - E_0 + \lambda)u = f,$$

where

$$\begin{aligned} H = &- \sum_{i=1}^{N} \frac{\hbar^2}{2m_e} \Delta_{\mathbf{x}_i} + \frac{e^2}{4\pi} \sum_{i<j} \int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\psi(y')}{|\mathbf{x}_i - \mathbf{x}_j - y + y'|} \\ &- \frac{e^2 Z_A}{4\pi} \sum_i \int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\psi(y')}{|\mathbf{x}_i - y + y'|} \end{aligned}$$

is self-adjoint with domain $D(H) = H^2(\mathbb{R}^{3N}) \cap L^2_{anti}(\mathbb{R}^{3N})$. Here E_0 is the smallest eigenvalue of H (see Proposition 2.5.1), $\lambda \geq 0$ is a constant, and $\psi \in C_0^\infty(\mathbb{R}^3)$ satisfies the assumptions (A1). Then there exist constants $C \geq 0$, $\gamma \geq 0$, independent of λ, such that

$$\rho_u(\mathbf{x}) \leq C \mathrm{e}^{-\gamma|\mathbf{x}|}.$$

Proof. For the sake of simplicity, we set $\hbar = m_e = e^2/4\pi = 1$. Set

$$V_\psi(\mathbf{x}) := -Z_A \int_{\mathbb{R}^6} dy dy' \frac{\psi(y)\psi(y')}{|\mathbf{x} - y + y'|}$$

and define the operator

$$L := -\frac{1}{2}\Delta_{\mathbf{x}} + V_\psi(\mathbf{x}) + (E_* - E_0) + \lambda$$

with domain $D(L) = H^2(\mathbb{R}^3)$. Here $E_* = \inf \operatorname{ess\,spec}(H)$ (with the analogous choice of units). By Proposition 2.5.1, we have $E_* - E_0 > 0$. Let ρ_u and ρ_f denote the one-particle densities of the antisymmetric functions u and f, and note that $\sqrt{\rho_u} \in H^2(\mathbb{R}^3)$, $\sqrt{\rho_f} \in L^2(\mathbb{R}^3)$.

Claim 1: $L\sqrt{\rho_u} \leq \sqrt{\rho_f}$.

Proof. The proof of the Schrödinger inequality for the one-particle density (see [Thi94] or [Fri]) yields

$$(L\sqrt{\rho_u})(\mathbf{x}) \leq \frac{N}{\sqrt{\rho_u}} \int_{\mathbb{R}^{3N-3}} f(\mathbf{x}, \ldots, \mathbf{x}_N)\overline{u}(\mathbf{x}, \ldots, \mathbf{x}_N) d\mathbf{x}_2 \ldots d\mathbf{x}_N \tag{A.2.3}$$

By assumption, f is real, and u is the preimage of f under the real operator H, so that u

is also real. Thus, using the Cauchy-Schwarz inequality on $L^2(\mathbb{R}^{3N-3})$, we can estimate

$$\begin{aligned}
&\frac{N}{\sqrt{\rho_u}}\int_{\mathbb{R}^{3N-3}} f(\mathbf{x}_1,\dots,\mathbf{x}_N)u(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_2\dots d\mathbf{x}_N\\
\leq&\frac{N}{\sqrt{\rho_u}}\int_{\mathbb{R}^{3N-3}} |f|(\mathbf{x}_1,\dots,\mathbf{x}_N)|u|(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_2\dots d\mathbf{x}_N\\
\leq&\frac{N}{\sqrt{\rho_u}}\left(\sqrt{\int_{\mathbb{R}^{3N-3}} |f|^2(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_2\dots d\mathbf{x}_N}\right)\left(\sqrt{\int_{\mathbb{R}^{3N-3}} |u|^2(\mathbf{x}_1,\dots,\mathbf{x}_N)d\mathbf{x}_2\dots d\mathbf{x}_N}\right)\\
=&\frac{N}{\sqrt{\rho_u}}\frac{1}{\sqrt{N}}\sqrt{\rho_f}\frac{1}{\sqrt{N}}\sqrt{\rho_u}\\
=&\sqrt{\rho_f},
\end{aligned}$$

proving Claim 1.
Next choose $d_0 > 0$ large enough such that $d_0 > 2/\Lambda$ (recall that Λ is the ultraviolet-cutoff parameter contained in the function ψ) and $-Z_A/d_0 + (E_* - E_0) \geq (E_* - E_0)/2$. Define the operator

$$\tilde{L} := -\frac{1}{2}\Delta_{\mathbf{x}} - \frac{Z_A}{d_0} + (E_* - E_0).$$

$\tilde{L}$ is self-adjoint on $L^2(\mathbb{R}^3)$ with domain $D(\tilde{L}) = H^2(\mathbb{R}^3)$. Furthermore, it is boundedly invertible by the choice of d_0 and the fact that $E_* - E_0 > 0$, and its inverse is given explicitly by convolution with the positive Green's function

$$G(\mathbf{x}) = \frac{1}{4\pi|\mathbf{x}|}\mathrm{e}^{-\sqrt{2((E_*-E_0)-Z_A/d_0)}|\mathbf{x}|}. \tag{A.2.4}$$

For $c \geq 1$, set

$$v_c := c\,\tilde{L}^{-1}[\sqrt{\rho_f}]$$

and note that $v_c \in H^2(\mathbb{R}^3)$ by elliptic regularity.

Claim 2: $v_c > 0$, and on $\{|\mathbf{x}| \geq d_0\}$, $(L\sqrt{\rho_u})(\mathbf{x}) \leq (Lv_c)(\mathbf{x})$.

Proof. The first assertion follows from (A.2.4). To prove the second assertion, we use Claim 1 and the definition of v_c to conclude

$$\begin{aligned}
&(L\sqrt{\rho_u})(\mathbf{x}) - (Lv_c)(\mathbf{x})\\
\leq&\sqrt{\rho_f}(\mathbf{x}) - \left[-\frac{1}{2}(\Delta_{\mathbf{x}}v_c)(\mathbf{x}) + V_\psi(\mathbf{x})v_c(\mathbf{x}) + (E_* - E_0)v_c(\mathbf{x}) + \lambda v_c(\mathbf{x})\right]\\
=&\sqrt{\rho_f}(\mathbf{x}) - \Bigg[\underbrace{\left(\left(-\frac{1}{2}\Delta_{\mathbf{x}} - \frac{Z_A}{d_0} + (E_* - E_0)\right)v_c\right)(\mathbf{x})}_{=(\tilde{L}v_c)(\mathbf{x})=c\sqrt{\rho_f}(\mathbf{x})} + \left(\frac{Z_A}{d_0} + V_\psi(\mathbf{x}) + \lambda\right)v_c(\mathbf{x})\Bigg]\\
=&(1-c)\sqrt{\rho_f}(\mathbf{x}) - \lambda v_c(\mathbf{x}) + \left(-V_\psi(\mathbf{x}) - \frac{Z_A}{d_0}\right)v_c(\mathbf{x}).
\end{aligned}$$

By the choice of c and the fact that ρ_f is non-negative, the first term is less or equal to zero for all $\mathbf{x} \in \mathbb{R}^3$. The same holds for the second term since $\lambda \geq 0$ and $v_c > 0$, as was

established above. By Newton's theorem (see e.g. [LL97], Theorem 9.7), we have

$$V_\psi(\mathbf{x}) = -Z_A \left((\psi * \psi) * \frac{1}{|\cdot|} \right)(\mathbf{x}) = -\frac{Z_A}{|\mathbf{x}|}$$

outside $\operatorname{supp}(\psi * \psi)$, and the latter is a subset of $B_{2/\Lambda}(0)$ by the assumptions (A1) on ψ. Thus by the choice of d_0, for any $\mathbf{x}$ with $|\mathbf{x}| \geq d_0$,

$$\left(-V_\psi(\mathbf{x}) - \frac{Z_A}{d_0} \right) v_c(\mathbf{x}) = \left(\frac{Z_A}{|\mathbf{x}|} - \frac{Z_A}{d_0} \right) v_c(\mathbf{x}) \leq 0,$$

which proves Claim 2.
Note that since $\sqrt{\rho_u}$, $v_c \in H^2(\mathbb{R}^3)$, the Sobolev inequalities imply $\sqrt{\rho_u}$, $v_c \in C(\mathbb{R}^3)$. Furthermore, $v_c > 0$ by the above, so that $\max\limits_{|\mathbf{x}| \leq d_0} \frac{\sqrt{\rho_u}(\mathbf{x})}{v_1(\mathbf{x})}$ is well-defined.

Claim 3: For $c \geq \max\limits_{|\mathbf{x}| \leq d_0} \frac{\sqrt{\rho_u}(\mathbf{x})}{v_1(\mathbf{x})}$, we have $\sqrt{\rho_u} \leq v_c$ on $\mathbb{R}^3$.

Proof. On $\{|\mathbf{x}| \leq d_0\}$ the choice of c implies

$$\sqrt{\rho_u}(\mathbf{x}) = v_1(\mathbf{x}) \frac{\sqrt{\rho_u}(\mathbf{x})}{v_1(\mathbf{x})} \leq v_1(\mathbf{x}) \max_{|\mathbf{x}| \leq d_0} \frac{\sqrt{\rho_u}(\mathbf{x})}{v_1(\mathbf{x})} \leq v_1(\mathbf{x}) c = v_c(\mathbf{x}).$$

This inequality holds in particular on the set $\{|\mathbf{x}| = d_0\} = \partial(\{|\mathbf{x}| \geq d_0\})$, so that the claim on the remaining set $\{|\mathbf{x}| \geq d_0\}$ follows from Claim 2 and the maximum principle (see e.g. [Eva98] or [GT01]).
In light of Claim 3, the assertion of the lemma follows once we show that v_c satisfies a pointwise exponential bound. To this end, recall that as noted above,

$$v_c = c\tilde{L}^{-1}[\sqrt{\rho_f}] = c\, G * \sqrt{\rho_f},$$

where

$$G(\mathbf{x}) = \frac{1}{4\pi|\mathbf{x}|} \mathrm{e}^{-\sqrt{2((E_* - E_0) - Z_A/d_0)}|\mathbf{x}|}.$$

Together with the assumed pointwise exponential decay of ρ_f, Lemma A.2.1 implies the existence of positive constants C and

$$\gamma < \min\{-\sqrt{2((E_* - E_0) - Z_A/d_0)}, \gamma'\}$$

such that

$$v_c(\mathbf{x}) \leq C\mathrm{e}^{-\gamma|\mathbf{x}|}.$$

□

A.3 Exponential bounds for H^1-norms

Lemma A.3.1. *For $d > 0$, set $\Omega_d := B_d(0) \times \dots B_d(0) \subset \mathbb{R}^{3N}$. Let $\chi_{\Omega_d}(\mathbf{x}_1, \dots, \mathbf{x}_N) = \Pi_{i=1}^N \chi_d(\mathbf{x}_i)$, where $0 \leq \chi_d \leq 1$, $\chi_d \in C_0^\infty(\mathbb{R}^3)$, $\chi_d(\mathbf{x}) = 1$ if $|\mathbf{x}| \leq d$, $\chi_d(\mathbf{x}) = 0$ if $|\mathbf{x}| > d + 1$, be a smooth characteristic function of Ω_d. Suppose that ψ is a measurable function which is (pointwise) exponentially decaying, i.e. there exist positive constants C and γ such that*

$$|\psi(\mathbf{x}_1, \dots, \mathbf{x}_N)| \leq C \mathrm{e}^{-\gamma(|\mathbf{x}_1| + \dots |\mathbf{x}_N|)}.$$

for almost all $(\mathbf{x}_1, \dots, \mathbf{x}_N) \in \mathbb{R}^{3N}$. Then for any $1 \leq p < \infty$,

i.

$$\|\psi\|_{L^p(\mathbb{R}^{3N} \setminus \Omega_d)} \leq C_1 \mathrm{e}^{-\gamma_1 d}$$

for suitable positive constants C_1, γ_1.

ii. If in addition, $\psi \in H^2(\mathbb{R}^{3N})$ is an eigenfunction of the elliptic operator $-\Delta_{3N} + V$, where $V = \sum_{i=1}^N V_i$ and $V_i \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$, then there exist positive constants C_2 and γ_2, independent of d, such that

$$\|(1 - \chi_{\Omega_d})\psi\|_{H^1(\mathbb{R}^{3N})}\| \leq C_2 \mathrm{e}^{-\gamma_2 d}.$$

Proof. i) Noting that there exists a decomposition $(\Omega_d)^c = \cup_J \Omega_J$ into $2^N - 1$ disjoint subsets, where every Ω_J is of the form $(\cdots \times \{|\mathbf{x}_{i_J}| > d\} \times \dots)$ for some $i_J \in \{1, \dots, N\}$, we find

$$\|\psi\|^p_{L^p(\mathbb{R}^{3N} \setminus \Omega_d)} = \sum_J \int_{\Omega_J} |\psi(\mathbf{x}_1, \dots, \mathbf{x}_N)|^p d\mathbf{x}_1 \dots d\mathbf{x}_N.$$

Since the integrand is positive, we can replace each Ω_J by the larger set

$$\mathbb{R}^3 \times \cdots \times \{|\mathbf{x}_{i_J}| > d\} \times \cdots \times \mathbb{R}^3$$

and use the fact that

$$\begin{aligned}\int_{x \in \mathbb{R}^3, |x| > d} \mathrm{e}^{-\tilde{\gamma}|x|} dx =& 4\pi \int_d^\infty r^2 \mathrm{e}^{-\tilde{\gamma} r} dr \\ =& 4\pi \, \mathrm{e}^{-\tilde{\gamma} d} \left(\frac{d^2}{\tilde{\gamma}} + \frac{2d}{\tilde{\gamma}^2} + \frac{2}{\tilde{\gamma}^3} \right) \\ \leq& c \, \mathrm{e}^{-c' d}\end{aligned}$$

for suitable constants c, c' to obtain

$$\begin{aligned}&\int_{\Omega_J} |\psi(\mathbf{x}_1, \dots, \mathbf{x}_N)|^p d\mathbf{x}_1 \dots d\mathbf{x}_N \\ \leq& C^p \int_{\Omega_J} \mathrm{e}^{-(p\gamma)(|\mathbf{x}_1| + \cdots + |\mathbf{x}_N|)} d\mathbf{x}_1 \dots d\mathbf{x}_N \\ \leq& C^p \int_{\mathbb{R}^3 \times \cdots \times \{|\mathbf{x}_{i_J}| > d\} \times \cdots \times \mathbb{R}^3} \mathrm{e}^{-(p\gamma)(|\mathbf{x}_1| + \cdots + |\mathbf{x}_N|)} d\mathbf{x}_1 \dots d\mathbf{x}_N \\ \leq& C^p \left(\frac{8\pi}{(p\gamma)^3} \right)^{N-1} c \, \mathrm{e}^{-c' d},\end{aligned}$$

the right-hand side being independent of J and thus yielding

$$\|\psi\|_{L^p(\mathbb{R}^{3N}\setminus\Omega_d)} \leq (2^N-1)C^p \left(\frac{8\pi}{(p\gamma)^3}\right)^{N-1} c\,\mathrm{e}^{-c'd},$$

proving the first assertion.
ii) Note that $(1-\chi_{\Omega_d})\psi \in H^2(\mathbb{R}^{3N})$ and $\operatorname{supp}(1-\chi_{\Omega_d})\psi \subset \mathbb{R}^{3N}\setminus\Omega_d$. Assume without loss of generality that ψ is real (otherwise, consider real and imaginary part separately). Using the Cauchy-Schwarz and Young inequalities, as well as the facts that $0 \leq (1-\chi_{\Omega_d}) \leq 1$ and $|\nabla[1-\chi_{\Omega_d}]|^2 \leq C_\chi$, we find

$$\begin{aligned}
&\|(1-\chi_{\Omega_d})\psi\|^2_{H^1(\mathbb{R}^{3N})}\\
&= \int_{\operatorname{supp}(1-\chi_{\Omega_d})} \Big[|(1-\chi_{\Omega_d})\psi|^2 + |\psi|^2\,|\nabla[1-\chi_{\Omega_d}]|^2 + (1-\chi_{\Omega_d})^2|\nabla\psi|^2\\
&\qquad\qquad + 2\left((1-\chi_{\Omega_d})\psi(\nabla[1-\chi_{\Omega_d}]\cdot\nabla\psi)\right)\Big]\\
&\leq \int_{\mathbb{R}^{3N}\setminus\Omega_d} \Big[|(1-\chi_{\Omega_d})\psi|^2 + |\psi|^2\,|\nabla[1-\chi_{\Omega_d}]|^2 + (1-\chi_{\Omega_d})^2|\nabla\psi|^2\\
&\qquad\qquad + (1-\chi_{\Omega_d})^2|\nabla\psi|^2 + |\nabla[1-\chi_{\Omega_d}]|^2|\psi|^2\Big]\\
&\leq \int_{\mathbb{R}^{3N}\setminus\Omega_d} \Big[(1+2C_\chi)|\psi|^2 + 2|\nabla\psi|^2\Big].
\end{aligned}$$

By part i), the first summand satisfies an exponential bound. To establish such a bound for the second term, let E denote the eigenvalue corresponding to ψ and set $\tilde{V} := V - E$. Testing the equation

$$(-\Delta_{3N} + \tilde{V})\psi = 0$$

with $(1-\chi_{\Omega_{d-1}})\psi$ and using partial integration yields

$$\begin{aligned}
0 &= \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} \nabla\psi\cdot\nabla[(1-\chi_{\Omega_{d-1}})\psi] + \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} \tilde{V}|\psi|^2(1-\chi_{\Omega_{d-1}})\\
&= \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} (1-\chi_{\Omega_{d-1}})|\nabla\psi|^2 - \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} \frac{1}{2}|\psi|^2\Delta[(1-\chi_{\Omega_{d-1}})]\\
&\quad + \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} \tilde{V}|\psi|^2(1-\chi_{\Omega_{d-1}}),
\end{aligned}$$

and the fact that $1-\chi_{\Omega_{d-1}} \equiv 1$ on $\mathbb{R}^{3N}\setminus\Omega_d$ implies

$$\begin{aligned}
\int_{\mathbb{R}^{3N}\setminus\Omega_d} |\nabla\psi|^2 &= \int_{\mathbb{R}^{3N}\setminus\Omega_d} (1-\chi_{\Omega_{d-1}})|\nabla\psi|^2\\
&\leq \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} (1-\chi_{\Omega_{d-1}})|\nabla\psi|^2\\
&= \int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} \left(\frac{1}{2}\Delta[(1-\chi_{\Omega_{d-1}})] - \tilde{V}(1-\chi_{\Omega_{d-1}})\right)|\psi|^2.
\end{aligned}$$

Since $\Delta[(1-\chi_{\Omega_{d-1}})]$ is bounded and $(1-\chi_{\Omega_{d-1}}) \leq 1$, the right-hand side can be bounded by

$$\int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} |C'_\chi + \tilde{V}||\psi|^2.$$

By the assumptions on V, there exists a representation $C'_\chi + \tilde{V} = \sum_{i=1}^N (\tilde{V}_{i,3/2} + \tilde{V}_{i,\infty})$, where $\tilde{V}_{i,3/2} \in L^{3/2}(\mathbb{R}^3)$ and $\tilde{V}_{i,\infty} \in L^\infty(\mathbb{R}^3)$, and Hölder's inequality yields

$$\begin{aligned}
&\int_{\mathbb{R}^{3N}\setminus\Omega_{d-1}} |C'_\chi + \tilde{V}||\psi|^2 \\
&\leq \sum_{i=1}^N \Big[\|\psi\|^2_{L^2(\mathbb{R}^{3N}\setminus\Omega_{d-1})} \|\tilde{V}_{i,\infty}\|_{L^\infty(\mathbb{R}^3)} + \|\psi\|^2_{L^6(\mathbb{R}^{3N}\setminus\Omega_{d-1})} \|\tilde{V}_{i,3/2}\|_{L^{3/2}(\mathbb{R}^3)} \Big],
\end{aligned}$$

so that the assertion of part ii) follows by applying part i) to the terms $\|\psi\|^2_{L^2(\mathbb{R}^{3N}\setminus\Omega_{d-1})}$ and $\|\psi\|^2_{L^6(\mathbb{R}^{3N}\setminus\Omega_{d-1})}$. □

A.4 Decay estimates for 1D and 2D oscillatory integrals

Lemma A.4.1. *Let $g \in C^N((0,\infty))$, and assume that g and all its derivatives up to order N are in $L^1((0,\infty))$ and have well-defined limits at 0 and ∞. Let $R \in \mathbb{R}$ and $N \in \mathbb{N}$. Then*

$$\begin{aligned}
&\int_0^\infty \sin(\xi R) g(\xi) d\xi \\
&= \sum_{k=0}^{k_{max}} (-1)^{k+1} \cos(\xi R) \left(\frac{d^{2k}}{d\xi^{2k}} g|_0^\infty \right) \frac{1}{R^{2k+1}} + (-1)^k \sin(\xi R) \left(\frac{d^{2k+1}}{d\xi^{2k+1}} g|_0^\infty \right) \frac{1}{R^{2k+2}} \\
&\quad + \begin{cases} (-1)^{N/2} \frac{1}{R^N} \int_0^\infty \sin(\xi R) (\frac{d^N}{d\xi^N} g) d\xi, & N \text{ even}, \\ (-1)^{(N-1)/2} \frac{1}{R^N} \int_0^\infty \cos(\xi R) (\frac{d^N}{d\xi^N} g) d\xi, & N \text{ odd}, \end{cases} \\
&\int_0^\infty \cos(\xi R) g(\xi) d\xi \\
&= \sum_{k=0}^{k_{max}} (-1)^{k} \cos(\xi R) \left(\frac{d^{2k+1}}{d\xi^{2k+1}} g|_0^\infty \right) \frac{1}{R^{2k+2}} + (-1)^k \sin(\xi R) \left(\frac{d^{2k}}{d\xi^{2k}} g|_0^\infty \right) \frac{1}{R^{2k+1}} \\
&\quad + \begin{cases} (-1)^{(N-1)/2+1} \frac{1}{R^N} \int_0^\infty \sin(\xi R) (\frac{d^N}{d\xi^N} g) d\xi, & N \text{ odd}, \\ (-1)^{N/2} \frac{1}{R^N} \int_0^\infty \cos(\xi R) (\frac{d^N}{d\xi^N} g) d\xi, & N \text{ even}, \end{cases}
\end{aligned}$$

where k_{max} is such that $2k_{max}+1 = N-1$ or $2k_{max} = N-1$.

Proof. Integration by parts. □

Lemma A.4.2. *Assume that $g \in C^\infty((0,\infty)\times(0,\infty))$ and that g and all its partial derivatives are continuous on $[0,\infty)\times[0,\infty)$ and rapidly decaying. Let $R>0$. Then for any $k,l\in\mathbb{N}_0$, there exist positive constants C_1,C_2,C_3, independent of R, such that*

$$\Big|\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\,\xi_2^l\,\sin(\xi_1 R)\,\cos(\xi_2 R)\,g(\xi_1,\xi_2)\Big| \le C_1\frac{1}{R^{k+l+2}}, \tag{A.4.1}$$

$$\Big|\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\,\xi_2^l\,\cos(\xi_1 R)\,\cos(\xi_2 R)\,g(\xi_1,\xi_2)\Big| \le C_2\frac{1}{R^{k+l+2}}, \tag{A.4.2}$$

$$\Big|\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\,\xi_2^l\,\sin(\xi_1 R)\,\sin(\xi_2 R)\,g(\xi_1,\xi_2)\Big| \le C_3\frac{1}{R^{k+l+2}}. \tag{A.4.3}$$

Moreover, for any $k\ge 0$, there exist positive constants C_4,C_5, such that

$$\Big|\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\,\sin(\xi_1 R)\,\frac{1}{\xi_2}\sin(\xi_2 R)\,g(\xi_1,\xi_2)\Big| \le C_4\frac{1}{R^{k+1}} \tag{A.4.4}$$

if k is odd, and

$$\Big|\int_0^\infty\int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\,\cos(\xi_1 R)\,\frac{1}{\xi_2}\sin(\xi_2 R)\,g(\xi_1,\xi_2)\Big| \le C_4\frac{1}{R^{k+1}} \tag{A.4.5}$$

if k is even.

Proof. We first prove (A.4.1) for the case that l and k are even. By Lemma A.4.1 (note that $g(\xi_1,\cdot)$ satisfies its assumptions),

$$\begin{aligned} I &:= \int_0^\infty\int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\,\xi_2^l\,\sin(\xi_1 R)\,\cos(\xi_2 R)\,g(\xi_1,\xi_2)\\ &= \int_0^\infty d\xi_1\xi_1^k\sin(\xi_1 R)\Bigg[(-1)^{l/2+1}\frac{1}{R^{l+1}}\int_0^\infty \sin(\xi_2 R)\left[\frac{\partial^{l+1}}{\partial\xi_2^{l+1}}(\xi_2^l g)\right](\xi_1,\xi_2)d\xi_2\\ &\qquad + (-1)^{l/2}\frac{1}{R^{l+1}}\underbrace{\Big[\sin(\xi_2 R)\Big[\frac{\partial^l}{\partial\xi_2^l}(\xi_2^l g)\Big](\xi_1,\xi_2)\Big]_{\xi_2=0}^\infty}_{=0}\Bigg], \end{aligned}$$

where we have used that $\lim_{\xi_2\to\infty}[\frac{\partial^s}{\partial\xi_2^s}(\xi_2^l g)](\xi_1,\xi_2)=0$ for any $\xi_1\in[0,\infty)$ and any $s,l\in\mathbb{N}_0$, and that $[\frac{\partial^s}{\partial\xi_2^s}(\xi_2^l g)](\xi_1,0)=0$ for any $\xi_1\in[0,\infty)$ and any $s<l$, which follows from the assumptions on the function g. Since $\xi_1^k\frac{\partial^{l+1}}{\partial\xi_2^{l+1}}(\xi_2^l g)\in L^1((0,\infty)\times(0,\infty))$, we can use Fubini's theorem to obtain

$$I = \frac{(-1)^{l/2+1}}{R^{l+1}}\int_0^\infty d\xi_2\sin(\xi_2 R)\int_0^\infty d\xi_1\sin(\xi_1 R)\,\xi_1^k\left[\frac{\partial^{l+1}}{\partial\xi_2^{l+1}}(\xi_2^l g)\right](\xi_1,\xi_2).$$

Again using Lemma A.4.1, we find

$$\begin{aligned}
&\int_0^\infty d\xi_1 \sin(\xi_1 R)\, \xi_1^k \left[\frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right](\xi_1,\xi_2) \\
=&\frac{(-1)^{k/2}}{R^{k+1}} \int_0^\infty \cos(\xi_1 R) \left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](\xi_1,\xi_2)\, d\xi_1 \\
&+ \frac{(-1)^{k/2+1}}{R^{k+1}} \left[\cos(\xi_1 R)\left[\frac{\partial^k}{\partial \xi_1^k}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](\xi_1,\xi_2)\right]_{\xi_1=0}^\infty \\
=&\frac{(-1)^{k/2}}{R^{k+1}} \int_0^\infty \cos(\xi_1 R) \left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](\xi_1,\xi_2)\, d\xi_1 \\
&+ \frac{(-1)^{k/2+2}}{R^{k+1}} \left[\frac{\partial^k}{\partial \xi_1^k}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](0,\xi_2),
\end{aligned}$$

where we have used that $\lim_{\xi_1\to\infty}\left[\frac{\partial^s}{\partial \xi_1^s}\left(\xi_1^k \frac{\partial^t}{\partial \xi_2^t}(\xi_2^l g)\right)\right](\xi_1,\xi_2) = 0$ for any $\xi_2 \in [0,\infty)$ and any $s,t,k,l \in \mathbb{N}_0$ by the assumptions on g and the fact that $\left[\frac{\partial^s}{\partial \xi_1^s}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](0,\xi_2) = 0$ if $s < k$. Note that

$$\left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](\xi_1,\xi_2) \in L^1((0,\infty)\times(0,\infty))$$

and

$$\left[\frac{\partial^k}{\partial \xi_1^k}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](0,\xi_2) \in L^1_{\xi_2}((0,\infty)),$$

which yields

$$\begin{aligned}
I =&\frac{(-1)^{l/2+k/2}}{R^{k+l+2}} \left[\int_0^\infty \int_0^\infty d\xi_1 d\xi_2\, \cos(\xi_1 R)\, \sin(\xi_2 R) \left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](\xi_1,\xi_2)\right. \\
&\left. + \int_0^\infty d\xi_2 \sin(\xi_2 R)\left[\frac{\partial^k}{\partial \xi_1^k}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](0,\xi_2)\right],
\end{aligned}$$

as well as the estimate

$$\begin{aligned}
I \leq&\frac{1}{R^{k+l+2}} \left[\left\|\left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](\xi_1,\xi_2)\right\|_{L^1((0,\infty)\times(0,\infty))}\right. \\
&\left. + \left\|\left[\frac{\partial^k}{\partial \xi_1^k}\left(\xi_1^k \frac{\partial^{l+1}}{\partial \xi_2^{l+1}}(\xi_2^l g)\right)\right](0,\xi_2)\right\|_{L^1_{\xi_2}((0,\infty))}\right],
\end{aligned}$$

proving (A.4.1) in the case k,l even. The other cases, as well as (A.4.2) and (A.4.3), are proven completely analogous, using the other cases occurring in Lemma A.4.1.
To prove (A.4.4), assume $k \in \mathbb{N}_0$ odd and use Lemma A.4.1 (note that $g(\cdot,\xi_2)$ satisfies its

assumptions) to obtain

$$\begin{aligned}
&\int_0^\infty d\xi_1\, \sin(\xi_1 R)\, \xi_1^k\, g(\xi_1,\xi_2)\\
=&\frac{(-1)^{(k+1)/2}}{R^{k+2}} \int_0^\infty d\xi_1 \cos(\xi_1 R) \left[\frac{\partial^{k+2}}{\partial \xi_1^{k+2}}(\xi_1^k g)\right](\xi_1,\xi_2)\\
&+ \frac{(-1)^{(k+1)/2+1}}{R^{k+2}} \left[\cos(\xi_1 R)\left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}(\xi_1^k g)\right](\xi_1,\xi_2)\right]_{\xi_1=0}^{\infty}\\
&+ \frac{(-1)^{(k-1)/2}}{R^{k+1}} \underbrace{\left[\sin(\xi_1 R)\left[\frac{\partial^{k}}{\partial \xi_1^{k}}(\xi_1^k g)\right](\xi_1,\xi_2)\right]_{\xi_1=0}^{\infty}}_{=0} \qquad \text{(A.4.6)}\\
=&\frac{(-1)^{(k+1)/2}}{R^{k+2}} \int_0^\infty d\xi_1 \cos(\xi_1 R) \left[\frac{\partial^{k+2}}{\partial \xi_1^{k+2}}(\xi_1^k g)\right](\xi_1,\xi_2)\\
&+ \frac{(-1)^{(k+1)/2}}{R^{k+2}} \left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}(\xi_1^k g)\right](0,\xi_2),
\end{aligned}$$

where we have used that the assumptions on g imply $\lim_{\xi_1\to\infty}\left[\frac{\partial^s}{\partial \xi_1^k}(\xi_1^k g)\right](\xi_1,\xi_2) = 0$ for any $\xi_2 \in [0,\infty)$, $s,k \in \mathbb{N}_0$ and $\left[\frac{\partial^s}{\partial \xi_1^k}(\xi_1^k g)\right](0,\xi_2) = 0$ for any $\xi_2 \in [0,\infty)$ and $s < k$. Note that the vanishing of the term (A.4.6) is exactly what allows us to gain the extra power of $1/R$ we need to balance the blowup of $\sin(\xi_2 R)/\xi_2$ at zero. Furthermore, noting that

$$\left[\frac{\partial^{k+2}}{\partial \xi_1^{k+2}}(\xi_1^k g)\right](\xi_1,\xi_2) \in L^1((0,\infty)\times(0,\infty))$$

and

$$\left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}(\xi_1^k g)\right](0,\xi_2) \in L^1_{\xi_2}((0,\infty)),$$

we find

$$\begin{aligned}
&\int_0^\infty \int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\, \sin(\xi_1 R)\, \frac{1}{\xi_2} \sin(\xi_2 R)\, g(\xi_1,\xi_2)\\
=&\frac{(-1)^{(k+1)/2}}{R^{k+1}} \left[\int_0^\infty \int_0^\infty \cos(\xi_1 R) \frac{\sin(\xi_2 R)}{\xi_2 R} \left[\frac{\partial^{k+2}}{\partial \xi_1^{k+2}}(\xi_1^k g)\right](\xi_1,\xi_2)\, d\xi_1 d\xi_2\right.\\
&\qquad \left. + \int_0^\infty \frac{\sin(\xi_2 R)}{\xi_2 R} \left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}(\xi_1^k g)\right](0,\xi_2) d\xi_2\right].
\end{aligned}$$

As $|\sin(\xi_2 R)/(\xi_2 R)| \le 1$, we arrive at the estimate

$$\begin{aligned}
&\left|\int_0^\infty \int_0^\infty d\xi_1 d\xi_2\, \xi_1^k\, \sin(\xi_1 R)\, \frac{1}{\xi_2} \sin(\xi_2 R)\, g(\xi_1,\xi_2)\right|\\
\le&\frac{1}{R^{k+1}} \left[\left\|\left[\frac{\partial^{k+2}}{\partial \xi_1^{k+2}}(\xi_1^k g)\right](\xi_1,\xi_2)\right\|_{L^1((0,\infty)\times(0,\infty))} + \left\|\left[\frac{\partial^{k+1}}{\partial \xi_1^{k+1}}(\xi_1^k g)\right](0,\xi_2)\right\|_{L^1_{\xi_2}((0,\infty))}\right],
\end{aligned}$$

establishing (A.4.4). The proof of (A.4.5) is completely analogous (just use the other case in Lemma A.4.1). □

A.5 Coulomb potential of a smeared charge density

Suppose that ψ satisfies the assumptions (A1). The Coulomb potential generated between N charges e_i at positions $x_i \in \mathbb{R}^3$, $i = 1, \dots N$, each described by the smeared charge density ψ, is given by (see e.g. [Spo04])

$$= \frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} dy dy' \frac{\psi(y)\, \psi(y')}{4\pi |x_i - x_j - y + y'|}.$$

Using the fact that ψ is real and even and that in the Fourier convention used by us, we have $\hat{\delta} = 1/(2\pi)^{3/2}$, $\widehat{(f * g)} = (2\pi)^{3/2} \hat{f}\, \hat{g}$ and $\widehat{1/|x|} = 4\pi/((2\pi)^{3/2}|k|^2)$, we find

$$\begin{aligned}
&Q(x_1, \dots, x_N) \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \int_{\mathbb{R}^3} dy \psi(y) \Big(\frac{1}{4\pi |x_i - x_j + \cdot|} * \psi \Big)(y) \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \int_{\mathbb{R}^3} dy \Big(\frac{1}{(2\pi)^{3/2}} \int_{\mathbb{R}^3} dk\ \hat{\psi}(k) \mathrm{e}^{\mathrm{i}ky} \Big) \Big(\frac{1}{(2\pi)^{3/2}} \int_{\mathbb{R}^3} dk' \widehat{\frac{1}{4\pi |x_i - x_j + \cdot|}} * \psi(k') \mathrm{e}^{\mathrm{i}k'y} \Big) \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \int_{\mathbb{R}^3} dy \Big(\frac{1}{(2\pi)^{3/2}} \int_{\mathbb{R}^3} dk\ \hat{\psi}(k) \mathrm{e}^{\mathrm{i}ky} \Big) \Big(\frac{(2\pi)^{3/2}}{(2\pi)^{3/2}} \int_{\mathbb{R}^3} dk' \widehat{\frac{1}{4\pi |x_i - x_j + \cdot|}} (k') \widehat{\psi}(k') \mathrm{e}^{\mathrm{i}k'y} \Big) \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \frac{1}{(2\pi)^{3/2}} \int dy dk dk' \widehat{\psi}(k) \widehat{\psi}(k') \mathrm{e}^{\mathrm{i}(x_i - x_j)k'} \widehat{(\frac{1}{4\pi |\cdot|})}(k') \mathrm{e}^{\mathrm{i}(k+k')y} \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \frac{1}{(2\pi)^{3}} \int dk dk' \widehat{\psi}(k) \widehat{\psi}(k') \mathrm{e}^{\mathrm{i}(x_i - x_j)k'} \frac{4\pi}{4\pi |k|^2} \Big(\int dy\ \mathrm{e}^{\mathrm{i}(k+k')y} \Big) \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \frac{1}{(2\pi)^{3}} \int dk dk' \widehat{\psi}(k) \widehat{\psi}(k') \mathrm{e}^{\mathrm{i}(x_i - x_j)k'} \frac{(2\pi)^3}{|k|^2} \delta(k + k') \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \int dk \widehat{\psi}(k) \widehat{\psi}(-k) \mathrm{e}^{\mathrm{i}(x_i - x_j)k} \frac{1}{|k|^2} \\
=&\frac{1}{2} \sum_{i,j=1}^{N} e_i e_j \int dk |\widehat{\psi}(k)|^2 \mathrm{e}^{\mathrm{i}(x_i - x_j)k} \frac{1}{|k|^2},
\end{aligned}$$

For two neutral atoms A and B of nuclear charges Z_A, Z_B, respectively, using the relative coordinates $\{\mathbf{x}_1, \dots, \mathbf{x}_{Z_A}, \mathbf{x}_{Z_A+1} + \mathbf{R}, \dots, \mathbf{x}_N + \mathbf{R}\}$ for the electrons of atom A and B,

respectively, the interatomic Coulomb potential takes the form

$$\begin{aligned}
&e^2 Q_R(x)\\
=&e^2 \sum_{i_A,j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \Big(\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}} - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{i_A}-\mathbf{R})} - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}+\mathbf{R})} + \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}-\mathbf{x}_{i_A}+\mathbf{R})} \Big)\\
=&e^2 \sum_{i_A,j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}} \Big(1 - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{j_B}} - \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}} + \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot(\mathbf{x}_{j_B}-\mathbf{x}_{i_A})} \Big)\\
=&e^2 \sum_{i_A,j_B} \int_{\mathbb{R}^3} d\mathbf{k} \frac{|\hat{\psi}(\mathbf{k})|^2}{|\mathbf{k}|^2} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}} \left[\Big(1 - \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}} \Big) \Big(1 - \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{j_B}} \Big) \right],
\end{aligned}$$

where in the second step we have used that $\hat{\psi}(-\mathbf{k}) = \hat{\psi}(\mathbf{k})$

A.6 Properties of the polarization vectors

Lemma A.6.1. *For $\mathbf{k}, \mathbf{l} \in \mathbb{R}^3$ and the polarization vectors $\mathbf{e}(\mathbf{k}, \lambda)$, the following relations hold:*

$$\sum_{\lambda,\mu=1,2} (\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{e}(\mathbf{l},\mu))^2 = 1 + (\hat{\mathbf{k}}\cdot\hat{\mathbf{l}})^2, \tag{A.6.1}$$

$$\sum_{\lambda=1,2} (\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{l})^2 = |\mathbf{l}|^2 \left(1 - (\hat{\mathbf{k}}\cdot\hat{\mathbf{l}})^2 \right),$$

$$\sum_{\lambda=1,2} |\mathbf{e}(\mathbf{k},\lambda)\rangle\langle\mathbf{e}(\mathbf{k},\lambda)| = I - |\hat{\mathbf{k}}\rangle\langle\hat{\mathbf{k}}|.$$

Proof. Follows from the fact that $\mathbf{k}$, $\mathbf{e}(\mathbf{k},1)$ and $\mathbf{e}(\mathbf{k},2)$ form an orthonormal basis of $\mathbb{R}^3$ for any $\mathbf{k} \in \mathbb{R}^3$. □

A.7 Commutator on eigenstates

Lemma A.7.1. *Consider an N-particle Schrödinger operator of the form*

$$H = \sum_{i=1}^{N} \frac{p_i^2}{2m_e} + V(x),$$

where $x = (x_1, \ldots, x_N) \in \mathbb{R}^{3N}$, $p_i = -\mathrm{i}\hbar\nabla_{x_i}$, and the potential V is such that H has a self-adjoint realization on (a subspace of) $L^2(\mathbb{R}^{3N})$. Then for any

$$\psi \in D(x_i H) \cap D(H x_i) \cap H^1(\mathbb{R}^{3N}),$$

the commutator relation

$$[H, x_i]\psi = H(x_i\psi) - x_i(H\psi) = \frac{\hbar}{\mathrm{i}m_e} p_i$$

holds. If in addition ψ is an eigenstate of H corresponding to the eigenvalue E, then

$$\mathbf{p}_i\psi = \frac{\mathrm{i}m_e}{\hbar}(H - E)\mathbf{x}_i\psi.$$

Proof. Under the above assumptions an easy calculation. □

A.8 $\mathbf{p}_\mathbf{x} \cdot \mathbf{A}(\mathbf{x})$ maps atomic ground states to orthogonal complements

Lemma A.8.1. *Assume (A1), (A2) and let $\Lambda \geq \Lambda_0$, with Λ_0 as in Proposition 2.5.2. Let $i_A \in \{1, \ldots, Z_A\}$, $j_B \in \{Z_{A+1}, \ldots, N\}$ and let Ψ_A^0, Ψ_B^0 be the ground states of H_A and H_B, respectively. Then*

$$\left(\mathbf{p}_{\mathbf{x}_{i_A}} \cdot \mathbf{A}_\sigma(\mathbf{x}_{i_A})\right)(\Psi_A^0 \otimes \Omega) \in \{\Psi_A^0\}^\perp \otimes \mathcal{F}^{(1)}, \tag{A.8.1}$$

$$\left(\mathbf{p}_{\mathbf{x}_{i_A}} \cdot \mathbf{A}_\sigma(\mathbf{x}_{i_A})\right)(\Psi_A^0 \otimes u) \in \{\Psi_A^0\}^\perp \otimes \mathcal{H}_B \otimes (\mathcal{F}^{(0)} \oplus \mathcal{F}^{(2)}), \tag{A.8.2}$$

$$\left(\mathbf{p}_{\mathbf{x}_{j_B}} \cdot \mathbf{A}_\sigma(\mathbf{x}_{j_B} + \mathbf{R})\right)(\Psi_B^0 \otimes \Omega) \in \{\Psi_B^0\}^\perp \otimes \mathcal{F}^{(1)}, \tag{A.8.3}$$

$$\left(\mathbf{p}_{\mathbf{x}_{j_B}} \cdot \mathbf{A}_\sigma(\mathbf{x}_{j_B} + \mathbf{R})\right)(\Psi_B^0 \otimes v) \in \{\Psi_B^0\}^\perp \otimes \mathcal{H}_A \otimes (\mathcal{F}^{(0)} \oplus \mathcal{F}^{(2)}) \tag{A.8.4}$$

for any $u \in \mathcal{H}_B \otimes \mathcal{F}^{(1)}$ and $v \in \mathcal{H}_A \otimes \mathcal{F}^{(1)}$.

Remark: The appearance of $\mathbf{R}$ in the argument of $\mathbf{A}$ is due to our using relative coordinates for the electrons of atom B. The assertion of the lemma holds in particular if we replace $\mathbf{x}_{i_A}$ and $\mathbf{x}_{j_B}$ by zero, as can be seen by an inspection of its proof below.

Proof. We give the proof of (A.8.1) and (A.8.2), the other cases being completely analogous. By definition of the vector potential,

$$\mathbf{A}_\sigma(\mathbf{x}) = a^\dagger(\mathbf{G}_\sigma^\mathbf{x}) + a(\mathbf{G}_\sigma^\mathbf{x}),$$

where

$$\mathbf{G}_\sigma^\mathbf{x}(\mathbf{k}, \lambda) = \chi_\sigma(\mathbf{k})\, c\, \rho(\mathbf{k}) \sqrt{\frac{\hbar}{2\omega(\mathbf{k})}} \mathbf{e}(\mathbf{k}, \lambda) \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}}.$$

Since $a(f)\Omega = 0$ for any $f \in W$, we conclude

$$\begin{aligned} f &:= \left(\mathbf{p}_{\mathbf{x}_{i_A}} \cdot \mathbf{A}_\sigma(\mathbf{x}_{i_A})\right)(\Psi_A^0 \otimes \Omega) \\ &= \left(\mathbf{p}_{\mathbf{x}_{i_A}} \cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_{i_A}})\right)(\Psi_A^0 \otimes \Omega) \\ &= \left(\mathbf{p}_{\mathbf{x}_{i_A}} \Psi_A^0 \cdot \mathbf{e}(\mathbf{k}, \lambda)\right) \chi_\sigma(\mathbf{k})\, c\, \rho(\mathbf{k}) \sqrt{\frac{\hbar}{2\omega(\mathbf{k})}} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}}. \end{aligned}$$

To verify $f \in \{\Psi_A^0\}^\perp \otimes \mathcal{F}^{(1)}$, it suffices to show that f is invariant under the corresponding orthogonal projection, i.e. that

$$\left(\overline{(I - P_{\{\Psi_A^0\}}) \otimes I_{\mathcal{F}^{(1)}}}\right) f = f,$$

which is equivalent to showing

$$\left(P_{\{\Psi_A^0\}} \otimes I_{\mathcal{F}^{(1)}}\right) f = 0 \tag{A.8.5}$$

(note that $\operatorname{Ran} P_{\{\Psi_A^0\}}$ is finite-dimensional). To this end, we observe

$$\begin{aligned}&\left((P_{\{\Psi_A^0\}}\otimes I_{\mathcal{F}^{(1)}})f\right)(\mathbf{x}_A;\mathbf{k},\lambda)\\ =&\Psi_A^0(\mathbf{x}_A)\left\langle\Psi_A^0\middle|\chi_\sigma(\mathbf{k})\,c\,\rho(\mathbf{k})\sqrt{\frac{\hbar}{2\omega(\mathbf{k})}}\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}}\left(\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{p}_{\mathbf{x}_{i_A}}\Psi_A^0\right)\right\rangle_{\mathcal{H}_A}(\mathbf{k},\lambda).\end{aligned}$$

Here we have used the shorthand $\mathbf{x}_A=(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})$ for the electron variables of atom A. The second factor can be written as

$$\chi_\sigma(\mathbf{k})\,c\,\rho(\mathbf{k})\sqrt{\frac{\hbar}{2\omega(\mathbf{k})}}\,\mathbf{e}(\mathbf{k},\lambda)\cdot\left\langle\Psi_A^0\middle|\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}}\mathbf{p}_{\mathbf{x}_{i_A}}\Psi_A^0\right\rangle_{\mathcal{H}_A}(\mathbf{k}),\tag{A.8.6}$$

and the inner product with respect to $\mathcal{H}_A$ equals

$$\int_{\mathbb{R}^{3Z_A}}\overline{\Psi_A^0(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A})}(-\mathrm{i}\hbar\nabla_{\mathbf{x}_{i_A}}\Psi_A^0(\mathbf{x}_1,\dots,\mathbf{x}_{Z_A}))\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}}d\mathbf{x}_1\dots d\mathbf{x}_{Z_A}.$$

By Proposition 2.5.2, Ψ_A^0 can be chosen to be a real function, which allows us to use the identity

$$\overline{\Psi_A^0}\nabla\Psi_A^0=\frac{1}{2}\nabla|\Psi_A^0|^2,$$

so that upon using antisymmetry and integrating out the remaining variables, we obtain

$$\left\langle\Psi_A^0\middle|\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}}\mathbf{p}_{\mathbf{x}_{i_A}}\Psi_A^0\right\rangle_{\mathcal{H}_A}(\mathbf{k})=-\frac{\mathrm{i}\hbar}{2Z_A}\int_{\mathbb{R}^3}\nabla_{\mathbf{x}}\rho_{\Psi_A^0}(\mathbf{x})\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}}d\mathbf{x},\tag{A.8.7}$$

where

$$\rho_{\Psi_A^0}(\mathbf{x})=Z_A\int_{\mathbb{R}^{3Z_A-3}}|\Psi_A^0|^2(\mathbf{x},\mathbf{x}_2,\dots,\mathbf{x}_{Z_A})d\mathbf{x}_2\dots,\mathbf{x}_{Z_A}$$

is the one-particle density of Ψ_A^0, which is an element of $L^1(\mathbb{R}^3)$. The right-hand side of (A.8.7) is now easily recognized to equal

$$\frac{\hbar(2\pi)^{3/2}}{2Z_A}\,\mathbf{k}\,\widehat{\rho_{\Psi_A^0}}(\mathbf{k}),$$

so that, using the property $\mathbf{e}(\mathbf{k},\lambda)\cdot\mathbf{k}=0$ of the polarization vectors, we conclude $(A.8.6)=0$ and thus $(A.8.5)$, proving (A.8.1).
To establish (A.8.2), we consider the terms

$$g_1:=\left(\mathbf{p}_{\mathbf{x}_{i_A}}\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_{i_A}})\right)(\Psi_A^0\otimes\,u)\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes\mathcal{F}^{(2)}$$

and

$$g_2:=\left(\mathbf{p}_{\mathbf{x}_{i_A}}\cdot a(\mathbf{G}_\sigma^{\mathbf{x}_{i_A}})\right)(\Psi_A^0\otimes u)\in\mathcal{H}_A\otimes\mathcal{H}_B\otimes\{\Omega\}$$

separately. As concerns the first one, it suffices to show that

$$\begin{aligned}&\left((P_{\{\Psi_A^0\}}\otimes I_{\mathcal{H}_B\otimes\mathcal{F}^{(2)}})g_1\right)(\mathbf{x}_A,\mathbf{x}_B;\mathbf{k}_1,\lambda_1,\mathbf{k}_2,\lambda_2)\\ =&\Psi_A^0(\mathbf{x}_A)\left\langle\Psi_A^0\middle|(\mathbf{p}_{\mathbf{x}_{i_A}}\Psi_A^0)\cdot a^\dagger(\mathbf{G}_\sigma^{\mathbf{x}_{i_A}})u\right\rangle_{\mathcal{H}_A}(\mathbf{x}_B;\mathbf{k}_1,\lambda_1,\mathbf{k}_2,\lambda_2)=0,\end{aligned}$$

which amounts to the same argument as above. Regarding g_2, note that

$$\begin{aligned} g_2(\mathbf{x}_A, \mathbf{x}_B) =& (\mathbf{p}_{\mathbf{x}_{i_A}} \Psi_A^0)(\mathbf{x}_A) \cdot \left(\sum_{\lambda=1,2} \int_{\mathbb{R}^3} \overline{\mathbf{G}_\sigma^{\mathbf{x}_{i_A}}(\mathbf{k}, \lambda)}\, u(\mathbf{x}_B; \mathbf{k}, \lambda) d\mathbf{k} \right) \\ =& \sum_{\lambda=1,2} \int_{\Omega_\sigma} \left(\overline{\mathbf{G}^{\mathbf{x}_{i_A}}(\mathbf{k}, \lambda)} \cdot (\mathbf{p}_{\mathbf{x}_{i_A}} \Psi_A^0)(\mathbf{x}_A) \right) u(\mathbf{x}_B; \mathbf{k}, \lambda) d\mathbf{k}, \end{aligned}$$

and we need to verify

$$\left(P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}^{(0)}} \right) g_2 = 0.$$

Using Fubini's theorem, we find

$$\begin{aligned} & \left((P_{\{\Psi_A^0\}} \otimes I_{\mathcal{H}_B \otimes \mathcal{F}^{(0)}}) g_2 \right) (\mathbf{x}_A, \mathbf{x}_B) \\ =& \Psi_A^0(\mathbf{x}_A) \langle \Psi_A^0 | g_2 \rangle_{\mathcal{H}_A}(\mathbf{x}_B) \\ =& \Psi_A^0(\mathbf{x}_A) \Bigg[\sum_{\lambda=1,2} \int_{\Omega_\sigma} u(\mathbf{x}_B; \mathbf{k}, \lambda)\, c\, \overline{\rho(\mathbf{k})} \sqrt{\frac{\hbar}{2\omega(\mathbf{k})}}\, \mathbf{e}(\mathbf{k}, \lambda) \\ & \cdot \left(\int_{\mathbb{R}^{3Z_A}} \overline{\Psi_A^0(\mathbf{x}_A)}\, (\mathbf{p}_{\mathbf{x}_{i_A}} \Psi_A^0)(\mathbf{x}_A)\, \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_{i_A}} \right) d\mathbf{k} \Bigg], \end{aligned}$$

which is found to vanish by the same argument involving the Fourier transform as above, finishing the proof. □

An immediate consequence of Lemma A.8.1 is the following.

Lemma A.8.2 (Vanishing of cross-terms w/o dipole approximation)**.** *Assume the hypotheses of Lemma A.8.1, let* $\Psi_0 = \Psi_A^0 \otimes \Psi_B^0 \otimes \Omega$ *and let*

$$H'_{\sigma,A} := -\frac{1}{m_e c} \sum_{j_A=1}^{Z_A} (\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(\mathbf{x}_{j_A}))$$

and

$$H'_{\sigma,B} := -\frac{1}{m_e c} \sum_{j_B=Z_A+1}^{N} (\mathbf{p}_{j_B} \cdot \mathbf{A}_\sigma(\mathbf{x}_{j_B} + \mathbf{R}))$$

be the first-order perturbation operators without the dipole approximation. Then

$$\langle H'_{\sigma,A} \Psi_0 | (T^\sigma)^\alpha | H'_{\sigma,B} \Psi_0 \rangle = 0 \quad \text{for } \alpha \in \mathbb{N}.$$

Proof. By Lemma A.8.1,

$$H'_{\sigma,A} \Psi_0 \in \{\Psi_B^0\} \widehat{\otimes} (\{\Psi_A^0\}^\perp \otimes f^{(1)}),$$

$$H'_{\sigma,B} \Psi_0 \in \{\Psi_A^0\} \widehat{\otimes} (\{\Psi_B^0\}^\perp \otimes \mathcal{F}^{(1)}),$$

and both of these subspaces are invariant under $(T^\sigma)^\alpha$ by Lemma 4.2.5. Since they are also mutually orthogonal, the assertion follows.

□

Remark A.8.3. The assertion of the above lemma holds in particular if we use the dipole-approximated perturbations

$$H'_{\sigma,A} = -\frac{1}{m_e c} \sum_{j_A=1}^{Z_A} (\mathbf{p}_{j_A} \cdot \mathbf{A}_\sigma(0))$$

and

$$H'_{\sigma,B} = -\frac{1}{m_e c} \sum_{j_B=Z_A+1}^{N} (\mathbf{p}_{j_B} \cdot \mathbf{A}_\sigma(\mathbf{R})).$$

Indeed, the non-degeneracy of the atomic ground states Ψ_A^0 and Ψ_B^0 implies that they are eigenfunctions of the parity operator, and the dipole-approximated operators $H'_{\sigma,A}$ and $H'_{\sigma,B}$ invert parity, see Lemma 4.2.11. In particular, $H'_{\sigma,A}\Psi_0 \in \{\Psi_B^0\}\widehat{\otimes}(\overline{\{\Psi_A^0\}^\perp \widehat{\otimes} \mathcal{F}})$ and $H'_{\sigma,B}\Psi_0 \in \{\Psi_A^0\}\widehat{\otimes}(\overline{\{\Psi_B^0\}^\perp \widehat{\otimes} \{\Psi_B^0\}^\perp \otimes \mathcal{F}})$. Since these are invariant subspaces for T^σ (Lemma 4.2.5), this implies $\langle H'_{\sigma,A}\Psi_0 | T^\sigma | H'_{\sigma,B}\Psi_0\rangle = \langle H'_{\sigma,B}\Psi_0 | T^\sigma | H'_{\sigma,A}\Psi_0\rangle = 0$ by orthogonality of Fock space levels.

A.9 Calculation of the quantity S_A

For the normalized ground state eigenfunction of the hydrogen atom in spherical coordinates,

$$\Psi(r,\theta,\varphi) = \sqrt{(\frac{2}{a_0})^3 \frac{1}{8\pi}} \mathrm{e}^{-\frac{2r}{2a_0}},$$

we calculate

$$(\mathbf{p}\Psi)(r,\theta,\varphi) = \sqrt{(\frac{2}{a_0})^3 \frac{1}{8\pi}} \frac{\hbar}{\mathrm{i}} (-\frac{2}{2a_0}) \frac{\mathbf{x}}{|x|} \mathrm{e}^{-\frac{2r}{2a_0}},$$

so that

$$\begin{aligned} S_A =& \langle \mathbf{x}\Psi_A^0 | \overline{H}_A | \mathbf{x}\Psi_A^0\rangle = \frac{\hbar}{\mathrm{i}m_e} \langle \mathbf{x}\Psi_A^0 | \mathbf{p}\Psi_A^0\rangle \\ =& \frac{\hbar^2}{m_e} \frac{1}{4} (\frac{2}{a_0})^4 (\frac{a_0}{2})^4 \Big(\int r^3 \mathrm{e}^{-r} dr \Big) \\ =& \frac{\hbar^2}{m_e} \frac{1}{4} \frac{3!}{1^4} = \frac{3}{2} \hbar\, a_0\, \alpha\, c, \end{aligned}$$

where we have used the formula

$$a_0 = \frac{\hbar}{\alpha m_e c}$$

for the Bohr radius. For general $Z_{A,B} \leq 1$, we have the following

Lemma A.9.1. *Assume that the ground state wave function wave functions $\Psi_A^0 \in H^2(\mathbb{R}^{3Z_A})$ and $\Psi_B^0 \in H^2(\mathbb{R}^{3Z_B})$ of the operators H_A and H_B are real. Then*

$$S_{A,B} = \frac{\hbar^2}{2m_e} n Z_{A,B} = \hbar\, (a_0/2)\, \alpha\, c\, n\, Z_{A,B}.$$

Proof. We prove the assertion for S_A. A simple calculation exploiting the fact that Ψ_A^0 is real shows

$$\begin{aligned}
S_A &= \sum_{i,j=1}^{Z_A} \langle \mathbf{x}_i \Psi_A^0 | \overline{H}_A | \mathbf{x}_j \Psi_A^0 \rangle = \sum_{i,j=1}^{Z_A} \frac{\hbar}{\mathrm{i} m_e} \langle \mathbf{x}_i \Psi_A^0 | \overline{H}_A | \mathbf{p}_j j \Psi_A^0 \rangle \\
&= \sum_{i,j=1}^{Z_A} \frac{\hbar^2}{\mathrm{i} m_e} \int_{\mathbb{R}^{3Z_A}} (\mathbf{x}_i \Psi_A^0) \cdot (-\mathrm{i}) \nabla_j \Psi_A^0 = \sum_{i,j=1}^{Z_A} \frac{\hbar^2}{\mathrm{i} m_e} (-\mathrm{i}) \int_{\mathbb{R}^{3Z_A}} (-\nabla_j) \cdot (\mathbf{x}_i \Psi_A^0) \Psi_A^0 \\
&= \sum_{i,j=1}^{Z_A} \frac{\hbar^2}{\mathrm{i} m_e} (-\mathrm{i}) \left(- \int_{\mathbb{R}^{3Z_A}} \left(\delta_{ij} n |\Psi_A^0|^2 + (\mathbf{x}_i \cdot \nabla_j \Psi_A^0) \Psi_A^0 \right) \right) \\
&= \frac{\hbar^2}{m_e} n Z_A \|pa\|^2 + \sum_{i,j=1}^{Z_A} \frac{\hbar^2}{\mathrm{i} m_e} \int_{\mathbb{R}^{3Z_A}} (\Psi_A^0 \mathbf{x}_i) \cdot (\mathrm{i} \nabla_j \Psi_A^0) \\
&= \frac{\hbar^2}{m_e} n Z_A - \sum_{i,j=1}^{Z_A} \frac{\hbar}{\mathrm{i} m_e} \int_{\mathbb{R}^{3Z_A}} (\Psi_A^0 \mathbf{x}_i) \cdot ((-\mathrm{i}) \hbar \nabla_j \Psi_A^0) \\
&= \frac{\hbar^2}{m_e} n Z_A - \sum_{i,j=1}^{Z_A} \frac{\hbar}{\mathrm{i} m_e} \langle \mathbf{x}_i \Psi_A^0 | \mathbf{p}_j \Psi_A^0 \rangle \\
&= \frac{\hbar^2}{m_e} n Z_A - S_A,
\end{aligned}$$

from which it follows that

$$2 S_A = \frac{\hbar^2}{m_e} n Z_A = \hbar \, a_0 \, \alpha \, c \, n \, Z_A.$$

□

A.10 Integral symmetry

Lemma A.10.1. *Let $f, g : \mathbb{R}^6 \to \mathbb{C}$ be measurable and suppose that $fg \in L^1(\mathbb{R}^6)$. Suppose further that $g(x_2, x_1) = g(x_1, x_2)$ for almost all $(x_1, x_2) \in \mathbb{R}^6$. Then*

$$\int_{\mathbb{R}^6} dx_1 dx_2 f(x_1, x_2) g(x_1, x_2) \int_{\mathbb{R}^6} dx_1 dx_2 f(x_2, x_1) g(x_1, x_2).$$

In particular, if $f(x_2, x_1) = -f(x_1, x_2)$, the integral is zero.

Proof. Change of variables. □

Appendix B

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