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Electromagnetic quantization in dispersive inhomogeneous nonlinear dielectrics

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A technique of canonical quantization in a general dispersive nonlinear dielectric medium is presented. The medium can be inhomogeneous and anisotropic. The fields are expanded in a slowly varying envelope approximation to allow quantization. An arbitrary number of envelopes is included, assuming lossless propagation in each relevant frequency band. The resulting Lagrangian and Hamiltonian agree with known propagation equations and expressions for the dispersive energy. The central result of the theory is an expansion of the quantum Hamiltonian in terms of annihilation and creation operators corresponding to group-velocity photon-polariton excitations in the dielectric.

I. INTRODUCTION

The classical theory of nonlinear optics is now well established.[1] However, the rapid growth of quantum-limited laser experiments[2] has led to increased interest in the quantum properties of dielectrics. A large range of dielectric fibers and waveguides are commonly used in applications where quantum limits are approached. This leads to the central problem of quantum nonlinear optics: how to quantize a dielectric that, as well as the usual inhomogeneities and anisotropy, can also have nonlinearities and dispersion. One approach is to simply model the dielectric by its microscopic components, which in principle is always possible. This approach is seldom used since typically dielectrics have a complex internal structure.

Instead, the essential similarities of nonlinear dielectrics can be used to generate a theory of minimum complexity. This alternative procedure uses the classical field equations extended to nonlinear dielectrics, to define a set of canonical variables that can be quantized. In fact, Born and Infeld[3] treated a theoretical model of nonlinear electrodynamics as early as 1934, although this theory did not attempt to treat the general problem of a nonlinear dielectric. The first treatment of a dielectric along these lines was apparently that of Jauch and Watson,[4] who gave a canonical theory of a homogeneous linear nondispersive dielectric. With the growth of laser physics came the requirement to extend these methods to nonlinear optics.[1] However, early attempts in this direction, while incorporating the known linear theory, did not fully reproduce the nonlinear field equations.[5] Other approaches to this problem include the treatment of evanescent fields in linear media,[6] the definition of the mode operators for a linear dispersive medium,[7] and the theory of a nondispersive inhomogeneous linear dielectric.[8] An innovative treatment by Hillery and Mlodinow[9] used the displacement field as the canonical variable for nonlinear quantization, and successfully quantized a homogeneous, nondispersive medium using a similar technique to that of Born and Infeld.

In real dielectrics, there is generally a combination of dispersion, inhomogeneity, and nonlinearity. This leads to the need for a unification of the Hillery-Mlodinow nonlinear theory with other techniques incorporating dispersion and inhomogeneity. It is important to note that relatively simple techniques using effective Hamiltonians are already in wide use in quantum optics.[10] They are able to explain the nonclassical features observed in squeezing experiments,[2,10] which also occur even in optical fiber media.[11] There is now a growing recognition of the similarities between the effective Hamiltonians of extended nonlinear dielectrics[12] and those of simple quantum field theories.[13] However, it is desirable to derive the relevant Hamiltonians from a canonical quantization scheme.

A theory of nonlinear dispersive quantization in media with a nonlinear refractive index was recently given for the soliton problem.[12] Another interesting approach to this problem led to the correct paraxial field equation, but with a Hamiltonian that was different from the energy of the system.[14] While this is possible in cases of time-dependent Hamiltonians, it can lead to nonuniqueness of the quantization scheme.[15,16] An approach of dividing a medium into alternatively dispersive and nonlinear segments is also known.[17] In the present paper, a detailed three-dimensional canonical quantization technique is proposed that extends the original treatment used in the soliton problem[12] to more general dielectric structures. It is shown to lead to a Hamiltonian that generates the correct field equations, and also agrees with the known classical results for the nonlinear and dispersive energy of a dielectric.[1,18] The present treatment is able to handle the case of dispersive, inhomogeneous, and anisotropic media, as well as treating nonlinearities of any given order. The canonical field is related to the electric displacement ($\mathbf{D}$), which is also used in the quantum treatment of multipolar expansions.[19]

The resulting Hamiltonian is relatively simple, the displacement field and magnetic fields being expanded in annihilation and creation operators that correspond to photon-polaritons or dressed photons, traveling at the dielectric group velocity. It should be emphasized that the theory is a minimal quantum theory, and is necessari-

ly restricted to regions of low absorption that occur between resonances in the dielectric transmission. As dispersion is related to absorption elsewhere in frequency space, absorbing reservoirs can be added to the theory, although these effects are small in frequency ranges that do not overlap absorption bands. An interesting result in the theory is the occurrence of an extra type of boson which originates from the causality and dispersion relations in the underlying local fields. However, as these extra bosons are generally neither phase matched nor resonant, they can be neglected to a first approximation for propagation problems. In fact, they are part of more general phase noise and absorbing reservoirs, which require a detailed microscopic theory for their treatment.

While dispersion is almost universal in dielectrics, its effects are largest in the propagation of wideband radiation. The theory presented here is therefore likely to be useful in treating quantum effects that occur in communications systems or measurements using wideband quantum-limited electromagnetic radiation. Since the theory does allow an arbitrary dielectric structure, it has a variety of applications. It is well suited for questions of nonlinear input-output problems. There is no restriction to slowly varying dielectric properties. The theory can treat surface or waveguide nonlinearities. A solid lattice structure can also be included, which allows treatment of quantum local-field corrections that are important in inhomogeneous devices.

The results are presented in the following order. In Sec. II a treatment of a classical dielectric is given, introducing the nonlinear response function in terms of the displacement field **D**. The expansion parameter is therefore an inverse dielectric permittivity tensor, which is denoted $\underline{\zeta}$. In Sec. III a canonical theory of a linear dielectric is obtained, using a causal, local Lagrangian. In Sec. IV the local Lagrangian method is used as the foundation of a nonlinear canonical Lagrangian and Hamiltonian. In Sec. V the quantization of the nonlinear medium is presented, using a treatment of modes defined relative to the new Lagrangian. In Sec. VI the results are interpreted in terms of group-velocity quanta or photon-polaritons, propagating in the medium and interacting via the Hamiltonian nonlinearities.

II. CLASSICAL THEORY OF THE NONLINEAR DIELECTRIC

For simplicity, the dielectric of interest is regarded as having a uniform linear magnetic susceptibility with an arbitrary induced polarization. In addition, since propagation is of interest, the charges present are assumed to occur only in the induced dipoles of the polarization. The field equations are therefore, using the notation $\dot{D} \equiv (\partial/\partial t)D$:

$$\begin{aligned} &\nabla\cdot\mathbf{D}(t,\mathbf{x})=0 , \\ &\nabla\times\mathbf{E}(t,\mathbf{x})=-\dot{\mathbf{B}}(t,\mathbf{x}) , \\ &\nabla\cdot\mathbf{B}(t,\mathbf{x})=0 , \\ &\nabla\times\mathbf{H}(t,\mathbf{x})=\dot{\mathbf{D}}(t,\mathbf{x}) , \end{aligned} \tag{2.1}$$

where

$$\mathbf{D}(t,\mathbf{x})=\epsilon_0\mathbf{E}(t,\mathbf{x})+\mathbf{P}(t,\mathbf{x}) ,$$

$$\mathbf{B}(t,\mathbf{x})=\mu\mathbf{H}(t,\mathbf{x}) .$$

The medium is completely defined by its polarization **P**, which in general depends on the local fields. It is known that the polarization in the dipole approximation or multipolar expansion can be expressed most readily by canonically transforming to the microscopic Hamiltonian that uses the electric displacement **D** as its canonical variable.[19] Thus, a specified electric displacement generates an electronic polarization in the medium. This approach is also useful here. The total electric-field vector is expanded as a local functional of the electric displacement,[9,12,17] including the finite response time of the medium to allow for dispersive effects:

$$\begin{aligned} E_i(t,\mathbf{x})=\sum_{n\ (>0)}\int_0^\infty \underline{\zeta}^{(n)}_{i;j_1,\ldots,j_n}(\tau_1,\ldots,\tau_n,\mathbf{x}) \\ \times D_{j_l}(t-\tau_1,\mathbf{x})\cdots \\ \times D_{j_n}(t-\tau_n,\mathbf{x})d\tau_1\cdots d\tau_n . \end{aligned} \tag{2.2}$$

Here the functions $\underline{\zeta}^{(n)}$ are the nth-order electric-field response functions to the local displacement field, and the Einstein summation convention is employed whereby repeated vector indices $j_1,\ldots,j_n$ are summed over. This technique of expanding the polarization and hence the electric field in terms of the displacement appears preferable, for quantization purposes, to the more traditional technique of expanding the polarization in terms of the local electric field.[1] The displacement technique was first suggested for a nondispersive, homogeneous medium,[9] while the present theory treats the general case of a dispersive, inhomogeneous medium. The new expansion coefficients have a clear operational meaning, since the electric displacement at time $(t-\tau)$ can be calculated from a known charge deposited using a longitudinal current, across a small element of the medium. The electric field is then obtained from the measured voltage induced at later times (t), which is a uniquely defined physical observable.

In general, there could also be higher-order multipolar or magnetic response functions as well. The present theory, for simplicity, only includes electric dipole interactions that can be treated using the above expansion.

Because the response function in Eq. (2.2) is time dependent, it is not easily possible to quantize the theory in time-integral form. Instead, the response function is approximated using a slowly varying envelope approximation[14] prior to quantization, so that a Taylor-expansion series can be used. In order to achieve this, **D** is expanded in terms of a series of complex envelope functions, each of which have restricted bandwidths. The relevant nonzero central frequencies are then $\omega^{-N},\ldots,\omega^{N}$, and the displacement is expanded in complex fields $\mathcal{D}^\nu$, with $\mathcal{D}^\nu\sim e^{-i\omega^\nu t}$. Thus

$$\mathbf{D}(t,\mathbf{x})=\sum_{\nu=-N}^{N}\mathcal{D}^\nu(t,\mathbf{x}) , \tag{2.3}$$

where

$$\mathcal{D}^{-\nu}=(\mathcal{D}^{\nu})^{*} .$$

The original displacement is now replaced effectively by $N+1$ real fields, each with a restricted bandwidth. A zero-frequency field is allowed in this notation, with $\nu=0$. There are N additional fields, introduced as dynamical variables corresponding to the imaginary parts of $\mathcal{D}^{\nu}$, giving $2N+1$ real fields in total.

Substituting Eq. (2.3) into Eq. (2.2) gives the result that, for completely monochromatic fields at frequencies ω^{ν}, the electric field is just

$$E_i(t,\mathbf{x})=\sum_{n\,(>0)}\left[\sum_{\nu_1=-N}^{N}\cdots\sum_{\nu_n=-N}^{N}\xi^{(n)}_{i;j_1,\ldots,j_n}(\omega^{\nu_1},\ldots,\omega^{\nu_n};\mathbf{x})\mathcal{D}^{\nu_1}_{j_1}(t,\mathbf{x})\cdots\mathcal{D}^{\nu_n}_{j_n}(t,\mathbf{x})\right] , \tag{2.4}$$

where

$$\xi^{(n)}_{i;j_1,\ldots,j_n}(\omega^1,\ldots,\omega^n;\mathbf{x})=\int_0^{\infty}e^{i(\omega^1\tau_1+\cdots+\omega^n\tau_n)}\xi^{(n)}_{i;j_1,\ldots,j_n}(\tau_1,\ldots,\tau_n;\mathbf{x}) .$$

Clearly, Eq. (2.4) is only valid when each field $\mathcal{D}^{\nu}$ is purely monochromatic, although a combination of frequencies $\omega^1,\ldots,\omega^n$ is possible. This will be generalized later to cases of slowly varying envelopes, in which the fields $\mathcal{D}^{\nu}$ are not purely monochromatic. In these expressions the sums extend over all positive and negative frequencies, with $\omega^{-\nu}\equiv-\omega^{\nu}$. The frequency-dependent coefficients $\underline{\xi}^{(n)}(\omega^1,\ldots,\omega^n;\mathbf{x})$ correspond to those more usually measured in high-frequency regions.

Next, the time-averaged linear dispersive energy will be treated for a classical monochromatic field at nonzero frequency ω. For a permittivity $\underline{\varepsilon}(\omega,\mathbf{x})$ this can be written in terms of a complex envelope function $\mathcal{E}$:[1,18]

$$\langle H\rangle=\int_V\left[\mathcal{E}^{*}(\mathbf{x})\cdot\frac{\partial}{\partial\omega}[\omega\underline{\varepsilon}(\omega,\mathbf{x})]\cdot\mathcal{E}(\mathbf{x})+\frac{1}{2\mu}\langle\mathbf{B}(t,\mathbf{x})\cdot\mathbf{B}(t,\mathbf{x})\rangle\right]d^3\mathbf{x} , \tag{2.5}$$

where

$$\mathbf{E}(t,\mathbf{x})=2\,\mathrm{Re}[\mathcal{E}(\mathbf{x})e^{-i\omega t}] .$$

In the present notation, for monochromatic frequencies near ω^{ν}, i.e., $\omega\simeq\omega^{\nu}$, the electric displacement is given by

$$\mathcal{D}^{\nu}(t,\mathbf{x})=\underline{\varepsilon}(\omega,\mathbf{x})\cdot\mathcal{E}(\mathbf{x})e^{-i\omega t} . \tag{2.6}$$

This relation can be inverted, using Eq. (2.4), to give the νth frequency component of the electric field in terms of the electric displacement. Thus the field $\mathcal{E}(\mathbf{x})$ is

$$\mathcal{E}(\mathbf{x})=\underline{\xi}(\omega,\mathbf{x})\cdot\mathcal{D}^{\nu}(t,\mathbf{x})e^{i\omega t} ,$$

where

$$\underline{\xi}(\omega,\mathbf{x})=\underline{\xi}^{(1)}(\omega,\mathbf{x})=\int_0^{\infty}\underline{\xi}^{(1)}(\tau,\mathbf{x})e^{i\omega\tau}d\tau . \tag{2.7}$$

Combining Eqs. (2.6) and (2.7) shows that $\underline{\xi}(\omega,\mathbf{x})$ is the inverse of the usual frequency-dependent permittivity tensor, which is presumed to be nonsingular:

$$\underline{\xi}(\omega,\mathbf{x})=[\underline{\varepsilon}(\omega,\mathbf{x})]^{-1} . \tag{2.8}$$

The energy integral in Eq. (2.5) then becomes, in terms of the displacement fields,

$$\langle H\rangle=\int\left[\tfrac{1}{2}\sum_{\nu=-N}^{N}\mathcal{D}^{-\nu}(t,\mathbf{x})\cdot[\underline{\xi}(\omega,\mathbf{x})-\omega\underline{\xi}'(\omega,\mathbf{x})]\cdot\mathcal{D}^{\nu}(t,\mathbf{x})+\frac{1}{2\mu}\langle\mathbf{B}(t,\mathbf{x})\cdot\mathbf{B}(t,\mathbf{x})\rangle\right]d^3\mathbf{x} . \tag{2.9}$$

Here $\underline{\xi}'$ indicates a frequency derivative. The permittivity tensor $\underline{\varepsilon}(\omega)$ must be real, symmetric, and equal to $\underline{\varepsilon}(-\omega)$ for a nonabsorptive medium. This must clearly also hold for the inverse permittivity $\underline{\xi}(\omega)$. Here Eq. (2.9) is valid in the general case of anisotropic, inhomogeneous, and dispersive dielectrics. It is correct even when $\underline{\xi}'(\omega)$ and $\underline{\xi}(\omega)$ do not commute, as could occur if the geometry of principle directions in the dielectric depended on the frequency.

It is useful to introduce a dual potential function[9] at this point. This is defined as $\mathbf{\Lambda}$, with a similar expansion to that of Eq. (2.3), so that

$$\mathbf{D}(t,\mathbf{x})=\nabla\times\mathbf{\Lambda}(t,\mathbf{x}) ,$$

$$\mathbf{B}(t,\mathbf{x})=\mu\dot{\mathbf{\Lambda}}(t,\mathbf{x}) , \tag{2.10}$$

$$\mathbf{\Lambda}(t,\mathbf{x})=\sum_{\nu=-N}^{N}\mathbf{\Lambda}^{\nu}(t,\mathbf{x}) .$$

Next, $\underline{\xi}(\omega,\mathbf{x})$ can be expanded in a quadratic Taylor series $\underline{\xi}_{\nu}(\omega,\mathbf{x})$ near ω^{ν}, which is rearranged to demonstrate the explicit dependence on ω. Hence $\underline{\xi}(\omega,\mathbf{x})$ becomes, near $\omega=\omega^{\nu}$,

$$\underline{\xi}(\omega,\mathbf{x})\simeq\underline{\xi}_{\nu}(\omega,\mathbf{x})=\underline{\xi}_{\nu}(\mathbf{x})+\omega\underline{\xi}'_{\nu}(\mathbf{x})+\tfrac{1}{2}\omega^2\underline{\xi}''_{\nu}(\mathbf{x})+O((\omega-\omega^{\nu})^3) , \tag{2.11}$$

where

$$\underline{\zeta}_\nu(\mathbf{x})=\underline{\zeta}_\nu(0,\mathbf{x})$$
$$\equiv\underline{\zeta}(\omega^\nu,\mathbf{x})-\omega^\nu\underline{\zeta}'(\omega^\nu,\mathbf{x})+\tfrac{1}{2}(\omega^\nu)^2\underline{\zeta}''(\omega^\nu,\mathbf{x}) ,$$
$$\underline{\zeta}'_\nu(\mathbf{x})=\frac{\partial}{\partial\omega}\underline{\zeta}_\nu(\omega,\mathbf{x})\bigg|_{\omega=0}\equiv\underline{\zeta}'(\omega^\nu,\mathbf{x})-\omega^\nu\underline{\zeta}''(\omega^\nu,\mathbf{x}) ,$$
$$\underline{\zeta}''_\nu(\mathbf{x})=\frac{\partial^2}{\partial\omega^2}\underline{\zeta}_\nu(\omega,\mathbf{x})\bigg|_{\omega=0}$$
$$\equiv\underline{\zeta}''(\omega^\nu,\mathbf{x})=\frac{\partial^2}{\partial\omega^2}\underline{\zeta}(\omega,\mathbf{x})\bigg|_{\omega=\omega^\nu} .$$

The odd term $\underline{\zeta}'_\nu(\mathbf{x})$, which appears in the expansion of $\underline{\zeta}(\omega,\mathbf{x})$, does not appear in the energy integral. The new functions $\underline{\zeta}_\nu(\mathbf{x})$ obtained here are quadratic approximations to $\underline{\zeta}(\omega,\mathbf{x})$ near ω^ν with derivatives $\underline{\zeta}'_\nu(\mathbf{x})$ and $\underline{\zeta}''_\nu(\mathbf{x})$. In order to simplify the energy expression in terms of the local fields and their time derivatives, these functions are formally evaluated at $\omega=0$, in Eq. (2.11). It should be noted that even though the Taylor series itself is not valid at $\omega=0$, the field envelopes $\mathbf{\Lambda}^\nu$ are defined to only have frequency components near $\omega=\omega^\nu$. Thus Eq. (2.11) is approximately valid—up to $O((\omega-\omega^\nu)^3)$—when used with $\mathbf{\Lambda}^\nu$. It therefore has the full accuracy of the more usual Taylor expansion expressed in terms of $\omega-\omega^\nu$.

Combining (2.9) with (2.11) gives the overall result for the linear dispersive energy after making a slowly varying envelope approximation at each frequency ω^ν:

$$\langle H\rangle=\tfrac{1}{2}\sum_{\nu=-N}^{N}\int\{[\nabla\times\mathbf{\Lambda}^{-\nu}(t,\mathbf{x})]\cdot\underline{\zeta}_\nu(\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^\nu(t,\mathbf{x})]-\tfrac{1}{2}[\nabla\times\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})]\cdot\underline{\zeta}''_\nu(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^\nu(t,\mathbf{x})]$$
$$+\mu\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})\cdot\dot{\mathbf{\Lambda}}^\nu(t,\mathbf{x})\}d^3\mathbf{x} . \tag{2.12}$$

This now has the form of an integral over local fields and their derivatives. By the superposition principle, it must be valid for arbitrary time-varying fields whose spectral components are in a range of frequencies near the central frequencies ω^ν in the nonabsorbing bands. It is possible, by using a large number of modes $\mathbf{\Lambda}^\nu(t,\mathbf{x})$, to obtain the average energy for a relatively wideband field in this form.

III. LOCAL FIELD THEORY FOR A LINEAR DISPERSIVE DIELECTRIC

In order to quantize the theory, a canonical Lagrangian must be found that corresponds to the Hamiltonian of Eq. (2.12), while generating the Maxwell equations of Eq. (2.1) as Lagrange's equations. The requirement that the Lagrangian has to generate a Hamiltonian that agrees with the classical energy is essential to obtaining a correct quantum theory. The reason for this is that generating *only* the equations of motion, without regard to the Hamiltonian, could lead to a wide variety of canonical momenta. Since these would not result in equivalent quantum theories, no unique quantum predictions would result.[15,16] It is similarly insufficient to only generate the correct energy, without regard to equations of motion, as pointed out by Hillery and Mlodinow.[9] In this section, a classical canonical theory is developed for the linear case using a Lagrangian technique. This is required in order to determine canonical variables for later quantization. Here a local Lagrangian theory is preferable. This allows straightforward extensions to the case of a nonlinear field theory. An alternative, nonlocal, Lagrangian technique is given in the Appendix for comparison purposes.

As a first step, the linear Maxwell equations will be recast as a wave equation. Using the dual potentials $\mathbf{\Lambda}^\nu(t,\mathbf{x})$, together with Maxwell's equations, Eq. (2.1), it is clear that the linear wave equation is

$$\nabla\times\left[\int_0^\infty\underline{\zeta}(\tau,\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^\nu(t-\tau,\mathbf{x})]d\tau\right]=-\mu\ddot{\mathbf{\Lambda}}^\nu(t,\mathbf{x}) . \tag{3.1}$$

For a monochromatic field at frequency $\omega\simeq\omega^\nu$, this becomes

$$\nabla\times\{\underline{\zeta}(\omega,\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^\nu(t,\mathbf{x})]\}=-\mu\ddot{\mathbf{\Lambda}}^\nu(t,\mathbf{x}) . \tag{3.2}$$

The wave equation for the νth envelope function with arbitrary frequency components near ω^ν can therefore be rewritten as

$$\mu\ddot{\mathbf{\Lambda}}^\nu(t,\mathbf{x})=-\nabla\times\{\underline{\zeta}_\nu(\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^\nu(t,\mathbf{x})]$$
$$+i\underline{\zeta}'_\nu(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^\nu(t,\mathbf{x})]$$
$$-\tfrac{1}{2}\underline{\zeta}''_\nu(\mathbf{x})\cdot[\nabla\times\ddot{\mathbf{\Lambda}}^\nu(t,\mathbf{x})]\} . \tag{3.3}$$

Here the combination $\mathbf{\Lambda}^\nu(t-\tau)e^{-i\omega^\nu\tau}$ is treated as a slowly varying function of τ that can be expanded in a Taylor series near $\tau=0$, up to order τ^2. Note that $\underline{\zeta}'_\nu=0$ for $\nu=0$, since $\underline{\zeta}(\omega)$ is an even function of ω. However, for $\nu\neq0$ there is in general a term in the wave equation proportional to ζ'_ν. This does not appear in the energy expression, and is a result of changes in phase velocity due to dispersion.

It is next necessary to derive a Lagrangian whose variational equations correspond to Eq. (3.3), and whose Hamiltonian corresponds to Eq. (2.12). In fact, since $\mathbf{\Lambda}^\nu$ can be specified to be a transverse field, the variations can be also restricted to be transverse. This is similar to the choice of Coulomb gauge often used to quantize the electromagnetic field in the presence of free charges. However, this gauge choice is not identical to the Coulomb gauge, since $\mathbf{\Lambda}^\nu$ is different to the usual vector potential $\mathbf{A}$. The use of restricted variations is well known in canonical quantization theory, and can be treated using transverse functional derivatives.[19]

Given a Lagrangian $L(\mathbf{\Lambda},\dot{\mathbf{\Lambda}})$, which is a functional of $\mathbf{\Lambda}$ and $\dot{\mathbf{\Lambda}}$, the resulting transverse Euler-Lagrange equations are

$$\frac{\partial}{\partial t}\frac{\delta L}{\delta\dot{\Lambda}_i^\perp}-\frac{\delta L}{\delta\Lambda_i^\perp}=0 , \tag{3.4}$$

where transverse derivatives of an arbitrary vector **a** have the property that

$$\frac{\delta}{\delta a_i^{\perp}}[(\nabla\times\mathbf{a})\cdot\mathbf{v}]=(\nabla\times\mathbf{v})_i \ .$$

A general class of Lagrangian will be treated here, using functions $\underline{\alpha}$, $\underline{\beta}$, $\underline{\gamma}$, and $\underline{\delta}$, which are symmetric real matrices. The most general type of Lagrangian to be treated is

$$L=L_0=\tfrac{1}{2}\int\sum_{\nu=-N}^{N}\{\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})\cdot\underline{\alpha}^{\nu}(\mathbf{x})\cdot\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})-[\nabla\times\mathbf{\Lambda}^{-\nu}(t,\mathbf{x})]\cdot\underline{\beta}^{\nu}(\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^{\nu}(t,\mathbf{x})]$$
$$-2i[\nabla\times\mathbf{\Lambda}^{-\nu}(t,\mathbf{x})]\cdot\underline{\gamma}^{\nu}(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]-[\nabla\times\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})]\cdot\underline{\delta}^{\nu}(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]\}d^3\mathbf{x} \ , \tag{3.5}$$

where

$$\underline{\alpha}^{\nu}(\mathbf{x})=\underline{\alpha}^{-\nu}(\mathbf{x}), \quad \underline{\beta}^{\nu}(\mathbf{x})=\underline{\beta}^{-\nu}(\mathbf{x}) \ ,$$
$$\underline{\gamma}^{\nu}(\mathbf{x})=-\underline{\gamma}^{-\nu}(\mathbf{x}), \quad \underline{\delta}^{\nu}(\mathbf{x})=\underline{\delta}^{-\nu}(\mathbf{x}) \ .$$

The canonical momenta are then, omitting the field arguments for clarity,

$$\mathbf{\Pi}_j^{\nu}=\frac{\delta L}{\delta\dot{\Lambda}_j^{\nu\perp}}=\{\underline{\alpha}^{\nu}(\mathbf{x})\cdot\dot{\mathbf{\Lambda}}^{-\nu}-\nabla\times[i\underline{\gamma}^{\nu}(\mathbf{x})\cdot(\nabla\times\mathbf{\Lambda}^{-\nu})+\underline{\delta}^{\nu}(\mathbf{x})\cdot(\nabla\times\dot{\mathbf{\Lambda}}^{-\nu})]\}_j \ . \tag{3.6}$$

These are clearly transverse also, as required for transverse functional derivatives, provided $\underline{\alpha}^{\nu}(\mathbf{x})$ is a uniform scalar quantity. Lagrange's equations follow immediately from (3.6), with the result that

$$\underline{\alpha}^{\nu}(\mathbf{x})\cdot\ddot{\mathbf{\Lambda}}^{\nu}=-\nabla\times[\underline{\beta}^{\nu}(\mathbf{x})\cdot(\nabla\times\underline{\Lambda}^{\nu})-\underline{\delta}^{\nu}(\mathbf{x})\cdot(\nabla\times\ddot{\mathbf{\Lambda}}^{\nu})+2i\underline{\gamma}^{\nu}(\mathbf{x})\cdot(\nabla\times\dot{\mathbf{\Lambda}}^{\nu})] \ . \tag{3.7}$$

The Hamiltonian generated can be greatly simplified by using the divergence theorem and assuming vanishing boundary terms. This gives the result that, in terms of $\mathbf{\Lambda}^{\nu}$ and $\dot{\mathbf{\Lambda}}^{\nu}$,

$$\mathcal{H}=\mathcal{H}_0=\tfrac{1}{2}\int\sum_{\nu=-N}^{N}\{\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})\cdot\underline{\alpha}^{\nu}(\mathbf{x})\cdot\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})+[\nabla\times\mathbf{\Lambda}^{-\nu}(t,\mathbf{x})]\cdot\underline{\beta}^{\nu}(\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^{\nu}(t,\mathbf{x})]$$
$$-[\nabla\times\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})]\cdot\underline{\delta}^{\nu}(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]\}d^3\mathbf{x} \ . \tag{3.8}$$

This can now be compared with the known form of the classical energy in Eq. (2.12), which immediately gives $\underline{\alpha}$, $\underline{\delta}$, and $\underline{\beta}$. The value of $\underline{\gamma}$ is obtained from comparing the wave equation of Eq. (3.3) with Lagrange's equations in Eq. (3.7). Complete agreement is obtained, provided

$$\begin{aligned}&\underline{\alpha}^{\nu}(\mathbf{x})=\mu \ ,\\ &\underline{\beta}^{\nu}(\mathbf{x})=\underline{\zeta}_{\nu}(\mathbf{x}) \ ,\\ &\underline{\gamma}^{\nu}(\mathbf{x})=\tfrac{1}{2}\underline{\zeta}_{\nu}'(\mathbf{x}) \ ,\\ &\underline{\delta}^{\nu}(\mathbf{x})=\tfrac{1}{2}\underline{\zeta}_{\nu}''(\mathbf{x}) \ .\end{aligned} \tag{3.9}$$

In summary, the results of this section give a local-field theory of a linear dispersive dielectric, with results that agree both in dynamics and energy with the known classical results for slowly varying envelope functions. The Hamiltonian has the form of Eq. (2.12), while the Lagrangian has the following structure:

$$L=L_0=\tfrac{1}{2}\int\sum_{\nu=-N}^{N}\{\mu\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})\cdot\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})-[\nabla\times\mathbf{\Lambda}^{-\nu}(t,\mathbf{x})]\cdot\underline{\zeta}_{\nu}(\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^{\nu}(t,\mathbf{x})]$$
$$-i[\nabla\times\mathbf{\Lambda}^{-\nu}(t,\mathbf{x})]\cdot\underline{\zeta}_{\nu}'(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]-\tfrac{1}{2}[\nabla\times\dot{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})]\cdot\underline{\zeta}_{\nu}''(\mathbf{x})\cdot[\nabla\times\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]\}d^3\mathbf{x} \ . \tag{3.10}$$

In the plane-wave case, in one dimension, these results are straightforward. In this case the dual potential Λ is just a scalar. The Hamiltonian is then given by

$$\mathcal{H}=\int[\mu|\dot{\Lambda}(t,\mathbf{x})|^2+\zeta(x)|\partial_x\Lambda(t,\mathbf{x})|^2-\tfrac{1}{2}\zeta''(x)|\partial_x\dot{\Lambda}(t,\mathbf{x})|^2]dx \ . \tag{3.11}$$

The corresponding Lagrangian is similarly obtained to be

$$L_0=\int\left[\mu|\dot{\Lambda}(t,\mathbf{x})|^2-\zeta(x)|\partial_x\Lambda(t,\mathbf{x})|^2-\tfrac{1}{2}\zeta''(x)|\partial_x\dot{\Lambda}(t,\mathbf{x})|^2-\frac{i}{2}\zeta'(x)[\partial_x\Lambda^*(t,\mathbf{x})\partial_x\dot{\Lambda}(t,\mathbf{x})-\partial_x\Lambda(t,\mathbf{x})\partial_x\dot{\Lambda}^*(t,\mathbf{x})]\right]dx \ . \tag{3.12}$$

Here the notation is restricted to just one scalar complex field $[\Lambda(t,\mathbf{x})=\Lambda^1(t,\mathbf{x})]$. In both the Hamiltonian and Lagrangian, the first two terms correspond to those normally found in the theory of massless scalar bosons. The remaining terms indicate the corrections due to dispersion. It is emphasized here that a correct classical Lagrangian should generate *both* the usual field equations *and* the classical energy; which is indeed true of Eq. (3.10). A physically realistic model of a dielectric would include vector fields and a periodic structure for $\zeta(\mathbf{x})$. This would then encompass the usual local field corrections.[18]

IV. NONLINEAR HAMILTONIAN AND LAGRANGIAN

In the case of a nonlinear medium, the full nonlinear polarizability should be used. For a nonlinearity that is relatively fast in its response time, the nonlinear polarizability terms in Eq. (2.2) can be approximated on replacing $\zeta^{(n)}$ by an equivalent infinitely fast δ-function response term, so that

$$P_j^N(t,\mathbf{x})=-\epsilon_0\sum_{n\,(>1)}\zeta^{(n)}_{i;j_1,\ldots,j_n}(\mathbf{x})D_{j_1}(t,\mathbf{x})\cdots D_{j_n}(t,\mathbf{x})\ , \tag{4.1}$$

where

$$\mathbf{P}(t,\mathbf{x})=\mathbf{P}^L(t,\mathbf{x})+\mathbf{P}^N(t,\mathbf{x})=\mathbf{D}(t,\mathbf{x})-\epsilon_0\mathbf{E}(t,\mathbf{x})$$

and

$$\underline{\zeta}^{(n)}(\mathbf{x})=\int_0^\infty\cdots\int_0^\infty\underline{\zeta}^{(n)}(\tau_1,\ldots,\tau_n;\mathbf{x})d\tau_1\cdots d\tau_n\ .$$

Using a standard result of classical nonlinear dielectric theory,[1,17] the increase in stored electrostatic energy density in a nonlinear dielectric is

$$U(t,\mathbf{x})=\int_0^{\mathbf{D}(t,\mathbf{x})}\mathbf{E}\cdot d\mathbf{D}\ . \tag{4.2}$$

As the linear energy is already calculated, including dispersive corrections, it remains only to calculate the additional energy stored due to the nonlinear response, assuming that dispersive corrections are negligible for the nonlinear energy. It is possible to distinguish linear and nonlinear components of the **E** field, as a functional of the electric displacement, from the expansion given in Eq. (2.2). That is, using

$$\mathbf{E}(t,x)=\frac{1}{\epsilon_0}[\mathbf{D}(t,\mathbf{x})-\mathbf{P}^L(t,\mathbf{x})-P^N(t,\mathbf{x})]\ , \tag{4.3}$$

the following result is obtained:

$$U(t,\mathbf{x})=U^L(t,\mathbf{x})+U^N(t,\mathbf{x})\ , \tag{4.4}$$

where

$$U^L(t,\mathbf{x})=\frac{1}{\epsilon_0}\int_0^{\mathbf{D}(t,\mathbf{x})}(\mathbf{D}-\mathbf{P}^L)\cdot d\mathbf{D}\ ,$$

$$U^N(t,\mathbf{x})=\frac{-1}{\epsilon_0}\int_0^{\mathbf{D}(t,\mathbf{x})}\mathbf{P}^N\cdot d\mathbf{D}\ .$$

The first of these terms corresponds to the electrostatic part of the linear dispersive energy already calculated. The second term is a new expression for the nonlinear energy density which is similar to one calculated by Hillery and Mlodinow:[9]

$$U^N(t,\mathbf{x})=\sum_{n\,(>1)}\frac{1}{n+1}\zeta^{(n)}_{j_1,\ldots,j_{n+1}}(\mathbf{x})\times D_{j_1}(t,\mathbf{x})\cdots D_{j_{n+1}}(t,\mathbf{x})\ . \tag{4.5}$$

Here the following symmetry restriction is required:

$$\zeta^{(n)}_{j_1,j_i,j_{n+1}}(\mathbf{x})=\zeta^{(n)}_{j_i,\ldots,j_1,\ldots,j_{n+1}}(\mathbf{x})\ . \tag{4.6}$$

This is necessary in order to allow integration of Eq. (4.4). Hence the total Lagrangian and Hamiltonian in terms of the dual potentials are

$$\begin{aligned}L&=L_0-\int U^N(t,\mathbf{x})d^3\mathbf{x}\ ,\\ \mathcal{H}&=\mathcal{H}_0+\int U^N(t,\mathbf{x})d^3\mathbf{x}\ ,\end{aligned} \tag{4.7}$$

where L_0 and $\mathcal{H}_0$ are as in Sec. III. Since the additional nonlinear terms are assumed to be nondispersive, they do not include the terms in $\dot{\Lambda}$.

Equation (4.5) has an obvious generalization to cases where different central frequencies ω_ν have distinct nonlinear couplings. In this case, new nonlinear coupling coefficients $\zeta^{\nu,\nu_1,\ldots,\nu_n}_{j,j_1,\ldots,j_n}$ are defined, to give a nonlinear energy density of [using (2.4)]

$$U^N(t,\mathbf{x})=\sum_{n\,(>1)}\frac{1}{n+1}\sum_{\nu_1,\ldots,\nu_{n+1}}\zeta^{\nu_1,\ldots,\nu_{n+1}}_{j_1,\ldots,j_{n+1}}(\mathbf{x})[\nabla\times\mathbf{\Lambda}^{\nu_1}(t,\mathbf{x})]_{j_1}\cdots[\nabla\times\mathbf{\Lambda}^{\nu_{n+1}}(t,\mathbf{x})]_{j_{n+1}}\ ,$$

where

$$\begin{aligned}\zeta^{\nu_1,\ldots,\nu_{n+1}}_{j_1,\ldots,j_{n+1}}(\mathbf{x})&=\delta^{(n)}_{j_1,\ldots,j_{n+1}}(\omega^{\nu_2},\ldots,\omega^{\nu_{n+1}};\mathbf{x})\\ &\quad\times\delta_{-\omega^{\nu_1},\omega^{\nu_2}+\omega^{\nu_3}+\cdots+\omega^{\nu_{n+1}}}\ ,\\ \zeta^{\nu_1,\ldots,\nu_i,\ldots,\nu_{n+1}}_{j_1,\ldots,j_i,\ldots,j_{n+1}}(\mathbf{x})&=\zeta^{\nu_i,\ldots,\nu_1,\ldots,\nu_{n+1}}_{j_i,\ldots,j_1,\ldots,j_{n+1}}(\mathbf{x})\ .\end{aligned} \tag{4.8}$$

The Lagrangian that generates Eq. (4.8) is simply that given in Eq. (4.7), with the new expression for the nonlinear energy density included. This expression is only correct when dispersive terms in the nonlinear energy are negligible, and when the above permutation symmetry exists. The Kronecker δ expression in Eq. (4.8) is used to remove nonresonant terms from the energy density in which the relevant carrier frequencies do not sum to zero, so that a slowly varying envelope approximation is implicit here.

The Lagrangian generates the correct equation of motion for the field in the present approximation, which is a generalization of the linear equation given by Eq. (3.3):

$$\mu\ddot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})=-\nabla\times\left[\underline{\zeta}_{\nu}(\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}^{\nu}(t,\mathbf{x})]+i\underline{\zeta}'_{\nu}\cdot[\nabla\times\dot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]-\tfrac{1}{2}\underline{\zeta}''_{\nu}(\mathbf{x})\cdot[\nabla\times\ddot{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})]\right.$$
$$\left.+\sum_{n\,(>1)}\sum_{\nu_1,\ldots,\nu_n}\underline{\zeta}^{\nu,\nu_1,\ldots,\nu_n}(\mathbf{x}):[\nabla\times\mathbf{\Lambda}^{\nu_1}(t,\mathbf{x})]\cdots[\nabla\times\mathbf{\Lambda}^{\nu_n}(t,\mathbf{x})]\right]. \tag{4.9}$$

This Lagrangian therefore gives a canonical theory of the dispersive nonlinear medium, with the correct equations of motion and Hamiltonian. The chief approximation is that the envelope functions are slowly varying, so that a Taylor-series expansion is possible. In addition, there is the restriction that absorption and nonlinear dispersion are neglected within each frequency band. The theory can be regarded as the canonical equivalent of the widely applied classical Bloembergen expansion[1] used in nonlinear optics. Thus the technique given here is the natural canonical field theory for nonabsorptive nonlinear optics. While it only describes those processes which are nondissipative in nature, it is possible to add reservoir couplings to describe dissipation, if required.

In order to give an example of this, a one-dimensional scalar case will be treated, as in Sec. III. The lowest nonlinearity of most universal interest is the nonlinear refractive index. This is a third order nonlinearity, due to $\zeta^{(3)}$ in the present notation. This can be seen on comparing the expansion of Eq. (2.2) with the commonly used expansion[1,20] of

$$P(t,\mathbf{x})=\epsilon_0\chi^{(3)}(-\omega,\omega,\omega)\mathcal{E}^*(\mathbf{x})\mathcal{E}(\mathbf{x})\mathcal{E}(\mathbf{x})e^{-i\omega t}+\text{H.c.} \tag{4.10}$$

Here, as in Eq. (2.5), $\mathcal{E}(\mathbf{x})$ is an envelope function with carrier frequency ω. This result can be used to obtain $\zeta^{(3)}$ in terms of the more usual $\chi^{(3)}$ coefficient on matching powers of $\mathcal{D}$ in the power-series expansion. Thus, on comparing the nonlinear polarizability equation Eq. (4.1) with Eq. (4.10), it is immediate that

$$\zeta^{(3)}(-\omega,\omega,\omega)=-\epsilon_0\chi^{(3)}(-\omega,\omega,\omega)/[\varepsilon(\omega)]^4 . \tag{4.11}$$

The overall Hamiltonian is then, for a complex field $\Lambda(t,\mathbf{x})$,

$$\mathcal{H}=\mathcal{H}_0+\tfrac{1}{4}\zeta^{(3)}\int|\partial_x\Lambda(t,\mathbf{x})|^4dx . \tag{4.12}$$

This model of a one-dimensional nonlinear refractive index medium corresponds to that used in recent calculations[11] of quantum soliton properties in optical media. However, in order to treat quantized fields, it is necessary to include commutators, and extend the classical canonical theory to a fully quantized field theory.

V. QUANTIZATION

The form of the nonlinear Lagrangian and Hamiltonian in Eq. (4.7) is similar to field theories used elsewhere to describe boson fields. The term in $\underline{\zeta}'_{\nu}(\mathbf{x})$ in the linear Lagrangian is an unusual feature, and gives rise to canonical momenta which have the structure

$$\Pi^{\nu}(t,\mathbf{x})=\mu\dot{\mathbf{\Lambda}}^{-\nu}-\tfrac{1}{2}\nabla\times[\underline{\zeta}''_{\nu}(\mathbf{x})\cdot(\nabla\times\dot{\mathbf{\Lambda}}^{-\nu})$$
$$+i\underline{\zeta}'_{\nu}(\mathbf{x})\cdot(\nabla\times\Lambda^{-\nu})] . \tag{5.1}$$

These canonical momenta combine terms in $\Lambda^{-\nu}$ with the usual derivative terms in $\dot{\Lambda}^{-\nu}$. This changes the nature of the mode expansion in terms of annihilation and creation operators. For a nondispersive medium, the linear Lagrangian has no terms of this type. Thus the nondispersive case, which is present as the low-frequency term in the envelope expansion with $\nu=0$, has a rather simpler mode structure than the dispersive case. In neither case do the nonlinear terms enter the canonical momenta, since the nonlinear terms have no time derivatives.

The corresponding quantum theory is obtained from the Dirac commutation relations, which are identical to those in the homogeneous case.[9] In this case, these are *transverse* commutation relations, and would usually have the form, where the corresponding field operators are denoted $\hat{\Lambda}$ and $\hat{\Pi}$,

$$[\hat{\Lambda}_i^{\nu}(\mathbf{x},t),\hat{\Pi}_j^{\mu}(\mathbf{x}',t)]=i\hbar\delta_{ij}^{\perp}(\mathbf{x}-\mathbf{x}')\delta_{\mu\nu} . \tag{5.2}$$

These commutation relations apply at equal times, and have spatial Fourier components of arbitrarily large frequency. In the current problem, this prescription is not appropriate,[21] as the fields are limited in bandwidth. Instead, the commutation relations should be viewed as only applying to spatial Fourier components of frequency within the bandwidth range of the νth frequency band. In practice it is usual to obtain commutation relations for annihilation and creation operators of the field eigenmodes. This is just as valid a quantization procedure as the field quantization defined by (5.2), and has the distinct advantage that it clarifies the questions of bandwidth.

In order to obtain mode functions, a set of Fourier transformed fields is defined. These Fourier components are indexed with $k=(\mathbf{k},\mathbf{e}_k)$. The momentum $\mathbf{k}$ satisfies periodic boundary conditions, and the polarization $\mathbf{e}_k$ is defined so that $\mathbf{k}\cdot\mathbf{e}_k=0$. Thus

$$\mathbf{\Lambda}^{\nu}(t,\mathbf{x})=\sqrt{1/V}\sum_{\mathbf{k}}\lambda_k^{\nu}(t)\mathbf{e}_k e^{i\mathbf{k}\cdot\mathbf{x}}\quad(\nu\geq 0) . \tag{5.3}$$

Here the sums over k include terms labeled $-k$, with $-k\equiv(-\mathbf{k},e_k^*)$, in a symmetrical combination. The linear Lagrangian of Eq. (4.10) for this set of fields is then

$$L_0=\sum_{\nu=0}^{N}\frac{1}{1+\delta_{\nu 0}}\{(\dot{\boldsymbol{\lambda}}^{\nu})^{\dagger}\cdot\underline{\alpha}^{\nu}\cdot\dot{\boldsymbol{\lambda}}^{\nu}-(\boldsymbol{\lambda}^{\nu})^{\dagger}\cdot\underline{\beta}^{\nu}\cdot\boldsymbol{\lambda}^{\nu}$$
$$-i[(\boldsymbol{\lambda}^{\nu})^{\dagger}\cdot\underline{\gamma}^{\nu}\cdot\dot{\boldsymbol{\lambda}}^{\nu}-(\dot{\boldsymbol{\lambda}}^{\nu})^{\dagger}\cdot\underline{\gamma}^{\nu}\cdot\boldsymbol{\lambda}^{\nu}]\} , \tag{5.4}$$

where

$$\lambda_{-k}^{-\nu} \equiv (\lambda_k^{\nu})^* \ .$$

The matrices $\underline{\alpha}^{\nu}$, $\underline{\beta}^{\nu}$, and $\underline{\gamma}^{\nu}$ are Hermitian matrices spanning the range of Fourier modes labeled k in each frequency range. These are defined explicitly as

$$\alpha_{k'k}^{\nu} = \left[\mu \mathbf{e}_{k'}^* \cdot \mathbf{e}_k \delta_{kk'} - \frac{1}{2V} \int [(\mathbf{k}' \times \mathbf{e}_{k'}^*) \cdot \underline{\zeta}_{\nu}''(\mathbf{x}) \cdot (\mathbf{k} \times e_k) \times e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}}] d^3\mathbf{x} \right] ,$$

$$\beta_{k'k}^{\nu} = \frac{1}{V} \int [(\mathbf{k}' \times \mathbf{e}_{k'}^*) \cdot \underline{\zeta}_{\nu}(\mathbf{x}) \cdot (\mathbf{k} \times \mathbf{e}_k) e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}}] d^3\mathbf{x} \ , \quad (5.5)$$

$$\gamma_{k'k}^{\nu} = \frac{1}{2V} \int [(\mathbf{k}' \times \mathbf{e}_{k'}^*) \cdot \underline{\zeta}_{\nu}'(\mathbf{x}) \cdot (\mathbf{k} \times \mathbf{e}_k) e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}}] d^3\mathbf{x} \ .$$

In general, $\underline{\alpha}$, $\underline{\beta}$, and $\underline{\gamma}$ are not diagonal, and the resulting linear Hamiltonian has the structure

$$\mathcal{H}_0 = \sum_{\nu=0}^{N} \frac{1}{1+\delta_{\nu 0}} \{ [\pi^{\nu} + i(\boldsymbol{\lambda}^{\nu})^{\dagger} \underline{\gamma}^{\nu}] \cdot (\underline{\alpha}^{\nu})^{-1} \cdot [(\pi^{\nu})^{\dagger} - i\underline{\gamma}^{\nu} \boldsymbol{\lambda}^{\nu}] + (\boldsymbol{\lambda}^{\nu})^{\dagger} \cdot \underline{\beta}^{\nu} \cdot \boldsymbol{\lambda}^{\nu} \} \ . \quad (5.6)$$

The canonical momenta π^{ν} of the Hamiltonian are given by

$$\begin{aligned} \pi^{\nu} &= [(\dot{\boldsymbol{\lambda}}^{\nu})^{\dagger} \cdot \underline{\alpha}^{\nu} - i(\boldsymbol{\lambda}^{\nu})^{\dagger} \cdot \underline{\gamma}^{\nu}] / (1+\delta_{\nu 0}) \quad (\nu \geq 0) \\ (\pi^{\nu})^{\dagger} &= (\underline{\alpha}^{\nu} \cdot \dot{\boldsymbol{\lambda}}^{\nu} + i\underline{\gamma}^{\nu} \cdot \boldsymbol{\lambda}^{\nu}) / (1+\delta_{\nu 0}) \quad (\nu \geq 0) \ . \end{aligned} \quad (5.7)$$

The theory is quantized by imposing the standard commutation relations between π^{ν} and $\boldsymbol{\lambda}^{\nu}$. These do not involve transversality restrictions, and can simply be written

$$[\hat{\lambda}_k^{\mu}, \hat{\pi}_{k'}^{\nu}] = i\hbar \delta_{\mu\nu} \delta_{kk'} \ . \quad (5.8)$$

It is now possible to reexpand $\hat{\mathcal{H}}_0$ using boson annihilation and creation operators. These are defined as two column vectors $\hat{\mathbf{a}}^{\nu}$ and $\hat{\mathbf{b}}^{\nu}$ relative to an arbitrary invertible complex matrix $\underline{A}^{\nu}$, for $\nu \geq 0$:

$$\begin{aligned} \hat{\mathbf{a}}^{\nu} &= \frac{1}{\sqrt{2\hbar}} \{ \underline{A}^{\nu} \cdot \hat{\boldsymbol{\lambda}}^{\nu} + i[(\underline{A}^{\nu})^{\dagger}]^{-1} \cdot (\hat{\pi}^{\nu})^{\dagger} \} \ , \\ (\hat{\mathbf{b}}^{\nu})^{\dagger} &= \frac{1}{\sqrt{2\hbar}} \{ \underline{A}^{\nu} \cdot \hat{\boldsymbol{\lambda}}^{\nu} - i[(\underline{A}^{\nu})^{\dagger}]^{-1} \cdot (\hat{\pi}^{\nu})^{\dagger} \} \ . \end{aligned} \quad (5.9)$$

It is straightforward to verify that $\hat{\mathbf{a}}_k^{\nu}$ and $\hat{\mathbf{b}}_k^{\nu}$ have the character of annihilation operators, and $(\hat{\mathbf{a}}_k^{\nu})^{\dagger}$ and $(\hat{\mathbf{b}}_k^{\nu})^{\dagger}$ are creation operators. In the nondispersive case with $\nu=0$ it is only necessary to use the plus sign in Eq. (5.9), as $\hat{\mathbf{a}}_k^{\nu}$ and $\hat{\mathbf{b}}_k^{\nu}$ are not independent operators. It is desirable to expand the Hamiltonian in terms of these operators, in a quasidiagonal form, as

$$\hat{\mathcal{H}}_0 = \hbar \sum_{\nu=0}^{N} (\hat{\mathbf{a}}^{\nu})^{\dagger} \cdot \underline{\Omega}^{\nu} \cdot \hat{\mathbf{a}}^{\nu} + \hbar \sum_{\nu=1}^{N} (\hat{\mathbf{b}}^{\nu})^{\dagger} \cdot \underline{\Omega}^{-\nu} \cdot \hat{\mathbf{b}}^{\nu} \ . \quad (5.10)$$

In the Appendix it is shown that this is only possible when $\underline{A}^{\nu}$ is the solution to a quartic matrix equation under appropriate conditions on $\underline{\alpha}$, $\underline{\beta}$, and $\underline{\gamma}$. The quartic equation for $\nu \geq 0$ is

$$[(\underline{A}^{\nu})^{\dagger} \underline{A}^{\nu} (\underline{\alpha}^{\nu})^{-1}]^2 \underline{\alpha}^{\nu} = [\underline{\beta}^{\nu} + \underline{\gamma}^{\nu} (\underline{\alpha}^{\nu})^{-1} \underline{\gamma}^{\nu}] \quad (5.11)$$

and the corresponding frequency matrices will be, using Eq. (A8),

$$\underline{\Omega}^{\pm\nu} = \{ \underline{A}^{\nu} \pm [(\underline{A}^{\nu})^{\dagger}]^{-1} \underline{\gamma}^{\nu} \} (\underline{\alpha}^{\nu})^{-1} (\underline{A}^{\nu})^{\dagger} \ . \quad (5.12)$$

If required, the Hamiltonian can then be further simplified to the completely diagonal form, on diagonalizing the matrices $\underline{\Omega}^{\pm\nu}$ within each frequency band; thus

$$\hat{\mathcal{H}}_0 = \hbar \left[\sum_{\nu=0}^{N} \sum_{n} \omega_n^{\nu} (\tilde{a}_n^{\nu})^{\dagger} \tilde{a}_n^{\nu} + \sum_{\nu=1}^{N} \sum_{n} \omega_n^{-\nu} (\tilde{b}_n^{\nu})^{\dagger} \tilde{b}_n^{\nu} \right] , \quad (5.13)$$

Here a complete diagonalization is performed within each envelope, to obtain an expansion of the free Hamiltonian in terms of uncoupled harmonic-oscillator modes. The diagonal operators $\tilde{a}_n^{\nu}$ and $\tilde{b}_n^{\nu}$, and frequencies $\omega_n^{\pm\nu}$, are given by

$$\begin{aligned} \tilde{\mathbf{a}}^{\nu} &= \underline{U}^{\nu} \cdot \hat{\mathbf{a}}^{\nu} \ , \\ (\tilde{\mathbf{b}}^{\nu})^{\dagger} &= \underline{U}^{-\nu} \cdot (\hat{\mathbf{b}}^{\nu})^{\dagger} \ , \end{aligned} \quad (5.14)$$

where

$$[\underline{U}^{\pm\nu} \underline{\Omega}^{\pm\nu} (\underline{U}^{\pm\nu})^{-1}]_{nm} = \omega_n^{\pm\nu} \delta_{nm} \ .$$

This generates a set of normal modal solutions corresponding to the diagonal operators $\tilde{\mathbf{a}}^{\nu}$ ($\nu \geq 0$) which typically have characteristic frequencies $\omega_n^{\nu} > 0$. In addition, there is a set of anomalous modes with $\nu < 0$ corresponding to the operators $\tilde{\mathbf{b}}^{\nu}$ ($\nu \geq 1$). These have a characteristic behavior in which the solutions generate envelopes $\boldsymbol{\Lambda}^{|\nu|}$ which vary with negative frequencies ($\boldsymbol{\Lambda}^{|\nu|} \sim e^{i|\omega|t}$). Although these modes are a necessary part of the local Lagrangian theory, they correspond to frequencies that are outside the range of validity of the Taylor expansion, since the original Taylor series expansion was only valid when $\boldsymbol{\Lambda}^{|\nu|} \sim e^{-i|\omega|t}$. The anomalous modes correspond to the physical absorption regions which must necessarily accompany any dispersive solution. It is necessary to assume that these modes are in the vacuum state, because they correspond to dynamics outside the range of validity of the approximations used. In order to accurately treat absorption, a microscopic theory of the interaction with the dielectric atoms is preferable.

Having obtained a modal expansion of $\hat{\mathcal{H}}_0$, the correct final nonlinear Hamiltonian can be written as in Eq. (4.7). This now has the following expanded form, written for simplicity in terms of the operators $\hat{\mathbf{a}}^{\nu}$:

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\sum_{n\,(>0)}\frac{1}{n+1}\sum_{\{\nu_1,\ldots,\nu_{n+1}\}}\int\zeta^{\nu_1,\ldots,\nu_{n+1}}_{j_1,\ldots,j_{n+1}}(\mathbf{x})[\nabla\times\hat{\mathbf{\Lambda}}^{\nu_1}(t,\mathbf{x})]_{j_1}\cdots[\nabla\times\hat{\mathbf{\Lambda}}^{\nu_{n+1}}(t,\mathbf{x})]_{j_{n+1}}d^3\mathbf{x}\ , \tag{5.15}$$

where

$$\hat{\mathbf{\Lambda}}^{\nu}(t,\mathbf{x})=\left[\frac{\hbar}{2V}\right]^{1/2}\sum_k\mathbf{e}_k e^{i\mathbf{k}\cdot\mathbf{x}}\left[\sum_{k'}(\underline{A}^{\nu})^{-1}_{kk'}\hat{\mathbf{a}}^{\nu(\dagger)}_{k'}\right]\quad(\nu>0)$$

and

$$\hat{\mathbf{\Lambda}}^{-\nu}(t,\mathbf{x})=[\mathbf{\Lambda}^{\nu}(t,\mathbf{x})]^{\dagger}\ ,$$

$$\hat{\mathbf{\Lambda}}^{0}(t,\mathbf{x})=\left[\frac{\hbar}{2V}\right]^{1/2}\sum_k\mathbf{e}_k e^{i\mathbf{k}\cdot\mathbf{x}}\left[\sum_{k'}(\underline{A}^{0})^{-1}_{kk'}\hat{\mathbf{a}}^{0(\dagger)}_{k'}\right]+\text{H.c.}$$

Here only the $\hat{\mathbf{a}}^{\nu}_k$ operators corresponding to the normal modes are included, as the $\hat{\mathbf{b}}^{\nu}_k$ operators generate anomalous localized modes. These would normally have an off-resonant coupling in calculations of lossless propagation. The corresponding terms are therefore omitted from Eq. (5.15). However, it is obvious from the theory presented here that additional quanta are a necessary element of a dispersive medium. In principle, these could become excited through the nonlinear coupling induced by the Hamiltonian. While this is certainly possible, a calculation of these effects requires more information on the dielectric structure, since the anomalous modes may not have the characteristics inferred from the present simplified model.

VI. SUMMARY

A well-defined Lagrangian-based canonical theory of a nonlinear, dispersive dielectric was obtained from the known linear and nonlinear dielectric properties. This can be quantized, provided care is taken to use mode functions whose characteristic frequencies are within the relevant bandwidth applicable to the dielectric transmission bands. The resulting modes have creation and annihilation operators that are very similar to normal photon operators. The nonlinearities in the Hamiltonian couple the linear quanta together. In order to demonstrate this more specifically, the plane-wave case will be treated for an isotropic, homogeneous medium, with a single carrier frequency ($\nu=1$).

Here $\underline{\alpha}$, $\underline{\beta}$, and $\underline{\gamma}$ are given by

$$\begin{aligned}\alpha_{k'k}&=\alpha_k\delta_{k'k}=(\mu-\tfrac{1}{2}|\mathbf{k}|^2\zeta_1'')\delta_{k'k}\ ,\\ \beta_{k'k}&=\beta_k\delta_{k'k}=|\mathbf{k}|^2\zeta_1\delta_{k'k}\ ,\\ \gamma_{k'k}&=\gamma_k\delta_{k'k}=\tfrac{1}{2}|\mathbf{k}|^2\zeta_1'\delta_{k'k}\ .\end{aligned}\tag{6.1}$$

This result implies that A, X, and Ω are all diagonal, with

$$\begin{aligned}A_{k'k}&=(\alpha_k\beta_k+\gamma_k^2)^{1/4}\cdot\delta_{k'k}\ ,\\ (\Omega^{\pm})_{k'k}&=\frac{1}{\alpha_k}[(\alpha_k\beta_k+\gamma_k^2)^{1/2}\pm\gamma_k]\delta_{k'k}\ .\end{aligned}\tag{6.2}$$

Taking these solutions it is straightforward to then show that the dispersion relation for propagating waves is satisfied. The expected relation is that $\mu\omega^2=|\mathbf{k}|^2\zeta(\omega)$. From the result of (6.2), it is possible to show that

$$\mu\omega^2=|\mathbf{k}|^2(\zeta_1\pm\omega\zeta_1'+\tfrac{1}{2}\omega^2\zeta_1'')\ . \tag{6.3}$$

The expression in parentheses is identical to the approximate expansion $\zeta_1(\omega)$ near $\omega=\omega^1$, provided the positive branch is used. It is clear that the solutions Ω^- from the negative branch do not correspond to frequencies near ω^1, and cannot be normal propagating solutions. In this sense, the modes described by the operators $\hat{\mathbf{b}}$ are unphysical, as they occur in frequency ranges for which the Taylor expansion used for the dispersion relation is no longer strictly valid.

Neglecting these unphysical modes, the results for the expansion are obtained as follows, in the free-field homogeneous case:

$$\begin{aligned}\mathbf{D}(t,\mathbf{x})&=i\sum_k\left[\frac{\hbar\dfrac{\partial\omega_k}{\partial k}}{2V|\mathbf{k}|\zeta_1(\omega_k)}\right]^{1/2}\\ &\quad\times(\mathbf{k}\times\mathbf{e}_k\hat{\mathbf{a}}_k e^{i\mathbf{k}\cdot\mathbf{x}-i\omega_k t}-\text{H.c.})\ ,\\ \mathbf{B}(t,\mathbf{x})&=-i\sum_k\mu\omega_k\left[\frac{\hbar\dfrac{\partial\omega_k}{\partial k}}{2V|\mathbf{k}|\zeta_1(\omega_k)}\right]^{1/2}\\ &\quad\times(\mathbf{e}_k\hat{\mathbf{a}}_k e^{i\mathbf{k}\cdot\mathbf{x}-i\omega_k t}-\text{H.c.})\end{aligned}\tag{6.4}$$

where

$$\begin{aligned}\zeta_1(\omega)&\equiv\zeta_1+\omega\zeta_1'+\tfrac{1}{2}\omega^2\zeta_1''\ ,\\ (\omega_k)^2&=|\mathbf{k}|^2\zeta_1(\omega_k)/\mu\ .\end{aligned}$$

The above expression agrees precisely with the nonlocal dispersive mode expansion obtained in the Appendix, apart from the Taylor expansion for $\zeta(\omega)$. This is approximated here by expanding around the carrier frequencies for the envelope function to give $\zeta_1(\omega)$, as in Eq. (3.2). Thus the new mode operators $\hat{\mathbf{a}}^{\nu}_k$ in each frequency band correspond closely to the operators found using a nonlocal Lagrangian.

It is useful to note here that the intensity or Poynting vector in the combined dielectric and radiation field system, neglecting rapidly varying terms, is just $\mathbf{E}\times\mathbf{H}$, even in a general dielectric.[18] This now has a quantum expectation value in the single-mode case of

$$\langle\mathbf{E}\times\mathbf{H}\rangle=\left[\frac{\omega_k'}{V}\right]\hbar\omega_k\langle\hat{\mathbf{a}}^{\dagger}_k\hat{\mathbf{a}}_k\rangle\ . \tag{6.5}$$

Letting $V=AL$ for a medium length L, the ratio L/ω_k' is the transit time for a wave to pass through the medium. Thus, there are $\mathcal{N}=\langle\hat{\mathbf{a}}^{\dagger}_k\hat{\mathbf{a}}_k\rangle$ quanta of energy $\hbar\omega_k$ in the quantization volume, being transported across the area A

per unit transit time of the medium. The quantized theory therefore clearly describes quanta of energy $\hbar\omega_k$, each traveling at their respective group velocity of ω_k' through the quantization volume V. The quanta themselves must be regarded as combined excitations of the field and of the polarized medium. They can therefore be termed photon polaritons or dressed photons using the terminology of Hopfield,[22] as they correspond to the photonlike branch of the polariton dispersion relation.

Thus there is a simple physical interpretation of these results. This is that the displacement field is expanded in annihilation and creation operators corresponding to the photon polaritons of the dielectric. Nonlinear couplings between these quanta are provided by the additional nonlinear terms in the Hamiltonian, when expressed as a function of the canonical electric displacement operator. The expansion of Eq. (5.15) omits the anomalous quanta, which model the far-off-resonant absorption bands needed to satisfy dispersion relations.[22] In most cases, the usual resonance conditions are arranged to prevent coupling to absorption bands. In those cases where resonant absorption does occur, a microscopic theory of the dielectric is necessary.

For this reason, the dynamics of the anomalous quanta are not included accurately within the present Taylor series expansion of the dielectric permittivity. Their origin is clearly related to the use of complex amplitudes for the envelope functions. These are classically necessary in order to correctly treat dispersion in dielectrics. The imaginary part of the complex amplitude is a new dynamical degree of freedom, which allows for the existence of polarization dynamics. This dynamical behavior is known in solid-state physics[23] to result in polariton dispersion relations having either photon *or* phononlike character. However, only the photon-polariton behavior can be accurately obtained from refractive-index-type information.

In quantum field theory, a complex field acquires a charge; in other words, the number of possible bosons is automatically doubled. Here, these do not appear symmetrically, owing to the Lagrangian structure. Instead, they have the useful effect of providing a minimal representation of the other degrees of freedom[24] that any dispersive dielectric with a local polarization must possess, and which are in fact observable in intracavity dynamics experiments.[25] In the present theory, only the photonlike branches have a physical significance. Nevertheless, it is clear that nonlinearities can in principle couple these different types of quanta together. To treat this would require a detailed microscopic model.

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APPENDIX

The algebraic properties of the boson annihilation and creation operators are obtained in this appendix. For simplicity, the classical Lagrangian from Eq. (5.4) is written in matrix form for one frequency component ν, as

$$L=\frac{1}{1+\delta_{\nu 0}}[\dot{\boldsymbol{\lambda}}^{\dagger}\underline{\alpha}\dot{\boldsymbol{\lambda}}-i(\boldsymbol{\lambda}^{\dagger}\underline{\gamma}\dot{\boldsymbol{\lambda}}-\dot{\boldsymbol{\lambda}}^{\dagger}\underline{\gamma}\boldsymbol{\lambda})-\boldsymbol{\lambda}^{\dagger}\underline{\beta}\boldsymbol{\lambda}] . \quad \text{(A1)}$$

Here the canonical momenta are clearly

$$\begin{aligned}\boldsymbol{\pi}&=(\dot{\boldsymbol{\lambda}}^{\dagger}\underline{\alpha}-i\boldsymbol{\lambda}^{\dagger}\underline{\gamma}) ,\\ \boldsymbol{\pi}^{\dagger}&=(\underline{\alpha}\dot{\boldsymbol{\lambda}}+i\underline{\gamma}\boldsymbol{\lambda}) .\end{aligned} \quad \text{(A2)}$$

Next, boson-type operators can be defined for the corresponding quantum theory after changing the classical fields to quantum fields, as

$$\begin{aligned}\hat{\mathbf{a}}&=\frac{1}{\sqrt{2\hbar}}[\underline{A}\hat{\boldsymbol{\lambda}}+i(\underline{A}^{\dagger})^{-1}\hat{\boldsymbol{\pi}}^{\dagger}] ,\\ \hat{\mathbf{b}}^{\dagger}&=\frac{1}{\sqrt{2\hbar}}[\underline{A}\hat{\boldsymbol{\lambda}}-i(\underline{A}^{\dagger})^{-1}\hat{\boldsymbol{\pi}}^{\dagger}] ,\end{aligned} \quad \text{(A3)}$$

so that (if $\nu\neq 0$)

$$\begin{aligned}&[\hat{a}_i,\hat{a}_j^{\dagger}]=[b_i,b_j^{\dagger}]=\delta_{ij} ,\\ &[\hat{a}_i,\hat{a}_j]=[\hat{b}_i,\hat{b}_j]=[\hat{a}_i^{\dagger},\hat{b}_j]=[\hat{a}_i,\hat{a}_j^{\dagger}]=0 .\end{aligned}$$

The quantum Hamiltonian for Eq. (A1) is to be written using the new operators in the form

$$\hat{\mathcal{H}}=\hbar(\hat{\mathbf{a}}^{\dagger}\underline{\Omega}\hat{\mathbf{a}}+\hat{\mathbf{b}}^{\dagger}\underline{\Omega}\hat{\mathbf{b}})/(1+\delta_{\nu 0}) . \quad \text{(A4)}$$

This must be equal to the original canonical Hamiltonian generated using Eq. (A1), which is

$$\hat{\mathcal{H}}=[(\hat{\boldsymbol{\pi}}+i\hat{\boldsymbol{\lambda}}^{\dagger}\underline{\gamma})\underline{\alpha}^{-1}(\hat{\boldsymbol{\pi}}^{\dagger}-i\underline{\gamma}\hat{\boldsymbol{\lambda}})+\hat{\boldsymbol{\lambda}}^{\dagger}\underline{\beta}\hat{\boldsymbol{\lambda}}]/(1+\delta_{\nu 0}) . \quad \text{(A5)}$$

Equating the two forms of Hamiltonian (and ignoring zero-point energy terms) results in the following identities:

$$\begin{aligned}&\tfrac{1}{2}\underline{A}^{-1}(\underline{\Omega}^{+}+\underline{\Omega}^{-})(\underline{A}^{\dagger})^{-1}=\underline{\alpha}^{-1} ,\\ &\tfrac{1}{2}\underline{A}^{\dagger}(\underline{\Omega}^{+}+\underline{\Omega}^{-})\underline{A}=(\underline{\beta}+\underline{\gamma}\,\underline{\alpha}^{-1}\underline{\gamma}) ,\\ &\tfrac{1}{2}\underline{A}^{\dagger}(\underline{\Omega}^{+}-\underline{\Omega}^{-})(\underline{A}^{\dagger})^{-1}=\underline{\gamma}\,\underline{\alpha}^{-1} .\end{aligned} \quad \text{(A6)}$$

This immediately leads to an equation for $\underline{A}^{\dagger}\underline{A}$, which is

$$\underline{X}^2=(\underline{\beta}+\underline{\gamma}\,\underline{\alpha}^{-1}\underline{\gamma})\underline{\alpha}^{-1} , \quad \text{(A7)}$$

where

$$\underline{X}\,\underline{\alpha}=\underline{A}^{\dagger}\underline{A} .$$

Once the solution to this matrix fourth root is obtained, the frequency matrices are

$$\underline{\Omega}^{\pm}=[\underline{A}\pm(\underline{A}^{\dagger})^{-1}\underline{\gamma}]\underline{\alpha}^{-1}(\underline{A}^{\dagger}) . \quad \text{(A8)}$$

Given this frequency matrix, $\underline{\Omega}$ can then be diagonalized, if it is Hermitian. The requirement of Hermiticity implies, from (A8), that $\underline{X}$ is chosen to commute with $\underline{\gamma}\,\underline{\alpha}^{-1}$ (and hence $\underline{\beta}\,\underline{\alpha}^{-1}$).

An important question here is the existence of the solu-

tion defined implicitly in Eq. (A7). Clearly $\underline{X}\,\underline{\alpha}$ must have a positive definite structure, to allow the Hermitian product $(\underline{A}^{\dagger}\underline{A})$ to be well defined. While any solution for $\underline{X}$ can be used to generate an alternate solution $-\underline{X}$, this does not necessarily guarantee the existence of a positive definite solution, since the matrix $\underline{X}\,\underline{\alpha}$ could have eigenvalues of alternating sign. Where $\underline{\alpha}$, $\underline{\beta}$, and $\underline{\gamma}$ do not allow positive definite solutions, for $A^{\dagger}A$, or if no Hermitian frequency matrix Ω exists, it would then become necessary to use more general forms of mode expansion than Eq. (A3), by coupling positive- and negative-frequency terms. This, however, implies that the original well-defined frequency ranges are necessarily violated. It is a requirement, therefore, of mode expansions of this type, that $\underline{\alpha}$, $\underline{\beta}$, and $\underline{\gamma}$ must generate positive definite solutions for $\underline{X}\,\underline{\alpha}$, and that $\underline{X}$ commutes with $\underline{\gamma}\,\underline{\alpha}^{-1}$. Under these conditions, the wave equation is equivalent to the following equation for Ω:

$$\underline{\alpha}\,\underline{A}^{-1}(\underline{\Omega}^{\pm})^2=\underline{\beta}\,\underline{A}^{-1}\pm 2\underline{\gamma}\,\underline{A}^{-1}\underline{\Omega}^{\pm}\ . \tag{A9}$$

In the case of $\nu=0$, only one type of boson exists (since $\underline{\gamma}=\underline{0}$), and the question of cross coupling does not arise. Apart from this and a factor of $\frac{1}{2}$ in the Hamiltonian and Lagrangian, the equations are identical.

An alternative procedure that can be used is to treat the eigenvalue problem of the linear wave equation directly. Since this does not involve the local Lagrangian theory, these can be regarded as nonlocal modes. The nonlocal eigenmodes are just the solutions to the Fourier transformed equation for the envelope functions. In fact, they are given by the solutions to

$$\omega_n^2\mu\mathbf{\Lambda}_n(\mathbf{x})=\nabla\times\{\underline{\xi}(\omega_n,\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}_n(\mathbf{x})]\}\ . \tag{A10}$$

Here the eigenfrequencies are labeled with subscripts ω_n to distinguish them from the carrier frequencies ω^{ν} used elsewhere. A Taylor-series expansion of $\underline{\xi}(\omega,\mathbf{x})$ is not necessary here. Of course, appropriate boundary conditions must be utilized at any cavity boundary. For an infinite medium, it is usual to define a periodic boundary condition on a large volume V, and then take limits as $V\to\infty$. This results in complex eigenmodes $\mathbf{\Lambda}_n$, with a frequency spacing determined by the nonlinear eigenvalue problem of Eq. (A10). In fact, (A10) simply generalizes (A9) to the case of more general frequency dependence in $\underline{\xi}(\omega,\mathbf{x})$, relative to the Taylor-series quadratic dependence on frequency of $\xi_1(\omega,\mathbf{x})$.

Given that a complete set of modal solutions $\mathbf{\Lambda}_n$ to Eq. (A10) is available, the total field can be expanded in terms of $\mathbf{\Lambda}_n$ as

$$\mathbf{\Lambda}(t,\mathbf{x})=2\,\mathrm{Re}\left[\sum_{n\,(>0)}a_n\mathbf{\Lambda}_n(\mathbf{x})e^{-i\omega_n t}\right]\ . \tag{A11}$$

Corresponding to each eigenmode $\mathbf{\Lambda}_n$ at frequency ω_n, there is also a solution $\mathbf{\Lambda}_{n'}$ at frequency $\omega_{n'}=-\omega_n$, since $\underline{\xi}(\omega,\mathbf{x})=\underline{\xi}(-\omega,\mathbf{x})$. Similarly, any solution $\mathbf{\Lambda}_n$ has a companion solution $\mathbf{\Lambda}_n^*$, since $\underline{\xi}(\omega,\mathbf{x})=\underline{\xi}^*(\omega,\mathbf{x})$. Of course, these results are only applicable in the time-reversible propagation that occurs in nonabsorbing dielectrics without external magnetic fields. In order to have a complete set of complex solutions, it is simplest to include the solutions $\mathbf{\Lambda}_n^*$ directly, by considering $\mathbf{\Lambda}_n^*$ as a distinct eigenmode with a different index n', in Eq. (A10). The coefficients a_n can themselves be used to generate time-dependent dynamical variables $a_n(t)$. They have an equation of motion given by

$$\dot{a}_n(t)=-i\omega_n a_n(t)\ . \tag{A12}$$

The trivial symmetry in Eq. (A10), where replacing ω_n by $-\omega_n$ gives an identical mode function, is included by taking the complex conjugate of a_n as a solution. These are not regarded here as distinct solutions, since they are already included in Eq. (A11) through taking the real part. Thus all the frequencies ω_n can be regarded as positive.

The Lagrangian that generates Eq. (A12) is now just the usual harmonic oscillator Lagrangian, where

$$L=\hbar\sum_n[i(\dot{a}_n a_n^*-\dot{a}_n^* a_n)-\omega_n a_n^* a_n]\ . \tag{A13}$$

From this Lagrangian, the quantum canonical momenta and canonical commutation relations are

$$\hat{\pi}_n=i\hbar\hat{a}_n^{\dagger}\ , \tag{A14}$$

where

$$[\hat{a}_n,\hat{a}_m^{\dagger}]=\delta_{nm}\ .$$

Hence the total system energy or quantum Hamiltonian is given by

$$\hat{\mathcal{H}}=\sum_n\hbar\omega_n\hat{a}_n^{\dagger}\hat{a}_n\ . \tag{A15}$$

An expression of the form (A15) is the usual one employed for vacuum fields also. However, the derivation here relies heavily on the reality and symmetry of the refraction index. These assumptions cannot in fact be valid over all frequencies, since dispersive dielectrics are also absorbers. For this reason (A15) is only an approximation, valid inside restricted frequency bands. The frequency band index ν is implicit here, as it is not necessary to distinguish the bands. Nevertheless, the expression is valid only near the nonabsorbing frequencies of the dielectric.

It is essential to have a correspondence between $\hat{\mathcal{H}}$ defined in Eq. (A15), and the usual classical dispersive energy expression. This is obtained by calculating $\langle H\rangle$ from Eq. (2.9), for a classical single-mode excitation, i.e.,

$$\begin{aligned}\langle H\rangle=a_n^*a_n\int\{&[\nabla\times\mathbf{\Lambda}_n^*(\mathbf{x})]\cdot[\underline{\xi}(\omega_n,\mathbf{x})\\&-\omega_n\underline{\xi}'(\omega_n,\mathbf{x})]\cdot[\nabla\times\mathbf{\Lambda}_n(\mathbf{x})]\\&+\mu\omega_n^2\mathbf{\Lambda}_n^*(\mathbf{x})\cdot\mathbf{\Lambda}_n(\mathbf{x})\}d^3\mathbf{x}\ .\end{aligned} \tag{A16}$$

The following result from vector analysis, using the divergence theorem, is now required:

$$\int\mathbf{v}\cdot(\nabla\times\mathbf{u})d^3\mathbf{x}=\int\mathbf{u}\cdot(\nabla\times\mathbf{v})d^3\mathbf{x}\ ,$$

provided

$$\int(\mathbf{u}\times\mathbf{v})\cdot d\,\mathbf{A}=0\ . \tag{A17}$$

Applying this result and assuming the boundary terms in fact vanish, together with the wave equation of Eq. (3.1), gives the necessary condition on mode normalization. For the Hamiltonian of Eq. (3.7) to agree with the classical energy, the result is

$$\frac{1}{\hbar}\int\{2\mu\omega_n\mathbf{\Lambda}_n^*(\mathbf{x})\cdot\mathbf{\Lambda}_n(\mathbf{x}) - [\nabla\times\mathbf{\Lambda}_n^*(\mathbf{x})]\cdot\underline{\zeta}'(\omega_n,\mathbf{x})\cdot[\nabla\times\mathbf{\Lambda}_n(\mathbf{x})]\}d^3\mathbf{x}=1\ . \tag{A18}$$

Thus a *linear* theory of a quantum dispersive dielectric is relatively straightforward in terms of nonlocal mode operators. In this case, there is no need even to use a Taylor expansion. This treatment generalizes a known homogeneous result[7] to the case of an inhomogeneous dispersive dielectric. A similar result for the nondispersive inhomogeneous case is also known.[8] However, the complete integral expression given in Eq. (A18) is required for mode-function normalization in this dispersive inhomogeneous case.

The usual orthogonality conditions of mode functions can also be very strongly modified in a dispersive medium. Despite this, in homogeneous cases, this form of mode expansion is similar to the usual free-field one (apart from the normalization). For an isotropic homogeneous medium, the relevant modes are plane waves. The normalization then reduces to a simple expression in terms of the group velocity $\omega'=\partial\omega/\partial k$, so that Eq. (3.2) becomes in the quantum case

$$\hat{\mathbf{\Lambda}}(t,\mathbf{x})=\sum_n\left[\frac{\hbar\omega_n'}{2\zeta(\omega_n)|\mathbf{k}_n|V}\right]^{1/2}(\hat{\mathbf{a}}_n\mathbf{e}_n e^{i\mathbf{k}_n\cdot\mathbf{x}-i\omega_n t}+\text{H.c.})\ , \tag{A19}$$

where

$$\omega_n'=\left[\frac{\partial}{\partial\omega}|\mathbf{k}(\omega_n)|\right]^{-1}=\left[\frac{\omega_n}{|k_n|}\right]\left[1-\frac{1}{2}\frac{\omega_n\zeta'(\omega_n)}{\zeta(\omega_n)}\right]^{-1}\ .$$

Here $\mathbf{e}_n$ is a unit polarization vector with $\mathbf{e}_n^*\cdot\mathbf{e}_n=1$ and $\mathbf{k}_n\cdot\mathbf{e}_n=0$. This is substantially identical to the expansion of Eq. (6.4) in the main text, also for the plane-wave case. In fact, the only difference, as expected, is that Eq. (6.4) uses the Taylor expansion approximation $\xi_1(\omega)$ for the inverse permittivity. By comparison, in (A19) the exact inverse permittivity is used. This *nonlocal* expansion therefore has the advantage that it is applicable over wide bandwidths. However, as it does not result from a local Lagrangian, it is less straightforward to generalize to nonlinear interactions, and requires the solution of a nonlinear eigenvalue problem.

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