Multipolar Quantum Electrodynamics of Localized Charge-Current Distributions: Theory and Renormalization

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We formulate a non-relativistic quantum field theory to model interactions between quantized electromagnetic fields and localized charge-current distributions. The electronic degrees of freedom are encoded in microscopic polarization and magnetization field operators whose moments are identified with the multipole moments of the charge-current distribution. The multipolar Hamiltonian is obtained from the minimal coupling Hamiltonian through a unitary transformation corresponding to the canonical “PZW transformation” of classical or semi-classical theory; we renormalize this Hamiltonian at leading-order in perturbation theory, the result of which is used to compute the shift of the electronic energy levels due to interactions between the electrons and quantum vacuum fluctuations in the electromagnetic field. Our renormalized energy shift constitutes a generalization of the Lamb shift in atomic hydrogen, valid for general localized assemblies of atoms and molecules, possibly with net charge but absent free current. By expanding the fields in a series of multipole moments, we study contributions to this energy shift coming from specific multipole moments.

I. INTRODUCTION

While relativistic quantum electrodynamics has been tremendously successful at describing scattering processes in high energy physics, it is cumbersome when applied to bound state problems and those with non-relativistic sources [1]. In part, this is because any process involving matter has an antimatter counterpart that must be included in calculations of probability amplitudes, e.g., to preserve unitarity of the S-matrix. However, after taking the non-relativistic limit the antimatter degrees of freedom decouple in the path integral and can be integrated out of the theory [2]. The result is an effective field theory, called non-relativistic quantum electrodynamics [3], that is particularly well-suited for describing interactions between many-body systems with a fixed number of particles and quantized radiation fields.

Non-relativistic quantum electrodynamics is an effective field theory obtained from an ultraviolet completion with only one characteristic mass scale (the electron mass), implying that the infinite sum of interaction terms in its Lagrangian can be ordered in reciprocal powers of this mass [4]; for applications in quantum optics and condensed matter physics, it is often sufficient to consider only the leading-order terms. In this Lagrangian, matter is described by charge and current densities (ρ, j) that couple to the U(1)-gauge potentials (φ, a) rather than directly to the electromagnetic field [5].

In many systems, however, an alternative approach is possible, provided one can identify specific points in space (such as the nucleus of an atom if that nucleus is assumed to be at rest, or the centre of mass of an atom or molecule) around which it is physically permissible to perform multipole expansions [6]. This approach is based on a formalism called multipolar electrodynamics, which has been specifically designed to study such systems. In this paradigm, the material degrees of freedom are encoded in polarization and magnetization fields (p, m) in place of charge and current densities (ρ, j), which couple to the “Maxwell fields” (d, b) rather than the gauge potentials (φ, a). These descriptions are related by

\[ \rho(x,t) = -\nabla \cdot p(x,t) + \rho_E(x,t), \]  \[ \tag{1} \]

\[ j(x,t) = \frac{\partial p(x,t)}{\partial t} + e \nabla \times m(x,t) + j_E(x,t), \]  \[ \tag{2} \]

allowing for the possibility of free charge and current densities, which would arise if the atom or molecule had a net charge and was free to move. In its Hamiltonian formulation, the standard procedure for obtaining the quantum theory of multipolar electrodynamics involves a unitary transformation of the minimal coupling Hamiltonian, which corresponds in classical or semi-classical theory to a canonical transformation that is often referred to as the “PZW transformation” after its originators [7–9]. Calculations based on the resulting multipolar Hamiltonian often afford more physical insight into the dynamics of the system than do those based on the minimal coupling Hamiltonian, and are free of the artificial divergences in the determination of response coefficients that can plague the latter [10].

Multipolar electrodynamics was originally formulated in terms of the many-body wavefunctions of “first-quantized” quantum mechanics [11]. While their use simplifies certain calculations, these wavefunctions become increasingly difficult to work with as the number of particles becomes large, and certainly prohibitively so if one envisions ultimately extending the treatment to condensed matter systems. Computations are vastly simplified when recast in terms of second-quantized field operators, which automatically account for the various combinatorial factors arising from particle exchange symmetry and permit the application of powerful Fock space methods from quantum field theory [12].
Even in second-quantized treatments, the multipolar Hamiltonian is usually simplified by making the ubiquitous “electric dipole approximation” [13–17], neglecting higher-order electric and magnetic multipole moments as well as spatial variations in the electromagnetic field. However, the electric dipole approximation is only suitable for smaller atoms and molecules; for larger systems and those with several distinct chromophores, spatial variations in optical fields are non-negligible and the electric dipole approximation is invalid [18]. Ultimately, to describe more general charge distributions than those of the simplest atoms and molecules, it is desirable to work with a second-quantized model of radiation-matter interactions that is also capable of including electric and magnetic multipole moments of arbitrary order.

In this paper we present such an approach. We reformulate multipolar electrodynamics within the framework of quantum field theory—a formalism that we call **multipolar quantum electrodynamics**—using a field-theoretic generalization of the PZW transformation applied to the minimal coupling Hamiltonian. The electronic degrees of freedom are encoded in second-quantized scalar field operators that are used to define microscopic polarization and magnetization field operators. These microscopic fields contain the full electric and magnetic multipole series, and couple directly to the quantized electromagnetic field through interaction terms of the same form as their first-quantized counterparts. While an analogous reformulation has been introduced previously [19, 20], the focus of those studies was the fields in the vicinity of an atom or molecule [21] and the impact of the associated local field corrections on intermolecular interactions [22–24]. Our focus instead concerns the vacuum structure of the theory, which is responsible for a vast landscape of phenomena in quantum electrodynamics including the Lamb shift [25], spontaneous emission [26], and the Casimir [27] and Casimir-Polder interactions [28].

In this first communication, we use perturbation theory to calculate the leading-order corrections to the electronic energy levels resulting from interactions between the electrons in an isolated atom or molecule and quantum vacuum fluctuations of the electromagnetic field. Such perturbative calculations generically yield ultraviolet divergences, necessitating regularization and renormalization. We renormalize the multipolar Hamiltonian at leading-order in Rayleigh-Schrödinger perturbation theory, using standard techniques from effective field theory [2]. Our renormalized energy shift constitutes a finite and observable shift of the electronic energy levels. We confirm that for a single electron atom this energy shift reduces to Bethe’s result for the Lamb shift in the electric dipole approximation [29], and its generalizations when spatial variations in the electromagnetic field over the atom are included agrees with the results of other authors [31, 32] who extended Bethe’s result in this direction. We conclude by expanding this renormalized energy shift in a sum of contributions coming from the first few electric and magnetic multipole moments, and quote the expressions for higher-order multipole contributions.

**II. HAMILTONIAN THEORY**

A. Minimal coupling

We consider a system of electrons interacting with one or more ions at fixed positions clustered around a specific point \( \mathbf{R} \) representing, e.g., the center of mass of a molecule. Denoting by \( Q_N \) the charge of the \( N^{th} \) ion located at \( \mathbf{d}_N \) with respect to \( \mathbf{R} \), the charge density of the ions is

\[
\rho_{\text{ion}}(\mathbf{x}) = \sum_N Q_N \delta(\mathbf{x} - \mathbf{R} - \mathbf{d}_N).
\]  

The ionic charge density \( \rho_{\text{ion}}(\mathbf{x}) \) leads to an electrostatic interaction between the ions that we ignore under the frozen-ion approximation, together with a fixed background potential through which the electrons propagate.
of the form

$$V(x) = e \sum_{N} \frac{Q_N}{|x - R - d_N|^4},$$

(4)

where $e = -|e|$ is the charge of an electron. The dynamics of non-interacting electrons propagating within this background potential are described as usual by the Schrödinger equation, which in the coordinate representation can be derived from the Lagrange density

$$L_F = \frac{i\hbar}{2} \left( \psi^\dagger(x,t) \dot{\psi}(x,t) - \dot{\psi}^\dagger(x,t) \psi(x,t) \right) - \frac{\hbar^2}{2m} \nabla \psi^\dagger(x,t) \cdot \nabla \psi(x,t) - \psi^\dagger(x,t) V(x) \psi(x,t).$$

(5)

To couple the electron field $\psi(x,t)$ to a classical electromagnetic field, we apply the standard minimal coupling prescription to the Lagrange density above [1]. In four-vector notation, this amounts to the replacement

$$\partial_\mu \rightarrow \partial_\mu - \frac{ie}{\hbar c} a_\mu(x,t),$$

(6)

where the object on the right is a U(1)–covariant derivative and $a_\mu(x,t)$ is the electromagnetic four-potential. In non-relativistic mechanics, this is equivalent to the addition of two terms to the Lagrange density

$$L_F \rightarrow L'_F = \int d^4x \left( \rho^e(x,t) \phi(x,t) + \frac{1}{c} j(x,t) \cdot a(x,t) \right),$$

(7)

where $(\phi(x,t), a(x,t))$ are the U(1)–gauge potentials and we have introduced the electronic charge density

$$\rho^e(x,t) = e \psi^\dagger(x,t) \psi(x,t)$$

(8)

and the current density

$$j(x,t) = \frac{\hbar e}{2mi} \left( \psi^\dagger(x,t) \nabla \psi(x,t) - \nabla \psi^\dagger(x,t) \psi(x,t) \right) - \frac{e^2}{2me} \psi^\dagger(x,t) a(x,t) \psi(x,t).$$

(9)

And to give dynamics to the electromagnetic field we add to (5) a kinetic term

$$L_B = \frac{1}{8\pi} \left( e(x,t) \cdot e(x,t) - b(x,t) \cdot b(x,t) \right)$$

(10)

associated with the free Maxwell equations. The gauge potentials are related to the electric and magnetic fields $(e(x,t), b(x,t))$ through the usual relations [6]

$$e(x,t) = -\nabla \phi(x,t) - \frac{1}{c} \frac{\partial a(x,t)}{\partial t},$$

(11)

$$b(x,t) = \nabla \times a(x,t).$$

(12)

The total Lagrangian for non-relativistic quantum electrodynamics in minimal coupling is a sum of three terms

$$L = L_B + L_F + L_{\text{int}},$$

(13)

where $L_B$ and $L_F$ are integrals over $\mathbb{R}^3$ of the Lagrange densities given in (10) and (5), and

$$L_{\text{int}} = -\int d^3x \rho^e(x,t) \phi(x,t) + \frac{1}{c} \int d^3x j(x,t) \cdot a(x,t).$$

(14)

To obtain the Hamiltonian field theory, we apply the Legendre transformation to $L$. The canonical momentum conjugate to the electron field $\psi(x,t)$ is

$$\pi_\psi(x,t) = \frac{\delta L}{\delta \dot{\psi}(x,t)} = \frac{i\hbar}{2} \psi^\dagger(x,t),$$

(15)

while the canonical momentum conjugate to the vector potential $a(x,t)$ is

$$\pi_a(x,t) = \frac{\delta L}{\delta \dot{a}(x,t)} = -\frac{1}{4\pi c} e(x,t).$$

(16)

However, the canonical momentum $\pi_\phi(x,t)$ conjugate to the scalar potential $\phi(x,t)$ vanishes. Thus the Lagrangian is degenerate, and the standard method for constructing a Hamiltonian theory involves the identification and classification of first- and second-class constraints using the Poisson bracket, elimination of the former by fixing a gauge, and enforcing the latter by replacing the Poisson bracket by a suitable generalization called the Dirac bracket [33]. The solution in the case of electrodynamics is well-known, and so we only summarize the main results [34, 35]. There are two constraints: The vanishing of $\pi_\phi(x,t)$, and Gauss’s law, $\nabla \cdot e(x,t) = 4\pi \rho^e(x,t)$. After fixing the transverse gauge

$$\nabla \cdot a(x,t) = 0,$$

(17)

the scalar potential is obtained from the electronic charge density through Poisson’s equation

$$\nabla^2 \phi(x,t) = 4\pi \rho^e(x,t)$$

(18)

and the canonical variable $\pi_\phi(x,t)$ can be discarded. The gauge-fixed Maxwell Lagrangian $L'_B$ is

$$L'_B = \frac{1}{8\pi} \int d^3x \left( \frac{1}{c^2} |\dot{a}(x,t)|^2 - |\nabla \times a(x,t)|^2 \right),$$

(19)

where $|x| = \sqrt{x \cdot x}$ is the Euclidean 2-norm, while the gauge-fixed interaction term is

$$L'_{\text{int}} = \frac{1}{c} \int d^3x j(x,t) \cdot a(x,t) - \frac{1}{2} \int d^3x \rho^e(x,t) \phi(x,t).$$

(20)

Using Poisson’s equation, the contribution to the Maxwell Lagrangian $L_B$ coming from the longitudinal electric field (absent in the gauge-fixed $L'_B$) is rewritten in terms of the electronic charge density and scalar potential; this is the origin of the factor $1/2$ in the second term above. After gauge-fixing, Gauss’s law becomes a second-class constraint and is enforced upon quantization by replacing Dirac brackets of the remaining canonical
variables with (anti)commutators, the result being the equal-time commutation relations

\[ [a_t^i(x,t), e_T^j(y,t)] = 4\pi i\hbar \delta^i_j(x - y), \]

\[ [a_t^i(x,t), a_t^j(y,t)] = [e_T^j(y,t), e_T^i(x,t)] = 0, \]

from which follows

\[ [e_T^j(x,t), b^j(y,t)] = 4\pi i\hbar \varepsilon^{ijk} \frac{\partial}{\partial y^k} \delta(x - y), \]

(21)

(22)

(23)

(24)

In (21) we have introduced the transverse delta function \( \delta^i_j(x - y) \), which is used together with the longitudinal delta function \( \delta^i_j(x - y) \) to decompose a given vector field \( O^i(x) \) into its transverse and longitudinal components

\[ O_T^i(x,t) = \int dy \delta^i_j(x - y)O^j(y,t), \]

\[ O_L^i(x,t) = \int dy \delta^i_j(x - y)O^j(y,t). \]

(25)

(26)

The Hamiltonian operator that we obtain from \( L \) after quantization is given in the Schrödinger picture by

\[ H = H_B \otimes \mathbb{1}_F + \int dx \psi^\dagger(x)\hat{H}(x)\psi(x), \]

(27)

where \( \hat{H}(x) = H_B \otimes \mathbb{1}_F + \int dy s(x; y, R)\rho(y) \)

is in a sense “replaced” by a freedom in precisely how the polarization and magnetization fields are defined; the moments of the polarization and magnetization fields are identified with the multipole moments of the charge-current distribution.

In the fully quantum theory, instead of a canonical transformation there is a corresponding unitary transformation that acts on the composite Hilbert space \( \mathcal{H}_B \otimes \mathcal{H}_F \), generated by

\[ S = \frac{1}{c} \int dx p(x) \cdot a(x), \]

(30)

where \( p(x) \) is the polarization field operator, which is related to the total charge density operator

\[ \rho(x) = \rho^c(x) + \rho^{ion}(x) \]

(31)

through the first of the identities in (1), namely

\[ \rho(x) = -\nabla \cdot p(x) + \rho_F(x). \]

(32)

The free charge density \( \rho_F(x) \) is nonzero only if the system has net charge, in which case

\[ \rho_F(x) = Q \delta(x - R) \]

(33)

with net charge

\[ Q = \int dx \rho^c(x) + \sum N Q_N. \]

(34)

We allow \( Q \) to be non-zero to include single ions or charged molecules. A suitable polarization field operator that satisfies (32) is

\[ p(x) = \int dy s(x; y, R)\rho(y), \]

(35)

where we have introduced a “relator” \( s(x; y, R) \) [36], defined by the distributional expression

\[ s(x; y, R) = \int_{C(y, R)} dz \delta(x - z) \]

(36)

with \( C(y, R) \) an arbitrary continuously differentiable curve in \( \mathbb{R}^3 \) that begins at \( R \) and ends at \( y \).

The original Hamiltonian operator (27) can be written as a functional of the canonical variables

\[ H = \mathcal{F}[a, \pi_a, \psi, \pi_\psi], \]

(37)

where \( \pi_a(x) \) and \( \pi_\psi(x) \) are the canonical momenta defined in (16) and (15), respectively. Under the unitary transformation \( U = \exp(iS/\hbar) \), the new canonical variables (denoted by a tilde) are

\[ \tilde{a}(x) = Ua(x)U^{-1}, \]

\[ \tilde{\pi}_a(x) = U\pi_a(x)U^{-1}, \]

\[ \tilde{\psi}(x) = U\psi(x)U^{-1}, \]

\[ \tilde{\pi}_\psi(x) = U\pi_\psi(x)U^{-1}. \]

(38)
The strategy then [36] is to write the Hamiltonian as a functional of the transformed fields,
\[ H = \mathcal{F}[\tilde{a}, \tilde{\pi}_a, \tilde{\psi}, \tilde{\pi}_\psi]. \] (39)
Equating (37) and (39) and using the general form of the transformations (38), the new functional \( \mathcal{F} \) is obtained from the old one \( \mathcal{F} \) through
\[ \mathcal{F}[a, \pi_a, \psi, \pi_\psi] = U^{-1} \mathcal{F}[a, \pi_a, \psi, \pi_\psi] U, \] (40)
where on both sides we use the non-transformed fields. Once the new functional \( \mathcal{F} \) is identified, the multipolar Hamiltonian is obtained by substituting the transformed field operators into this new functional. The explicit transformations of the field operators are
\[ \tilde{a}(x) = a(x), \]
\[ \tilde{\pi}_a(x) = \pi_a(x) - \frac{i}{c} p_T(x), \]
\[ \tilde{\psi}(x) = e^{-i \Phi(x, R)} \psi(x), \]
\[ \tilde{\pi}_\psi(x) = e^{+i \Phi(x, R)} \pi_\psi(x), \] (41)
where
\[ \Phi(x, R) = \frac{e}{\hbar c} \int dr \, s^i(r; x, R)a^i(r). \] (42)
In classical or semiclassical theory the quantity \( \Phi(x, R) \) is a generalized Peierls phase [37]; in the fully quantum theory, the exponential of this quantity in (41) is the Wilson line operator of Abelian gauge theory [38]. With the canonical momentum conjugate to the vector potential given in (16), the transformed transverse electric field is \( \hat{e}_T(x) = \hat{e}_T(x) + 4\pi \mathbf{p}_T(x) \), which we identify with the transverse part of the displacement field \( \mathbf{d}_T(x) \), while the magnetic field is unchanged, \( \hat{b}(x) = b(x) \). The multipolar Hamiltonian operator (39) is
\[ H = H_B \otimes \mathbb{I}_F + \mathbb{I}_B \otimes H_F + H_{\text{int}}, \] (43)
where \( H_B \) is now written in terms of the transverse displacement field instead of the transverse electric field
\[ H_B = \frac{1}{8\pi} \int dx \left( \mathbf{d}_T(x) \cdot \mathbf{d}_T(x) + \mathbf{b}(x) \cdot \mathbf{b}(x) \right), \] (44)
the electronic term is
\[ H_F = \frac{\hbar^2}{2m} \int dx \, \nabla \tilde{\psi}^\dagger(x) \cdot \nabla \tilde{\psi}(x) + 2\pi \int dx \| \mathbf{p}_L(x) \|^2 - \int dx \mathbf{d}_L(x) \cdot \left( \mathbf{p}(x) - \frac{1}{8\pi} \mathbf{d}_L(x) \right), \] (45)
and the interaction term is
\[ H_{\text{int}} = 2\pi \int dx \| \mathbf{d}_T(x) \|^2 - \int dx \, \mathbf{p}(x) \cdot \mathbf{d}_T(x) - \int dx \, \hat{\mathbf{m}}_P(x) \cdot \mathbf{b}(x) - \frac{1}{2} \int dx \, \hat{\mathbf{m}}_D(x) \cdot \mathbf{b}(x). \] (46)

The longitudinal displacement field, defined through Gauss’ law, \( \nabla \cdot \mathbf{d}_L(x) = 4\pi \rho_F(x) \), is given by
\[ \mathbf{d}_L(x) = \frac{1}{4\pi} \int dy \, \rho_F(y) \left( \frac{x - y}{|x - y|^3} \right). \] (47)

In the interaction term (46), \( \hat{\mathbf{m}}_P(x) \) and \( \hat{\mathbf{m}}_D(x) \) refer to the paramagnetic and diamagnetic contributions to the (orbital) magnetization field operator
\[ \hat{\mathbf{m}}(x) = \hat{\mathbf{m}}_P(x) + \hat{\mathbf{m}}_D(x). \] (48)
Explicitly, the field operators \( \hat{\mathbf{m}}_P(x) \) and \( \hat{\mathbf{m}}_D(x) \) are
\[ \hat{\mathbf{m}}_P(x) = \frac{1}{c} \int dy \, \alpha^{ij}(x; y, R) \hat{\mathbf{j}}_P(y), \] (49)
\[ \hat{\mathbf{m}}_D(x) = \frac{1}{c} \int dy \, \alpha^{ij}(x; y, R) \hat{\mathbf{j}}_D(y), \] (50)
where the paramagnetic and diamagnetic current density operators are
\[ \hat{\mathbf{j}}_P(x) = \frac{\hbar c}{2m} \left( \hat{\psi}^\dagger(x) \nabla \hat{\psi}(x) - \nabla \hat{\psi}^\dagger(x) \hat{\psi}(x) \right), \] (51)
\[ \hat{\mathbf{j}}_D(x) = -\frac{e^2}{mc} \hat{\psi}^\dagger(x) \Omega_R(x) \hat{\psi}(x), \] (52)
and we have introduced another “relator,”
\[ \alpha^{ij}(x; y, R) = \varepsilon^{ijmn} \int_{c(y, R)} dz_m \frac{\partial z^n}{\partial y^l} \delta(x - z). \] (53)
In multipolar electrodynamics, the diamagnetic current density (52) depends not on the vector potential, but instead on the magnetic field, through
\[ \Omega_R(x) = \int dy \, \alpha^{ij}(y; x, R) b^j(y). \] (54)
To summarize the above discussion we repeat the full Hamiltonian of multipolar quantum electrodynamics, where now and henceforth we drop the tilde over the transformed variables:
\[ H = H_B \otimes \mathbb{I}_F + \mathbb{I}_B \otimes H_F + H_{\text{int}}, \] (55)
where the Maxwell term is
\[ H_B = \frac{1}{8\pi} \int dx \left( \mathbf{d}_T(x) \cdot \mathbf{d}_T(x) + \mathbf{b}(x) \cdot \mathbf{b}(x) \right), \] (56)
the electronic term is
\[ H_F = \frac{\hbar^2}{2m} \int dx \, \nabla \tilde{\psi}^\dagger(x) \cdot \nabla \tilde{\psi}(x) + 2\pi \int dx \| \mathbf{p}_L(x) \|^2 - \int dx \mathbf{d}_L(x) \cdot \left( \mathbf{p}(x) - \frac{1}{8\pi} \mathbf{d}_L(x) \right), \] (57)
and the interaction term is
\[ H_{\text{int}} = 2\pi \int \mathbf{dx} \, |\mathbf{p}_T(x)|^2 - \int \mathbf{dx} \, \mathbf{p}(x) \cdot \mathbf{d}_T(x) \]
\[ - \int \mathbf{dx} \, \mathbf{m}_T(x) \cdot \mathbf{b}(x) - \frac{1}{2} \int \mathbf{dx} \, \mathbf{m}_D(x) \cdot \mathbf{b}(x). \]  \hspace{1cm} (58)

The charge and current densities are related to the microscopic polarization and magnetization fields through
\[ \rho(x,t) = -\nabla \cdot \mathbf{p}(x,t) + \rho_F(x), \]  \hspace{1cm} (59)
\[ j(x,t) = \frac{\partial \mathbf{p}(x,t)}{\partial t} + e \nabla \times \mathbf{m}(x,t). \]  \hspace{1cm} (60)

III. RENORMALIZATION STRATEGY

The composite Hilbert space of multipolar quantum electrodynamics is the tensor product \( \mathcal{H}_B \otimes \mathcal{H}_F \) of the Hilbert spaces \( \mathcal{H}_B \) and \( \mathcal{H}_F \) associated with the Bose and Fermi sectors, respectively. Since implicit in the Hamiltonian \( H \) is the assumption that the electron particle number is conserved, we introduce an ultraviolet (UV) cutoff \( \Lambda \), excluding contributions to Fourier integrals coming from electromagnetic field modes of \( |\mathbf{k}| > \Lambda \); we denote by \( \tilde{H}^\Lambda \) our Hamiltonian \( H \) subject to this cut-off, which we will henceforth refer to as the \textit{regularized Hamiltonian}. Choosing \( \Lambda \) to be on the order of the inverse (reduced) Compton wavelength of the electron, this guarantees that we are restricting ourselves to energies for which electron-positron pair production that we neglect would not be present in the full quantum theory of electrodynamics [39], of which the multipolar quantum electrodynamics that we consider here is an approximation.

In the next section we will use Rayleigh-Schrödinger perturbation theory to compute the leading-order corrections to the electronic energy levels resulting from interactions between the electrons and quantum vacuum fluctuations in the electromagnetic field. Were \( \Lambda \) taken to infinity, the typical ultraviolet divergences would result. With the “hard cut-off” imposed each regularized energy shift, denoted by \( \Delta E_n^\Lambda \), is finite, albeit explicitly dependent on \( \Lambda \). However, this dependence on \( \Lambda \) is unphysical and should not appear in observable quantities; it arises because \( \Delta E_n^\Lambda \) contains an unobservable, \( \Lambda \)-dependent contribution coming from \textit{free electrons} interacting with the electromagnetic vacuum. Since this contribution is unobservable, we should subtract it from \( \Delta E_n^\Lambda \); what remains will be the observable energy shift of the \textit{bound} state electronic energy levels.

This can be implemented by adding one or more local interactions \( \delta H(\Lambda) \), depending explicitly on \( \Lambda \), to the regularized Hamiltonian \( \tilde{H}^\Lambda \) [40]. In general, \( \delta H(\Lambda) \) will be a sum of many terms, each summand being called a \textit{counterterm}. A central tenet of renormalization theory is that it should be possible to choose the counterterms in \( \delta H(\Lambda) \) such that the \textit{renormalized Hamiltonian}
\[ H^R = \tilde{H}^\Lambda + \delta H(\Lambda) \]  \hspace{1cm} (61)
accurately represents the full range of energies in the interval \( 0 \leq |\mathbf{k}| < \infty \) and should \textit{not} depend on \( \Lambda \). The \textit{renormalized} energy shift, denoted by \( \Delta E_n^R \) and computed using \( H^R \), should be finite in the limit \( \Lambda \to \infty \) and represents the observable correction to the electronic energy levels in the electromagnetic vacuum.

This is the usual strategy we implement here. However, values of \(|\mathbf{k}|\) above the inverse reduced Compton wavelength of the electron are treated improperly by the theory, and so extensions of \( \Lambda \) greater than that go beyond the validity of the Hamiltonian. And, indeed, non-relativistic QED is a \textit{non-renormalizable} field theory, so we only expect \( \Delta E_n^R \) to be finite to the order of perturbation theory at which we renormalized the Hamiltonian, since additional divergences are generated in non-renormalizable theories as one moves to higher orders, as well as divergences that are generated by the counterterms in \( \delta H(\Lambda) \). If we want to compute observables at next-to-leading-order, for example, we must check if additional divergences or \( \Lambda \)-dependent terms are generated. In this paper we will only renormalize the multipolar Hamiltonian at leading-order in the fine structure constant.

IV. ENERGY SHIFTS

A. Preliminaries

We begin by identifying the contributions to the energy shifts arising from the free fields. For the radiation field this is standard, but we take the opportunity to introduce notation we will use later in the paper; for the electrons the computation is done in terms of fermionic scalar fields rather than the single or few-electron states that are usually used. Finally, we identify the two contributions that arise from the interaction of the radiation and electron fields. They are worked out in detail in Sections IV.B and IV.C below.

1. Free fields

The dynamical evolution of the transverse Maxwell fields \( \mathbf{d}_T(x,t) \) and \( \mathbf{b}(x,t) \) is generated by \( H_B \) through the Heisenberg equations
\[ \frac{\partial \mathbf{d}_T(x,t)}{\partial t} = \frac{1}{i\hbar} [\mathbf{d}_T(x,t), H_B]_-, \]  \hspace{1cm} (62)
\[ \frac{\partial \mathbf{b}(x,t)}{\partial t} = \frac{1}{i\hbar} [\mathbf{b}(x,t), H_B]_-, \]  \hspace{1cm} (63)
which, with the explicit form (56) for \( H_B \), lead to the familiar Maxwell equations
\[ \frac{\partial \mathbf{d}_T(x,t)}{\partial t} = +e \nabla \times \mathbf{b}(x,t), \]  \hspace{1cm} (64)
\[ \frac{\partial \mathbf{b}(x,t)}{\partial t} = -e \nabla \times \mathbf{d}_T(x,t), \]  \hspace{1cm} (65)
subject to the constraints
\[ \nabla \cdot d_F(x, t) = 0, \quad \nabla \cdot b(x, t) = 0. \]

We quantize in a box of volume \( V = L^3 \), in which case the allowed wavevectors are
\[ k = \frac{2\pi}{L} (n_x, n_y, n_z) \in \frac{2\pi}{L} \mathbb{Z}^3, \]
and expand \( d_F(x, t) \) and \( b(x, t) \) in a Fourier series of transverse spatial modes, which can themselves be written as a sum of \( \mathbb{C} \)-valued circular polarization vectors \( e_{lk} \) (\( I = L, R \)) defined by
\[
e_{lk} = -\frac{1}{\sqrt{2}} (\varepsilon_{1k} + i\varepsilon_{2k}),
e_{lk} = +\frac{1}{\sqrt{2}} (\varepsilon_{1k} - i\varepsilon_{2k}),
\]
where \( \varepsilon_{1k} \) and \( \varepsilon_{2k} \) are the standard Cartesian polarization vectors \([18]\). The circular polarization vectors satisfy
\[ e_{lk} \cdot e_{lj(-k)} = \delta_{ll}, \]
\[ i\hbar \times e_{lk} = s_l e_{lk}, \]
where \( s_L = +1 \) and \( s_R = -1 \), and we choose the convention \( e_{l(-k)} = e_{lk}^\dagger \). Moreover, these vectors satisfy the polarization sum rule
\[ \sum_i e_{il(-k)}^i e_{lk}^j = \delta_{ij} - \frac{k^i k^j}{|k|^2}. \]

The Maxwell field operators are
\[ d_F(x, t) = i \sum_{lk} \left( \frac{2\pi \hbar \omega_k}{V} \right)^{1/2} e_{lk} a^\dagger_{lk} e^{i(k \cdot x - \omega_k t)} + \text{H.c.}, \]
\[ b(x, t) = \sum_{lk} \left( \frac{2\pi \hbar \omega_k}{V} \right)^{1/2} s_l e_{lk} a_{lk} e^{i(k \cdot x - \omega_k t)} + \text{H.c.}, \]
where \( \omega_k = c|k| \). To enforce the commutation relations (17-23), the photonic creation and annihilation operators \( a^\dagger_{lk} \) and \( a_{lk} \) must obey the equal-time canonical commutation relations
\[ [a_{lk}, a_{lj}^\dagger] = \delta_{lj} \delta_{kk'}, \]
\[ [a_{lk}, a_{lj}^\dagger] = [a_{lk}^\dagger, a_{lj}^\dagger] = 0. \]

After regularization, the Maxwell Hamiltonian \( H_B \) is given in the Schrödinger picture by
\[ H_B^\Lambda = \sum_l \sum_{|k| \leq \Lambda} \hbar \omega_k \left( a_{lk}^\dagger a_{lk} + \frac{1}{2} \right). \]

The second term is the well-known divergence associated with the zero-point energy in free quantum electrodynamics \([1, 26]\). Summing over polarization states, the energy of the vacuum state \( |\text{vac}\rangle \in \mathcal{H}_B \) is exactly equal to this zero-point energy
\[ E_{0,B}^\Lambda \equiv \sum_{|k| \leq \Lambda} \hbar c|k| \rightarrow \frac{\hbar V}{8\pi^2} \Lambda^4, \]
where the right side follows from the continuum limit
\[ \sum_k \rightarrow \frac{V}{8\pi^3} \int dk. \]

Therefore, the regularized Maxwell Hamiltonian is
\[ H_B^\Lambda = \sum_l \sum_{|k| \leq \Lambda} \hbar \omega_k a_{lk}^\dagger a_{lk} + E_{0,B}^\Lambda. \]

Since we are primarily interested in vacuum effects, we take as our basis for \( \mathcal{H}_B \) the photon number states \( |\{n_{lk}\}\rangle \in \mathcal{H}_B \). The spectral problem for \( H_B^\Lambda \) in terms of these eigenstates is
\[ H_B^\Lambda |\{n_{lk}\}\rangle = E_B^\Lambda |\{n_{lk}\}\rangle, \]
where the energy eigenvalues are
\[ E_B^\Lambda = \sum_l \sum_{|k| \leq \Lambda} \hbar \omega_k n_{lk} + E_{0,B}^\Lambda. \]

Turning to the electron field, time evolution in the electronic Hilbert space \( \mathcal{H}_F \) is generated by \( H_F \) through the Heisenberg equation
\[ \frac{\partial}{\partial t} \psi(x, t) = \frac{i}{\hbar} \left[ \psi(x, t), H_F \right], \]
with \( H_F \) given by (57). As shown in Appendix A, the regularized electronic Hamiltonian \( H_F \) is given in the Schrödinger picture by
\[ H_F^\Lambda = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + \frac{1}{2} \int dx dx' \psi^\dagger(x) \psi^\dagger(x') \frac{e^2}{|x - x'|} \psi(x') \psi(x) + \int dx \psi^\dagger(x) V(x) \psi(x) + E_{0,F}^\Lambda, \]
where \( E_{0,F}^\Lambda \) is a \( \Lambda \)-dependent term associated with the \( r \rightarrow 0 \) limit of the electrostatic Coulomb potential
\[ E_{0,F}^\Lambda = \frac{1}{\pi} \left[ e^2 N_e + 2e N_e \sum_N Q_N + \sum_N Q_N^2 \right] \Lambda. \]

We take as our basis for \( \mathcal{H}_F \) the set \( \{|\psi_n\rangle\}_n \) of many-body eigenstates of \( H_F^\Lambda \). The spectral problem for \( H_F^\Lambda \) in terms of these eigenstates is
\[ H_F^\Lambda |\psi_n\rangle = E_n^\Lambda |\psi_n\rangle, \]
where the energy eigenvalues are

\[ E^\Lambda_n = \frac{\hbar^2}{2m} \int dx \left\langle \nabla \psi^\dagger(x) \cdot \nabla \psi(x) \right\rangle_n + \frac{1}{2} \int dxdx' \left\langle \psi^\dagger(x)\psi^\dagger(x') \frac{e^2}{|x - x'|} \psi(x')\psi(x) \right\rangle_n + \int dx \left\langle \psi^\dagger(x)V(x)\psi(x) \right\rangle_n + E^\Lambda_{0,F}, \tag{86} \]

where the expectation value is over the state \( |\psi_n\rangle \). There are a host of techniques in atomic and molecular physics, and in condensed matter physics, that have been designed to solve spectral problems of the form given in (86) [41].

2. Interactions

In the interacting theory, the dynamical evolution of the field operators is generated by the full Hamiltonian \( H \) given in (55-60). After regularization, we write this Hamiltonian as

\[ H^\Lambda = H^\Lambda_0 + H^\Lambda_{\text{int}}, \tag{87} \]

where

\[ H^\Lambda_0 = H^\Lambda_B \otimes I_F + I_B \otimes H^\Lambda_F \tag{88} \]

with \( H^\Lambda_B \) and \( H^\Lambda_F \) given in (79) and (83), while \( H^\Lambda_{\text{int}} \) is given by (58) with all of the Fourier series (implicit in the mode expansions (73)) subject to the cutoff \( |k| \leq \Lambda \).

We take as our basis for the composite Hilbert space \( \mathcal{H}_B \otimes \mathcal{H}_F \) the product states

\[ |\{n_{1k}\};\psi_n\rangle \equiv |\{n_{1k}\} \rangle \otimes |\psi_n\rangle. \tag{89} \]

The spectral problem for the regularized free Hamiltonian \( H^\Lambda_0 \) in terms of these eigenstates is

\[ H^\Lambda_0 |\{n_{1k}\};\psi_n\rangle = \left( E^\Lambda_B + E^\Lambda_n \right) |\{n_{1k}\};\psi_n\rangle, \tag{90} \]

with \( E^\Lambda_B \) and \( E^\Lambda_n \) given by (81) and (86), respectively. We want to compute the regularized energy shift \( \Delta E^\Lambda_n \) of the electronic energy levels \( E^\Lambda_n \) in the electromagnetic vacuum, resulting from interactions described by \( H^\Lambda_{\text{int}} \). Since we will do so using perturbation theory at \( \mathcal{O}(\alpha) \), it is useful to split up the interaction term

\[ H^\Lambda_{\text{int}} = H^\Lambda_{\text{int}(1)} + H^\Lambda_{\text{int}(2)}, \tag{91} \]

where

\[ H^\Lambda_{\text{int}(1)} = 2\pi \int dx \| p_T(x) \|^2 + \frac{1}{2} \int dx \mathbf{m}_D(x) \cdot \mathbf{b}(x) \tag{92} \]

are the interactions that are already \( \mathcal{O}(\alpha) \) and will be computed at the first order, while

\[ H^\Lambda_{\text{int}(2)} = -\int dx \mathbf{p}(x) \cdot \mathbf{d}_T(x) - \int dx \mathbf{m}_P(x) \cdot \mathbf{b}(x) \tag{93} \]

are the interactions that do not contribute to \( H^\Lambda_{\text{int}(1)} \), since the vacuum expectation values of \( \mathbf{d}_T(x) \) and \( \mathbf{b}(x) \) vanish; these will be computed at the second order. Define

\[ |\Psi_n\rangle \equiv |\text{vac};\psi_n\rangle, \tag{94} \]
\[ |\Psi_\delta\rangle \equiv |1_{1k};\psi_\delta\rangle. \tag{95} \]

In terms of these states, the regularized correction to the electronic energy levels \( E^\Lambda_n \) is

\[ \Delta E^\Lambda_n = \Delta E^\Lambda_{n,(1)} + \Delta E^\Lambda_{n,(2)}, \tag{96} \]

where

\[ \Delta E^\Lambda_{n,(1)} = \langle \Psi_n | H^\Lambda_{\text{int}(1)} | \Psi_n \rangle \tag{97} \]

is the first-order correction, while

\[ \Delta E^\Lambda_{n,(2)} = \sum_{\delta \neq n} \left| \langle \Psi_\delta | H^\Lambda_{\text{int}(2)} | \Psi_n \rangle \right|^2 \tag{98} \]

is the second-order correction.

B. First-order correction

The first-order correction is

\[ \Delta E^\Lambda_{n,(1)} = -\frac{1}{2} \int dx \langle \Psi_n | \mathbf{m}_D(x) \cdot \mathbf{b}(x) | \Psi_n \rangle + 2\pi \int dx \langle \Psi_n | \| \mathbf{p}_T(x) \|^2 | \Psi_n \rangle. \tag{99} \]

The first term is the diamagnetic contribution to the regularized energy shift, which is more commonly written as

\[ + \frac{1}{2} \int dxdx' \langle \Psi_n | \mathbf{b}^\dagger(x) \mathbf{O}^{ij}(x, x') \mathbf{b}(x') | \Psi_n \rangle, \tag{100} \]

where

\[ \mathbf{O}^{ij}(x, x') = \int dy \psi^\dagger(y) \mathbf{o}^{ij}(y; x', y) \psi(y) \tag{101} \]

is the second-quantized diamagnetization field [36]

\[ \mathbf{o}^{ij}(x, x'; y) = \frac{e^2}{2mc^2} \alpha^{ik}(x; y, \mathbf{R}) \alpha^{jk}(x'; y, \mathbf{R}). \tag{102} \]

We show in Appendix B that the diamagnetic contribution to the first-order correction in the continuum limit (78) is given by

\[ -\frac{1}{2} \int dx \langle \Psi_n | \mathbf{m}_D(x) \cdot \mathbf{b}(x) | \Psi_n \rangle = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 \]

\[ - \frac{\hbar}{mc} \int dx \rho_{mn}(x) \langle \mathbf{a}(x) \cdot \nabla \Phi(x, \mathbf{R}) \rangle_{\text{vac}} \]

\[ + \frac{\hbar^2}{2mc} \int dx \rho_{mn}(x) \langle \| \nabla \Phi(x, \mathbf{R}) \|^2 \rangle_{\text{vac}}. \tag{103} \]
where $N_e$ is the total number of electrons, and the expectation value is taken in the vacuum state of the radiation field. We will see below that when the full interaction shift is calculated the latter two terms are cancelled by terms in the second-order correction $\Delta E_{n,(2)}^\Lambda$.

Consider next the second term in (99). Inserting a resolution of the identity in $\mathcal{H}_F$ with respect to the many-body eigenstates $\{ |\psi_m \rangle \}_m$, we can (after regularization) write this term as

$$\mathcal{I}_F = \sum_m |\psi_m \rangle \langle \psi_m |,$$  

the expectation value

$$\langle \psi_n | p^j(x') p^i(x) | \psi_n \rangle = \sum_m p^j_{nm}(x') p^i_{mn}(x),$$

where $p^i_{nm}(x) \equiv \langle \psi_n | p^i(x) | \psi_m \rangle$. Using the Fourier integral representation of the transverse delta function $\delta_{ij}$

$$\delta_{ij}(x - x') = \int \frac{dk}{(2\pi)^3} \left( \delta_{ij} - \frac{k^i k^j}{|k|^2} \right) e^{ik(x' - x)},$$

we can (after regularization) write this term as

$$2\pi \int dx \int p^j_{nm}(x') p^i_{mn}(x) e^{ik(x' - x)}.$$

To proceed, it is convenient to recast the angular part of the Fourier integral into an expression in coordinate space. Explicitly, we write [19]

$$\frac{1}{2\pi} \int d\Omega_k \left( \delta_{ij} - \frac{k^i k^j}{|k|^2} \right) e^{ik(x' - x)} = \frac{1}{|k|^2} \left( -\delta_{ij} \partial^2 + \partial^i \partial^j \right) F(x, x'; k),$$

where $d\Omega_k = \sin \theta d\theta d\phi$ and $k = |k|$, we have introduced the abbreviated notation $\partial^i \equiv \partial/\partial x^i$ and $\partial^2 \equiv \nabla^2$, and

$$F(x, x'; k) \equiv \frac{\sin |k| x - x'|}{k|x - x'|}.$$ Relating the polarization field operators to the total charge density through (59), the contribution to $\Delta E_{n,(1)}^\Lambda$ coming from the transverse polarization fields is

$$2\pi \int dx \int \Psi_n | p_T(x) |^2 \Psi_n = \frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dxdx' \left[ -\delta_{ij} \partial^2 + \partial^i \partial^j \right] F(x, x'; k) p^i_{mn}(x') p^j_{nm}(x).$$

Hence the total first-order correction is

$$\Delta E_{n,(1)}^\Lambda = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 + \frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dxdx' \left[ -\delta_{ij} \partial^2 + \partial^i \partial^j \right] F(x, x'; k) p^i_{mn}(x') p^j_{nm}(x)$$

$$- \frac{\hbar}{mc} \int dx \rho^\text{c}_{nm}(x) \langle a(x) \cdot \nabla \Phi(x, R) \rangle_{\text{vac}} + \frac{\hbar^2}{2mc} \int dx \rho^\text{c}_{nm}(x) \langle \| \nabla \Phi(x, R) \|^2 \rangle_{\text{vac}}.$$

C. Second-order correction

To compute the second-order correction, we begin with the matrix element

$$\langle \Psi_\delta | H_{\text{int}(2)}^\Lambda | \Psi_n \rangle = - \int dx \int dT_{nm}(x) \langle 1_{lk} | d_T(x) | \text{vac} \rangle$$

$$- \int dx \int m^j_{mn}(x) \langle 1_{lk} | b^j(x) | \text{vac} \rangle,$$

where $m^j_{mn}(x) \equiv \langle \psi_m | m^j(x) | \psi_n \rangle$ denotes the paramagnetic contribution to the orbital magnetization. Inserting the mode expansions (73), these matrix elements become

$$\langle \Psi_\delta | H_{\text{int}(2)}^\Lambda | \Psi_n \rangle = i \left( \frac{2\pi \hbar \omega_k}{V} \right)^{1/2} e^{i\delta_{lk}(-k)} \int dx \int m^j_{mn}(x) e^{-ik\cdot x}$$

$$+ \left( \frac{2\pi \hbar \omega_k}{V} \right)^{1/2} s_i e^{i\delta_{lk}(-k)} \int dx \int m^j_{mn}(x) e^{-ik\cdot x}.$$
The energy difference between the states $|\Psi_\delta\rangle$ and $|\Psi_n\rangle$ is $E_\delta - E_n = E_{mn} + \hbar \omega k$, where $E_{mn} \equiv E_m - E_n$. The regularized energy shift is

$$\Delta E_{n,(2)}^\Lambda = -\frac{\hbar c}{4\pi^2} \sum_m \int_{|k| \leq \Lambda} dk |k| \int dx dx' \frac{e^{i k \cdot (x' - x)}}{E_{mn} + \hbar c |k|} \left\{ \delta_{ij} - \frac{k_i k_j}{|k|^2} \right\} \left( p_{i,m}^j(x') p_{j,m}^i(x) + m_{P,nn}^i(x') m_{P,nn}^j(x) \right)$$

and after taking the continuum limit (78), we find

$$\sum_{\delta} \sum_m \sum_l \sum_{|k| \leq \Lambda},$$

The simplification of this expression is largely formulaic and has been relegated to Appendix C. There it is shown that it can be written in terms of (109) as

$$\Delta E_{n,(2)}^\Lambda = -\frac{1}{\pi} \sum_m \int_0^\Lambda dk \frac{\hbar c k}{E_{mn} + \hbar c |k|} \int dx dx' \left[ (\delta_{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{i,m}^j(x') p_{j,m}^i(x) + m_{P,nn}^i(x') m_{P,nn}^j(x) \right)$$

Further simplifications can be made using the relations (59,60). After a tedious but straightforward calculation, the total second-order correction is

$$\Delta E_{n,(2)}^\Lambda = -\frac{\hbar c}{4\pi^2} \sum_m \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \int dx dx' j_{P,nn}^i(x') j_{P,nn}^j(x) \left( \delta_{ij} - \frac{k_i k_j}{|k|^2} \right) e^{i k \cdot (x' - x)}$$

Hence, combining the first-order and second-order corrections, the regularized energy shift is

$$\Delta E_n^\Lambda = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{4\pi^2} \sum_m \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \int dx dx' j_{P,nn}^i(x') j_{P,nn}^j(x) \left( \delta_{ij} - \frac{k_i k_j}{|k|^2} \right) e^{i k \cdot (x' - x)}.$$
where

\[ P = \frac{\hbar}{i} \int dx \psi^\dagger(x) \nabla \psi(x) \]  

(122)

is the total (second-quantized) momentum operator. This distinction highlights the difference between our approach and existing treatments, which generally only apply to hydrogenic atoms. If one is interested in many-electron atoms, molecules, or more general charge-current distributions, then it is necessary to couple the radiation field to the current density rather than the total momentum. In doing so, our result becomes valid to all orders in the multipole expansion.

V. RENORMALIZATION

A. Renormalization of \( H_0^\Lambda \)

We first renormalize the free Hamiltonian \( H_0^\Lambda \), which produces the vacuum divergence \( E_{0,B}^\Lambda \) of free quantum electrodynamics given in (77) and the divergence \( E_{0,F}^\Lambda \) computed in Appendix A. Since both of these divergences are static, we can trivially remove them from \( H_0^\Lambda \) by addition of static counterterms \( \delta H_B(\Lambda) \) and \( \delta H_F(\Lambda) \) to \( H_B^\Lambda \) and \( H_F^\Lambda \). The renormalized free Hamiltonian is

\[ H_R^0 = H_B^\Lambda \otimes 1_F + 1_B \otimes H_F^\Lambda, \]  

(123)

where the renormalized Maxwell and electronic terms are

\[ H_B^\Lambda = H_B + \delta H_B(\Lambda), \]  

(124)

\[ H_F^\Lambda = H_F + \delta H_F(\Lambda), \]  

(125)

and the counterterms (in minimal subtraction) are

\[ \delta H_B(\Lambda) = - \left[ \frac{\hbar c V}{8\pi^2\Lambda^4} \right] 1_B, \]  

(126)

\[ \delta H_F(\Lambda) = - \left[ \frac{1}{\Lambda} \left( e^2 N_e + 2e N_e \sum N + \sum Q_N^2 \right) \right] 1_F. \]  

(127)

The energy eigenvalues computed from \( H_B^\Lambda \) and \( H_F^\Lambda \) are finite in the limit \( \Lambda \to \infty \). In particular, the renormalized electronic energy levels are

\[ E_n^R = \frac{\hbar^2}{2m} \int dx \left( \langle \nabla \psi^\dagger(x) \cdot \nabla \psi(x) \rangle_n + \frac{1}{2} \right) \int \int dx dx' \left( \langle \psi^\dagger(x) \psi(x') \rangle_n \right) + \int \int dx dx' \langle \psi^\dagger(x) V(x) \psi(x') \rangle_n. \]  

(128)

B. Renormalization of \( H_{\text{int}}^\Lambda \)

The renormalized interaction term is defined by

\[ H_{\text{int}}^R = H_{\text{int}}^\Lambda + \delta H_{\text{int}}(\Lambda), \]  

(129)

where \( \delta H_{\text{int}}(\Lambda) \) contains one or more counterterms that we will determine below (in minimal subtraction). As outlined in Section III, our renormalization condition is based on the observation that the \( \Lambda \)-dependent contribution to the regularized energy shift (119) coming from a free electron gas, described by the Hamiltonian

\[ H_F^{(0)} = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x), \]  

(130)

is unobservable and should be subtracted from \( \Delta E_n^\Lambda \). What remains will be the energy shift of the bound state electronic energy levels.

We must first determine the contribution to \( \Delta E_n^\Lambda \) coming from a free electron gas. Denote by \( \{ |\varphi_n\rangle \}_n \) the set of many-body eigenstates of \( H_F^{(0)} \). Because this Hamiltonian commutes with the total momentum operator \( P \), we can choose each energy eigenstate \( |\varphi_n\rangle \) to be an eigenstate of \( P \). Then for an arbitrary many-body eigenstate \( |\varphi_n\rangle \) we have

\[ \Delta E_{\text{free}}^\Lambda \equiv \langle \varphi_n | H_{\text{int}}^\Lambda | \varphi_n \rangle, \]  

(131)

\[ \Delta E_{\text{free}}^R \equiv \langle \varphi_n | H_{\text{int}}^R | \varphi_n \rangle. \]  

(132)

We choose the expectation values of the counterterms in \( \delta H_{\text{int}}(\Lambda) \) to cancel the \( \Lambda \)-dependent terms in \( \Delta E_{\text{free}}^\Lambda \), so that the renormalized energy shift \( \Delta E_{\text{free}}^R \) vanishes for an arbitrary state \( |\varphi_n\rangle \) (i.e., an arbitrary distribution of electron momenta in the gas). From \( \delta H_{\text{int}}(\Lambda) \), we will infer the form of the operator \( \delta H_{\text{int}}(\Lambda) \).

To compute \( \Delta E_{\text{free}}^R \), it is useful to expand the electron field operators in a basis of plane waves

\[ \psi(x) = \frac{1}{\sqrt{V}} \sum q e^{iqx} b_q, \]  

(133)

where we return to considering a system in volume \( V \), and where the anticommutation relation (24) implies that the electronic creation and annihilation operators satisfy the canonical anticommutation relations

\[ [b_q, b_q^\dagger]_+ = \delta_{qq'}, \]  

\[ [b_q, b_q^\dagger]_+ = [b_q^\dagger, b_q^\dagger]_+ = 0. \]  

(134)

In this plane-wave expansion, the Fourier transform of the paramagnetic current density operator is

\[ \vec{j}_\mu(k) = \frac{\hbar e}{2m} \sum_q \left( 2k^2 - k^2 \right) b_q^\dagger \delta_{k,k} b_q. \]  

(135)

We obtain \( \Delta E_{\text{free}}^\Lambda \) by replacing the many-body eigenstates of \( H_F \) in (119) with those of \( H_R^{(0)} \), and replacing \( E_{mn} \) with \( E_{mn}^{(0)} \), where \( E_{mn}^{(0)} = \langle \varphi_n | H_F^{(0)} | \varphi_m \rangle \). Thus, for a free electron gas the regularized energy shift is
\[ \Delta E_{\text{free}}^\Lambda = \frac{\alpha}{2\pi} \left( \frac{h^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{4\pi^2 c} \sum_{m} \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \sum_{l} |e_I(k) \cdot \tilde{j}_{P,mn}(k)|^2. \]  

(136)

To simplify the second term, consider the expression

\[ \sum_m \tilde{j}_P^I(-k) \frac{1}{E_{mn}^0 + \hbar c |k|} \tilde{j}_P^J(k) = \left\langle \tilde{j}_P^I(-k) \left( \frac{1}{H_p^0 - E_{mn}^0 + \hbar c |k|} \right) \tilde{j}_P^J(k) \right\rangle_n, \]  

(137)

where the subscript \( mn \) indicates the matrix element between state \(|\phi_m\rangle\) and state \(|\phi_n\rangle\). We simplify the right side of this equality using the Baker-Campbell-Hausdorff (BCH) formula [12]

\[ \frac{1}{H_p^0 - E_{mn}^0 + \hbar c |k|} = -i \int_0^\infty ds e^{isH_p^0} \left\langle \tilde{j}_P^I(-k) e^{-isH_p^0} \right\rangle_{mn}, \]  

(138)

with which (137) can be written

\[ \int_0^\infty ds e^{isH_p^0} \frac{1}{H_p^0 - E_{mn}^0 + \hbar c |k|} = \sum_{m} \int_0^\infty ds e^{isH_p^0} \left\langle \tilde{j}_P^I(-k) e^{-isH_p^0} \right\rangle_{mn}, \]  

(139)

It is straightforward to compute the \( \ell \)th nested commutator above, the result of which is

\[ \left[ H_p^0, \left[ H_p^0, \ldots, \left[ H_p^0, \tilde{j}_P^J(k) \right] \ldots \right] \right] = \frac{\hbar c}{2m} \sum_q \left( 2q^j - k^j \right) b_{q-k}^\dagger b_q \left( E_{hk} - \frac{\hbar k \cdot q}{m} \right)^{(\ell)}, \]  

(140)

where \( E_{hk} = \hbar^2 |k|^2 / 2m \). Thus, the BCH series can be resummed

\[ e^{iH_p^0} \tilde{j}_P^I(k) e^{-iH_p^0} = \frac{\hbar c}{2m} \sum_q \left( 2q^j - k^j \right) b_{q-k}^\dagger b_q e^{i(E_{hk} - \hbar k \cdot q/m)}, \]  

(142)

and so

\[ \sum_m \tilde{j}_P^I(-k) \frac{1}{E_{mn}^0 + \hbar c |k|} \tilde{j}_P^J(k) = \frac{\hbar^2 c^2}{4m^2} \sum_q \left( 2q^i - k^i \right) \left( 2q^j - k^j \right) \left( E_{hk} + \hbar c |k| - \hbar k \cdot q/m \right) \]  

\[ \times \sum_m \langle \phi_n \left| b_q^\dagger b_{q-k}^\dagger \phi_m \right| \left( \phi_m \left| b_{q-k} \phi_n \right. \right). \]

(143)

Since we are working with electrons that are non-relativistic \(|q| \ll mc\), we can drop the \( \hbar k \cdot q/m \) term in the denominator. Recollecting the Fourier transforms of the paramagnetic current operators using (177) we find

\[ \sum_m \tilde{j}_P^I(-k) \frac{1}{E_{mn}^0 + \hbar c |k|} \tilde{j}_P^J(k) = \sum_m \tilde{j}_P^I(-k) \left( \frac{1}{E_{hk} + \hbar c |k|} \right) \tilde{j}_P^J(k), \]  

(144)

from which follows

\[ \Delta E_{\text{free}}^\Lambda = \frac{\alpha}{2\pi} \left( \frac{h^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{4\pi^2 c} \sum_{m} \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{hk} + \hbar c |k|} \sum_{l} |e_I(k) \cdot \tilde{j}_{P,mn}(k)|^2. \]  

(145)

As discussed in Section III, we add to the regularized interaction term a counterterm \( \delta H_{\text{int}}(\Lambda) \) whose \( \Lambda- \)
dependence is chosen so that the renormalized interaction term
\[ H^R_{\text{int}} = H^\Lambda_{\text{int}} + \delta H_{\text{int}}(\Lambda) \] (146)
is finite in the limit \( \Lambda \to \infty \). However, instead of fixing the counterterm itself, we first fix its expectation value in the state \( |\varphi_n\rangle \), after which we will infer its form as an operator. Enforcing our renormalization condition — that is, requiring that \( \Delta E^R_{\text{free}} \) vanishes in an arbitrary eigenstate \( |\varphi_n\rangle \), and therefore an arbitrary distribution of electron momenta — uniquely fixes the expectation value of the counterterm

\[
\langle \delta H_{\text{int}}(\Lambda) \rangle_n = -\frac{\alpha}{2\pi} \left( \frac{h^2 N_e}{m} \right) \Lambda^2 + \frac{h}{4\pi^2 c} \sum_m \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{hk} + \hbar c |k|} \sum_i |e_I(k) \cdot \hat{j}_{P,mn}(k)|^2,
\]

and we therefore conclude that the operator \( \delta H_{\text{int}}(\Lambda) \) is

\[
\delta H_{\text{int}}(\Lambda) = -\left[ \frac{\alpha}{2\pi} \left( \frac{h^2 N_e}{m} \right) \Lambda^2 \right] I_B \otimes I_F + \frac{h}{4\pi^2 c} \sum_m \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{hk} + \hbar c |k|} \sum_i \left( e_I(-k) \cdot \hat{j}_{P,mn}^\dagger(k) \right) \left( e_I(k) \cdot \hat{j}_P(k) \right).
\] (148)

We have now collected all of the counterterms necessary to obtain the full renormalized Hamiltonian from

\[
H^\Lambda = H^\Lambda_{\text{F}} \otimes I_F + I_B \otimes H^\Lambda_{\text{F}} + H^\Lambda_{\text{int}}.
\] (149)
The renormalized Hamiltonian is

\[
H^R = H^\Lambda + \delta H(\Lambda),
\] (150)

where \( H^\Lambda \) is given by (87) and

\[
\delta H(\Lambda) = \delta H_B(\Lambda) \otimes I_F + I_B \otimes \delta H_F(\Lambda) + \delta H_{\text{int}}(\Lambda).
\] (151)

Using (126,127) and (148), the explicit form of the operator \( \delta H(\Lambda) \) is

\[
\delta H(\Lambda) = -\left[ \frac{\alpha}{2\pi} \left( \frac{h^2 N_e}{m} \right) \Lambda^2 \right] I_B \otimes I_F + \frac{h}{4\pi^2 c} \sum_m \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{hk} + \hbar c |k|} \sum_i \left( e_I(-k) \cdot \hat{j}_{P,mn}^\dagger(k) \right) \left( e_I(k) \cdot \hat{j}_P(k) \right).
\] (152)

C. Renormalized energy shift

We are now in a position to calculate the renormalized energy shift
\[
\Delta E^R_n = \langle \text{vac}; \psi_n | H^R_{\text{int}} | \text{vac}; \psi_n \rangle,
\] (153)
where \( |\psi_n\rangle \) is an eigenstate of the renormalized electronic Hamiltonian \( H^R_{\text{elec}} |\psi_n\rangle = E^R_n |\psi_n\rangle \) with eigenvalue \( E^R_n \) given in (128). For later purposes, write \( H^B_{\text{elec}} = H^B_{\text{elec}}^{(0)} + U^R \) with

\[
U^R = \frac{1}{2} \int dx \, dx' \psi^\dagger(x) \psi^\dagger(x') \frac{e^2}{x - x'} \psi(x') \psi(x) + \int dx \, \psi^\dagger(x) V(x) \psi(x).
\] (154)
The static counterterm in \( \langle \delta H_{\text{int}}(\Lambda) \rangle_n \equiv \langle \psi_n | \delta H_{\text{int}}(\Lambda) | \psi_n \rangle \) trivially cancels the static self-energy in \( \Delta E^\Lambda_n \) and we are left with

\[
\Delta E^R_n = -\frac{\hbar}{4\pi^2 c} \sum_m \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E^R_{mn} + \hbar c |k|} \sum_i |e_I(k) \cdot \hat{j}_{P,mn}(k)|^2
\]
\[+ \frac{\hbar}{4\pi^2 c} \sum_m \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{hk} + \hbar c |k|} \sum_i \left( \langle e_I(-k) \cdot \hat{j}_{P,mn}^\dagger(k) \rangle \langle e_I(k) \cdot \hat{j}_P(k) \rangle \right)_n,
\] (155)
the second line being the dynamical counterterm in $\delta H_{\text{int}}(\lambda)$. Introducing a resolution of the identity in $\mathcal{H}_F$ with respect to the states $\{|\psi_m\rangle\}_m$, we can collect these terms together

$$\Delta E_n^R = -\frac{\hbar}{4\pi^2 c} \sum_m \int_{|k| \leq \Lambda} dk \left( \frac{|k|^{-1}}{E_{mn}^R + \hbar c|k|} - \frac{|k|^{-1}}{E_{hk} + \hbar c|k|} \right) \sum_I e_I(k) \cdot \tilde{j}_{P,mn}(k) \right|^2 . \quad (156)$$

To simplify the expression in the parentheses, we use the following identity (proven in Appendix E)

$$\sum_m \tilde{j}_{P,mn}(-k) \frac{1}{E_{mn}^R + \hbar c|k|} \tilde{j}_{P,mn}(k) = \sum_m \tilde{j}_{P,mn}(-k) \frac{1}{E_{hk} + \hbar c|k|} \left( 1 - \frac{U_{mn}^R}{E_{mn}^R + \hbar c|k|} \right). \quad (157)$$

The first term on the right of the above equality exactly cancels the second term in the parentheses of (156), and so the renormalized energy shift is

$$\Delta E_n^R = \frac{\hbar}{4\pi^2 c} \sum_m \int dk \left( \frac{|k|^{-1}}{E_{hk} + \hbar c|k|} \right) \sum_I e_I(k) \cdot \tilde{j}_{P,mn}(k) \right|^2 , \quad (158)$$

where we have removed the regulator, since this expression is finite in the limit $\Lambda \to \infty$. 

Before moving back to the multipole formalism, we first simplify the energy shift in the form above and thereby demonstrate that it is indeed finite. To begin, we approximate the expression (158) by replacing $E_{mn}^R$ in the denominator by a “reference” or average value $E_{mn}^R \to E_n^R$. Through a short calculation we have

$$j_{P,mn}(x)U_{mn}^R = \frac{\hbar e}{m_i} \langle \psi^\dagger(x) \partial^i U^R(x) \psi(x) \rangle_{nm} , \quad (159)$$

where $U^R(x) = \phi(x) + V(x)$ is the total electrostatic potential of the electrons and the ions. Since we have replaced $E_{mn}^R$ in the denominator of (158) by the reference value $E_n^R$, we can evaluate the sum using the identity [18]

$$\sum_m j_{P,mn}(x)j_{P,mn}(x')U_{mn}^R = \frac{1}{2} \left( \left[ j_{P}(x), U_{mn}^R \right] \right)_{nm} \langle \frac{\hbar e}{m_i} \rangle . \quad (160)$$

Evaluating this expression directly is cumbersome. It is easier to evaluate the complete expression

$$\frac{1}{4\pi} \int d\Omega_k \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) \sum_m \tilde{j}_{P,mn}(-k) \tilde{j}_{P,mn}^\dagger(k) U_{mn}^R. \quad (161)$$

The solid angle integral yields $(2/3)\delta_{ij}$, and using

$$\nabla^2 \left( \frac{1}{|x' - x|} \right) = -4\pi \delta(x' - x) \quad (162)$$

we find (after a lengthy calculation)

$$\frac{1}{4\pi} \int d\Omega_k \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) \sum_m \tilde{j}_{P,mn}(-k) \tilde{j}_{P,mn}^\dagger(k) U_{mn}^R \quad (163)$$

$$= -\frac{\hbar^2 c^2}{3m^2} \int dx \langle \psi^\dagger(x) \nabla^2 V(x) \psi(x) \rangle_{nm} . \quad (163)$$

To arrive at this equality we have used the fact that $\psi(x)^2 = 0$ and $\psi^\dagger(x)^2 = 0$. Using the definition (4) of the background ionic potential,

$$\frac{1}{4\pi} \int d\Omega_k \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) \sum_m \tilde{j}_{P,mn}(-k) \tilde{j}_{P,mn}^\dagger(k) U_{mn}^R \quad (164)$$

$$= -\frac{4\pi \hbar^2 c^2}{m^2} \sum_{N} \langle \rho^N(R + d_N) \rangle_n ,$$

where $R + d_N$ is the location of the $N^{th}$ ion. Inserting this back into $\Delta E_n^R$, we find that our “generalized Lamb shift” is given by

$$\Delta E_n^R = -\frac{4\alpha \hbar^3}{3m^2 c} \sum_{N} \langle \rho^N(R + d_N) \rangle_n \times \int_0^\infty \frac{dk}{\hbar k/2mc + 1} \left( \frac{1}{E_n^R + \hbar c k} \right) . \quad (165)$$

Notice that this expression is finite, and so the integral can be evaluated. The “$\hbar k/2mc$” term in the denominator accounts for spatial variations in the electromagnetic field. We can compute this integral using techniques from complex analysis. Define $\beta_n^R = E_n^R/2mc^2$ and consider the change of variables $x = \hbar k/2mc$. Then the second line is

$$\int_0^\infty \frac{dk}{\hbar k/2mc + 1} \left( \frac{1}{E_n^R + \hbar c k} \right) = \frac{1}{\hbar c} \int_0^\infty dx \frac{1}{(x + 1)(x + \beta_n^R)} . \quad (166)$$

Consider the contour integral

$$\int_{\mathcal{C}} dz \frac{\log(z)}{(z + 1)(z + \beta_n^R)} . \quad (167)$$

where $\log(z)$ is the complex logarithm and $\mathcal{C}$ is the standard “keyhole” contour that avoids the branch cut (at
\(\theta = 0\) of \(\log(z)\) on the positive real axis. The contour integrals over the circular arcs vanish by application of the “ML-estimate” method for complex integration [42]. The other two integrals yield the identity

\[
\int dz \frac{\log(z)}{(z+1)(z+\beta_n)} = -2\pi i \int_0^\infty dx \frac{1}{(x+1)(x+\beta_n)}.
\]

Evaluating the contour integral on the left using the calculus of residues, we obtain

\[
\int_0^\infty dx \frac{1}{(x+1)(x+\beta_n^R)} = \left( 1 - \frac{E_R^R}{2mc^2} \right) \ln \left( \frac{2mc^2}{|E_R^R|} \right),
\]

and so our renormalized energy shift is

\[
\Delta E_n^R = -\frac{40\hbar^2}{3mc^2} \sum_N Q_N \frac{\langle \rho^2(R + \mathbf{d}_N) \rangle_n}{1 - \frac{E_R^R}{2mc^2}} \ln \left( \frac{2mc^2}{|E_R^R|} \right).
\]

Consider a single hydrogenic atom at \(R\). We can replace \(Q_N \langle \rho^2(R + \mathbf{d}_N) \rangle_n\) with \(-e^2 Z |\phi_{n00}(R)|^2\), where \(\phi_{n0m}(x)\) are the usual hydrogen wavefunctions. Moreover, using \(|\phi_{n00}(R)|^2 = Z^3/3n^3\pi a_0^3\) we have

\[
\Delta E_n^R = -\frac{40\hbar^2}{3mc^2} \sum_N \frac{mc^2}{1 - \frac{E_R^R}{2mc^2}} \ln \left( \frac{2mc^2}{|E_R^R|} \right).
\]

For the hydrogen atom and we take the average excitation energy for the \(2s\) level to be \(E_R^R = 16.64\ \text{Ryd}\) [18], from which we find the energy shift to be \(\Delta E_n^R \approx 1051\ \text{MHz}\), which is in good agreement with the experimental value of 1054 MHz [18]. To make the electric dipole approximation we replace the denominator with unity (since \(E_R^R \ll 2mc^2\)), through which we obtain Bethe’s result [29] (for \(Z = 1\))

\[
\Delta E_n^R = \frac{40\hbar^2}{3mc^2} \ln \left( \frac{2mc^2}{|E_R^R|} \right).
\]

## D. Multipole expansion

A primary advantage of using the multipole formalism over minimal coupling is the ability to perform localized multipole expansions. To do so, we use the Heisenberg equation

\[
\frac{\partial \rho(x, t)}{\partial t} = \frac{1}{\hbar} [\rho(x, t), H_F^R] = -\nabla \cdot \mathbf{j}_P(x, t),
\]

and so

\[
\mathbf{j}_P(x, t) = \frac{\partial \rho(x, t)}{\partial t} + c \nabla \times \mathbf{m}_P(x, t),
\]

\[
\mathbf{j}_D(x, t) = c \nabla \times \mathbf{m}_D(x, t).
\]

Then, using the identity for the matrix elements

\[
\langle \frac{\partial \rho(x, t)}{\partial t} \rangle_{nm} = (i\hbar)^{-1} E_{nm}^R p_{nm}(x, t),
\]

the matrix elements of the paramagnetic current density in the Schrödinger picture are

\[
\mathbf{j}_P(x, t) = (i\hbar)^{-1} E_{nm}^R p_{nm}(x, t) + c \nabla \times \mathbf{m}_P(x, t),
\]

and so the renormalized energy shift is

\[
\frac{\partial \rho(x, t)}{\partial t} = \frac{1}{\hbar} [\rho(x, t), H_F^R] = -\nabla \cdot \mathbf{j}_P(x, t),
\]

\[
\mathbf{j}_D(x, t) = c \nabla \times \mathbf{m}_D(x, t).
\]

This is our general expression for the renormalized energy shift in the multipole formalism. To study specific multipole contributions, we expand the polarization and magnetization field operators in a sum of multipole moments at \(R\) of the form

\[
p^i(x) = \left( \mu^i_R - q^i_R \delta^i + \ldots \right) \delta(x - R),
\]

\[
m^i_P(x) = \left( \nu^i_R + \ldots \right) \delta(x - R),
\]

where the electric dipole, quadrupole, and paramagnetic dipole moment operators are given (respectively) by

\[
\mu^i_R = \int dr \ (r^i - R^i) \rho(r),
\]

\[
q^{ij}_R = \frac{1}{2} \int dr \ (r^i - R^i)(r^j - R^j) \rho(r),
\]

\[
\nu^i_R = \frac{1}{2c} e^{jab} \int dr \ (r^a - R^a)^b \rho(r).
\]

To obtain these expressions, we take as the defining curve for the “relators” \(s^i(x; y, R)\) and \(a^i(x; y, R)\) a straight-
line path
\[ z(\lambda) = R + \lambda(y - R) \quad (184) \]
with unit parametrization \( \lambda \in [0, 1] \), in which case [36]
\[ s^i(x; y, R) = (y^i - R^i) \int_0^1 d\lambda \delta(x - R - \lambda(y - R)), \]
\[ \alpha^{ij}(x; y, R) = \varepsilon^{ijk}(y^k - R^k) \int_0^1 d\lambda \lambda \delta(x - R - \lambda(y - R)). \quad (185) \]

We then expand these expressions in a formal Taylor series with appropriate radius of convergence. Inserting the result back into the polarization and magnetization fields, the full multipole expansions are
\[ p^i(x) = \sum_{n=0}^{\infty} (-1)^n \mu_R^{i_1 \ldots i_n} \partial^{i_1} \ldots \partial^{i_n} \delta(x - R), \]
\[ m^i_p(x) = \sum_{n=0}^{\infty} (-1)^n \nu_R^{i_1 \ldots i_n} \partial^{i_1} \ldots \partial^{i_n} \delta(x - R), \quad (186) \]
where the \( n \)-th order electric and magnetic multipole moment operators are
\[
\mu_R^{i_1 \ldots i_n} = \frac{1}{n!} \int dr \left( r^{i_1} - R^{i_1} \right) \ldots \left( r^{i_n} - R^{i_n} \right) \left( r^i - R^i \right) \rho(r),
\]
\[
m_R^{i_1 \ldots i_n} = \frac{1}{c^n (n+1)!} \int dr \left( r^{i_1} - R^{i_1} \right) \ldots \left( r^{i_n} - R^{i_n} \right) \varepsilon^{iab} \left( r^a - R^a \right)^{i} \mu_p(r).
\]

In the renormalized energy shift (178) we need to evaluate Fourier integrals such as
\[
\int dk \frac{|k|}{E_{\hat{k}k} + \hbar c |k|} \left( \frac{1}{E_{\hat{k}k}^R + \hbar c |k|} \right) \sum_i e_i(k) \cdot \hat{p}_{mn}(k)|^2,
\]
with analogous ones for the magnetization and cross-term contributions. Write this integral in the form
\[
\int d\Omega_k \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) \int dx p^i_{mn}(x) \int dx' p^j_{mn}(x') \int_0^\infty dk \frac{e^{i\hat{k} \cdot (x - x')}}{(\hbar^2 k/2m + \hbar c)(E_{\hat{k}k}^R + \hbar c k)}.
\]

Inserting the multipole expansion for \( p^j_{mn}(x') \) in (186),
\[
\int dx' p^j_{mn}(x') \int_0^\infty dk \frac{e^{i\hat{k} \cdot (x - x')}}{(\hbar^2 k/2m + \hbar c)(E_{\hat{k}k}^R + \hbar c k)} = \sum_{b=0}^{\infty} (-i)^b \mu_{mn}^{j_1 \ldots j_b} \hat{k}^{j_1} \ldots \hat{k}^{j_b} \mathcal{J}(\hat{k}; x, R),
\]
where
\[
\mathcal{J}(\hat{k}; x, R) = \int_0^\infty dk \frac{k^b}{(\hbar^2 k/2m + \hbar c)(E_{\hat{k}k}^R + \hbar c k)} e^{i\xi(\hat{k}; x, R) k}
\]
and \( \xi(\hat{k}; x, R) = \hat{k} \cdot (x - R) \). Thus, the full integral (188) can be written
\[
\sum_{b=0}^{\infty} (-i)^b \mu_{mn}^{j_1 \ldots j_b} \int d\Omega_k \left( \delta^{ij} - \hat{k}^i \hat{k}^j \right) \hat{k}^{j_1} \ldots \hat{k}^{j_b} \left[ \lim_{r \to 0^+} \int_{V_r(R)} dx p^i_{mn}(x) \mathcal{J}(\hat{k}; x, R) \right],
\]
where \( V_r(R) \) denotes \( \mathbb{R}^3 \) minus a small closed ball of radius \( r \) centered on \( x = R \), which is necessary to avoid the singularity in \( \mathcal{J}(\hat{k}; x, R) \) at \( x = R \). By avoiding this singular point we can hold \( \hat{k} \cdot (x - R) \) fixed and thereby replace \( \xi(\hat{k}; x, R) \) in (191) with a constant value \( \xi \). Without loss of generality, we will assume \( \xi > 0 \). As \( |k| \to \infty \), the integral \( \mathcal{J}(\hat{k}; x, R) \) oscillates with divergent amplitude. We make this integral well-behaved using a “heat kernel regulator,” which in the continuum limit amounts to [1, 18]
\[
\int_0^\infty dk \frac{k^b e^{-|k|/\Lambda}}{(\hbar^2 k/2m + \hbar c)(E_{\hat{k}k}^R + \hbar c k)} e^{i\xi k},
\]
and the we take the limit \( \Lambda \to \infty \) (i.e., remove the regulator) after evaluating the integral. Consider the related
contour integral
\[ \oint_C dz \frac{z^b e^{-|z|^4/A} e^{i z}}{(z+1)(z+\beta R)} \log(z), \] (194)
where \( C \) is the “keyhole” contour as before. Simplifying this contour integral yields an identity analogous to (168), left side of which can be evaluated using the calculus of residues. After removing the regulator
\[ \mathcal{J}(k; x, R) = \frac{1}{\hbar c} \left( \frac{E_R^m}{\hbar c} \right)^b \left( \frac{1}{1 - E_R^{mn}/2mc^2} \right) \mathcal{J}(k; x, R), \] (195)
where
\[ \mathcal{J}(k; x, R) = \ln \left( \frac{2mc^2}{E_R^{mn}} \right) e^{i(E_R^{mn}/\hbar c)k \cdot (x-R)} - i \pi \left[ e^{i(E_R^{mn}/\hbar c)k \cdot (x-R)} - \left( \frac{2mc^2}{E_R^{mn}} \right)^b e^{i(2mc/\hbar)c k \cdot (x-R)} \right]. \] (196)
Thus we have
\[ \lim_{r \to 0^+} \int_{V_r(R)} dx p_{nm}^i(x) \mathcal{J}(k; x, R) = \frac{1}{\hbar c} \sum_{a=0}^{\infty} (i)^a \mu_{nm}^{a+} \left( \frac{E_R^m}{\hbar c} \right)^a \mathcal{J}(a+b), \] (197)
and so (188) is
\[ \frac{1}{\hbar c} \left( \frac{1}{1 - E_R^{mn}/2mc^2} \right) \sum_{a=0}^{\infty} \sum_{b=0}^{\infty} (i)^a (-i)^b \left[ \mu_{nm}^{a+} \mathcal{T}_{(1)}^{1 \ldots a ij1 \ldots js} \mu_{nm}^{j1 \ldots js} \left( \frac{E_R^m}{\hbar c} \right)^{a+b} \mathcal{J}(a+b), \right. \] (198)
where
\[ \mathcal{J}(a+b) = \ln \left( \frac{2mc^2}{E_R^{mn}} \right) - i \pi \left[ 1 - \left( \frac{2mc^2}{E_R^{mn}} \right)^{a+b} \right]. \] (199)
Evaluating the other two terms in \( \Delta E_n^R \) following a similar procedure, the total renormalized energy shift is
\[ \Delta E_n^R = \frac{1}{\pi} \sum_{m} \left( \frac{U_R^m}{1 - E_R^{mn}/2mc^2} \right) \left( \frac{E_R^m}{\hbar c} \right)^2 \sum_{a=0}^{\infty} \sum_{b=0}^{\infty} (-1)^b a + b \left( \frac{E_R^m}{\hbar c} \right)^{a+b} \left[ \mu_{nm}^{a+} \mathcal{T}_{(1)}^{1 \ldots a+ij1 \ldots js} \mu_{nm}^{j1 \ldots js} \right] \mathcal{J}(a+b) \]
\[ + \frac{1}{\pi} \sum_{m} \left( \frac{U_R^m}{1 - E_R^{mn}/2mc^2} \right) \left( \frac{E_R^m}{\hbar c} \right)^2 \sum_{a=0}^{\infty} \sum_{b=0}^{\infty} (-1)^b a + b \left( \frac{E_R^m}{\hbar c} \right)^{a+b} \left[ \mu_{nm}^{a+} \mathcal{T}_{(1)}^{1 \ldots a+ij1 \ldots js} \mu_{nm}^{j1 \ldots js} \right] \mathcal{J}(a+b+2) \]
\[ - \frac{2}{\pi} \sum_{m} \left( \frac{U_R^m}{1 - E_R^{mn}/2mc^2} \right) \left( \frac{E_R^m}{\hbar c} \right)^2 \sum_{a=0}^{\infty} \sum_{b=0}^{\infty} (-1)^b \left( \frac{E_R^m}{\hbar c} \right)^{a+b} \left[ \mu_{nm}^{a+} \mathcal{T}_{(2)}^{1 \ldots a+ij1 \ldots js} \mu_{nm}^{j1 \ldots js} \right] \mathcal{J}(a+b+1). \] (200)
Above have defined the isotropic Cartesian tensors
\[ \mathcal{I}_{(1)}^{1 \ldots a+ij1 \ldots js} = \frac{d \Omega_R}{4 \pi} \left( \delta^i k^j - k^i k^j \right) \hat{k}^i \ldots \hat{k}^a \hat{k}^j \ldots \hat{k}^b, \]
\[ \mathcal{I}_{(2)}^{1 \ldots a+ij1 \ldots js} = \frac{d \Omega_R}{4 \pi} \left( e^{ij p \hat{k}^p} \right) \hat{k}^i \ldots \hat{k}^a \hat{k}^j \ldots \hat{k}^b. \] (201)
The tensor \( \mathcal{I}_{(1)} \) vanishes unless \( a+b = 2n \) for some \( n \in \mathbb{N} \), while the tensor \( \mathcal{I}_{(2)} \) vanishes unless \( a+b = 2n-1 \) for some \( n \in \mathbb{N} \). But if \( a+b = 2n-1 \), then in the cross-term contribution
\[ \text{Re} \left[ \mu_{nm}^{a+} \mathcal{T}_{(2)}^{1 \ldots a+ij1 \ldots js} \mu_{nm}^{j1 \ldots js} \right] = 0, \] (202)
since the matrix elements of self-adjoint operators are real, and so we only obtain contributions to \( \Delta E_n^R \) coming from the first two lines of (201). The condition \( a+b = 2n \) for \( n \in \mathbb{N} \) precludes a number of electric and magnetic terms, such as the product of the electric dipole and quadrupole moments, while allowing others like the product of the electric dipole and octopole moments. The latter behave as interference terms, since they come with an overall minus sign, while those involving the square of a given electric or magnetic multipole moment are positive. The first several contributions to \( \Delta E_n^R \) are given below...
\[
\Delta E_n^R = \frac{2}{3\pi} \sum_m \left( \frac{U_{mn}^R}{1 - E_{mn}^R/2mc^2} \right) \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \left[ |\mu_{nm}|^2 \mathcal{J}(0) + |\nu_{nm}|^2 \mathcal{J}(2) + \frac{1}{10} \left( E_{mn}^R/\hbar c \right)^2 (3q_{nm}^{ab}q_{mn}^{ab} - q_{nm}^{aa}q_{mn}^{bb}) \mathcal{J}(2) \right] - \frac{1}{5} \left( E_{mn}^R/\hbar c \right)^2 \left( \mu_{nm}^{aa} + \nu_{nm}^{ab} \right) \mathcal{J}(2) + \ldots \right] ,
\]

(203)

where \( o_{ijk}^R \) is the electric octopole moment operator and the ellipses denote the higher-order multipole contributions. Consider only the electric dipole term

\[
\Delta E_n^R = \frac{2}{3\pi} \sum_m \left( \frac{U_{mn}^R}{1 - E_{mn}^R/2mc^2} \right) \left( \frac{E_{mn}^R}{\hbar c} \right)^2 |\mu_{nm}|^2 \mathcal{J}(0) .
\]

Using the identity

\[
(E_{mn}^R)^2 |\mu_{nm}|^2 = - |E_{mn}^R \mu_{nm}|^2 = \frac{\hbar^2 e^2}{m^2} |\mathbf{P}_{nm}|^2 \tag{205}
\]

this can be written

\[
\Delta E_n^R = \frac{2\alpha}{3\pi} \frac{\hbar}{m^2 c} \sum_m \frac{U_{mn}^R}{1 - E_{mn}^R/2mc^2} |\mathbf{P}_{nm}|^2 \mathcal{J}(0) . \tag{206}
\]

As before, we replace \( E_{mn}^R \) with an average value \( \bar{E}_n^R \), and so the electric dipole contribution is

\[
\Delta E_n^R = \frac{2\alpha}{3\pi} \frac{\hbar}{m^2 c} \sum_m \frac{U_{mn}^R |\mathbf{P}_{nm}|^2}{1 - E_n^R/2mc^2} \ln \left( \frac{2mc^2}{|E_n^R|} \right) . \tag{207}
\]

The electric dipole approximation is made in the limit \( E_n^R/2mc^2 \ll 1 \), so that we can replace the denominator with unity

\[
\Delta E_n^R = \frac{2\alpha}{3\pi} \frac{\hbar}{m^2 c} \sum_m U_{mn}^R |\mathbf{P}_{nm}|^2 \ln \left( \frac{2mc^2}{|E_n^R|} \right) . \tag{208}
\]

In a first-quantized treatment, this is precisely Bethe’s result for the Lamb shift in the electric dipole approximation [29]. The higher-order multipole moments yield additional contributions to \( \Delta E_n^R \), weighted by successively higher reciprocal powers of \( c \).

\[\text{VI. CONCLUSION}\]

We have introduced a field-theoretic reformulation of multipolar electrodynamics to model interactions between quantized electromagnetic fields and localized charge-current distributions. The electronic degrees of freedom were encoded in microscopic polarization and magnetization field operators — defined through second-quantized scalar field operators — whose moments we identified with the multipole moments of the charge-current distribution. These field operators couple to

the quantized electromagnetic field in the multipolar Hamiltonian, which was obtained from the minimal coupling Hamiltonian through a unitary transformation that corresponds in classical or semi-classical theory to the canonical “PZW transformation.” Our reformulation generalizes existing work [7, 20] insofar that a field-theoretic model allows us to treat very general charge-current distributions — provided they are sufficiently localized — including those of large assemblies of atoms and molecules, and those in which the number of charge carriers may change over time.

The microscopic polarization and magnetization fields were defined through line integrals of the charge and current densities along a fixed curve \( C(x, \mathbf{R}) \), and the multipolar Hamiltonian was written in terms of these and the Maxwell fields \((\mathbf{d}, \mathbf{b})\). It is not clear, a priori, what happened to the gauge freedom that was present in minimal coupling. The minimal coupling Lagrangian is invariant under the usual U(1)—gauge transformations

\[
\mathbf{a}(x, t) \rightarrow \mathbf{a}(x, t) - \nabla g(x, t), \nonumber
\]

\[
\phi(x, t) \rightarrow \phi(x, t) + \frac{1}{c} \frac{\partial g(x, t)}{\partial t}, \nonumber
\]

\[
\psi(x, t) \rightarrow e^{-\frac{i}{\hbar c} g(x, t)} \psi(x, t), \tag{209}
\]

where \( g(x, t) \) is a real-valued function that parametrizes the group of gauge transformations. To implement a change of variables in the minimal coupling Lagrangian that will be equivalent to a suitable canonical transformation of the associated Hamiltonian, we add to it a total time derivative of the form

\[
L \rightarrow L - \frac{1}{c} \frac{d}{dt} \int dx \lambda(x; t) \rho(x, t) . \tag{210}
\]

In particular, we obtain the multipolar Lagrangian by setting

\[
\lambda(x; t) = \int dy s^i(y; x, \mathbf{R}) a^i(y, t) . \tag{211}
\]

After implementing this transformation, one finds that the multipolar Lagrangian \( L \) depends on the electric and magnetic fields \((\mathbf{E}, \mathbf{B})\) and the polarization and magnetization fields \((\mathbf{P}, \mathbf{M})\), and not on the gauge potentials. (We have restored the tilde accents here to distinguish the multipolar fields from the minimal coupling ones, as we did in Section II.) Therefore, we do not have the same
manifest gauge freedom that we did in the minimal coupling Lagrangian $L$. However, there is still a kind of “gauge freedom” in the choice of curve used in $\lambda(x; t)$ [36], since changing from one curve $C(x, R)$ to another $C'(x, R)$ will lead to the “gauge transformations”

$$\tilde{p}(x, t) \to \tilde{p}(x, t) + \nabla \times \mathbf{v}(x, t),$$

$$\tilde{m}(x, t) \to \tilde{m}(x, t) - \frac{1}{c} \frac{\partial \mathbf{v}(x, t)}{\partial t},$$

$$\tilde{\psi}(x, t) \to \tilde{\psi}(x, t),$$

where $\mathbf{v}(x, t)$ is a vector field whose exact form depends on the initial and final curves, and

$$W[\Gamma_R(x); t] = \exp \left( -\frac{ie}{\hbar c} \oint_{\Gamma_R(x)} \mathbf{a}(z, t) \cdot dz \right)$$

is the Wilson loop operator of Abelian gauge theory with the connection 1-form $\mathbf{a}(z, t) \cdot dz$ [35]; the loop $\Gamma_R(x)$ is obtained by concatenating the curves $C'(x, R)$ and $C(R, x)$. In this sense, the gauge freedom in $L$ is “replaced” in $\tilde{L}$ by a freedom in the choice of curve through which $\lambda(x; t)$ and thus the polarization and magnetization fields are defined. In the Hamiltonian framework, just as the minimal coupling Hamiltonian is invariant under the transformations (209), the multipolar Hamiltonian is independent of the choice of curve in $\lambda(x; t)$; this can be shown by relating the canonical variables (38) with one choice of curve to those of another, and substituting this relationship into the multipolar Hamiltonian.

Following the reformulation of multipolar electrodynamics in Section II, our focus thereafter concerned the vacuum structure of the theory, as outlined in Section III. Using Rayleigh-Schrödinger perturbation theory we computed the regularized shift $\Delta E_n^R$ of the electronic energy levels in the electromagnetic vacuum state, described by the regularized interaction term $H_{\text{int}}^\Lambda$. This energy shift, computed in Section IV, depended explicitly on the ultraviolet cutoff $\Lambda$, an artifact of our choice of “hard cutoff” regulator. To remove this $\Lambda$-dependence, we renormalized the regularized Hamiltonian $H^\Lambda$ at leading-order by addition of appropriate counterterms. In Section V we used the renormalized Hamiltonian that followed to compute the finite and observable correction $\Delta E_n^R$ to the (renormalized) electronic energy levels $E_n^R$.

We obtained a closed form expression for the renormalized energy shift $\Delta E_n^R$ when written in terms of the matrix elements of the current density operator. For hydrogenic atoms, our expression reproduces existing results, including Bethe’s calculation [29] of the Lamb shift in the electric dipole approximation, and extensions thereof [31, 32] to include spatial variations in the electromagnetic field. However, our expression is valid for more general assemblies of atoms and molecules (possibly with net charge), consisting of a sum of contributions centered on each ion core in the assembly, each of which is weighted by the expectation value of the electronic charge density evaluated at that location. To compute these expectation values, one could expand the electron field operators in terms of an appropriate set of single-particle wavefunctions that transform in an irreducible representation of the symmetry group of the system [43]. Because of how complicated this symmetry group may be, we would no longer expect the contributions to $\Delta E_n^R$ to be restricted to wavefunctions with full SO(3)-symmetry like the $s$-type hydrogen orbitals; instead, we expect that these expectation values will depend in a complicated way on the electronic structure of the system. Numerically evaluating these expectation values and the expression for $\Delta E_n^R$ would be an interesting direction for future work.

At the end of Section V we rewrote the renormalized energy shift in terms of the polarization and magnetization fields, permitting us to expand $\Delta E_n^R$ in a sum of contributions coming from products of specific electric and magnetic multipole moments at $\mathbf{R}$. We derived the general such expression, and then gave explicit form of the contributions coming from the first several multipole moments, namely the electric dipole, quadrupole, and magnetic dipole moments, together with the lowest-order interference term from the product of the electric dipole and octopole moments. Existing treatments of the vacuum energy shift have focused on hydrogenic atoms, and therefore include only the electric dipole term. As far as we can tell, ours is the first explicit expression for the renormalized energy shift that includes the full sum of multipole contributions.

This work lays the foundation for a broader research project wherein we intend on applying multipolar quantum electrodynamics, as formulated in Section II, to more complex solid-state dielectrics such as molecular crystals and crystalline solids. In this paper our description of the material degrees of freedom was sufficiently general to be applied to an isolated unit cell in a periodic lattice, in which case $\mathbf{R}$ would label a particular unit cell and the set $\{d_{\mathbf{R}}\}_N$ consists of the locations of the ions within that cell. Recent work indicates that a description of crystalline solids using multipolar quantum electrodynamics should be possible [37, 44]; there the authors introduced a semi-classical formalism based on microscopic polarization and magnetization fields that could be associated with individual lattice sites, together with itinerant contributions due to the presence of free charge and current. In future work, we intend on extending their model into the fully quantum regime following a formulation of multipolar QED analogous to that which we have presented above, thereby permitting us to study quantum optical effects in crystals from this microscopic perspective.

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Appendix A: Electrostatic interactions in the multipolar Hamiltonian

We demonstrate the equivalence between the electronic Hamiltonian $H_F^\Lambda$ in multipolar QED

$$H_F = \frac{\hbar^2}{2m} \int dx \left( \nabla \psi(x) \cdot \nabla \psi(x) + 2\pi \int dx \|p_L(x)\|^2 - \int dx \mathbf{d}_L(x) \cdot \left( \mathbf{p}(x) - \frac{1}{8\pi} \mathbf{d}_L(x) \right) \right), \quad (A1)$$

and the electronic Hamiltonian

$$H_F^\Lambda = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + \frac{1}{2} \int dx dx' \psi^\dagger(x) \psi^\dagger(x') \frac{e^2}{|x-x'|} \psi(x') \psi(x) + \int dx \psi^\dagger(x) \nabla \psi(x) + E_F^\Lambda, \quad (A2)$$

where $E_F^\Lambda$ is a $\Lambda$-dependent term that we compute here. To begin, we use the following formula

$$\delta^{ij}_L(x - x') = \frac{1}{(2\pi)^3} k^i k^j e^{i k \cdot (x - x')} = -\frac{1}{2\pi^2} \int_0^\infty dk \partial^i \partial^j F(x, x'; k), \quad (A3)$$

where $F(x, x'; k) = \text{sinc}(k|x - x'|)$. After regularizing the Fourier integrals in the representation (A3) of the longitudinal delta functions, the second term on the first line of (A1) is

$$2\pi \int dx \|p_L(x)\|^2 = -\frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \left( \rho(x') - \rho_F(x') \right) F(x, x'; k) \left( \rho(x) - \rho_F(x) \right),$$

$$= -\frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \rho(x') F(x, x'; k) \rho(x) + \frac{2Q_R}{\pi} \int_0^\Lambda \int dx \rho(x) F(x, R; k) + \frac{Q^2_R}{\pi} \int_0^\Lambda \int dx.$$

Meanwhile, using the Gauss law $\nabla \cdot \mathbf{d}_L(x) = 4\pi \rho_F(x)$, a straightforward calculation leads to

$$-\int dx \mathbf{d}_L(x) \cdot \left( \mathbf{p}(x) - \frac{1}{8\pi} \mathbf{d}_L(x) \right) = -\frac{Q^2_R}{\pi} \int_0^\Lambda \int dx \rho(x) F(x, R; k), \quad (A5)$$

and therefore

$$H_F = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + \frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \rho(x') F(x, x'; k) \rho(x). \quad (A6)$$

Next, using $\rho(x) = \rho^e(x) + \rho^{en}(x)$,

$$\frac{1}{\pi} \int_0^\Lambda \int dx dx' \rho(x') F(x, x'; k) \rho(x) = \frac{1}{\pi} \int_0^\Lambda \int dx dx' \rho^e(x') F(x, x'; k) \rho^e(x) + \frac{2}{\pi} \sum_N Q_N \int_0^\Lambda \int dx F(x, R + d_N; k) \rho^e(x) + \sum_{N,M} \frac{Q_N Q_M}{\pi} \int_0^\Lambda \int dx F(d_N, d_M; k), \quad (A7)$$

and so, isolating the divergences in $E_F^\Lambda$, we have

$$\frac{1}{\pi} \int_0^\Lambda \int dx dx' \rho(x') F(x, x'; k) \rho(x) = \frac{1}{\pi} \int_0^\Lambda \int dx dx' \psi^\dagger(x) \psi^\dagger(x') F(x, x'; k) \psi(x') \psi(x)$$

$$+ \frac{2}{\pi} \sum_N Q_N \int_0^\Lambda \int dx F(x, R + d_N; k) \rho^e(x) + \sum_{N \neq M} \frac{Q_N Q_M}{\pi} \int_0^\Lambda \int dx F(d_N, d_M; k) + E_F^\Lambda, \quad (A8)$$

where in the integral in the second term on the right side of the above equality it should be understood that a small sphere centered at $x = R + d_N$ is excluded, and the $\Lambda$-dependent term is given by

$$E_F^\Lambda = \frac{1}{\pi} \left[ e^2 N_e + 2e N_e \sum_N Q_N + \sum_N Q^2_N \right] \Lambda, \quad (A9)$$
and the second term on the right-hand-side of this expression comes from the integration over that small sphere. Since we have isolated the $\Lambda$-dependent terms in $E_F^\Lambda$, we can take the limit $\Lambda \to \infty$ in the remaining terms and use the identity (valid for $x \neq x'$)

$$\int_0^\infty dk F(x, x'; k) = \frac{\pi}{2} \frac{1}{|x - x'|}.$$  \hspace{1cm} (A10)

Dropping the ion-ion interaction as in Section II, we obtain the desired result

$$H_D^\Lambda = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + \frac{e^2}{2} \int dx dx' \psi^\dagger(x) \psi^\dagger(x') \frac{e^2}{|x - x'|} \psi(x) \psi(x') + \int dx \psi^\dagger(x)V(x)\psi(x) + E_F^\Lambda. \hspace{1cm} (A11)$$

\section*{Appendix B: First-order diamagnetic contribution}

The diamagnetic term in the Hamiltonian (27) is

$$H_D^{(1)} = \frac{1}{2} \int dx m_D(x) \cdot b(x). \hspace{1cm} (B1)$$

As noted in the main text, we can rewrite $H_D^{(1)}$ explicitly in terms of the magnetic field

$$H_D^{(1)} = + \frac{1}{2} \int dx dx' b^i(x) O^{ij}(x, x') b^j(x'), \hspace{1cm} (B2)$$

where

$$O^{ij}(x, x') = \int dy \psi^\dagger(y) o^{ij}(x, x'; y) \psi(y) \hspace{1cm} (B3)$$

is the second-quantized diamagnetization field [36]

$$o^{ij}(x, x'; y) = \frac{e^2}{2mc^2} \alpha^{ik}(x; y, R) \alpha^{jk}(x'; y, R). \hspace{1cm} (B4)$$

Use (12) to write the above expression in terms of the vector potential

$$H_D^{(1)} = \frac{1}{2} \int dx dx' a^a(x) \tilde{O}^{ab}(x, x') a^b(x'), \hspace{1cm} (B5)$$

where $\tilde{O}^{ab}(x, x')$ is the second-quantized form of

$$\tilde{o}^{ab}(x, x'; y) = \varepsilon^{api} \varepsilon^{bjq} \partial \partial x^p \partial x^q o^{ij}(x, x'; y). \hspace{1cm} (B6)$$

With the identity [37]

$$\varepsilon^{abc} \partial \partial x^a \alpha^{cd}(x; y, R) = - \partial \partial y^d \delta^{cd}(x; y, R) + \delta^{cd} \delta(x - y) \hspace{1cm} (B7)$$

the field $\tilde{o}^{ab}(x, x'; y)$ is

$$\tilde{o}^{ab}(x, x'; y) = \frac{e^2}{2mc^2} \left( \delta^{ab} \delta(x - y) \delta(x' - y) - \delta^{ca} \delta(x - y) \partial \partial y^b s^i(x'; y, R) - \delta^{cb} \delta(x' - y) \partial \partial y^a s^i(x; y, R) + \left[ \partial \partial y^a s^i(x; y, R) \right] \left[ \partial \partial y^b s^i(x'; y, R) \right] \right). \hspace{1cm} (B8)$$

Substituting this expression back into (B5), the diamagnetic term is

$$H_D^{(1)} = \frac{e}{2mc^2} \int dx \rho^c(x) \|a(x)\|^2 - \frac{\hbar}{mc} \int dx \rho^c(x) a(x) \cdot \nabla \Phi(x, R) + \frac{\hbar^2}{2mc} \int dx \rho^c(x) \|\nabla \Phi(x, R)\|^2. \hspace{1cm} (B9)$$

Define

$$\Delta E_D^\Lambda \equiv \langle \Psi_n | H_D^{(1)} | \Psi_n \rangle, \hspace{1cm} (B10)$$

with $|\Psi_n\rangle$ given by (94). The regularized mode expansion for the vector potential $a(x)$ that leads to the mode expansion (73) for $b(x)$ is

$$a(x, t) = \sum_I \sum_{|k| \leq \Lambda} \left( \frac{2\pi\hbar c}{V|k|} \right)^{1/2} e_{1k} a_{1k} e^{i(k \cdot x - \omega_k t)} + H.c. \hspace{1cm} (B11)$$
A straightforward calculation of the vacuum expectation value of $||a(x)||^2$ leads to

$$e^{mc^2} \int \! dx \, \rho_{nn}^e(x) \langle ||a(x)||^2 \rangle_{\text{vac}} = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_c}{m} \right) \Lambda^2.$$  
(B12)

Focusing on the latter two terms of (B9), the expectation value of the first is

$$- \frac{\hbar}{mc} \int \! dx \, \rho_{nn}^e(x) \langle a(x) \cdot \nabla \Phi(x, R) \rangle_{\text{vac}},$$  
(B13)

while the expectation value of the second is

$$+ \frac{\hbar^2}{2mc} \int \! dx \, \rho_{nn}^e(x) \langle ||\nabla \Phi(x, R)||^2 \rangle_{\text{vac}}.$$  
(B14)

With the regularized mode expansion for $a(x)$, we have

$$\langle a^a(x)a^b(x') \rangle_{\text{vac}} = \sum_{|k| \leq \Lambda} \left( \frac{2\pi \hbar c}{|k|} \right) \left( \delta_{ab} - \frac{k^a k^b}{|k|^2} \right) e^{ik \cdot (x - x')}.$$  
(B15)

or in the continuum limit

$$\langle a^a(x)a^b(x') \rangle_{\text{vac}} = \frac{\hbar c}{4\pi^2} \int \! dk |k|^{-1} \left( \delta_{ab} - \frac{k^a k^b}{|k|^2} \right) e^{ik \cdot (x - x')}.$$  
(B16)

Thus, the vacuum expectation value in (B13) is

$$- \frac{\hbar c}{4\pi^2 mc} \int \! dk |k|^{-1} \left( \delta_{ij} - \frac{k^i k^j}{|k|^2} \right) \iiint \! dx dx' e^{i k \cdot (x' - x)} \rho_{nn}^e(x) \left[ \frac{\partial}{\partial x^i} s^j(x'; x, R) \right],$$  
(B17)

while the vacuum expectation value in (B14) is

$$\frac{\hbar c}{8\pi^2 mc} \int \! dk |k|^{-1} \left( \delta_{ij} - \frac{k^i k^j}{|k|^2} \right) \iiint \! dx dx' e^{i k \cdot (x' - x)} \int \! dy \, \rho_{nn}^e(y) \left[ \frac{\partial}{\partial y^i} s^j(x; y, R) \right],$$  
(B18)

so that the expectation values of the latter two terms of (B9) are

$$- \frac{\hbar}{mc} \int \! dx \, \rho_{nn}^e(x) \langle a(x) \cdot \nabla \Phi(x, R) \rangle_{\text{vac}} + \frac{\hbar^2}{2mc} \int \! dx \, \rho_{nn}^e(x) \langle ||\nabla \Phi(x, R)||^2 \rangle_{\text{vac}} = - \frac{\hbar c}{4\pi^2 mc} \int \! dk |k|^{-1} \left( \delta_{ij} - \frac{k^i k^j}{|k|^2} \right) \times \iiint \! dx dx' e^{i k \cdot (x' - x)} \rho_{nn}^e(x) \left[ \frac{\partial}{\partial x^i} s^j(x'; x, R) \right] - \frac{1}{2} \int \! dy \, \rho_{nn}^e(y) \left[ \frac{\partial}{\partial y^i} s^j(x; y, R) \right] \left[ \frac{\partial}{\partial y^j} s^i(x' ; y, R) \right].$$  
(B19)

As shown in Appendix C, these terms exactly cancel with terms in the second-order correction $\Delta E^\Lambda_{n,(2)}$. In total, the diamagnetic contribution to the first-order correction is

$$\Delta E^\Lambda_D = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_c}{m} \right) \Lambda^2 - \frac{\hbar}{mc} \int \! dx \, \rho_{nn}^e(x) \langle a(x) \cdot \nabla \Phi(x, R) \rangle_{\text{vac}} + \frac{\hbar^2}{2mc} \int \! dx \, \rho_{nn}^e(x) \langle ||\nabla \Phi(x, R)||^2 \rangle_{\text{vac}}.$$  
(B20)

Appendix C: Second-order simplifications

Begin with the full expression for the second-order correction

$$\Delta E^\Lambda_{n,(2)} = - \frac{\hbar c}{4\pi^2} \sum_m \int \! dk |k| \iint \! dx dx' \frac{e^{i k \cdot (x' - x)}}{E_{mn} + \hbar c |k|} \left\{ \left( \delta_{ij} - \frac{k^i k^j}{|k|^2} \right) \left( p_{i mn}^j(x') p_{mn}^j(x) + m^i_{p mn}(x') m^j_{p mn}(x) \right) \right. \left. - (\epsilon^{ijp} k^p |k|) \left( p_{i mn}^j(x') m^j_{p mn}(x) + m^i_{p mn}(x') p_{mn}^j(x) \right) \right\}.$$  
(C1)

We can simplify the second line using the identity [19]

$$\frac{1}{4\pi} \int \! d\Omega_k \, \epsilon^{ijp} \frac{k^p}{|k|} e^{\pm i k \cdot (x' - x)} = \pm \frac{i}{|k|} \epsilon^{ijp} \partial^p F(x, x'; k),$$  
(C2)
where $F(x, x'; k)$ is given by (109). Together with the identity (108), we have

$$
\Delta E_{n,(2)}^\Lambda = -\frac{1}{\pi} \sum_m \int_0^\Lambda dk \frac{\hbar k c}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) + m_{P, nm}^i(x') m_{P, mn}^j(x) \right) 
$$

$$
\quad - \frac{i}{\pi} \sum_m \int_0^\Lambda dk \frac{\hbar k^2}{E_m + \hbar k c} \int dxdx' \left[ \epsilon^{ijp} \partial^p F(x, x'; k) \right] \left( p_{nm}^i(x') m_{P, mn}^j(x) + m_{P, nm}^i(x') p_{mn}^j(x) \right). \tag{C3}
$$

To simplify $\Delta E_{n,(2)}^\Lambda$, we break up its summands and process them in turn. Denote the first term by $\Delta E_{pp}^\Lambda$, the second term by $\Delta E_{mn}^\Lambda$, and the second line collectively by $\Delta E_{mm}^\Lambda$.

Consider first the contribution $\Delta E_{pp}^\Lambda$. Using twice the algebraic identity

$$
\frac{1}{E_m + \hbar c|k|} = \frac{1}{\hbar c|k|} - \frac{1}{E_m + \hbar c|k|}, \tag{C4}
$$

we have

$$
\Delta E_{pp}^\Lambda = -\frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad + \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \frac{k}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad - \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \frac{k^2}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right). \tag{C5}
$$

Then, using

$$
p_{nm}^i(x) = (ih)^{-1} E_m p_{nm}^i(x), \tag{C6}
$$

the last line can be rewritten

$$
\Delta E_{pp}^\Lambda = -\frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad + \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \frac{k}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( E_m p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad - \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \frac{k^2}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right). \tag{C7}
$$

To simplify the last line, we use (177) to rewrite the time derivatives of the polarization fields in terms of the magnetization and paramagnetic current density. After a lengthy calculation, the full contribution $\Delta E_{pp}^\Lambda$ is

$$
\Delta E_{pp}^\Lambda = -\frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad + \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \frac{k}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( E_m p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad - \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \frac{k^2}{E_m + \hbar k c} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] \left( p_{nm}^i(x') p_{mn}^j(x) \right) 
$$

$$
\quad + \frac{h}{\pi} \sum_m \int_0^\Lambda dk \frac{k}{E_m + \hbar k c} \int dxdx' \left[ \epsilon^{ijp} \partial^p F(x, x'; k) \right] \left( j_{P, nm}^i(x') m_{P, mn}^j(x) + m_{P, nm}^i(x') j_{P, mn}^j(x) \right). \tag{C8}
$$
We can combine the third line above with the contribution $\Delta E_{mm}^\Lambda$, which is

$$\Delta E_{mm}^\Lambda = -\frac{1}{\pi} \sum_m \int_0^\Lambda \frac{dk}{E_{mm} + \hbar k} \int dxdx' \left[ (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k) \right] m_{i,mm}^x(x') m_{j,mm}^x(x).$$  \hspace{1cm} (C9)

Consider last the contribution $\Delta E_{pm}^\Lambda$. Using (C6) along with the algebraic identity (C4), we have

$$\Delta E_{pm}^\Lambda = -\frac{i}{\pi} \int_0^\Lambda dkk \int dxdx' \left[ \varepsilon^{ipj} \partial^p F(x, x'; k) \right] \left( p^j(x') m_{j,p}^x(x) - m_{i,p}^x(x') p^i(x) \right).$$

Then, through a trivial relabelling

$$\Delta E_{pm}^\Lambda = -\frac{i}{\pi} \int_0^\Lambda dkk \int dxdx' \left[ \varepsilon^{ipj} \partial^p F(x, x'; k) \right] \left( p^i(x') m_{j,p}^x(x) - m_{i,p}^x(x') p^j(x) \right).$$

Using the identities

$$(\varepsilon^{ipj} \partial^p F(x, x'; k) \left\{ m_{j,p}^x(x'), p^i(x) \right\}^\Lambda_n = k^2 \varepsilon^{ipj} \partial^p F(x, x'; k) \left\{ m_{j,p}^x(x'), p^i(x) \right\}^\Lambda_n$$}

and

$$\sum_m E_{mm} p_{mm}^x(x') p_{mm}^x(x) = \frac{i\hbar}{2} \left\{ j_{j,p}^x(x'), p^i(x) \right\}^\Lambda_n - c \varepsilon^{ipj} \partial^p \left\{ m_{j,p}^x(x'), p^i(x) \right\}^\Lambda_n,$$
we can collect the second and fourth lines together. Restoring the angular parts of the Fourier integrals, we the
second-order correction is
\[
\Delta E_{n,(2)}^{\Lambda} = \frac{i}{8\pi^2 c} \sum_{m} \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \int dx \int dx' \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) e^{i k \cdot (x' - x)} \left\langle \left[ j^i_{P}(x'), p^j_{i}(x) \right] \right\rangle_n + c e^{ik} \partial^p m^l_{P}(x') \partial^p m^l_{P}(x) \left\langle \left[ j^i_{P}(x'), p^j_{i}(x) \right] \right\rangle_n
\]
\[
- \frac{\hbar}{4\pi^2 c} \sum_{m} \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \int dx \int dx' \left( j^i_{P,mn}(x') j^i_{P,mn}(x) \right) \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) e^{i k \cdot (x' - x)}
\]
\[
- \frac{1}{\pi} \sum_{m} \int_{0}^{\Lambda} dk \int dx \int dx' \left[ \left( -\delta^{ij} \partial^2 + \partial^i \partial^j \right) F(x, x'; k) p^i_{n,m}(x') p^j_{n,m}(x) \right].
\]

(C16)

Working out the commutators in the first line, we find
\[
\frac{1}{2} \left\langle \left[ j^i_{P}(x'), p^j_{i}(x) \right] \right\rangle_n + c e^{ik} \partial^p m^l_{P}(x') \partial^p m^l_{P}(x) \left\langle \left[ j^i_{P}(x'), p^j_{i}(x) \right] \right\rangle_n = \frac{\hbar c}{m i} \left( \frac{\partial}{\partial x^i} s^i(x; x, R) \right) \rho_{n,m}(x)
\]
\[
- \frac{1}{2} \int dy \rho_{n,m}(y) \left[ \frac{\partial}{\partial y^k} s^i(x; y, R) \right] \left[ \frac{\partial}{\partial y^k} s^j(x; y, R) \right],
\]

(C17)

and so, comparing to (B19), the second-order correction is
\[
\Delta E_{n,(2)}^{\Lambda} = - \frac{\hbar}{4\pi^2 c} \sum_{m} \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \int dx \int dx' \left( j^i_{P,mn}(x') j^i_{P,mn}(x) \right) \left( \delta^{ij} - \frac{k^i k^j}{|k|^2} \right) e^{i k \cdot (x' - x)}
\]
\[
+ \frac{\hbar}{mc} \int dx \rho_{n,m}(x) \left( a(x) \cdot \nabla \Phi(x, R) \right)_{\text{vac}} - \frac{\hbar^2}{2mc} \int dx \rho_{n,m}(x) \left( \| \nabla \Phi(x, R) \|^2 \right)_{\text{vac}}
\]
\[
- \frac{1}{\pi} \sum_{m} \int_{0}^{\Lambda} dk \int dx \int dx' \left[ \left( -\delta^{ij} \partial^2 + \partial^i \partial^j \right) F(x, x'; k) p^i_{n,m}(x') p^j_{n,m}(x) \right].
\]

(C18)

### Appendix D: Energy shift in minimal coupling

For comparison with the regularized energy shift (119) computed with the multipolar Hamiltonian, we work out the same energy shift in minimal coupling. Since these formulations of non-relativistic quantum electrodynamics are unitarily equivalent, we should obtain the same result. The minimal coupling Hamiltonian is given in the Schrödinger picture by

\[
H = H_B \otimes I_F + \int dx \psi^\dagger(x) H(x) \psi(x),
\]

(D1)

where the free Maxwell term \( H_B \) is

\[
H_B = \frac{1}{8\pi} \int dx \left( e_T(x) \cdot e_T(x) + b(x) \cdot b(x) \right),
\]

(D2)

while

\[
H(x) = \frac{1}{2m} \left( \frac{\hbar}{i} \nabla - \frac{e}{c} a(x) \right)^2 + V(x) + e \phi(x).
\]

(D3)

Split up the Hamiltonian into free and interaction terms

\[
H = H_0 + H_{\text{int}},
\]

(D4)

where the free term is

\[
H_0 = H_B \otimes I_F + I_B \otimes H_F
\]

with the electronic Hamiltonian

\[
H_F = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x)
\]
\[
+ \frac{1}{2} \int dx \rho^e(x) \phi(x) + \int dx \psi^\dagger(x) V(x) \psi(x),
\]

(D6)

and the interaction term is

\[
H_{\text{int}} = - \frac{1}{c} \int dx j(x) \cdot a(x).
\]

(D7)

Here \( j(x) \) is the total current density (9). As in Section IV, we split up the interaction term

\[
H_{\text{int}} = H_{\text{int}(1)} + H_{\text{int}(2)},
\]

(D8)

where

\[
H_{\text{int}(1)} = \frac{e}{2mc^2} \int dx \psi^\dagger(x) \| a(x) \|^2 \psi(x)
\]

(D9)

is already \( O(\alpha) \) and is treated at the first order, while

\[
H_{\text{int}(2)} = - \frac{1}{c} \int dx j_P(x) \cdot a(x)
\]

(D10)
is $O(\sqrt{\alpha})$ and is treated at the second order. The first-order correction is

$$\Delta E_{n,(1)}^\Lambda = \frac{e^2}{2mc^2} \int dx \langle \Psi_n | \hat{\psi}^\dagger(x) \hat{\alpha}(x) \hat{\psi}(x) | \Psi_n \rangle,$$

with the initial state (94). In terms of the mode expansion (B11) for the vector potential, a straightforward calculation in the continuum limit leads to

$$\Delta E_{n,(1)}^\Lambda = \frac{\hbar^2 \alpha N_e}{\pi m} \int_0^\Lambda k dk = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2.$$

To obtain the second-order correction, consider the matrix element

$$\langle \Psi_\delta | H_{\text{int}}^{(2)} | \Psi_n \rangle = -\frac{1}{c} \int dx \langle \Psi_\delta | j_P(x) \cdot \alpha(x) | \Psi_n \rangle.$$

Again using the mode expansion (B11)

$$\langle \Psi_\delta | H_{\text{int}}^{(2)} | \Psi_n \rangle = -\left( \frac{2\pi \hbar}{c |k| V} \right)^{1/2} e^{i(-k)} j_P^{(2)}(x),$$

where $j^{(2)}_P(x) \equiv (j_P(x))_{mn}$. Therefore, we have

$$\left| \langle \Psi_\delta | H_{\text{int}}^{(2)} | \Psi_n \rangle \right|^2 = \left( \frac{2\pi \hbar}{c |k| V} \right)^{1/2} e^{i(-k)} j_P^{(2)}(x).$$

Introducing the sums (114) and taking the continuum limit, the regularized second-order correction is

$$\Delta E_{n}^{(2)} = -\frac{\hbar}{\pi c} \sum_m \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \sum_I |e_I(k) \cdot j_{P, mn}(k)|^2.$$

Collecting the first-order and second-order corrections together, the regularized energy shift at $O(\alpha)$ in minimal coupling is

$$\Delta E_{n}^{\Lambda} = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{\pi c} \sum_m \int_{|k| \leq \Lambda} dk \frac{|k|^{-1}}{E_{mn} + \hbar c |k|} \sum_I |e_I(k) \cdot j_{P, mn}(k)|^2,$$

which is exactly the result (119) that we obtained using the multipolar Hamiltonian, as expected.

### Appendix E: Proof of identity (157)

We prove the following identity

$$\sum_m j_{P, mn}^j(-k) E_{mn}^R + \hbar c |k| j_{P, mn}^j(k) = \sum_m j_{P, mn}^j(-k) 2 j_{P, mn}^j(k) E_{kk} + \hbar c |k| \left( 1 - \frac{U_{mn}^R}{E_{mn}^R + \hbar c |k|} \right).$$

To begin, consider the operator expression

$$H_{F}^R - E_n + \hbar c |k| = \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} - \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} U^{R} \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|},$$

from which follows

$$\frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} j_{P, mn}^j(k) = \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} j_{P, mn}^j(k) - \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} U^{R} \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} j_{P, mn}^j(k).$$

To simplify, we use our energy shift result for $H_{F}^{(0)}$, namely

$$\frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|} j_{P, mn}^j(k) = -i \int_0^\infty ds e^{i \delta E_n + H_{F}^{(0)}} j_{P, mn}^j(k)$$

$$= -i \int_0^\infty ds e^{i \delta E_n + H_{F}^{(0)}} j_{P, mn}^j(k) e^{-i \delta E_n + H_{F}^{(0)}} e^{i \delta E_n + H_{F}^{(0)}}$$

$$= j_{P, mn}^j(k) \frac{1}{H_{F}^{(0)} - E_n + \hbar c |k|}.$$
where we have dropped the $|q| \ll mc$ terms, as before. Therefore we have

$$\frac{1}{H_F^R - E_n + \hbar c |k|} \tilde{j}_P^R (k) = \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|} \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|}$$

$$= \frac{1}{H_F^{(0)} - E_n + \hbar c |k|} \left[ U_R^{(1)} \tilde{j}_P^R (k) \right] \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|}$$

and so bringing the last term to the left side

$$\frac{1}{H_F^{R} - E_n + \hbar c |k|} \tilde{j}_P^R (k) \left( H_F^{R} - E_n + E_{hk} + \hbar c |k| \right) \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|}$$

$$= \tilde{j}_P^R (k) \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|} - \frac{1}{H_F^{(0)} - E_n + \hbar c |k|} \left[ U_R^{(1)} \tilde{j}_P^R (k) \right] \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|}$$

(E5)

Therefore, cancelling the factor $(H_F^{(0)} - E_n + E_{hk} + \hbar c |k|)^{-1}$ that features on both sides and multiplying the result by $(H_F^{R} - E_n + E_{hk} + \hbar c |k|)^{-1}$ on the right, we end up with

$$\frac{1}{H_F^{R} - E_n + \hbar c |k|} \tilde{j}_P^R (k) = \frac{1}{H_F^{(0)} - E_n + E_{hk} + \hbar c |k|} - \frac{1}{H_F^{(0)} - E_n + \hbar c |k|} \left[ U_R^{(1)} \tilde{j}_P^R (k) \right]$$

(E6)

Using that $H_F^{R} |\psi_{n,m} \rangle = E_{n,m} |\psi_{n,m} \rangle$, this implies

$$\left\langle \frac{1}{H_F^{R} - E_n + \hbar c |k|} \tilde{j}_P^R (k) \right\rangle_{m,n} = \frac{1}{E_{n,m} + \hbar c |k|} \tilde{j}_P^{R,m} (k)$$

$$= \tilde{j}_P^{R,m} (k) \frac{1}{E_{h,k} + \hbar c |k|} - \frac{1}{E_{m,n} + \hbar c |k|} \left\langle \left[ U_R^{(1)} \tilde{j}_P^R (k) \right] \right\rangle_{m,n} \frac{1}{E_{h,k} + \hbar c |k|}$$

(E7)

For simplicity, we define

$$\tilde{j}_P^{R,m} (k) U_R^{R,m} \equiv \left\langle \left[ U_R^{(1)} \tilde{j}_P^R (k) \right] \right\rangle_{m,n}$$

(E8)

with which our result follows.

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