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Comparison of quantum and classical local-field effects on two-level atoms in a dielectric

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The macroscopic quantum theory of the electromagnetic field in a dielectric medium interacting with a dense collection of embedded two-level atoms fails to reproduce a result that is obtained from an application of the classical Lorentz local-field condition. Specifically, macroscopic quantum electrodynamics predicts that the Lorentz redshift of the resonance frequency of the atoms will be enhanced by a factor of the refractive index n of the host medium. However, an enhancement factor of $(n^2+2)/3$ is derived using the Bloembergen procedure in which the classical Lorentz local-field condition is applied to the optical Bloch equations. Both derivations are short and uncomplicated and are based on well-established physical theories, yet lead to contradictory results. Microscopic quantum electrodynamics confirms the classical local-field-based results. Then the application of macroscopic quantum electrodynamic theory to embedded atoms is proved false by a specific example in which both the correspondence principle and microscopic theory of quantum electrodynamics are violated.

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I. INTRODUCTION

The effect of a dielectric host medium on the spontaneous-emission rate of a two-level atom remains an interesting and challenging problem in quantum optics with an importance that befits a rigorous test of our understanding of the interaction of light with matter. The essential characteristic of the problem is its multiscale nature with the atom being a creature of microscopic quantum electrodynamics and the dielectric manifesting in the realm of classical continuum electrodynamics. In principle, it is possible to represent both the atom and dielectric microscopically, although, in practice, the spontaneous-emission rate is calculated using a macroscopic quantum electrodynamic theory [1–6] in which the continuous dielectric medium is incorporated into a medium-assisted electromagnetic field. While the dielectric renormalization of the spontaneous-emission rate of an embedded atom generates considerable interest, a mere handful of papers discuss the effect of a dielectric on the Lorentz redshift of the resonance frequency of two-level atoms. Knoester and Mukamel [7] used a Hopfield [8] polariton model of macroscopic quantum electrodynamics and found that the Lorentz redshift is enhanced by a factor of the refractive index n compared to the vacuum value. Crenshaw and Bowden [9] derived the enhancement factor $(n^2+2)/3$ using the Bloembergen [10] procedure in which the classical Lorentz local-field condition is applied to the optical Bloch equations. The two procedures also produce different values for the renormalization of the field (Rabi frequency) that drives the dynamics of atoms in a dielectric.

In this paper, we derive the generalized optical Bloch equations for a dense collection of two-level atoms in a dielectric host material using, first, the Bloembergen procedure based on the classical Lorentz local-field condition and, second, Ginzburg macroscopic quantum electrodynamics. The derivations are simple and direct and based on well-established physical theories, yet produce contradictory results for (i) the Lorentz redshift of the resonance frequency of two-level atoms densely embedded in the dielectric, and (ii) the renormalization of the field that drives the dynamics of the embedded atoms. The microscopic quantum electrodynamic procedure described by Crenshaw and Bowden [11] confirms the Bloembergen-based results. Then, we must conclude that the macroscopic quantum electrodynamic theory applied to dielectrically embedded atoms violates both the correspondence principle and microscopic quantum electrodynamics.

II. DIELECTRIC LOCAL-FIELD EFFECTS

In the view of Lorentz, classical continuum electrodynamics is better expressed in terms of an atomistic model of discrete particles embedded in the vacuum that interact with the microscopic electromagnetic field at the point of the particle [12,13]. The local field that acts on that particle,

$$\mathbf{E}_L = \mathbf{E} + \frac{4\pi}{3}\mathbf{P}, \tag{2.1}$$

is comprised of the macroscopic Maxwell field $\mathbf{E}$ and the reaction field of all other particles, expressed in terms of the macroscopic polarization $\mathbf{P}$. For a linearly polarizable material, the polarization $\mathbf{P}=pN_d\mathbf{E}_L$ is the product of the microscopic polarizability p, the dipole number density N_d, and the local field. Using the Lorentz local-field condition (2.1) to eliminate the microscopic local field produces the Clausius-Mossotti-Lorentz-Lorenz relation

$$\frac{4\pi}{3}pN_d = \frac{\varepsilon - 1}{\varepsilon + 2} \tag{2.2}$$

between the polarizability and the macroscopic dielectric constant $\varepsilon = 1 + 4\pi P/E$.

The local-field principle also applies to nonlinear media. Bloembergen [10] investigated nonlinear optics in the presence of a linear host medium and found that a local-field dielectric enhancement factor of

$$\ell = \frac{n^2+2}{3} \tag{2.3}$$

accompanies each appearance of a macroscopic field in the nonlinear susceptibility. In our notation, $\varepsilon = n^2$ for a dielectric and ℓ refers to the specific quantity in the preceding equation and not to any other constant or variable representing a local-field factor. Bowden and co-workers [14–16] predicted intrinsic optical bistability in a dense collection of vacuum-embedded two-level atoms due to an inversion-dependent local-field shift of the resonance frequency. Later work [9] reported the effect of embedding the dense two-level systems in a host dielectric. The dynamics of the two-level systems are described by the optical Bloch equations [17]. Building on Bloembergen's work [10], Bowden and Dowling [18] showed that the field that drives the atoms is the local field (2.1). With that substitution, one obtains the generalized optical Bloch equations [18],

$$\frac{\partial R_{21}}{\partial t} = i(\omega-\omega_0)R_{21} - \frac{i\mu}{2\hbar}\left(\mathcal{E} + \frac{4\pi}{3}\mathcal{P}\right)W - \gamma_\perp R_{21}, \tag{2.4a}$$

$$\frac{\partial W}{\partial t} = -\frac{i\mu}{\hbar}\left[\left(\mathcal{E}^* + \frac{4\pi}{3}\mathcal{P}^*\right)R_{21} - \left(\mathcal{E} + \frac{4\pi}{3}\mathcal{P}\right)R_{21}^*\right] - \gamma_\parallel (W - W_{\mathrm{eq}}). \tag{2.4b}$$

Here, fields are represented in the plane-wave limit by envelope functions such that $P = \frac{1}{2}(\mathcal{P}e^{-i\omega t} + \text{c.c.})$, $E = \frac{1}{2}(\mathcal{E}e^{-i\omega t} + \text{c.c.})$, and $E_L = \frac{1}{2}(\mathcal{E}_L e^{-i\omega t} + \text{c.c.})$. The macroscopic spatially averaged atomic variables in a rotating frame of reference are $R_{21} = \langle \rho_{21} e^{i\omega t}\rangle_{\mathrm{sp}}$, $R_{12} = \langle \rho_{12} e^{-i\omega t}\rangle_{\mathrm{sp}}$, and $W = R_{22} - R_{11} = \langle \rho_{22}\rangle_{\mathrm{sp}} - \langle \rho_{11}\rangle_{\mathrm{sp}}$, where $\langle \cdots \rangle_{\mathrm{sp}}$ corresponds to a spatial average over a volume of the order of a resonance wavelength cubed and the ρ_{ij} are the density-matrix elements for a two-level system with a lower state $|1\rangle$ and an upper state $|2\rangle$. Also, μ is the matrix element of the transition dipole moment, assumed real, $\gamma_\perp$ is a phenomenological dipole dephasing rate, $\gamma_\parallel$ is a phenomenological population relaxation rate, and W_{eq} is the population difference at equilibrium.

For a linearly polarizable material, the polarization is $\mathbf{P} = pN_d \mathbf{E}_L$. For atoms embedded in a linearly polarizable material, the polarization is the sum of the linear and nonlinear components. Substituting the local field (2.1) into the linear component, we have the polarization envelope

$$\mathcal{P} = pN_d\left(\mathcal{E} + \frac{4\pi}{3}\mathcal{P}\right) + 2N\mu R_{21},$$

where N is the number density of atoms. Collecting terms in $\mathcal{P}$ and using the Clausius-Mossotti-Lorentz-Lorenz relation (2.2) yields

$$\mathcal{P} = \frac{\varepsilon - 1}{4\pi}\mathcal{E} + \frac{\varepsilon + 2}{3} 2N\mu R_{21}. \tag{2.5}$$

Substituting the polarization envelope (2.5) into the generalized Bloch equations (2.4) produces

$$\frac{\partial R_{21}}{\partial t} = i\left(\omega - \omega_0 - \frac{4\pi}{3\hbar}N\mu^2 \ell W\right)R_{21} - \frac{i\mu}{2\hbar}\ell\mathcal{E}W - \gamma_\perp R_{21}, \tag{2.6a}$$

$$\frac{\partial W}{\partial t} = -\frac{i}{\hbar}[\mu\ell\mathcal{E}^* R_{21} - \mu\ell\mathcal{E}R_{21}^*] - \gamma_\parallel (W - W_{\mathrm{eq}}). \tag{2.6b}$$

For now, we assume that ℓ is real. Then, the local-field effect of the dielectric is simply an enhancement of the driving field $\mathcal{E}$, or Rabi frequency $\mu\mathcal{E}/\hbar$, and the inversion-dependent Lorentz redshift by ℓ [9]. The decay rates remain phenomenological.

The optical Bloch equations [17] are the basic semiclassical equations of motion for an isolated two-level system in the vacuum. The two-level system interacts with its environment through the local field. Using the classical Lorentz local-field condition shows the effects of the environment to be (i) a Bowden-Lorentz redshift of the resonance frequency by $(4\pi/3\hbar)N\mu^2 W$ due to nearby atoms, (ii) a Bloembergen enhancement of the field $\mathcal{E}$ by ℓ due to the linear host, and (iii) a Bloembergen-type enhancement of the Bowden-Lorentz redshift by ℓ due to the linear host. Further consideration of the generalized Bloch equations, derived using the classical Lorentz local-field condition, is suspended until quantum electrodynamic equations of motion have been derived. At that time, Eq. (2.6) will serve as the basis for quantum-classical correspondence.

III. MACROSCOPIC QUANTUM ELECTRODYNAMICS

Quantum electrodynamics can be viewed as the quantized version of Lorentzian electrodynamics in which discrete quantum particles interact with the vacuum field modes. When applied to dielectrics, the practice has been to create a macroscopic version of quantum electrodynamics along the lines of a quantized version of continuum electrodynamics. The macroscopic theory can be derived either by quantizing the classical Maxwell fields or by applying a continuum approximation to the microscopic quantum electrodynamic Hamiltonian. Ginzburg [1] pioneered the procedure of canonical quantization of the field in a dielectric and applied it to Cherenkov radiation. The macroscopic quantization procedure was limited to dielectrics with negligible dispersion and absorption. Jauch and Watson [2,19] continued Ginzburg's work and extended macroscopic quantization to dispersive dielectrics [5,20], while other researchers have treated absorption [21,22] and nonlinear dielectrics [23,24]. Knoester and Mukamel [7] and Huttner, Baumberg, and Barnett [25] start with the fundamental microscopic Hamiltonian and transform from coordinate space to wave-number space in the continuum approximation to derive the macroscopic Hopfield Hamiltonian [26]. Recent work [27–31] includes macroscopic quantization of fields in magnetodielectric media, including left-handed negative-index materials.

The spontaneous-emission rate of an atom in a dielectric is readily derived by the macroscopic quantum electrodynamic theory. In 1976, Nienhuis and Alkemade [6] used a macroscopic version of Fermi's golden rule to derive the dielectrically enhanced spontaneous-emission rate with the macroscopic Ginzburg fields. Huttner, Barnett, and Loudon [32], among others, have combined the macroscopic Fano-Hopfield [8,33] theory with Fermi's golden rule to derive the dielectric renormalization of the spontaneous-emission rate of an impurity atom, while other studies begin with macroscopic Green's functions [34] or auxiliary fields [35]. For the most part, the collective attention is focused on the dielectric renormalization of the spontaneous-emission rate. Knoester and Mukamel [7] obtained the dielectric effect on both the Lorentz redshift and the spontaneous decay rate by deriving operator equations of motion from the Hopfield model. In this section, we derive equations of motion using Weisskopf-Wigner theory applied to the macroscopic Hamiltonian in terms of the Ginzburg field operators [36]. A term-by-term comparison with the generalized Bloch equations that were derived in the preceding section under the Lorentz local-field condition exposes an extraordinary degree of disagreement between two known and accepted treatments of the effect of a dielectric host on the electrodynamics of two-level atoms.

The principal product of the macroscopic quantization theory is the medium-assisted field operator

$$\bar{\mathbf{E}} = \frac{i\hbar}{n}\sum_{l\lambda}\sqrt{\frac{2\pi\omega_l}{\hbar V}}(\bar{a}_l e^{i\mathbf{k}_l\cdot\mathbf{r}} - \text{H.c.})\hat{\mathbf{e}}_{\mathbf{k}_l\lambda}, \tag{3.1}$$

where $\bar{a}_l^\dagger$ and $\bar{a}_l$ are the macroscopic creation and destruction operators for the field modes and ω_l is the frequency of the field in the mode l. Also, V is the quantization volume, $\hat{\mathbf{e}}_{\mathbf{k}_l\lambda}$ is a unit vector in the direction of the polarization, and λ denotes the state of polarization. The spontaneous-emission rate of an impurity atom in a dielectric can then be obtained by applying Fermi's golden rule,

$$\Gamma = \frac{2\pi}{\hbar}|\langle f|H_{\text{int}}|i\rangle|^2 D, \tag{3.2}$$

to the effective interaction Hamiltonian $H_{\text{int}} = -\mu_a\cdot\bar{\mathbf{E}}$, where i labels the initial state and f denotes all available final states. As is typically calculated, the dielectric renormalization of the vacuum spontaneous-emission rate of an atom is found to be n [4–6] due to the dielectric renormalization of the electric field operator (3.1) by $1/n$, which is squared, and the $D=n^3$ density-of-states factor.

Local-field effects of a dielectric are suppressed in the macroscopic quantization procedure, and such local-field effects must be introduced phenomenologically. The paradigm that emerged from the propagation studies of Hopf and Scully [37] and Bloembergen's work [10] in nonlinear optics is that the effect of a dielectric host is to multiply each occurrence of the dipole moment of a two-level atom by a local-field enhancement factor [3,7,17]. Using the Lorentz virtual cavity model of the local field, the spontaneous-emission rate

$$\Gamma = n\ell^2\frac{4\omega_b^3|\mu_b|^2}{3c^3\hbar} = n\left(\frac{n^2+2}{3}\right)^2\Gamma_0 \tag{3.3}$$

for atoms in a dielectric scales as n^5 for large n. For an atom in a real cavity, the local-field enhancement factor is based on the Onsager model, and the modified spontaneous-emission rate

$$\Gamma = n\left(\frac{3n^2}{2n^2+1}\right)^2\Gamma_0 \tag{3.4}$$

scales as n. A study of local-field effects by de Vries and Lagendijk [38] found that the Lorentz virtual-cavity model is appropriate if the atom goes into a crystal substitutionally but that the Onsager real-cavity model should be used for interstitial impurities.

The macroscopic quantum electrodynamic theory can be used to derive additional consequences of the dielectric host for two-level atoms. Taking the field in a coherent state, the effective Hamiltonian is

$$\begin{aligned} H_{\text{eff}} = {} & \sum_{js}\frac{\hbar\omega_a}{2}\sigma_3^j + \hbar\sum_{l\lambda}\omega_l\bar{a}_l^\dagger\bar{a}_l \\ & - \frac{i\hbar}{n}\sum_{js}\sum_{l\lambda}(g_l^j\sigma_+^j\bar{a}_l e^{i\mathbf{k}_l\cdot\mathbf{r}_j} - g_l^{j*}\bar{a}_l^\dagger\sigma_-^j e^{-i\mathbf{k}_l\cdot\mathbf{r}_j}) \\ & - \frac{i\mu_a}{2}\sum_j(\sigma_+^j\bar{\mathcal{E}}e^{-i(\omega_p t-\mathbf{k}_p\cdot\mathbf{r}_j)} - \bar{\mathcal{E}}^*\sigma_-^j e^{i(\omega_p t-\mathbf{k}_p\cdot\mathbf{r}_j)}). \end{aligned} \tag{3.5}$$

For species a, σ_3^j is the inversion operator and $\sigma_\pm^j$ are the raising and lowering operators for the jth atom, $g_l^j = (2\pi\omega_l/\hbar V)^{1/2}\mu_a(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{e}}_{\mathbf{k}_l\lambda})$ is the coupling between the atom at position $\mathbf{r}_j$ and the radiation field, $\hat{\mathbf{x}}_j$ is a unit vector in the direction of the dipole moment at $\mathbf{r}_j$, ω_a is the transition frequency, and μ_a is the matrix element of the transition dipole moment.

Except for coefficients of $1/n$, and macroscopic field-mode operators, the effective Hamiltonian is the same as the microscopic Hamiltonian for identical two-level atoms in the vacuum. Equations of motion can then be derived in the same manner, which is the primary reason for adopting the macroscopic formalism. The formal integral of the Heisenberg equation of motion for the field-mode operators is used to eliminate these operators from the remaining Heisenberg equations of motion. One obtains

$$\begin{aligned} \frac{d\sigma_-^j}{dt} = {} & -i\omega_a\sigma_-^j(t) + \frac{\mu_a}{2\hbar}\sigma_3^j(t)\bar{\mathcal{E}}(t)e^{-i(\omega_p t-\mathbf{k}_p\cdot\mathbf{r}_j)} \\ & + \frac{1}{n}\sum_{l\lambda}g_l^j\sigma_3^j(t)\bar{a}_l(0)e^{-i(\omega_l t-\mathbf{k}_l\cdot\mathbf{r}_j)} \\ & + \frac{1}{n^2}\sum_{l\lambda}g_l^j\sigma_3^j(t)\int_0^t dt' e^{-i\omega_l(t-t')}\sum_{i\neq j,s}g_l^{i*}\sigma_-^i(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_j-\mathbf{r}_i)} \\ & + \frac{1}{n^2}\sum_{l\lambda}g_l^j\sigma_3^j(t)\int_0^t dt' e^{-i\omega_l(t-t')}g_l^{j*}\sigma_-^j(t') \end{aligned} \tag{3.6}$$

and a similar equation of motion for the inversion operator. The procedure to evaluate these terms for two-level atoms in the vacuum is generally known and will be considered in detail in the next section. For now it is sufficient to note that the only differences from the vacuum case are the coefficients of powers of n and the n^3 renormalization of the density of states in a dielectric in which the sum over modes is evaluated as

$$\sum_l \{\,\} \rightarrow n^3 \left(\frac{L}{2\pi c}\right)^3 \int_0^\infty d\omega_l \omega_l^2 \int d\Omega \{\,\} \tag{3.7}$$

in the mode continuum limit. Transforming to a rotating frame of reference and performing a local spatial average, one obtains the Bloch-like equations of motion

$$\frac{\partial R_{21}}{\partial t} = i\left(\omega_p - \omega_a - \frac{4\pi}{3\hbar} N \mu_a^2 n W\right) R_{21} - \frac{i\mu_a}{2\hbar} \bar{\mathcal{E}} W - n\frac{\Gamma_0}{2} R_{21}, \tag{3.8a}$$

$$\frac{\partial W}{\partial t} = -\frac{i}{\hbar}[\mu_a \bar{\mathcal{E}}^* R_{21} - \mu_a \bar{\mathcal{E}} R_{21}^*] - n\Gamma_0 (W+1), \tag{3.8b}$$

where $W = \langle \sigma_3 \rangle_{\mathrm{sp}}$ and $R_{21} = \langle -i\sigma_- \rangle_{\mathrm{sp}}$.

Equations (3.8) are the equations of motion for a two-level atom in a dielectric host medium that are derived using macroscopic quantum electrodynamics. The effects of the host appear as an enhancement of the decay rates and the Bowden-Lorentz redshift by a factor of the refractive index n, when compared to the vacuum $n=1$ case. However, the equations of motion derived using the classical Lorentz local-field condition, Eqs. (2.6), display an enhancement of both the Bowden-Lorentz redshift and the field by a factor of ℓ.

The fact that local-field effects are suppressed in the macroscopic theory is well known. The accepted practice [3,7,17] is to phenomenologically associate a local-field factor of ℓ with each occurrence of the dipole moment μ based on the propagation studies of Hopf and Scully [37] and Bloembergen's work [10] in nonlinear optics. In this case, Eqs. (3.8) become

$$\frac{\partial R_{21}}{\partial t} = i\left(\omega_p - \omega_a - \frac{4\pi}{3\hbar} N \mu_a^2 n \ell^2 W\right) R_{21} - \frac{i\mu_a}{2\hbar} \ell \bar{\mathcal{E}} W - n\ell^2 \frac{\Gamma_0}{2} R_{21}, \tag{3.9a}$$

$$\frac{\partial W}{\partial t} = -\frac{i}{\hbar}[\mu_a \ell \bar{\mathcal{E}}^* R_{21} - \mu_a \ell \bar{\mathcal{E}} R_{21}^*] - n\ell^2 \Gamma_0 (W+1). \tag{3.9b}$$

The ad hoc local-field correction gives the Bloembergen enhancement of the field that was derived classically in Eq. (2.6), although the redshift is overcorrected. More significantly, the Bowden-Lorentz redshift retains the extraneous factor of the refractive index due to the macroscopically quantized fields. Then the equations of motion for two-level atoms in a dielectric host, Eqs. (3.8) and (3.9), that were derived using the macroscopically quantized fields are inconsistent with the generalized Bloch equations (2.6) of the preceding section, and the application of quantized macroscopic fields to the electrodynamics of two-level atoms is contraindicated by classical Lorentz local-field theory.

IV. MICROSCOPIC QUANTUM ELECTRODYNAMICS

In the preceding two sections, we derived generalized Bloch equations of motion for two-level atoms in a dielectric host by two well-known methods and obtained contradictory results. The most fundamental theoretical approach is to represent both the atoms and the dielectric microscopically [7,23,25,32,39–44] and derive equations of motion from first principles. We show that the results of microscopic quantum electrodynamics affirm the Lorentz local-field theory with respect to the field renormalization and the Lorentz redshift. The macroscopic quantum electrodynamic theory produces different results for these effects and is therefore not valid.

A dielectric host containing one or more two-level atoms is modeled quantum electromagnetically as a mixture of two species of atoms, a and b, embedded in the vacuum. To emphasize the symmetry of the local-field interaction, both species of atoms are initially treated as two-level systems. Species b is later taken in the harmonic-oscillator limit that is associated with a relatively large detuning from resonance. Then, the total Hamiltonian is comprised of the Hamiltonians for the free atoms, the free-space quantized radiation field, and the interaction of the two-level systems with the free-space quantized electromagnetic field. The multipolar and minimal-coupling Hamiltonians are related by a canonical transformation and either can be used. However, due to the canonical transformation, the circumstances of the rotating-wave approximation (RWA) are different for the two Hamiltonians [26]. In the typical derivation of the dielectric susceptibility from the minimal-coupling Hamiltonian, the RWA is invoked implicitly by replacing polariton eigenenergies with photon energies [7,8,33]. We take the direct route and use the multipolar Hamiltonian in the RWA. Using a plane-wave expansion of the electromagnetic field

$$\mathbf{E} = i\hbar \sum_{l\lambda} \sqrt{\frac{2\pi\omega_l}{\hbar V}} (a_l e^{i\mathbf{k}_l \cdot \mathbf{r}_j} - \mathrm{H.c.}) \hat{\mathbf{e}}_{\mathbf{k}_l \lambda}, \tag{4.1}$$

the multipolar RWA Hamiltonian is [11,45]

$$\begin{aligned} H = {} & \sum_{js} \frac{\hbar\omega_a}{2} \sigma_3^j + \sum_{ns} \frac{\hbar\omega_b}{2} \varsigma_3^n + \sum_{l\lambda} \hbar\omega_l a_l^\dagger a_l \\ & - i\hbar \sum_{js} \sum_{l\lambda} (g_l^j \sigma_+^j a_l e^{i\mathbf{k}_l \cdot \mathbf{r}_j} - g_l^{j*} a_l^\dagger \sigma_-^j e^{-i\mathbf{k}_l \cdot \mathbf{r}_j}) \\ & - i\hbar \sum_{ns} \sum_{l\lambda} (h_l^n \varsigma_+^n a_l e^{i\mathbf{k}_l \cdot \mathbf{r}_n} - h_l^{n*} a_l^\dagger \varsigma_-^n e^{-i\mathbf{k}_l \cdot \mathbf{r}_n}), \end{aligned} \tag{4.2}$$

where $a_l^\dagger$ and a_l are the creation and destruction operators for the field modes and ω_l is the frequency of the field in the mode l with $k_l = \omega_l / c$. For species a, σ_3^j is the inversion operator and $\sigma_\pm^j$ are the raising and lowering operators for the jth atom, $g_l^j = (2\pi\omega_l/\hbar V)^{1/2} \mu_a (\hat{\mathbf{x}}_j \cdot \hat{\mathbf{e}}_{\mathbf{k}_l \lambda})$ is the coupling between the atom at position $\mathbf{r}_j$ and the radiation field, $\hat{\mathbf{x}}_j$ is a unit vector in the direction of the dipole moment at $\mathbf{r}_j$, ω_a is the transition frequency, and μ_a is the dipole moment. For species b, ς_3^n, $\varsigma_\pm^n$, h_l^n, $\mathbf{r}_n$, ω_b, and μ_b perform the same functions. Also, V is the quantization volume, $\hat{\mathbf{e}}_{\mathbf{k}_l\lambda}$ is the polarization vector, and λ denotes the state of polarization. The polarization indices on the variables have been suppressed for clarity. In the two-level approximation, the transitions $s=\Delta m \in (-1,0,+1)$ are treated separately and the operators need not carry a specific value for the magnetic sublevel [46].

Equations of motion for the material and field-mode operators are developed in a straightforward manner from the Hamiltonian using the Heisenberg equation. We have

$$\frac{da_l}{dt} = -i\omega_l a_l + \sum_{js} g_l^{j*}\sigma_-^j e^{-i\mathbf{k}_l\cdot\mathbf{r}_j} + \sum_{ns} h_l^{n*}\varsigma_-^n e^{-i\mathbf{k}_l\cdot\mathbf{r}_n} \tag{4.3a}$$

$$\frac{d\sigma_-^j}{dt} = -i\omega_a\sigma_-^j + \sum_{l\lambda} g_l^j\sigma_3^j a_l e^{i\mathbf{k}_l\cdot\mathbf{r}_j}, \tag{4.3b}$$

$$\frac{d\sigma_3^j}{dt} = -2\sum_{l\lambda}\left(g_l^j\sigma_+^j a_l e^{i\mathbf{k}_l\cdot\mathbf{r}_j} + g_l^{j*}a_l^\dagger\sigma_-^j e^{-i\mathbf{k}_l\cdot\mathbf{r}_j}\right), \tag{4.3c}$$

$$\frac{d\varsigma_-^n}{dt} = -i\omega_b\varsigma_-^n + \sum_{l\lambda} h_l^n\varsigma_3^n a_l e^{i\mathbf{k}_l\cdot\mathbf{r}_n}, \tag{4.3d}$$

$$\frac{d\varsigma_3^n}{dt} = -2\sum_{l\lambda}\left(h_l^n\varsigma_+^n a_l e^{i\mathbf{k}_l\cdot\mathbf{r}_n} + h_l^{n*}a_l^\dagger\varsigma_-^n e^{-i\mathbf{k}_l\cdot\mathbf{r}_n}\right). \tag{4.3e}$$

Bloch-like operator equations of motion are obtained by substituting the formal integral of the field-mode operator equation (4.3a),

$$a_l(t) = a_l(0)e^{-i\omega_l t} + \int_0^t dt'\, e^{-i\omega_l(t-t')} \times \left(\sum_{js} g_l^{j*}\sigma_-^j(t')e^{-i\mathbf{k}_l\cdot\mathbf{r}_j} + \sum_{ns} h_l^{n*}\varsigma_-^n(t')e^{-i\mathbf{k}_l\cdot\mathbf{r}_n}\right), \tag{4.4}$$

into the material operator equations of motion (4.3b), (4.3c), (4.3d), and (4.3e) [47]. We transform operator variables to different rotating frames of reference in which $\tilde{\sigma}_-^j = \sigma_-^j e^{i\omega_a t}$ and $\breve{\varsigma}_-^n = \varsigma_-^n e^{i\omega_b t}$ are slowly varying quantities. Performing the indicated substitution into Eq. (4.3b) produces

$$\begin{aligned}\frac{d\tilde{\sigma}_-^j}{dt} = {} & \sum_{l\lambda} g_l^j\sigma_3^j(t)e^{i\mathbf{k}_l\cdot\mathbf{r}_j}a_l(0)e^{-i(\omega_l-\omega_a)t} + \sigma_3^j(t)\sum_{l\lambda} g_l^j \\ & \times \int_0^t dt'\, e^{-i(\omega_l-\omega_a)(t-t')}\sum_{is} g_l^{i*}\tilde{\sigma}_-^i(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_j-\mathbf{r}_i)} \\ & + e^{-i(\omega_b-\omega_a)t}\sum_{l\lambda} g_l^j\sigma_3^j(t)\int_0^t dt'\, e^{-i(\omega_l-\omega_b)(t-t')} \\ & \times \sum_{ns} h_l^{n*}\breve{\varsigma}_-^n(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_j-\mathbf{r}_n)}\end{aligned} \tag{4.5}$$

in normal ordering with $\mathbf{r}_{ji}=\mathbf{r}_j-\mathbf{r}_i$. Likewise, one obtains

$$\begin{aligned}\frac{d\breve{\varsigma}_-^n}{dt} = {} & \sum_{l\lambda} h_l^n\varsigma_3^n e^{i\mathbf{k}_l\cdot\mathbf{r}_n}a_l(0)e^{-i(\omega_l-\omega_b)t} \\ & + e^{i(\omega_b-\omega_a)t}\sum_{l\lambda} h_l^n\varsigma_3^n(t)\int_0^t dt'\, e^{-i(\omega_l-\omega_a)(t-t')} \\ & \times \sum_{js} g_l^{j*}\tilde{\sigma}_-^j(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_n-\mathbf{r}_j)} \\ & + \sum_{l\lambda} h_l^n\varsigma_3^n(t)\int_0^t dt'\, e^{-i(\omega_l-\omega_b)(t-t')} \\ & \times \sum_{ms} h_l^{m*}\breve{\varsigma}_-^m(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_n-\mathbf{r}_m)}\end{aligned} \tag{4.6}$$

from Eq. (4.3d). Equations of motion for the inversion operators are obtained from Eqs. (4.3c) and (4.3e) in a similar fashion.

The field that drives an atom, Eq. (4.4), consists of the vacuum field, the self-field, and the reaction field and we can identify the terms on the right-hand side of Eqs. (4.5) and (4.6) with fluctuations due to the vacuum field, spontaneous decay from the self-field, near-dipole-dipole interactions between same-species atoms associated with the reaction field, and near-dipole-dipole interactions of an atom with the atoms of the other species, also associated with the reaction field. The usual procedure is to limit consideration to only the spontaneous decay rate of a single impurity atom by dropping the fluctuations and the single-species interactions for both the bath and impurity atoms. These terms are retained here because they contain significant information about local-field effects in dielectrics. For concreteness, we take species b to be the bath atoms and species a to be the two-level impurity atoms.

V. NEAR-DIPOLE-DIPOLE INTERACTION

The near-dipole-dipole interaction is the basic mechanism of the action of the local field. The Weisskopf-Wigner-based procedure to evaluate the dipole-dipole interaction for a dense collection of identical two-level atoms was developed by Ben-Aryeh, Bowden, and Englund [14], with corrections by Benedict, Malyshev, Trifonov, and Zaitsev [48], to investigate single-species intrinsic optical bistability. The results apply to both species of two-level atoms, individually, but we work with species b in order to maintain consistent notation when we take the harmonic-oscillator limit of a two-level atom and derive the Lorentz local-field correction in a dielectric.

We consider a dense collection of identical two-level atoms of species b in which the atoms are evenly distributed in the vacuum with a number density N_b. The same-species interaction

$$I_1 = \sum_{l\lambda} h_l^n\varsigma_3^n(t)\int_0^t dt'\, e^{-i(\omega_l-\omega_b)(t-t')}\sum_{ms} h_l^{m*}\breve{\varsigma}_-^m(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_n-\mathbf{r}_m)} \tag{5.1}$$

can be extracted from Eq. (4.6).

The self-interaction of the nth atom with its own reaction field is characterized by the term $\mathbf{r}_m=\mathbf{r}_n$ in the interaction as a consequence of the relation $\varsigma_3^n(t)\breve{\varsigma}_-^n(t)=-\breve{\varsigma}_-^n(t)$ between Pauli spin operators for the same atom. Then

$$I_1^{\mathrm{self}}=\sum_{l\lambda} h_l^n \varsigma_3^n(t)\int_0^t dt' e^{-i(\omega_l-\omega_b)(t-t')} h_l^{n*}\breve{\varsigma}_-^n(t'). \tag{5.2}$$

Applying the typical Weisskopf-Wigner procedure [49–51] in the mode continuum limit, one obtains

$$I_1^{\mathrm{self}}=-\frac{2\omega_b^3|\mu_b|^2}{3\hbar c^3}\breve{\varsigma}_-^n(t)=-\frac{\gamma_b}{2}\breve{\varsigma}_-^n(t), \tag{5.3}$$

where

$$\gamma_b=\frac{4\omega_b^3|\mu_b|^2}{3\hbar c^3} \tag{5.4}$$

is the spontaneous decay rate. For an atom of species b, initially in the excited state, γ_b is the spontaneous-emission rate Γ_0 into the vacuum.

The pairwise interaction of atoms is carried in the remaining $m\neq n$ part of the summation. In the Milonni-Knight [46] model of the interaction of two identical two-level atoms, the strength of the interaction depends on the separation distance and the magnetic sublevel transition. Then [46],

$$F_1(R)=e^{iR}\left(-\frac{i}{R}+\frac{i}{R^3}+\frac{1}{R^2}\right) \tag{5.5}$$

for $\Delta m=\pm 1$ transitions and

$$F_2(R)=e^{iR}\left(-\frac{2i}{R^3}-\frac{2}{R^2}\right) \tag{5.6}$$

for $\Delta m=0$ transitions, where

$$\beta_b=\frac{2\omega_b^3|\mu_b|^2}{3\hbar c^3}, \tag{5.7}$$

$R=k_b r_{nm}=\omega_b r_{nm}/c$, $k_b=|\mathbf{k}_b|$, $\mathbf{r}_{nm}=\mathbf{r}_n-\mathbf{r}_m$, and $r_{nm}=|\mathbf{r}_{nm}|$. Performing the summation over the magnetic sublevels, the pairwise dipole-dipole interaction can be written as

$$I_1^{\mathrm{dd}}=\varsigma_3^n(t)\frac{3}{2}\beta_b\sum_{m\neq n}B_{nm}\breve{\varsigma}_-^m(t-r_{nm}/c), \tag{5.8}$$

where

$$B_{nm}=[(\hat{\mathbf{x}}_m\cdot\hat{\mathbf{x}}_n)-(\hat{\mathbf{x}}_m\cdot\hat{\mathbf{n}}_{nm})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{nm})]F_1(k_b r_{nm})+(\hat{\mathbf{x}}_m\cdot\hat{\mathbf{n}}_{nm})\times(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{nm})F_2(k_b r_{nm}) \tag{5.9}$$

and $\hat{\mathbf{n}}_{nm}=\mathbf{r}_{nm}/r_{nm}$ is a unit vector in the direction of $\mathbf{r}_{nm}=\mathbf{r}_n-\mathbf{r}_m$ [14,48]. Further, B_{nm} incorporates a view factor to account for the arrangement of the dipoles in the volume.

The atoms are evenly distributed with a number density N_b. For the nth atom, the single-species dipole-dipole interaction is obtained in a summation over all other atoms of species b. In the region near $\mathbf{r}_n$, the interaction is evaluated by taking the location of dipoles as discrete, while the continuum approximation is applied elsewhere. Then

$$I_1^{\mathrm{dd}}=\varsigma_3^n(t)\frac{3}{2}\beta_b\sum_{m:r_{nm}<\delta}B_{nm}\breve{\varsigma}_-^m(t-r_{nm}/c)+\varsigma_3^n(t)\frac{3}{2}\beta_b N_b\int_{V-V_\delta}B\breve{\varsigma}_-(t-|\mathbf{r}|/c)d^3\mathbf{r}, \tag{5.10}$$

where δ is the radius of a small spherical volume V_δ, larger than a cubic wavelength, about the point $\mathbf{r}_n$. For cubic symmetry, the field generated by the localized atoms $\mathbf{r}_m\neq\mathbf{r}_n$ in the virtual cavity is zero at the center [12,14,48].

The atom n is located at the origin of a cylindrical volume of thickness L and radius R_0. The near-dipole-dipole interaction is obtained by evaluating the integral

$$I_1^{\mathrm{dd}}\approx\frac{3}{2}\beta_b N_b\varsigma_3^n(t)\int_0^{2\pi}d\phi\int_{-L/2}^{L/2}dz\int_{\rho_{\min}}^{\rho_{\max}}\rho d\rho\breve{\varsigma}_-\times\left\{\left[1-\left(\frac{\rho}{r}\right)^2\cos^2\phi\right]F_1+\left(\frac{\rho}{r}\right)^2\cos^2\phi F_2\right\}, \tag{5.11}$$

excluding a volume $(4/3)\pi\delta^3$ about the origin from the range of integration, resulting in

$$I_1^{\mathrm{dd}}\approx\frac{-4\pi i}{k_b^3}\frac{N_b\omega_b^3|\mu_b|^2}{\hbar c^3}e^{ik_b\delta}\varsigma_3^n(t)\langle\breve{\varsigma}_-(t-|\mathbf{r}|/c)\rangle_{\mathrm{sp}}. \tag{5.12}$$

In the limit $\delta\rightarrow 0$, the near-dipole-dipole interaction

$$I_1^{\mathrm{dd}}=-i\nu_b\varsigma_3^n(t)\bar{\varsigma}_- \tag{5.13}$$

remains finite. Here

$$\nu_b=\frac{4\pi}{3\hbar}N_b|\mu_b|^2 \tag{5.14}$$

is the strength of the near-dipole-dipole interaction and $\bar{\varsigma}_-=\langle\breve{\varsigma}_-(t-|\mathbf{r}|/c)\rangle_{\mathrm{sp}}$ represents a spatially averaged quantity. The details of this calculation can be found in the articles by Ben-Aryeh, Bowden, and Englund [14] and by Benedict, Malyshev, Trifonov, and Zaitsev [48].

The atoms of species b can be treated as harmonic oscillators if all excitation frequencies are far from resonance with ω_b. In this limit, the atom essentially remains in the ground state such that $\varsigma_3^n\rightarrow -1$. Then the near-dipole-dipole interaction reduces to the Lorentz local-field correction, shifting the resonance frequency by $4\pi N_b|\mu_b|^2/(3\hbar)$. The microscopic result is in full agreement with the classical Lorentz local-field correction and has been experimentally validated [52,53] by selective reflection of Rb from a sapphire window.

Finally, all of the results of this section can be applied to the other species of atom. Repeating for species a yields

$$I_1^{\mathrm{self}}=-\frac{2\omega_a^3|\mu_a|^2}{3\hbar c^3}\tilde{\sigma}_-^j(t)=-\frac{\gamma_a}{2}\tilde{\sigma}_-^j(t), \tag{5.15}$$

$$I_1^{\mathrm{dd}} \approx -i\nu_a \sigma_3^j(t)\langle\tilde{\sigma}_-(t-|\mathbf{r}|/c)\rangle_{\mathrm{sp}} = -i\nu_a\sigma_3^j(t)\bar{\sigma}_-. \tag{5.16}$$

In addition,

$$\gamma_a = 2\beta_a = \frac{4\omega_a^3|\mu_a|^2}{3\hbar c^3} \tag{5.17}$$

and

$$\nu_a = \frac{4\pi}{3\hbar}N_a|\mu_a|^2 \tag{5.18}$$

are defined for later use.

VI. INTERSPECIES INTERACTION

The effect of the interspecies near-dipole-dipole interaction can also be evaluated microscopically using Weisskopf-Wigner theory. A single rotating frame of reference is used for both species of atoms by making the transformation $\tilde{\varsigma}_-^n = \breve{\varsigma}_-^n e^{-i(\omega_b-\omega_a)t}$. Applying the results of the preceding section, we have

$$\frac{d\tilde{\varsigma}_-^n}{dt} = -i(\omega_b-\omega_a)\tilde{\varsigma}_-^n - \frac{\mu_b}{\hbar}f_b^- + i\nu_b\bar{\varsigma}_- - \frac{\gamma_b}{2}\tilde{\varsigma}_-^n - \sum_{l\lambda} h_l^n \int_0^t dt' e^{-i(\omega_l-\omega_a)(t-t')}\sum_{js} g_l^{j*}\tilde{\sigma}_-^j(t')e^{i\mathbf{k}_l\cdot(\mathbf{r}_n-\mathbf{r}_j)} \tag{6.1}$$

in the harmonic-oscillator limit $\varsigma_3^n(t)\rightarrow -1$, where the fluctuating field

$$f_b^- = \sum_{l\lambda}(2\pi\omega_l\hbar/V)^{1/2}(\hat{\mathbf{x}}_n\cdot\mathbf{e}_k)e^{i\mathbf{k}_l\cdot\mathbf{r}_n}a_l(0)e^{-i(\omega_l-\omega_b)t}$$

is associated with the spontaneous decay rate by the Kramers-Kronig relations.

The interspecies interaction in Eq. (6.1) describes how a specific host atom n interacts pairwise with each of the impurity atoms. The formal integral of the equation of motion of the host atoms, Eq. (6.1), is

$$\tilde{\varsigma}_-^n(t) = -\int_0^t dt' e^{-i\alpha(t-t')}\frac{\mu_b}{\hbar}f^- - \int_0^t dt' e^{-i\alpha(t-t')} \times \sum_{l\lambda} h_l^n \int_0^{t'} dt'' e^{-i(\omega_l-\omega_a)(t'-t'')}\sum_{is} g_l^{i*}\tilde{\sigma}_-^i(t'')e^{i\mathbf{k}_l\cdot\mathbf{r}_{ni}}, \tag{6.2}$$

where $\alpha=\omega_b-\omega_a-\nu_b-i\gamma_b/2$. Substituting Eq. (6.2) into Eq. (4.5), one obtains

$$\frac{d\tilde{\sigma}_-^j}{dt} = \frac{\mu_a}{\hbar}\sigma_3^j f^- - i\nu_a\sigma_3^j\bar{\sigma}_- - \frac{\gamma_a}{2}\tilde{\sigma}_-^j + I_2, \tag{6.3}$$

where

$$I_2 = -\sum_{nsl\lambda} g_l^j\sigma_3^j(t)\int_0^t dt' e^{-i(\omega_l-\omega_a)(t'-t)}h_l^{n*} \times e^{i\mathbf{k}_l\cdot\mathbf{r}_{jn}}\int_0^{t'} dt'' e^{-i\alpha(t'-t'')}\sum_{l'\lambda'} h_{l'}^n \int_0^{t''} dt''' \times e^{-i(\omega_{l'}-\omega_a)(t''-t''')}\sum_{is'} g_{l'}^{i*}\tilde{\sigma}_-^i(t''')e^{i\mathbf{k}_{l'}\cdot\mathbf{r}_{ni}}. \tag{6.4}$$

The term containing the fluctuations has been dropped from consideration because the procedures presented here apply only to slowly varying quantities and because there will be no contribution from the random fluctuations after averaging.

The direct dipole-dipole interactions between impurity atoms were derived in Sec. V. Equation (6.4) contains two such dipole-dipole interactions between nonidentical atoms that are integrated over the different subspaces corresponding to (i) bath atoms and (ii) two-level impurity atoms. Due to the relation $\sigma_3^j(t)\sigma_-^j(t) = -\sigma_-^j(t)$ between Pauli spin operators, the term $i=j$ is the special case that is associated with the renormalization of the spontaneous decay rate. This separates the interaction $I_2 = I_2^{\mathrm{ndd}} + I_2^{\mathrm{self}}$ into I_2^{self} for the case $i=j$ and I_2^{ndd} for the summation over the rest of the impurity atoms. The two parts of the interaction will be considered separately.

A. Dielectric mediated dipole-dipole interaction

The nth atom of the dielectric interacts pairwise with every impurity atom. The term

$$I_2^{\mathrm{dd}^A} = \sum_{l'\lambda'} h_{l'}^n \int_0^{t''} dt''' e^{-i(\omega_{l'}-\omega_a)(t''-t''')}\sum_{i\neq j,s} g_{l'}^{i*}\tilde{\sigma}_-^i(t''')e^{i\mathbf{k}_{l'}\cdot(\mathbf{r}_n-\mathbf{r}_i)}, \tag{6.5}$$

extracted from Eq. (6.4), can be evaluated in the same manner as in Sec. V, except that the atoms are of different species. The summation represents the effect of all the impurity atoms on a single atom of the host material. The sum over the $i\neq j$ impurity atoms is performed (i) in the near region by the discrete summation over the impurity atoms and (ii) elsewhere by treating the impurity atoms in the continuum limit. For nonidentical atoms, the pairwise interaction goes as [54,55]

$$I_2^{\mathrm{dd}^A} = \frac{3}{2}\sqrt{\beta_a\beta_b}\sum_{i:r_{ni}<\delta} B_{ni}\tilde{\sigma}_-^i(t''-r_{ni}/c) + \frac{3}{2}\sqrt{\beta_a\beta_b}N_a\int_{V-V_\delta} B\tilde{\sigma}_-(t''-|\mathbf{r}|/c)d^3\mathbf{r}. \tag{6.6}$$

Then, Eq. (6.6) is evaluated as in Sec. V to obtain [14]

$$I_2^{\mathrm{dd}^A} = -\frac{4\pi i}{3\hbar}N_a\mu_a^*\mu_b\langle\tilde{\sigma}_-(t''-|\mathbf{r}|/c)\rangle_{\mathrm{sp}} = -\frac{4\pi i}{3\hbar}N_a\mu_a^*\mu_b\bar{\sigma}_-. \tag{6.7}$$

The quantity $\bar{\sigma}_- = \langle\tilde{\sigma}_-(t''-|\mathbf{r}|/c)\rangle_{\mathrm{sp}}$ is slowly varying in time, and the temporal integral

$$I_2^{\mathrm{dd}^B}(t')=\int_0^{t'}dt''e^{-i\alpha(t'-t'')}\left(\frac{-4\pi i}{3\hbar}\right)N_a\mu_a^*\mu_b\bar{\sigma}_- \tag{6.8}$$

can be performed in the adiabatic-following approximation. Repeatedly integrating Eq. (6.8) by parts [56], the series can be truncated at the first term in the expansion if the time rate of change of $\bar{\sigma}_-$ is much smaller than $\alpha\bar{\sigma}_-$ yielding

$$I_2^{\mathrm{dd}^B}(t')=\left(\frac{-1}{\alpha}\right)\left(\frac{-4\pi i}{3\hbar}\right)N_a\mu_a^*\mu_b\bar{\sigma}_-. \tag{6.9}$$

The remaining part of the I_2^{dd} integration is another interspecies dipole-dipole interaction. In this case, the summation imparts the effect of all the atoms of the host dielectric, modified by interspecies interaction with the impurity atoms, on the jth impurity atom. Combining terms,

$$I_2^{\mathrm{dd}}(t')=\frac{-4\pi i}{3\hbar}N_b\mu_b^*\mu_a\frac{1}{\alpha}\frac{4\pi i}{3\hbar}N_a\mu_a^*\mu_b\sigma_3^j\bar{\sigma}_- \tag{6.10}$$

becomes

$$I_2^{\mathrm{dd}}(t')=-\frac{4\pi i}{3}\chi_b\nu_a\sigma_3^j\bar{\sigma}_-, \tag{6.11}$$

where χ_b is the linear susceptibility of species b. Adding the direct near-dipole-dipole interaction from Eq. (6.3), we obtain

$$\begin{aligned}I_2^{\mathrm{dd}}-i\nu_a\sigma_3(t)\bar{\sigma}_-(t)&=-i\left(1+\frac{4\pi}{3}\chi_b\right)\nu_a\sigma_3(t)\bar{\sigma}_-(t)\\&=-i\frac{n^2+2}{3}\nu_a\sigma_3(t)\bar{\sigma}_-(t).\end{aligned} \tag{6.12}$$

Comparison of Eq. (6.12) with the single species dipole-dipole interaction, Eq. (5.16), shows that the effect of the dielectric host is to enhance the interaction by a factor of $\ell=(n^2+2)/3$. Taking the local spatial average, $W=\langle\sigma_3\rangle_{\mathrm{sp}}$ and $R_{21}=\langle -i\sigma_-\rangle_{\mathrm{sp}}$, one finds that the Lorentz redshift

$$\frac{n^2+2}{3}\frac{4\pi}{3\hbar}N\mu^2 \tag{6.13}$$

is consistent with the Lorentz local-field calculation, Eq. (2.6a), while the macroscopic quantum electrodynamic result, Eq. (3.9a), is not.

B. Dielectric-enhanced spontaneous decay rate

Most of the elements of the microscopic theory of the spontaneous decay rate of an atom in a dielectric are common to the treatment of the dipole-dipole interaction. In order to show this clearly, we consider an equivalent derivation of the dielectric mediated dipole-dipole interaction. Performing the temporal integrations first, the interspecies interaction (6.4) can be written as

$$\begin{aligned}I_2=&-\sum_{nsl\lambda}g_l^j\sigma_3^j(t)\pi\delta(\omega_l-\omega_a)h_l^{n*}e^{i\mathbf{k}_l\cdot(\mathbf{r}_j-\mathbf{r}_n)}\\&\times\left(\frac{-i}{\alpha}\right)\sum_{is'l'\lambda'}h_{l'}^n\pi\delta(\omega_{l'}-\omega_a)\sum_{is'}g_{l'}^{i*}\tilde{\sigma}_-^i(t')e^{i\mathbf{k}_{l'}\cdot(\mathbf{r}_n-\mathbf{r}_i)}.\end{aligned} \tag{6.14}$$

Applying the Milonni-Knight interaction with a view factor results in

$$\begin{aligned}I_2=&-\frac{-i}{\alpha}\frac{9}{4}\beta_a\beta_b\sigma_3^j\sum_n\{[(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{n}}_{nj})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{nj})]F_2(k_ar_{nj})\\&+[(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{x}}_n)-(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{n}}_{nj})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{nj})]F_1(k_ar_{nj})\}\\&\times\sum_i\{[(\hat{\mathbf{x}}_i\cdot\hat{\mathbf{n}}_{ni})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{ni})]F_2(k_ar_{ni})\\&+[(\hat{\mathbf{x}}_i\cdot\hat{\mathbf{x}}_n)-(\hat{\mathbf{x}}_i\cdot\hat{\mathbf{n}}_{ni})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{ni})]F_1(k_ar_{ni})\}\sigma_-^i.\end{aligned} \tag{6.15}$$

Converting the sums, excluding $i=j$, to integrals and integrating over the subspace of two-level atoms and then over the subspace of oscillators is equivalent to the derivation of the dielectric mediated dipole-dipole interaction that was presented in the preceding subsection.

The renormalization of the spontaneous decay rate of a dielectric-embedded two-level atom is derived from the interspecies interaction (6.4) in the same fashion by taking the target atom to be the same as the source atom. The summation over the two-level atoms is evaluated with the use of the delta function δ_{ij}, rather than the integration over the subspace of two-level atoms. Likewise, the summation over the magnetic sublevels invokes $\delta_{ss'}$. Then,

$$\begin{aligned}I_2^{\mathrm{self}}=&\frac{-i}{\alpha}\frac{9}{4}\beta_a\beta_b\sigma_-^j\sum_n\{[(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{n}}_{nj})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{nj})]^2F_2^2(k_ar_{nj})\\&+[(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{x}}_n)-(\hat{\mathbf{x}}_j\cdot\hat{\mathbf{n}}_{nj})(\hat{\mathbf{x}}_n\cdot\hat{\mathbf{n}}_{nj})]^2F_1^2(k_ar_{nj})\}.\end{aligned} \tag{6.16}$$

The microscopic treatments of the spontaneous decay rate in a dielectric [11,45] are missing elements of Eq. (6.16) and can neither affirm nor contradict the macroscopic theory of quantum electrodynamics. Because the dielectric renormalization of the spontaneous decay rate does not have a classical local-field condition-based analog, we do not consider it further.

VII. DIELECTRIC-ENHANCED FIELD

The dielectric has an effect on an applied electromagnetic field that can also be evaluated microscopically. Taking the field in a coherent state, the partial Hamiltonian is

$$\begin{aligned}H_f=&-\frac{i\hbar}{2}\sum_j(\Omega_a\sigma_+^je^{-i(\omega_pt-\mathbf{k}_p\cdot\mathbf{r}_j)}-\Omega_a^*\sigma_-^je^{i(\omega_pt-\mathbf{k}_p\cdot\mathbf{r}_j)})\\&-\frac{i\hbar}{2}\sum_n(\Omega_bs_+^ne^{-i(\omega_pt-\mathbf{k}_p\cdot\mathbf{r}_n)}-\Omega_a^*s_-^ne^{i(\omega_pt-\mathbf{k}_p\cdot\mathbf{r}_n)}),\end{aligned} \tag{7.1}$$

where ω_p is the nominal frequency of the field and Ω_a

$=\mu_a\mathcal{E}/\hbar$ and $\Omega_b=\mu_b\mathcal{E}/\hbar$ are Rabi frequencies. The total Hamiltonian is now comprised of the Hamiltonians (7.1) and (4.2). Developing Heisenberg equations of motion and eliminating the field-mode operators and the dielectric operators results in the appearance of

$$I_3=\frac{\mu_a}{2\hbar}\sigma_3^j(t)\mathcal{E}(t)-\sum_{nsl\lambda}g_l^j\sigma_3^j(t)\int_0^t dt'\,e^{-i(\omega_l-\omega_p)(t-t')}\times h_l^{n*}e^{i(\mathbf{k}_l-\mathbf{k}_p)\cdot\mathbf{r}_{jn}}\int_0^{t'}dt''e^{\alpha(t'-t'')}\frac{\mu_a}{2\hbar}\mathcal{E}(t'') \qquad (7.2)$$

as an addition to Eq. (6.3). Equation (7.2) contains the same type of interaction that was evaluated in the previous section. Performing the adiabatic-following approximation and the sum over polarizations, bath atoms, and magnetic sublevels in the mode continuum limit, we obtain

$$I_3=\frac{n^2+2}{3}\frac{\mu_a}{2\hbar}\mathcal{E}\sigma_3^j. \qquad (7.3)$$

The electromagnetic field is enhanced by the same factor of ℓ as the reaction field.

VIII. OPTICAL BLOCH EQUATIONS FOR EMBEDDED ATOMS

The macroscopic optical Bloch equations can be derived from the quantum electrodynamic equations of motion in the limit of large numbers. Combining Eqs. (6.3), (6.11), and (7.2), one obtains

$$\frac{d\tilde{\sigma}_-^j}{dt}=-i\ell\nu_a\sigma_3^j\tilde{\sigma}_-+\ell\frac{\mu_a}{2\hbar}\mathcal{E}e^{-i(\omega_p-\omega_a)t}\sigma_3^j, \qquad (8.1)$$

neglecting the Gaussian noise source with zero mean and absorption. The equation of motion for the inversion operator

$$\frac{d\sigma_3^j}{dt}=2\left[i\ell\nu_a\tilde{\sigma}_+^j\tilde{\sigma}_--\frac{\mu_a}{2\hbar}\tilde{\sigma}_+^j\ell\mathcal{E}e^{-i(\omega_p-\omega_a)t}+\text{H.c.}\right] \qquad (8.2)$$

is derived in a similar manner. Optical Bloch equations of motion are obtained by transforming to a frame rotating at the frequency of the field and taking a local-spatial average, as in Sec. III. We compare the optical Bloch equations

$$\frac{\partial R_{21}}{\partial t}=i\left(\omega_p-\omega_a-\frac{4\pi}{3\hbar}N\mu_a^2\ell W\right)R_{21}-\frac{i\mu_a}{2\hbar}\ell\mathcal{E}W, \qquad (8.3a)$$

$$\frac{\partial W}{\partial t}=-\frac{i}{\hbar}[\mu_a\ell^*\mathcal{E}^*R_{21}-\mu_a\ell\mathcal{E}R_{21}^*], \qquad (8.3b)$$

that were derived from first principles, to the Lorentz local-field-based equations (2.6). Based on a favorable comparison of the local-field enhancement of the Lorentz redshift and the Rabi frequency with the classically derived result, we can reasonably assert that the microscopic theory, unlike the macroscopic quantum electrodynamic theory, satisfies the correspondence principle.

The microscopic theory allows us to consider the more general case of a complex local-field enhancement factor. Separating the real and imaginary parts of ℓ, the optical Bloch equations can be written as

$$\frac{\partial R_{21}}{\partial t}=i\left(\omega_p-\omega_a-\frac{4\pi}{3\hbar}N\mu_a^2\ell_rW\right)R_{21}-\frac{i\mu_a}{2\hbar}\ell\mathcal{E}W-\frac{4\pi}{3\hbar}N\mu_a^2\ell_iWR_{21}, \qquad (8.4a)$$

$$\frac{\partial W}{\partial t}=-\frac{i}{\hbar}[\mu_a\ell^*\mathcal{E}^*R_{21}-\mu_a\ell\mathcal{E}R_{21}^*]-4\ell_i\nu_a|R_{21}|^2 \qquad (8.4b)$$

with $\ell=\ell_r+i\ell_i$. The microscopic theory justifies the use of a complex refractive index in the classical Lorentz local-field condition. Then Eqs. (8.4), with phenomenological damping, can be derived by substituting the polarization (2.5) with complex n into the generalized Bloch equations (2.4). The imaginary part of the Lorentz redshift, derived in this manner, was found to be associated with an intrinsic cooperative decay for two-level atoms in an absorptive host [9]. This result is confirmed by the microscopic theory.

The optical Bloch equations (8.4) for dielectric-embedded two-level atoms are derived from the microscopic description of quantum electrodynamics using vacuum-based fields that are known to satisfy the equal-time commutation relations. Because the field-mode operators have been eliminated, the equal-time commutation relations cannot be discussed in the context of the optical Bloch equations (8.4) or Heisenberg equations (8.1) and (8.2). Instead, the optical Bloch equations, generalized for a dielectric host, must demonstrate conservation of probability. The total population is $W^2+4|R_{21}|^2$. Direct substitution from Eqs. (8.4) shows that the temporal derivative of this quantity is nil, as required, in the limit that absorption by the atoms and the host dielectric can be neglected.

IX. SUMMARY

The interest in the dielectric renormalization of the spontaneous-emission rate of an atom embedded in a dielectric material has obscured the inconsistencies in the macroscopic theory of quantum electrodynamics. The dielectric renormalization of the Lorentz redshift and the Rabi frequency, but not the spontaneous decay rate, can be derived using the classical Lorentz local-field condition providing an independent check on the validity of macroscopic quantum electrodynamics. The optical Bloch equations for a dense collection of two-level atoms in a dielectric host medium were derived using the classical Lorentz local-field condition

$$\mathbf{E}_L=\mathbf{E}+\frac{4\pi}{3}\mathbf{P}$$

in Sec. II, while in Sec. III, a different set of optical Bloch equations were derived using the macroscopic quantum electrodynamic theory. Both derivations are short and uncomplicated and are based on well-established physical theories, yet lead to contradictory results for the Lorentz redshift and the Rabi frequency. If we assume the validity of the Lorentz local-field condition, then the macroscopic procedure is proven to be incorrect. Conversely, the validity of the macroscopic quantum electrodynamic theory would imply that the Lorentz local-field condition is incorrect. One deciding factor is that the Lorentz local-field correction has been validated experimentally [52,53], while the experimental record for the macroscopic quantum theory has been inconclusive. We applied the more fundamental microscopic theory of quantum electrodynamics to the same problem and demonstrated complete agreement with classical theory. The differences in the Rabi frequencies can be reconciled with a phenomenological local-field factor applied in the macroscopic case, providing the virtual-cavity model is used. However, no such facile reconciliation can be provided for the Lorentz redshift. We conclude that both the correspondence principle and microscopic quantum electrodynamics are violated by the macroscopic quantum electrodynamic theory.

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