Multipolar quantum electrodynamics of localized charge-current distributions: Spectral 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, often referred to as the Power-Zienau-Woolley transformation; we renormalize this Hamiltonian using perturbation theory, the result of which is used to compute the leading-order radiative corrections to 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, our results can be used to study contributions to this energy shift coming from specific multipole moments of arbitrary order.

I. INTRODUCTION

While relativistic quantum electrodynamics has been tremendously successful in describing scattering processes in high energy physics, it is cumbersome when applied to bound state problems and those with non-relativistic sources [1, 2]. 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 [3]. 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 [4, 5].

The result is an effective field theory, called non-relativistic quantum electrodynamics [6], that is particularly well-suited for describing interactions between many-body systems with a fixed number of particles and quantized radiation fields. It is obtained from an ultraviolet completion with only one characteristic energy scale (set by the electron mass), implying that the infinite sum of interaction terms in its Lagrangian can be ordered in reciprocal powers of this mass [7]; 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 \((\rho, j)\) that couple to the \(\textnormal{U}(1)\)-gauge potentials \((\phi, a)\) rather than directly to the electromagnetic field.

In many systems, however, an alternative approach is possible, provided one can identify specific points in space around which it is physically permissible to perform multipole expansions. This approach is based on a formalism called multipolar electrodynamics [8–11], which has been specifically designed to study such systems. In this paradigm, the material degrees of freedom are encoded in polarization and magnetization fields \((m, j)\) in place of charge and current densities \((\rho, j)\), which couple to the “Maxwell fields” \((\mathbf{d} + \mathbf{b})\) rather than the gauge potentials \((\phi, a)\). These descriptions are related by

\[
\rho(x, t) = -\nabla \cdot \mathbf{p}(x, t) + \rho_F(x, t),
\]

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

allowing for the possibility of free charge and current densities \((\rho_F, j_F)\), which would arise if the atom or molecule had a net charge and was free to move [12]. In its Hamiltonian formulation, the standard procedure for obtaining the quantum theory of multipolar electrodynamics involves a unitary transformation of the minimal coupling Hamiltonian [13, 14], often referred to as the Power-Zienau-Woolley (PZW) transformation after its originators [8, 9, 15]. 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 [16].

Multipolar electrodynamics was originally formulated in terms of the many-body wavefunctions of “first-quantized” quantum mechanics [17]. 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 [18].

Even in second-quantized treatments, the multipolar Hamiltonian is usually simplified by making the ubiq-
uitious “electric dipole approximation” [19–23], neglecting higher-order electric and magnetic multipole moments that become important when spatial variations of the electromagnetic field over the atom or molecule are non-negligible. And, indeed, for larger systems, such as molecules with several distinct chromophores, spatial variations in optical fields are non-negligible, and the electric dipole approximation is invalid [12]. Cases involving multipole moments beyond the electric dipole have been investigated previously [24], notably in calculations of intermolecular dispersion interactions [25] and resonance energy transfer rates [26]; however, the formulation of second-quantized models of radiation-matter interactions that are also capable of including electric and magnetic multipole moments of arbitrary order has not yet been addressed.

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 [10, 27, 28], the focus of those studies was the fields in the vicinity of an atom or molecule [29] and the impact of the associated local field corrections on intermolecular interactions [30–32]. 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 [33], spontaneous emission [34], resonance energy transfer rates [35], and the Casimir [36] and Casimir-Polder interactions [25, 37].

In this first communication, we use perturbation theory to calculate the leading-order radiative 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 generally 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 [5]. Our renormalized energy shift constitutes a generalization of Bethe’s original calculation of the Lamb shift in atomic hydrogen [38], valid for general localized assemblies of atoms and molecules, with or without net charge. In its multipolar form, this renormalized energy shift can be expanded in a sum of contributions coming from an arbitrary number of electric and magnetic multipole moments, so that vacuum effects can be studied order-by-order in the multipole series.

We begin in Section II by recalling the essential features of minimal coupling electrodynamics. The minimal coupling Lagrangian is obtained directly from the Schrödinger Lagrangian by replacing partial derivatives with U(1)-covariant derivatives and adding to the result a kinetic term yielding the free Maxwell equations [39]. To obtain the Hamiltonian field theory, one employs Dirac’s extended Hamiltonian formalism [40], since the minimal coupling Lagrangian is degenerate and the associated Hamiltonian will therefore be subject to constraints. Briefly summarizing this standard analysis in Appendix A.1, we obtain the quantized minimal coupling Hamiltonian, together with the commutation/anticommutation relations for the field operators. Applying the unitary PZW transformation to the canonical variables and Hamiltonian of minimal coupling (summarized in Appendix A.2), the quantized Hamiltonian theory of multipolar electrodynamics follows.

In Section III we present a sketch of the procedure we follow to regularize and renormalize the multipolar Hamiltonian, and in Section IV we compute the regularized energy shift of the electronic energy levels in the electromagnetic vacuum state. We regularize the divergent integrals with a hard cutoff \( |k| \leq \Lambda \), introducing a second energy scale into the theory. However, this scale is artificial and should not appear in expressions for measurable quantities; to remove this \( \Lambda \)-dependence from our energy shift, we renormalize the multipolar Hamiltonian following the procedure outlined in Section III. This is done in Section V, the result of which is 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 [38], and its generalizations when spatial variations in the electromagnetic field over the atom are included agree with the results of other authors [41, 42] 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. We summarize our results in Section VI, and some of the details are relegated to the Appendices.

II. HAMILTONIAN THEORY

A. Minimal coupling

We consider a system of electrons interacting with one or more point-like ions at fixed positions clustered around a specific point \( \mathbf{R} \in \mathbb{R}^3 \). 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 $\rho_{\text{ion}}(x)$ 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

$$V(x) = e \sum_N \frac{q_N}{\|x - R - d_N\|^4},$$

where $\|x\| = \sqrt{x \cdot x}$ is the Euclidean 2-norm and $e = -|e|$ 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 Lagrangian density

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

(3)

To couple the electron field $\psi(x, t)$ to a classical electromagnetic field, we apply the standard minimal coupling prescription to the Lagrangian density above [39]. In the four-vector notation of relativistic mechanics, this amounts to the replacement

$$\partial_\mu \rightarrow \partial_\mu + \frac{ie}{\hbar c} a_\mu(x, t) \equiv \mathcal{D}_\mu(x, t),$$

(4)

where the object on the right is a $\text{U}(1)$–covariant derivative and $a_\mu(x, t)$ is the electromagnetic four-potential. And to give dynamics to the electromagnetic field we add to the result a kinetic term

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

(5)

associated with the free Maxwell equations. The $\text{U}(1)$–gauge potentials $(\phi(x, t), a(x, t))$ are related to the electric and magnetic fields $(e(x, t), b(x, t))$ through the usual relations [43]

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

(6)

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

(7)

Then the total Lagrangian density for non-relativistic quantum electrodynamics in minimal coupling is

$$\mathcal{L} = \frac{1}{8\pi} \left( e(x, t) \cdot e(x, t) - b(x, t) \cdot b(x, t) \right) + \frac{i\hbar}{2} \left( \psi^\dagger(x, t) \dot{\psi}(x, t) - \dot{\psi}^\dagger(x, t) \psi(x, t) \right) - \psi^\dagger(x, t) V(x) \psi(x, t)$$

$$- \frac{\hbar^2}{2m} (\mathcal{D}(x, t) \psi(x, t))^\dagger \cdot (\mathcal{D}(x, t) \psi(x, t)) - \rho^e(x, t) \phi(x, t),$$

(8)

where $\mathcal{D}(x, t)$ are the spatial components of the covariant derivative defined in (4) and we have introduced the electronic charge density

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

(9)

Expanding the covariant derivative, we can write the total Lagrangian associated to $\mathcal{L}$ as a sum of three terms

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

(10)

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

$$L_{\text{int}} = - \int dx \, \rho^e(x, t) \phi(x, t) + \frac{\hbar e}{2mc} \int dx \left( \dot{\psi}^\dagger(x, t) \nabla \psi(x, t) - \nabla \dot{\psi}^\dagger(x, t) \psi(x, t) \right) \cdot a(x, t)$$

$$- \frac{e^2}{2mc^2} \int dx \, \psi^\dagger(x, t) ||a(x, t)||^2 \psi(x, t).$$

(11)

To obtain the Hamiltonian field theory we apply the Legendre transformation to $L$, which is summarized in Appendix A.1. The Hamiltonian operator that we obtain from $L$ after quantization is given in the Schrödinger picture by

$$H = H_B \otimes \mathbb{I}_F + \mathbb{I}_B \otimes H_F + H_{\text{int}}$$

(12)

generating unitary dynamics in a composite Hilbert space $\mathcal{H}_B \otimes \mathcal{H}_F$ constructed from Hilbert spaces $\mathcal{H}_B$ and $\mathcal{H}_F$ associated with the Bose and Fermi sectors, respectively.
In terms of the transverse electric and magnetic field operators, the Maxwell Hamiltonian is

$$H_B = \frac{1}{8\pi} \int dx \left( e_T(x) \cdot e_T(x) + b(x) \cdot b(x) \right),$$  \hspace{1cm} (13)$$
the electronic term is

$$H_F = \frac{\hbar^2}{2m} \int dx \left( \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + \frac{1}{2} \int dx \rho^\dagger(x) \phi(x) \right. \right.$$ 
$$\left. + \int dx \psi^\dagger(x) V(x) \psi(x), \right)$$  \hspace{1cm} (14)
and the interaction term is

$$H_{\text{int}} = -\frac{1}{c} \int dx \ j_F(x) \cdot a(x) - \frac{1}{2c} \int dx \ j_D(x) \cdot a(x).$$  \hspace{1cm} (15)$$
Here we have introduced the paramagnetic and diamagnetic current density operators

$$j_F(x) = \frac{e}{2mc} \psi^\dagger(x) \nabla \psi(x) - \nabla \psi^\dagger(x) \psi(x),$$  \hspace{1cm} (16)$$
$$j_D(x) = -\frac{e^2}{mc} \psi^\dagger(x) a(x) \psi(x),$$  \hspace{1cm} (17)$$
such that the total current density operator is

$$j(x) = j_F(x) + j_D(x),$$  \hspace{1cm} (18)$$
which satisfies the Heisenberg-picture equation

$$\frac{\partial \rho^\dagger(x,t)}{\partial t} + \nabla \cdot j(x,t) = 0$$  \hspace{1cm} (19)$$
associated with local U(1)–charge conservation. The electromagnetic field operators satisfy the (equal-time) commutation relations

$$\left[ e^\dagger_T(x), a^\dagger(y) \right]_+ = 4\pi i \hbar c \delta^{ij}_{\text{tr}}(x-y),$$  \hspace{1cm} (20)$$
$$\left[ a^\dagger(x), a^\dagger(y) \right]_+ = \left[ e^\dagger_T(x), e^\dagger_T(y) \right]_+ = 0, $$  \hspace{1cm} (20)$$
from which follows

$$\left[ e^\dagger_T(x), b^\dagger(y) \right]_+ = 4\pi i \hbar c \epsilon^{ijk} \frac{\partial}{\partial y^k} \delta(x-y),$$  \hspace{1cm} (21)$$
while the electron field operator and its adjoint satisfy the (equal-time) anticommutation relations

$$\left[ \psi(x), \psi^\dagger(y) \right]_+ = \delta(x-y),$$  \hspace{1cm} (22)$$
$$\left[ \psi(x), \psi^\dagger(y) \right]_+ = \left[ \psi^\dagger(x), \psi^\dagger(y) \right]_+ = 0.$$  \hspace{1cm} (22)$$
In the commutation relations (20) we have introduced the transverse delta function $\delta^{ij}_{\text{tr}}(x-y)$, which is used together with the longitudinal delta function $\delta^{ij}_{\text{lo}}(x-y)$ to decompose a given vector field $\mathcal{O}^j(x)$ into its transverse and longitudinal components [12]

$$\mathcal{O}^\dagger_T(x) = \int dy \ \delta^{ij}_{\text{tr}}(x-y) \mathcal{O}^j(y),$$  \hspace{1cm} (23)$$
$$\mathcal{O}^\dagger_L(x) = \int dy \ \delta^{ij}_{\text{lo}}(x-y) \mathcal{O}^j(y).$$  \hspace{1cm} (24)$$

B. Multipolar quantum electrodynamics

In classical or semiclassical theory, the multipolar Hamiltonian is obtained from the minimal coupling Hamiltonian through a canonical transformation. After the transformation, the Hamiltonian involves the electric and magnetic fields rather than the scalar and vector potentials, together with polarization and magnetization fields that are the microscopic analogues of the polarization and magnetization fields appearing in elementary classical electrodynamics [43]. 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, called the PZW transformation [17], that acts on the composite Hilbert space $\mathcal{H}_B \otimes \mathcal{H}_F$, generated by

$$S = \frac{1}{c} \int dx \ p(x) \cdot a(x),$$  \hspace{1cm} (25)$$
where $p(x)$ is the polarization field operator, which is related to the total charge density operator

$$\rho(x) = \rho^c(x) + \rho^{\text{ion}}(x)$$  \hspace{1cm} (26)$$
through the identity

$$\rho(x) = -\nabla \cdot p(x) + \rho_F(x).$$  \hspace{1cm} (27)$$
The free charge density $\rho_F(x)$ is nonzero only if the system has net charge, in which case

$$\rho_F(x) = Q \delta(x - R)$$  \hspace{1cm} (28)$$
with net charge

$$Q = \int dx \ \rho^c(x) + \sum_N q_N.$$  \hspace{1cm} (29)$$
We allow $Q$ to be non-zero to include single ions or charged molecules. A suitable polarization field operator that satisfies (27) is

$$p(x) = \int dy \ s(x; y, R) \rho(y),$$  \hspace{1cm} (30)$$
where we have introduced a “relator” $s(x; y, R)$ [6], defined by the distributional expression

$$s(x; y, R) = \int_{C(y, R)} dz \ \delta(x - z)$$  \hspace{1cm} (31)$$
with $C(y, R)$ an arbitrary continuously differentiable curve in $\mathbb{R}^3$ that begins at $R$ and ends at $y$. If a straight-line path is chosen between $R$ and $y$, then the usual multipole expansion of the polarization field follows.

After applying the PZW transformation, which is summarized in Appendix A2, the multipolar Hamiltonian that follows is

$$H = H_B \otimes 1 + 1_B \otimes H_F + H_{\text{int}},$$  \hspace{1cm} (32)$$
where $H_B$ is now written in terms of the transverse displacement field instead of the transverse electric field

$$H_B = \frac{1}{8\pi} \int dx \left( d_T(x) \cdot d_T(x) + b(x) \cdot b(x) \right), \quad (33)$$

the electronic term is

$$H_F = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + 2\pi \int dx \|p_L(x)\|^2 - \int dx d_L(x) \cdot \left(p(x) - \frac{1}{8\pi} d_L(x)\right), \quad (34)$$

and the interaction term is

$$H_{\text{int}} = 2\pi \int dx \|p_T(x)\|^2 - \int dx p(x) \cdot d_T(x) - \int dx m_P(x) \cdot b(x) - \frac{1}{2} \int dx m_D(x) \cdot b(x). \quad (35)$$

Importantly, here and henceforth the electron field operator $\psi(x)$ is the *transformed* operator (see Appendix A 2). The longitudinal displacement field, defined through Gauss’s law, $\nabla \cdot d_L(x) = 4\pi \rho_F(x)$, is given by

$$d_L(x) = \frac{1}{4\pi} \int dy \rho_F(y) \left( \frac{\hat{x} - \hat{y}}{||x - y||^2} \right). \quad (36)$$

In the interaction term (35) $m_P(x)$ and $m_D(x)$ refer to the paramagnetic and diamagnetic contributions to the (orbital) magnetization field operator

$$m(x) = m_P(x) + m_D(x). \quad (37)$$

Explicitly, the field operators $m_P(x)$ and $m_D(x)$ are

$$m_P^\dagger(x) = \frac{1}{c} \int dy \alpha^{ij}(x; y, R) j_P^\dagger(y), \quad (38)$$

$$m_D^\dagger(x) = \frac{1}{c} \int dy \alpha^{ij}(x; y, R) j_D^\dagger(y), \quad (39)$$

where the paramagnetic and diamagnetic current density operators are

$$j_P(x) = \frac{\hbar e}{2mi} \left( \psi^\dagger(x) \nabla \psi(x) - \nabla \psi^\dagger(x) \psi(x) \right), \quad (40)$$

$$j_D(x) = -\frac{e^2}{mc} \psi^\dagger(x) \Omega_R(x) \psi(x), \quad (41)$$

and we have introduced another “relator” [6]

$$\alpha^{ij}(x; y, R) = \epsilon^{ijn} \int_C(y, R) d\xi^m \frac{\partial z^n}{\partial y^j} \delta(x - z). \quad (42)$$

As is the case for the polarization field (30), taking a straight-line path for $C(y, R)$ yields the multipole expansion of the magnetization fields (38,39). In multipolar electrodynamics, the diamagnetic current density (41) depends not on the vector potential, but instead on the magnetic field through

$$\Omega_R^\dagger(x) = \int dy \alpha^{ij}(y, x, R) b^\dagger(y). \quad (43)$$

The charge and current density operators are related to the polarization and magnetization field operators through the Heisenberg-picture identities

$$\rho(x, t) = -\nabla \cdot p(x, t) + \rho_F(x), \quad (44)$$

$$j(x, t) = -\frac{\partial p(x, t)}{\partial t} + c \nabla \times m(x, t). \quad (45)$$

### 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 $||k|| > \Lambda$. We denote by $H^\Lambda$ our Hamiltonian $H$ subject to this cutoff, which we will henceforth refer to as the *regularized Hamiltonian*, and using the same superscript to denote regularized versions of the different contributions to $H$. 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 the electron-positron pair production that we neglect would not be present in the full theory of QED [44].

In the next section we will use Rayleigh-Schrödinger perturbation theory to compute the leading-order radiative 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 cutoff” 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 *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 correction to the *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 $H^\Lambda$, with the purpose of simulating virtual processes associated with the high energy ($||k|| > \Lambda$) degrees of freedom [45]. In general, $\delta H(\Lambda)$ will be a sum of many terms, each summand being called a *counterterm*. A central tenet of renormalization theory is that it should be possible to choose the counterterms...
in $\delta H(\Lambda)$ such that the renormalized Hamiltonian

$$H^R = H^A + \delta H(\Lambda)$$

(46)

accurately represents the full range of energies in the interval $0 \leq ||k|| < \infty$ and should not depend on $\Lambda$ [46]. The renormalized energy shift, denoted by $\Delta E_R^\delta$ 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 $||k||$ 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 non-renormalizable field theory, so we only expect $\Delta H$ [39]. If we want to compute observables at next-to-leading-order, for example, we must check if additional UV divergences are generated in non-renormalizable theories as one moves to higher orders, as well as divergences that are generated by the counterterms in $\delta H(\Lambda)$ [39]. If we want to compute observables at next-to-leading-order, for example, we must check if additional UV divergences 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 that 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 typically used. Finally, we identify the two contributions that arise from the interaction of the radiation and electron fields. They are then worked out in detail in Sections IV B and IV C below.

1. Free fields

The dynamical evolution of the transverse Maxwell fields $d_T(x,t)$ and $b(x,t)$ is generated by $H_B$ through the Heisenberg equations

$$\frac{\partial d_T(x,t)}{\partial t} = \frac{1}{\hbar} [d_T(x,t), H_B]_-, \quad (47)$$

$$\frac{\partial b(x,t)}{\partial t} = \frac{1}{\hbar} [b(x,t), H_B]_-, \quad (48)$$

which, with the explicit form (33) for $H_B$, lead to the familiar Maxwell equations

$$\frac{\partial d_T(x,t)}{\partial t} = +c \nabla \times b(x,t), \quad (49)$$

$$\frac{\partial b(x,t)}{\partial t} = -c \nabla \times d_T(x,t), \quad (50)$$

subject to the constraints

$$\nabla \cdot d_T(x,t) = 0, \quad (51)$$

$$\nabla \cdot b(x,t) = 0. \quad (52)$$

We quantize in a box of volume $V = L^3$, in which case the allowed wavevectors are

$$k = \frac{2\pi}{L} (n_x, n_y, n_z), \quad (53)$$

and expand $d_T(x,t)$ 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

$$e_{lk} = -\frac{1}{\sqrt{2}} (e_{1k} + ie_{2k}),$$

$$e_{lk} = +\frac{1}{\sqrt{2}} (e_{1k} - ie_{2k}), \quad (54)$$

where $e_{1k}$ and $e_{2k}$ are the standard Cartesian polarization vectors [12]. The circular polarization vectors satisfy

$$e_{lk} \cdot e_{l(-k)} = \delta_{ll'},$$

$$i \hat{k} \cdot e_{lk} = s_l e_{lk}, \quad (56)$$

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

$$\sum_I e_{I(-k)} e_{Ik}^\dagger = \frac{k^I k^j}{||k||^2} \equiv \delta_{Ij}^k(k). \quad (57)$$

The Maxwell field operators are

$$d_T(x,t) = i \sum_{lk} \left( \frac{2\pi \hbar \omega_k}{V} \right)^{1/2} e_{lk} a_{lk} e^{i(k \cdot x - \omega_k t)} + \text{h.c.},$$

$$b(x,t) = \sum_{lk} \left( \frac{2\pi \hbar \omega_k}{V} \right)^{1/2} s_l e_{lk} a_{lk} e^{i(k \cdot x - \omega_k t)} + \text{h.c.}, \quad (58)$$

where $\omega_k = c ||k||$. To enforce the commutation relations (20,21), the photonic creation and annihilation operators $a_{lk}^\dagger$ and $a_{lk}$ must obey the equal-time canonical commutation relations

$$[a_{lk}, a_{lk'}^\dagger]_0 = \delta_{lj} \delta_{kk'}, \quad (59)$$

$$[a_{lk}, a_{lk'}]_0 = [a_{lk}^\dagger, a_{lk'}^\dagger]_0 = 0. \quad (60)$$
After regularization, the Maxwell Hamiltonian $H_B$ is given in the Schrödinger picture by

$$H_B^\Lambda = \sum_I \sum_{|k| \leq \Lambda} \hbar \omega_k \left( a^\dagger_{1k} a_{1k} + \frac{1}{2} \right).$$  \hspace{1cm} (61)

The second term is the well-known divergence associated with the zero-point energy in free quantum electrodynamics [34, 39]. 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

$$E_{0,B}^\Lambda \equiv \sum_{|k| \leq \Lambda} \hbar \Omega \|k\| \rightarrow \frac{\hbar c V}{8\pi^2} \Lambda^4,$$  \hspace{1cm} (62)

where the right side follows in the continuum limit

$$\sum_k \rightarrow \frac{V}{8\pi^2} \int d\mathbf{k}.$$  \hspace{1cm} (63)

Therefore, the regularized Maxwell Hamiltonian is

$$H_B^\Lambda = \sum_I \sum_{|k| \leq \Lambda} \hbar \omega_k a^\dagger_{1k} a_{1k} + E_{0,B}^\Lambda.$$  \hspace{1cm} (64)

Since we are primarily interested in vacuum effects, we take as our basis for $\mathcal{H}_B$ the photon number states $|\{n_{1k}\}\rangle \in \mathcal{H}_B$. The spectral problem for $H_B^\Lambda$ in terms of these eigenstates is

$$H_B^\Lambda \left| \{n_{1k}\}\right\rangle = E_{B}^\Lambda \left| \{n_{1k}\}\right\rangle,$$  \hspace{1cm} (65)

where the energy eigenvalues are

$$E_{B}^\Lambda = \sum_I \sum_{|k| \leq \Lambda} \hbar \omega_k n_{1k} + E_{0,B}^\Lambda.$$  \hspace{1cm} (66)

Turning to the electron field, time evolution in the electronic Hilbert space $\mathcal{H}_E$ is generated by $H_F$ through the Heisenberg equation

$$\frac{\partial \psi(x,t)}{\partial t} = \frac{1}{i\hbar} \left[ \psi(x,t), H_F \right],$$  \hspace{1cm} (67)

with $H_F$ given by (34). As shown in Appendix B, the regularized electronic Hamiltonian $H_F^\Lambda$ is given in the Schrödinger picture by

$$H_F^\Lambda = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) + \frac{1}{2} \sum_{\mathbf{k}} \psi^\dagger(\mathbf{x}) \psi^\dagger(\mathbf{x}') \frac{e^2}{\|x - x'\|^2} \psi(\mathbf{x}') \psi(\mathbf{x}) + \int dx \psi^\dagger(x) V(x) \psi(x) + E_{0,F}^\Lambda,$$  \hspace{1cm} (68)

where $E_{0,F}^\Lambda$ is a $\Lambda$-dependent term associated with the $\|x - x'\| \rightarrow 0$ limit of the electrostatic Coulomb potential, and is given by

$$E_{0,F}^\Lambda = \frac{1}{\pi} \left[ e^2 N_e + 2e N_e \sum_N q_N + \sum_N q_N^2 \right] \Lambda,$$  \hspace{1cm} (69)

where $N_e$ is the total number of electrons. 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

$$E_{n}^\Lambda = E_{n}^\Lambda \left| \psi_n\right\rangle,$$  \hspace{1cm} (70)

where the energy eigenvalues are

$$E_{n}^\Lambda = \frac{\hbar^2}{2m} \int dx \left( \nabla \psi^\dagger(x) \cdot \nabla \psi(x) \right)_n + \frac{1}{2} \sum_{\mathbf{k}} \psi^\dagger(\mathbf{x}) \psi^\dagger(\mathbf{x}') \frac{e^2}{\|x - x'\|^2} \psi(\mathbf{x}') \psi(\mathbf{x})_n + \int dx \left( \psi^\dagger(x) V(x) \psi(x) \right)_n + E_{0,F}^\Lambda,$$  \hspace{1cm} (71)

and where the expectation value is over the state $|\psi_n\rangle$.

2. Interactions

In the interacting theory the dynamical evolution of the field operators is generated by the full Hamiltonian $H$ given in (32-35). After regularization, we write this Hamiltonian as

$$H^\Lambda = H_0^\Lambda + H_{\text{int}},$$  \hspace{1cm} (72)

where

$$H_0^\Lambda = H_0^\Lambda \otimes 1_F + 1_B \otimes H_F^\Lambda,$$  \hspace{1cm} (73)

with $H_B^\Lambda$ and $H_F^\Lambda$ given in (64) and (68), while $H_{\text{int}}$ is given by (35) with all of the Fourier series (implicit in the mode expansions (58)) subject to the cutoff $|\mathbf{k}| \leq \Lambda$.

We take as our basis for the composite Hilbert space $\mathcal{H}_E \otimes \mathcal{H}_F$ the product states

$$|\{n_{1k}\}; \psi_n\rangle \equiv |\{n_{1k}\}\rangle \otimes |\psi_n\rangle.$$  \hspace{1cm} (74)

The spectral problem for the regularized free Hamiltonian $H_0^\Lambda$ in terms of these eigenstates is

$$H_0^\Lambda \{|\psi_n\rangle; \{n_{1k}\}\} = E_0^\Lambda + E_{\{n_{1k}\}; \rho_n}\{|\psi_n\rangle; \{n_{1k}\}\},$$  \hspace{1cm} (75)

with $E_0^\Lambda$ and $E_{\{n_{1k}\}; \rho_n}$ given by (66) and (71), respectively. We want to compute the regularized energy shift $\Delta E_{\{n\}}^\Lambda$ of the electronic energy levels $E_{\{n\}}^\Lambda$ in the electromagnetic vacuum, resulting from interactions described by $H_{\text{int}}$, and we consider only bound states $|\psi_n\rangle$ in the Fermi sector. Since we will do so using perturbation theory at $O(\alpha)$, it is useful to split up the interaction term

$$H_{\text{int}} = H_{\text{int}(1)} + H_{\text{int}(2)},$$  \hspace{1cm} (76)

where

$$H_{\text{int}(1)} = 2\pi \int dx \left| p_F(x) \right|^2 - \frac{1}{2} \int dx \mathbf{m}_D(x) \cdot \mathbf{b}(x)$$  \hspace{1cm} (77)
are the interactions that are already $\mathcal{O}(\alpha)$ and will be computed at the first order, while

$$H_{\text{int}(2)} = -\int dx \mathbf{p}(x) \cdot \mathbf{d}_T(x) - \int dx \mathbf{m}_P(x) \cdot \mathbf{b}(x)$$  

(78)

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

$$|\Psi_n\rangle \equiv |\text{vac}; \psi_n\rangle, \quad (79)$$

$$|\Psi_\delta\rangle \equiv |1_k; \psi_m\rangle.$$  

(80)

In terms of these states, the regularized correction to the electronic energy levels $E^A_n$ is

$$\Delta E^A_n = \Delta E^A_{n(1)} + \Delta E^A_{n(2)},$$

where

$$\Delta E^A_{n(1)} = \langle \Psi_n | H^A_{\text{int}(1)} | \Psi_n \rangle$$

(82)

is the first-order correction, while

$$\Delta E^A_{n(2)} = \sum_{\delta \neq n} \frac{\left| \langle \Psi_\delta | H^A_{\text{int}(2)} | \Psi_n \rangle \right|^2}{E_n - E_\delta}$$

(83)

is the second-order correction.

B. First-order correction

The first-order correction is

$$\Delta E^A_{n(1)} = -\frac{1}{2} \int dx \langle \Psi_n | \mathbf{m}_D(x) \cdot \mathbf{b}(x) | \Psi_n \rangle$$

$$+ 2\pi \int dx \langle \Psi_n | \|\mathbf{p}_T(x)\|^2 | \Psi_n \rangle,$$  

(84)

where we understand that the right-hand-side is to be regularized with a cutoff at $\|k\| = \Lambda$. The first term is the diamagnetic contribution to the regularized energy shift, which is more commonly written as

$$+ \frac{1}{2} \int dx dx' \langle \Psi_n | b^i(x) O^{ij}(x, x') b^j(x') | \Psi_n \rangle,$$  

(85)

where second-quantized diamagnetization field is [6]

$$O^{ij}(x, x') = \frac{e}{2mc^2} \int dy \alpha^i_k(x; y, R) \alpha^{jk}(x'; y, R) \rho^c(y).$$

(86)

We show in Appendix C that in the continuum limit (63) the diamagnetic contribution to the first-order correction is given by

$$\Delta E^A_{n(2)} = \frac{1}{2} \int dx \langle \Psi_n | \mathbf{m}_D(x) \cdot \mathbf{b}(x) | \Psi_n \rangle$$

$$= \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar^2}{mc^2} \int dx \rho^c_{nm}(x) \langle \mathbf{a}(x) \cdot \nabla \Psi(|\mathbf{x}, R\rangle)_{\text{vac}}$$

$$+ \frac{\hbar^2}{2mc^2} \int dx \langle \psi_n | \mathbf{b}(x) | \psi_n \rangle \langle \|\nabla \Psi(|\mathbf{x}, R\rangle)\|^2 \rangle_{\text{vac}},$$

(87)

where the expectation value is taken in the vacuum state of the radiation field. We will see below that when the full energy shift is calculated the latter two terms on the right-hand-side are cancelled by terms in the second-order correction $\Delta E^A_{n(2)}$.

Consider next the second term on the right-hand-side of (84). Inserting a resolution of the identity in $\mathcal{H}_F$ with respect to the many-body eigenstates $\{|\psi_m\rangle\}_m$, the expectation value

$$\langle \psi_n | p^i(x') p^j(x) | \psi_n \rangle = \sum_m \langle \psi_n | p^i(x') \rangle \langle p^j(x) | \psi_m \rangle,$$

(88)

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 [12]

$$\delta^T_{ij}(x' - x) = \int \frac{d\mathbf{k}}{(2\pi)^3} \delta^T_{ij}(\mathbf{k}) e^{i\mathbf{k} \cdot (x' - x)},$$

(89)

(recall (57)) we can (after regularization) write

$$2\pi \int dx \|\mathbf{p}_T(x)\|^2 = \frac{1}{4\pi^2} \sum_m \int_{\|k\| \leq \Lambda} d\mathbf{k} \delta^T_{ij}(\mathbf{k})$$

$$\times \int dx dx' p^i_{nm}(x') p^j_{nm}(x) e^{i\mathbf{k} \cdot (x' - x)},$$

(90)

To proceed, it is convenient to recast the angular part of the Fourier integral into an expression in coordinate space. Explicitly, we write [27]

$$\frac{1}{4\pi} \int d\Omega_\mathbf{k} \delta^T_{ij}(\mathbf{k}) e^{i\mathbf{k} \cdot (x' - x)} = \frac{1}{\|k\|^2} \tau^{ij}(k \|x' - x\|),$$

(91)

where we have defined

$$\tau^{ij}(k \|x' - x\|) \equiv (-\delta^{ij} \partial^2 + \partial^i \partial^j) F(x, x'; k),$$

(92)

We have introduced the abbreviated notation $d\Omega_\mathbf{k} =$
Hence the total first-order correction (84), the sum of (87) and (94), is
\[
\Delta E_{n(1)}^\Lambda = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right)^2 \Lambda^2 + \frac{1}{\pi} \sum_m \left( \frac{\hbar^2}{2mc} \right)^2 \int dk \int d\mathbf{x} d\mathbf{x}' \tau^{ij}(k) (|\mathbf{x}' - |\mathbf{x}|) \right) p_{nm}(\mathbf{x}') p_{nm}(\mathbf{x}).
\]

Hence the total first-order correction (84), the sum of (87) and (94), is
\[
\Delta E_{n(1)}^\Lambda = \alpha \frac{\hbar^2 N_e}{m} \Lambda^2 + \frac{1}{\pi} \sum_m \frac{\hbar^2}{2mc} \int d\mathbf{x} \rho_{nn}(\mathbf{x}) \langle \mathbf{a}(\mathbf{x}) \cdot \nabla \Phi(\mathbf{R}) \rangle_{\text{vac}} + \frac{\hbar^2}{2mc} \int d\mathbf{x} \rho_{nn}(\mathbf{x}) \langle \| \nabla \Phi(\mathbf{R}) \|^2 \rangle_{\text{vac}}.
\]

C. Second-order correction

To compute the second-order correction (83), we begin with the matrix element
\[
\langle \Psi_\delta | H_{\text{int}(2)}^\Lambda | \Psi_n \rangle = - \int d\mathbf{x} p_{mn}(\mathbf{x}) \langle 1_{\mathbf{k}} | d_T^j(\mathbf{x}) | \text{vac} \rangle - \int d\mathbf{x} m_{mn}(\mathbf{x}) \langle 1_{\mathbf{k}} | b^j(\mathbf{x}) | \text{vac} \rangle,
\]

\[
\Delta E_{n(2)}^\Lambda = - \frac{\hbar^2}{4\pi^2} \sum_m \int \frac{dk}{k} \int \frac{d\mathbf{x} d\mathbf{x}'}{E_{mn} + \hbar \omega_k} \left\{ \delta_{ij} \left( \frac{p_{mn}(\mathbf{x}') p_{mn}(\mathbf{x})}{E_{mn} + \hbar \omega_k} + m_{mn}(\mathbf{x}') m_{mn}(\mathbf{x}) \right) \right. \\
- \left. \left( e^{ipj} \frac{k^p}{k} \right) \left( p_{mn}(\mathbf{x}') m_{mn}(\mathbf{x}) + m_{mn}(\mathbf{x}') p_{mn}(\mathbf{x}) \right) \right\},
\]

where \( p_{mn}(\mathbf{x}) \equiv \langle \Psi_\delta | m^p(\mathbf{x}) | \Psi_n \rangle \) denotes the paramagnetic contribution to the magnetization. Inserting the mode expansions (58), and using that the energy difference between the states \( |\Psi_\delta \rangle \) and \( |\Psi_n \rangle \) is \( E_\delta - E_n = E_{mn} + \hbar \omega_k \), where \( E_{mn} = E_m - E_n \), the regularized sum in expression (83) is
\[
\sum_\delta \sum_m \sum_l \sum_{||k|| \leq \Lambda} (l),
\]

and after taking the continuum limit (63), we find
\[
\Delta E_{n(2)}^\Lambda = - \frac{1}{\pi} \sum_m \int_0^\Lambda dk \left( \frac{\hbar^2}{E_{mn} + \hbar \omega_k} \right)^2 \int d\mathbf{x} d\mathbf{x}' \tau^{ij}(k) |\mathbf{x}' - |\mathbf{x}|) \right) p_{mn}(\mathbf{x}) (p_{mn}(\mathbf{x}') + m_{mn}(\mathbf{x}) m_{mn}(\mathbf{x})),
\]

\[
\Delta E_{n(2)}^\Lambda = - \frac{1}{\pi} \sum_m \int_0^\Lambda dk \left( \frac{\hbar^2}{E_{mn} + \hbar \omega_k} \right) \int d\mathbf{x} d\mathbf{x}' \tau^{ij}(k) |\mathbf{x}' - |\mathbf{x}|) \right) p_{mn}(\mathbf{x}) (p_{mn}(\mathbf{x}') + m_{mn}(\mathbf{x}) m_{mn}(\mathbf{x})),
\]

where \( \sigma^{ij}(k) |\mathbf{x}' - |\mathbf{x}|\rangle \equiv \frac{i}{4\pi} \int d\Omega_k e^{ipj} k^p e^{ik \mathbf{x}} \). Further simplifications can be made using the relation (44) and the Schrödinger-picture identity
\[
\mathbf{j}_{P,nm}(\mathbf{x}) = (ih)^{-1} E_{mn}^R p_{mn}(\mathbf{x}) + c \nabla \times \mathbf{m}_{P,nm}(\mathbf{x}),
\]
proven in Appendix D. After a tedious but straightforward calculation, the total second-order correction is
\[
\Delta E_{n(2)}^A = -\frac{\hbar}{4\pi^2 e} \sum_{m} \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{mn} + \hbar \omega_k} \int dxdx' j^i_{P,nm}(x) j^j_{P,mn}(x) \delta^{ij}(k) e^{ik(x'-x)} \\
+ \frac{\hbar}{m c} \int dx \rho^{e}_{nn}(x) (\alpha(x) \cdot \nabla \Phi(x,R))_{\text{vac}} - \frac{\hbar^2}{2mc} \int dx \rho^{e}_{nn}(x) \langle \| \nabla \Phi(x,R) \|^2 \rangle_{\text{vac}} \\
- \frac{1}{\pi} \sum_{m} \int_{0}^{\Lambda} \frac{d\ell}{d\ell} \int dxdx' \tau^{ij}(k|\vec{x}' - \vec{x}|) p_{nm}^i(x') p_{mn}^j(x).
\]  

Hence, combining the first-order (95) and second-order (102) corrections, the regularized energy shift is
\[
\Delta E_{n}^A = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{4\pi^2 e} \sum_{m} \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{mn} + \hbar \omega_k} \int dxdx' j^i_{P,nm}(x) j^j_{P,mn}(x) \delta^{ij}(k) e^{ik(\vec{x}' - \vec{x})}. 
\]  

We can also write \( \Delta E_{n}^A \) in terms of the polarization vectors \( \{ \epsilon_i(k) \} \):
\[
\Delta E_{n}^A = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{4\pi^2 e} \sum_{m} \int_{|k| \leq \Lambda} \frac{|k|^{-1}}{E_{mn} + \hbar \omega_k} \sum_{i} \left| \epsilon_i(k) \cdot \hat{j}_{P,mn}(k) \right|^2, 
\]  

where \( \hat{j}_{P}(k) \) is the Fourier transform
\[
\hat{j}_{P}(\pm k) = \int dx \ j_{P}(x) e^{\mp ik \cdot x}. 
\]  

Existing treatments \([38, 41, 42]\) differ from ours either because they are computed within a first-quantized framework, or because they do not include spatial variations of the electromagnetic field over the molecule. If those variations would be neglected, the polarization sum in (104) would reduce to
\[
\sum_{i} \left| \epsilon_i(k) \cdot \hat{P}_{nm} \right|^2, 
\]  

where
\[
\hat{P} = \frac{\hbar}{i} \int dx \psi^\dagger(x) \nabla \psi(x) 
\]  
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 \( H_{0}^A \)

We first renormalize the free Hamiltonian \( H_{0}^A \), which produces the vacuum divergence \( E_{0,F}^A \) of free quantum electrodynamics given in (62) and the divergence \( E_{0,F}^A \) computed in Appendix B. Since both of these divergences are static, we can trivially remove them from \( H_{0}^A \) by addition of static counterterms \( \delta H_B(\Lambda) \) and \( \delta H_F(\Lambda) \) to \( H_{0}^A \) and \( H_{F}^A \). The renormalized free Hamiltonian is
\[
H_{0}^R = H_{B}^R \otimes \mathbb{1}_F + \mathbb{1}_B \otimes H_{F}^R, 
\]  

where the renormalized Maxwell and electronic terms are
\[
H_{B}^R = H_{B}^A + \delta H_B(\Lambda), \\
H_{F}^R = H_{F}^A + \delta H_F(\Lambda), 
\]  

and the counterterms (in minimal subtraction [39]) are
\[
\delta H_B(\Lambda) = - \left[ \frac{\hbar c V}{8\pi^2 \Lambda^4} \right] \mathbb{1}_B, \\
\delta H_F(\Lambda) = - \left[ \frac{1}{\pi} \left( e^2 N_e + 2e N_e \sum_N q_N + \sum_N q_N^2 \right) \Lambda \right] \mathbb{1}_F. 
\]  

The energy eigenvalues computed from \( H_{B}^R \) and \( H_{F}^R \) are finite in the limit \( \Lambda \to \infty \). In particular, the renormalized Maxwell term is
\[
H_{B}^R = \sum_{ik} \hbar \omega_k a_{ik}^\dagger a_{ik}, 
\]  

while the renormalized electronic term is
\[
H_{F}^R = T + U^R, 
\]  

where the kinetic term is
\[
T = \frac{\hbar^2}{2m} \int dx \nabla \psi^\dagger(x) \cdot \nabla \psi(x) 
\]
and the renormalized electrostatic interaction term is
\[ U^R = \frac{1}{2} \int \int dx \, dx' \, \psi(x) \psi(x') \left( \frac{e^2}{||x - x'||} \right) \psi(x') \psi(x) + \int dx \, \psi(x) V(x) \psi(x). \]

The renormalized electronic energy levels are the expectation values \( E_n^R = \langle H_n^R \rangle \). There are a host of techniques in atomic and molecular physics, and in condensed matter physics, that have been designed to compute expectation values of the operator given in (116) [48].

B. Renormalization of \( H^\Lambda_{\text{int}} \)

The renormalized interaction term is defined by
\[ H^R_{\text{int}} = H^\Lambda_{\text{int}} + \delta H_{\text{int}}(\Lambda), \]
where \( \delta H_{\text{int}}(\Lambda) \) will be determined below. As outlined in Section III, our renormalization scheme is based on the observation that the \( \Lambda \)-dependent contribution to the regularized energy shift (104) coming from free electrons, described by the Hamiltonian \( H_F^{(0)} \equiv T \) (see (115)), is unobservable and should be subtracted from \( \Delta E^\Lambda \). What remains will be the observable correction to the bound state electronic energy levels.

Thus we must first determine the contribution to \( \Delta E^\Lambda \) coming from free electrons, described by \( H_F^{(0)} \); denote by \( \{\langle \varphi_{n'} \rangle \}_{n'} \) the set of many-body eigenstates thereof. Because this Hamiltonian commutes with the total momentum operator \( \mathbf{P} \), we can choose each energy eigenstate \( \langle \varphi_{n'} \rangle \) to be an eigenstate of \( \mathbf{P} \). Then for an arbitrary many-body eigenstate \( \langle \varphi_{n' \lambda} \rangle \) we have
\[ \Delta E^\Lambda_{\text{free}} = \langle \varphi_{n' \lambda} | H^\Lambda_{\text{int}} | \varphi_{n' \lambda} \rangle, \]
\[ \Delta E^R_{\text{free}} = \langle \varphi_{n' \lambda} | H^R_{\text{int}} | \varphi_{n' \lambda} \rangle. \]

We choose the counterterms in \( \delta H_{\text{int}}(\Lambda) \) to cancel the \( \Lambda \)-dependent terms in \( \Delta E^R_{\text{free}} \), so that the renormalized energy shift \( \Delta E^R_{\text{free}} \) vanishes for an arbitrary state \( \langle \varphi_{n' \lambda} \rangle \), i.e., an arbitrary distribution of electron momenta.

To compute \( \Delta E^R_{\text{free}} \), it is useful to expand the electron field operators in a basis of plane waves
\[ \psi(x) = \frac{1}{\sqrt{V}} \sum_q e^{i \mathbf{q} \cdot \mathbf{x}} b_q, \]
where we return to considering a system in volume \( V \), and where the anticommutation relations (22) imply that the canonical anticommutation relations
\[ \{b_q, b^\dagger_{q'}\} = \delta_{qq'}, \]
\[ \{b_q, b_{q'}\} = [b_q, b^\dagger_{q'}] = 0. \]

In this plane-wave expansion, the Fourier transform of the paramagnetic current density operator is
\[ \tilde{j}_P(k) = \frac{\hbar e}{2m} \sum_q \left( 2q^j - k^j \right) b_{q-k}^\dagger b_q. \]

We obtain \( \Delta E^\Lambda_{\text{free}} \) by replacing the many-body eigenstates of \( H_F \) involved in (104) with those of \( H_F^{(0)} \), and replacing \( E_{mn} \) with \( E_{mn}' \), where \( E_{mn}' = \langle \varphi_{n' \lambda} | H_F^{(0)} | \varphi_{n' \lambda} \rangle \). Thus, for free electrons the regularized energy shift is
\[ \Delta E^\Lambda_{\text{free}} = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_e}{m} \right) \Lambda^2 - \frac{\hbar}{4\pi^2} \sum_{q} \int_{||k|| \leq \Lambda} \sum_{m'} \left| \tilde{e}_l(k) \cdot \tilde{j}_{P,m'n'}(k) \right|^2. \]

To simplify the second term, consider the expression
\[ \sum_{m'} \tilde{j}_{P,m'n'}(-k) \left( \frac{1}{E_{mn}^{(0)} + \hbar \omega_k} \right) \tilde{j}_{P,m'n'}^\dagger(k), \]
which features in the integrand of (123). To proceed, we
\[ \sum_{m'} \tilde{j}_{P,m'n'}(-k) \left( \frac{1}{E_{mn}' + \hbar \omega_k + i\delta} \right) \tilde{j}_{P,m'n'}^\dagger(k) = -i \sum_{m'} \int_0^\infty ds e^{i s (E_{mn}' + \hbar \omega_k + i\delta)} j_{P,m'n'}(k) e^{i s H_F^{(0)} j_{P}(k)} e^{-i s H_F^{(0)}}_{m'n'}, \]
where the subscript \( m'n' \) indicates the matrix element between state \( \langle \varphi_{m'n'} \rangle \) and state \( \langle \varphi_{n' \lambda} \rangle \). The operator in the
angular brackets on the right-hand-side is simply the paramagnetic current density operator in the interaction picture with \( s = t/\hbar \) (which acts trivially on \( H_B \)), and so we have

\[
e^{isH_F^{(0)}} j_p^{(0)} e^{-isH_F^{(0)}} = \frac{\hbar e}{2m} \sum_q \left( 2q^j - k^j \right) b_{q-k}^{\dagger} b_q e^{is(E_{\omega_k} - \hbar k \cdot q/m)},
\]

(127)

where \( E_{\omega_k} = \hbar^2 \omega_k^2/2mc^2 \). Therefore, using the identity (125) in reverse (and taking \( \delta \to 0^+ \))

\[
\sum_{m'} \tilde{j}_{p,m'}^{(0)}(k) \left( \frac{1}{E_{m'}^{(0)} + \hbar \omega_k} \right) \tilde{j}_{p,m'}^{(0)}(k) = \frac{\hbar^2 e^2}{4m^2} \sum_{qq'} \left( 2q^j - k^j \right) \left( 2q'^j - k'^j \right) \left( \frac{1}{E_{\omega_k} + \hbar \omega_k - \hbar k \cdot q/m} \right) \times \sum_{m'} \langle \varphi_{m'} | b_{q}^{\dagger} b_{q'-\hbar k} | \varphi_{m'} \rangle.
\]

(128)

Since we are working with electrons that are non-relativistic, \( ||q|| \ll mc \), and we can drop the “\( \hbar k \cdot q/m \)” term in the denominator. Recollecting the Fourier transforms of the paramagnetic current density operators using (122) we find

\[
\sum_{m'} \tilde{j}_{p,m'}^{(0)}(-k) \left( \frac{1}{E_{m'}^{(0)} + \hbar \omega_k} \right) \tilde{j}_{p,m'}^{(0)}(k) = \sum_{m'} \tilde{j}_{p,m'}^{(0)}(-k) \left( \frac{1}{E_{\omega_k} + \hbar \omega_k} \right) \tilde{j}_{p,m'}^{(0)}(k),
\]

(129)

from which follows

\[
\Delta E_{\text{free}}^\Lambda = \alpha \frac{\hbar^2 N_c}{m} \Lambda^2 - \frac{\hbar}{4\pi^2 e} \sum_{m'} \int_{||k|| \leq \Lambda} \frac{dk}{E_{\omega_k} + \hbar \omega_k} \sum_l \left| e_l(k) \cdot \tilde{j}_{p,m'}(k) \right|^2,
\]

(130)

where we have dropped the principal value notation, since the integrand has no poles (or infrared divergences).

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 (117) is finite in the limit \( \Lambda \to \infty \). However, instead of setting the counterterm directly, we proceed by enforcing the condition that \( \Delta E_{\text{free}}^\Lambda \) vanishes. Ensuring this condition requires that \( \langle \delta H_{\text{int}}(\Lambda) \rangle_{\varphi_{m'}} \) is the negative of the right-hand-side of (130) for all \( ||\varphi_{m'}|| \), and this can be satisfied by taking

\[
\delta H_{\text{int}}(\Lambda) = - \left[ \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_c}{m} \right) \Lambda^2 \right] \mathbb{I}_B \otimes \mathbb{I}_F + \frac{\hbar}{4\pi^2 e} \int_{||k|| \leq \Lambda} \frac{dk}{E_{\omega_k} + \hbar \omega_k} \sum_l \left( e_l(-k) \cdot \tilde{j}_p(k) \right) \left( e_l(k) \cdot \tilde{j}_p(k) \right).
\]

(131)

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

\[
H^\Lambda = H^\Lambda_B \otimes \mathbb{I}_F + \mathbb{I}_B \otimes H^\Lambda_F + H^\Lambda_{\text{int}}.
\]

(132)

The renormalized Hamiltonian is

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

(133)

where \( H^\Lambda \) is given by (72) and

\[
\delta H(\Lambda) = \delta H_B(\Lambda) \otimes \mathbb{I}_F + \mathbb{I}_B \otimes \delta H_F(\Lambda) + \delta H_{\text{int}}(\Lambda).
\]

(134)

Using (111,112) and (131), the counterterm \( \delta H(\Lambda) \) is

\[
\delta H(\Lambda) = - \left[ \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_c}{m} \right) \Lambda^2 + \frac{\hbar e V}{8\pi^2} \Lambda^4 + \frac{1}{\pi} \left( \frac{e^2 N_c}{2} + 2e^2 N_c \sum_q \frac{q^2}{q^2} \right) \right] \mathbb{I}_B \otimes \mathbb{I}_F
\]

\[
+ \frac{\hbar}{4\pi^2 e} \int_{||k|| \leq \Lambda} \frac{dk}{E_{\omega_k} + \hbar \omega_k} \sum_l \left( e_l(-k) \cdot \tilde{j}_p(k) \right) \left( e_l(k) \cdot \tilde{j}_p(k) \right).
\]

(135)
C. Renormalized energy shift

We are now in a position to calculate the renormalized energy shift

$$\Delta E_n^R = \langle \text{vac} | H_{\text{int}}^R | \text{vac} ; \psi_n \rangle,$$

(136)

where $|\psi_n\rangle$ is a (bound) eigenstate of the renormalized electronic Hamiltonian $H_{\text{int}}^R |\psi_n\rangle = E_n^R |\psi_n\rangle$ with eigenvalue $E_n^R$ given by the expectation value of (116). The first counterterm in $\langle \delta H_{\text{int}}(\Lambda) | \psi_n \rangle \equiv \langle \psi_n | \delta H_{\text{int}}(\Lambda) | \psi_n \rangle$ (cf. the first term in (131)) trivially cancels the static self-energy in $\Delta E_n^R$ and we are left with

$$\Delta E_n^R = -\frac{\hbar}{4\pi^2 c} \text{p.v.} \sum_m \int_{||k|| \leq \Lambda} dk \frac{||k||^{-1}}{E_{mn}^R + \hbar \omega_k} \sum_J |e_I(k) \cdot \tilde{j}_{P,mn}(k)|^2 + \frac{\hbar}{4\pi^2 c} \int_{||k|| \leq \Lambda} dk \frac{||k||^{-1}}{E_{\text{haw}}^R + \hbar \omega_k} \sum_J \left( \langle e_I(-k) \cdot \tilde{j}_P(k) \rangle \langle e_I(k) \cdot \tilde{j}_P(k) \rangle \right),$$

(137)

the second line being the second counterterm in (131). Introducing a resolution of the identity in $H_F$ with respect to the states $\{|\psi_n\rangle\}_m$, we can collect these terms together

$$\Delta E_n^R = -\frac{\hbar}{4\pi^2 c} \text{p.v.} \sum_m \int_{||k|| \leq \Lambda} dk \left( \frac{||k||^{-1}}{E_{mn}^R + \hbar \omega_k} - \frac{||k||^{-1}}{E_{\text{haw}}^R + \hbar \omega_k} \right) \sum_J |e_I(k) \cdot \tilde{j}_{P,mn}(k)|^2.$$  

(138)

To simplify the expression in the parentheses, we use the following identity, which we prove in Appendix F:

$$\sum_m \tilde{j}_{P,mn}(-k) \frac{1}{E_{mn}^R + \hbar \omega_k} \tilde{j}_{P,mn}(k) = \sum_m \left( \frac{\tilde{j}_{P,mn}(-k) \tilde{j}_{P,mn}(k)}{E_{\text{haw}}^R + \hbar \omega_k} - \frac{\tilde{j}_{P,mn}(-k) \langle [U^R, \tilde{j}_P(k)] \rangle_{mn}}{(E_{\text{haw}}^R + \hbar \omega_k)(E_{mn}^R + \hbar \omega_k)} \right).$$

(139)

The first term on the right of the above equality exactly cancels the second term in the parentheses of (138), and so the renormalized energy shift is

$$\Delta E_n^R = \frac{\hbar}{4\pi^2 c} \text{p.v.} \sum_m \int dk ||k||^{-1} \frac{\delta_{j,m}^R(k)}{(E_{\text{haw}}^R + \hbar \omega_k)(E_{mn}^R + \hbar \omega_k)} \left( \tilde{j}_{P,mn}(-k) \langle [U^R, \tilde{j}_P(k)] \rangle_{mn} \right).$$

(140)

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

Before returning to the multipole formalism to study contributions to $\Delta E_n^R$ order-by-order in the multipole expansions, we first quote the result that one obtains by directly calculating this expression in terms of the paramagnetic current density; the details can be found in Appendix G 1. Replacing $E_{mn}^R$ by a “reference” or average value $\bar{E}_R$ [12, 38], the result of this calculation is

$$\Delta E_n^R = -\frac{4\alpha \hbar^2}{3m^2 c^2} \sum_N \langle R | \tilde{d}_N \rangle^2 \ln \left( \frac{2mc^2}{\bar{E}_R} \right).$$

(141)

To compare with existing results, we consider the case of a single hydrogenic atom at $R$. We can thereby replace $q_N \langle \tilde{d}_N \rangle_R$ with $-e^2 Z |\phi_{n00}(R)|^2$, where $\phi_{n\ell m}(x)$ are the usual hydrogen wavefunctions. Moreover, using $|\phi_{n00}(R)|^2 = Z^3/n^3a_0^3$ we have [41, 49]

$$\Delta E_n^R = \frac{4\alpha Z^4}{3\pi m^3} \left( \frac{mc^2}{1 - \bar{E}_R/2mc^2} \right) \ln \left( \frac{2mc^2}{\bar{E}_R} \right).$$

(142)

For a hydrogen atom ($Z = 1$) we take the average excitation energy for the $2s$ level to be $\bar{E}_{2s} = 16.64$ Ryd, from which we find the energy shift to be $\Delta E_n^R \approx 1051$ MHz, which is in good agreement with the experimental value of $1054$ MHz [12, 39]. To make the electric dipole approximation, we replace the denominator with unity and thereby obtain the standard result [12, 38, 50]

$$\Delta E_n^R = \frac{4\alpha^5}{3\pi m^2 c^2} \ln \left( \frac{2mc^2}{\bar{E}_R} \right).$$  

(143)

D. Multipole expansion

A primary advantage of using the multipole formalism over minimal coupling is the ability to perform localized multipole expansions of the polarization and magnetization fields. While renormalization is better done with the minimal coupling form of $\Delta E_n^R$ as above, we now rewrite it in terms of these polarization and magnetization fields. As we discuss below, the multipole expansions are only valid for small $||k||$, and in particular for $||k|| < \Lambda$. This means that we can drop the factor of $E_{\text{haw}}$ in the denom-
inator of (138), and use a modified form of the identity in the parentheses of (139), namely

\[
\frac{1}{E_{mn}^R + \hbar \omega_k} - \frac{1}{\hbar \omega_k} = -\frac{E_{mn}^{R}}{(\hbar \omega_k)(E_{mn}^R + \hbar \omega_k)}, \quad (144)
\]

\[
\Delta E_n^R = \frac{1}{4\pi^2} \sum_{m} \int \frac{d|k|}{2\pi} \sum_{l} \left[ \left( \frac{E_{mn}^{R}}{E_{mn}^R + \hbar \omega_k} \right)^2 e_{I}(k) \cdot \hat{p}_{mn}(k) \right]^2 + \frac{1}{2\pi^2} \sum_{m} \int \frac{d|k|}{2\pi} \sum_{l} s_l \text{Im} \left[ \left( e_{I}(-k) \cdot \hat{p}_{mn}(-k) \right) \left( e_{I}(k) \cdot \hat{m}_{P,mn}(k) \right) \right]. \quad (145)
\]

Then to rewrite \( \Delta E_n^R \) in terms of the polarization and magnetization fields, we use the identity (101) to write the renormalized energy shift as

\[
\Delta E_n^R = \frac{1}{4\pi^2} \sum_{m} \int \frac{d|k|}{2\pi} \sum_{l} \left[ \left( \frac{E_{mn}^{R}}{E_{mn}^R + \hbar \omega_k} \right)^2 e_{I}(k) \cdot \hat{p}_{mn}(k) \right]^2 + \frac{1}{2\pi^2} \sum_{m} \int \frac{d|k|}{2\pi} \sum_{l} s_l \text{Im} \left[ \left( e_{I}(-k) \cdot \hat{p}_{mn}(-k) \right) \left( e_{I}(k) \cdot \hat{m}_{P,mn}(k) \right) \right]. \quad (146)
\]

This is our general expression for the renormalized energy shift in the multipole formalism. As we discuss at the end of this section, making the electric dipole approximation for the ground state \( n = 0 \) of atomic hydrogen yields the usual Bethe result [38]. However, our result above holds for ground and excited states of a general localized charge-current distribution, and includes contributions from the full electric and magnetic multipole series. And it accounts for spatial variations in the electromagnetic field, which are usually neglected by replacing the polarization and magnetization fields with the electric and magnetic dipole moments. Of course, in dropping the factors of \( E_{\hbar \omega_k} \) above we have reintroduced the “Bethe log” divergences, but in any case the multipole expansions introduce stronger ultraviolet divergences that must be regularized, as discussed below.

To obtain the multipole expansions, we take as the defining curve for the “relators” \( s^I(x; y, R) \) and \( \alpha^I(x; y, R) \) a straight-line path

\[
z(\lambda) = R + \lambda(y - R) \quad (147)
\]

with unit parametrization \( \lambda \in [0, 1] \), in which case \[6\]

\[
s^I(x; y, R) = (y^I - R^I) \int_0^1 d\lambda \delta(x - R - \lambda(y - R)),
\]

\[
\alpha^I(x; y, R) = \varepsilon^{ijk}(y^k - R^k) \int_0^1 d\lambda \delta(x - R - \lambda(y - R)). \quad (148)
\]

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 electric and paramagnetic multipole expansions are then given by

\[
p^I(x) = \sum_{n=0}^{\infty} (-1)^n \mu^{i_1\ldots i_n} \partial^{i_1} \ldots \partial^{i_n} \delta(x - R),
\]

\[
m^I_p(x) = \sum_{n=0}^{\infty} (-1)^n \nu^{i_1\ldots i_n} \partial^{i_1} \ldots \partial^{i_n} \delta(x - R), \quad (149)
\]

where the \( n \)-th-order electric and paramagnetic multipole moment operators are

\[
\mu^{i_1\ldots i_n} = \frac{1}{n!} \int dr \left( r^{i_1} - R^{i_1} \right) \ldots \left( r^{i_n} - R^{i_n} \right) \rho(r),
\]

\[
\nu^{i_1\ldots i_n} = \frac{1}{n!} \int dr \left( r^{i_1} - R^{i_1} \right) \ldots \left( r^{i_n} - R^{i_n} \right) \varepsilon^{abh} (r^a - R^a) j^b_p(r). \quad (150)
\]

We can write (with \( k = ||k|| \))

\[
\Delta E_n^R = \frac{\hbar c}{\pi} \sum_{m} E_{mn} \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \sum_{a,b=0}^{\infty} (-i)^a (i)^b \left( \int_0^\infty \frac{dk}{E_{mn}^R + \hbar \omega_k} \right) \bigg( |\mu_{mn}^{i_1\ldots i_a i_1^1\ldots i_{i_1^1}j_1}\nu_{mn}^{j_1}\bigg)_1 + \left( r_{mn}^{i_1\ldots i_a i_1^1\ldots i_{i_1^1}j_1}\nu_{mn}^{j_1}\bigg)_2 \bigg) + 2 \text{Im} \left( |\mu_{mn}^{i_1\ldots i_a i_1^1\ldots i_{i_1^1}j_1}\nu_{mn}^{j_1}\bigg)_2 \right). \quad (151)
\]
Here we have defined the isotropic Cartesian tensors

\[ T^{i_1 \ldots i_n j_1 \ldots j_n}_{(1)} = \int \frac{d\Omega_k}{4\pi} (\delta^{i_1}_{i_2} - \hat{k}^{i_1} \hat{k}^{i_2}) \hat{k}^{j_1} \ldots \hat{k}^{j_n} \]

\[ T^{i_1 \ldots i_n j_1 \ldots j_n}_{(2)} = \int \frac{d\Omega_k}{4\pi} (\epsilon^{i_1 \nu_1}_{k p} \hat{k}^{\nu_1}) \hat{k}^{j_1} \ldots \hat{k}^{j_n} \]

(152)

The tensor \( T^{(1)} \) vanishes unless \( a + b = 2n \) for some \( n \in \mathbb{N} \), while the tensor \( T^{(2)} \) vanishes unless \( a + b = 2n + 1 \) for some \( n \in \mathbb{N} \). These conditions preclude a number of electric, magnetic, and magnetoelectric 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.

Unless \( a = 0 \) and \( b = 0 \) the integrals above diverge as \( \|k\| \to \infty \), even though the same expression written in terms of the current density was finite. This particular ultraviolet divergence is an artifact of our extrapolation of the Fourier transforms of the multipole expansions (149) to all values of \( \|k\| \). Indeed, for the straight-line paths considered above one can explicitly compute the Fourier transforms \( \tilde{p}(k) \) and \( \tilde{\mathbf{n}}_F(k) \), after which one finds that they vanish in the limit \( \|k\| \to \infty \) \cite{13}. But the Fourier transforms of the multipole expansions (149) clearly diverge in the ultraviolet limit for all \( n \in \mathbb{N} \), and so the resulting power series expansions in \( k \) must have a finite radius of convergence \( \|k\| \leq \Lambda' \) (not equal to the ultraviolet cutoff \( \Lambda = (\hbar/mc)^{-1} \) used in previous sections). The exact value of \( \Lambda' \) depends on the charge-current distribution, and it is therefore undesirable to regulate the Fourier integrals with a hard cutoff. We instead make these integrals well-behaved by introducing a “heat kernel regulator” \cite{12, 39}, which in the continuum limit amounts to evaluating the integral

\[ \int_0^\infty dk \frac{k^{a+b}}{E_{mn}^R + \hbar \mathcal{E}_k} e^{-k/\Lambda'}, \]

(153)

where we take the limit \( \Lambda' \to \infty \) to remove the regulator at the end of the calculation. This integral is evaluated in Appendix G.2. After removing the regulator, we find the general expression

\[
\Delta E_n^R = \frac{1}{\hbar \pi c} \sum_m E_{mn}^R \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \ln \left( \frac{2mc^2}{|E_{mn}^R|} \right) \sum_{a,b=0}^\infty (-1)^{a+b} \left( \frac{E_{mn}^R}{\hbar c} \right)^{a+b} \left[ \left( \frac{\mu_{mn}^1 \ldots \mu_{mn}^j \ldots \mu_{mn}^j \ldots \mu_{mn}^j}{\mu_{mn}^1 \ldots \mu_{mn}^j \ldots \mu_{mn}^j \ldots \mu_{mn}^j} \right) \right] + \left( \frac{\nu_{mn}^1 \ldots \nu_{mn}^j \ldots \nu_{mn}^j \ldots \nu_{mn}^j}{\nu_{mn}^1 \ldots \nu_{mn}^j \ldots \nu_{mn}^j \ldots \nu_{mn}^j} \right) + \frac{2\text{Im}}{5} \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \right],
\]

(154)

The first several contributions to \( \Delta E_n^R \) are given by

\[
\Delta E_n^R = \frac{2}{3\hbar \pi c} \sum_m E_{mn}^R \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \ln \left( \frac{2mc^2}{|E_{mn}^R|} \right) \left[ |\mu_{nm}|^2 + |\mu_{mn}|^2 + \frac{1}{10} \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \left( \frac{3\nu_{nm}^0 \nu_{mn}^0 - \nu_{nm}^0 \nu_{mn}^0}{\nu_{nm}^0 \nu_{mn}^0} \right) \right],
\]

(155)

where \( \mu^i \), \( q^{ij} \), and \( o^{ijk} \) are the electric dipole, quadrupole, and octopole moment operators, \( \nu^i \) is the magnetic dipole moment operator, and the ellipses denote the higher-order multipole contributions. To check the validity of this result, consider only the electric dipole term

\[
\Delta E_n^R = \frac{2}{3\hbar \pi c} \sum_m E_{mn}^R \left( \frac{E_{mn}^R}{\hbar c} \right)^2 \ln \left( \frac{2mc^2}{|E_{mn}^R|} \right) |\mu_{nm}|^2.
\]

(156)

To recover Bethe’s result in the electric dipole approximation, we first replace \( |E_{mn}^R| \) in the logarithm with an average value \( \bar{E}_{mn}^R \), in which case

\[
\Delta E_n^R = \frac{2\alpha}{3\pi} \frac{1}{m^2 c^2} \sum_m \left( \frac{E_{mn}^R}{\bar{E}_{mn}^R} \right)^3 |\mu_{nm}|^2.
\]

(157)

In a first-quantized treatment this is precisely Bethe’s result for the Lamb shift in the electric dipole approximation \cite{38}. To compare with our previous result (143), we use the identity \cite{12}

\[
(E_{mn}^R)^2 |\mu_{nm}|^2 = - E_{mn}^R |\mu_{nm}|^2 = \left( \frac{\hbar e}{m} \right)^2 |p_{nm}|^2,
\]

(158)
from which follows

\[ \Delta E^R_n = \frac{2 \alpha}{3 \pi} \frac{1}{m^2 c^2} \ln \left( \frac{2mc^2}{|E^R_n|} \right) \sum_m E^R_{mn} \| \mathbf{P}_{nm} \|^2. \]  

We recover the expression (143) by evaluating the sum over states \( |\psi_m\rangle \) using an expression analogous to (G2).

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 are encoded in microscopic polarization and magnetization field operators — defined through second-quantized scalar field operators — with moments that are identified with the multipole moments of the charge-current distribution. These field operators couple to the quantized electromagnetic field in the multipolar Hamiltonian, obtained from the minimal coupling Hamiltonian through a unitary transformation, often referred to as the PZW transformation. Our reformulation generalizes existing work [15, 28] in 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. Restricting oneself to the electric dipole approximation is not required.

Following a reformulation of multipolar electrodynamics from minimal coupling electrodynamics, our focus thereafter concerned the vacuum structure of the theory. Using Rayleigh-Schrödinger perturbation theory we computed the regularized shift \( \Delta E^A_n \) of the electronic energy levels in the electromagnetic vacuum state, described by the regularized interaction term \( H^A_{\text{int}} \). This energy shift depends explicitly on the ultraviolet cutoff \( \Lambda \), an artifact of our choice of a “hard cutoff” regulator. To remove this \( \Lambda \)-dependence, we renormalized the regularized Hamiltonian \( H^A \) at leading-order in the fine structure constant by addition of appropriate counterterms. We could then use the renormalized Hamiltonian that followed to compute the finite and observable correction \( \Delta E^R_n \) to the (renormalized) electronic energy levels \( E^R_n \).

We obtain a closed form expression for the renormalized energy shift \( \Delta E^R_n \) 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 [38] of the Lamb shift in the electric dipole approximation, and extensions thereof [41, 42] to include spatial variations in the electromagnetic field. However, our expression (141) is valid for more general assemblies of atoms and molecules (possibly with net charge), consisting of a sum of contributions centered on each ion 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 [51]. Because of how complicated this symmetry group may be, we would no longer expect the contributions to \( \Delta E^R_n \) 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 \( \Delta E^R_n \) is an interesting direction for future work.

We then rewrote the renormalized energy shift in terms of the polarization and magnetization fields, permitting us to expand \( \Delta E^R_n \) in a sum of contributions coming from products of specific electric and magnetic multipole moments of the molecule. We derived the general form of such expressions, and then identified the contributions coming from the first several multipole moments, namely the electric dipole, quadrupole, and magnetic dipole moments. Existing treatments of the vacuum energy shift have focused on hydrogenic atoms, and therefore include only the electric dipole term. Ours appears to be the first explicit expression for the renormalized energy shift that includes the full sum of multipole contributions — they can now be investigated in detail.

This work lays the foundation for a broader research program involving microscopic polarization and magnetization fields in the quantum regime. Recent work indicates that a description of crystalline solids using multipolar quantum electrodynamics should be possible [52–56]: 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. The extension of this into the fully quantum regime, by generalizing the work presented here to crystal lattices, should provide a microscopic underpinning for the study of quantum optical effects in crystals.

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Appendix A: Transformations in Section II

1. Legendre transformation in minimal coupling

We begin with the minimal coupling Lagrangian \( L \) given in (10). The canonical momentum density con-
jugate to the electron field $\psi(x, t)$ is
\[
\pi_\psi(x, t) = \frac{\delta L}{\delta \dot{\psi}(x, t)} = \frac{i\hbar}{2} \dot{\psi}^\dagger(x, t),
\] (A1)
while the canonical momentum density conjugate to the vector potential $a(x, t)$ is
\[
\pi_a(x, t) = \frac{\delta L}{\delta \dot{a}(x, t)} = -\frac{1}{4\pi c} e(x, t).
\] (A2)

However, the canonical momentum density $\pi_\phi(x, t)$ conjugate to the scalar potential $\phi(x, t)$ vanishes. Thus the Lagrangian is degenerate, and the standard method for constructing the corresponding 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 [57]. The solution in the case of electrodynamics is well-known, and so we only summarize the main results [11, 58]. There are two constraints: The vanishing of $\pi_\phi(x, t)$, and Gauss’s law, $\nabla \cdot e(x, t) = 4\pi \rho^F(x, t)$. After fixing the transverse gauge
\[
\nabla \cdot a(x, t) = 0,
\] (A3)
the scalar potential is obtained from the electronic charge density through Poisson’s equation
\[
\nabla^2 \phi(x, t) = 4\pi \rho^F(x, t),
\] (A4)
and the canonical variable $\pi_\phi(x, t)$ can be discarded. The gauge-fixed Maxwell Lagrangian $L_B'$ is
\[
L_B' = \frac{1}{8\pi} \int dx \left( \frac{1}{c^2} \|a(x, t)\|^2 - \|\nabla \times a(x, t)\|^2 \right),
\] (A5)
while the gauge-fixed interaction term is
\[
L_{\text{int}}' = L_{\text{int}} + \frac{1}{2} \int dx \rho^F(x, t) \phi(x, t).
\] (A6)
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 (20) for the electromagnetic field operators and the equal-time anticommutation relations (22) for the electron field operators. Then the minimal coupling Hamiltonian (12) follows.

2. The PZW transformation

We begin with the minimal coupling Hamiltonian (12), which can be written as a functional of the canonical variables
\[
H = \mathcal{F}[a, \pi_a, \psi, \pi_\psi],
\] (A7)
where $\pi_a(x)$ and $\pi_\psi(x)$ are the canonical momentum densities defined in (A2) and (A1), respectively. Under the unitary transformation $U = \exp(iS/\hbar)$, the new canonical variables (indicated by a breve) are
\[
\breve{a}(x) = a(x)U, \quad \breve{\pi}_a(x) = \pi_a(x)U^\dagger,
\]
\[
\breve{\psi}(x) = \psi(x)U^\dagger, \quad \breve{\pi}_\psi(x) = \pi_\psi(x)U^\dagger.
\] (A8)

The strategy then [6] is to write the Hamiltonian as a functional of the transformed fields,
\[
H = \mathcal{G}[\breve{a}, \breve{\pi}_a, \breve{\psi}, \breve{\pi}_\psi].
\] (A9)
Equating (A7) and (A9) and using the general form of the transformations (A8), the new functional $\mathcal{G}$ is obtained from the old one $\mathcal{F}$ through
\[
\mathcal{G}[a, \pi_a, \psi, \pi_\psi] = U^\dagger \mathcal{F}[a, \pi_a, \psi, \pi_\psi] U,
\] (A10)
where on both sides we use the non-transformed fields. Once the new functional $\mathcal{G}$ 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
\[
\breve{a}(x) = a(x),
\]
\[
\breve{\pi}_a(x) = \pi_a(x) - \frac{1}{e} F_T(x),
\]
\[
\breve{\psi}(x) = e^{-i\phi(x, R)} \psi(x),
\]
\[
\breve{\pi}_\psi(x) = e^{+i\phi(x, R)} \pi_\psi(x),
\] (A11)
where
\[
\Phi(x, R) = \frac{e}{\hbar c} \int dw s^t(w; x, R) a^t(w).
\] (A12)

In semiclassical theory the quantity $\Phi(x, R)$ is a generalized Peierls phase [53]; in the fully quantum theory, the exponential of this quantity in (A11) is the Wilson line operator of Abelian gauge theory [13]. The original gauge freedom involving the scalar and vector potentials has been replaced [13] by a freedom in choosing the relators ((31) and (42)); here this involves a choice of the paths $C(y, R)$, although other classes of relators are possible [59].

With the canonical momentum density conjugate to the vector potential given in (A2), the transformed transverse electric field is $\breve{e}_T(x) = e_T(x) + 4\pi \breve{p}_T(x)$, which we identify with the transverse part of the displacement field $d_T(x)$, while the magnetic field is unchanged, $\breve{b}(x) = b(x)$. Then, after dropping the breve accent on the transformed electron field operators, the multipolar Hamiltonian (32) follows.
Appendix B: Electrostatic interactions in the multipolar Hamiltonian

We demonstrate the equivalence between the regularized form of (34) and (68). To begin, we use [12]

\[
\delta_{ij}^\Lambda(x - x') = \int \frac{dk}{(2\pi)^3} \hat{k}^i \hat{k}^j e^{i k \cdot (x - x')} = -\frac{1}{2\pi^2} \int_0^\infty dk \partial_i \partial_j F(x, x'; k),
\]

(B1)

where \(F(x, x'; k)\) is given by (93). After regularizing the Fourier integrals in the representation (B1) of the longitudinal delta functions, the second term on the first line of (34) is

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

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

(B2)

Meanwhile, using Gauss's law \( \nabla \cdot d_L(x) = 4\pi \rho_F(x) \), a straightforward calculation leads to

\[- \int dx d_L(x) \cdot \left( p(x) - \frac{1}{8\pi} d_L(x) \right) = -\frac{Q^2}{\pi} \int_0^\Lambda dk + \frac{2Q}{\pi} \int_0^\Lambda dk \int dx \rho(x) F(x, R; k), \]

(B3)

and therefore

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

(B4)

Next, using \( \rho(x) = \rho^c(x) + \rho^{\text{ion}}(x) \),

\[ \frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \rho(x') F(x, x'; k) \rho(x) = \frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \rho^c(x') F(x, x'; k) \rho^c(x) + \frac{2}{\pi} \sum_N q_N \int_0^\Lambda dk \int dx F(x, R + d_N; k) \rho^c(x) + \sum_{N,M} \frac{q_N q_M}{\pi} \int_0^\Lambda dk F(d_N, d_M; k), \]

(B5)

and so, isolating the divergences in \( E^\Lambda_{0,F} \), we have

\[ \frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \rho(x') F(x, x'; k) \rho(x) = \frac{1}{\pi} \int_0^\Lambda dk \int dx dx' \rho^c(x') F(x, x'; k) \rho^c(x) + \frac{2}{\pi} \sum_N q_N \int_0^\Lambda dk \int dx F(x, R + d_N; k) \rho^c(x) + \sum_{N\neq M} \frac{q_N q_M}{\pi} \int_0^\Lambda dk F(d_N, d_M; k) + E^\Lambda_{0,F}, \]

(B6)

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; the \( \Lambda \)-dependent term is given by

\[ E^\Lambda_{0,F} = \frac{1}{\pi} \left[ e^2 N_e + 2eN_e \sum_N q_N + \sum_N q_N^2 \right] \Lambda, \]

(B7)

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 \( E^\Lambda_{0,F} \), we can take the limit \( \Lambda \to \infty \) in the remaining terms and use

\[ \int_0^\infty dk F(x, x'; k) = \frac{\pi}{2} \frac{1}{\| x - x' \|}, \]

(B8)

which is valid for \( x \neq x' \). Dropping the ion-ion interaction as in Section II, we obtain the desired result (68).

Appendix C: First-order diamagnetic contribution

Here we simplify the diamagnetic contribution to \( \Delta E^\Lambda_{n(1)} \). The diamagnetic term is

\[ H_D \equiv -\frac{1}{2} \int dx m_D(x) \cdot b(x). \]

(C1)
As noted in the main text, we can rewrite \( H_D \) explicitly as (85); using (7) we write this as

\[
H_D = \frac{1}{2} \int dxdx' \, a^a(x) \tilde{O}^{ab}(x, x') a^b(x'),
\]

where

\[
\tilde{O}^{ab}(x, x') = \varepsilon^{api} \varepsilon^{bjq} \frac{\partial}{\partial x^p} \frac{\partial}{\partial x'^q} O^{ij}(x, x').
\]

and where \( O^{ij}(x, x') \), the second-quantized diamagnetization field, is defined in (86). With [53]

\[
\varepsilon^{abc} \frac{\partial}{\partial x^a} \alpha^{cd}(x; y, R) = -\frac{\partial}{\partial y^a} s^d(x; y, R) + \delta^{ad}(x - y)
\]

we have

\[
\tilde{O}^{ab}(x, x') = \frac{e}{2mc^2} \int dy \left( \delta^{ab} \delta(x - y) \delta(x' - y) - \delta^{ia} \delta(x - y) \frac{\partial}{\partial y^a} s^i(x'; y, R) - \delta^{ib} \delta(x' - y) \frac{\partial}{\partial y^a} s^i(x; y, R) + \left[ \frac{\partial}{\partial y^a} s^i(x; y, R) \left[ \frac{\partial}{\partial y^a} s^i(x'; y, R) \right] \right] \rho^c(y). \]

Substituting this expression back into (C2), the diamagnetic term is

\[
H_D = \frac{e}{2mc^2} \int dxd\rho^c(x) \| a(x) \|^2 - \frac{h}{mc} \int dxd\rho^c(x) a(x) \cdot \nabla \Phi(x, R) + \frac{\hbar^2}{2mc} \int dxd\rho^c(x) \| \nabla \Phi(x, R) \|^2. \quad (C6)
\]

Define

\[
\Delta E_D^\Lambda \equiv \langle \Psi_n | H_D^\Lambda | \Psi_n \rangle, \quad (C7)
\]

with \( |\Psi_n\rangle \) given by (79). The regularized mode expansion for the vector potential \( a(x) \) that leads to the mode expansion (58) for \( b(x) \) is

\[
a(x) = \sum_I \sum_{\|k\| \leq \Lambda} \left( \frac{2\pi \hbar c}{V \|k\|} \right)^{1/2} e^{ik_a x} a_I e^{i/x} + \text{h.c.} \quad (C8)
\]

A straightforward calculation of the vacuum expectation value of \( \|a(x)\|^2 \) leads to

\[
\frac{e}{2mc^2} \int dxd\rho^c_{nn}(x) \left( \| a(x) \|^2 \right)_{\text{vac}} = \frac{\alpha}{2\pi} \left( \frac{\hbar^2 N_c}{m} \right) \Lambda^2. \quad (C9)
\]

Focusing on latter two terms of (C6), the expectation value of the first is

\[
-\frac{\hbar}{mc} \int dxd\rho^c_{nn}(x) \left( a(x) \cdot \nabla \Phi(x, R) \right)_{\text{vac}}, \quad (C10)
\]

while the expectation value of the second is

\[
+\frac{\hbar^2}{2mc} \int dxd\rho^c_{nn}(x) \langle \| \nabla \Phi(x, R) \|^2 \rangle_{\text{vac}}. \quad (C11)
\]

With the regularized mode expansion for \( a(x) \), in the continuum limit we have

\[
\langle a^a(x) a^b(x') \rangle_{\text{vac}} = \frac{\hbar c}{4\pi^2} \int_{\|k\| \leq \Lambda} dk \|k\|^{-1} \delta^{ab}(k) e^{ik(x' - x)}. \quad (C12)
\]

Thus, the vacuum expectation value in (C10) is

\[
-\frac{\hbar c}{4\pi^2 mc} \int_{\|k\| \leq \Lambda} dk \|k\|^{-1} \delta^{ij}(k) \int dxdx' e^{ik(x' - x)} \rho^c_{nn}(x) \left[ \frac{\partial}{\partial x^i} s^j(x'; x, R) \right], \quad (C13)
\]

while the vacuum expectation value in (C11) is

\[
\frac{\hbar c}{8\pi^2 mc} \int_{\|k\| \leq \Lambda} dk \|k\|^{-1} \delta^{ij}(k) \int dxdx' e^{ik(x' - x)} \int dy \rho^c_{nn}(y) \left[ \frac{\partial}{\partial y^i} s^j(x; y, R) \right] \left[ \frac{\partial}{\partial y^k} s^l(x'; y, R) \right], \quad (C14)
\]
so that the expectation values of the latter two terms of (C6) are

\[ -\frac{\hbar}{mc} \int dx \rho_{nn}(x) \langle \alpha(x) \cdot \nabla \Phi(x, R) \rangle_{\text{vac}} + \frac{\hbar^2}{2mc} \int dx \rho_{nn}^2(x) \langle \nabla^2 \Phi(x, R) \rangle_{\text{vac}} = -\frac{\hbar^2}{4\pi^2 mc} \int d\mathbf{k} \frac{1}{|\mathbf{k}|^2} \delta_{ij}^2(\mathbf{k}). \]

In total, the diamagnetic contribution to the first-order correction is then (87).

### Appendix D: Second-order correction

#### 1. A useful identity

We first prove a useful identity concerning matrix elements of field operators. Let \( \mathcal{O}(x) \) denote a field operator in the Schrödinger picture which acts trivially on the Hilbert space \( \mathcal{H}_B \) of the Bose sector. Separating the free and interaction terms of the multipolar Hamiltonian as in (72), we define an interaction picture state by taking the time-dependence of the field operator in the interaction picture to be

\[ \mathcal{O}_I(x, t) = e^{iH_0 t/\hbar} \mathcal{O}(x) e^{-iH_0 t/\hbar}, \]

where

\[ H_0 = H_B \otimes 1_F + 1_B \otimes H_F \]

with \( H_B \) and \( H_F \) given by (33) and (34), while an interaction picture state \( |\Psi_n\rangle_I \) is related to the corresponding Schrödinger picture state \( |\Psi_n\rangle_S \) through

\[ |\Psi_n\rangle_I = e^{iH_0 t/\hbar} |\Psi_n\rangle_S. \]

The expression (D1) is equivalent to

\[ \frac{\partial \mathcal{O}_I(x, t)}{\partial t} = \frac{1}{\imath \hbar} \mathcal{O}_I(x, t), \]

which is used in the main text and below.

#### 2. Simplifications

Begin with the full expression for the second-order correction (98). We can simplify the second line using (12)

\[ \frac{1}{4\pi} \int d\mathbf{k} e^{i\mathbf{p} \cdot \mathbf{x}} \langle \mathcal{O}(x) \rangle_{\text{vac}} = \frac{i}{|\mathbf{k}|} e^{i\mathbf{p} \cdot \mathbf{x}} \nabla \cdot \mathbf{F}(x, x'; k), \]

where \( F(x, x'; k) \) is given by (93). Together with the identity (91), we have

\[ \Delta E_{\text{n(2)}} = -\frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dk \frac{\hbar c \kappa E_{mn} + \hbar c k}{E_{mn}} \int dxdx' \tau^{ij} (k || x' - x ||) \left( \sum_{ij} (p_{mn}^i(x') p_{mn}^j(x) + m_{P,mn}^i(x') m_{P,mn}^j(x)) \right) \]

\[ -\frac{i}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dk \frac{\hbar c k^2 E_{mn} + \hbar c k}{E_{mn}} \int dxdx' \sigma^{ij} (k || x' - x ||) \left( \sum_{ij} (p_{mn}^i(x') m_{P,mn}^j(x) + m_{P,mn}^i(x') p_{mn}^j(x)) \right). \]
To simplify $\Delta E_{n(n)}^A$, we break up its summands and process them in turn. We denote by $\Delta E_{pp}^A$ the first term involving the product of polarization fields, by $\Delta E_{nm}^A$ the second term involving the product of the magnetization fields, and by $\Delta E_{pm}^A$ the second line above. Using twice the algebraic identity

$$\frac{1}{E_{mn} + \hbar k c} = \frac{1}{\hbar k c} - \frac{1}{\hbar k E_{mn} + \hbar k c}.$$  

we have

$$\Delta E_{pp}^A = -\frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dk \int dxdx' \tau^{ij}(k \| x' - x \|) \rho^i_{p,m}(x') \rho^j_{p,m}(x)$$

$$+ \frac{1}{\hbar \pi c} \sum_m \int_0^\Lambda dkk^{-1} E_{mn} \int dxdx' \tau^{ij}(k \| x' - x \|) \rho^i_{p,m}(x') \rho^j_{p,m}(x)$$

$$- \frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dk \frac{\hbar k}{E_{mn} + \hbar k c} \int dxdx' \tau^{ij}(k \| x' - x \|) \rho^i_{p,m}(x') \rho^j_{p,m}(x)$$

$$+ \frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dkk^{-1} \left( \frac{E_{mn}}{E_{mn} + \hbar k c} \right)^2 \int dxdx' \tau^{ij}(k \| x' - x \|) \rho^i_{p,m}(x') \rho^j_{p,m}(x).$$

(D13)

Using our identity (D9) this can be written as

$$\Delta E_{pp}^A = -\frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dk \int dxdx' \tau^{ij}(k \| x' - x \|) \rho^i_{p,m}(x') \rho^j_{p,m}(x)$$

$$+ \frac{1}{\hbar \pi c} \sum_m \int_0^\Lambda dkk^{-1} \int dxdx' \tau^{ij}(k \| x' - x \|) E_{mn} \rho^i_{p,m}(x') \rho^j_{p,m}(x)$$

$$- \frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dk \frac{\hbar k}{E_{mn} + \hbar k c} \int dxdx' \tau^{ij}(k \| x' - x \|) m^i_{p,m,n}(x') m^j_{p,m,n}(x)$$

$$- \frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dkk^{-1} \int dxdx' \tau^{ij}(k \| x' - x \|) j^i_{p,m,n}(x') j^j_{p,m,n}(x)$$

$$+ \frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda dkk^{-1} \int dxdx' \tau^{ij}(k \| x' - x \|) \rho^i_{p,m}(x') \rho^j_{p,m}(x) + m^i_{p,m,n}(x') m^j_{p,m,n}(x).$$

(D14)

We can combine the third line above with the contribution

$$\Delta E_{nm}^A = -\frac{1}{\pi} \text{p.v.} \sum_m \int_0^\Lambda \frac{\hbar k c}{E_{mn} + \hbar k c} \int dxdx' \tau^{ij}(k \| x' - x \|) m^i_{n,m}(x') m^j_{n,m}(x).$$

(D15)

Consider the last line of (D11). Using the algebraic identity (D12) along with the identity (D9), that term can be written

$$\Delta E_{pm}^A = -\frac{i}{\pi} \text{p.v.} \sum_m \int_0^\Lambda \frac{\hbar k^2}{E_{mn} + \hbar k c} \int dxdx' \sigma^{ij}(k \| x' - x \|) \left( \rho^i_{p,m}(x') m^j_{p,m}(x) + m^i_{p,m}(x') \rho^j_{p,m}(x) \right)$$

$$= -\frac{i}{\pi} \int_0^\Lambda dkk \int dxdx' \sigma^{ij}(k \| x' - x \|) \left( \rho^i(x') m^j_{p}(x) - m^i_{p}(x') \rho^j(x) \right)$$

$$+ \frac{2}{\pi} \text{p.v.} \sum_m \int_0^\Lambda \frac{\hbar k c}{E_{mn} + \hbar k c} \int dxdx' \tau^{ij}(k \| x' - x \|) m^i_{p,m,n}(x') m^j_{p,m,n}(x)$$

$$- \frac{\hbar}{\pi} \text{p.v.} \sum_m \int_0^\Lambda \frac{k}{E_{mn} + \hbar k c} \int dxdx' \sigma^{ij}(k \| x' - x \|) \left( j^i_{p,m,n}(x') m^j_{p,m,n}(x) + m^i_{p,m,n}(x') j^j_{p,m,n}(x) \right).$$

(D16)
Then, through a trivial relabelling,

\[ \Delta E_{\text{pm}}^A = -\frac{i}{\pi} \int_0^\Lambda dk \int dk' \int dx dx' \sigma^i(k||x' - x||) \left\langle \left[ m^k_P(x'), p^i(x) \right]_n \right\rangle_n \]

\[ + \frac{2}{\pi} \sum_m \int_0^\Lambda \frac{dk}{E_{mn} + \hbar k} \int dx dx' \tau^i(j(k||x' - x||)m^k_{P,nn}(x')m^j_{P,nn}(x)) \]

\[ - \frac{\hbar}{\pi} \sum_m \int_0^\Lambda \frac{dk}{E_{mn} + \hbar k} \int dx dx' \sigma^i(k||x' - x||) \left( j^k_{P,nn}(x')m^j_{P,nn}(x) + m^i_{P,nn}(x')j^j_{P,nn}(x) \right), \] (D17)

With the expressions (D14) for \( \Delta E_{\text{pp}}^A \), (D15) for \( \Delta E_{\text{mm}}^A \), and (D17) for \( \Delta E_{\text{pm}}^A \), the total second-order correction is

\[ \Delta E^A_{(2n)} = -\frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dx dx' \tau^i(k||x' - x||)p^i_{mn}(x')p^j_{mn}(x) \]

\[ + \frac{1}{\hbar c} \sum_m \int_0^\Lambda dk \int dx dx' \tau^i(k||x' - x||)E_{mn}p^i_{mn}(x')p^j_{mn}(x) \]

\[ - \frac{\hbar}{\pi} \sum_m \int_0^\Lambda \frac{dk}{E_{mn} + \hbar k} \int dx dx' \tau^i(k||x' - x||)j^i_{P,nn}(x')j^j_{P,nn}(x) \]

\[ - \frac{i}{\pi} \int_0^\Lambda dk \int dx dx' \sigma^i(k||x' - x||) \left\langle \left[ m^k_P(x'), p^i(x) \right]_n \right\rangle_n. \] (D18)

Using that the curl of a gradient vanishes, through some index manipulations one can show that

\[ (-\delta^i_j \partial^2 + \partial^i \partial^j) \varepsilon^{jpk} \partial^p F(x, x'; k) \left\langle \left[ m^k_P(x'), p^i(x) \right]_n \right\rangle_n = k^2 \varepsilon^{jpk} \partial^p F(x, x'; k) \left\langle \left[ m^k_P(x'), p^i(x) \right]_n \right\rangle_n \] (D19)

and

\[ \sum_m E_{mn}p^i_{mn}(x')p^j_{mn}(x) = \frac{i\hbar}{2} \left\langle \left[ j^i_{P,nn}(x'), p^j(x) \right]_n - c \varepsilon^{jpk} \partial^p \left[ m^k_P(x'), p^j(x) \right]_n \right\rangle_n, \] (D20)

we can collect the second and fourth lines together. Restoring the angular parts of the Fourier integrals, the second-order correction is

\[ \Delta E^A_{(2n)} = \frac{i}{8\pi^2 c} \sum_m \int_{||k||\leq \Lambda} dk \frac{||k||^{-1}}{E_{mn} + \hbar \omega_k} \int dx dx' \delta_{ij}^k(k) e^{ik(x' - x)} \left\langle \left[ j^i_{P,nn}(x'), p^j(x) \right]_n + c \varepsilon^{jpk} \partial^p \left[ m^k_P(x'), p^j(x) \right]_n \right\rangle_n \]

\[ - \frac{\hbar}{4\pi^2 c} \sum_m \int_{||k||\leq \Lambda} dk \frac{||k||^{-1}}{E_{mn} + \hbar \omega_k} \int dx dx' \tau^i_j(k) j^i_{P,nn}(x')j^j_{P,nn}(x) \delta_{ij}^k(k) e^{ik(x' - x)} \]

\[ - \frac{1}{\pi} \sum_m \int_0^\Lambda dk \int dx dx' \tau^i_j(k||x' - x||)p^i_{mn}(x')p^j_{mn}(x). \] (D21)

Working out the commutators in the first line, we find

\[ \frac{1}{2} \left\langle \left[ j^i_{P,nn}(x'), p^j(x) \right]_n + c \varepsilon^{jpk} \partial^p \left[ m^k_P(x'), p^j(x) \right]_n \right\rangle_n = \frac{\hbar e}{m_i} \left( \left[ \frac{\partial}{\partial x^i} s^j(x; x', R) \right] \right) \rho_{nn}(x) \]

\[ - \frac{1}{2} \int dy \rho_{nn}(y) \left( \frac{\partial}{\partial y^i} s^j(x; y, R) \right) \left[ \frac{\partial}{\partial y^j} s^i(x; y, R) \right], \] (D22)

and so, comparing to (C15), the second-order correction is (102).

**Appendix E: Energy shift in minimal coupling**

For comparison with the regularized energy shift (104) computed with the multipolar Hamiltonian, we work out the same energy shift in minimal coupling. The minimal coupling Hamiltonian is given in the Schrödinger picture.
by equations (12-15). Split up the Hamiltonian into free and interaction terms

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

(E1)

where the free term is

\[ H_0 = H_B \otimes 1_F + \mathbb{1}_B \otimes H_F \]  

(E2)

and the interaction term is

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

(E3)

The paramagnetic and diamagnetic current densities are defined in (16) and (17), respectively. As in Section IV, we split up the interaction term

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

(E4)

where now

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

(E5)

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

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

(E6)

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

\[ \Delta E_{n(1)}^\Lambda = \frac{e^2}{2mc^2} \int dx \, \langle \Psi_n | \dot{\psi}^\dagger(x) \| a(x) \|^2 \psi(x) | \Psi_n \rangle, \]  

(E7)

with the initial state (79). In terms of the mode expansion (C8) for the vector potential,

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

(E8)

To obtain the second-order correction, begin with

\[ \langle \Psi_\delta | H_{\text{int}(2)} | \Psi_n \rangle = -\frac{1}{c} \int dx \, \langle \Psi_\delta | j_F(x) \cdot a(x) | \Psi_n \rangle. \]  

(E9)

Again using the mode expansion (C8), we have

\[ \left| \langle \Psi_\delta | H_{\text{int}(2)} | \Psi_n \rangle \right|^2 = \left( \frac{2\pi\hbar}{V\omega_k} \right) \left| e_{jF}(k) \cdot \tilde{j}_{P,nm}(k) \right|^2, \]  

(E10)

where \( \tilde{j}_{P,nm}(x) \equiv (j_{p}(x))_{nm} \). Introducing the sums (97) and taking the continuum limit, the regularized second-order correction is

\[ \Delta E_{n(2)}^\Lambda = -\frac{\hbar}{4\pi^2 c} \mathcal{P.V.} \sum_m \int_{\|k\| \leq \Lambda} dk \, \frac{\|k\|^{-1}}{E_{mn} + \hbar\omega_k} \sum_l \left| e_{jF}(k) \cdot \tilde{j}_{P,nm}(k) \right|^2. \]  

(E11)

Collecting the first-order (E8) and second-order (E11) corrections together, the regularized energy shift at \( O(\alpha) \) in minimal coupling is exactly the result (104).

**Appendix F: Proof of identity (139)**

We prove the identity (139). To begin, consider the operator expression

\[ \frac{1}{H_F - E_F^R + \hbar\omega_k} = \frac{1}{H_F^{(0)} - E_F^R + \hbar\omega_k} - \frac{1}{H_F^{(0)} - E_F^R + \hbar\omega_k} \frac{U_F^{(R)}}{H_F^{(0)} - E_F^R + \hbar\omega_k}, \]  

(F1)

from which follows

\[ \frac{1}{H_F - E_F^R + \hbar\omega_k} \gamma^\dagger_{jF}(k) = \frac{1}{H_F^{(0)} - E_F^R + \hbar\omega_k} \gamma^\dagger_{jF}(k) - \frac{1}{H_F^{(0)} - E_F^R + \hbar\omega_k} \frac{U_F^{(R)}}{H_F^{(0)} - E_F^R + \hbar\omega_k} \gamma^\dagger_{jF}(k). \]  

(F2)

To simplify, we will use the identity (127). Introducing a convergence factor “+i\( \delta \)” (which will be removed below), we use an identity analogous to (125), namely

\[ \frac{1}{H_F^{(0)} - E_F^R + \hbar\omega_k + i\delta} \gamma^\dagger_{jF}(k) = -i \int_0^\infty ds \, e^{i \omega_k + H_F^{(0)}} e^{i \delta} \gamma^\dagger_{jF}(k) \]  

\[ = -i \int_0^\infty ds \, e^{i \omega_k + E_F^R + i\delta} e^{i H_F^{(0)}} e^{i \delta} \gamma^\dagger_{jF}(k) \]  

\[ = \gamma^\dagger_{jF}(k) \frac{1}{H_F^{(0)} - E_F^R + \hbar\omega_k + i\delta}. \]  

(F3)
where we have dropped the \( \|q\| \ll mc \) terms as before. We can trivially take the limit \( \delta \to 0^+ \), since upon forming matrix elements there will never be poles in this denominator. Therefore we have

\[
\frac{1}{H_F^R - E_n^R + \hbar \omega_k} \tilde{\gamma}_p^j(k) = \frac{\tilde{\gamma}_p^j(k)}{H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k} \frac{1}{H_F^R - E_n^R + \hbar \omega_k} - \frac{1}{H_F^R - E_n^R + \hbar \omega_k} \left[ U^R \tilde{\gamma}_p^j(k) \right] - \frac{1}{H_F^R - E_n^R + \hbar \omega_k} U^R \frac{1}{H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k}.
\]  

(F4)

and so bringing the last term to the left side

\[
\frac{1}{H_F^R - E_n^R + \hbar \omega_k} \tilde{\gamma}_p^j(k) \left( H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k \right) = \frac{\tilde{\gamma}_p^j(k)}{H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k} \frac{1}{H_F^R - E_n^R + \hbar \omega_k} - \frac{1}{H_F^R - E_n^R + \hbar \omega_k} \left[ U^R \tilde{\gamma}_p^j(k) \right] - \frac{1}{H_F^R - E_n^R + \hbar \omega_k} U^R \frac{1}{H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k}.
\]  

(F5)

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

\[
\frac{1}{H_F^R - E_n^R + \hbar \omega_k} \tilde{\gamma}_p^j(k) = \frac{\tilde{\gamma}_p^j(k)}{H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k} \frac{1}{H_F^R - E_n^R + \hbar \omega_k} - \frac{1}{H_F^R - E_n^R + \hbar \omega_k} \left[ U^R \tilde{\gamma}_p^j(k) \right] - \frac{1}{H_F^R - E_n^R + \hbar \omega_k} U^R \frac{1}{H_F^R - E_n^R + E_{\omega_k} + \hbar \omega_k}.
\]  

(F6)

Taking matrix elements in the states \( \psi_{n,m} \) and using that \( H_F^R |\psi_{n,m}\rangle = E_{n,m}^R |\psi_{n,m}\rangle \), the desired result (139) follows.

Appendix G: Computations in Section V

1. Direct calculation of \( \Delta E_n^R \)

To begin, we replace \( E_{nm}^R \) in the denominator of (140) by a “reference” or average value \( E_{nm}^R \to E_n^R [12, 38] \). Through a short calculation we have

\[
\langle [U^R, \tilde{\gamma}_p^j(x)] \rangle_{nm} = \frac{\hbar e}{mc} \left\langle \psi^\dagger(x) \partial^j \phi(x) \psi(x) \right\rangle_{nm} + \frac{\hbar e}{mc} \left\langle \psi^\dagger(x) \partial^j V(x) \psi(x) \right\rangle_{nm}.
\]  

(G1)

Since we have replaced \( E_{nm}^R \) in the denominator of (140) by the reference value \( E_n^R \), we can evaluate the sum using the identity [12]

\[
\sum_m \tilde{\gamma}_p^j_{nm}(x) \langle [U^R, \tilde{\gamma}_p^j(x')] \rangle_{nm} = \frac{1}{2} \left\langle [\tilde{\gamma}_p^j(x), U^R] \right\rangle_{n}. \tag{G2}
\]

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

\[
\frac{1}{4\pi} \int d\Omega_k \delta^j_k(k) \sum m \tilde{\gamma}_p^j_{nm}(k) \langle [U^R, \tilde{\gamma}_p^j(k)] \rangle_{nm}.
\]  

(G3)

The angular integral yields \( (2/3) \delta_{ij} \), and after a lengthy calculation we find

\[
\frac{1}{4\pi} \int d\Omega_k \delta^j_k(k) \sum m \tilde{\gamma}_p^j_{nm}(k) \langle [U^R, \tilde{\gamma}_p^j(k)] \rangle_{nm} = -\frac{\hbar^2 e^2}{3mc} \int dx \left\langle \psi^\dagger(x) \nabla^2 V(x) \psi(x) \right\rangle_n.
\]  

(G4)

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

\[
\frac{1}{4\pi} \int d\Omega_k \delta^j_k(k) \sum m \tilde{\gamma}_p^j_{nm}(k) \langle [U^R, \tilde{\gamma}_p^j(k)] \rangle_{nm} = -\frac{4\pi \hbar^2 e^2}{3mc} \sum_N q_N \langle \phi(r) (R + d_N) \rangle_n,
\]  

(G5)
where \( \mathbf{R} + \mathbf{d}_N \) 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

\[
\Delta E_n^R = -\frac{4\alpha \hbar^3}{3\mc} \sum_N q_N \langle \rho^x(\mathbf{R} + \mathbf{d}_N) \rangle_n \times \int_0^\infty dk \, \frac{1}{\hbar k/2\mc + 1} \left( \frac{1}{E_n^R + \hbar k} \right). \quad (G6)
\]

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

\[
\int_0^\infty dk \, \frac{1}{\hbar k/2\mc + 1} \left( \frac{1}{E_n^R + \hbar k} \right) = \left( \frac{1}{\hbar c} \right)^2 \int_0^\infty dx \, \frac{1}{(x + 1)(x + \beta_n^R)}. \quad (G7)
\]

Consider the contour integral

\[
\oint \frac{dz}{z} \frac{\log(z)}{(z + 1)(z + \beta_n^R)} \quad (G8)
\]

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 [60]. The other two integrals yield the identity

\[
\int_0^\infty dx \, \frac{1}{(x + 1)(x + \beta_n^R)} = \frac{1}{1 - E_n^R/2\mc} \ln \left( \frac{2mc^2}{|E_n^R|} \right). \quad (G10)
\]

and the result follows.

**2. Calculation of the integral (153)**

We calculate the integral

\[
\int_0^\infty dk \, \frac{k^{a+b}}{E_{mn}^R + \hbar k} e^{-k/\Lambda'} \quad (G11)
\]

using techniques from complex analysis. Introducing a change of variables \( x = \hbar k/2\mc \), we have

\[
\left( \frac{1}{\hbar c} \right)^2 \left( \frac{2mc}{\hbar} \right)^{a+b} \int_0^\infty dx \, \frac{x^{a+b} e^{-x/\Lambda'}}{x + \beta_{mn}^R} \quad (G12)
\]

where \( \beta_{mn}^R = E_{mn}^R/2\mc \). To evaluate this integral, consider the related contour integral

\[
\oint \frac{dz}{z} \frac{z^{a+b} e^{-|z|/\Lambda'}}{z + \beta_{mn}^R} \log(z) \quad (G13)
\]

where \( \mathcal{C} \) is the standard “keyhole” contour. The integrals over the circular arcs \( z = Re^{i\phi} \) and \( z = \epsilon e^{i\phi} \) vanish in the respective limits \( R \to \infty \) and \( \epsilon \to 0^+ \), by the “ML estimate” for complex integration [60]. Meanwhile, the remaining contour integrals, with the contours parametrized by \( z = x \pm i\epsilon \), can be combined together in the limit \( \epsilon \to 0^+ \) and thereby yield the identity

\[
\int_0^\infty dx \, \frac{x^{a+b} e^{-x/\Lambda'}}{x + \beta_{mn}^R} = -\frac{1}{2\pi i} \oint \frac{dz}{z} \frac{z^{a+b} e^{-|z|/\Lambda'}}{z + \beta_{mn}^R} \log(z). \quad (G14)
\]

We evaluate the integral on the right-hand-side using the calculus of residues: The pole is located at \( z = -\beta_{mn}^R \) in the complex plane, and with the identity above we end up with

\[
\int_0^\infty dx \, \frac{x^{a+b} e^{-x/\Lambda'}}{x + \beta_{mn}^R} = -(\beta_{mn}^R)^{a+b} e^{-|\beta_{mn}^R|/\Lambda'} \ln(|\beta_{mn}^R|), \quad (G15)
\]

and so the integral \( (G12) \) is

\[
\left( \frac{1}{\hbar c} \right)^2 (-1)^{a+b} \frac{E_{mn}^R}{\hbar c} \frac{a+b}{\Lambda'} \ln \left( \frac{2mc^2}{|E_{mn}^R|} \right), \quad (G16)
\]
where we have removed the regulator ($\Lambda \to \infty$). With this result the desired expression (154) for the renormalized energy shift in its multipolar form follows.