Quantum optics in a dielectric: macroscopic electromagnetic-field and medium operators for a linear dispersive lossy medium—a microscopic derivation of the operators and their commutation relations

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We derive the macroscopic electromagnetic-field and medium operators for a linear dispersive medium with a microscopic model. As an alternative to the previous treatments in the literature, we show that the canonical momentum for the macroscopic field can be chosen to be $-\epsilon_0\hat{E}$ instead of $-\hat{D}$ with the standard minimal-coupling Hamiltonian. We find that, despite the change in the field operator normalization constants, the equal-time commutators among the macroscopic electric-field, magnetic-field, and medium operators have the same values as their microscopic counterparts under a coarse-grained approximation. This preservation of the equal-time commutator is important from a fundamental standpoint, such as the preservation of microcausality for macroscopic quantities. The existence of more than one normal frequency mode at each k vector in a realistic causal-response medium is shown to be responsible for the commutator preservation. The process of macroscopic averaging is discussed in our derivation. The macroscopic field operators we derive are valid for a wide range of frequencies below, above, and around resonances. Our derivation covers the lossless, slightly lossy, and dispersionless as well as dispersive regimes of the medium. The local-field correction is also included in the formalism by inclusion of dipole–dipole interactions. Comparisons are made with other derivations of the macroscopic field operators. Using our theory, we discuss the questions of field propagation across a dielectric boundary and the decay rate of an atom embedded in a dielectric medium. We also discuss the question of squeezing in a linear dielectric medium and the extension of our theory to the case of a nonuniform medium.

1. INTRODUCTION

A proper understanding of the macroscopic electromagnetic field operators in a medium is important in quantum optics. For example, the field operator commutation relations give us the various uncertainty relations for the measurement of the electromagnetic field. The electromagnetic-field amplitude for each quantized mode is important in determining the spontaneous decay rate of an atom embedded in the medium. There have also been questions raised about the squeezing of the electromagnetic-field fluctuation in a linear dielectric medium. To provide proper answers to quantum-optic-related questions in a material medium, it is important to derive properly the macroscopic electromagnetic-field and medium operators. Although there have been a number of treatments of medium-field quantization, there has not been a direct microscopic derivation of the commutation relations for the macroscopic field and medium operators. Here we provide an understanding of the macroscopic electromagnetic-field operators based on a microscopic model, which we hope will help to resolve some questions regarding macroscopic field and medium operators that are often not fully answered in the literature.

There are many treatments in the literature on quantizing the macroscopic electromagnetic field in a linear dielectric medium. A few of these treatments are presented in Refs. 1–8. Pantell and Puthoff,[1] Marcuse,[2] Abram,[3] Yariv,[4] and Glauber–Lewenstein[5] obtained the macroscopic field operators by quantizing a macroscopic Hamiltonian or an equivalent Lagrangian. The macroscopic Hamiltonian that they used is for a linear, homogeneous, lossless, and dispersionless dielectric medium given by

$$\mathcal{H}^{\epsilon} = \int d^3x \frac{1}{2}(\epsilon \mathbf{E}^2 + \mu_0 \mathbf{H}^2). \qquad (1.1)$$

Later Hillery and Mlodinow[6] extended the macroscopic treatment to the case of a dispersionless nonlinear medium, Drummond and Carter[7] extended the macroscopic treatment to the case of soliton quantization in a dispersive nonlinear medium, and Drummond[8] extended the macroscopic treatment to the case of a general dispersive, nonlinear, and inhomogeneous medium. The treatment of Drummond and Carter[7] was successful in explaining soliton squeezing in a nonlinear dielectric medium. The treatment of Drummond[8] is interesting because of its generality but suffers from the appearance of unphysical photon modes that have to be neglected.

As is discussed below, while the above-mentioned macroscopic treatments of dielectric media are successful for their own purposes, the problem we find with the field operators derived from these treatments is that the commutation relations for the macroscopic field and medium operators suffer from a number of difficulties, including the violation of causality. The problem could be due to the association of one frequency mode with one k-vector mode in all these treatments. The refractive-index dispersion for a realistic medium obeys causality imposed by

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the Kramers–Kronig relation so that one k-vector mode is associated with more than one frequency mode. The use of a microscopic theory that automatically satisfies causality can resolve these problems and is the main subject of this paper. Specifically, we show that, if one looks at coarse-grained spacing that is large compared with the distance between two adjacent atoms, the equal-time commutators among the macroscopic electric-field, magnetic-field, and medium operators have the same values as their microscopic counterparts. Furthermore no unphysical photon modes exist in our model, and we do not foresee them in an extension of our microscopic model to the case of a nonlinear medium either. It would be interesting to see whether the existence of the unphysical photon modes in the general theory of Drummond[8] could be due to the fact that causality is not explicitly imposed in his theory.

There are many microscopic treatments of dielectric media given in the condensed-matter literature, including the pioneering work of Hopfield[9] and the recent work of Knoester and Mukamel.[10] The main subject of Hopfield's work is the problem of polaritons, and Knoester and Mukamel's work is on the decay rates of atoms embedded in a bulk dielectric. The main subject of our paper, however, is the properties of the macroscopic field and medium operators. Recently we learned that Huttner and coworkers[11,12] quantized the macroscopic electromagnetic field with an oscillator model for the medium and obtained operator results similar to those given here (we learned about this after the submission of our paper and the presentation of our results at a conference[13]). The main approach of their paper is similar to that of our paper. The minor difference between their approach and ours is that they do not use a fully microscopic method, as they have no lattice and no dipole–dipole Coulomb interaction. Also, they solved for the results by a method different from ours in that they worked on diagonalizing the Hamiltonian, while we worked on transforming the operator equations of motion. One advantage of our method may be that it permits a straightforward extension to the nonuniform medium case, as discussed in Appendix C.

Currently there are three main areas of interest in the application of the macroscopic electromagnetic field operators. The first area is the propagation of electromagnetic-field operators across a dielectric boundary. This problem was first raised by Abram.[4] Abram considered the case of a dispersionless dielectric medium. We discuss this problem briefly in Appendix A for the more general case of a dispersive dielectric medium. We show that the quantum-field-mode amplitudes we derive are consistent with the picture that the strength of the vacuum field fluctuation is altered as the field propagates from free space into the dielectric. Specifically, we show that one can derive the mode amplitudes for the macroscopic-field operators in a dielectric medium by using an argument based on the dielectric boundary conditions for the vacuum field.

The second area is the squeezing of the polariton modes in a linear dielectric postulated by Ben-Aryeh and Mann,[14] Artoni and Birman,[15] and Abram.[4] We point out in our theory that in the real ground state of the medium-field system there is no squeezing phenomena.

The third area is the decay rates of atoms embedded in a bulk dielectric. This problem was treated on the basis of macroscopic models by Pantell and Puthoff,[1] by Marcuse,[2] and by Glauber and Lewenstein[5] for the simple case of linear, homogeneous, lossless, and dispersionless medium. However, in these macroscopic models the local-field correction factor crucial for the calculation of the decay rate is derived by use of either a model of a real cavity or a virtual cavity around the embedded atom. As a result, the decay rates obtained can be very different, depending on whether a real or a virtual cavity is used. For example, Glauber and Lewenstein[5] obtained a theoretical decay rate that is different from that given by Marcuse[2] because of their implicit assumption of a real cavity. The question of whether a real or a virtual cavity should be used has been resolved by Knoester and Mukamel,[10] who used a microscopic model. They showed that the use of a virtual cavity is correct. Compared with the previous treatments, the Knoester–Mukamel theory is valid for the more general case of a dispersive medium. Later, Barnett *et al.*[16] treated this problem for the even more general case of a dispersive and lossy medium. However, in their theory the local-field correction factor was inserted into Fermi's golden-rule formula and was not derived from a microscopic model. In Appendix D we apply our formalism to obtain the decay rate by using an operator Langevin equation approach instead of the master-equation approach used by Knoester and Mukamel. In our theory the local-field correction factor is introduced with a microscopic approach. The decay-rate result that we obtain agrees with that given by Knoester and Mukamel,[10] Marcuse,[2] and Barnett *et al.*[16]

There are several issues that we wish to address in this paper regarding operator properties of the macroscopic field and medium operators. To clarify the issues, let us first discuss the type of result that one would obtain with the simple macroscopic Hamiltonian given by Eq. (1.1). Using this Hamiltonian, one finds that a quantum model of the dielectric medium can be built by canonically quantizing the fields so that $\mathcal{H}^\epsilon$ becomes an operator $\hat{\mathcal{H}}^\epsilon$. Although there have been variations in the methods, they invariably end up with the following expressions for the field operators:

$$\hat{\mathbf{E}}'(z,t) = \sum_{m,\sigma} i\left(\frac{\hbar\Omega_{pm}}{2\epsilon V_Q}\right)^{1/2} \times \mathbf{e}_{m\sigma}[\hat{a}_{m\sigma}(t) - \hat{a}_{-m\sigma}^{\dagger}(t)]\exp(ik_m z), \tag{1.2}$$

$$\mu_0\hat{\mathbf{H}}'(z,t) = \sum_{m,\sigma}\left(\frac{\hbar}{2\epsilon\Omega_{pm}V_Q}\right)^{1/2} \times (i\mathbf{k}_m \times \mathbf{e}_{m\sigma})[\hat{a}_{m\sigma}(t) + \hat{a}_{-m\sigma}^{\dagger}(t)]\exp(ik_m z), \tag{1.3}$$

where for discussion we specialize to modes with $\mathbf{k}_m$ along the z direction so that $\mathbf{k}_m = k_m\mathbf{e}_z = (2\pi m/L_z)\mathbf{e}_z$ in the sum. This specialization is denoted by primes on the field operators. In our notation, $\Omega_{pm} = |\mathbf{k}_m|c/n$, $\epsilon = n^2$, where n is the medium refractive index. We used $V_Q = L_x L_y L_z$ to denote the volume of quantization and $\mathbf{e}_{m\sigma}(\sigma = 1, 2)$ to denote the two polarization vectors for the mode m. Note that Eqs. (1.2) and (1.3) differ from the free-space field operators in that ϵ_0 has been replaced by ϵ, and Ω_{pm} is the physical frequency for mode $\mathbf{k}_m$ in the medium. One can show that $\hat{\mathcal{H}}^\epsilon = \Sigma_{m\sigma}\hbar\Omega_{pm}(\hat{a}_{m\sigma}^{\dagger}\hat{a}_{m\sigma} + 1/2)$, from which the time evolution of $\hat{a}_{m\sigma}$ can readily be deduced to be $\hat{a}_{m\sigma}(t) = \hat{a}_{m\sigma}(0)\exp(-i\Omega_{pm}t)$. Moreover, one can obtain the Maxwell equations. For example, Ampère's law,

$$\epsilon \frac{\partial \hat{\mathbf{E}}'(z,t)}{\partial t} = \nabla \times \hat{\mathbf{H}}'(z,t), \tag{1.4}$$

can be derived.[2] The field quantization thus seems to be self-consistent. Nevertheless, it is not clear whether a microscopic model of the medium, after proper macroscopic averaging, would give the same macroscopic field operators as in Eqs. (1.2) and (1.3). Even without a microscopic model, there are a number of questions that one can raise about Eqs. (1.1)–(1.3). For example, the operator version of Eq. (1.1) is equivalent to

$$\mathcal{H}^\epsilon = \int d^3x \tfrac{1}{2}(\hat{\mathbf{E}} \cdot \hat{\mathbf{D}} + \mu_0 \hat{\mathbf{H}}^2), \qquad \hat{\mathbf{D}} = \epsilon \hat{\mathbf{E}}.$$

The polarization operator associated with the medium (denoted $\hat{\mathbf{P}}_{\text{pol}}$), which is proportional to $\hat{\mathbf{E}}$, will not commute with the magnetic-field operator $\hat{\mathbf{H}}$. On the other hand, in a microscopic model we usually assume that the atomic operators commute with the field operators at equal time. Thus it is puzzling that $\hat{\mathbf{P}}_{\text{pol}}$ would not commute with the macroscopic magnetic-field operator. A more serious question that we wish to raise, however, has to do with the commutator between the electric- and magnetic-field operators. With Eqs. (1.2) and (1.3) the equal-time commutator between the noncommuting components of the electric and magnetic fields can be computed readily. For example, we have

$$[\hat{E}_x'(z,t), \hat{H}_y'(z',t)] = \frac{i\hbar}{\epsilon}\frac{\partial}{\partial z}\delta(z - z'). \tag{1.5}$$

We see that the commutator value is altered from its free-space value by the factor $1/\epsilon$. The dependence of this field commutator on ϵ has led many to suggest that the canonical field quantization of a macroscopic medium should be done[6] with the displacement field $\mathbf{D}$ rather than the electric field $\mathbf{E}$ [as the commutation between $\hat{\mathbf{D}} = \epsilon\hat{\mathbf{E}}$ and $\hat{\mathbf{H}}$ is independent of ϵ according to Eq. (1.5)]. It will be made clear here that such a step is not necessary. Instead, from our point of view it is the commutator given by Eq. (1.5) that is to be questioned. We find the dependence of this field commutator on a medium parameter ϵ puzzling because it is a basic property of the standard minimal-coupling QED Hamiltonian that the equal-time commutator between the microscopic electric and magnetic fields is independent of the presence of atoms. To see this property, let us consider the following standard minimal-coupling Hamiltonian describing a collection of atoms interacting with the electromagnetic field:

$$\mathcal{H}_{\text{MC}} = \sum_j \frac{1}{2m_e}[\hat{\mathbf{P}}_j(t) + e\hat{\mathbf{A}}(\mathbf{r}_j,t)]^2 + \frac{1}{2}\int d^3x[\epsilon_0 \hat{\mathbf{E}}^2(\mathbf{x},t) + \mu_0 \hat{\mathbf{H}}^2(\mathbf{x},t)] + \sum_j \hat{\phi}_j. \tag{1.6}$$

In the standard quantization procedure in which the Coulomb gauge is used, we express $\hat{\mathbf{A}}(\mathbf{x},t)$ in terms of a set of generalized coordinates $\{\hat{q}_m(t)\}$. The electric- and magnetic-field operators can be derived from $\hat{\mathbf{A}}(\mathbf{x},t)$, giving $\hat{\mathbf{E}}(\mathbf{x},t) \propto (\partial/\partial t)\hat{q}_m \equiv \dot{\hat{q}}_m$ and $\hat{\mathbf{H}}(\bar{x},t) \propto \hat{q}_m$. The generalized momentum conjugate to $\hat{q}_m$ is then given by $\hat{p}_m = \partial\hat{L}/\partial\dot{\hat{q}}_m$, where $\hat{L}$ is the Lagrangian operator corresponding to $\mathcal{H}_{\text{MC}}$. The Lagrangian has the form[17]

$$\hat{L}(\hat{\mathbf{r}}_j, \dot{\hat{\mathbf{r}}}_j, \hat{q}_m, \dot{\hat{q}}_m) = \sum_j \frac{\dot{\hat{\mathbf{r}}}_j^2}{2m_e} - \sum_j \hat{\phi}_j + e\hat{\mathbf{A}} \cdot \dot{\hat{\mathbf{r}}} + \frac{1}{2}\int d^3x(\epsilon_0 \hat{\mathbf{E}}^2 + \mu_0 \hat{\mathbf{H}}^2), \tag{1.7}$$

where

$$\dot{\hat{\mathbf{r}}}_j = (\hat{\mathbf{P}}_j + e\hat{\mathbf{A}})/m_e. \tag{1.8}$$

Since the atom–field interaction term in $\mathcal{H}_{\text{MC}}$ (or $\hat{L}$) depends only on $\hat{\mathbf{A}}(\mathbf{x},t)$ [this is not so for $\mathcal{H}^\epsilon$, in which $\tfrac{1}{2}(\epsilon - \epsilon_0)\hat{\mathbf{E}}^2$ is the interaction part], which in turn depends only on $\hat{q}_m$ (and not on $\dot{\hat{q}}_m$), the generalized momentum $\hat{p}_m(\partial L/\partial\dot{\hat{q}}_m)$ is independent of the presence of atoms and is always given by $\hat{p}_m = \dot{\hat{q}}_m$. Quantization then imposes $[\hat{p}_m, \hat{q}_n] = -i\hbar\delta_{nm}$, giving $[\dot{\hat{q}}_m, \hat{q}_n] = -i\hbar\delta_{nm}$. Since $\hat{\mathbf{E}}(\mathbf{x},t) \propto \dot{\hat{q}}_m$, the commutation between $\hat{\mathbf{E}}(\mathbf{x},t)$ and $\hat{\mathbf{H}}(\mathbf{x}',t)$ can readily be computed and be shown to have a fixed value irrespective of the presence of atoms. Thus it is clear that in the presence of interaction the microscopic electric- and magnetic-field operators in $\mathcal{H}_{\text{MC}}$ retain their free-space equal-time commutator value. For the same reason, the field variable canonical to $\hat{\mathbf{A}}$, given by $\partial\hat{L}/\partial\dot{\hat{\mathbf{A}}}$, is $-\epsilon_0\hat{\mathbf{E}}$, which is also independent of whether the medium is present.

The main theme of this paper is that the macroscopic-field operators derived with our microscopic model retain their free-space commutator value and the field variable canonical to the macroscopic $\hat{\mathbf{A}}$ remains as $-\epsilon_0\hat{\mathbf{E}}$, where $\hat{\mathbf{E}}$ is the macroscopic electric field. We show that the commutator given by Eq. (1.5) does not hold for this medium. Moreover, we show that all the macroscopic field operators commute with all the macroscopic medium operators.

Another question often raised on the literature is, How does one quantize a linear medium when there is dispersion? On the basis of an energy argument (see Appendix A), there have been suggestions that the field operators in that case should be given by[7,8]

$$\hat{\mathbf{E}}'(z,t) = \sum_{m\sigma}\left(\frac{\hbar\Omega_{pm}v_m}{2\epsilon_0 V_Q n_m c}\right)^{1/2} \times \mathbf{e}_{m\sigma}[\hat{a}_{m\sigma}(t) - \hat{a}_{-m\sigma}{}^\dagger(t)]\exp(ik_m z), \tag{1.9}$$

$$\mu_0\hat{\mathbf{H}}'(z,t) = \sum_{m\sigma}\left(\frac{\hbar v_m}{2\epsilon_0 V_Q n_m \Omega_{pm} c}\right)^{1/2} \times (i\mathbf{k}_m \times \mathbf{e}_{m\sigma})[\hat{a}_{m\sigma}(t) + \hat{a}_{-m\sigma}{}^\dagger(t)]\exp(ik_m z), \tag{1.10}$$

where v_m is the group velocity and n_m is the refractive index at frequency Ω_{pm}. Note that Eqs. (1.9) and (1.10) reduce to Eqs. (1.2) and (1.3) when there is no dispersion. We show in Appendix A that one can also obtain the mode amplitudes in Eq. (1.9) simply by using the dielectric boundary conditions as well as the energy argument.

However, such expressions for the field operators raise further questions. For example, one finds that the equal-time commutator of the fields in Eqs. (1.9) and (1.10) would, in general, not be a delta function in space because the product of the normalization constants in $\hat{\mathbf{E}}'$ and $\hat{\mathbf{H}}'$ is proportional to (v_m/n_m), which, in general, is frequency dependent. A delta-function equal-time field commutator is often required in field theory to satisfy microcausality.[18] Should one then conclude that the macroscopic field operators violate microcausality when there is dispersion?

Because the macroscopic field operators are derived in this paper from a microscopic theory, they will be valid in the dispersionless and lossless regime of low frequency, the dispersive and lossless regime far above resonances, or the dispersive and lossy regime around a resonance frequency. The results we obtain show that there is nothing wrong with the field operators of Eqs. (1.2) and (1.3) or those of Eqs. (1.9) and (1.10). The macroscopic field operators we obtain are of a form similar to Eqs. (1.9) and (1.10), except that they explicitly include the fact that for each k vector there is more than one normal-frequency mode in the case of a realistic medium. Specifically, there are two modes in our model. Our derivation shows that it is the existence of these two normal-frequency modes for each k vector that helps preserve the delta-function equal-time commutator between the macroscopic field operators, makes the field operators commute with the medium operators, and preserves the equal-time commutator between the macroscopic medium operators. The functional form of v_m and n_m in a realistic medium cannot be arbitrary and must obey causality constraints such as the Kramers–Kronig relation. We conclude that it is the causal dispersion curve for a realistic medium that answers the questions mentioned above.

In addition to resolving the various questions, our microscopic derivation of the macroscopic field operators includes the local-field correction. It also gives macroscopic field operators valid in the lossy regime around a resonance, where Langevin forces have to be included properly to preserve the operator commutations. In Appendix C we also discuss an extension for treating a nonuniform medium.

2. METHOD OF TREATMENT

The relationship among different sections of this paper is as follows: In Section 3 we consider a microscopic medium consisting of a uniform distribution of atoms. In the low-excitation limit of interest here, we approximate these atoms by quantum-harmonic oscillators. The atoms are coupled to the electromagnetic field by means of the standard minimal-coupling assumption. In Sections 4 and 5 the Hamiltonian so obtained is expressed in terms of the creation and annihilation operators for the modes of the free field and the free transverse polarization. In the condensed-matter literature, one often transforms the resulting Hamiltonian directly into a diagonal form, such as in Ref. 9. We take a different approach. First, instead of working directly with the Hamiltonian, we work with the equations of motion for the creation and annihilation operators. Second, we show that the diagonalization procedure can be carried out in three different steps.

The first step, carried out in Sections 4 and 5, is a Bogoliubov transformation of the type

$$\hat{a}_m^{(f)} \to (\mu \hat{a}_m^{(f)} + v \hat{a}_{-m}^{(f)\dagger}) \equiv \hat{a}_m,$$

$$\hat{B}_m^{(f)} \to (\mu \hat{B}_m^{(f)} + v \hat{B}_{-m}^{\dagger(f)}) \equiv \hat{B}_m,$$

where $\hat{a}_m^{(f)}$ and $\hat{B}_m^{(f)}$ are the annihilation operators for the free field and the free polarization wave, respectively. $\hat{a}_m$ and $\hat{B}_m$ are the resulting operators. We show that this transformation has the effect of giving an apparent frequency change to both the field and the polarization wave. The second step, carried out in Section 6, is a dressing transformation of the type

$$\hat{a}_m \to \hat{a}_m + i\zeta_m G(\hat{B}_m + \hat{B}_{-m}^{\dagger}) \equiv \tilde{a}_m,$$

$$\hat{B}_m \to \hat{B}_m + i\zeta_m G(\hat{a}_m + \hat{a}_{-m}^{\dagger}) \equiv \tilde{b}_m.$$

This transformation mixes the field and the medium operator. In spite of this mixing, the resulting dressed operators, denoted $\tilde{a}_m$ and $\tilde{B}_m$, respectively, can still be clearly identified with either the field or the medium. One property of this transformation is that it preserves $\hat{a}_m + \hat{a}_{-m}^{\dagger}$ and $\hat{B}_m + \hat{B}_{-m}^{\dagger}$ (i.e., $\hat{a}_m + \hat{a}_{-m}^{\dagger} \to \tilde{a}_m + \tilde{a}_{-m}^{\dagger}$, $\hat{B}_m + \hat{B}_{-m}^{\dagger} \to \tilde{B}_m + \tilde{B}_{-m}^{\dagger}$). The preservation of $\hat{a}_m + \hat{a}_{-m}^{\dagger}$ and $\hat{B}_m + \hat{B}_{-m}^{\dagger}$ results in the preservation of the form of the coordinate operators for the field and the medium, which are proportional to $\hat{a}_m + \hat{a}_{-m}^{\dagger}$ and $\hat{B}_m + \hat{B}_{-m}^{\dagger}$, respectively (the coordinate operator for the field is just the vector potential operator). The transformation does, however, change the form of the momentum operators for both the field and the medium so that they each have a partial contribution from $\{\tilde{a}_m\}$ and a partial contribution from $\{\tilde{B}_m\}$.

The net transformation generated by the above two steps removes the counterrotating terms from the Hamiltonian (i.e., bilinear products such as $\hat{a}_m^{(f)}\hat{a}_{-m}^{(f)}$, $\hat{B}_m^{(f)}\hat{B}_{-m}^{(f)}$, $\hat{a}_m^{(f)}\hat{B}_{-m}^{(f)}$, and their conjugates). It, however, preserves the physical picture of the coupling between the medium and the field in that the coupling terms stay in the form $\tilde{a}_m^{\dagger}\tilde{B}_m$ and $\tilde{B}_m^{\dagger}\tilde{a}_m$. The new $\{\tilde{a}_m\}$ and $\{\tilde{B}_m\}$ are purely positive-frequency operators. We note that they are similar to a transformation used by Drummond in a different context.[19]

In the third step, carried out in Section 9, the coupled equations for $\tilde{a}_m$ and $\tilde{B}_m$ are solved in terms of two normal modes, $\tilde{c}_m$ and $\tilde{d}_m$, which rotate at different frequencies. Operators $\hat{c}_m^{\dagger}$ and $\hat{d}_m^{\dagger}$ are the commonly known polariton creation operators, each of which evokes a partial excitation of the medium and the field. Thus each k-vector field mode $\tilde{a}_m$ is a linear combination of two normal-frequency modes $\hat{c}_m$ and $\hat{d}_m$. The same can be said of the polarization mode $\tilde{B}_m$. The existence of two normal-frequency modes for each k vector helps preserve the equal-time commutator between the macroscopic electric and magnetic field operators. The equal-time commutator between the macroscopic medium operators and the macroscopic field operators is also preserved. In our treatment the preservation of the equal-time commutator requires no extra proof, since it can be clearly seen as a direct consequence of the canonical nature of the transformation (i.e., a transformation that preserves equal-time commutators) of the free-field and free-polarization-wave operators $\hat{a}_m^{(f)}$ and $\hat{B}_m^{(f)}$ to $\hat{c}_m$ and $\hat{d}_m$. However, we note that because of the macroscopic averaging involved in solving for the mode operators (see Section 5), the macroscopic field operators are meaningful only if one looks at a distance that is large compared with the medium atomic spacing. In other words, because of the approximation, we can meaningfully say only that the equal-time commutators preserve their microscopic form until they reach the coarse-grained distance involved in the averaging. But that is exactly what is meaningful when we talk about macroscopic field operators.

We note that the vacuum state $|0\rangle_{\tilde{a},\tilde{B}}$ annihilated by $\tilde{a}_m$ and $\tilde{B}_m$ (also by $\hat{c}_m$ and $\hat{d}_m$) is the lowest state of energy for the entire medium-field system. Hence there is a net energy change when the atoms are brought close to one another from infinity. The original vacuum state $|0\rangle_{\hat{a},\hat{B}}$ annihilated by $\hat{a}_m^{(f)}$ and $\hat{B}_m^{(f)}$ is not the state of lowest energy for the entire medium-field system. The transformations in the first two steps are similar to the well-known squeezing transformation in that they create particles.[20,21] When the atoms are brought close together, the lowering of total energy creates particles. Nevertheless, in steady state the entire system should relax to its ground state $|0\rangle_{\tilde{a},\tilde{B}}$. There is thus no squeezing of the polariton modes in steady state. This is in contrast to some suggestions in the literature that these modes are squeezed.[4,14,15] It can be shown that the polariton modes are the ones detected by a detector in the medium.[22]

As is explained in Section 5, the transformations employed do not straightforwardly give rise to damping of the medium by means of spontaneous decay. A proper treatment of spontaneous decay requires careful treatment of the field modes near the atomic resonance frequency, which is complicated and beyond the scope of this paper. Instead we introduce damping to the harmonic oscillators composing the medium by means of phenomenological method that is quantum mechanically consistent. A theory with damping introduced into the medium is given in Sections 8 and 10, where the medium operator $\hat{\mathbf{x}}_\perp(\mathbf{r},t)$ is coupled to a thermal-field reservoir. Since the medium operator $\hat{\mathbf{x}}_\perp(\mathbf{r},t)$ depends solely on positive-frequency modes $\{\tilde{B}_m\}$, this treatment gives us the damping for the polarization modes $\{\tilde{B}_m\}$ directly. In Sections 7 and 9 we discuss the macroscopic Maxwell equations and the macroscopic field operators. For completeness, in Section 11 we introduce the local-field correction by including dipole–dipole interactions. A cubic lattice is assumed in this local-field treatment. We show that the local-field correction does not affect the equal-time commutators among the macroscopic field and the medium operators, which is also an obvious result of the canonical transformations employed. By showing preservation of the macroscopic field equal-time commutators, we thus reaffirm that, in a realistic causal-response medium, $-\epsilon_0\hat{\mathbf{E}}$ can be interpreted as the canonical momentum operator for the macroscopic field.

The macroscopic averaging that brings the microscopic model into the macroscopic realm is discussed in detail in Section 5. There we show that the macroscopic averaging involves neglecting modes that have wavelengths smaller than the separation between the atoms. We also show that it has no effect on $-\epsilon_0\hat{\mathbf{E}}$'s being the field momentum operator. We further point out that the macroscopic averaging is equivalent to simply replacing the sums with the integrals in the Hamiltonian.

3. MEDIUM HAMILTONIAN

We model the medium as a collection of uniformly distributed harmonic dipole oscillators. This harmonic dipole oscillator model is valid in the limit of weak excitation as discussed by Fano.[23] The resonance frequency of these oscillators in the absence of coupling with the electromagnetic field is assumed to be ω_a. The momentum and position operators of the jth oscillator at position $\mathbf{r}_j$ are denoted $\hat{\mathbf{P}}_j$ and $\hat{\mathbf{x}}_j$, respectively. We couple these dipoles to the electromagnetic field by means of the minimal-coupling prescription of replacing $\hat{\mathbf{P}}_j(t)$ with $\hat{\mathbf{P}}_j(t) + e\hat{\mathbf{A}}[\mathbf{r}_j(t),t]$, where $\hat{\mathbf{A}}$ is the vector-potential operator and e ($e > 0$) is the electron charge. The total Hamiltonian of this medium–field system is then given by

$$\hat{\mathcal{H}}_{\mathrm{TOT}}(t) = \sum_j \frac{[\hat{\mathbf{P}}_j(t) + e\hat{\mathbf{A}}_\perp(\mathbf{r}_j,t)]^2}{2m_e} + \sum_j \frac{m_e\omega_a{}^2\hat{\mathbf{x}}_j{}^2(t)}{2} + \int_{V_Q} \mathrm{d}\mathbf{r}\frac{\epsilon_0\hat{\mathbf{E}}_\perp{}^2(\mathbf{r},t) + \mu_0\hat{\mathbf{H}}_\perp{}^2(\mathbf{r},t)}{2} + \hat{\mathcal{H}}_{RC} + \hat{\mathcal{H}}_D, \tag{3.1}$$

where m_e is the electron mass, $\hat{\mathcal{H}}_{RC}$ is a Hamiltonian that couples the atoms to a thermal-field reservoir, and $\hat{\mathcal{H}}_D$ describes the dipole–dipole Coulomb interaction energy. The expressions for $\hat{\mathcal{H}}_{RC}$ *and* $\hat{\mathcal{H}}_D$ are given below. We have chosen the Coulomb gauge so that $\hat{\mathbf{A}}_\perp$, $\hat{\mathbf{E}}_\perp$, and $\hat{\mathbf{H}}_\perp$ in Eq. (3.1) are the transverse vector-potential, electric-field, and magnetic-field operators, respectively, and the Coulomb interaction between the electric charges is described by the scalar potential. Following the usual prescription of expressing $\hat{\mathbf{P}}_j$ and $\hat{\mathbf{x}}_j$ in terms of creation and annihilation operators, we have

$$\hat{\mathbf{P}}_j = \sum_\alpha i\left(\frac{m_e\hbar\tilde{\omega}}{2}\right)^{1/2}(\hat{b}_{j\alpha}{}^\dagger - \hat{b}_{j\alpha})\mathbf{e}_\alpha,$$

$$\hat{\mathbf{x}}_j = \sum_\alpha \left(\frac{\hbar}{2m_e\tilde{\omega}}\right)^{1/2}(\hat{b}_{j\alpha}{}^\dagger + \hat{b}_{j\alpha})\mathbf{e}_\alpha, \tag{3.2}$$

where $\alpha \in \{x,y,z\}$, with $\mathbf{e}_x$, $\mathbf{e}_y$, and $\mathbf{e}_z$ being the coordinate unit vectors. The operators $\{\hat{b}_{j\alpha}, \hat{b}_{j\alpha}{}^\dagger\}$ obey the commutator $[\hat{b}_{j\alpha}, \hat{b}_{k\alpha'}{}^\dagger] = \delta_{jk}\delta_{\alpha\alpha'}$ so that $[\hat{\mathbf{P}}_j\cdot\mathbf{e}_\alpha, \hat{\mathbf{x}}_k\cdot\hat{\mathbf{e}}_{\alpha'}] = -i\hbar\delta_{\alpha\alpha'}\delta_{jk}$. The variable $\tilde{\omega}$ is commonly chosen to be ω_a, but that is not necessary. In fact $\tilde{\omega}$ can take any value without affecting the commutation between $\hat{b}_{j\alpha}$ and $\hat{b}_{k\alpha'}{}^\dagger$. In the case of free-dipole oscillators the choice of $\tilde{\omega} = \omega_a$ is necessary to give $\hat{b}_{j\alpha}{}^\dagger(t)$ the negative-frequency solution $\hat{b}_{j\alpha}{}^\dagger(0)\exp(+i\omega_a)$ that makes $\hat{b}_{j\alpha}{}^\dagger(t)\hat{b}_{j\alpha}(t)$ a stationary operator. This allows one to interpret $\hat{b}_{j\alpha}{}^\dagger(t)$ as the creation operator of a long-lived quanta of excitation. However, in the presence of coupling to the electromagnetic field, the choice $\tilde{\omega} = \omega_a$ no longer gives $\hat{b}_{j\alpha}{}^\dagger(t)$ a negative-frequency solution. In such a case we shall treat $\tilde{\omega}$ as a parameter with an appropriate value to be determined later. We shall find that a different value must be chosen for $\tilde{\omega}$ in order to give $\hat{b}_{j\alpha}{}^\dagger(t)$ a negative-frequency solution [See Eq. (6.16) and the paragraph after Eq. (5.5)].

The electromagnetic field is quantized in the Coulomb gauge. In the usual method of canonical quantization of the electromagnetic field in a box of volume V_Q, one expresses the vector-potential operator $\hat{\mathbf{A}}_\perp(\mathbf{r},t)$ in terms of a set of generalized coordinate operators $\{\hat{q}_m(t)\}$ such that

$$\hat{\mathbf{A}}_\perp(\mathbf{r},t) = \frac{1}{\sqrt{\epsilon_0}}\sum_m \hat{q}_m(t)\mathbf{u}_m(\mathbf{r}). \tag{3.3}$$

Here $\{\mathbf{u}_m(\mathbf{r})\}$ is a set of complete orthonormal spatial modes defined over the quantization volume V_Q. The electric- and magnetic-field operators are then given by

$$\hat{\mathbf{E}}_\perp(\mathbf{r},t) = -\frac{\partial}{\partial t}\hat{\mathbf{A}}_\perp(\mathbf{r},t), \tag{3.4}$$

$$\mu_0\hat{\mathbf{H}}_\perp(\mathbf{r},t) = \nabla \times \hat{\mathbf{A}}_\perp(\mathbf{r},t). \tag{3.5}$$

After imposing the commutation condition

$$[\hat{p}_n(t), \hat{q}_m(t)] = -i\hbar\delta_{nm} \tag{3.6}$$

on $\{\hat{q}_m(t)\}$ and the corresponding generalized momentum operators $\{\hat{p}_m(t)\}$, one then transforms them into sets of annihilation and creation operators $\{\hat{a}_m(t)\}$ and $\{\hat{a}_m{}^\dagger(t)\}$, respectively. The transverse fields are then expressed in terms of these creations and annihilation operators as

$$\hat{\mathbf{A}}_\perp(\mathbf{r},t) = \sum_{m,\sigma} \sqrt{\hbar/\tilde{\Omega}_m}\, g_F \mathbf{e}_{m\sigma}[\hat{a}_{m\sigma}(t) + \hat{a}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{3.7}$$

$$\hat{\mathbf{E}}_\perp(\mathbf{r},t) = \sum_{m,\sigma} i\sqrt{\hbar\tilde{\Omega}_m}\, g_F \mathbf{e}_{m\sigma}[\hat{a}_{m\sigma}(t) - \hat{a}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{3.8}$$

$$\mu_0\hat{\mathbf{H}}_\perp(\mathbf{r},t) = \sum_{m,\sigma} \sqrt{\hbar/\tilde{\Omega}_m}\, g_F i\mathbf{k}_m \times \mathbf{e}_{m\sigma}[\hat{a}_{m\sigma}(t) + \hat{a}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{3.9}$$

where $g_F = (1/2\epsilon_0 V_Q)^{1/2}$. We imposed periodic boundary conditions for the spatial modes $\{\mathbf{u}_m(\mathbf{r})\}$ over the volume V_Q. For reasons similar to $\tilde{\omega}$ in Eq. (3.2) the variables $\{\tilde{\Omega}_m\}$ are also parameters to be fixed later [see Eq. (6.11)], though in free space they are usually given by $\tilde{\Omega}_m = |\mathbf{k}_m|c$. We note that, through the use of $\hat{p}_m(t) = \partial\hat{q}_m(t)/\partial t$, the electric-field operator has been expressed in terms of $\{\hat{a}_{m\sigma}(t)\}$ rather than $\{(\partial\hat{a}_{m\sigma}(t)/\partial t)\}$. Substituting Eq. (3.2) and Eqs. (3.7)–(3.9) into Eq. (3.1), we obtain

$$\hat{\mathscr{H}}_{\mathrm{TOT}} = \hat{\mathscr{H}}_M + \hat{\mathscr{H}}_{A^2} + \hat{\mathscr{H}}_F + \hat{\mathscr{H}}_{\mathrm{MF}} + \hat{\mathscr{H}}_{\mathrm{RC}}, \tag{3.10}$$

$$\begin{aligned}\hat{\mathscr{H}}_M &= \sum_j \left(\frac{\hat{\mathbf{P}}_j{}^2}{2m_e} + \frac{m_e\omega_a{}^2\hat{\mathbf{x}}_j{}^2}{2}\right) \\ &= i\hbar\sum_{j\alpha}\left[-\frac{i\tilde{\omega}}{2}\left(1+\frac{\omega_a{}^2}{\tilde{\omega}^2}\right)\left(\hat{b}_{j\alpha}{}^\dagger\hat{b}_{j\alpha}+\frac{1}{2}\right) - \frac{i\tilde{\omega}}{4}\left(\frac{\omega_a{}^2}{\tilde{\omega}^2}-1\right)(\hat{b}_{j\alpha}{}^\dagger\hat{b}_{j\alpha}{}^\dagger + \hat{b}_{j\alpha}\hat{b}_{j\alpha})\right],\end{aligned} \tag{3.11}$$

$$\begin{aligned}\hat{\mathscr{H}}_{A^2} &= \sum_j \frac{e^2\hat{A}_\perp{}^2(\mathbf{r}_j,t)}{2m_e} \\ &= i\hbar\sum_m -\frac{iG^2N}{\tilde{\Omega}_m}(\hat{a}_{m\sigma}+\hat{a}_{-m\sigma}{}^\dagger)(\hat{a}_{m\sigma}{}^\dagger+\hat{a}_{-m\sigma}),\end{aligned} \tag{3.12}$$

$$\begin{aligned}\hat{\mathscr{H}}_F &= \frac{1}{2}\int_{V_Q} d\mathbf{r}(\epsilon_0\hat{\mathbf{E}}_\perp{}^2 + \mu_0\hat{\mathbf{H}}_\perp{}^2) \\ &= i\hbar\sum_m\left[-\frac{i\tilde{\Omega}_m{}^2}{2}\left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2}+1\right)\left(\hat{a}_{m\sigma}{}^\dagger\hat{a}_{m\sigma}+\frac{1}{2}\right) - \frac{i\tilde{\Omega}_m{}^2}{4}\left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2}-1\right)(\hat{a}_{m\sigma}\hat{a}_{-m\sigma}+\hat{a}_{m\sigma}{}^\dagger\hat{a}_{-m\sigma}{}^\dagger)\right],\end{aligned} \tag{3.13}$$

$$\begin{aligned}\hat{\mathscr{H}}_{MF} &= \frac{e}{m_e}\sum_j \hat{\mathbf{P}}_j\cdot\hat{\mathbf{A}}_\perp(\mathbf{r}_j,t) \\ &= i\hbar\sum_{j\alpha}\sum_{m\sigma} G\sqrt{\tilde{\omega}/\tilde{\Omega}_m}\,\mathbf{e}_\sigma\cdot\mathbf{e}_\alpha(\hat{b}_{j\alpha}{}^\dagger - \hat{b}_{j\alpha})(\hat{a}_{m\sigma}+\hat{a}_{-m\sigma}{}^\dagger) \times\exp(i\mathbf{k}_m\cdot\mathbf{r}_j),\end{aligned} \tag{3.14}$$

where $\Omega_m \equiv |\mathbf{k}_m|c$ (not to be confused with the parameter $\tilde{\Omega}_m$) and $G = eg_Mg_F$ with $g_M = (1/2m_e)^{1/2}$

In Eq. (3.10) we may regard $\hat{\mathscr{H}}_M$ as the free-medium energy, $\hat{\mathscr{H}}_{A^2}$ as the medium plasma energy, $\hat{\mathscr{H}}_F$ as the free-energy, and $\hat{\mathscr{H}}_{\mathrm{MF}}$ as the medium-field interaction energy. The energy $\hat{\mathscr{H}}_{A^2}$ can be identified as the medium plasma energy because it is responsible for the dielectric response of the medium far above resonance frequency where the electrons behave as a free-electron plasma.[24,25] In deriving Eq. (3.12) we have taken the sum over all the dipole positions and have used the formula

$$\sum_j \exp[i(\mathbf{k}_m - \mathbf{k}_n)\cdot\mathbf{r}_j] = N\delta_{mn}, \tag{3.15}$$

where N is the total number of dipoles in volume V_Q. This formula is valid for $|\mathbf{k}_m - \mathbf{k}_n| < (N/V_Q)^{1/3}$. Before we leave this section, we note that leaving $\tilde{\Omega}_m$ as a free parameter is equivalent to making the transformation $\hat{a}_{m\alpha} \to \hat{a}_{m\sigma}{}'$, where

$$\hat{a}_{m\sigma}{}'(t) = \mu\hat{a}_{m\sigma}(t) + \nu\hat{a}_{-m\sigma}{}^\dagger(t). \tag{3.16}$$

For example, the substitution of Eq. (3.16) into

$$\hat{\mathbf{A}}_\perp(\mathbf{r},t) = \sum_{m,\sigma}\sqrt{\hbar/\Omega_m}\,g_F\mathbf{e}_{m\sigma}[\hat{a}_{m\sigma}{}'(t) + \hat{a}_{m\sigma}{}'^\dagger(t)]\exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{3.17}$$

with

$$\mu = \tfrac{1}{2}(\sqrt{\Omega_m/\tilde{\Omega}_m} + \sqrt{\tilde{\Omega}_m/\Omega_m}), \tag{3.18}$$

$$\nu = \tfrac{1}{2}(\sqrt{\Omega_m/\tilde{\Omega}_m} - \sqrt{\tilde{\Omega}_m/\Omega_m}), \tag{3.19}$$

will transform Eq. (3.17) into Eq. (3.7). Similar substitution of Eq. (3.16) into the $\hat{\mathbf{E}}$ and $\mu_0\hat{\mathbf{H}}$ operators corresponding to Eq. (3.17) will also transform them into Eqs. (3.8) and (3.9), respectively.

4. EQUATIONS OF MOTION FOR BARE OPERATORS

We shall refer to $\{\hat{a}_{m\sigma}, \hat{a}_{m\sigma}{}^\dagger\}$ and $\{\hat{b}_{j\alpha}, \hat{b}_{j\alpha}{}^\dagger\}$ as the bare creation and annihilation operators for the field and the atoms, respectively. They reduce to the free-field and free-dipole operators with the choice of $\tilde{\Omega}_m = |\mathbf{k}_m|c$ and $\tilde{\omega} = \omega_a$. The equations of motion for these bare operators can be found by use of the Heisenberg equation of motion, giving

$$\begin{aligned}\frac{\partial\hat{a}_{m\sigma}}{\partial t} &= -i\frac{\tilde{\Omega}_m}{2}\left[\left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2}+1\right)\hat{a}_{m\sigma} + \left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2}-1\right)\hat{a}_{-m\sigma}{}^\dagger\right] - \frac{2iNG^2}{\tilde{\Omega}_m}(\hat{a}_{m\sigma}+\hat{a}_{-m\sigma}{}^\dagger) \\ &\quad - \sum_{j\alpha}\sqrt{\tilde{\omega}/\tilde{\Omega}_m}\,G_{\sigma\alpha}(\hat{b}_{j\alpha}-\hat{b}_{j\alpha}{}^\dagger)\exp(-i\mathbf{k}_m\cdot\mathbf{r}_j),\end{aligned} \tag{4.1}$$

$$\begin{aligned}\frac{\partial\hat{b}_{j\alpha}}{\partial t} &= -\frac{i\tilde{\omega}}{2}\left[\left(\frac{\omega_a{}^2}{\tilde{\omega}^2}+1\right)\hat{b}_{j\alpha} + \left(\frac{\omega_a{}^2}{\tilde{\omega}^2}-1\right)\hat{b}_{j\alpha}{}^\dagger\right] \\ &\quad + \sum_{m,\sigma}\sqrt{\tilde{\omega}/\tilde{\Omega}_m}\,G_{\sigma\alpha}(\hat{a}_{m\sigma}+\hat{a}_{-m\sigma}{}^\dagger)\exp(i\mathbf{k}_m\cdot\mathbf{r}_j),\end{aligned} \tag{4.2}$$

where $G_{\sigma\alpha} = G\mathbf{e}_\sigma\cdot\mathbf{e}_\alpha$.

5. TRANSVERSE POLARIZATION WAVE OPERATORS

From Eq. (4.1) it is obvious that the photon operator $\hat{a}_{m\sigma}$ couples directly to the operator $\Sigma_{j\alpha}\mathbf{e}_\alpha \cdot \mathbf{e}_\sigma \hat{b}_{j\alpha} \exp(-i\mathbf{k}_m \cdot \mathbf{r}_j)$. Here we define

$$\hat{B}_{m\alpha}{}^{\tilde{\omega}} \equiv \frac{1}{\sqrt{N}} \sum_j \hat{b}_{j\alpha}{}^{\tilde{\omega}} \exp(-i\mathbf{k}_m \cdot \mathbf{r}_j), \tag{5.1}$$

$$\hat{B}_{m\sigma}{}^{\tilde{\omega}} \equiv \sum_\alpha \mathbf{e}_{m\sigma} \cdot \mathbf{e}_\alpha \hat{B}_{m\alpha}{}^{\tilde{\omega}}, \tag{5.2}$$

where we have put a superscript $\tilde{\omega}$ on $\hat{b}_{j\alpha}$ because the operator $\hat{b}_{j\alpha}$ is really a function of $\tilde{\omega}$, which is apparent from its definition in Eq. (3.2). This superscript will be omitted when the context is clear. The relation between $\hat{b}_{j\alpha}{}^{\tilde{\omega}}$ and $\hat{b}_{j\alpha}{}^{\tilde{\omega}'}$ for different values of $\tilde{\omega}$ and $\tilde{\omega}'$ is given in Appendix B. The operator $\hat{B}_{m\alpha}$ is the spatial Fourier transform of $\hat{b}_{j\alpha}$. Its inverse transform is given by

$$\hat{b}_{j\alpha}{}^{\tilde{\omega}} = \frac{1}{\sqrt{N}} \sum_m{}' \hat{B}_{m\alpha}{}^{\tilde{\omega}} \exp(i\mathbf{k}_m \cdot \mathbf{r}_j). \tag{5.3}$$

The prime in the preceding summation denotes a restricted sum over $\mathbf{k}_m = (2\pi m_x/L_x)\mathbf{e}_x + (2\pi m_y/L_y)\mathbf{e}_y + (2\pi m_z/L_z)\mathbf{e}_z$ with $0 \le m_x, m_y, m_z < N$ and $L_xL_yL_z = V_Q$. The operators $\{\hat{B}_{m\sigma}\}$ are the transverse components of $\{\hat{B}_{m\sigma}\}$. They have the commutation $[\hat{B}_{m\sigma}, \hat{B}_{n\sigma'}{}^\dagger] = \delta_{mn}\delta_{\sigma\sigma'}$.

In terms of $\{\hat{B}_{m\sigma}\}$ the equations of motion corresponding to Eqs. (4.1)–(4.2) are

$$\begin{aligned}\frac{\partial \hat{a}_{m\sigma}}{\partial t} = &-i\frac{\tilde{\Omega}_m}{2}\left[\left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2}+1\right)\hat{a}_{m\sigma} + \left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2}-1\right)\hat{a}_{-m\sigma}{}^\dagger\right] \\ &- \frac{2iNG^2}{\tilde{\Omega}_m}(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger) \\ &- \sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}\,G(\hat{B}_{m\sigma} - \hat{B}_{-m\sigma}{}^\dagger),\end{aligned} \tag{5.4}$$

$$\begin{aligned}\frac{\partial \hat{B}_{m\sigma}}{\partial t} = &-\frac{i\tilde{\omega}_m}{2}\left[\left(\frac{\omega_a{}^2}{\tilde{\omega}_m{}^2}+1\right)\hat{B}_{m\sigma} + \left(\frac{\omega_a{}^2}{\tilde{\omega}_m{}^2}-1\right)\hat{B}_{-m\sigma}{}^\dagger\right] \\ &+ \sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}\,G(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger),\end{aligned} \tag{5.5}$$

where we have denoted $\tilde{\omega}$ with an additional mode subscript m as $\tilde{\omega}_m$. We do so because we will choose its value later, which will be different for different modes.

We note that the last term of Eq. (5.5) is also obtained by use of Eq. (3.15), which is valid only for those $\{\hat{a}_n\}$ modes with $|\mathbf{k}_m - \mathbf{k}_n| < (N/V_Q)^{1/3}$. Thus Eq. (5.5) is in a sense not exact, as we neglected the coupling of $\hat{B}_{m\sigma}$ to modes $\{\hat{a}_{n\sigma}\}$ with $|\mathbf{k}_n| > (NV_Q)^{-1/3}$. In general these modes would be rotating at a much higher frequency so that it is a good approximation to neglect them. However, for those $\{\hat{a}_{n\sigma}\}$ modes with frequencies near the resonance frequency ω_a, their refractive index n_n can assume a large value, so that their wavelengths can be shorter than the atomic distance. Put another way, their $|\mathbf{k}_n|$ value, with $|\mathbf{k}_n| \simeq \omega_a n_n/c$, can be larger than $(NV_Q)^{-1/3}$. Although these near-resonance modes have $|\mathbf{k}_n| > (NV_Q)^{-1/3}$, it is not a good approximation to neglect them if ω_a is not very far from the frequency of $\hat{B}_{m\sigma}$ of interest given by $\tilde{\omega}_m$. Thus the right-hand side of Eq. (5.5) should really have many of these near-resonance modes. Coupling of the atomic operator to the near-resonance modes is responsible for causing radiative damping. We note that the summation over j performed on the $\exp(i\mathbf{k}_m \cdot \mathbf{r}_j)$ terms for these near-resonance modes when one transforms Eq. (4.2) to Eq. (5.5) can affect the number of near-resonance modes effectively coupled to $\hat{B}_{m\sigma}$ and can, as a result, cause the radiative damping to be altered at high enough atomic density.[26] However, a full treatment of radiative decay with these near-resonance modes is rather complicated and is beyond the scope of this paper. Hence, although these near-resonance modes are in principle not negligible, we shall nevertheless neglect them here. Instead, we shall treat the decay of $\hat{B}_{m\sigma}$ later by using a phenomenological method that is quantum mechanically consistent.

All the modes that we neglect have wavelengths shorter than the lattice constant or the separation between any two atoms. As a result we are smoothing out those field components whose spatial variations are smaller than the lattice constant. This is the process of macroscopic averaging that takes the microscopic model given by $\hat{\mathscr{H}}_{\mathrm{TOT}}$ in Section 3 to a macroscopic model. The same result of macroscopic averaging can also be obtained by replacing all the $\Sigma_j f(\mathbf{r}_j)$ in the Hamiltonian $\hat{\mathscr{H}}_{\mathrm{TOT}}$ of Eq. (3.1) with $\int d^3\mathbf{x} f(\mathbf{x})\rho_A$, where ρ_A is the atomic number density. In that case Eqs. (5.4) and (5.5) will be an exact result of the replaced Hamiltonian[27] because, under the continuous integral, terms that couple $\hat{B}_{m\sigma}$ to $\{\hat{a}_n\}$ with $|\mathbf{k}_n| > (NV_Q)^{-1/3}$ will simply drop out. It is clear that this replacement does not change the reasoning in Section 1 as to why the momentum canonical to $\hat{q}_m(t)$ is $\hat{p}_m(t) = (\partial\hat{q}_m/\partial t)$. Hence macroscopic averaging does not change the fact that the momentum canonical to $\hat{\mathbf{A}}$ is $-\epsilon_0\hat{\mathbf{E}}$. We see from this discussion that a quantum formulation of macroscopic medium can be obtained from a microscopic formalism with a simple replacement of the discrete sum over atoms with an integral. Such a replacement constitutes the process of averaging.

For later use it is also of interest to obtain the equations of motion for the operators $\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger, \hat{a}_{m\sigma} - \hat{a}_{-m\sigma}{}^\dagger$, $\hat{B}_{m\sigma} + \hat{B}_{-m\sigma}{}^\dagger$, and $\hat{B}_{m\sigma} - \hat{B}_{-m\sigma}{}^\dagger$, which can be obtained by adding Eqs. (5.4) and (5.5) to their own complex conjugated versions, giving

$$\frac{\partial}{\partial t}(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger) = -i\tilde{\Omega}_m(\hat{a}_{m\sigma} - \hat{a}_{-m\sigma}{}^\dagger), \tag{5.6}$$

$$\begin{aligned}\frac{\partial}{\partial t}(\hat{a}_{m\sigma} - \hat{a}_{-m\sigma}{}^\dagger) = &-\frac{i\Omega_m{}^2}{\tilde{\Omega}_m}(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger) \\ &- \frac{4iNG^2}{\tilde{\Omega}_m}(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger), \\ &- 2\sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}\,G(\hat{B}_{m\sigma} - \hat{B}_{-m\sigma}{}^\dagger),\end{aligned} \tag{5.7}$$

$$\begin{aligned}\frac{\partial}{\partial t}(\hat{B}_{m\sigma} + \hat{B}_{-m\sigma}{}^\dagger) = &-i\tilde{\omega}_m(\hat{B}_{m\sigma} - \hat{B}_{-m\sigma}{}^\dagger), \\ &+ 2\sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}\,G(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger),\end{aligned} \tag{5.8}$$

$$\frac{\partial}{\partial t}(\hat{B}_{m\sigma} - \hat{B}_{-m\sigma}{}^\dagger) = -\frac{i\omega_a{}^2}{\tilde{\omega}_m}(\hat{B}_{m\sigma} + \hat{B}_{-m\sigma}{}^\dagger). \tag{5.9}$$

The following identity will be of interest below:

$$\sum_{\alpha'} \hat{b}_{j\alpha'}{}^{\tilde{\omega}}(t)\mathbf{e}_{\alpha'} = \sum_{\alpha'}\sum_{m}{}' \exp(i\mathbf{k}_m\cdot\mathbf{r}_j)\hat{B}_{m\alpha'}{}^{\tilde{\omega}}(t) \times \left[\sum_{\sigma}(\mathbf{e}_{\alpha'}\cdot\mathbf{e}_{m\sigma})\mathbf{e}_{m\sigma} + (\mathbf{e}_{\alpha'}\cdot\mathbf{k}_m)\mathbf{k}_m/|\mathbf{k}_m|^2\right] = \sum_{\sigma}\sum_{m}{}' \exp(i\mathbf{k}_m\cdot\mathbf{r}_j)\hat{B}_{m\sigma}{}^{\tilde{\omega}}(t)\mathbf{e}_{m\sigma} + \sum_{m}{}' \hat{O}_{m\|}{}^{\tilde{\omega}}\mathbf{k}_m/|\mathbf{k}_m|, \quad (5.10)$$

$$\hat{O}_{m\|}{}^{\tilde{\omega}} = \sum_{\alpha'} \exp(i\mathbf{k}_m\cdot\mathbf{r}_j)\hat{B}_{m\alpha'}{}^{\tilde{\omega}}(t)(\mathbf{e}_{\alpha'}\cdot\mathbf{k}_m/|\mathbf{k}_m|), \quad (5.11)$$

where $\hat{O}_{m\|}{}^{\tilde{\omega}}$ is an operator whose equation of motion is not coupled to the transverse electromagnetic field.

6. DRESSED OPERATORS

Here we define the following dressed operators:

$$\tilde{a}_{m\sigma}{}^{\tilde{\omega}} = \hat{a}_{m\sigma}{}^{\tilde{\omega}} + i\zeta_m G(\hat{B}_{m\sigma}{}^{\tilde{\omega}} + \hat{B}_{-m\sigma}{}^{\tilde{\omega}\dagger}), \quad (6.1)$$

$$\tilde{B}_{m\sigma}{}^{\tilde{\omega}} = \hat{B}_{m\sigma}{}^{\tilde{\omega}} + i\zeta_m G(\hat{a}_{m\sigma}{}^{\tilde{\omega}} + \hat{a}_{-m\sigma}{}^{\tilde{\omega}\dagger}). \quad (6.2)$$

The superscript $\tilde{\omega}$ will be omitted when the context is clear. These operators have the following commutations:

$$[\tilde{a}_{m\sigma}, \tilde{a}_{n\sigma'}{}^{\dagger}] = [\tilde{B}_{m\sigma}, \tilde{B}_{n\sigma'}{}^{\dagger}] = \delta_{mn}\delta_{\sigma\sigma'}, \quad (6.3)$$

$$[\tilde{B}_{m\sigma}, \tilde{a}_{n\sigma'}{}^{\dagger}] = [\tilde{B}_{m\sigma}, \tilde{a}_{n\sigma'}{}^{\dagger}] = 0. \quad (6.4)$$

They have the following properties:

$$\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^{\dagger} = \tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}, \quad (6.5)$$

$$\hat{a}_{m\sigma} - \hat{a}_{-m\sigma}{}^{\dagger} = \tilde{a}_{m\sigma} - \tilde{a}_{-m\sigma}{}^{\dagger} - 2i\zeta_m G(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^{\dagger}), \quad (6.6)$$

$$\hat{B}_{m\sigma} + \hat{B}_{-m\sigma}{}^{\dagger} = \tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^{\dagger}, \quad (6.7)$$

$$\hat{B}_{m\sigma} - \hat{B}_{-m\sigma}{}^{\dagger} = \tilde{B}_{m\sigma} - \tilde{B}_{-m\sigma}{}^{\dagger} - 2i\zeta_m G(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}). \quad (6.8)$$

The equations of motion for $\tilde{a}_{m\sigma}$ is given by

$$\frac{d\tilde{a}_{m\sigma}}{dt} = -\frac{i\tilde{\Omega}_m}{2}\left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2} + 1\right)\tilde{a}_{m\sigma} - \frac{i\tilde{\Omega}_m}{2}\left(\frac{\Omega_m{}^2}{\tilde{\Omega}_m{}^2} - 1\right)\tilde{a}_{-m\sigma}{}^{\dagger} - \tilde{\Omega}_m\zeta_m G(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^{\dagger}) - \frac{2iG^2N}{\tilde{\Omega}_m}(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}) - \sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}\,G(\tilde{B}_{m\sigma} - \tilde{B}_{-m\sigma}{}^{\dagger}) + 2i\zeta_m\sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}\,G^2(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}) + \zeta_m\tilde{\omega}_m G(\tilde{B}_{m\sigma} - \tilde{B}_{-m\sigma}{}^{\dagger}) - 2i\zeta_m{}^2\tilde{\omega}_m G^2(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}) + 2i\zeta_m G^2\sqrt{\tilde{\omega}_m N/\tilde{\Omega}_m}(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}), \quad (6.9)$$

where the first three terms come from the first term of Eq. (5.4) and the use of Eq. (6.6). The fourth term comes from the second term of Eq. (5.4), which arises from the $\mathscr{H}_{A^2}$ part of the Hamiltonian. The fifth and sixth terms come from the third term of Eq. (5.4) and the use of Eq. (6.8). The seventh through ninth terms come from differentiating the $(\hat{B}_{m\sigma} + \hat{B}_{-m\sigma}{}^{\dagger})$ term in Eq. (6.1) and the use of Eqs. (5.8) and (6.8).

We want to choose the value of ζ_m such that the $\tilde{B}_{-m\sigma}{}^{\dagger}$ terms are nulled in Eq. (6.9). This gives us

$$\zeta_m = \frac{\sqrt{N\tilde{\omega}_m/\tilde{\Omega}_m}}{\tilde{\Omega}_m + \tilde{\omega}_m}. \quad (6.10)$$

The value of $\tilde{\Omega}_m$ is to be chosen so that the $\tilde{a}_{-m\sigma}{}^{\dagger}$ terms in Eq. (6.9) are nulled. After some algebra we get the condition

$$\tilde{\Omega}_m{}^2 - \Omega_m{}^2 = \frac{4NG^2\tilde{\Omega}_m{}^2}{(\tilde{\Omega}_m + \tilde{\omega}_m)^2}. \quad (6.11)$$

With these choices of ζ_m and $\tilde{\Omega}_m$, Eq. (6.9) reduces to the simple form:

$$\frac{d\tilde{a}_{m\sigma}}{dt} = -i\tilde{\Omega}_m\tilde{a}_{m\sigma} - \frac{2G\sqrt{N\tilde{\Omega}_m\tilde{\omega}_m}}{\tilde{\Omega}_m + \tilde{\omega}_m}\tilde{B}_{m\sigma}. \quad (6.12)$$

The equation of motion for $\tilde{B}_{m\sigma}$ is likewise given by

$$\frac{\partial\tilde{B}_{m\sigma}}{\partial t} = -\frac{i\tilde{\omega}_m}{2}\left(\frac{\omega_a{}^2}{\tilde{\omega}_m{}^2} + 1\right)\tilde{B}_{m\sigma} - \frac{i\tilde{\omega}_m}{2}\left(\frac{\omega_a{}^2}{\tilde{\omega}_m{}^2} - 1\right)\tilde{B}_{-m\sigma}{}^{\dagger} - \tilde{\omega}_m\zeta_m G(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}) + \sqrt{N\tilde{\omega}_m/\tilde{\Omega}_m}\,G(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^{\dagger}) + \zeta_m\tilde{\Omega}_m G(\tilde{a}_{m\sigma} - \tilde{a}_{-m\sigma}{}^{\dagger}) - 2i\zeta_m{}^2\tilde{\Omega}_m G^2(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^{\dagger}). \quad (6.13)$$

We see that the value of ζ_m given by Eq. (6.10) also leads to the cancellation of the $\tilde{a}_{-m\sigma}{}^{\dagger}$ terms in Eq. (6.13). The cancellation of the $\tilde{b}_{-m\sigma}{}^{\dagger}$ terms, however, can be achieved by choosing $\tilde{\omega}_m$ to satisfy

$$\tilde{\omega}_m{}^2 - \omega_a{}^2 = \frac{4NG^2\tilde{\omega}_m{}^2}{(\tilde{\Omega}_m + \tilde{\omega}_m)^2}. \quad (6.14)$$

Using Eqs. (6.11) and (6.14), we get the relation $\tilde{\Omega}_m{}^2(\tilde{\omega}_m{}^2 - \omega_a{}^2) = \tilde{\omega}_m{}^2(\tilde{\Omega}_m{}^2 - \Omega_m{}^2)$, which gives us $\tilde{\omega}_m = \tilde{\Omega}_m\omega_a/\Omega_m$. This can be used to eliminate $\tilde{\omega}_m$ in Eq. (6.11) and to solve for $\tilde{\Omega}_m$, giving

$$\tilde{\Omega}_m = \Omega_m\left[1 + \frac{4NG^2}{(\Omega_m + \omega_a)^2}\right]^{1/2}, \quad (6.15)$$

which can then be used to solve for $\tilde{\omega}_m$, giving

$$\tilde{\omega}_m = \omega_a\left[1 + \frac{4NG^2}{(\Omega_m + \omega_a)^2}\right]^{1/2}. \quad (6.16)$$

With the above values for ζ_m and $\tilde{\omega}_m$, Eq. (6.13) reduces to

$$\frac{d\tilde{B}_{m\sigma}}{dt} = -i\tilde{\omega}_m\tilde{B}_{m\sigma} + \frac{2G\sqrt{N\tilde{\Omega}_m\tilde{\omega}_m}}{\tilde{\Omega}_m + \tilde{\omega}_m}\tilde{a}_{m\sigma}. \quad (6.17)$$

We note that the right-hand side of Eq. (6.12) can be written in terms of $(\partial\tilde{B}_{m\sigma}/\partial t)$ by using Eq. (6.17), giving

$$\frac{\partial\tilde{a}_{m\sigma}}{\partial t} = -i\tilde{\Omega}_m\tilde{a}_{m\sigma} + i\frac{2GN\tilde{\Omega}_m}{(\tilde{\Omega}_m + \tilde{\omega}_m)^2}\tilde{a}_{m\sigma} - \frac{i2G\sqrt{N\tilde{\Omega}_m\tilde{\omega}_m}}{\tilde{\omega}_m(\tilde{\Omega}_m + \tilde{\omega}_m)}\frac{\partial\tilde{B}_{m\sigma}}{\partial t}, \tag{6.18}$$

which is used below to show Ampère's law.

7. MACROSCOPIC FIELD OPERATORS AND THE MACROSCOPIC MAXWELL EQUATIONS

It is straightforward to derive the macroscopic Maxwell equations. Before we do that, we need to express the field operators in terms of the dressed operators $\{\tilde{a}_{m\sigma}\}$ and $\{\tilde{B}_{m\sigma}\}$. Using Eqs. (3.7)–(3.9), (6.5), and (6.6), we have

$$\hat{\mathbf{A}}_\perp(\mathbf{r}, t) = \sum_{m\sigma}\sqrt{\hbar/\tilde{\Omega}_m}\, g_F\, \mathbf{e}_{m\sigma}[\tilde{a}_{m\sigma}(t) + \tilde{a}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{7.1}$$

$$\hat{\mathbf{E}}_\perp(\mathbf{r}, t) = \sum_{m\sigma}\sqrt{\hbar/\tilde{\Omega}_m}\, g_F\, \mathbf{e}_{m\sigma}\{\tilde{a}_{m\sigma}(t) - \tilde{a}_{-m\sigma}{}^\dagger(t) - 2i\zeta_m G[\tilde{B}_{m\sigma}(t) + \tilde{B}_{-m\sigma}{}^\dagger(t)]\}\exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{7.2}$$

$$\mu_0\hat{\mathbf{H}}_\perp(\mathbf{r}, t) = \sum_{m\sigma}\sqrt{\hbar/\tilde{\Omega}_m}\, g_F\, i\mathbf{k}_m \times \mathbf{e}_{m\sigma}(t) \times [\tilde{a}_{m\sigma}(t) + \tilde{a}_{-m\sigma}{}^\dagger(t)]\exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{7.3}$$

$$g_F = \sqrt{1/2\epsilon_0 V_Q}, \tag{7.4}$$

$$\tilde{\Omega}_m = |\mathbf{k}_m|c\left[1 + \frac{4NG^2}{(|\mathbf{k}_m|c + \omega_a)^2}\right]^{1/2}, \tag{7.5}$$

$$2iG\zeta_m = i\frac{(4NG^2\tilde{\omega}_m/\tilde{\Omega}_m)^{1/2}}{(\tilde{\Omega}_m + \tilde{\omega}_m)}, \tag{7.6}$$

$$\tilde{\omega}_m = \omega_a\left[1 + \frac{4NG^2}{(|\mathbf{k}_m|c + \omega_a)^2}\right]^{1/2}, \tag{7.7}$$

where we have expressed all variables in terms of the physical parameters $|\mathbf{k}_m|c$ and ω_a. We can also express the atomic operator in terms of the dressed operators by using Eqs. (3.2), (5.3), (5.10), (6,7), (6.8), and (B1)–(B5). Doing so, we have

$$\hat{\mathbf{x}}_j = \hat{\mathbf{x}}_{j\perp} + \hat{\mathbf{x}}_{j\parallel} \tag{7.8}$$

$$\hat{\mathbf{x}}_{j\perp} = \sum_{m\sigma}{}' \left(\frac{\hbar}{2m_e\tilde{\omega}_m N}\right)^{1/2}\mathbf{e}_{m\sigma}[\tilde{B}_{m\sigma}(t) + \tilde{B}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m\cdot\mathbf{r}_j), \tag{7.9}$$

$$\hat{\mathbf{x}}_{j\parallel} = \sum_{m}{}' \left(\frac{\hbar}{2m_e\tilde{\omega}_m N}\right)^{1/2}(\mathbf{k}_m/|\mathbf{k}|)(\hat{O}_{m\parallel} + \hat{O}_{m\parallel}{}^\dagger), \tag{7.10}$$

$$\hat{\mathbf{P}}_j = \hat{\mathbf{P}}_{j\perp} + \hat{\mathbf{P}}_{j\parallel}, \tag{7.11}$$

$$\hat{\mathbf{P}}_{j\perp} = \sum_{m\sigma}{}' i\left(\frac{\hbar m_e\tilde{\omega}_m}{2N}\right)^{1/2}\mathbf{e}_{m\sigma}[\tilde{B}_{-m\sigma}{}^\dagger(t) - \tilde{B}_{m\sigma}(t)]\exp(i\mathbf{k}_m\cdot\mathbf{r}_j), \tag{7.12}$$

$$\hat{\mathbf{P}}_{j\parallel} = \sum_{m}{}' i\left(\frac{\hbar m_e\tilde{\omega}_m}{2N}\right)^{1/2}(\mathbf{k}_m/|\mathbf{k}|)(\hat{O}_{m\parallel}{}^\dagger - \hat{O}_{m\parallel}). \tag{7.13}$$

We note that in Eqs. (7.1)–(7.13), $\tilde{B}_{m\sigma} = \tilde{B}_{m\sigma}{}^{\tilde{\omega}_m}$, $\hat{O}_{m\parallel} = \hat{O}_{m\parallel}{}^{\tilde{\omega}_m}$. Note that the free-field frequencies $\{\Omega_m = |\mathbf{k}_m|c\}$ and the free harmonic-oscillator frequency ω_a play similar roles in Eqs. (7.1)–(7.13). We can define continuous version of $\hat{\mathbf{x}}_j$ and $\hat{\mathbf{P}}_j$ by replacing $\mathbf{r}_j$ with the continuous position variable $\mathbf{r}$. In particular, we are interested in the continuous version of $\hat{\mathbf{x}}_{j\perp}$, and we define

$$\hat{\mathbf{x}}_\perp(\mathbf{r}, t) \equiv \sum_{m\sigma}{}' \left(\frac{\hbar}{2m_e\tilde{\omega}_m N}\right)^{1/2}\mathbf{e}_{m\sigma}[\tilde{B}_{m\sigma}(t) + \tilde{B}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m\cdot\mathbf{r}). \tag{7.14}$$

One can also define $\hat{\mathbf{P}}_\perp(\mathbf{r}, t)$ in a similar way as the continuous version of $\hat{\mathbf{P}}_{j\perp}$. We note that Eqs. (7.1)–(7.3) and Eq. (7.14) are really macroscopic operators. It is clear from the above that the macroscopic operators $\hat{\mathbf{x}}_\perp(\mathbf{r}, t)$ and $\hat{\mathbf{P}}_\perp(\mathbf{r}, t)$ retain the commutation relation of their microscopic counterpart until they reach the coarse grain defined by the smallest separation between atoms. That this is so is easily seen from the fact that their commutation is independent on the values of $\{\tilde{\omega}_m\}$. The coarse-grained nature comes in because the sum in Eq. (7.14) is only a restricted sum over small-k modes. The set of small-k modes forms a complete set only for functions with spatial variations larger than the coarse-grained spacing between atoms. With these macroscopic operators defined, it is straightforward to derive the macroscopic Maxwell equations. In fact, one can do it by using either the original Eqs. (4.1) and (4.2) or the transformed Eqs. (6.12), (6.17), and (6.18). Here we do it by using the transformed Eqs. (6.12), (6.17), and (6.18). From Eq. (6.12) we get

$$\frac{\partial}{\partial t}(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^\dagger) = -i\Omega_m(\tilde{a}_{m\sigma} - \tilde{a}_{m\sigma}{}^\dagger) - \frac{2G\sqrt{N\tilde{\omega}_m/\tilde{\Omega}_m}}{\tilde{\Omega}_m + \tilde{\omega}_m}(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^\dagger), \tag{7.15}$$

which gives us the Faraday's law[2]:

$$\frac{\partial}{\partial t}\mu_0\hat{\mathbf{H}}_\perp(\mathbf{r}, t) = -\nabla\times\hat{\mathbf{E}}_\perp(\mathbf{r}, t). \tag{7.16}$$

Using Eq. (6.18), we get

$$\begin{aligned}\frac{\partial}{\partial t}\left[\tilde{a}_{m\sigma} - \tilde{a}_{-m\sigma}{}^\dagger - i\frac{2G\sqrt{N\tilde{\omega}_m/\tilde{\Omega}_m}}{\tilde{\Omega}_m + \tilde{\omega}_m}(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^\dagger)\right] &= i\tilde{\Omega}_m\left(1 - \frac{2GN}{(\tilde{\Omega}_m + \tilde{\omega}_m)^2}\right)(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^\dagger) - i\left(\frac{4G^2N}{\tilde{\omega}_m\tilde{\Omega}_m}\right)^{1/2}\frac{\partial}{\partial t}(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^\dagger) \\ &= -i(\Omega_m{}^2/\tilde{\Omega}_m)(\tilde{a}_{m\sigma} + \tilde{a}_{-m\sigma}{}^\dagger) - i\frac{2eg_Fg_MN}{\sqrt{\tilde{\omega}_m\tilde{\Omega}_m N}}\frac{\partial}{\partial t}(\tilde{B}_{m\sigma} + \tilde{B}_{-m\sigma}{}^\dagger),\end{aligned} \tag{7.17}$$

where we have used Eq. (6.11). This gives us Ampère's law:

$$\frac{\partial}{\partial t}\epsilon_0\hat{\mathbf{E}}_\perp(\mathbf{r}, t) = \nabla\times\hat{\mathbf{H}}_\perp(\mathbf{r}, t) - \frac{N}{V_Q}\frac{\partial}{\partial t}[-e\hat{\mathbf{x}}_\perp(\mathbf{r}, t)]. \tag{7.18}$$

We note that in the dressed picture the field coordinate operator $\hat{\mathbf{A}}_\perp(\mathbf{r}, t)$ [see Eq. (7.1)] is dependent solely on $\{\tilde{a}_{m\sigma}(t)\}$ and the atomic coordinate operator $\hat{\mathbf{x}}_\perp(\mathbf{r}, t)$ [see Eq. (7.14)] is dependent solely on $\{\tilde{B}_{m\sigma}(t)\}$. Both $\{\tilde{a}_{m\sigma}(t)\}$ and $\{\tilde{B}_{m\sigma}(t)\}$ are purely positive-frequency operators, which is obvious from their equations of motion given by Eqs. (6.12) and (6.17). Hence we can interpret $\tilde{a}_{m\sigma}(t)$ as the amplitude operator for the light wave with wave vector $\bar{k}_m$ and $\tilde{B}_{m\sigma}(t)$ as the amplitude operator for the polarization wave with wave vector $\mathbf{k}_m$. We note, however, that the field momentum operator $\hat{\mathbf{E}}_\perp(\mathbf{r}, t)$ and the atomic momentum operator $\hat{\mathbf{P}}_\perp(\mathbf{r}, t)$ are dependent on both $\{\tilde{a}_{m\sigma}(t)\}$ and $\{\tilde{B}_{m\sigma}(t)\}$, showing clearly the dynamical coupling between the medium and the field.

8. DAMPING OF THE MEDIUM OPERATORS

Let us incorporate damping of the medium operators $\tilde{B}_{m\sigma}(t)$ by coupling them to separate thermal field reservoirs.[28] This would provide a simple, consistent quantum model of damping sufficient for our purpose. We assume the reservoir coupling Hamiltonian $\hat{\mathcal{H}}_{RC}$ in Eq. (3.1) to be of the following form:

$$\hat{\mathcal{H}}_{RC} = i\hbar \sum_{m\sigma} \mathcal{K}(\tilde{B}_{m\sigma}{}^\dagger \hat{\Gamma}_{m\sigma} - \tilde{B}_{m\sigma}\hat{\Gamma}_{m\sigma}{}^\dagger) + \hat{\mathcal{H}}_R \tag{8.1}$$

$$\hat{\Gamma}_{m\sigma}(t) = \sum_{l\sigma} \hat{c}_{lm\sigma}(t), \tag{8.2}$$

$$\hat{\mathcal{H}}_R = \sum_{ml\sigma} \hbar\omega_{lR} \hat{c}_{lm\sigma}{}^\dagger \hat{c}_{lm\sigma}, \tag{8.3}$$

where $\{\hat{c}_{lm\sigma}{}^\dagger, \hat{c}_{lm\sigma}\}$ are creation and annihilation operators for the reservoir quanta and $\omega_{lR} = 2\pi l/T$, with l being an integer. T is taken to be longer than any time period of interest. We note that Eq. (8.1) is a generic form. A more realistic form should be given by $\Sigma_j \hat{\mathbf{x}}_{j\perp} \cdot \hat{\mathbf{R}}_j$, with $\hat{\mathbf{R}}_j$ being a reservoir field operator.[29] Near the resonance frequency ω_a, these more realistic forms can always be reduced to the generic form of Eq. (8.1) by making the rotating-wave approximation. These results obtained with the generic form of Eq. (8.1) should be valid near resonance. If the damping is weak, then it will have little effect away from the resonance frequency, and whichever form we use will not have much effect on the off-resonance results. Hence we expect the results obtained with the generic form to be valid at all frequencies if $\gamma \ll \omega_a$. Following the standard technique in quantum optics,[28] we obtain decay terms additional to Eq. (6.17) because of the damping reservoir, giving

$$\frac{\partial \tilde{B}_{m\sigma}}{\partial t} = -i\tilde{\omega}_m \tilde{B}_{m\sigma} + \alpha_m \tilde{a}_{m\sigma} - \gamma \tilde{B}_{m\sigma} + \hat{\Gamma}_{Lm\sigma}, \tag{8.4}$$

$$\gamma = (T/2)\mathcal{K}^2, \tag{8.5}$$

$$\hat{\Gamma}_{Lm\sigma} = \sum_l \left(\frac{2\gamma}{T}\right)^{1/2} \hat{c}_{lm\sigma}(0)\exp(-i\omega_{lR}t), \tag{8.6}$$

where $\hat{\Gamma}_{Lm\sigma}$ is a Langevin force term with the commutation $[\hat{\Gamma}_{Lm\sigma}(t), \hat{\Gamma}_{Ln\sigma}{}^\dagger(t')] = 2\gamma\delta_{mn}\delta(t - t')$ and

$$\alpha_m = (2G\sqrt{\tilde{\Omega}_m \tilde{\omega}_m N})/(\tilde{\Omega}_m + \tilde{\omega}_m).$$

Equation (6.12) is then

$$\frac{\partial \tilde{a}_{m\sigma}}{\partial t} = -i\tilde{\Omega}_m \tilde{a}_{m\sigma} - \alpha_m \tilde{B}_{m\sigma}. \tag{8.7}$$

In the absence of field the damping introduced will lead to the following form of equation describing the usual damped Harmonic oscillators [by using Eqs. (7.9) and (8.4)]:

$$\frac{\partial^2 \hat{\mathbf{x}}_{j\perp}(t)}{\partial t^2} = -2\gamma \frac{\partial \hat{\mathbf{x}}_{j\perp}(t)}{\partial t} - \omega_a{}^2 \hat{\mathbf{x}}_{j\perp}(t) + \hat{\Gamma}(t), \tag{8.8}$$

where $\hat{\Gamma}(t)$ is a Langevin force operator and we have taken $\gamma \ll \omega_a$.

9. NORMAL-MODE SOLUTIONS AND THE MACROSCOPIC FIELDS

The coupled equations (8.4) and (8.7) can be solved by direct integration. Before we give the general solution, let us solve the simple lossless case in which $\gamma = 0$. In this case the coupled equations can be solved by straightforwardly diagonalizing them in terms of new normal modes. Let us define

$$\hat{c}_{m\sigma} = \tilde{\mu}_m \tilde{a}_{m\sigma} + i\tilde{\nu}_m \tilde{B}_{m\sigma}, \tag{9.1}$$

$$\hat{d}_{m\sigma} = \tilde{\mu}_m \tilde{a}_{m\sigma} - i\tilde{\nu}_m \tilde{B}_{m\sigma}, \tag{9.2}$$

$$\tilde{\mu}_m{}^2 + \tilde{\nu}_m{}^2 = 1, \tag{9.3}$$

where $\tilde{\mu}_m$ and $\tilde{\nu}_m$ are real constants. These new modes obey the commutations $[\hat{c}_{m\sigma}, \hat{c}_{n\sigma'}{}^\dagger] = \delta_{mn}\delta_{\sigma\sigma'} = [\hat{d}_{m\sigma}, \hat{d}_{n\sigma'}{}^\dagger]$ and $[\hat{c}_{m\sigma}, \hat{d}_{n\sigma'}{}^\dagger] = [\hat{c}_{m\sigma}, \hat{d}_{n\sigma'}] = 0$. When we use the equations of motion for $\hat{c}_{m\sigma}$ and $\hat{d}_{m\sigma}$, it is straightforward to show that, with the following choice of $\tilde{\mu}_m$ and $\tilde{\nu}_m$ (the sign of $\tilde{\mu}_m$ is arbitrarily chosen to be positive),

$$\tilde{\mu}_m = +\{½ + ½\,[1 - (4\alpha_m{}^2/y_m{}^2)]^{1/2}\}^{1/2} \tag{9.4}$$

$$y_m{}^2 = 4\alpha_m{}^2 + (\tilde{\omega}_m - \tilde{\Omega}_m)^2, \tag{9.5}$$

$$\tilde{\nu}_m = \frac{\alpha_m(2\tilde{\mu}_m{}^2 - 1)}{\tilde{\mu}_m(\tilde{\omega}_m - \tilde{\Omega}_m)}, \tag{9.6}$$

we will have the normal-frequency form for the equations of motion for $\hat{c}_{m\sigma}$ and $\hat{d}_{m\sigma}$:

$$\frac{\partial \hat{c}_{m\sigma}}{\partial t} = -i\Omega_{pm}{}^c \hat{c}_{m\sigma}, \tag{9.7}$$

$$\Omega_{pm}{}^c = \tilde{\Omega}_m \tilde{\mu}_m{}^2 + \tilde{\omega}_m \tilde{\nu}_m{}^2 - 2\tilde{\mu}_m \tilde{\nu}_m \alpha_m, \tag{9.8}$$

$$\frac{\partial \hat{d}_{m\sigma}}{\partial t} = -i\Omega_{pm}{}^d \hat{d}_{m\sigma}, \tag{9.9}$$

$$\Omega_{pm}{}^d = \tilde{\Omega}_m \tilde{\nu}_m{}^2 + \tilde{\omega}_m \tilde{\mu}_m{}^2 + 2\tilde{\mu}_m \tilde{\nu}_m \alpha_m. \tag{9.10}$$

Equations (9.7) and (9.9) give the solutions

$$\hat{c}_{m\sigma}(t) = \hat{c}_{m\sigma}(0)\exp(-i\Omega_{pm}{}^c t), \tag{9.11}$$

$$\hat{d}_{m\sigma}(t) = \hat{d}_{m\sigma}(0)\exp(-i\Omega_{pm}{}^d t). \tag{9.12}$$

Note that we used a subscript p to denote the physical frequencies, such as in $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$. One can show alge-

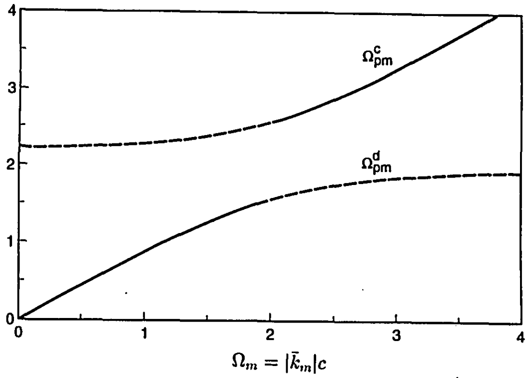


Fig. 1. Plot of $\Omega_{pm}{}^c$ (solid curves) and $\Omega_{pm}{}^d$ (dashed curves) as a function of $\Omega_m = |\mathbf{k}_m|c$ with $4NG^2 = 1$ and $\omega_a = 2$.

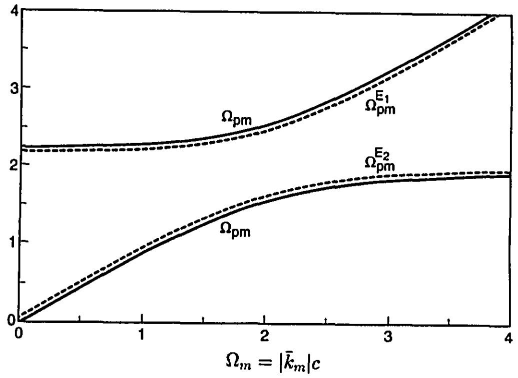


Fig. 2. Plot of Ω_{pm} (solid curves), $\Omega_{pm}{}^{E_1}$ (lower dashed curve), and $\Omega_{pm}{}^{E_2}$ (upper dashed curve) as a function of Ω_m with $4NG^2 = 1$ and $\omega_a = 2$. $\gamma = 0.5$ for the $\Omega_{pm}{}^{E_1}$ and $\Omega_{pm}{}^{E_2}$ curves.

braically that the combined function of $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$ is equivalent to Ω_{pm}, where

$$\Omega_{pm} = \Omega_m / n_m, \tag{9.13}$$

$$n_m = \sqrt{1 \times \chi_m}, \tag{9.14}$$

$$\chi_m = \frac{4NG^2}{\omega_a{}^2 - \Omega_{pm}{}^2}. \tag{9.15}$$

The function Ω_{pm} given by Eq. (9.13) is the familiar dispersion relation for a linear medium with resonance frequency ω_a. The algebra that is necessary to show that the combined function of $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$ is equivalent to Ω_{pm} is, however, very tedious. Instead, let us illustrate their equality numerically for the special case of $4N\tilde{G}^2 = 1$, and $\omega_a = 2$. First, we plot the values of $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$ as a function of $|\mathbf{k}_m|c = \Omega_m$, using Eqs. (9.8), (9.10), (9.4)–(9.6), (6.15), and (6.16). Figure 1 shows the functions $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$ as the solid curves and the dashed curves, respectively. Then we plot Ω_{pm} also as a function of Ω_m, which is shown in Fig. 2 as the solid curves. It is clear from Figs. 1 and 2 that $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$ are just separate branches of Ω_{pm}. $\Omega_{pm}{}^c$ is the photonlike branch, while $\Omega_{pm}{}^d$ is the polarization-quanta-like branch.

The inverse relations for Eqs. (9.1) and (9.2) are

$$\tilde{a}_{m\sigma} = \tilde{\mu}_m \hat{c}_{m\sigma} + \tilde{\nu}_m \hat{d}_{m\sigma}, \tag{9.16}$$

$$\tilde{B}_{m\sigma} = -i(\tilde{\nu}_m \hat{c}_{m\sigma} - \tilde{\mu}_m \hat{d}_{m\sigma}). \tag{9.17}$$

In Fig. 3 we plot the constants $\tilde{\mu}_m$ and $\tilde{\nu}_m$ as functions of Ω_m; they are shown as the solid curve and the dashed curve, respectively. We do so again for the case with $4NG^2 = 1$ and $\omega_a = 2$. We see that near resonance $|\tilde{\mu}_m| \simeq |\tilde{\nu}_m| \simeq 1/\sqrt{2}$, so that $\hat{c}_{m\sigma}$ and $\hat{d}_{m\sigma}$ have equal weightings in $\tilde{a}_{m\sigma}$. At far above or below resonance, $\tilde{\mu}_m$ is much larger than $\tilde{\nu}_m$, so that $\tilde{a}_{m\sigma}$ is determined mainly by the photonlike operator $\hat{c}_{m\sigma}$, whereas $\tilde{B}_{m\sigma}$ is determined mainly by the polarization-quanta-like operator $\hat{d}_{m\sigma}$. We can express the macroscopic field operators in terms of the normal modes, $\{\hat{c}_{m\sigma}\}$ and $\{\hat{d}_{m\sigma}\}$. For example, $\hat{\mathbf{A}}_\perp(\mathbf{r}, t)$ will be

$$\hat{\mathbf{A}}_\perp(\mathbf{r}, t) = \sum_{m\sigma} \sqrt{\hbar/\tilde{\Omega}_m}\, g_F \mathbf{e}_{m\sigma}[\tilde{\mu}_m \hat{c}_{m\sigma}(t) + \tilde{\nu}_m \hat{d}_{m\sigma}(t) + \tilde{\mu}_m \hat{c}_{-m\sigma}{}^\dagger(t) + \tilde{\nu}_m \hat{d}_{-m\sigma}{}^\dagger(t)]\exp(i\mathbf{k}_m \cdot \mathbf{r}). \tag{9.18}$$

We see that at each k vector there are two normal-mode operators under the sum of Eq. (9.18). We can rewrite Eq. (9.18) by defining

$$\hat{\mathbf{A}}_\perp{}^c(\mathbf{r}, t) = \sum_{m\sigma} C_m \sqrt{\hbar} g_F \mathbf{e}_{m\sigma}[\hat{c}_{m\sigma}(t) + \hat{c}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m \cdot \mathbf{r}), \tag{9.19}$$

$$\hat{\mathbf{A}}_\perp{}^d(\mathbf{r}, t) = \sum_{m\sigma} D_m \sqrt{\hbar} g_F \mathbf{e}_{m\sigma}[\hat{d}_{m\sigma}(t) + \hat{d}_{-m\sigma}{}^\dagger(t)] \times \exp(i\mathbf{k}_m \cdot \mathbf{r}), \tag{9.20}$$

$$C_m = \sqrt{1/\tilde{\Omega}_m}\, \tilde{\mu}_m, \tag{9.21}$$

$$D_m = \sqrt{1/\tilde{\Omega}_m}\, \tilde{\nu}_m, \tag{9.22}$$

so that

$$\hat{\mathbf{A}}_\perp(\mathbf{r}, t) = \hat{\mathbf{A}}_\perp{}^c(\mathbf{r}, t) + \hat{\mathbf{A}}_\perp{}^d(\mathbf{r}, t). \tag{9.23}$$

The normalization constants C_m and D_m depend on both $\{\tilde{\mu}_m, \tilde{\nu}_m\}$ and $\tilde{\Omega}_m$. It turns out that, like $\Omega_m{}^c$ and $\Omega_{pm}{}^d$, C_m and D_m can also be simplified. It is again algebraically complicated to show the simplification. Let us state the answer here and show the simplification graphically. We define $C_m' \equiv \sqrt{v_m{}^c/\Omega_{pm}{}^c n_m{}^c c}$, where $n_m{}^c$ is the refractive index and $v_m{}^c$ is the group velocity given by

$$(c/v_m{}^c) = n_m{}^c\left[1 + \frac{(\Omega_{pm}{}^c)^2 \chi_m{}^c}{(n_m{}^c)^2[\omega_a{}^2 - (\Omega_{pm}{}^c)^2]}\right], \tag{9.24}$$

$$n_m{}^c = \sqrt{1 + \chi_m{}^c}, \tag{9.25}$$

$$\chi_m{}^c = \frac{4NG^2}{\omega_a{}^2 - (\Omega_{pm}{}^c)^2}. \tag{9.26}$$

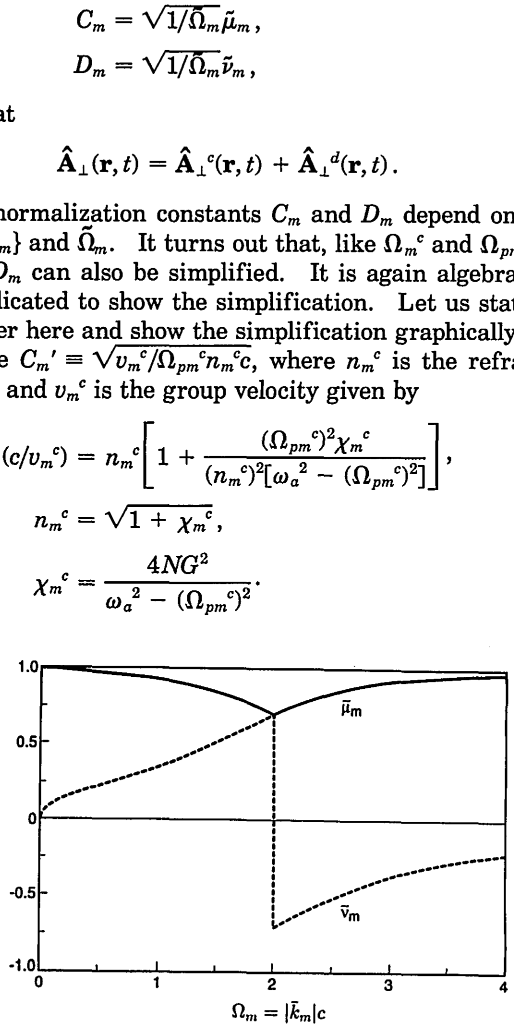
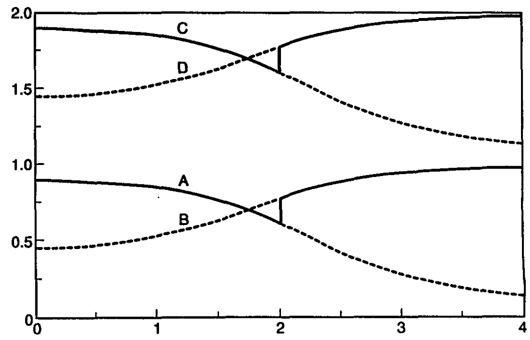
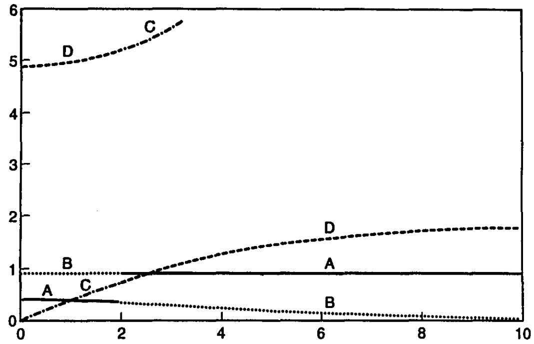


Fig. 3. Plot of $\tilde{\mu}_m$ (solid curve) and $\tilde{\nu}_m$ (dashed curve) as a function of Ω_m.



$\Omega_m = |\bar{k}_m|c$

Fig. 4. Plot of $(\Omega_{pm}{}^c)^{1/2}C_m$ (lower solid curve A), $(\Omega_{pm}{}^d)^{1/2}D_m$ (lower dashed curve B), $1 + (\Omega_{pm}{}^c)^{1/2}C_m'$ (upper solid curve C), and $1 + \sqrt{\Omega_{pm}{}^d}D_m'$ (upper dashed cruve D) as a function of Ω_m with $4NG^2 = 1$ and $\omega_a = 2$.



$\Omega_m = |\bar{k}_m|c$

Fig. 5. Plot of $(\Omega_{pm}{}^c)^{1/2}C_m$ (curve A), $\sqrt{\Omega_{pm}{}^d}D_m$ (curve B), $\Omega_{pm}{}^c$ (curve C), and $\Omega_{pm}{}^d$ (curve D) as a function of Ω_m with $4NG^2 = 20$ and $\omega_a = 2$.

Similarly, we define $D_m' \equiv [v_m{}^d/(\Omega_{pm}{}^d n_m{}^d c)]^{1/2}$, where $v_m{}^d$ and $n_m{}^d$ are the same as $v_m{}^c$ and $n_m{}^c$ but with $\Omega_{pm}{}^c$ replaced by $\Omega_{pm}{}^d$. We propose that $C_m = C_m'$ and $D_m = D_m'$. To demonstrate that, in Fig. 4 we plot $(\Omega_{pm}{}^c)^{1/2}C_m$ and $(\Omega_{pm}{}^d)^{1/2}D_m$ as a function of Ω_m for the case of $4NG^2 = 1$ and $\omega_a = 2$, which are shown as curve A (solid curve) and curve B (dashed curve), respectively. We also plot $(\Omega_{pm}{}^c)^{1/2}C_m'$ and $(\Omega_{pm}{}^d)^{1/2}D_m'$ as functions of Ω_m for the same case; they are shown displaced upward by 1 as curve C (solid curve) and curve D (dashed curve), respectively. The equality of C_m and C_m' and D_m and D_m' is then apparent. This proves the statement in Section 1 about the basic correctness of Eqs. (1.9) and (1.10) provided that we properly include two normal-frequency modes at each k vector as in Eq. (9.18). In Fig. 5 we show the case of high atomic density with $4NG^2 = 20$ and $\omega_a = 2$. Curves A and B are the coupling coefficients $(\Omega_{pm}{}^c)^{1/2}C_m$ and $(\Omega_{pm}{}^d)^{1/2}D_m$, while curves C and D are the frequency dispersion curves $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$. Note that $(\Omega_{pm}{}^c)^{1/2}C_m$ and $(\Omega_{pm}{}^d)^{1/2}D_m$ are bounded below unity. We see that there is a broad polariton frequency band gap inside which there is no mode. As is pointed out below, this photonic band gap may close up when loss is included.

Thus we have shown that the vector potential operator is given by

$$\hat{\mathbf{A}}_\perp(\mathbf{r},t) = \sum_{m\sigma}\mathbf{e}_{m\sigma}\left\{\left(\frac{\hbar v_m}{2\epsilon_0 V_Q \Omega_{pm}{}^c n_m c}\right)^{1/2}[\hat{c}_{m\sigma}(t) + \hat{c}_{-m\sigma}{}^\dagger(t)] + \left(\frac{\hbar v_m}{2\epsilon_0 V_Q \Omega_{pm}{}^d n_m c}\right)^{1/2}[\hat{d}_{m\sigma}(t) + \hat{d}_{-m\sigma}{}^\dagger(t)]\right\} \times \exp(i\mathbf{k}_m\cdot\mathbf{r}). \tag{9.27}$$

The other macroscopic operators can similarly be shown to be given by

$$\hat{\mathbf{E}}_\perp(\mathbf{r},t) = \sum_{m\sigma} i\sqrt{\hbar}g_F\mathbf{e}_{m\sigma}\{C_m'\Omega_{pm}{}^c[\hat{c}_{m\sigma}(t) - \hat{c}_{-m\sigma}{}^\dagger(t)] + D_m'\Omega_{pm}{}^d(\hat{d}_{m\sigma}(t) - \hat{d}_{-m\sigma}{}^\dagger(t)]\}\exp(i\mathbf{k}_m\cdot\mathbf{r}) \tag{9.28}$$

$$= \sum_{m\sigma} i\mathbf{e}_{m\sigma}\left\{\left(\frac{\hbar v_m\Omega_{pm}{}^c}{2\epsilon_0 V_Q n_m c}\right)^{1/2}[\hat{c}_{m\sigma}(t) - \hat{c}_{-m\sigma}{}^\dagger(t)] + \left(\frac{\hbar v_m\Omega_{pm}{}^d}{2\epsilon_0 V_Q n_m c}\right)^{1/2}[\hat{d}_{m\sigma}(t) - \hat{d}_{-m\sigma}{}^\dagger(t)]\right\}\exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{9.29}$$

$$\mu_0\hat{\mathbf{H}}_\perp(\mathbf{r},t) = \sum_{m\sigma}\sqrt{\hbar}g_F(i\mathbf{k}_m\times\mathbf{e}_{m\sigma})\{C_m'[\hat{c}_{m\sigma}(t) + \hat{c}_{-m\sigma}{}^\dagger(t)] + D_m'(\hat{d}_{m\sigma}(t) + d_{-m\sigma}{}^\dagger(t)]\}\exp(i\mathbf{k}_m\cdot\mathbf{r}) \tag{9.30}$$

$$= \sum_{m\sigma}(i\mathbf{k}_m\times\mathbf{e}_{m\sigma})\left\{\left(\frac{\hbar v_m}{2\epsilon_0 V_Q\Omega_{pm}{}^c n_m c}\right)^{1/2}[\hat{c}_{m\sigma}(t) + \hat{c}_{-m\sigma}{}^\dagger(t)] + \left(\frac{\hbar v_m}{2\epsilon_0 V_Q\Omega_{pm}{}^d n_m c}\right)^{1/2}[\hat{d}_{m\sigma}(t) + \hat{d}_{-m\sigma}{}^\dagger(t)]\right\}\exp(i\mathbf{k}_m\cdot\mathbf{r}), \tag{9.31}$$

$$\frac{-N}{V_Q}e\hat{\mathbf{x}}_\perp(\mathbf{r},t) = \epsilon_0\sum_{m\sigma} i\sqrt{\hbar}g_F\mathbf{e}_{m\sigma}\{C_m'\chi_m{}^c\Omega_{pm}{}^c[\hat{c}_{m\sigma}(t) - \hat{c}_{-m\sigma}{}^\dagger(t)] + D_m'\chi_m{}^d\Omega_{pm}{}^d[\hat{d}_{m\sigma}(t) - \hat{d}_{-m\sigma}{}^\dagger(t)]\}\exp(i\mathbf{k}_m\cdot\mathbf{r})$$

$$= \epsilon_0\sum_{m\sigma} i\mathbf{e}_{m\sigma}\left\{\left(\frac{\hbar v_m\Omega_{pm}{}^c}{2\epsilon_0 V_Q n_m c}\right)^{1/2}\chi_m{}^c[\hat{c}_{m\sigma}(t) - \hat{c}_{-m\sigma}{}^\dagger(t)] + \left(\frac{\hbar v_m\Omega_{pm}{}^d}{2\epsilon_0 V_Q n_m c}\right)^{1/2}\chi_m{}^d[\hat{d}_{m\sigma}(t) - \hat{d}_{-m\sigma}{}^\dagger(t)]\right\}\exp(i\mathbf{k}_m\cdot\mathbf{r}). \tag{9.32}$$

From Fig. 1 we see that, when $\Omega_{pm}{}^c$ is below resonance, then $\Omega_{pm}{}^d$ is above resonance and vice versa, so that $\chi_m{}^c$ and $\chi_m{}^d$ at the same $\mathbf{k}_m$ always have opposite signs [see Eq. (9.26)]. Because of this difference in signs, it can be shown numerically that the quantity $C_m'^2\chi_m{}^c\Omega_{pm}{}^c + D_m'^2\chi_m{}^d\Omega_{pm}{}^d = 0$, which is the quantity one will get by commuting $\hat{\mathbf{x}}_\perp$ with $\hat{\mathbf{H}}_\perp$. That is why the atomic operator $\hat{\mathbf{x}}_\perp(\mathbf{r},t)$ in Eq. (9.30) would commute with the magnetic-field operator $\mu_0\hat{\mathbf{H}}_\perp(\mathbf{r},t)$ at equal time. Its commutation with $\hat{\mathbf{E}}_\perp(\mathbf{r},t)$ is obvious.

10. LOSSY REGIME

In the lossy regime near resonance the coupled-mode equations (8.4) and (8.7) can be solved by direct integration by use of the Caley–Hamilton theorem, a standard procedure in linear algebra.[30] The solution is

$$\tilde{B}_{m\sigma}(t) = T_m{}^b(t)\tilde{B}_{m\sigma}(0) + U_m{}^b(t)\tilde{a}_{m\sigma}(0) + \int_0^t T_m{}^b(t - t')\hat{\Gamma}_{m\sigma}(t')\mathrm{d}t', \tag{10.1}$$

$$\tilde{a}_{m\sigma}(t) = T_m{}^a(t)\tilde{a}_{m\sigma}(0) + U_m{}^a(t)\tilde{B}_{m\sigma}(0) + \int_0^t U_m{}^a(t - t')\hat{\Gamma}_{m\sigma}(t')\mathrm{d}t', \tag{10.2}$$

$$T_m{}^b(t) = [E_1(1 + Q_m)/2 + E_2(1 - Q_m)/2], \tag{10.3}$$

$$U_m{}^b(t) = [E_1\alpha_m/(2W_m) - E_2\alpha_m/(2W_m)], \tag{10.4}$$

$$T_m{}^a(t) = [E_1(1 - Q_m)/2 + E_2(1 + Q_m)/2], \tag{10.5}$$

$$U_m{}^a(t) = [-E_1\alpha_m/(2W_m) + E_2\alpha_m/(2W_m)], \tag{10.6}$$

$$Q_m = [i(\tilde{\omega}_m - \tilde{\Omega}_m) + \gamma]/(2W_m), \tag{10.7}$$

$$W_m = \frac{iy_m}{2}\left[1 - \frac{i2\gamma(\tilde{\omega}_m - \tilde{\Omega}_m)}{{y_m}^2}\right]^{1/2} \equiv W_{mR} + iW_{mI}, \tag{10.8}$$

$$y_m = +[4{\alpha_m}^2 + (\tilde{\omega}_m - \tilde{\Omega}_m)^2 - \gamma^2]^{1/2}, \tag{10.9}$$

$$E_1 = \exp(-S_m t + W_m t), \tag{10.10}$$

$$E_2 = \exp(-S_m t - W_m t), \tag{10.11}$$

$$S_m = ½[i(\tilde{\Omega}_m + \tilde{\omega}_m) + \gamma]. \tag{10.12}$$

We can rewrite E_1 and E_2 as

$$E_1 = \exp(-i\Omega_{pm}{}^{E_1}t - \gamma_{E_1}t), \tag{10.13}$$

$$E_2 = \exp(-i\Omega_{pm}{}^{E_2}t - \gamma_{E_2}t), \tag{10.14}$$

$$\Omega_{pm}{}^{E_1} = ½(\tilde{\Omega}_m + \tilde{\omega}_m) - W_{mI}, \tag{10.15}$$

$$\gamma_{E_1} = ½\gamma - W_{mR}, \tag{10.16}$$

$$\Omega_{pm}{}^{E_2} = ½(\tilde{\Omega}_m + \tilde{\omega}_m) + W_{mI}, \tag{10.17}$$

$$\gamma_{E_2} = ½\gamma + W_{mR}. \tag{10.18}$$

The normal frequencies $\Omega_{pm}{}^{E_1}$ and $\Omega_{pm}{}^{E_2}$ are plotted as a function of Ω_m in Fig. 2 for the case of $4NG^2 = 1$, $\gamma = 0.5$, and $\omega_a = 2$. They are shown as the lower dashed curve and the upper dashed curve, respectively. The solid curves in Fig. 2 are Ω_{pm} plots of Eq. (9.13) without loss. We see that loss induces a frequency shift. The loss coefficients γ_{E_1} and γ_{E_1} are plotted as functions of Ω_{pm} in Fig. 6. This lossy limit can be shown to be well approximated by a complex χ_m of the form

$$\chi_m = \frac{4NG^2}{({\omega_a}^2 - {\Omega_{pm}}^2) - 2i\gamma\Omega_{pm}}. \tag{10.19}$$

The approximation is good when $\gamma \ll \omega_a$. We see from Fig. 2 that when there is high enough loss, the polariton frequency band gap between $\Omega_{pm}{}^{E_1}$ and $\Omega_{pm}{}^{E_2}$ curves will close up. Last, we note that modes $\Omega_{pm}{}^{E_1}$ and $\Omega_{pm}{}^{E_2}$ are not the same as modes $\Omega_{pm}{}^c$ and $\Omega_{pm}{}^d$, as they correspond to different subsections of the two branches of Ω_{pm}. Their difference is only an artifact of how their solutions are obtained and written.

The actual expressions for the various macroscopic field and medium operators can be obtained from the solutions given in this section. Using them, one can show that the macroscopic-field and medium operators in the lossy regime take on complex coefficients when expressed in terms of the normal-mode operators. In addition, there are Langevin forces in the field and medium operators, which help to preserve their commutation relations.

11. LOCAL-FIELD CORRECTIONS

Following Ref. 9, we include the local-field corrections by including the dipole–dipole Coulomb interaction energy $\hat{\mathcal{H}}_L$. We let $\hat{\mathcal{H}}_L$ be

$$\hat{\mathcal{H}}_L = i\hbar \sum_{k\neq j\alpha'}\sum_{j\alpha}\left(\frac{+ie^2}{4\pi\hbar\epsilon_0}\right) \times \left[\frac{\hat{\mathbf{x}}_{k\alpha'}\cdot\hat{\mathbf{x}}_{j\alpha}}{|\mathbf{R}_{kj}|^3} - \frac{3(\hat{\mathbf{x}}_{k\alpha'}\cdot\mathbf{R}_{kj})(\hat{\mathbf{x}}_{j\alpha}\cdot\mathbf{R}_{kj})}{|\mathbf{R}_{kj}|^5}\right], \tag{11.1}$$

where R_{kj} is the distance vector between dipole k and dipole j. We assume that the atoms are in a cubic lattice. This gives us the following term additional to $\partial\hat{B}_{m\sigma}/\partial t$:

$$\frac{\partial\hat{B}_{m\sigma}}{\partial t} \sim \frac{ie^2}{4\pi\hbar\epsilon_0}\sum_{j\alpha}\mathbf{e}_\sigma\cdot\mathbf{e}_\alpha \times \exp(-\mathbf{k}_m\cdot\mathbf{r}_j)\sum_{k\neq j}\sum_{\alpha'}\left(\frac{\hbar}{2m_e\tilde{\omega}_m}\right)\left\{\frac{(\hat{b}_{k\alpha'}{}^\dagger + \hat{b}_{k\alpha'})\mathbf{e}_{\alpha'}\cdot\mathbf{e}_\alpha}{|R_{kj}|^3} - \frac{(\hat{b}_{k\alpha'}{}^\dagger + \hat{b}_{k\alpha'})3(\mathbf{e}_\alpha\cdot\mathbf{R}_{kj})(\mathbf{e}_{\alpha'}\cdot\mathbf{R}_{kj})}{|R_{kj}|^5}\right\}. \tag{11.2}$$

We will need the following formula:

$$\sum_{j\neq k}\exp[i\mathbf{k}_m(\mathbf{r}_j - \mathbf{r}_k)]\left[\frac{\mathbf{e}_{\alpha'}\cdot\mathbf{e}_\alpha}{|\mathbf{R}_{kj}|^5} - \frac{3(\mathbf{e}_{\alpha'}\cdot\mathbf{R}_{kj})(\mathbf{e}_{\alpha'}\cdot\mathbf{R}_{kj})}{|R_{kj}|^5}\right] = \frac{4\pi}{3}\frac{N}{V_Q}\left(\delta_{\alpha'\alpha} - \frac{3k_{m\alpha}k_{m\alpha'}}{|k_m|^2}\right), \tag{11.3}$$

which is valid for a cubic lattice and for $|k_m| < 1/d$ (d is the lattice constant). This formula is well discussed in the condensed-matter literature. Inserting the formula given by Eq. (11.3) into relation (11.2), we obtain

$$\frac{\partial\hat{B}_{m\sigma}}{\partial t} \sim \frac{i}{2}\left(\frac{e^2N}{3\epsilon_0 m_e\tilde{\omega}_m V_Q}\right)(\hat{B}_{m\sigma} + \hat{B}_{-m\sigma}{}^\dagger). \tag{11.4}$$

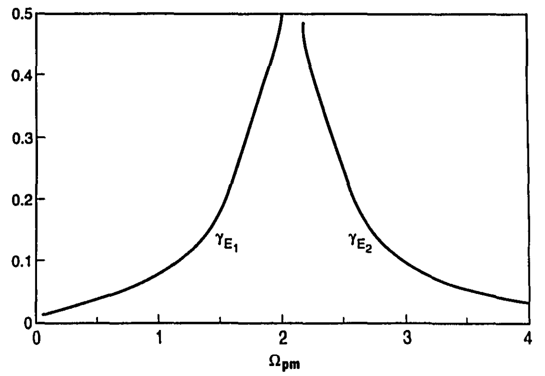


Fig. 6. Plot of γ_{E_1} (left solid curve) and γ_{E_2} (right solid curve) as a function of the frequency Ω_{pm} with $4NG^2 = 1$, $\gamma = 0.5$, and $\omega_a = 2$.

In doing the sum in Eq. (11.2) we have also used the fact that

$$\sum_{j,\alpha}\sum_{k\neq j,\alpha'}\exp(-i\mathbf{k}_m\cdot\mathbf{r}_j)$$

$$=\sum_{k,\alpha'}\sum_{j\neq k,\alpha}\exp(-i\mathbf{k}_m\cdot\mathbf{r}_j) \quad (11.5)$$

$$=\sum_{k,\alpha'}\sum_{j\neq k,\alpha}\exp[-i\mathbf{k}_m\cdot(\mathbf{r}_j-\mathbf{r}_k)]\exp(-i\mathbf{k}_m\cdot\mathbf{r}_k). \quad (11.6)$$

The extra term given by Eq. (11.4) can be generated by ${\omega_{a'}}^2$ in Eq. (5.5) if we substitute ${\omega_{a'}}^2$ for ${\omega_a}^2$ in Eq. (5.5), where

$$\begin{aligned}{\omega_{a'}}^2 &= {\omega_a}^2-\frac{e^2N}{3\epsilon_0 m_e V_Q}\\ &= {\omega_a}^2-\tfrac{4}{3}NG^2. \end{aligned}\quad (11.7)$$

Thus χ_m in Eq. (9.15) is modified to

$$\chi_m=\frac{N\chi_a}{1-(N\chi_a/3)}=\frac{4NG^2}{{\omega_a}^2-{\Omega_{pm}}^2-\tfrac{4}{3}NG^2},$$

$$\chi_a=\frac{4G^2}{({\omega_a}^2-{\Omega_{pm}}^2)}, \quad (11.8)$$

where χ_a can be recognized as the electric susceptibility of a single isolated dipole. Since the effect of local-field correction is a simple change of the value of ω_a, it does not alter the commutation relations for the field and medium operators.

12. CONCLUSIONS

In conclusion, we have given a rigorous microscopic approach for deriving all the macroscopic field and medium operators for a linear medium. We show that one can readily obtain a macroscopic Hamiltonian for the medium-field system from a microscopic Hamiltonian by simply replacing sums with integrals, and there is no need to change the field canonical momentum from electric field to displacement field. Our results show that the electric- and magnetic-field operators for a lossless dispersive medium are given basically by Eqs. (1.9) and (1.10), except that there is more than one normal-frequency mode at each k vector. (In our simplified model there are two normal-frequency modes.) In discussing the ground state of the medium considered we concluded in Section 2 that the polariton modes in a stationary linear medium are not squeezed.

We show that the macroscopic-field and medium operators retain the equal-time commutation relations of their microscopic counterparts (when below the coarse grains of the macroscopic averaging), implying that they carry the same quantum uncertainty relations as their microscopic cousins. We think that such results are a consequence of the causal property of the realistic medium that we consider, which may not be true for fictitious noncausal media. We also rigorously derive the macroscopic field operators in the dispersive and the lossy regimes. In the lossy regime Langevin-force operators are needed to preserve the operator commutations. In our model these Langevin-force operators are derived by coupling the atomic operators to a thermal-field reservoir. Although we treat a linear medium from the general discussion in this paper, we have reasons to believe that the preservation of field and medium commutators and the fact that $-\epsilon_0\hat{E}$ is the field momentum operator should hold for a nonlinear medium such as an atomic medium. They should also hold for the nonuniform medium discussed in Appendix C. We apply our theory in Appendix D to compute the decay rates of an atom embedded in a dielectric medium. The question of field propagation across an air–dielectric interface is discussed in Appendix B, where we show that the mode amplitudes for the macroscopic-field operators in a dielectric can be derived by an argument based on the dielectric boundary conditions for the vacuum field.

We note that the normalization constants that we obtained for the field operators are in agreement with those obtained by a more sophisticated macroscopic approach such as that given by Drummond.[8] This shows that there is basically nothing wrong with a macroscopic approach, provided that one carefully considers causality in order to avoid unphysical results, such as the unphysical behavior of field commutators.

APPENDIX A

We obtain the mode amplitudes in the electric-field operator [Eq. (1.9)] for a dispersive medium by an argument based on the dielectric boundary conditions for the vacuum fields. We also give a brief energy argument. For simplicity, we talk about the vacuum fields as stochastic fluctuating fields. The derivation remains valid for operator fields. Consider a dielectric boundary between air and a dielectric medium (Fig. 7). Let the frequency-dependent dielectric constant of the medium be $\epsilon(\omega)$. Let E_0 and E_i be the incident vacuum field amplitudes in air and in the medium, respectively, as shown in Fig. 7. Let E_r be the vacuum field amplitude in the medium propagating away from the boundary. These fields are assumed to be at frequency ω. We may assume a narrow frequency bandwidth $\delta\omega$ for all these fields, so that their vacuum field energies are nonzero. From the boundary conditions we have

$$E_r=TE_0+RE_i, \quad (A1)$$

where $T=2/(1+n)$, $R=(n-1)/(1+n)$, $n^2=\epsilon(\omega)$. Since the fluctuations in E_0 and E_i should be independent, the expectation value for the field amplitude square of E_r

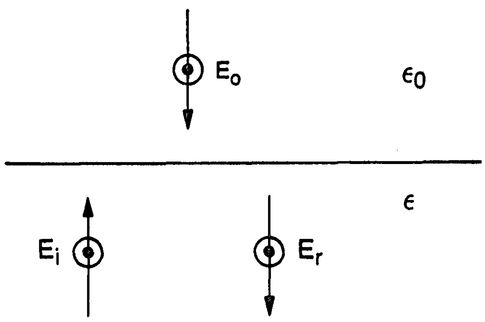


Fig. 7. Schematic diagram showing the various vacuum fields at the air–dielectric boundary.

can be easily computed, giving

$$\langle E_r^2\rangle = T^2\langle E_0^2\rangle + R^2\langle E_i^2\rangle. \tag{A2}$$

Since E_r and E_i are both steady-state vacuum fields in the medium ϵ, they must have the same fluctuating properties so that $\langle E_r^2\rangle = \langle E_i^2\rangle$; Eq. (A2) then gives

$$\langle E_r^2\rangle = (1/n)\langle E_0^2\rangle. \tag{A3}$$

Equation (A3) gives us the relation between the vacuum field amplitudes in air and in the medium. These fields are assumed to span the same frequency bandwidth. However, in Eq. (1.9) we want to compare the field amplitudes that span the same k-vector bandwidth δk. Here we denote the field amplitudes E_r' and E_0'. It is straightforward to show that they are related to E_r and E_0 by means of the respective group velocities in the medium and in air as follows:

$$\langle E_r'^2\rangle = \langle E_r^2\rangle(\mathrm{d}\omega/\mathrm{d}k)\delta k, \tag{A4}$$

$$\langle E_r'^2\rangle = \langle E_r^2\rangle c\delta k, \tag{A5}$$

where $(\mathrm{d}\omega/\mathrm{d}k)$ is the group velocity in the medium. Equations (A3) to (A5) then give

$$\langle E_r'^2\rangle = \frac{1}{n(\mathrm{d}k/\mathrm{d}\omega)/c}\langle E_0'^2\rangle. \tag{A6}$$

By taking $\langle E_0^2\rangle = \hbar\omega/(2V_Q\epsilon_0)$ in Eq. (A6) we obtain the mode amplitudes of Eq. (1.9). Note that V_Q is the volume of quantization whose value determines δk for each mode.

The energy argument can be described briefly as follows. The total energy in a particular field mode for a dispersive dielectric medium is given by[17]

$$\mathcal{H} = \int \mathrm{d}^3x \frac{1}{2}\left[\frac{\mathrm{d}\omega\epsilon}{\mathrm{d}\omega}E_m^2 + \mu_0 H_m^2\right]$$

$$\mathcal{H} = \int \mathrm{d}^3x \frac{cn_m}{v_m}\epsilon_0 E_m^2, \tag{A7}$$

where E_m and H_m are the electric and magnetic fields for that mode. The normalization constant in the electric-field operator is equal to the electric-field strength carried by half a photon energy. It can be found by setting $\mathcal{H} = \hbar\Omega_{pm}/2$, giving

$$E_m = \left(\frac{\hbar\Omega_{pm}v_m}{2\epsilon_0 V_Q n_m c}\right)^{1/2}, \tag{A8}$$

which is to be compared with Eq. (1.2).

APPENDIX B

We give the relations between $\hat{b}_{j\alpha}{}^{\bar{\omega}}$ and $\hat{b}_{j\alpha}{}^{\bar{\omega}'}$, as well as between $\hat{B}_{m\sigma}{}^{\bar{\omega}}$ and $\hat{B}_{m\sigma}{}^{\bar{\omega}'}$, with $\bar{\omega} \neq \bar{\omega}'$. From the defining equation, Eq. (3.2), and the commutation relations between $\hat{b}_{j\alpha}{}^{\bar{\omega}}$ and $\hat{b}_{j\alpha}{}^{\bar{\omega}\dagger}$, it is clear that

$$\hat{b}_{j\alpha}{}^{\bar{\omega}'} = \mu_b \hat{b}_{j\alpha}{}^{\bar{\omega}} + \nu_b \hat{b}_{j\alpha}{}^{\bar{\omega}\dagger}, \tag{B1}$$

$$\mu_b = \tfrac{1}{2}(\sqrt{\bar{\omega}'/\bar{\omega}} + \sqrt{\bar{\omega}/\bar{\omega}'}), \tag{B2}$$

$$\nu_b = \tfrac{1}{2}(\sqrt{\bar{\omega}'/\bar{\omega}} - \sqrt{\bar{\omega}/\bar{\omega}'}), \tag{B3}$$

which can be verified simply by substitution of them into Eq. (3.2). Similarly,

$$\hat{B}_{m\sigma}{}^{\bar{\omega}'} = \mu_b \hat{B}_{m\sigma}{}^{\bar{\omega}} + \nu_b \hat{B}_{-m\sigma}{}^{\bar{\omega}\dagger}, \tag{B4}$$

$$\hat{B}_{m\sigma}{}^{\bar{\omega}'} = \mu_b \hat{B}_{m\sigma}{}^{\bar{\omega}} + \nu_b \hat{B}_{-m\sigma}{}^{\bar{\omega}\dagger}. \tag{B5}$$

We note that the derivations of the above relations are similar to that of Eq. (3.16) for $\hat{a}_{m\sigma}'$.

APPENDIX C

We discuss the extension of our formalism to the case of a nonuniform medium. A complete treatment will be complicated, and we intend to point out here only the major difference between a uniform and a nonuniform medium. For a nonuniform medium the required transformations to $\tilde{a}_{m\sigma}$ and $\tilde{b}_{j\alpha}$ are

$$\begin{aligned}\tilde{a}_{m\sigma} = \hat{a}_{m\sigma} &+ \sum_{jn\sigma'} z_{mn}G^2(\hat{a}_{n\sigma'} + \hat{a}_{-n\sigma'}{}^\dagger) \\ &\times \exp[i(\mathbf{k}_n - \mathbf{k}_m)\cdot\mathbf{r}_j]\mathbf{e}_{n\sigma'}\cdot\mathbf{e}_{m\sigma} \\ &+ ab\sum_{jn\alpha} i\eta_m G(\hat{b}_{j\alpha} + \hat{b}_{j\alpha}{}^\dagger)\exp(-i\mathbf{k}_m\cdot\mathbf{r}_j)\mathbf{e}_{m\sigma}\cdot\mathbf{e}_\alpha,\end{aligned} \tag{C1}$$

$$\begin{aligned}\tilde{b}_{j\alpha} = \hat{b}_{j\alpha} &+ \sum_{\alpha'} P_\alpha G^2(\hat{b}_{j\alpha'} + \hat{b}_{j\alpha'}{}^\dagger)\mathbf{e}_\alpha\cdot\mathbf{e}_{\alpha'} \\ &+ \sum_\sigma i\eta_m G(\hat{a}_{m\sigma} + \hat{a}_{-m\sigma}{}^\dagger)\exp(i\mathbf{k}_m\cdot\mathbf{r}_j)\mathbf{e}_{m\sigma}\cdot\mathbf{e}_\alpha.\end{aligned} \tag{C2}$$

It can be shown that these transformations reduce to the transformations given earlier in this paper when the medium is uniform. With the choice

$$P_\alpha = \sum_{m\sigma}(\eta_m{}^2\Omega_{pm}/\omega_a)(\mathbf{e}_{m\sigma}\cdot\mathbf{e}_\alpha)^2, \tag{C3}$$

$$\eta_m = \sqrt{\omega_a/\bar{\Omega}_m}/(\omega_a + \Omega_{pm}), \tag{C4}$$

$$z_{mn} = \frac{2\Omega_{pm}\Omega_{pn}}{\sqrt{\bar{\Omega}_m\bar{\Omega}_n}(\Omega_{pm} + \omega_a)(\Omega_{pn} + \omega_a)(\Omega_{pn} + \Omega_{pm})}, \tag{C5}$$

one can show that to the order of G^2 the equation of motion for $\tilde{a}_{m\sigma}$ reduces to

$$\begin{aligned}\frac{\partial}{\partial t}\tilde{a}_{m\sigma} = &-i\Omega_{pm}\tilde{a}_{m\sigma} - i\Omega_{pm}\sum_{jn\sigma'}4G^2(z_{mn}\Omega_{pn}/\Omega_{pm}) \\ &\times \tilde{a}_{n\sigma'}\mathbf{e}_{n\sigma'}\cdot\mathbf{e}_{m\sigma}\exp[i(\mathbf{k}_n - \mathbf{k}_m)\cdot\mathbf{r}_j] \\ &- \frac{2\Omega_{pm}G}{(\Omega_{pm} + \omega_a)}\left(\frac{\omega_a}{\bar{\Omega}_m}\right)^{1/2}\sum_{j\alpha}\mathbf{e}_{m\sigma}\cdot\mathbf{e}_\alpha\tilde{b}_{j\alpha}\exp(-i\mathbf{k}_m\cdot\mathbf{r}_j).\end{aligned} \tag{C6}$$

We see that for the case of a uniform medium the second term on the right-hand side of Eq. (C6) will be reduced to only the $\tilde{a}_{m\sigma}$ term after the sum is carried out over j. However, for a nonuniform medium there will be terms $\tilde{a}_{n\sigma'}$ with $n \neq m$ on the right-hand side of Eq. (C6). These terms will give coupling between modes of different wave vectors and will account for Rayleigh scattering when the medium is not uniform. Such is the major feature for a nonuniform medium.

APPENDIX D

We outline the calculation of the decay rate for a two-level atom substituting at one of the lattice sites of the dielectric medium. The embedded atom is assumed to have a resonance frequency ω_A that is different from the resonance frequency of the dielectric medium. Let the upper- and the ground-level eigenfunctions of the embedded atom be $|u\rangle$ and $|g\rangle$, respectively. We define the atomic transition operator $\hat{V} \equiv |g\rangle\langle u|$, the upper-level population operator $\hat{N}_u = |u\rangle\langle u|$, and the ground-level population operator $\hat{N}_g$. The total Hamiltonian of the embedded atom plus the dielectric medium is given by

$$\hat{\mathcal{H}}(t) = \hat{\mathcal{H}}_{\mathrm{TOT}}(t) + \hat{\mathcal{H}}_A(t), \tag{D1}$$

where $\mathcal{H}_{\mathrm{TOT}}(t)$ is given by Eq. (3.10) and $\mathcal{H}_A(t)$ is given by

$$\hat{\mathcal{H}}_A(t) = \hbar\omega_A\hat{N}_u(t) + i\omega_A(\boldsymbol{\mu}\hat{V}^\dagger - \boldsymbol{\mu}^*\hat{V}) \cdot \hat{\mathbf{A}}_\perp(\mathbf{r}_A, t) + \frac{e^2}{m_e}\hat{\mathbf{A}}_\perp(\mathbf{r}_A, t) \cdot \hat{\mathbf{A}}_\perp(\mathbf{r}_A, t) + \hat{\mathcal{H}}_D(t), \tag{D2}$$

where $\boldsymbol{\mu} = \langle u|e\hat{\mathbf{x}}_A|g\rangle$, $\hat{\mathbf{x}}_A$ is the electron displacement operator for the embedded atom, $\mathbf{r}_A$ is the position vector of the embedded atom, and $\hat{\mathcal{H}}_D(t)$ is the interaction energy of the embedded atom with the dielectric medium, given by

$$\hat{\mathcal{H}}_D(t) = i\hbar\sum_{j\alpha}\left(\frac{ie^2}{4\pi\hbar\epsilon_0}\right)\left(\frac{\hat{\mathbf{x}}_A \cdot \hat{\mathbf{x}}_{j\alpha}}{|\mathbf{R}_{Aj}|^3} - \frac{3(\hat{\mathbf{x}}_A \cdot \mathbf{R}_{Aj})(\hat{\mathbf{x}}_{j\alpha} \cdot \mathbf{R}_{Aj})}{|\mathbf{R}_{Aj}|^5}\right), \tag{D3}$$

where $e\hat{\mathbf{x}}_A = [\boldsymbol{\mu}\hat{V}^\dagger(t) + \boldsymbol{\mu}^*\hat{V}(t)]$ and $|\mathbf{R}_{Aj}|$ is the distance between the medium dipole at $\mathbf{r}_j$ and the embedded atom at $\mathbf{r}_A$.

Using $\hat{\mathcal{H}}(t)$ and the expression for $\mathbf{A}_\perp(\mathbf{r}_A, t)$ given by $\hat{\mathbf{A}}_\perp{}^c(\mathbf{r}_A, t)$ of Eq. (9.20) [or $\hat{\mathbf{A}}_\perp{}^d(\mathbf{r}_A, t)$ of Eq. (9.23) depending on the frequency of ω_A], one can derive the following equations of motion by making the rotating-wave approximation:

$$\frac{\partial\hat{c}_{m\sigma}}{\partial t} = -i\Omega_{pm}\hat{c}_{m\sigma} - \frac{\mathscr{C}_{m\sigma}{}^*}{\hbar}\hat{V}\exp(-i\mathbf{k}_m \cdot \mathbf{r}_A), \tag{D4}$$

$$\frac{\partial\hat{V}}{\partial t} = -i\omega_A\hat{V} + \frac{(\hat{N}_g - \hat{N}_u)}{\hbar} \times \left\{\sum_{m,\sigma}\mathscr{C}_{m\sigma}\hat{c}_{m\sigma} + \sum_{m,\sigma}\frac{ie}{3\epsilon_0 V_Q}\left(\frac{\hbar N}{2m_e\tilde{\omega}_m}\right)^{1/2}\boldsymbol{\mu} \cdot \mathbf{e}_{m\sigma}\beta_{m\sigma}\right\} \times \exp(i\mathbf{k}_m \cdot \mathbf{r}_A), \tag{D5}$$

$$\frac{\partial\tilde{\beta}_{m\sigma}}{\partial t} = -i\tilde{\omega}_m\tilde{\beta}_{m\sigma} - \gamma\tilde{\beta}_{m\sigma} + \alpha_m\tilde{a}_{m\sigma} + \hat{\Gamma}_{m\sigma} + \sum_\alpha\sum_j\frac{\mathbf{e}_\alpha \cdot \mathbf{e}_\sigma}{\sqrt{N}}\exp(-i\mathbf{k}_m \cdot \mathbf{r}_j)\left(\frac{ie}{4\pi\hbar\epsilon_0}\right)\left(\frac{\hbar}{2m_e\tilde{\omega}_m}\right)^{1/2} \times \left\{\frac{(\boldsymbol{\mu}^*\hat{V} + \boldsymbol{\mu}\hat{V}^\dagger) \cdot \mathbf{e}_\alpha}{|\mathbf{R}_{Aj}|^3} - \frac{[3(\boldsymbol{\mu}^*\hat{V} + \boldsymbol{\mu}\hat{V}^\dagger) \cdot \mathbf{R}_{Aj}](\mathbf{e}_\alpha \cdot \mathbf{R}_{Aj})}{|\mathbf{R}_{Aj}|^5}\right\}, \tag{D6}$$

where $\hat{N}_g = \hat{V}\hat{V}^\dagger$, $\hat{N}_u = \hat{V}^\dagger\hat{V}$, $\mathscr{C}_{m\sigma} = (\mathbf{e}_{m\sigma} \cdot \boldsymbol{\mu})[(\hbar v_m\omega_A{}^2)/(2\epsilon_0 V_Q\Omega_{pm}n_m c)]^{1/2}$, v_m is the group velocity, and n_m is the medium refractive index. The frequency Ω_{pm} is the physical frequency given by $\Omega_{pm}{}^c$ (or $\Omega_{pm}{}^d$). The field modes $\tilde{\beta}_{m\sigma}$ responsible for the decay of the atom are the field modes close to the resonance frequency ω_A. The operator $\tilde{\beta}_{m\sigma}$ in Eq. (D6) can be solved adiabatically at frequency ω_A by setting $\partial\tilde{\beta}_{m\sigma}/\partial t = -i\omega_A\tilde{\beta}_{m\sigma}$, giving

$$\tilde{\beta}_{m\sigma} = -\frac{i\alpha_m\tilde{a}_{m\sigma}}{(\tilde{\omega}_m - \omega_A - i\gamma)} + \hat{\mathscr{O}}_\beta, \tag{D7}$$

where $\hat{\mathscr{O}}_\beta$ is an operator associated with the last two terms of Eq. (D6) and $\tilde{a}_{m\sigma} = \tilde{\mu}_m\hat{c}_{m\sigma}$ [see Eq. (9.17)] with $\tilde{\mu}_m = (v_m\tilde{\Omega}_m)/(\Omega_{pm}n_m c)^{1/2}$. It turns out that $\hat{\mathscr{O}}_\beta$ does not contribute to the decay rate if ω_A is far from the resonance frequency of the dielectric medium ω_a. (It gives only a frequency shift to the decaying atom.) Here we assume that ω_A is far from ω_a. By substituting Eq. (D7) into Eqs. (D4) and (D5) we can show that

$$\frac{\partial\hat{V}}{\partial t} = -i\omega_A\hat{V} + (\hat{N}_g - \hat{N}_u)\sum_{m,\sigma}\frac{\mathscr{C}_{m\sigma}}{\hbar}\hat{c}_{m\sigma}\left(1 + \frac{\chi_m}{3}\right) \times \exp(i\mathbf{k}_m \cdot \mathbf{r}_A), \tag{D8}$$

$$\frac{\partial\hat{c}_{m\sigma}}{\partial t} = -i\Omega_{pm}\hat{c}_{m\sigma} - \frac{\mathscr{C}_{m\sigma}{}^*}{\hbar}\hat{V}\left(1 + \frac{\chi_m}{3}\right)\exp(-i\mathbf{k}_m \cdot \mathbf{r}_A), \tag{D9}$$

where we have used the relation

$$\frac{\tilde{\Omega}_m 4NG^2}{(\tilde{\Omega}_m + \tilde{\omega}_m)(\tilde{\omega}_m - \Omega_{pm})} = \frac{\Omega_{pm}4NG^2}{(\omega_a{}^2 - \Omega_{pm}{}^2)} = \Omega_{pm}\chi_m.$$

In Eq. (D8) we have dropped an operator that gives only the frequency shift of $\hat{V}$. The modes of interest in Eq. (D5) are those with the physical frequency Ω_{pm} close to ω_A. One can find the decay rate of $\hat{V}$ by solving for $\tilde{a}_{m\sigma}$, $\hat{N}_u$, and $\hat{N}_g$ formally (the equations of motion for $\hat{N}_u$ and $\hat{N}_g$ can be similarly derived), then substituting them into Eq. (D8). For example, the formal solution for $\hat{c}_{m\sigma}$ is simply

$$\hat{c}_{m\sigma}(t) = \hat{c}_{m\sigma}(0)\exp(-i\Omega_{pm}t) - \int_0^t \mathrm{d}t'\frac{\mathscr{C}_{m\sigma}{}^*}{\hbar}\hat{V}(0)\left(1 + \frac{\chi_m}{3}\right) \times \exp\{-i[(\omega_A - \Omega_{pm})t' + \Omega_{pm}t]\}\exp(-i\mathbf{k}_m \cdot \mathbf{r}_A), \tag{D10}$$

where we have approximated $\hat{V}(t')$ by $\hat{V}(0)\exp(-i\omega_A t')$. The solution after we discard a frequency shift term corresponding to Lamb shift gives

$$\frac{\partial\hat{V}}{\partial t} = -i\omega_A\hat{V} - \frac{\gamma}{2}\hat{V} + \hat{\Gamma}_V. \tag{D11}$$

where $\hat{\Gamma}_V$ is a zero-mean Langevin force operator and $\gamma/2$ is given by

$$\frac{\gamma}{2} = \sum_{m,\sigma}|e_{m\sigma} \cdot \mu|^2\frac{v_m\omega_A{}^2}{2\hbar\epsilon_0 V_Q\Omega_{pm}n_m c}\left(1 + \frac{\chi_m}{3}\right)^2\pi\delta(\omega_A - \Omega_{pm}). \tag{D12}$$

This expression can be reduced to

$$\gamma = \left[\frac{2 + (\epsilon/\epsilon_0)}{3}\right]^2\sqrt{\epsilon/\epsilon_0}\,\gamma_{\mathrm{FS}}, \tag{D13}$$

where γ_{FS} is the decay rate of the same atom in free space. Note that the group velocity v_m [in Eq. (D12)] is canceled by another group-velocity term when one changes the sum Σ_m to the integral $\int d^3k = \int d\omega |dk/d\omega| (\omega/c)^2 n_m{}^2$ in Eq. (D12) to arrive at Eq. (D13). We assume that the vacuum state is the state annihilated by $\hat{c}_{m\sigma}$ (or $\hat{d}_{m\sigma}$) in this decay-rate calculation. The above decay-rate result is correct in the regime where the embedded atom sees a transparent dielectric. In the regime where the embedded atom sees an absorbing medium, the decay-rate result can be altered as a result of the $\hat{\mathcal{O}}_\beta$ term mentioned above.

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