

Quantization of the electromagnetic field in dielectrics

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We present a fully canonical quantization scheme for the electromagnetic field in dispersive and lossy linear dielectrics. This scheme is based on a microscopic model, in which the medium is represented by a collection of interacting matter fields. We calculate the exact eigenoperators for the coupled system and express the electromagnetic field operators in terms of them. The dielectric constant of the medium is explicitly derived and is shown to satisfy the Kramers-Kronig relations. We apply these results to treat the propagation of light in dielectrics and obtain simple expressions for the electromagnetic field in the medium in terms of space-dependent creation and annihilation operators. These operators satisfy a set of equal-space commutation relations and obey spatial Langevin equations of evolution. This justifies the use of such operators in phenomenological models in quantum optics. We also obtain two interesting relationships between the group and the phase velocity in dielectrics.

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I. INTRODUCTION

In classical electromagnetism, the analysis of the interaction of light with a continuous medium is performed in two stages [1, 2]. In the first, an explicit model of the medium is introduced and its response to an electromagnetic field is calculated. At this stage, the emphasis is put on the matter and the light field is considered as a probe of the medium properties. For an insulating and nonmagnetic medium, the properties relevant to the interaction with light are embodied in the linear and nonlinear susceptibilities $\chi^{(1)}$, $\chi^{(2)}$, ... which can be calculated in terms of the parameters of the model. In the second stage, these susceptibilities are used to obtain the characteristics of the electromagnetic wave in the matter. The emphasis is now put on the light field and on the way it is modified by the interaction with the matter. At this stage, the explicit model of the matter is no longer needed. Indeed, in many cases in nonlinear optics, calculation of the susceptibilities from the structure of the medium is not practical and they have to be obtained as experimental parameters. A fundamental requirement is that the dielectric constant should satisfy the Kramers-Kronig relations [3].

The usual approach to quantum optics in dielectric media is to start from the second stage and introduce the medium only by its linear or nonlinear susceptibilities. In one procedure, the macroscopic fields are used to build an effective Lagrangian density [4-11] whose Euler-Lagrange equations are identical to the macroscopic Maxwell equations in the dielectric:

$$\begin{aligned} \left(\frac{1}{\mu_0}\right) \nabla \times \mathbf{B} &= \frac{\partial \mathbf{D}}{\partial t}, & \nabla \cdot \mathbf{B} &= 0, \\ \nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t}, & \nabla \cdot \mathbf{D} &= 0, \end{aligned} \quad (1.1)$$

together with the constitutive relation linking the displacement field \mathbf{D} to the electric field \mathbf{E} : $\mathbf{D} = \hat{\epsilon} \mathbf{E}$, where $\hat{\epsilon}$, the dielectric response operator, is in general an integral operator acting on \mathbf{E} . This procedure has been used extensively in dispersionless media. It was first developed in a homogeneous infinite linear medium [4] and later extended to nonlinear ones [5-7]. More recently, it was applied to inhomogeneous media as well [8] and to paraxial propagation [9]. However, attempts to add dispersion to this effective scheme have run into difficulties [10, 11]. The reason for this is that inclusion of dispersion leads to a temporally nonlocal relationship between the electric field \mathbf{E} and the displacement field \mathbf{D} [3]. The effective Lagrangian, which includes both, is therefore also nonlocal in time and cannot be used directly in a quantization scheme [5]. In an attempt to overcome this problem, Watson and Jauch [10] have shown that, under some restrictive assumptions on the refractive index of the medium, it is still possible to quantize the field in the reciprocal space. Indeed, in this space, the integral operators appearing in the theory are replaced by multiplicative factors, thus making the Lagrangian density local in reciprocal space. However, this approach relies on a one-to-one correspondence between the frequency ω and the wave vector k . Therefore it is restricted to indices of refraction which are monotonic functions of the frequency, which makes it inapplicable in many physical situations. Much more recently, a similar type of approach has been used by Drummond [11]. In this approach, the total fields are decomposed into a sum of fields with restricted bandwidths, which are taken as the fundamental variables of the theory. As above, quantization is performed in the reciprocal space and relies on the same one-to-one correspondence between k and ω within each frequency band. As the field bandwidths can be chosen to be arbitrarily small, this limitation is no longer very restrictive. A

problem associated with this method is the appearance in the theory of extra unphysical boson fields which have eventually to be discarded. Very recently, we showed [12] that, by introducing the medium explicitly in the theory, one could overcome all these problems and quantize the electromagnetic field in a dielectric in a completely standard fashion. However, as with the effective methods, our approach was limited to a nonabsorbing dielectric. In this work, we shall lift this restriction and show how our method can be extended to include lossy media.

Other, more phenomenological procedures which do not rely on a Lagrangian formalism have been used by other workers to treat the case of a nondispersive but inhomogeneous medium [13] and to quantize the field within the slowly varying envelope approximation [14]. In a popular approach [15–19], which closely follows classical electromagnetism, the fields are decomposed into space-dependent monochromatic amplitudes and Maxwell equations are used to obtain spatial evolution equations. Quantization of the field is performed by replacing these amplitudes by operators and imposing a set of equal-*space* commutation relations (ESCR). This technique can be used in linear as well as nonlinear media and its simplicity has led to its wide application to propagation problems in quantum optics. However, it is not derived from a Lagrangian and therefore has not been justified in terms of a canonical scheme. As the main difference between the classical and the quantum case lies in the commutation relations, which are responsible, for example, for the vacuum fluctuations, we believe it is of great theoretical importance to justify the use of such ESCR by deriving them from a canonical quantization scheme. One of the aims of this work is to provide such a derivation in the case of a linear dielectric.

Another problem related to *all* the above phenomenological approaches is the inclusion of losses into the system. In classical electromagnetism, it is well known that if we consider the full frequency spectrum, including the absorbing regions always associated with a dispersive medium, then the dielectric constant will be a complex quantity, whose real and imaginary parts are related by the Kramers-Kronig relations [3]. To our knowledge, this fundamental identity between dispersion and losses has never before been derived for a quantized electromagnetic field. The reason why an effective theory cannot accommodate losses is that, in contrast to the classical case, losses in quantum mechanics imply a coupling to a reservoir whose degrees of freedom have to be added to the Hamiltonian. This suggests that, in order to quantize the electromagnetic field in a dielectric in a way that is consistent with the Kramers-Kronig relations, one has to introduce the medium into the formalism explicitly. This should be done in such a way that the interaction between light and matter will generate both dispersion and damping of the light field. Indeed it is widely agreed that a fully canonical theory, valid for all the frequency spectrum including the regions close to the resonances of the medium, has to be based on a microscopic model [5, 11]. Very recently, we presented an outline of a one-dimensional version of such a theory [20]. In this paper, we shall expand the model to three dimensions, in-

clude the polarization of the electromagnetic field and give more details of the calculations. A rigorous approach to quantization in linear dielectrics has very recently acquired experimental significance with the work of Steinberg, Kwiat, and Chiao [21] on the propagation of single photons in dielectrics. Moreover, by solving the linear case exactly, our work provides a step towards a complete canonical theory, including dispersion, losses, and nonlinearities.

The paper is organized as follows. In Sec. II, we present our model of the dielectric, perform the canonical quantization, and obtain the Hamiltonian of the combined system. We diagonalize this Hamiltonian exactly in Sec. III and express the field operators in terms of its eigenoperators and of the complex dielectric constant of the medium in Sec. IV. In Sec. V we analyze the propagation of the light in the dielectric, express the fields in terms of space-dependent amplitudes, and obtain their spatial equations of evolution. In Sec. VI, we derive two interesting relationships between the group and phase velocity in the dielectric. We discuss the main results and conclude in Sec. VII. Finally, we present an outline of the somewhat lengthy calculations in the Appendixes.

II. CANONICAL QUANTIZATION

Our microscopic model is based on the Hopfield model of a dielectric [22], where the matter is represented by a harmonic polarization field. This model was introduced by Fano [23], who justified it in terms of an atomic medium. However, to our knowledge, it was only applied to solid-state physics [24], where the main interest is in the properties of the medium and where the electromagnetic field is introduced as a probe of these properties. Here, we shall change the emphasis and use the model to obtain the properties of the light field in the medium. We also introduce an extra coupling between the polarization field and other harmonic fields and show that this additional element is responsible for the absorption of light.

The approach to canonical quantization that we use in this section follows closely the one adopted by Cohen-Tannoudji, Dupont-Roc, and Grynberg [25] and we refer the reader to this book for a more complete derivation. In order to emphasize a few significant differences and to set the notation, we shall nevertheless repeat the main points.

A. Separation into transverse and longitudinal parts

Following the standard approach in quantum electrodynamics [25], we start from a Lagrangian density in real space [all the vectors are written in boldface characters and, for notational simplicity, we do not write the (\mathbf{r}, t) dependence of the fields explicitly]:

$$\mathcal{L} = \mathcal{L}_{\text{em}} + \mathcal{L}_{\text{mat}} + \mathcal{L}_{\text{res}} + \mathcal{L}_{\text{int}} , \quad (2.1)$$

where

$$\mathcal{L}_{\text{em}} = \frac{\epsilon_0}{2} \mathbf{E}^2 - \frac{1}{2\mu_0} \mathbf{B}^2 \quad (2.2)$$

is the electromagnetic part which can be expressed in terms of the vector potential \mathbf{A} and the scalar potential U ($\mathbf{E} = -\dot{\mathbf{A}} - \nabla U$ and $\mathbf{B} = \nabla \times \mathbf{A}$);

$$\mathcal{L}_{\text{mat}} = \frac{\rho}{2} \dot{\mathbf{X}}^2 - \frac{\rho\omega_0^2}{2} \mathbf{X}^2 \quad (2.3)$$

is the polarization part, modeled by a harmonic oscillator field \mathbf{X} of frequency ω_0 ;

$$\mathcal{L}_{\text{res}} = \int_0^\infty d\omega \left(\frac{\rho}{2} \dot{\mathbf{Y}}_\omega^2 - \frac{\rho\omega^2}{2} \mathbf{Y}_\omega^2 \right) \quad (2.4)$$

is the reservoir part, comprising a continuum of harmonic oscillators, used to model the losses; and

$$\mathcal{L}_{\text{int}} = -\alpha \left(\mathbf{A} \cdot \dot{\mathbf{X}} + U \nabla \cdot \mathbf{X} \right) - \int_0^\infty d\omega v(\omega) \mathbf{X} \cdot \dot{\mathbf{Y}}_\omega \quad (2.5)$$

is the interaction part, which includes the interaction between the light and the polarization field, with coupling constant α , and the interaction between the polarization field and the other oscillator fields used to model the losses with coupling constant $v(\omega)$. In this paper, we shall restrict ourselves to a nonsingular, square-integrable coupling. Moreover, we shall make the following assumptions: (i) the analytic continuation of $|v(\omega)|^2$ to negative frequencies is an even function and (ii) $v(\omega) \neq 0$ for all nonzero frequencies. The first assumption is needed in order to extend the frequency integrals to the negative real axis and use integration in the complex plane, while the second one ensures that all the reservoir fields are coupled to the system. As we are primarily interested in the optical region, which means that $v(\omega)$ is significantly nonzero only far from the origin, the choice of an even function is not really restrictive. The choice of a particular $\mathbf{X} \cdot \dot{\mathbf{Y}}_\omega$ coupling for the loss term is not essential (any linear coupling would also lead to a loss term), but leads to some simplification in the calculations. For future use, we also define the displacement field $\mathbf{D}(\mathbf{r}, t)$, which is given by the following combination of the electric field and the material polarization:

$$\mathbf{D}(\mathbf{r}, t) = \epsilon_0 \mathbf{E}(\mathbf{r}, t) - \alpha \mathbf{X}(\mathbf{r}, t). \quad (2.6)$$

As \dot{U} does not appear in the Lagrangian, U is not a proper dynamical variable, but it can be written in terms of the proper dynamical variables \mathbf{A} , \mathbf{X} , and \mathbf{Y}_ω . The easiest way to do so is to go to the reciprocal space and write all the fields in terms of their spatial Fourier transforms. For example the electric field is written (to differentiate between the fields in real and reciprocal space, we shall underline the latter):

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{(2\pi)^{3/2}} \int d^3k \underline{\mathbf{E}}(\mathbf{k}, t) e^{i\mathbf{k} \cdot \mathbf{r}}. \quad (2.7)$$

The introduction of a complex field $\underline{\mathbf{E}}(\mathbf{k}, t)$ to represent a real field $\mathbf{E}(\mathbf{r}, t)$ implies that we double the number of variables. However, as $\underline{\mathbf{E}}^*(\mathbf{k}, t) = \underline{\mathbf{E}}(-\mathbf{k}, t)$, we can recover the correct number of variables by restricting the k integration to a half space [25]. The total Lagrangian is then obtained as

$$L = \int' d^3k \left(\underline{\mathcal{L}}_{\text{em}} + \underline{\mathcal{L}}_{\text{mat}} + \underline{\mathcal{L}}_{\text{res}} + \underline{\mathcal{L}}_{\text{int}} \right), \quad (2.8)$$

where the prime means that the integration is restricted to half the reciprocal space and the Lagrangian densities in this space are defined by

$$\begin{aligned} \underline{\mathcal{L}}_{\text{em}} &= \epsilon_0 \left(|\underline{\mathbf{E}}|^2 - c^2 |\underline{\mathbf{B}}|^2 \right), \\ \underline{\mathcal{L}}_{\text{mat}} &= \rho |\underline{\dot{\mathbf{X}}}|^2 - \rho\omega_0^2 |\underline{\mathbf{X}}|^2, \end{aligned} \quad (2.9)$$

$$\begin{aligned} \underline{\mathcal{L}}_{\text{res}} &= \int_0^\infty d\omega \left(\rho |\underline{\dot{\mathbf{Y}}}_\omega|^2 - \rho\omega^2 |\underline{\mathbf{Y}}_\omega|^2 \right), \\ \underline{\mathcal{L}}_{\text{int}} &= -\alpha \left[\underline{\mathbf{A}}^* \cdot \underline{\dot{\mathbf{X}}} + \underline{\mathbf{A}} \cdot \underline{\dot{\mathbf{X}}}^* + i\mathbf{k} \cdot (\underline{U}^* \underline{\mathbf{X}} - \underline{U} \underline{\mathbf{X}}^*) \right] \\ &\quad - \int_0^\infty d\omega v(\omega) \left(\underline{\mathbf{X}}^* \cdot \underline{\dot{\mathbf{Y}}}_\omega + \underline{\mathbf{X}} \cdot \underline{\dot{\mathbf{Y}}}_\omega^* \right). \end{aligned}$$

Following the standard approach to QED for non-relativistic phenomena, we choose the Coulomb gauge $\mathbf{k} \cdot \underline{\mathbf{A}}(\mathbf{k}, t) = 0$, so that the vector potential \mathbf{A} is a purely transverse field and use the Euler-Lagrange equation for \underline{U}^* to eliminate \underline{U} from the Lagrangian. We obtain

$$\underline{U}(\mathbf{k}, t) = i \frac{\alpha}{\epsilon_0} \left(\frac{\boldsymbol{\kappa} \cdot \underline{\mathbf{X}}(\mathbf{k}, t)}{\mathbf{k}} \right), \quad (2.10)$$

where $\boldsymbol{\kappa}$ is a unit vector in the direction of \mathbf{k} . Next we decompose the matter fields $\underline{\mathbf{X}}$ and $\underline{\mathbf{Y}}_\omega$ into transverse and longitudinal parts. For example $\underline{\mathbf{X}}$ can be written

$$\underline{\mathbf{X}}(\mathbf{k}, t) = \underline{\mathbf{X}}^\top(\mathbf{k}, t) + \underline{\mathbf{X}}^\parallel(\mathbf{k}, t) \boldsymbol{\kappa}, \quad (2.11)$$

with a similar expression for $\underline{\mathbf{Y}}_\omega$. The total Lagrangian can then be written as the sum of two independent parts. The transverse part, containing only transverse fields, is given by

$$L^\top = \int' d^3k \left(\underline{\mathcal{L}}_{\text{em}}^\top + \underline{\mathcal{L}}_{\text{mat}}^\top + \underline{\mathcal{L}}_{\text{res}}^\top + \underline{\mathcal{L}}_{\text{int}}^\top \right), \quad (2.12)$$

with

$$\begin{aligned} \underline{\mathcal{L}}_{\text{em}}^\top &= \epsilon_0 \left(|\underline{\dot{\mathbf{A}}}|^2 - c^2 k^2 |\underline{\mathbf{A}}|^2 \right), \\ \underline{\mathcal{L}}_{\text{mat}}^\top &= \left[\rho |\underline{\dot{\mathbf{X}}}^\top|^2 - \rho\omega_0^2 |\underline{\mathbf{X}}^\top|^2 \right], \end{aligned} \quad (2.13)$$

$$\begin{aligned} \underline{\mathcal{L}}_{\text{res}}^\top &= \int_0^\infty d\omega \left(\rho |\underline{\dot{\mathbf{Y}}}_\omega^\top|^2 - \rho\omega^2 |\underline{\mathbf{Y}}_\omega^\top|^2 \right), \\ \underline{\mathcal{L}}_{\text{int}}^\top &= - \left[\alpha \underline{\mathbf{A}} \cdot \underline{\dot{\mathbf{X}}}^{\top*} + \int_0^\infty d\omega v(\omega) \underline{\mathbf{X}}^{\top*} \cdot \underline{\dot{\mathbf{Y}}}_\omega^\top + \text{c.c.} \right], \end{aligned}$$

and the longitudinal part, containing only longitudinal fields, is written

$$L^\parallel = \int' d^3k \underline{\mathcal{L}}^\parallel, \quad (2.14)$$

where

$$\begin{aligned} \underline{\mathcal{L}}^{\parallel} = & \rho |\dot{\underline{\mathbf{X}}}^{\parallel}|^2 - \rho \omega_L^2 |\underline{\mathbf{X}}^{\parallel}|^2 + \int_0^{\infty} d\omega \left(\rho |\dot{\underline{\mathbf{Y}}}_{\omega}^{\parallel}|^2 - \rho \omega^2 |\underline{\mathbf{Y}}_{\omega}^{\parallel}|^2 \right) \\ & - \int_0^{\infty} d\omega v(\omega) \left(\underline{\mathbf{X}}^{\parallel*} \cdot \dot{\underline{\mathbf{Y}}}_{\omega}^{\parallel} + \underline{\mathbf{X}}^{\parallel} \cdot \dot{\underline{\mathbf{Y}}}_{\omega}^{\parallel*} \right), \end{aligned} \quad (2.15)$$

where ω_L , the longitudinal frequency, is defined by $\omega_L = \sqrt{\omega_0^2 + \omega_c^2}$ and $\omega_c^2 = \frac{\alpha^2}{\rho \epsilon_0}$. In contrast with the more standard approach [25], the decomposition of the *matter* field into transverse and longitudinal parts allows a complete separation between the transverse and the longitudinal parts of the Lagrangian. The link between the two is given by the total electric field, which is written

$$\underline{\mathbf{E}}(\mathbf{k}, t) \equiv \underline{\mathbf{E}}^{\top}(\mathbf{k}, t) + \underline{\mathbf{E}}^{\parallel}(\mathbf{k}, t) \boldsymbol{\kappa} = -\dot{\underline{\mathbf{A}}}(\mathbf{k}, t) + \frac{\alpha}{\epsilon_0} \underline{\mathbf{X}}^{\parallel} \boldsymbol{\kappa}. \quad (2.16)$$

Using (2.11), (2.16), and the definition of the displacement field $\underline{\mathbf{D}}$ given in (2.6), we recover the fact that, as expected, $\underline{\mathbf{D}}(\mathbf{r}, t)$ is also a purely transverse field.

From (2.12)–(2.15), it is easy to see that the longitudinal Lagrangian is similar to one component of the *matter* part of the transverse Lagrangian (including the polarization field, the reservoir and their interaction) with a change of frequency from ω_0 to ω_L . Its quantization can thus be performed in exactly the same way as the quantization of the transverse part, but without introducing the vector potential $\underline{\mathbf{A}}$. In this work, we are mainly interested in the transverse fields representing the propagating photons in the dielectric and shall only present the detailed quantization of the transverse part of the Lagrangian. Quantization of the longitudinal part follows exactly the same lines, but we shall not do it here. In all that follows, we shall therefore restrict ourselves to transverse fields and omit the superscript \top .

B. Quantization of the transverse fields

We introduce unit polarization vectors $\mathbf{e}_{\lambda}(\mathbf{k})$, $\lambda = 1, 2$, which are orthogonal to $\boldsymbol{\kappa}$ and to one another, and decompose the transverse fields along them to get

$$\underline{\mathbf{A}}(\mathbf{k}, t) = \sum_{\lambda=1,2} \underline{\mathbf{A}}^{\lambda}(\mathbf{k}, t) \mathbf{e}_{\lambda}(\mathbf{k}), \quad (2.17)$$

with similar expressions for the other fields. $\underline{\mathcal{L}}$ can now be used to obtain the components of the conjugate variables for the fields

$$-\epsilon_0 \underline{\mathbf{E}}^{\lambda} \equiv \frac{\partial \underline{\mathcal{L}}}{\partial \dot{\underline{\mathbf{A}}}^{\lambda*}} = \epsilon_0 \dot{\underline{\mathbf{A}}}^{\lambda}, \quad (2.18a)$$

$$\underline{\mathbf{P}}^{\lambda} \equiv \frac{\partial \underline{\mathcal{L}}}{\partial \dot{\underline{\mathbf{X}}}^{\lambda*}} = \rho \dot{\underline{\mathbf{X}}}^{\lambda} - \alpha \underline{\mathbf{A}}^{\lambda}, \quad (2.18b)$$

$$\underline{\mathbf{Q}}_{\omega}^{\lambda} \equiv \frac{\partial \underline{\mathcal{L}}}{\partial \dot{\underline{\mathbf{Y}}}_{\omega}^{\lambda*}} = \rho \dot{\underline{\mathbf{Y}}}_{\omega}^{\lambda} - v(\omega) \underline{\mathbf{X}}^{\lambda}. \quad (2.18c)$$

One important point is that with this particular type of coupling between light and matter, the conjugate of $\underline{\mathbf{A}}$ is the transverse electric field $-\epsilon_0 \underline{\mathbf{E}}$. A change of gauge, leading to an $\underline{\mathbf{E}} \cdot \underline{\mathbf{X}}$ type of coupling, would give $-\underline{\mathbf{D}}$,

the displacement field, as the conjugate to $\underline{\mathbf{A}}$. Naturally, these two possibilities lead to the same results and we have used the second one in a recent work dealing with a lossless medium [12]. We choose here the first possibility in order to keep as close as possible to the classical theory, where $\underline{\mathbf{E}}$ is usually considered as the fundamental variable and $\underline{\mathbf{D}}$ is written in terms of $\underline{\mathbf{E}}$ and the dielectric response operator $\hat{\epsilon}$.

Using the Lagrangian (2.12) defined in (2.13) and the expression for the conjugate variables in (2.18), we obtain the Hamiltonian for the transverse fields,

$$H = \int d^3k \left(\mathcal{H}_{\text{em}} + \mathcal{H}_{\text{mat}} + \mathcal{H}_{\text{int}} \right), \quad (2.19)$$

where

$$\mathcal{H}_{\text{em}} = \left[\epsilon_0 |\underline{\mathbf{E}}|^2 + \epsilon_0 c^2 \tilde{k}^2 |\underline{\mathbf{A}}|^2 \right] \quad (2.20a)$$

is the electromagnetic energy density, \tilde{k} being defined by $\tilde{k} = \sqrt{k^2 + k_c^2}$ with $k_c \equiv \frac{\omega_c}{c} = \sqrt{\alpha^2 / \rho c^2 \epsilon_0}$;

$$\begin{aligned} \mathcal{H}_{\text{mat}} = & \frac{|\underline{\mathbf{P}}|^2}{\rho} + \rho \tilde{\omega}_0^2 |\underline{\mathbf{X}}|^2 \\ & + \int_0^{\infty} d\omega \left[\frac{|\underline{\mathbf{Q}}_{\omega}|^2}{\rho} + \rho \omega^2 |\underline{\mathbf{Y}}_{\omega}|^2 \right. \\ & \left. + \frac{v(\omega)}{\rho} \left(\underline{\mathbf{X}}^* \cdot \underline{\mathbf{Q}}_{\omega} + \text{c.c.} \right) \right], \end{aligned} \quad (2.20b)$$

is the energy density of the matter fields, including the interaction between the polarization and the reservoir and $\tilde{\omega}_0^2 \equiv \omega_0^2 + \int_0^{\infty} d\omega \frac{v(\omega)^2}{\rho^2}$ is the renormalized frequency of the polarization field;

$$\mathcal{H}_{\text{int}} = \frac{\alpha}{\rho} (\underline{\mathbf{A}}^* \cdot \underline{\mathbf{P}} + \text{c.c.}) \quad (2.20c)$$

is the interaction energy between the electromagnetic field and the polarization. We note that in this approach the electromagnetic energy in (2.20a) already includes part of the interaction energy with the matter, namely $\frac{\alpha^2}{\rho} |\underline{\mathbf{A}}|^2$. In standard quantization schemes, this energy is included in the interaction Hamiltonian. Here, as our aim is to include the matter from the beginning and derive its influence exactly, our decomposition is preferable.

The fields are quantized in a standard fashion [25] by demanding equal-time commutation relations (ETCR) between the variables and their conjugates. For the electromagnetic field components, we get

$$\left[\hat{\underline{\mathbf{A}}}^{\lambda}(\mathbf{k}, t), \hat{\underline{\mathbf{E}}}^{\lambda*}(\mathbf{k}', t) \right] = -\frac{i\hbar}{\epsilon_0} \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}'), \quad (2.21a)$$

and for the matter fields

$$\left[\hat{\underline{\mathbf{X}}}^{\lambda}(\mathbf{k}, t), \hat{\underline{\mathbf{P}}}^{\lambda*}(\mathbf{k}', t) \right] = i\hbar \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}'), \quad (2.21b)$$

$$\left[\hat{\underline{\mathbf{Y}}}_{\omega}^{\lambda}(\mathbf{k}, t), \hat{\underline{\mathbf{Q}}}_{\omega'}^{\lambda*}(\mathbf{k}', t) \right] = i\hbar \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'), \quad (2.21c)$$

with all the other equal-time commutators between fields within the half k space we are integrating over being zero. From now on, we shall denote all quantized operators by a caret.

To facilitate calculations, we introduce three sets of annihilation operators:

$$\hat{a}(\lambda, \mathbf{k}, t) = \sqrt{\frac{\epsilon_0}{2\hbar\tilde{k}c}} \left[\tilde{k}c\hat{A}^\lambda(\mathbf{k}, t) - i\hat{E}^\lambda(\mathbf{k}, t) \right], \quad (2.22a)$$

$$\hat{b}(\lambda, \mathbf{k}, t) = \sqrt{\frac{\rho}{2\hbar\tilde{\omega}_0}} \left[\tilde{\omega}_0\hat{X}^\lambda(\mathbf{k}, t) + \frac{i}{\rho}\hat{P}^\lambda(\mathbf{k}, t) \right], \quad (2.22b)$$

$$\hat{b}_\omega(\lambda, \mathbf{k}, t) = \sqrt{\frac{\rho}{2\hbar\omega}} \left[-i\omega\hat{Y}^\lambda(\mathbf{k}, t) + \frac{1}{\rho}\hat{Q}^\lambda(\mathbf{k}, t) \right], \quad (2.22c)$$

where \tilde{k} and $\tilde{\omega}_0$ are defined in (2.20a) and (2.20b). The different definitions for \hat{b} and \hat{b}_ω only amount to a change of phase and have been chosen for future simplicity.

From the ETCR for the fields (2.21), we obtain the ETCR for the creation and annihilation operators:

$$\begin{aligned} [\hat{a}(\lambda, \mathbf{k}, t), \hat{a}^\dagger(\lambda', \mathbf{k}', t)] &= \delta_{\lambda\lambda'}\delta(\mathbf{k} - \mathbf{k}'), \\ [\hat{b}(\lambda, \mathbf{k}, t), \hat{b}^\dagger(\lambda', \mathbf{k}', t)] &= \delta_{\lambda\lambda'}\delta(\mathbf{k} - \mathbf{k}'), \\ [\hat{b}_\omega(\lambda, \mathbf{k}, t), \hat{b}_\omega^\dagger(\lambda', \mathbf{k}', t)] &= \delta_{\lambda\lambda'}\delta(\omega - \omega')\delta(\mathbf{k} - \mathbf{k}'). \end{aligned} \quad (2.23)$$

We emphasize that, in contrast to the previous ETCR between the conjugate fields (2.21), which were correct only in half k space, Eqs. (2.23) are valid in the whole reciprocal space. Inverting (2.22) to express the field operators in terms of the creation and annihilation operators, inserting these into the Hamiltonian (2.19), and performing the integration, we obtain the normally ordered Hamiltonian for the transverse fields

$$H \equiv H_{\text{em}} + H_{\text{mat}} + H_{\text{int}}, \quad (2.24)$$

where

$$\hat{H}_{\text{em}} = \int d^3k \sum_{\lambda=1,2} \hbar\tilde{k}c\hat{a}^\dagger(\lambda, \mathbf{k}, t)\hat{a}(\lambda, \mathbf{k}, t), \quad (2.25a)$$

$$\begin{aligned} H_{\text{mat}} = \int d^3k \sum_{\lambda=1,2} & \left[\hbar\tilde{\omega}_0\hat{b}^\dagger(\lambda, \mathbf{k}, t)\hat{b}(\lambda, \mathbf{k}, t) + \int_0^\infty d\omega \hbar\omega\hat{b}_\omega^\dagger(\lambda, \mathbf{k}, t)\hat{b}_\omega(\lambda, \mathbf{k}, t) \right. \\ & \left. + \frac{\hbar}{2} \int_0^\infty d\omega V(\omega)[\hat{b}^\dagger(\lambda, -\mathbf{k}, t) + \hat{b}(\lambda, \mathbf{k}, t)][\hat{b}_\omega^\dagger(\lambda, -\mathbf{k}, t) + \hat{b}_\omega(\lambda, \mathbf{k}, t)] \right], \end{aligned} \quad (2.25b)$$

$$H_{\text{int}} = i\frac{\hbar}{2} \int d^3k \sum_{\lambda=1,2} \Lambda(k)[\hat{a}^\dagger(\lambda, -\mathbf{k}, t) + \hat{a}(\lambda, \mathbf{k}, t)][\hat{b}^\dagger(\lambda, -\mathbf{k}, t) - \hat{b}(\lambda, \mathbf{k}, t)], \quad (2.25c)$$

where $V(\omega) \equiv [v(\omega)/\rho]\sqrt{\omega/\tilde{\omega}_0}$, $\Lambda(k) \equiv \sqrt{\tilde{\omega}_0ck_c^2/\tilde{k}}$ and the k integration has been extended to the full reciprocal space. From the original assumptions (i) and (ii) on $v(\omega)$, we see that the analytic continuation of $|V(\omega)|^2$ to negative frequencies is an odd function and is nonzero everywhere except at the origin.

The above equations (2.24) and (2.25), together with the commutation relations in (2.23), complete the quantization procedure of the transverse fields. It is interesting to note that the explicit introduction of the matter in our model has enabled us to perform the quantization in a perfectly standard fashion and to avoid the problem of nonlocality in time present in effective quantum schemes, where the medium is only introduced by its response operator $\hat{\epsilon}$. The apparent drawback of our procedure is that it is still purely formal. Indeed, it is easily shown that the Heisenberg equations of evolution based on the Hamiltonian (2.24) and the ETCR (2.23) are identical to the Maxwell-Lorentz equations for the coupled matter-electromagnetic field system. In order to extract useful information about the system, we need to solve these equations. This is the aim of Sec. III, where we present the exact diagonalization of the Hamiltonian (2.24).

III. DIAGONALIZATION OF THE HAMILTONIAN

A. Diagonalization of \hat{H}_{mat}

The polarization and reservoir part of the Hamiltonian \hat{H}_{mat} (2.25b) can be diagonalized by a Fano type of technique (discrete mode coupled to a continuum) [26, 27] to give a dressed matter field. An important point here is that, as we are interested in the full frequency spectrum, we cannot make the rotating-wave approximation but have to keep all the contributions. The calculations leading to the diagonalization are lengthy and we outline them in Appendix A. For notational simplicity, we shall omit the polarization index λ and the explicit time dependence of the operators. The diagonalized expression for \hat{H}_{mat} is

$$\hat{H}_{\text{mat}} = \int d^3k \int_0^\infty d\omega \hbar\omega\hat{B}^\dagger(\mathbf{k}, \omega)\hat{B}(\mathbf{k}, \omega), \quad (3.1)$$

where $\hat{B}^\dagger(\mathbf{k}, \omega)$ and $\hat{B}(\mathbf{k}, \omega)$ are the dressed matter field creation and annihilation operator which satisfy the usual ETCR

$$[\hat{B}(\mathbf{k}, \omega), \hat{B}^\dagger(\mathbf{k}', \omega')] = \delta(\mathbf{k} - \mathbf{k}')\delta(\omega - \omega'). \quad (3.2)$$

They are expressed in terms of the initial creation and annihilation operators by

$$\begin{aligned} \hat{B}(\mathbf{k}, \omega) = & \alpha_0(\omega)\hat{b}(\mathbf{k}) + \beta_0(\omega)\hat{b}^\dagger(-\mathbf{k}) \\ & + \int_0^\infty d\omega' \left[\alpha_1(\omega, \omega')\hat{b}_{\omega'}(\mathbf{k}) \right. \\ & \left. + \beta_1(\omega, \omega')\hat{b}_{\omega'}^\dagger(-\mathbf{k}) \right], \end{aligned} \quad (3.3)$$

and all the coefficients $\alpha_0(\omega)$, $\beta_0(\omega)$, $\alpha_1(\omega, \omega')$, and $\beta_1(\omega, \omega')$ are defined in Appendix A. We emphasize that, as the parameters $\tilde{\omega}_0$ and $V(\omega)$ in the Hamiltonian \hat{H}_{mat} (2.25b) do not depend on \mathbf{k} , the above coefficients are also independent of \mathbf{k} .

It is also important to note that, in the above diagonalization, we have not proven that our set of eigenoperators is complete. In particular, it is well known that the coupling of even two harmonic oscillators can result in eigenmodes with negative energies when the coupling is too strong. Here, we have assumed that only positive frequencies appear in the expression for the diagonalized Hamiltonian (3.1). This assumption was needed in order to avoid problems such as unboundedness that appear in harmonic oscillators with negative energies. However, we still have to check its validity. A consistency check, which will in fact give us an upper limit for the coupling factor $V(\omega)$, can be found by inverting (3.3) to write the free creation and annihilation operators \hat{b} and \hat{b}_ω and their conjugates in terms of the eigen- or dressed operators \hat{B} and \hat{B}^\dagger . If the set is complete, then it will be possible to construct \hat{b} and \hat{b}_ω as well as their commutators in terms of the dressed operators. Using the commutators of \hat{b} and \hat{b}_ω with \hat{B} and \hat{B}^\dagger together with (3.3), it is easily seen that

$$\hat{b}(\mathbf{k}) = \int_0^\infty d\omega \left[\alpha_0^*(\omega)\hat{B}(\mathbf{k}, \omega) - \beta_0(\omega)\hat{B}^\dagger(-\mathbf{k}, \omega) \right], \quad (3.4)$$

and similarly

$$\hat{b}_\omega(\mathbf{k}) = \int_0^\infty d\omega' \left[\alpha_1^*(\omega', \omega)\hat{B}(\mathbf{k}, \omega') \right. \\ \left. - \beta_1(\omega', \omega)\hat{B}^\dagger(-\mathbf{k}, \omega') \right]. \quad (3.5)$$

The consistency of the diagonalization procedure is checked by verifying that the initial commutation relations between $\hat{b}(\mathbf{k})$ and $\hat{b}^\dagger(\mathbf{k})$ and between $\hat{b}_\omega(\mathbf{k})$ and $\hat{b}_\omega^\dagger(\mathbf{k})$ are conserved. Using (3.4), (3.5), and the commutation relations for the \hat{B} 's (3.2), we obtain

$$[\hat{b}(\mathbf{k}), \hat{b}^\dagger(\mathbf{k}')] = \delta(\mathbf{k} - \mathbf{k}') \int_0^\infty d\omega [|\alpha_0(\omega)|^2 - |\beta_0(\omega)|^2], \quad (3.6)$$

$$[\hat{b}_\omega(\mathbf{k}), \hat{b}_{\omega'}^\dagger(\mathbf{k}')] = \delta(\mathbf{k} - \mathbf{k}') \int_0^\infty d\nu \left[\alpha_1^*(\nu, \omega)\alpha_1(\nu, \omega') \right. \\ \left. - \beta_1(\nu, \omega)\beta_1^*(\nu, \omega') \right]. \quad (3.7)$$

Equations (3.6) and (3.7) show that the commutation relations are conserved by the transformation from the free operators to the dressed ones if and only if the two following conditions are satisfied:

$$I \equiv \int_0^\infty d\omega [|\alpha_0(\omega)|^2 - |\beta_0(\omega)|^2] = 1, \quad (3.8)$$

$$I(\omega, \omega') \equiv \int_0^\infty d\nu \left[\alpha_1^*(\nu, \omega)\alpha_1(\nu, \omega') - \beta_1(\nu, \omega)\beta_1^*(\nu, \omega') \right] \\ = \delta(\omega - \omega'). \quad (3.9)$$

These integrals are calculated in Appendix B where it is shown that the scheme is consistent: $I = 1$ and $I(\omega, \omega') = \delta(\omega - \omega')$ if and only if $|V(\omega)|^2$ satisfies the inequality:

$$\int_0^\infty d\omega \frac{|V(\omega)|^2}{\omega} < \tilde{\omega}_0. \quad (3.10)$$

Using the definition of $\tilde{\omega}_0$ and of $V(\omega)$ in terms of the coupling $v(\omega)$ given in (2.20b) and (2.25b), it is easily seen that (3.10) is indeed satisfied for any square-integrable coupling $v(\omega)$, provided that the frequency of the polarization field ω_0 is not zero. This shows that the above procedure is restricted to dielectric material in which there are no free charges. The treatment of a conducting medium would require an extension of the model to account for the longitudinal component of \mathbf{D} arising from the free charges.

The main result of this section is that, subject to the above restriction, the Hamiltonian H_{mat} is diagonalizable for any square-integrable coupling $v(\omega)$ satisfying the original assumptions (i) and (ii). Moreover, the above diagonalization procedure, which was performed here for a one-resonance model of a dielectric, can be iterated to treat any number of resonances. Indeed, the original technique which forms the basis of our scheme has already been extended to treat any number of discrete modes coupled to one continuum [26, 27].

B. Diagonalization of \hat{H}

The diagonalization of the total Hamiltonian \hat{H} (2.24) is formally very similar to the above diagonalization of the matter part \hat{H}_{mat} . We write \hat{H}_{mat} (2.25b) in diagonal form (3.1) and use (3.4) to replace the $\hat{b}(\mathbf{k})$ operators in the interaction part of the Hamiltonian \hat{H}_{int} (2.25b) by their expression in terms of the dressed matter operators $\hat{B}(\mathbf{k}, \omega)$ to give

$$\begin{aligned} \hat{H} = & \int d^3k \left\{ \hbar \tilde{\omega}_0 \hat{a}^\dagger(\mathbf{k}) \hat{a}(\mathbf{k}) + \int_0^\infty d\omega \hbar \omega \hat{B}^\dagger(\mathbf{k}, \omega) \hat{B}(\mathbf{k}, \omega) \right. \\ & \left. + \frac{\hbar}{2} \Lambda(\mathbf{k}) \int_0^\infty d\omega \{ g(\omega) \hat{B}^\dagger(\mathbf{k}, \omega) [\hat{a}(\mathbf{k}) + \hat{a}^\dagger(-\mathbf{k})] + \text{H.c.} \} \right\}, \end{aligned} \quad (3.11)$$

where $g(\omega) \equiv i\alpha_0(\omega) + i\beta_0(\omega)$. It will prove useful to define a dimensionless coupling constant $\zeta(\omega) = \sqrt{\omega_0}g(\omega)$. The Hamiltonian is then written as

$$\begin{aligned} \hat{H} = \int d^3k \left\{ \hbar \tilde{k}c \hat{a}^\dagger(\mathbf{k})\hat{a}(\mathbf{k}) + \int_0^\infty d\omega \hbar\omega \hat{B}^\dagger(\mathbf{k}, \omega)\hat{B}(\mathbf{k}, \omega) \right. \\ \left. + \frac{\hbar}{2} \sqrt{\frac{\omega_c^2}{\tilde{k}c}} \int_0^\infty d\omega \{ \zeta(\omega)\hat{B}^\dagger(\mathbf{k}, \omega) [\hat{a}(\mathbf{k}) + \hat{a}^\dagger(-\mathbf{k})] + \text{H.c.} \} \right\}. \end{aligned} \quad (3.12)$$

Direct comparison between the above Hamiltonian (3.12) and the expression for \hat{H}_{mat} in (2.25b) shows that the two have the same structure. Moreover, we show in Appendix C that the coupling $\zeta(\omega)$, which corresponds to $V(\omega)$ in (2.25b), satisfies the same condition as $V(\omega)$, namely that the analytic continuation of $|\zeta(\omega)|^2$ to negative frequencies is an odd function, nonzero everywhere on the axis, except at $\omega = 0$. We also derive the following normalization condition:

$$\int_0^\infty d\omega |\zeta(\omega)|^2 / \omega = 1. \quad (3.13)$$

The diagonalization procedure of \hat{H} follows the one given in Sec. III A and in Appendix A, with the relevant changes in the parameters. The final expression for \hat{H} is

$$\hat{H} = \int d^3k \int_0^\infty d\omega \hbar\omega \hat{C}^\dagger(\mathbf{k}, \omega)\hat{C}(\mathbf{k}, \omega), \quad (3.14)$$

where \hat{C}^\dagger and \hat{C} are the creation and annihilation operators for the eigenmodes of the system, the excitations of which are known as polaritons. They are defined in terms of the $\hat{a}(\mathbf{k})$ and $\hat{B}(\mathbf{k}, \omega)$ operators and their Hermitian conjugates by

$$\begin{aligned} \hat{C}(\mathbf{k}, \omega) = \tilde{\alpha}_0(k, \omega)\hat{a}(\mathbf{k}) + \tilde{\beta}_0(k, \omega)\hat{a}^\dagger(-\mathbf{k}) \\ + \int_0^\infty d\omega' [\tilde{\alpha}_1(k, \omega, \omega')\hat{B}(\mathbf{k}, \omega') \\ + \tilde{\beta}_1(k, \omega, \omega')\hat{B}^\dagger(-\mathbf{k}, \omega')], \end{aligned} \quad (3.15)$$

where the coefficients $\tilde{\alpha}_0(\mathbf{k}, \omega)$, $\tilde{\beta}_0(\mathbf{k}, \omega)$, $\tilde{\alpha}_1(\mathbf{k}, \omega, \omega')$, and $\tilde{\beta}_1(\mathbf{k}, \omega, \omega')$ are given in Appendix D. The $\hat{C}(\mathbf{k}, \omega)$ and $\hat{C}^\dagger(\mathbf{k}, \omega)$ also satisfy the usual commutation relations (CR)

$$[\hat{C}(\mathbf{k}, \omega), \hat{C}^\dagger(\mathbf{k}', \omega')] = \delta(\mathbf{k} - \mathbf{k}')\delta(\omega - \omega'), \quad (3.16)$$

and, being operators for eigenmodes, they have a harmonic time dependence

$$\hat{\mathbf{A}}(\mathbf{r}, t) = \frac{1}{(2\pi)^{3/2}} \int d^3k \sqrt{\frac{\hbar}{2\epsilon_0 \tilde{k}c}} \sum_{\lambda=1,2} \mathbf{e}_\lambda(\mathbf{k}) [\hat{a}(\lambda, \mathbf{k}, t)e^{i\mathbf{k}\cdot\mathbf{r}} + \hat{a}^\dagger(\lambda, \mathbf{k}, t)e^{-i\mathbf{k}\cdot\mathbf{r}}]. \quad (4.1)$$

Following the method used in (3.4) to derive the polarization field operators \hat{b} and \hat{b}^\dagger in terms of the dressed matter operators \hat{B} and \hat{B}^\dagger , we invert (3.15) to write the photon creation and annihilation operators \hat{a}^\dagger and \hat{a} in terms of the polariton operators \hat{C} and \hat{C}^\dagger to give

$$\hat{a}(\lambda, \mathbf{k}, t) = \int_0^\infty d\omega [\tilde{\alpha}_0^*(k, \omega)\hat{C}(\lambda, \mathbf{k}, \omega)e^{-i\omega t} - \tilde{\beta}_0(k, \omega)\hat{C}^\dagger(\lambda, -\mathbf{k}, \omega)e^{i\omega t}], \quad (4.2)$$

$$\hat{C}(\mathbf{k}, \omega, t) = \hat{C}(\mathbf{k}, \omega, 0)e^{-i\omega t}. \quad (3.17)$$

Finally, following the results of Sec. III A, we find the upper bound on the coupling $\zeta(\omega)$, corresponding to (3.10) in Sec. III A, which reads

$$\frac{\omega_c^2}{\tilde{k}c} \int_0^\infty d\omega \frac{|\zeta(\omega)|^2}{\omega} < \tilde{k}c. \quad (3.18)$$

The definition of \tilde{k} , $\tilde{k}^2c^2 = k^2c^2 + \omega_c^2$ (2.20a), together with (3.13), ensures that (3.18) is indeed satisfied for all nonzero k 's [28]. Therefore, subject only to the initial assumptions on $v(\omega)$, the above procedure is exact; the total Hamiltonian is diagonalizable and its eigenoperators are given by (3.15). Moreover, the above consistency check leads to the following equality:

$$\int_0^\infty d\omega [|\tilde{\alpha}_0(k, \omega)|^2 - |\tilde{\beta}_0(k, \omega)|^2] = 1, \quad (3.19)$$

which corresponds to (3.8) in Sec. III A and is satisfied for all $k \neq 0$. We shall show in Sec. VI that (3.19) leads to two interesting relationships between the group and the phase velocities in the dielectric. We shall use our results to derive expressions for the electromagnetic field operators in the dielectric in terms of the eigenoperators.

IV. EXPRESSION FOR THE ELECTROMAGNETIC FIELD OPERATORS

In order to obtain the electromagnetic field operators in terms of the polariton creation and annihilation operators \hat{C}^\dagger and \hat{C} , we first use the results of Sec. II and in particular (2.22a) to express them in terms of the photon creation and annihilation operators $\hat{a}^\dagger(\lambda, \mathbf{k}, t)$ and $\hat{a}(\lambda, \mathbf{k}, t)$. The vector potential operator $\hat{\mathbf{A}}(\mathbf{r}, t)$ is given by (note that we now write the explicit polarization and time dependence)

where we have included the time dependence of \hat{C} and its conjugate explicitly. Using (4.1), (4.2), and the expression for the coefficients $\tilde{\alpha}_0$ and $\tilde{\beta}_0$ derived in Appendix D, we obtain the vector potential operator in terms of the eigenoperators:

$$\hat{\mathbf{A}}(\mathbf{r}, t) = \frac{1}{(2\pi)^{3/2}} \int d^3k \sqrt{\frac{\hbar\omega_c^2}{2\epsilon_0}} \sum_{\lambda=1,2} \mathbf{e}_\lambda(\mathbf{k}) \int_0^\infty d\omega \left[\frac{\zeta^*(\omega)}{\omega^2\epsilon(\omega) - k^2c^2} \hat{C}(\mathbf{k}, \omega) e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})} + \text{H.c.} \right], \quad (4.3)$$

where $\epsilon(\omega)$, which we shall interpret as the complex dielectric constant of the medium, is defined by

$$\epsilon(\omega) = 1 + \frac{\omega_c^2}{2\omega} \left[P \int_{-\infty}^\infty d\omega' \frac{\xi(\omega')}{\omega' - \omega} + i\pi\xi(\omega) \right], \quad (4.4)$$

with $\xi(\omega) \equiv \frac{|\zeta(\omega)|^2}{\omega}$ for positive frequencies and its analytic continuation for negative frequencies. The full justification of this interpretation shall be given later in this section and in Sec. V. It is shown in Appendix E that $\epsilon(\omega)$ has the usual properties of a dielectric constant [3], namely it is an analytic function in the upper half of the complex plane and it tends to one as ω tends to infinity and to a number larger than one when ω tends to zero. Most importantly, we show that it satisfies the Kramers-Kronig relations.

We can also calculate the transverse electric field ($\mathbf{E} = -\dot{\mathbf{A}}$)

$$\hat{\mathbf{E}}(\mathbf{r}, t) = \frac{i}{(2\pi)^{3/2}} \int d^3k \sqrt{\frac{\hbar\omega_c^2}{2\epsilon_0}} \sum_{\lambda=1,2} \mathbf{e}_\lambda(\mathbf{k}) \int_0^\infty d\omega \left[\frac{\omega\zeta^*(\omega)}{\omega^2\epsilon(\omega) - k^2c^2} \hat{C}(\mathbf{k}, \omega) e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})} - \text{H.c.} \right] \quad (4.5)$$

and the magnetic field ($\mathbf{B} = \nabla \times \mathbf{A}$)

$$\hat{\mathbf{B}}(\mathbf{r}, t) = \frac{i}{(2\pi)^{3/2}} \int d^3k \sqrt{\frac{\hbar\omega_c^2}{2\epsilon_0}} \sum_{\lambda=1,2} \mathbf{k} \times \mathbf{e}_\lambda(\mathbf{k}) \int_0^\infty d\omega \left[\frac{\zeta^*(\omega)}{\omega^2\epsilon(\omega) - k^2c^2} \hat{C}(\mathbf{k}, \omega) e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})} - \text{H.c.} \right]. \quad (4.6)$$

The remaining important field appearing in the Maxwell equations (1.1) is the displacement field \mathbf{D} , defined in (2.6). The calculation of $\mathbf{D}(\mathbf{r}, t)$ is straightforward but lengthy, as we need first to express the polarization field $\hat{\mathbf{X}}(\mathbf{r}, t)$ in terms of the \hat{C} 's and then to insert the resulting expression in (2.6). After some algebra, we find

$$\hat{\mathbf{D}}(\mathbf{r}, t) = \frac{i}{(2\pi)^{3/2}} \int d^3k \sqrt{\frac{\hbar\omega_c^2}{2\epsilon_0}} \sum_{\lambda=1,2} \mathbf{e}_\lambda(\mathbf{k}) \int_0^\infty d\omega \left[\epsilon_0\epsilon(\omega) \frac{\omega\zeta^*(\omega)}{\omega^2\epsilon(\omega) - k^2c^2} \hat{C}(\mathbf{k}, \omega) e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})} - \epsilon_0 \frac{\zeta^*(\omega)}{\omega} \hat{C}(\mathbf{k}, \omega) e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})} - \text{H.c.} \right]. \quad (4.7)$$

The first term of (4.7) is equivalent to the classical result, where the link between \mathbf{D} and \mathbf{E} is given by the dielectric susceptibility. This justifies the interpretation of $\epsilon(\omega)$ as the dielectric constant of the medium. The second term, which has no classical equivalent, represents a Langevin fluctuation term as we shall show in Sec. V.

The set of equations (4.3), (4.5), (4.6), and (4.7) form a complete solution to the the problem of quantization in the dielectric. The electromagnetic field operators are written in terms of the eigenoperators of the system, with a known spatial and temporal dependence. An interesting point is that by coupling the light to an infinite number of oscillator fields, in the form of a dressed medium polarization, we lose the relationship between ω and k . Each of them has to be considered as an independent real variable. This is the natural limit to the phenomenon leading to different branches in the $\omega - k$ dispersion curve in the undamped Hopfield model [12, 22, 24]. There, the coupling between the field and a single-frequency harmonic polarization leads to two real values of ω for each real k . In our model, the coupling between the field and a material polarization with a *continuous*

spectrum of frequencies leads to a *continuum* of allowed real values of ω for each k , that is, independent real values for ω and k . Another interesting point is that the only parameters in the model are the coupling frequency ω_c , which characterizes the strength of the coupling between the electromagnetic field and the dressed matter field, and the dimensionless coupling $\zeta(\omega)$ which characterizes the frequency dependence of the coupling and is normalized by (3.13). This shows that the model is *not restricted to a single resonance* of the medium. Indeed, as the electromagnetic field is coupled to an infinite number of matter fields by a very general coupling constant $\zeta(\omega)$, the Hamiltonian (3.12) can represent the interaction of the electromagnetic field with *any* linear medium. The restrictions on the coupling $\zeta(\omega)$, namely that the analytic continuation of $|\zeta(\omega)|^2$ to negative frequencies has to be an odd function, nonzero everywhere except at the origin, are in fact necessary to get a dielectric constant $\epsilon(\omega)$ which satisfies causality requirement as expressed in the Kramers-Kronig relations. In this work, we decided to start from a Lagrangian density for a one-resonance model of the dielectric and to

perform explicitly a fully canonical quantization scheme. Our aim is to emphasize that this procedure is possible and that the difficulties associated with previous methods were only due to the use of effective schemes, which did not introduce the matter degrees of freedom. It is clear that the introduction of many resonances of the medium would lead to exactly the same type of Hamiltonian as (3.12), but the calculation of the coupling constant $\zeta(\omega)$ in terms of the parameters of the Lagrangian would be quite complicated and we believe unnecessary. For practical applications, it is now possible to follow the approach of classical electromagnetism, where the susceptibility of the medium is obtained as an experimental parameter, and to start from the Hamiltonian (3.12), with coupling constant ω_c and $\zeta(\omega)$ chosen to match the dielectric function $\epsilon(\omega)$. Very recently, we used this approach to calculate the spontaneous-emission rate of an excited atom embedded in a dielectric [29]. The set of equations (4.3)–(4.7) can also be used as a basis for treating various types of polariton-mediated interactions between embedded atoms or molecules [30].

V. PROPAGATION IN THE DIELECTRIC

Having solved the problem of quantization in the dielectric, we now apply our results to treat the propagation of a beam of light in the dielectric. The fundamental difficulty related to the propagation of a quantized electromagnetic field is that the usual decomposition of the field into spatial modes whose time evolution is given by Heisenberg equation of motion is not well adapted to this problem. In propagation, the significant property is frequency rather than wavelength. For example, at an interface, we impose boundary conditions on components with the same frequency. The natural quantities to use are therefore space and frequency dependent amplitudes or, in quantum optics, operators. This is the reason why, in most of the recent literature, authors have used an approach based on decomposing the field into temporal modes whose spatial evolution has to be calculated [15–19]. This approach closely follows clas-

sical electromagnetism, where the fields are decomposed into monochromatic amplitudes whose spatial evolution is obtained from Maxwell equations. However, quantization of these space-dependent amplitudes has been only phenomenological rather than derived from a canonical scheme. In this section, we shall show how this approach can indeed be justified in terms of the canonical scheme that was developed in the preceding sections. As a first step, we shall begin by restricting ourselves to a one-dimensional case.

A. Reduction to a one-dimensional model

To perform the reduction from a three-dimensional to a one-dimensional model, we follow the approach given by Blow *et al.* [31]. We consider the propagation in the x direction of plane waves polarized in the y direction. Excitations of this type will have $k_y = k_z = 0$ and a cross-sectional area S [32]. We have chosen this particular model for its simplicity and to facilitate comparison with earlier results [20]. However, it is worth noting that our expressions for the fields in three dimensions are complete and that we can extract components for propagation in a single direction with *any* transverse mode pattern. Such a reduction is suitable for modeling propagation in a monomode optical fiber [33]. For our plane wave model, the required conversions are [31]

$$\begin{aligned} \int d^3k \sum_{\lambda=1,2} &\rightarrow \frac{(2\pi)^2}{S} \int dk, \\ \delta(\mathbf{k} - \mathbf{k}') &\rightarrow \frac{S}{(2\pi)^2} \delta(k - k'), \\ \hat{C}(\lambda, \mathbf{k}, \omega) &\rightarrow \frac{S^{1/2}}{2\pi} \hat{C}(k, \omega), \end{aligned} \quad (5.1)$$

where k is in the x direction. The above set of equations (5.1) gives us the one-dimensional form of the system and can be used to write any of the previous operators. For example, the vector potential (4.3) becomes

$$\hat{A}(x, t) = \sqrt{\frac{\hbar\omega_c^2}{4\pi\epsilon_0 S}} \int_{-\infty}^{\infty} dk \int_0^{\infty} d\omega \left[\frac{\zeta^*(\omega)}{\omega^2\epsilon(\omega) - k^2c^2} \hat{C}(k, \omega) e^{-i(\omega t - kx)} + \text{H.c.} \right], \quad (5.2)$$

with similar expressions for the other fields. The ETCR for the polariton creation and annihilation operators \hat{C}^\dagger and \hat{C} (3.16) is transformed into

$$[\hat{C}(k, \omega), \hat{C}^\dagger(k', \omega')] = \delta(k - k') \delta(\omega - \omega'), \quad (5.3)$$

and the Hamiltonian (3.14) becomes

$$\hat{H} = \int_{-\infty}^{\infty} dk \int_0^{\infty} d\omega \hbar\omega \hat{C}^\dagger(k, \omega) \hat{C}(k, \omega). \quad (5.4)$$

These equations are similar to the ones derived in our previous work on quantization of a one-dimensional elec-

tromagnetic field [20] and we shall use them to calculate the one-dimensional propagation of a beam of light in the dielectric.

B. Propagation of the fields

In order to decompose the fields into space-dependent amplitudes, we use the fact that each eigenvalue of \hat{H} (5.4) is infinitely degenerate. This allows us to perform the k integration in (5.2) while keeping the same harmonic time dependence. The vector potential operator is thus decomposed into

$$\hat{A}(x, t) = \frac{1}{\sqrt{2\pi}} \int_0^\infty d\omega \mathcal{A}(\omega) [\hat{c}(x, \omega)e^{-i\omega t} + \text{H.c.}] , \quad (5.5)$$

where

$$\mathcal{A}(\omega) = \left(\frac{\hbar\eta(\omega)}{\epsilon_0 S c \omega |n(\omega)|^2} \right)^{1/2} ,$$

$n(\omega) \equiv \eta(\omega) + i\kappa(\omega)$ is the complex refractive index, defined as the square root of the dielectric constant $\epsilon(\omega)$ with a positive real part $\eta(\omega)$. Some of the properties of $n(\omega)$ are discussed in Appendix E. The space-dependent operators $\hat{c}(x, \omega)$ are defined in terms of the $\hat{C}(k, \omega)$ by

$$\hat{c}(x, \omega) = \sqrt{\frac{\omega_c^2 c \omega |n(\omega)|^2}{2\eta(\omega)}} \int_{-\infty}^\infty dk \frac{\zeta^*(\omega)}{\omega^2 \epsilon(\omega) - k^2 c^2} \times \hat{C}(k, \omega) e^{ikx} . \quad (5.6)$$

The form of this operator is chosen to lead to the ESCR for $\hat{c}(x, \omega)$:

$$[\hat{c}(x, \omega), \hat{c}^\dagger(x, \omega')] = \delta(\omega - \omega') . \quad (5.7)$$

From the one-dimensional reduction of (4.5) and (4.7), it is straightforward to obtain the electric field

$$\hat{E}(x, t) = \frac{i}{\sqrt{2\pi}} \int_0^\infty d\omega \mathcal{A}(\omega) \omega [\hat{c}(x, \omega)e^{-i\omega t} - \text{H.c.}] \quad (5.8)$$

and the displacement field

$$\hat{d}(x, \omega) = \sqrt{\frac{\omega_c^2 c \omega |n(\omega)|^2}{2\eta(\omega)}} \int_{-\infty}^\infty dk \left(\frac{kc}{n(\omega)\omega} \right) \frac{\zeta^*(\omega)}{\omega^2 \epsilon(\omega) - k^2 c^2} \hat{C}(k, \omega) e^{ikx} , \quad (5.13)$$

and, as in (5.5), the coefficients are chosen to give the ESCR

$$[\hat{d}(x, \omega), \hat{d}^\dagger(x, \omega')] = \delta(\omega - \omega') . \quad (5.14)$$

Moreover, from (5.6) and (5.13), we can verify that the \hat{c} and \hat{c}^\dagger operators commute with the \hat{d} and \hat{d}^\dagger operators. Let us emphasize that, because of the extra k factor in the integration, the magnetic-field operator cannot be expressed in terms of the same operators $\hat{c}(x, \omega)$ used in (5.5), (5.8), and (5.9). However, using the definition of \hat{B} [in this one-dimensional restriction, it reads $\hat{B}(x, t) = \partial_x \hat{A}(x, t)$], (5.5) and (5.12), we can obtain the relationship between $\hat{c}(x, \omega)$ and $\hat{d}(x, \omega)$

$$\frac{\partial \hat{c}(x, \omega)}{\partial x} = iK(\omega) \hat{d}(x, \omega) , \quad (5.15)$$

where $K(\omega)$ is the complex wave vector defined by $K(\omega) = n(\omega)\omega/c$. From the properties of $n(\omega)$ given in Appendix E, we see that both the real part of $K(\omega)$, $K_r(\omega)$, and the imaginary one, $K_i(\omega)$, are positive. Using Maxwell's equation for the displacement field $\hat{D}(x, t)$

$$\begin{aligned} \hat{D}(x, t) = & \frac{i}{\sqrt{2\pi}} \int_0^\infty d\omega \mathcal{A}(\omega) \omega [\epsilon_0 \epsilon(\omega) \hat{c}(x, \omega) e^{-i\omega t} - \text{H.c.}] \\ & + \frac{1}{\sqrt{2\pi}} \int_0^\infty d\omega \sqrt{2\hbar\epsilon_0\epsilon_i(\omega)} \left[\frac{n(\omega)}{|n(\omega)|} \hat{f}(x, \omega) e^{-i\omega t} \right. \\ & \left. + \text{H.c.} \right] , \quad (5.9) \end{aligned}$$

where $\epsilon_i(\omega)$ is the imaginary part of the dielectric constant defined in (4.4), $\hat{f}(x, \omega)$ is defined by

$$\hat{f}(x, \omega) = \frac{-i}{\sqrt{2\pi}} e^{i\phi(\omega)} \int_{-\infty}^\infty dk \hat{C}(k, \omega) e^{ikx} , \quad (5.10)$$

and the phase factor $e^{i\phi(\omega)} = \frac{\zeta^*(\omega) |n(\omega)|}{|\zeta(\omega)| n(\omega)}$ is chosen for future convenience. The coefficients are again chosen in order to give simple commutation relations

$$[\hat{f}(x, \omega), \hat{f}^\dagger(x', \omega')] = \delta(x - x') \delta(\omega - \omega') . \quad (5.11)$$

This type of CR is characteristic of a Langevin noise operator which is consistent with the way they are introduced in the system in (4.7). It is easily verified that any average of one of these operators is zero for nonsingular excitations [i.e., when the quantum average of $\hat{C}(k, \omega)$ is a nonsingular function]. We can use (4.6) to give the magnetic field operator

$$\hat{B}(x, t) = \frac{i}{\sqrt{2\pi}} \int_0^\infty d\omega \mathcal{A}(\omega) \omega \left[\frac{n(\omega)}{c} \hat{d}(x, \omega) e^{-i\omega t} - \text{H.c.} \right] , \quad (5.12)$$

where the operators $\hat{d}(x, \omega)$ are defined by

(1.1), we obtain from (5.9) and (5.12) the propagation equation

$$\frac{\partial \hat{d}(x, \omega)}{\partial x} = iK(\omega) \hat{c}(x, \omega) + 2\sqrt{K_i(\omega)} \hat{f}(x, \omega) . \quad (5.16)$$

The coupled system of equations (5.15) and (5.16) can be solved by introducing the linear combinations

$$\hat{c}_\pm(x, \omega) = \frac{1}{\sqrt{2}} [\hat{c}(x, \omega) \pm \hat{d}(x, \omega)] , \quad (5.17)$$

which clearly satisfy the same ESCR as the \hat{c} 's and \hat{d} 's (5.7) and (5.14):

$$[\hat{c}_\pm(x, \omega), \hat{c}_\pm^\dagger(x, \omega')] = \delta(\omega - \omega') , \quad (5.18)$$

$$[\hat{c}_\pm(x, \omega), \hat{c}_\mp^\dagger(x, \omega')] = 0 .$$

Using (5.6), (5.13), and (5.17), we write them in terms of the eigenoperators of the system:

$$\hat{c}_{\pm}(x, \omega) = \sqrt{\frac{K_i(\omega)}{\pi}} e^{i\phi(\omega)} \int_{-\infty}^{\infty} dk \frac{\hat{C}(k, \omega) e^{ikx}}{K(\omega) \mp k}, \quad (5.19)$$

where $e^{i\phi(\omega)}$ is the phase factor defined in (5.10). From (5.15) and (5.16), we find their spatial equation of evolution

$$\frac{\partial \hat{c}_{\pm}(x, \omega)}{\partial x} = \pm iK(\omega) \hat{c}_{\pm}(x, \omega) \pm \sqrt{2K_i(\omega)} \hat{f}(x, \omega). \quad (5.20)$$

This equation is the equivalent to the classical equation for the forward- and backward-propagating fields amplitudes. In addition to the classically expected part,

$$\begin{aligned} \hat{A}(x, t) &= \frac{1}{\sqrt{4\pi}} \int_0^{\infty} d\omega \mathcal{A}(\omega) [\hat{c}_+(x, \omega) e^{-i\omega t} + \hat{c}_-(x, \omega) e^{-i\omega t} + \text{H.c.}], \\ \hat{E}(x, t) &= \frac{i}{\sqrt{4\pi}} \int_0^{\infty} d\omega \mathcal{A}(\omega) \omega [\hat{c}_+(x, \omega) e^{-i\omega t} + \hat{c}_-(x, \omega) e^{-i\omega t} - \text{H.c.}], \\ \hat{B}(x, t) &= \frac{i}{\sqrt{4\pi}} \int_0^{\infty} d\omega \mathcal{A}(\omega) \omega \left[\frac{n(\omega)}{c} \hat{c}_+(x, \omega) e^{-i\omega t} - \frac{n(\omega)}{c} \hat{c}_-(x, \omega) e^{-i\omega t} - \text{H.c.} \right], \\ \hat{D}(x, t) &= \frac{i}{\sqrt{4\pi}} \int_0^{\infty} d\omega \mathcal{A}(\omega) \omega \epsilon_0 [\epsilon(\omega) \hat{c}_+(x, \omega) e^{-i\omega t} + \epsilon(\omega) \hat{c}_-(x, \omega) e^{-i\omega t} - \text{H.c.}] \\ &\quad + \frac{1}{\sqrt{2\pi}} \int_0^{\infty} d\omega \sqrt{2\hbar \epsilon_0 \epsilon_i(\omega)} \left[\frac{n(\omega)}{|n(\omega)|} \hat{f}(x, \omega) e^{-i\omega t} + \text{H.c.} \right], \end{aligned} \quad (5.21)$$

where $\mathcal{A}(\omega)$ is defined in (5.5). These expressions are similar to the ones used in the phenomenological approach to quantization in a continuous medium in the lossless case ($\eta = n$ and $\epsilon_i = 0$) [19, 31]. In this work, they emerge directly from a canonical quantization scheme and include the Langevin operators needed in order to treat the absorption. Let us emphasize that, for the case of a linear dielectric, the ESCR for the \hat{c}_{\pm} (5.18) have been derived from the canonical quantization scheme and that they are fully consistent with the usual ETCR for the conjugate fields \hat{A} and $-\epsilon_0 \hat{E}$. However, as this derivation relies on the explicit diagonalization of the total Hamiltonian (2.24), the extension to nonlinear media, where such a diagonalization is not always possible, is still problematic. The agreement between experiments and theories based on the ESCR [15, 16, 18, 34] seems to indicate that they are at least a good approximation, but further work is needed in order to understand their domain of validity in the nonlinear case [35].

VI. TWO INTERESTING RELATIONSHIPS

The principal aim of this paper is to provide a rigorous quantization of the electromagnetic field in a dielectric medium and to apply the results to the problem of propagation. In this section, we shall use one of the results arising from our quantization scheme to find two interesting relationships between the group velocity and the phase velocity in the dielectric. These relationships hold for both quantum and classical models. They can be

given by the wave vector $K(\omega)$, we have an added Langevin noise operator corresponding to the absorption of the electromagnetic field. Using (5.10) and (5.19) it is easily seen that $\hat{c}_+(x, \omega)$ commutes with all the Langevin operators $\hat{f}(x', \omega')$ and $\hat{f}^\dagger(x', \omega')$ for all $x' > x$, while $\hat{c}_-(x, \omega)$ commutes with all the Langevin operators $\hat{f}(x', \omega')$ and $\hat{f}^\dagger(x', \omega')$ for all $x' < x$. This again is consistent with the interpretation of these operators as the creation-annihilation operators for forward- and backward-propagating waves. The operator for a forward-propagating wave at a given point will commute with Langevin noise operators corresponding to positions *ahead* of the point at which the operator is calculated.

Finally, we write the expression of the various fields in terms of the \hat{c}_{\pm} operators to get

derived by using well-known properties of the dielectric constant.

Our derivation is based on the consistency check for the quantization procedure presented in Sec. III. This consistency check led to the equality (3.19), which we rewrite here for clarity:

$$\int_0^{\infty} d\omega [|\tilde{\alpha}_0(k, \omega)|^2 - |\tilde{\beta}_0(k, \omega)|^2] = 1. \quad (6.1)$$

We use the results of Appendix D [(D3)–(D5)] and of Appendix E [(E2)] to write it in terms of the dielectric constant $\epsilon(\omega)$

$$\frac{2}{\pi} \int_0^{\infty} d\omega \frac{\omega^3 \epsilon_i(\omega)}{|\omega^2 \epsilon(\omega) - k^2 c^2|^2} = 1, \quad (6.2)$$

where $\epsilon_i(\omega)$ is the imaginary part of $\epsilon(\omega)$. We emphasize that the derivation of this equality only relies on the analyticity of $\epsilon(\omega)$ in the upper half plane and on the fact that the solutions of the dispersion equation

$$\omega^2 \epsilon(\omega) - k^2 c^2 = 0 \quad (6.3)$$

are all in the lower half plane. These two properties were derived in Appendix E for our model of a dielectric. However, as is well known from classical electromagnetism [3], they are satisfied by any dielectric constant which is consistent with causality requirements. For this reason, (6.2) is indeed correct in any linear dielectric. Let us emphasize that here k is a *real* parameter and that the zeros

will be at complex frequencies. This is in contrast to Sec. V, where we used a complex wave vector and a real frequency. As shown in Sec. IV, the electromagnetic field operators are given as double integrals over both real frequencies and wave vectors. It is only when performing one of the integrals that we obtain either a real frequency and a complex wave vector (as in Sec. IV) or a real wave vector and a complex frequency. The first possibility is preferable in order to treat the problem of propagation in the medium in a way that is similar to the classical analysis. However, here we need to use the second one. We denote the zeros of $\omega n(\omega) - kc$ at a given k by Ω_j , $j = 1, \dots, m$ (for notational simplicity, we will not write the k dependence explicitly). From the results of Appendix E, we find that the real part of Ω_j is positive and its imaginary part negative. The zeros of $|\omega^2 \epsilon(\omega) - k^2 c^2|^2$ are then $\Omega_j, -\Omega_j, \Omega_j^*, -\Omega_j^*$, $j = 1, \dots, m$. We define the complex group velocity at frequency Ω_j by

$$V_g^j = \frac{\partial \Omega_j}{\partial k} \quad (6.4)$$

and the complex phase velocity

$$V_p^j = \frac{\Omega_j}{k}. \quad (6.5)$$

These two quantities reduce to the usual definitions when the imaginary part is negligible. We note that using a real wave vector and a complex frequency enables us to define the group velocity in a lossy dielectric in a straightforward manner. However, its physical interpretation is still unclear. Indeed, the definition of the group and phase velocity in a lossy dielectric has been the subject of some difficulties [36] and we shall not discuss these problems in detail, but rather use our approach to present some interesting new results. The interpretation and discussion of these results will be left for future work.

To obtain the first relation, we decompose (6.2) into partial fractions in the following way:

$$\frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega \left[\frac{\omega}{\omega^2 \epsilon^*(\omega) - k^2 c^2} - \frac{\omega}{\omega^2 \epsilon(\omega) - k^2 c^2} \right] = 1. \quad (6.6)$$

We close the integration contour in the lower half plane (contour C^-), so that the first term in the integral does not contribute and decompose the second term into partial fractions again to get

$$\frac{1}{2\pi i} \oint_{C^-} d\Omega \frac{\Omega}{2kc} \left[\frac{1}{\Omega n(\Omega) - kc} - \frac{1}{\Omega n(\Omega) + kc} \right] = -1. \quad (6.7)$$

Using integration in the complex plane to calculate the integral in terms of the residues at Ω_j , we obtain the first relation

$$\sum_j \text{Re} [V_g^j V_p^j] = c^2, \quad (6.8)$$

where Re means the real part.

To prove our second relation, we use the following equation:

$$\frac{1}{2\pi i} \oint_{C^-} d\Omega \left[\frac{\omega \epsilon(\omega)}{\omega^2 \epsilon(\omega) - k^2 c^2} \right] = -1. \quad (6.9)$$

This equation can be proven by the same techniques used in Appendix B 2 and is verified in any linear dielectric. We again use integration in the complex plane to calculate the integral in terms of the residues at Ω_j and $-\Omega_j^*$ and obtain the second relation

$$\sum_j \text{Re} \left[\frac{V_g^j}{V_p^j} \right] = 1. \quad (6.10)$$

We emphasize that these two relations (6.8) and (6.10) apply in any linear dielectric. The only requirements on the dielectric constant ϵ are similar to the ones used in deriving the Kramers-Kronig relations and are equivalent to demanding that causality should be satisfied in the dielectric. Further work is needed in order to find a good physical interpretation of these relations.

It is interesting to note that a truly elementary derivation of these results can be given for a lossless dielectric. In this case, the dielectric constant is real and the Kramers-Kronig relations do not apply. Causality is preserved by the appearance of forbidden band gaps in the polariton dispersion spectrum [12, 22]. Here, we shall discuss only a simple one-resonance model of the dielectric, but the extension to more complicated media is straightforward. The dielectric constant of the medium is given by

$$\epsilon(\omega) = 1 + \frac{\omega_c^2}{\omega_0^2 - \omega^2}, \quad (6.11)$$

where ω_0 is the resonance frequency and ω_c is the coupling constant. The dispersion equation is thus

$$\omega^4 + (\omega_L^2 + k^2 c^2) \omega^2 - k^2 c^2 \omega_0^2 = 0, \quad (6.12)$$

where $\omega_L^2 \equiv \omega_0^2 + \omega_c^2$ is the longitudinal resonance frequency. There are two solutions ω_{\pm} for each value of k one in each of the two branches of the polariton spectrum. The forbidden band gap corresponds to frequencies between ω_0 and ω_L . We do not need to calculate ω_- and ω_+ explicitly, but shall only use their properties (sum and product of their squares) obtained from the dispersion equation (6.12). The equivalent to (6.8) is here

$$V_g(\omega_-) V_p(\omega_-) + V_g(\omega_+) V_p(\omega_+) = \frac{1}{2k} \frac{d}{dk} (\omega_-^2 + \omega_+^2) = c^2 \quad (6.13)$$

and the equivalent to (6.10) is

$$\frac{V_g(\omega_-)}{V_p(\omega_-)} + \frac{V_g(\omega_+)}{V_p(\omega_+)} = k \frac{d}{dk} (\ln \omega_- \omega_+) = 1. \quad (6.14)$$

To our knowledge, the first equation (6.13) has not been derived before, while the second one (6.14) was derived by us very recently [12]. They can be easily generalized to a lossless medium with any number of resonances. Moreover, we now know that the same type of relations also hold for any linear lossy medium.

VII. CONCLUSION

To conclude this work, we shall summarize our main results. By using a microscopic model for the medium, we performed a fully canonical quantization scheme for the electromagnetic field in a dispersive and lossy dielectric. We explicitly derived the dielectric constant of the medium in terms of the parameters of the model and showed that it satisfies the Kramers-Kronig relations. In this work, we treated the case of a dielectric with a single resonance, but showed that this can be easily extended to more complicated media. In fact, it is possible to use the Hamiltonian of the coupled system in (3.12) as a starting point and choose the coupling constant $\zeta(\omega)$ to give any dielectric constant $\epsilon(\omega)$ which is consistent with causality. We calculated the eigenoperators of the coupled matter-field system and expressed the electromagnetic field operators in terms of them. The most significant result obtained by this approach is the loss of the one-to-one correspondence between the wave vector and the frequency of the field in the dielectric. In a lossy dielectric medium, the fields have to be expressed as double integrals, over both the frequency and the wave vector, of the polariton creation-annihilation operators $\hat{C}(\mathbf{k}, \omega)$, as shown in Eqs. (4.3)–(4.7). It is, however, possible to perform the integration on the wave vector, to obtain the electromagnetic field operators in terms of monochromatic space-dependent amplitudes, which satisfy a set of equal-space commutation relations. We showed that the spatial equations of evolution of these amplitudes are very similar to the ones obtained in the classical case, with a frequency-dependent complex wave vector and an extra Langevin operator needed to accommodate the losses. This justifies the use of such operators to treat propagation problems in quantum optics at least in the case of a linear, dispersive, and lossy medium.

Finally, we derived two interesting relations between the group velocity and the phase velocity in a lossy dielectric. Further work is still needed to clarify the interpretation of these relations. We are currently working on extending our model to treat boundary conditions and to include nonlinearities perturbatively.

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APPENDIX A: FANO DIAGONALIZATION

In this appendix, we perform the diagonalization of \hat{H}_{mat} defined in (2.25b). The same calculation can be used with only minor changes to the diagonalization of the total Hamiltonian H (2.24). The starting point is (2.25b), which we reproduce here for clarity

$$H_{\text{mat}} = \int d^3k \left[\hbar\tilde{\omega}_0 \hat{b}^\dagger(\mathbf{k})\hat{b}(\mathbf{k}) + \int_0^\infty d\omega \hbar\omega \hat{b}_\omega^\dagger(\mathbf{k})\hat{b}_\omega(\mathbf{k}) + \frac{\hbar}{2} \int_0^\infty d\omega V(\omega) [\hat{b}^\dagger(-\mathbf{k}) + \hat{b}(\mathbf{k})] \times [\hat{b}_\omega^\dagger(-\mathbf{k}) + \hat{b}_\omega(\mathbf{k})] \right], \quad (\text{A1})$$

where, following the convention of Sec. III, we do not write the explicit polarization and time dependence of the operators. The only assumption we make on $V(\omega)$ is that the analytic continuation of $|V(\omega)|^2$ to negative frequencies, which we denote by $\mathcal{V}(\omega)$, should be an odd function and be nonzero except at the origin. The diagonalization of \hat{H}_{mat} is performed by introducing the operators $\hat{B}(\mathbf{k}, \omega)$ defined by

$$\hat{B}(\mathbf{k}, \omega) = \alpha_0(\omega)\hat{b}(\mathbf{k}) + \beta_0(\omega)\hat{b}^\dagger(-\mathbf{k}) + \int_0^\infty d\omega' [\alpha_1(\omega, \omega')\hat{b}_{\omega'}(\mathbf{k}) + \beta_1(\omega, \omega')\hat{b}_{\omega'}^\dagger(-\mathbf{k})], \quad (\text{A2})$$

where the coefficients are chosen so that the operators satisfy the eigenoperator equation [27]

$$[\hat{B}(\mathbf{k}, \omega), \hat{H}_{\text{mat}}] = \hbar\omega\hat{B}(\mathbf{k}, \omega). \quad (\text{A3})$$

This equation, together with the expansion of the Hamiltonian (A1) and the definition of $\hat{B}(\mathbf{k}, \omega)$ in (A2), leads to a set of linear equations between the coefficients:

$$\alpha_0(\omega)\omega = \alpha_0(\omega)\tilde{\omega}_0 + \frac{1}{2} \int_0^\infty d\omega' [\alpha_1(\omega, \omega')V(\omega') - \beta_1(\omega, \omega')V^*(\omega')], \quad (\text{A4a})$$

$$\beta_0(\omega)\omega = -\beta_0(\omega)\tilde{\omega}_0 + \frac{1}{2} \int_0^\infty d\omega' [\alpha_1(\omega, \omega')V(\omega') - \beta_1(\omega, \omega')V^*(\omega')], \quad (\text{A4b})$$

$$\alpha_1(\omega, \omega')\omega = \frac{1}{2} [\alpha_0(\omega) - \beta_0(\omega)] V^*(\omega') + \alpha_1(\omega, \omega')\omega', \quad (\text{A4c})$$

$$\beta_1(\omega, \omega')\omega = \frac{1}{2} [\alpha_0(\omega) - \beta_0(\omega)] V(\omega') - \beta_1(\omega, \omega')\omega'. \quad (\text{A4d})$$

This set of equations can easily be solved to obtain $\beta_0(\omega)$, $\alpha_1(\omega, \omega')$, and $\beta_1(\omega, \omega')$ in terms of $\alpha_0(\omega)$. Subtracting (A4a) from (A4b) we obtain

$$\beta_0(\omega) = \frac{\omega - \tilde{\omega}_0}{\omega + \tilde{\omega}_0} \alpha_0(\omega). \quad (\text{A5a})$$

We now use (A5a) to replace β_0 in (A4c) and (A4d) to give

$$\alpha_1(\omega, \omega') = \left[P \left(\frac{1}{\omega - \omega'} \right) + y(\omega) \delta(\omega - \omega') \right] \times V^*(\omega') \frac{\tilde{\omega}_0}{\omega + \tilde{\omega}_0} \alpha_0(\omega), \quad (\text{A5b})$$

$$\beta_1(\omega, \omega') = \left[\frac{1}{\omega + \omega'} \right] V(\omega') \frac{\tilde{\omega}_0}{\omega + \tilde{\omega}_0} \alpha_0(\omega), \quad (\text{A5c})$$

where P means principal part. The function $y(\omega)$ is obtained by replacing the expressions for α_1 and β_1 in (A5b) and (A5c) into (A4a). After some algebra, we get

$$y(\omega) = \frac{2(\omega^2 - \tilde{\omega}_0^2)}{\tilde{\omega}_0 |V(\omega)|^2} + \frac{1}{|V(\omega)|^2} P \int_{-\infty}^{\infty} d\omega' \frac{\mathcal{V}(\omega')}{\omega' - \omega}, \quad (\text{A5d})$$

where we used the assumption that $\mathcal{V}(\omega)$, defined in (A1), is an odd function to extend the integral in the negative frequency region. In order to calculate $\alpha_0(\omega)$, we impose the standard commutation relations for $\hat{B}(\mathbf{k}, \omega)$:

$$\left[\hat{B}(\mathbf{k}, \omega), \hat{B}^\dagger(\mathbf{k}', \omega') \right] = \delta(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'). \quad (\text{A6})$$

Using the expression for $\hat{B}(\omega)$ given by (A2) and the set of equations defining all the coefficients in terms of α_0 (A5a), the above equation (A6) defines $\alpha_0(\omega)$ up to a phase factor. We choose this phase factor to give after a somewhat lengthy but straightforward calculation:

$$\alpha_0(\omega) = \frac{\omega + \tilde{\omega}_0}{\tilde{\omega}_0 V^*(\omega)} \frac{1}{y(\omega) - i\pi}. \quad (\text{A7})$$

Using the expression for $y(\omega)$ derived in (A5d), this equation can be written in a final form

$$\alpha_0(\omega) = \left(\frac{\omega + \tilde{\omega}_0}{2} \right) \frac{V(\omega)}{\omega^2 - \tilde{\omega}_0^2 z(\omega)}, \quad (\text{A8})$$

where $z(\omega)$ is defined by

$$z(\omega) = 1 - \frac{1}{2\tilde{\omega}_0} \left[\int_{-\infty}^{\infty} d\omega' \frac{\mathcal{V}(\omega')}{\omega' - \omega + i\varepsilon} \right], \quad (\text{A9})$$

with $\varepsilon \rightarrow 0^+$. As $\mathcal{V}(\omega)$ is an analytic function on the real axis, it is easily seen that $z(\omega)$ is also an analytic function on the real axis. The final expression for β_0 is now readily derived from (A5a):

$$\beta_0(\omega) = \left(\frac{\omega - \tilde{\omega}_0}{2} \right) \frac{V(\omega)}{\omega^2 - \tilde{\omega}_0^2 z(\omega)}. \quad (\text{A10})$$

Finally, we use (A7) to obtain y in terms of α_0 and replace in (A5b) to give

$$\alpha_1(\omega, \omega') = \delta(\omega - \omega') + \left(\frac{\tilde{\omega}_0}{2} \right) \left(\frac{V^*(\omega')}{\omega - \omega' - i\varepsilon} \right) \times \frac{V(\omega)}{\omega^2 - \tilde{\omega}_0^2 z(\omega)} \quad (\text{A11})$$

and similarly for β_1 ,

$$\beta_1(\omega, \omega') = \left(\frac{\tilde{\omega}_0}{2} \right) \left(\frac{V(\omega')}{\omega + \omega'} \right) \frac{V(\omega)}{\omega^2 - \tilde{\omega}_0^2 z(\omega)}. \quad (\text{A12})$$

The expansion of the dressed matter field annihilation operators $\hat{B}(\mathbf{k}, \omega)$ in terms of the initial creation and annihilation operators given in Eqs. (A8)–(A12) completes the diagonalization of \hat{H}_{mat} (3.3).

APPENDIX B: A CONSISTENCY CHECK

In this appendix, we calculate the two integrals I and $I(\omega, \omega')$ defined in (3.8) and (3.9). The main idea in this calculation is to extend the integrals to negative frequencies and use contour integration in the complex plane. As can be expected from the definition of the expansion coefficients α_0 , β_0 , α_1 , and β_1 given in (A8)–(A12), the zeros of $\omega^2 - \tilde{\omega}_0^2 z(\omega)$, corresponding to poles of α_0 , β_0 , α_1 , and β_1 , will play an important role in this calculation. Therefore we shall begin this appendix by analyzing their position.

1. Zeros of $\omega^2 - \tilde{\omega}_0^2 z(\omega)$

We define

$$Z(\omega) \equiv \omega^2 - \tilde{\omega}_0^2 z(\omega), \quad (\text{B1})$$

where $z(\omega)$ is given in (A9), and look for a condition on $V(\omega)$ such that all the zeros of Z are in the upper half plane. We already know that $z(\omega)$ is an analytic function on the real axis. As $\mathcal{V}(\omega)$ is an odd function, $z(\omega)$ satisfies the condition $z(-\omega) = z^*(\omega)$. Extending the definition of $z(\omega)$ to complex frequencies (which we shall denote by Ω), it is easily seen that

$$z(-\Omega^*) = z^*(\Omega), \quad (\text{B2})$$

and that $z(\Omega)$ is an analytic function in the lower half plane ($\text{Im}\Omega \leq 0$) and can be written

$$z(\Omega) = 1 - \frac{1}{2\tilde{\omega}_0} \int_{-\infty}^{\infty} d\omega' \frac{\mathcal{V}(\omega')}{\omega' - \Omega + i\varepsilon} \quad \text{for } \text{Im}\Omega \leq 0. \quad (\text{B3})$$

In the rest of the Appendix B 1 we shall restrict ourselves to the lower half plane. Writing $\Omega = \Omega_r - i\Omega_i$ ($\Omega_i \geq 0$) and using (B3), we can now calculate the real and imaginary part of $Z(\Omega)$ and show that the imaginary part is zero on the imaginary axis only. Therefore, in order to find the zeros of $Z(\omega)$, we only need to calculate it on the imaginary axis. After some algebra, we get

$$Z(-i\mu) = -(\mu^2 + \tilde{\omega}_0^2) + \tilde{\omega}_0 \int_0^{\infty} d\omega \frac{\omega |V(\omega)|^2}{\omega^2 + (\mu + \varepsilon)^2} \quad (\text{B4})$$

for $\mu \geq 0$. Equation (B4) shows that $Z(-i\mu)$ is a strictly decreasing function of μ , which tends to $-\infty$ as μ tends to ∞ . Its maximum value is therefore reached at $\mu = 0$ and is

$$Z(0) = -\tilde{\omega}_0^2 + \tilde{\omega}_0 \int_0^{\infty} d\omega \frac{|V(\omega)|^2}{\omega}. \quad (\text{B5})$$

It is now clear that $Z(\Omega)$ has no zeros in the lower half plane when

$$\int_0^\infty d\omega \frac{|V(\omega)|^2}{\omega} < \tilde{\omega}_0, \quad (\text{B6})$$

and that it has one and only one zero (which is on the imaginary axis) when (B6) is not satisfied. In the rest of this section, we assume that (B6) is satisfied so that $Z(\omega)$ has no zeros in the lower half plane.

2. Calculation of I

From the expressions for $\alpha_0(\omega)$ and $\beta_0(\omega)$ derived in Appendix A, (A8) and (A10), we obtain

$$I = \int_0^\infty d\omega \omega \tilde{\omega}_0 \frac{|V(\omega)|^2}{|\omega^2 - \tilde{\omega}_0^2 z(\omega)|^2}. \quad (\text{B7})$$

We decompose I into partial fractions and use the assumption that both $z(\omega)$ and $\mathcal{V}(\omega)$, the analytic continuation of $|V(\omega)|^2$ to negative frequencies, are odd functions. This allows us to write I in a way that is suitable for integration in the complex plane:

$$I = \frac{1}{\pi i} \int_{-\infty}^\infty d\omega \frac{\omega}{\omega^2 - \tilde{\omega}_0^2 z(\omega)}. \quad (\text{B8})$$

From Appendix B 1, we know that the integrand has no

pole in the lower half of the complex plane, so that the integral on the contour based on the real axis and closing in the lower half plane is zero. The integral on the real axis is therefore equal to the opposite of the integral along the closing circle C_- :

$$I = \frac{-1}{\pi i} \int_{C_-} d\Omega \frac{\Omega}{\Omega^2 - \tilde{\omega}_0^2 z(\Omega)}. \quad (\text{B9})$$

Writing the variable $\Omega = R e^{-i\theta}$, where θ varies from zero to π , and letting R tend to ∞ gives

$$I = 1. \quad (\text{B10})$$

It is also straightforward to check that if (B6) is not satisfied, which means that $Z(\Omega)$ has one pole in the lower half plane, $I \neq 1$, and the diagonalization scheme is not consistent.

3. Calculation of $I(\omega, \omega')$

The calculation of $I(\omega, \omega')$ follows the same lines as the previous one. Using the expression of α_1 and β_1 from (A11) and (A12), and the fact that $\mathcal{V}(\omega)$ and $z(\omega)$ are odd functions, we get

$$I(\omega, \omega') = \delta(\omega - \omega') + \frac{V(\omega)V^*(\omega')}{\omega - \omega' - i\varepsilon} \left[\frac{1}{\omega^2 - \tilde{\omega}_0^2 z(\omega)} - \frac{1}{\omega'^2 - \tilde{\omega}_0^2 z^*(\omega')} \right] + \frac{\tilde{\omega}_0}{2} V(\omega)V^*(\omega') \int_{-\infty}^\infty d\nu \frac{\mathcal{V}(\nu)}{|\nu^2 - \tilde{\omega}_0^2 z(\nu)|^2} \frac{1}{(\nu - \omega - i\varepsilon)(\nu - \omega' - i\varepsilon)}, \quad (\text{B11})$$

where $\varepsilon \rightarrow 0^+$. The calculation of the integral on the right-hand side of the equation presents no difficulties. We again use integration in the complex plane and the results of Appendix B 1 to obtain

$$I(\omega, \omega') = \delta(\omega - \omega'). \quad (\text{B12})$$

To conclude this appendix, we emphasize its main result. When (3.10) is satisfied, the diagonalization scheme is consistent and the two equations (B10) and (B12) are also satisfied. We use this result in the diagonalization of the total Hamiltonian in Sec. III B.

APPENDIX C: PROPERTIES OF THE COUPLING CONSTANT $\zeta(\omega)$

The coupling constant $\zeta(\omega)$ was defined in (3.12) as $\zeta(\omega) = i\sqrt{\tilde{\omega}_0} [\alpha_0(\omega) + \beta_0(\omega)]$. Using the expressions of α_0 and β_0 derived in Appendix A, in (A8) and (A10), we write

$$\zeta(\omega) = i\sqrt{\tilde{\omega}_0} \frac{\omega V(\omega)}{\omega^2 - \tilde{\omega}_0^2 z(\omega)}, \quad (\text{C1})$$

where $z(\omega)$ is defined in (A9). As the analytic continuation of $|V(\omega)|^2$ to negative frequencies is an odd function, nonzero for all nonzero frequencies, and $\omega^2 - \tilde{\omega}_0^2 z(\omega)$

is nonzero for all real frequencies (as derived in Appendix B 1), the analytic continuation of $|\zeta(\omega)|^2$ to negative frequencies is also an odd function, nonzero for all frequencies except at the origin, and analytic on the real axis. Moreover, using the integral derived in Appendix B 2, it is easy to see that $\zeta(\omega)$ satisfies the following normalization condition:

$$\int_0^\infty d\omega \frac{|\zeta(\omega)|^2}{\omega} = 1. \quad (\text{C2})$$

We define the function $\xi(\omega)$ by $\xi(\omega) \equiv \frac{|\zeta(\omega)|^2}{\omega}$ for positive frequencies and its analytic continuation for negative frequencies. From the above expression for $\zeta(\omega)$ (C1) and the definition of $V(\omega)$ in (2.25a), we obtain the expression of $\xi(\omega)$ for positive frequencies

$$\xi(\omega) = \frac{1}{\rho^2} \frac{\omega^2 |v(\omega)|^2}{|\omega^2 - \tilde{\omega}_0^2 z(\omega)|^2}, \quad (\text{C3})$$

where ρ is the original polarization field density, defined in (2.3) and $v(\omega)$ is the coupling between this field and the reservoir fields defined in (2.5). As the analytic continuation of $|v(\omega)|^2$ to negative frequencies is an even function, nonzero everywhere except possibly at the origin, $\xi(\omega)$ is clearly an even function, analytic on the real axis and strictly positive except at $\omega = 0$. From Eq. (C2), it is also normalized. Finally, (C3) can be used to show

that $\frac{\xi(\omega)}{\omega^2}$ is an analytic function on the real axis and that it tends to a finite limit when ω tends to zero. The above results will be needed in the diagonalization of the total Hamiltonian in Sec. III B and in deriving some properties of the dielectric function in Appendix E.

APPENDIX D: EXPANSION COEFFICIENTS OF THE \hat{C} 'S

The expression of the total Hamiltonian of the coupled system (3.12) in terms of the electromagnetic and dressed matter operators has the same form as the expression of the matter part of the Hamiltonian (2.25b) in terms of the initial creation and annihilation operators, when the parameters of the model in (2.25b): $\tilde{\omega}_0$ and $V(\omega)$ are replaced by $\tilde{k}c$ and $\sqrt{\omega_c^2/\tilde{k}c}\zeta(\omega)$ and the initial matter and reservoir operators \hat{b} and \hat{b}_ω are replaced by the photons and dressed matter operators \hat{a} and $\hat{B}(\omega)$. Moreover, from the results of Appendix C, the coupling $\zeta(\omega)$ satisfies the same conditions as $V(\omega)$. The calculation of the expansion coefficients of the polariton creation and annihilation operators is therefore similar to the one performed in Appendix A. The only noticeable difference is that, as the parameters in the Hamiltonian (3.12) are explicitly k dependent, the expansion coefficients of the polariton operators also become functions of k . Of course, as the medium is isotropic, there is no angular dependence. Thus we can use the expressions for α_0 , β_0 , α_1 , and β_1 derived in (A8), (A10), (A11), and (A12) to obtain the new coefficients $\tilde{\alpha}_0$, $\tilde{\beta}_0$, $\tilde{\alpha}_1$, and $\tilde{\beta}_1$ directly. However, for future use, we write these expressions in a slightly modified form. We obtain $\tilde{\alpha}_0(k, \omega)$ as the analog of (A8):

$$\tilde{\alpha}_0(k, \omega) = \sqrt{\frac{\omega_c^2}{\tilde{k}c}} \left(\frac{\omega + \tilde{k}c}{2} \right) \frac{\zeta(\omega)}{\omega^2 - \tilde{k}^2 c^2 \tilde{z}(k, \omega)}, \quad (\text{D1})$$

where $\tilde{z}(k, \omega)$ is defined by

$$\tilde{z}(k, \omega) = 1 - \frac{\omega_c^2}{2(\tilde{k}c)^2} \left[\int_{-\infty}^{\infty} d\omega' \frac{|\zeta(\omega')|^2}{\omega' - \omega + i\varepsilon} \right], \quad (\text{D2})$$

with $\varepsilon \rightarrow 0^+$. We use the definition of \tilde{k} , $\tilde{k}^2 = k^2 + k_c^2$, and the normalization condition on $\zeta(\omega)$ (3.13) to rewrite:

$$\tilde{\alpha}_0(k, \omega) = \sqrt{\frac{\omega_c^2}{\tilde{k}c}} \left(\frac{\omega + \tilde{k}c}{2} \right) \frac{\zeta(\omega)}{\varepsilon^*(\omega)\omega^2 - k^2 c^2}, \quad (\text{D3})$$

where $\varepsilon(\omega)$ is defined by

$$\varepsilon(\omega) = 1 + \frac{\omega_c^2}{2\omega} \int_{-\infty}^{\infty} d\omega' \frac{\xi(\omega')}{\omega' - \omega - i\varepsilon}, \quad (\text{D4})$$

with $\varepsilon \rightarrow 0^+$ and $\xi(\omega)$ is defined in Appendix C. This expression is similar to (4.4) in Sec. IV. The advantage of (D3) with respect to the initial expression (D1) is that, in contrast to the function $\tilde{z}(k, \omega)$ defined in (D2), the function $\varepsilon(\omega)$ in (D3) is independent of k . We interpret

$\varepsilon(\omega)$ as the dielectric constant of the medium.

It is now straightforward to obtain all the other expansion coefficients by comparison with (A10), (A11), and (A12):

$$\tilde{\beta}_0(k, \omega) = \sqrt{\frac{\omega_c^2}{\tilde{k}c}} \left(\frac{\omega - \tilde{k}c}{2} \right) \frac{\zeta(\omega)}{\varepsilon^*(\omega)\omega^2 - k^2 c^2}, \quad (\text{D5})$$

$$\tilde{\alpha}_1(k, \omega, \omega') = \delta(\omega - \omega') + \frac{\omega_c^2}{2} \left(\frac{\zeta^*(\omega')}{\omega - \omega' - i\varepsilon} \right) \times \frac{\zeta(\omega)}{\varepsilon^*(\omega)\omega^2 - k^2 c^2}, \quad (\text{D6})$$

$$\tilde{\beta}_1(k, \omega, \omega') = \frac{\omega_c^2}{2} \left(\frac{\zeta^*(\omega')}{\omega - \omega' - i\varepsilon} \right) \frac{\zeta(\omega)}{\varepsilon^*(\omega)\omega^2 - k^2 c^2}. \quad (\text{D7})$$

APPENDIX E: SOME PROPERTIES OF THE DIELECTRIC CONSTANT $\varepsilon(\omega)$

In this appendix, we shall derive some properties of the dielectric constant $\varepsilon(\omega)$ that was defined in Appendix D (D4). We use the fact that $\xi(\omega)$ is an even function to rewrite $\varepsilon(\omega)$ as

$$\varepsilon(\omega) = 1 + \frac{\omega_c^2}{2} \int_{-\infty}^{\infty} d\omega' \frac{\xi(\omega')}{\omega'(\omega' - \omega - i\varepsilon)}, \quad (\text{E1})$$

where $\varepsilon \rightarrow 0^+$. Equation (E1), together with the properties of $\xi(\omega)$ derived in Appendix C, ensures that the extension of ε to complex frequencies is an analytic function in the upper half plane. This shows that ε satisfies the Kramers-Kronig relations. From Appendix C, we know that $\frac{\xi(\omega)}{\omega^2}$ is positive and finite on the real axis, so that $\varepsilon(\omega)$ tends to a limit larger than one when ω tends to zero. Moreover, as $\xi(\omega)$ is normalized, $\varepsilon(\omega)$ tends to one when ω tends to infinity. Using a method similar to the one presented in Appendix B 1, we can also use the expression of $\varepsilon(\omega)$ in (E1) to show that the dispersion equation $\varepsilon(\omega)\omega^2 - k^2 c^2$ has no zeros in the upper half plane. This last result is used in Sec. VI in order to derive the two relations between the group and the phase velocity.

For real frequencies, we separate the dielectric constant into real and imaginary parts $\varepsilon(\omega) \equiv \varepsilon_r(\omega) + i\varepsilon_i(\omega)$, where

$$\varepsilon_r(\omega) = 1 + \frac{\omega_c^2}{2\omega} \text{P} \int_{-\infty}^{\infty} d\omega' \frac{\xi(\omega')}{\omega' - \omega}, \quad (\text{E2})$$

$$\varepsilon_i(\omega) = \frac{\pi\omega_c^2}{2} \frac{\xi(\omega)}{\omega}.$$

We introduce the index of refraction $n(\omega)$ as the square root of $\varepsilon(\omega)$ with a positive real part and separate it into real and imaginary parts by

$$n(\omega) \equiv \eta(\omega) + i\kappa(\omega), \quad (\text{E3})$$

where η and κ are two real functions, given by the following set of equations:

$$\begin{aligned}\eta^2(\omega) - \kappa^2(\omega) &= \epsilon_r(\omega), \\ 2\eta(\omega)\kappa(\omega) &= \epsilon_i(\omega).\end{aligned}\tag{E4}$$

As, from (E2), ϵ_i is an odd function, positive for positive frequencies, and as we have chosen η to be positive, (E4) shows that $\kappa(\omega)$ is positive for positive frequencies and negative for negative frequencies.

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