Quantum optics of dispersive dielectric media

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We quantize the electromagnetic field in a polar medium starting with the fundamental equations of motion. In our model the medium is described by a Lorenz–type dielectric function $\epsilon(\mathbf{r},\omega)$ appropriate, e.g., for ionic crystals, metals, and inert dielectrics. There are no restrictions on the spatial behavior of the dielectric function, i.e., there can be many different polar media with arbitrary shapes. We assume no losses in our system so the dielectric function for the whole space is assumed as real. The quantization procedure is based on an expansion of the total field (transverse and longitudinal) in terms of the coupled (*polariton*) eigenmodes, and this approach incorporates all previous results derived for similar but restricted systems (e.g., without spatial or frequency dependence of coupled modes). Within the same model, we also quantize the Hamiltonian of a nonretarded electromagnetic field in polar media. Particular attention is paid to the derivation of the orthogonality and closure relations, which are used in a discussion of the fundamental (equal-time) commutation relations between the conjugate field operators.

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I. INTRODUCTION

The quantization of electromagnetic field in a dispersive dielectric medium represents one of the most interesting problems in quantum optics because it gives a rigorous and simply achievable test of our understanding of the interaction of light with matter. Large number of articles that have discussed that problem in the past few decades can be roughly divided according to their microscopic or macroscopic point of view. Starting from the fundamental equations of motion, the microscopic theories give, as one of the key results, averaged operators [1] and a dielectric function ϵ [2] that describe the macroscopic properties of a medium. Macroscopic theories take appropriate field operators, construct the Hamiltonian of the coupled system, and try to diagonalize it. In both cases different gauge requirements and different couplings of matter and light were discussed depending on the problem involved, so there are also specific analyses that try to connect various approaches [3,5,4].

In the macroscopic theories that are concentrated mainly on the basic interaction between the particle (polarization) field and the electromagnetic field, the part of Hamiltonian that would describe losses is often neglected (in principle it can be added later [6,7]). In such approach the system is generally described by a real dielectric function of Lorentz-type [8]. It has a good experimental verification when applied to polar dielectrics (ionic crystals or metals) in a frequency region for which the wavelength of light is much greater than a lattice constant.

Using that model, Hopfield [9] was the first who has quantized the interaction between the polarization and the electromagnetic field in a homogeneous dielectric. In his approach he started from the equations of motion for the fields and derived the eigenfrequencies of the coupled modes—*polaritons*. Based on this theory, the model was extended to dielectric layers and quantum wells [10,11] and to a medium with a number of different resonance frequencies [12]. Alternatively, one can start with the classical Hamiltonian [13] appropriate for an arbitrary frequency dependence of a dielectric function, and quantize it by applying the standard procedure [14,15]. However, in this approach the dielectric function is assumed to be frequency dependent only and therefore this is appropriate for the discussion of light properties (dispersion, radiation, etc.) in the bulk. On the other hand, a theory has been developed for a dielectric function with an arbitrary space, but without a frequency dependence [16,17]. Such approach was successful in describing some interesting properties (atomic decay, local-field corrections, etc.) of a dielectric medium [18]. In our paper we wish to give a theory which will, starting from the first principles, quantize the Hamiltonian by taking into account both the frequency and the (arbitrary) space dependence of a dielectric function. As limiting cases, this theory will then recover the above-mentioned approaches.

In all the theories that apply quantum optics to a dielectric medium one essential question arises: What are the fundamental *equal-time commutation relations* between the field operators and the conjugate momenta? The clear answer is obtained in a case where ϵ does not depend upon frequency. The dynamics of the system is then described by the vector potential $\mathbf{A}$, and its conjugate momentum is proportional to the displacement $\mathbf{D}=\epsilon(\mathbf{r})\mathbf{E}$, where $\mathbf{E}$ is the electric field. The corresponding commutation relations are proportional to the δ function, as expected [16,17]. The situation becomes more complicated in the case where ϵ depends upon a frequency ω, because it also involves the nontrivial dynamics of the system, described by a polarization $\mathbf{P}$. That problem can *a priori* be solved by expanding $\mathbf{A}$ and $\mathbf{P}$ in terms of their own eigenmodes and then by adding the interaction term to the Hamiltonian [9]. The commutation relations are satisfied from the beginning, but the Hamiltonian must be diagonalized in terms of new operators, which could be difficult [10]. If one starts with the semiclassical Hamiltonian [13], the polarization is implicitly included in the form of $\epsilon(\omega)$, so the system is described only by the (renormalized) vector potential. The Hamiltonian is diagonalized by using the imposed commutation relations between the creation and annihilation operators. *A posteriori*, one can define the con-

jugate momentum to be proportional to $\mathbf{D}=\Sigma_\omega \epsilon(\omega)\mathbf{E}(\omega)$ [3]. The correct fundamental commutation relations were derived for the infinite ionic crystal, while there were some difficulties in the metallic case [15]. The difficulties are also present in models that start with the classical density of electromagnetic field, using the arbitrary (one-dimensional) dielectric function [19,20].

In fact, all problems in deriving the required commutation relations for the conjugate variables of the electromagnetic field can be avoided if the medium is described by a *complex* dielectric function which satisfies Kramers-Kronig relations and takes into account the absorption [2,21]. However, in that case there is no dispersion relation and, e.g., in a homogeneous dielectric, for each wave vector $\mathbf{k}$ there is a continuum of allowed frequencies ω. Since one cannot determine the eigenfrequencies of the system, the quantization scheme requires the integration over all frequencies and obviously it becomes more complicated. For instance, the classical constitutive equation $\mathbf{D}(\mathbf{r},\omega)=\epsilon(\omega)\mathbf{E}(\mathbf{r},\omega)$ can be consistently used in the quantum approach only if the losses are neglected [22].

In a model that neglects absorption, an interesting situation occurs when one starts from the equations of motion for the fields $\mathbf{A}$ and $\mathbf{P}$ and tries to quantize the Hamiltonian from the beginning in terms of the *coupled* eigenmodes, i.e., polaritons. Such a model was developed, e.g., for a semi-infinite medium [23] or a dielectric layer [24], but the appropriate commutation relations were not discussed. In our paper we shall use that approach and, as we have pointed out, generalize it to include a dielectric function of the form $\epsilon(\mathbf{r},\omega)$. In particular, in our model the momentum conjugate to $\mathbf{A}$ can be chosen proportional to $\mathbf{E}$ as well as to $\mathbf{D}$ [25]. This generalization will enable us to analyze the fundamental commutation relations with more flexibility and explain some of the present controversies in models with a real dielectric function.

The paper is organized as follows. In Sec. II we start from the equations of motion for the photon field propagating in the polar dielectric. We have derived appropriate Lagrangian and Hamiltonian of the system in the generalized Coulomb gauge. The Hamiltonian is quantized in Sec. III with the help of the general orthonormality relation developed for the eigenmodes of the coupled system. Some special cases are discussed in Sec. IV and the connection with the existing theories is pointed out. In Sec. V we discuss the validity of the equal-time commutation relations between the field operators and the conjugate momenta. The conclusion is given in Sec. VI. In the Appendix, we analyze the nonretarded limit that turns out to be important in the discussion of the general particle field.

II. MODEL HAMILTONIAN

We investigate the behavior of an electromagnetic field in a polar medium. The electromagnetic field is described by the following Maxwell equations:

$$\nabla\cdot\mathbf{D}(\mathbf{r},t)=4\pi\rho(\mathbf{r},t),\quad \nabla\times\mathbf{E}(\mathbf{r},t)+\frac{1}{c}\frac{\partial\mathbf{B}(\mathbf{r},t)}{\partial t}=\mathbf{0},$$

$$\nabla\cdot\mathbf{B}(\mathbf{r},t)=0,\quad \nabla\times\mathbf{B}(\mathbf{r},t)-\frac{1}{c}\frac{\partial\mathbf{D}(\mathbf{r},t)}{\partial t}=\frac{4\pi}{c}\mathbf{j}(\mathbf{r},t). \tag{1}$$

In these equations the electric field $\mathbf{E}$, the displacement $\mathbf{D}$, and the magnetic field $\mathbf{B}$ are determined by the external charge density ρ and the current density $\mathbf{j}$, and by the polarization which describes, e.g., the dynamics of ions in ionic crystals or conducting electrons in metals [8]. Usually, the total polarization $\mathbf{P}_{tot}$ of the medium is divided in two parts:

$$\mathbf{P}_{tot}=\mathbf{P}+\mathbf{P}_e,$$

where $\mathbf{P}$ describes the dynamics of *point ions* and $\mathbf{P}_e$ represents the *electronic* contribution to the polarization.

The equation of motion for the point-ion polarization is given by

$$\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2}+\omega_T^2(\mathbf{r})\mathbf{P}(\mathbf{r},t)=\frac{\omega_P^2(\mathbf{r})}{4\pi}\mathbf{E}(\mathbf{r},t), \tag{2}$$

where $\omega_T(\mathbf{r})$ and $\omega_P(\mathbf{r})$ are transverse and plasma frequency of ions, respectively. One chooses $\omega_T=0$ for conducting electrons in metals and $\omega_T=\omega_P=0$, i.e., $\mathbf{P}=\mathbf{0}$ for the inert dielectrics or for the empty space.

Assuming a periodic solution in time with a frequency ω, we find the following for the point-ion susceptibility χ:

$$\mathbf{P}(\mathbf{r},\omega)=\chi(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega), \tag{3}$$

$$\chi(\mathbf{r},\omega)=\frac{\omega_P^2(\mathbf{r})}{4\pi}\frac{1}{[\omega_T^2(\mathbf{r})-\omega^2]}. \tag{4}$$

At high frequencies ($\omega\gg\omega_T$) $\mathbf{P}(\mathbf{r},\omega)\to\mathbf{0}$ and therefore the electronic polarization $\mathbf{P}_e$ becomes particularly important. We shall denote its contribution to the total susceptibility by χ_∞ so we can define the total polarization of the system as

$$\mathbf{P}_{tot}(\mathbf{r},\omega)=[\chi_\infty(\mathbf{r})+\chi(\mathbf{r},\omega)]\mathbf{E}(\mathbf{r},\omega).$$

The displacement $\mathbf{D}$ which enters the Maxwell equations (1) is related to the total polarization, so for the periodical solution we can write

$$\mathbf{D}(\mathbf{r},\omega)=\mathbf{E}(\mathbf{r},\omega)+4\pi\mathbf{P}_{tot}(\mathbf{r},\omega)=\epsilon(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega),$$

where we have introduced the standard Lorentz-type dielectric function for a polar dielectric

$$\epsilon(\mathbf{r},\omega)=\epsilon_\infty(\mathbf{r})+4\pi\chi(\mathbf{r},\omega), \tag{5}$$

$$\epsilon_\infty(\mathbf{r})=1+4\pi\chi_\infty(\mathbf{r}).$$

Now we can write the displacement $\mathbf{D}(\mathbf{r},t)$ in the form

$$\mathbf{D}(\mathbf{r},t)=\epsilon_\infty(\mathbf{r})\mathbf{E}(\mathbf{r},t)+4\pi\mathbf{P}(\mathbf{r},t) \tag{6}$$

which clearly distinguishes the electronic (ϵ_∞) and ionic ($\mathbf{P}$) contribution of the total polarization.

Let us quote two simple relations, which we shall use in later calculations,

$$\frac{\partial(\omega\epsilon)}{\partial\omega}=\epsilon_\infty+\frac{\omega_P^2(\omega_T^2+\omega^2)}{(\omega_T^2-\omega^2)^2},\quad \frac{\partial(\omega^2\epsilon)}{\partial\omega^2}=\epsilon_\infty+\frac{\omega_P^2\omega_T^2}{(\omega_T^2-\omega^2)^2}. \tag{7}$$

We introduce the vector $\mathbf{A}(\mathbf{r},t)$ and the scalar $\Phi(\mathbf{r},t)$ potentials in the standard way,

$$\mathbf{B}(\mathbf{r},t)=\boldsymbol{\nabla}\times\mathbf{A}(\mathbf{r},t),\quad \mathbf{E}(\mathbf{r},t)=-\boldsymbol{\nabla}\Phi(\mathbf{r},t)-\frac{1}{c}\frac{\partial\mathbf{A}(\mathbf{r},t)}{\partial t}, \tag{8}$$

which leads to the following equations of motion for the potentials:

$$\boldsymbol{\nabla}\cdot(\epsilon_\infty\boldsymbol{\nabla}\Phi)+\frac{1}{c}\frac{\partial}{\partial t}\boldsymbol{\nabla}\cdot(\epsilon_\infty\mathbf{A})=4\pi(\boldsymbol{\nabla}\cdot\mathbf{P}-\rho), \tag{9}$$

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{A}+\frac{1}{c^2}\epsilon_\infty\frac{\partial^2\mathbf{A}}{\partial t^2}+\frac{1}{c}\epsilon_\infty\frac{\partial\boldsymbol{\nabla}\Phi}{\partial t}=\frac{4\pi}{c}\left(\mathbf{j}+\frac{\partial\mathbf{P}}{\partial t}\right). \tag{10}$$

Here we have omitted denoting explicit dependence of potentials on $(\mathbf{r},t)$, and we shall use this short notation when such a dependence is obvious.

The potentials $\mathbf{A}$, Φ are usually solved in the Coulomb gauge,

$$\boldsymbol{\nabla}\cdot(\epsilon_\infty\mathbf{A})=0, \tag{11}$$

in which the vector potential $\mathbf{A}$ essentially describes the transverse and the scalar potential Φ the longitudinal field. This gauge is dominantly used when one discusses the radiation (transverse) field where the scalar potential can be neglected. In this paper we wish to discuss both (transverse and longitudinal) fields. Besides giving the complete theory of polaritons in dispersive dielectrics, this will enable us to make a close connection with the nonretarded limit.

Let us perform the standard gauge transformation,

$$\mathbf{A}=\mathbf{A}_0-c\boldsymbol{\nabla}\lambda,\quad \Phi=\phi+\frac{\partial\lambda}{\partial t}, \tag{12}$$

and choose the parameter λ so that a new vector potential $\mathbf{A}_0$ satisfies the following gauge requirement:

$$\boldsymbol{\nabla}\cdot\left(\epsilon_\infty\frac{\partial\mathbf{A}_0}{\partial t}-4\pi c\mathbf{P}\right)=0. \tag{13}$$

In this gauge, $\mathbf{A}_0$ satisfies Eq. (10) with the obvious replacement, $\mathbf{A}\to\mathbf{A}_0$, $\Phi\to\phi$, while the scalar potential ϕ is determined by the external charges only:

$$\boldsymbol{\nabla}\cdot(\epsilon_\infty\boldsymbol{\nabla}\phi)=-4\pi\rho. \tag{14}$$

The electromagnetic field,

$$\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{A}_0}{\partial t}-\boldsymbol{\nabla}\phi,\quad \mathbf{B}=\boldsymbol{\nabla}\times\mathbf{A}_0, \tag{15}$$

is generally determined by both potentials $\mathbf{A}_0$ and ϕ, but in the absence of external charges, the field (transverse and longitudinal) is completely described by $\mathbf{A}_0$.

The equations of motion for the renormalized potentials $\mathbf{A}_0$, ϕ, along with the Lorentz force equation for external charges, can be derived from the Lagrangian

$$L=L_f+L_e+L_i, \tag{16}$$

where the electromagnetic and polarization fields are described by

$$L_f=\int d\mathbf{r}\left\{\frac{1}{8\pi}\left[\epsilon_\infty\left(\frac{1}{c}\dot{\mathbf{A}}_0\right)^2-(\boldsymbol{\nabla}\times\mathbf{A}_0)^2\right]+\frac{2\pi}{\omega_P^2}(\dot{\mathbf{P}}^2-\omega_T^2\mathbf{P}^2)-\frac{1}{c}(\mu\mathbf{P}\cdot\dot{\mathbf{A}}_0-\nu\dot{\mathbf{P}}\cdot\mathbf{A}_0)\right\}. \tag{17}$$

External particles, with a mass m_i, a charge e_i, and at a position $\mathbf{r}_i$, are represented by

$$L_e=\frac{1}{2}\sum_i m_i\dot{\mathbf{r}}_i^2$$

and the interaction between the fields and the external particles is given by

$$L_i=\int d\mathbf{r}\left(\frac{1}{c}\mathbf{j}\cdot\mathbf{A}_0-\rho\phi\right)+\int d\mathbf{r}\left[\frac{1}{8\pi}\epsilon_\infty(\boldsymbol{\nabla}\phi)^2+\frac{1}{4\pi}\boldsymbol{\nabla}\phi\cdot\left(\frac{1}{c}\epsilon_\infty\dot{\mathbf{A}}_0-4\pi\mathbf{P}\right)\right].$$

Let us point out the following.

(i) In the Coulomb gauge (11), even in the absence of external charges, the polariton field is described by both (vector and scalar) potentials, so that the second integral in L_i should be the part of L_f, which would make the quantization of the field Hamiltonian more complicated.

(ii) The field Lagrangian L_f contains two parameters μ and ν. They are not quite arbitrary but must satisfy

$$\mu+\nu=1$$

so that L_f contains a term $\partial(\mathbf{P}\cdot\mathbf{A}_0)/\partial t$ which does not influence the equations of motion or the Hamiltonian of the system. The transformation from $(\mu=1,\nu=0)$ to $(\mu=0,\nu=1)$ can be viewed as a gauge transformation [5], but our approach enables us to treat both types of Lagrangian in the same form, which we shall extensively use in Sec. V.

The momenta conjugate to the dynamical variables $\mathbf{A}_0$, $\mathbf{P}$, $\mathbf{r}_i$ are derived from Eq. (16):

$$\mathbf{A}_0,\quad \boldsymbol{\Pi}_\mathbf{A}=-\frac{1}{4\pi c}\left[\epsilon_\infty\left(-\frac{1}{c}\dot{\mathbf{A}}_0\right)+\mu 4\pi\mathbf{P}\right]+\frac{1}{4\pi c}\epsilon_\infty\boldsymbol{\nabla}\phi; \tag{18}$$

$$\mathbf{P}, \quad \mathbf{\Pi_P} = \frac{4\pi}{\omega_P^2}\dot{\mathbf{P}} + \nu\frac{1}{c}\mathbf{A}_0; \tag{19}$$

$$\mathbf{r}_i, \quad \mathbf{p}_i = m_i\dot{\mathbf{r}}_i + \frac{e_i}{c}\mathbf{A}_{0i}. \tag{20}$$

Usually, one takes $(\mu=1,\nu=0)$, which gives $\mathbf{\Pi_A}\sim\mathbf{D}$, but the opposite choice $(\mu=0,\nu=1)$, i.e., $\mathbf{\Pi_A}\sim\mathbf{E}$, is sometimes also preferred [2].

Now we are able to obtain the Hamiltonian of the system which can be factorized in the similar way as the Lagrangian (16):

$$H = H_f + H_e + H_i. \tag{21}$$

The field Hamiltonian

$$H_f = \int d\mathbf{r}\left\{\frac{1}{8\pi}\left[\frac{\epsilon_\infty}{c^2}\dot{\mathbf{A}}_0^2 + (\boldsymbol{\nabla}\times\mathbf{A}_0)^2\right] + \frac{2\pi}{\omega_P^2}(\dot{\mathbf{P}}^2 + \omega_T^2\mathbf{P}^2)\right\} \tag{22}$$

is determined by the fields $\mathbf{A}_0$, $\mathbf{P}$. The particle Hamiltonian

$$H_e = \frac{1}{2}\sum_i\ [m_i\dot{\mathbf{r}}_i^2 + e_i\phi_i]$$

besides the kinetic-energy operator also includes the self-energy term. The interaction Hamiltonian describes a linear coupling of the polarization with the external potential:

$$H_i = \int d\mathbf{r}\mathbf{P}\cdot\boldsymbol{\nabla}\phi. \tag{23}$$

III. QUANTIZATION OF THE FIELD HAMILTONIAN

The Hamiltonian H_f describes the energy of the polariton field, i.e., the energy of electromagnetic and polarization fields coupled through the equations of motion (1) and (2). In order to quantize that Hamiltonian, we shall expand field operators $\mathbf{A}_0$ and $\mathbf{P}$ in terms of their eigenfunctions $\mathbf{A}_{Ks}$ and $\mathbf{P}_{Ks}$, respectively, defined so as to satisfy Eqs. (2), (10), and (13) in the absence of the external charges:

$$\ddot{\mathbf{P}}_{Ks} + \omega_T^2\mathbf{P}_{Ks} = \frac{\omega_P^2}{4\pi}\left(-\frac{1}{c}\dot{\mathbf{A}}_{Ks}\right), \tag{24}$$

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{A}_{Ks} + \frac{1}{c^2}\epsilon_\infty\ddot{\mathbf{A}}_{Ks} = \frac{4\pi}{c}\dot{\mathbf{P}}_{Ks}, \tag{25}$$

$$\boldsymbol{\nabla}\cdot(\epsilon_\infty\dot{\mathbf{A}}_{Ks}) = 4\pi c\boldsymbol{\nabla}\cdot\mathbf{P}_{Ks}. \tag{26}$$

In order to determine the corresponding eigenfrequencies of a coupled system, we assume

$$\mathbf{A}_{Ks} = \mathbf{A}_{Ks}(\mathbf{r})\exp(-i\omega_K t),$$
$$\mathbf{P}_{Ks} = \mathbf{P}_{Ks}(\mathbf{r})\exp(-i\omega_K t). \tag{27}$$

Here indices (K,s) denote all linearly independent solutions of the field eigenmodes $\mathbf{A}_{Ks}$, $\mathbf{P}_{Ks}$. Particularly, index $s(K)$ denotes all degenerate eigenmodes that have the same eigenfrequency ω_K.

Now we can define the susceptibility $\chi_K(\mathbf{r})\equiv\chi(\mathbf{r},\omega_K)$ in the same way as in Eq. (3),

$$\mathbf{P}_{Ks}(\mathbf{r}) = i\frac{\omega_K}{c}\chi_K(\mathbf{r})\mathbf{A}_{Ks}(\mathbf{r}), \tag{28}$$

and with the notation $\epsilon_K(\mathbf{r})\equiv\epsilon(\mathbf{r},\omega_K)$ we can rewrite the gauge condition (26) and the eigenequation for the vector potential (25) in the following transparent form:

$$\boldsymbol{\nabla}\cdot(\epsilon_K\mathbf{A}_{Ks}) = 0, \tag{29}$$

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{A}_{Ks} - \frac{1}{c^2}\epsilon_K\omega_K^2\mathbf{A}_{Ks} = \mathbf{0}. \tag{30}$$

This is not a standard Hermitian problem [19], but Eqs. (29) and (30) would reduce to it if a dielectric function ϵ would not depend upon the eigenfrequency ω_K [16,17]. Otherwise we may have different eigenfrequencies ω for the same "eigenvalue" $\epsilon\omega^2$ (see Sec. IV C).

Let us make a standard assumption that a dielectric function ϵ_K does not depend upon the position within a given dielectric. Therefore, e.g., in the volume of a dielectric j we find $\epsilon_K^j\boldsymbol{\nabla}\cdot\mathbf{A}_{Ks}=0$, which gives the following.

(i) *Longitudinal modes.* $\epsilon_K^j=0$, $\boldsymbol{\nabla}\times\mathbf{A}_{Ks}=\mathbf{0}$.

All modes $K=j$ are degenerate, with the longitudinal frequency

$$\omega_j^2 = \omega_L^{j2} \equiv \omega_T^{j2} + \frac{\omega_P^{j2}}{\epsilon_\infty^j}, \tag{31}$$

and they all vanish outside the dielectric j. Because the modes satisfy $\boldsymbol{\nabla}\times\mathbf{A}_0=\mathbf{0}$, they can also be determined by the scalar potential Φ_0, introduced as $\mathbf{E}=-(1/c)\dot{\mathbf{A}}_0=-\boldsymbol{\nabla}\Phi_0$.

(ii) *Transverse modes.* $\epsilon_K^j\neq 0$, $\boldsymbol{\nabla}\cdot\mathbf{A}_{Ks}=0$. The modes satisfy the wave equation

$$\Delta\mathbf{A}_{Ks}(\mathbf{r}) + \epsilon_K^j\frac{\omega_K^2}{c^2}\mathbf{A}_{Ks}(\mathbf{r}) = 0 \tag{32}$$

and the eigenfrequencies ω_K depend upon the boundaries between dielectrics.

Generally, we can expand the vector potential and the polarization over their eigenmodes,

$$\mathbf{A}_0(\mathbf{r},t) = \frac{1}{\sqrt{V}}\sum_{Ks}\ [r_{Ks}(t)\mathbf{A}_{Ks}(\mathbf{r}) + r_{Ks}^\dagger(t)\mathbf{A}_{Ks}^*(\mathbf{r})], \tag{33}$$

$$\mathbf{P}(\mathbf{r},t) = \frac{1}{\sqrt{V}}\sum_{Ks}\ [r_{Ks}(t)\mathbf{P}_{Ks}(\mathbf{r}) + r_{Ks}^\dagger(t)\mathbf{P}_{Ks}^*(\mathbf{r})], \tag{34}$$

where V is the normalization volume and $r_{Ks}(t)$, which enters into both expansions, is the polariton field operator, with the following standard time dependence:

$$r_{Ks}(t)=r_{Ks}\exp(-i\omega_K t). \tag{35}$$

Using this expression one can easily calculate the time derivatives of the fields $\mathbf{A}_0$ and $\mathbf{P}$ and expand the conjugate momenta (18) and (19) in the way analogous to the expansions (33) and (34):

$$\mathbf{\Pi}_{\mathbf{A}}(\mathbf{r},t)=\frac{(-i)}{(4\pi c)^2}\frac{1}{\sqrt{V}}\sum_{Ks}4\pi\omega_K\left(\epsilon_\infty+\mu\frac{\omega_P^2}{\omega_T^2-\omega_K^2}\right)[r_{Ks}(t)\mathbf{A}_{Ks}(\mathbf{r})-r_{Ks}^\dagger(t)\mathbf{A}_{Ks}^*(\mathbf{r})], \tag{36}$$

$$\mathbf{\Pi}_{\mathbf{P}}(\mathbf{r},t)=\frac{(-i)}{\omega_P^2}\frac{1}{\sqrt{V}}\sum_{Ks}4\pi\omega_K\left(\mu+\nu\frac{\omega_T^2}{\omega_K^2}\right)[r_{Ks}(t)\mathbf{P}_{Ks}(\mathbf{r})-r_{Ks}^\dagger(t)\mathbf{P}_{Ks}^*(\mathbf{r})]. \tag{37}$$

Now we shall proceed with the quantization of the Hamiltonian H_f. In Eq. (22), we shall rewrite the magnetic term as

$$\int d\mathbf{r}(\nabla\times A_0)^2=\frac{1}{2}\int d\mathbf{r}[(\nabla\times\nabla\times\mathbf{A}_0)\cdot\mathbf{A}_0+\mathbf{A}_0\cdot(\nabla\times\nabla\times\mathbf{A}_0)] \tag{38}$$

in order to obtain all terms in the Hamiltonian in the symmetric form, which will enable us an appropriate factorization. Inserting the expansions (33) and (34) into Eq. (22) and using Eqs. (30) and (38) we find

$$H_f=\int d\mathbf{r}\frac{1}{V}\sum_{Ks}\sum_{K's'}[X^-_{KsK's'}(\mathbf{r},t)Y^-_{KK'}(\mathbf{r})+X^+_{KsK's'}(\mathbf{r},t)Y^+_{KK'}(\mathbf{r})], \tag{39}$$

where we have introduced the following abbreviations:

$$\begin{aligned}X^-_{KsK's'}&=r_{Ks}r_{K's'}\mathbf{A}_{Ks}\cdot\mathbf{A}_{K's'}+r^\dagger_{Ks}r^\dagger_{K's'}\mathbf{A}^*_{Ks}\cdot\mathbf{A}^*_{K's'},\\ X^+_{KsK's'}&=r_{Ks}r^\dagger_{K's'}\mathbf{A}_{Ks}\cdot\mathbf{A}^*_{K's'}+r^\dagger_{Ks}r_{K's'}\mathbf{A}^*_{Ks}\cdot\mathbf{A}_{K's'},\end{aligned} \tag{40}$$

$$Y^\mp_{KK'}=\frac{1}{4c^2}(\omega_K\mp\omega_{K'})\left[\frac{1}{4\pi}(\epsilon_K\omega_K\mp\epsilon_{K'}\omega_{K'})\mp\frac{4\pi}{\omega_P^2}(\omega_K\mp\omega_{K'})\omega_K\chi_K\omega_{K'}\chi_{K'}\right].$$

Inserting the relations (4) for χ_K and (5) for ϵ_K we finally obtain

$$Y^\mp_{KK'}(\mathbf{r})=\frac{1}{16\pi c^2}(\omega_K\mp\omega_{K'})^2Z_{KK'}(\mathbf{r}), \tag{41}$$

$$Z_{KK'}(\mathbf{r})=\epsilon_\infty(\mathbf{r})+\frac{\omega_P^2(\mathbf{r})\omega_T^2(\mathbf{r})}{[\omega_T^2(\mathbf{r})-\omega_K^2][\omega_T^2(\mathbf{r})-\omega_{K'}^2]}. \tag{42}$$

The function $Z_{KK'}$ will have an important role in further considerations. Let us note that we can write it in a more symmetric form,

$$Z_{KK'}(\mathbf{r})=\frac{1}{(\omega_K^2-\omega_{K'}^2)}[\omega_K^2\epsilon_K(\mathbf{r})-\omega_{K'}^2\epsilon_{K'}(\mathbf{r})]. \tag{43}$$

From Eq. (43) we find a finite value of function Z when both indices are equal:

$$Z_{KK}(\mathbf{r})=\epsilon_\infty(\mathbf{r})+\frac{\omega_P^2(\mathbf{r})\omega_T^2(\mathbf{r})}{[\omega_T^2(\mathbf{r})-\omega_K^2]^2}=\frac{\partial(\omega_K^2\epsilon_K(\mathbf{r}))}{\partial\omega_K^2}. \tag{44}$$

The last relation is derived with the help of Eq. (7).

In order to perform the integration in Eq. (39) we must derive a suitable relation for the eigenvectors $\mathbf{A}_{Ks}$. We shall start with the vector identity

$$\begin{aligned}&\mathbf{A}_2^{(*)}\cdot\nabla\times\nabla\times\mathbf{A}_1-\mathbf{A}_1\cdot\nabla\times\nabla\times\mathbf{A}_2^{(*)}\\&\quad=\nabla\cdot[(\nabla\times\mathbf{A}_1)\times\mathbf{A}_2^{(*)}-(\nabla\times\mathbf{A}_2^{(*)})\times\mathbf{A}_1],\end{aligned} \tag{45}$$

where $(*)$ means that the equation holds with or without the complex conjugation. Equation (45), applied to the eigenvectors $\mathbf{A}_{Ks}$, $\mathbf{A}_{K's'}$ that satisfy Eq. (30), leads to the following general form of the orthogonality relation for the vector potential eigenmodes:

$$\int d\mathbf{r}\mathbf{A}^{(*)}_{K's'}(\mathbf{r})\cdot\mathbf{A}_{Ks}(\mathbf{r})[\omega_K^2\epsilon_K(\mathbf{r})-\omega_{K'}^2\epsilon_{K'}(\mathbf{r})]=0. \tag{46}$$

Note that the orthogonality relation is derived with the assumption.

$$\mathbf{A}_{Ks}(\mathbf{r}\to\infty)=\mathbf{0}. \tag{47}$$

Such a behavior is typical for surface modes, and in Sec. IV C we shall show that Eq. (46) also holds for plane waves. Using the definition of the function $Z_{KK'}$, Eq. (43), we can rewrite Eq. (46) as

$$\int d\mathbf{r} Z_{KK'}(\mathbf{r}) \mathbf{A}^{(*)}_{K's'}(\mathbf{r}) \cdot \mathbf{A}_{Ks}(\mathbf{r}) = 0, \quad K \neq K'. \tag{48}$$

For the degenerate modes ($K=K', s \neq s'$), function Z_{KK}, Eq. (44), is finite, positive, and independent of s. In the subspace determined by the degenerate eigenvectors we can choose eigenvectors $\mathbf{A}_{Ks}$ as orthogonal, so we can write the orthonormality relation in the form

$$\frac{1}{V}\int d\mathbf{r} Z_{KK'}(\mathbf{r}) \mathbf{A}^{*}_{K's'}\mathbf{r}) \cdot \mathbf{A}_{Ks}(\mathbf{r}) = N_K \delta_{K,K'} \delta_{s,s'}, \tag{49}$$

where $N_K > 0$ is a suitably chosen normalization parameter, e.g., $N_K = 1$. Note that from the orthogonality relations (49) we cannot derive a suitable closure relation because the eigenfunctions (30) are not solutions of a standard Hermitian problem [19,25].

The quantization of the Hamiltonian H_f now becomes simple. The integration over $\mathbf{r}$ in Eq. (39) includes $\mathbf{A}^{(*)}_{K's'} \cdot \mathbf{A}_{Ks}$ terms from the functions $X^{\pm}_{KsK's'}$, Eq. (40), and $Z_{KK'}$ terms from the functions $Y^{\pm}_{KK'}$, Eq. (41). From the orthogonality relation (48) it follows that only terms with $K=K'$ contribute to the Hamiltonian (39). But in that case we have $\omega_K = \omega_{K'}$, which gives $Y^{-}_{KK}=0$, so in Eq. (39) only terms $X^{+}_{KsKs'} Y^{+}_{KK}$ remain. With the orthonormality condition (49), we arrive at

$$H_f = \frac{1}{4\pi c^2} \sum_{Ks} N_K \omega_K^2 [r_{Ks} r^{\dagger}_{Ks} + r^{\dagger}_{Ks} r_{Ks}]. \tag{50}$$

Let us now introduce in the standard way the following creation and annihilation operators:

$$r_{Ks} = \zeta_K a_{Ks}, \tag{51}$$

$$[a_{Ks}, a^{\dagger}_{K's'}] = \delta_{K,K'} \delta_{s,s'}, \quad [a_{Ks}, a_{K's'}] = 0. \tag{52}$$

With the choice

$$\zeta_K = \sqrt{hc^2 \omega_K N_K} \tag{53}$$

we can write the Hamiltonian (50) in the following second-quantized form:

$$H_f = \sum_{Ks} \hbar \omega_K \left(a^{\dagger}_{Ks} a_{Ks} + \frac{1}{2} \right). \tag{54}$$

The expansion of the fundamental field operators $\mathbf{A}_0$, Eq. (33), and $\mathbf{P}$, Eq. (34), in terms of the polariton creation and annihilation operators $(a_{Ks}, a^{\dagger}_{Ks})$, Eqs. (52), takes the form

$$\mathbf{A}_0(\mathbf{r},t) = \frac{1}{\sqrt{V}} \sum_{Ks} \zeta_K [a_{Ks} \exp(-i\omega_K t) \mathbf{A}_{Ks}(\mathbf{r}) + a^{\dagger}_{Ks} \exp(i\omega_K t) \mathbf{A}^{*}_{Ks}(\mathbf{r})], \tag{55}$$

$$\mathbf{P}(\mathbf{r},t) = \frac{1}{\sqrt{V}} \sum_{Ks} \zeta_K [a_{Ks} \exp(-i\omega_K t) \mathbf{P}_{Ks}(\mathbf{r}) + a^{\dagger}_{Ks} \exp(i\omega_K t) \mathbf{P}^{*}_{Ks}(\mathbf{r})], \tag{56}$$

and the equivalent expansion of the electromagnetic field operators $\mathbf{E}$, $\mathbf{B}$, Eqs. (15), or the interaction of polaritons with external sources (23), becomes obvious.

IV. SPECIAL CASES

In this section we shall apply our general results to some specific problems that have been already discussed in the literature. It will enable us not only to compare the existing results with ours, but also to give more detailed explanation of some intriguing questions.

A. Planar symmetry

Let us suppose that our system consists of thin dielectric layers with plates parallel in the $\boldsymbol{\rho}$ direction. In that case ϵ_∞, ω_P, ω_T are only functions of z so we can put

$$\epsilon_K(\mathbf{r}) = \epsilon_K(z) = \epsilon_\infty(z) + 4\pi\chi_K(z).$$

The eigenfunctions of the vector potential can be factorized as

$$\mathbf{A}_{Ks}(\mathbf{r}) = \mathbf{A}_{Ks}(z) \exp(i\mathbf{k}\cdot\boldsymbol{\rho}), \tag{57}$$

where $\mathbf{k}$ is a two-dimensional wave vector. The normalization volume V is connected with the normalization length L as $V = L\int d\boldsymbol{\rho}$ so, e.g., the orthonormality relation (49) becomes

$$\frac{1}{L}\int dz \frac{1}{(\omega_K^2 - \omega_{K'}^2)} [\omega_K^2 \epsilon_K(z) - \omega_{K'}^2 \epsilon_{K'}(z)] \mathbf{A}^{*}_{K's'}(z) \cdot \mathbf{A}_{Ks}(z) = N_K \delta_{K,K'} \delta_{s,s'}.$$

The equivalent relation was derived by Santos and Loudon [19] who analyzed the electromagnetic field in a one-dimensional dielectric with an arbitrary dielectric function $\epsilon_K(z)$.

B. Frequency independent dielectric function

Let us assume that the dielectric function is independent of a frequency ω_K, i.e., $\epsilon = \epsilon(\mathbf{r})$. In that case we have $\chi_K = 0$, i.e., we neglect the time-dependent influence of the ionic polarization $\mathbf{P}$. It is correct at high frequencies ($\omega_K \gg \omega_P$) where only electronic polarizability remains. In our model we can simply put $\mathbf{P}=\mathbf{0}$ or $\omega_P = 0$.

The vector potential eigenmodes $\mathbf{A}_{Ks}$ now satisfy Eqs. (29) and (30) with the substitution $\epsilon_K(\mathbf{r}) = \epsilon_\infty(\mathbf{r})$. The momentum $\boldsymbol{\Pi}_{\mathbf{A}}$, Eq. (18), conjugate to the vector potential is

$$\boldsymbol{\Pi}_{\mathbf{A}} = \frac{1}{4\pi c^2} \epsilon_\infty \dot{\mathbf{A}}_0$$

and the Hamiltonian of the electromagnetic field takes the standard form

$$H_{em}=\int d\mathbf{r}\frac{1}{8\pi}\left[\epsilon_\infty\frac{1}{c^2}\dot{\mathbf{A}}_0^2+(\boldsymbol{\nabla}\times\mathbf{A}_0)^2\right]. \tag{58}$$

In the $\omega_P=0$ limit, Eq. (43) gives

$$Z_{KK'}(\mathbf{r})=\epsilon_\infty(\mathbf{r}) \tag{59}$$

so the orthonormality relation (49) becomes

$$\frac{1}{V}\int d\mathbf{r}\epsilon_\infty(\mathbf{r})\mathbf{A}^*_{K's'}(\mathbf{r})\cdot\mathbf{A}_{Ks}(\mathbf{r})=N_K\delta_{K,K'}\delta_{s,s'}. \tag{60}$$

It now contains the weighting factor $\epsilon_\infty(\mathbf{r})$ which does not depend upon the eigenmode index K so we can derive the closure relation in the simple form

$$\frac{1}{V}\sum_{Ks}\frac{1}{N_K}\epsilon_\infty(\mathbf{r})A^\alpha_{Ks}(\mathbf{r})A^{\beta*}_{Ks}(\mathbf{r}')=\delta^T_{\alpha,\beta}(\mathbf{r}-\mathbf{r}'), \tag{61}$$

where from now on we shall use (α,β) to denote the Cartesian coordinates.

The relations equivalent to Eqs. (60) and (61) were derived in Ref. [17], where the detailed explanation of the properties of the transverse δ function (with the notation $\delta^T_{\alpha,\beta}=\epsilon\delta^\epsilon_{\alpha,\beta}$) is also given; it appears here because of the absence of the longitudinal modes. In fact, our results can be directly connected to the results of Ref. [17] because, with the assumption $\epsilon_K=\epsilon_\infty$, we are using the same gauge condition (29): $\boldsymbol{\nabla}\cdot[\epsilon_\infty(\mathbf{r})\mathbf{A}_{Ks}(\mathbf{r})]=0$.

C. Spatially independent dielectric function

Let us assume that the whole space is homogeneous, i.e., occupied by the same dielectric with the spatially independent dielectric function ϵ_K. Then the vector potential eigenmodes $\mathbf{A}_{Ks}$ satisfy Eqs. (31) and (32) with the obvious substitution $\epsilon^j_K=\epsilon_K$. The solutions for the eigenmodes are plane waves

$$\mathbf{A}^p_{\mathbf{K}n}(\mathbf{r})=C^p_{Kn}\mathbf{e}^p_{\mathbf{K}}\exp(i\mathbf{K}\cdot\mathbf{r}) \tag{62}$$

determined by the three-dimensional wave vector $\mathbf{K}$, index p which denotes one longitudinal $(p=L)$ and two transverse $(p=T1,T2)$ polarizations with the unit vectors $\mathbf{e}^p_{\mathbf{K}}$, and index $n(p)$ which counts polariton branches for each polarization p. C^p_{Kn} is a normalization constant. All the modes with the *longitudinal* polarization have the same frequency ω_L, Eq. (31), while the *transverse* modes have two branches $(n=+,-)$ for $\omega_T>0$ and one branch $(n=+)$ for $\omega_T=0$, determined by the well-known dispersion relation

$$\epsilon_{Kn}\omega^2_{Kn}=K^2c^2. \tag{63}$$

Since the plane waves do not satisfy boundary condition (47) we cannot take the orthogonality relation (48) for granted. To prove that it holds for plane waves, we first note that they satisfy the following orthogonality and closure relations:

$$\frac{1}{V}\int d\mathbf{r}e^{i(\mathbf{K}-\mathbf{K}')\cdot\mathbf{r}}=\delta_{\mathbf{K},\mathbf{K}'},$$

$$\frac{1}{V}\sum_{\mathbf{K}}e^{i(\mathbf{r}-\mathbf{r}')\cdot\mathbf{K}}=\delta(\mathbf{r}-\mathbf{r}').$$

The same holds for the polarization vectors:

$$\mathbf{e}^p_{\mathbf{K}}\cdot\mathbf{e}^{p'}_{\mathbf{K}}=\delta_{p,p'},$$

$$\sum_p e^{p\alpha}_{\mathbf{K}}e^{p\beta}_{\mathbf{K}}=\delta_{\alpha,\beta}.$$

In principle, we can denote the polarization vectors $\mathbf{e}^p_{\mathbf{K}}$ as $\mathbf{e}^p_{\mathbf{K}n}$ and (for $\omega_T>0$) choose, e.g., $\mathbf{e}^{T1}_{\mathbf{K}-}$ and $\mathbf{e}^{T1}_{\mathbf{K}+}$ as different vectors. However, the polarization of two branches are not correlated and we can simply take the same polarization vector $\mathbf{e}^{T1}_{\mathbf{K}}$ for both $(+,-)$ modes. Note that the polarization vectors of different branches cannot be chosen as mutually orthogonal because there are four vectors $\{\mathbf{e}^T_{\mathbf{K}n}\}$ in the same two-dimensional space.

In order to derive orthogonality relation for different polariton branches we shall rewrite the factor $Z^p_{KnK'n'}$, Eq. (43), using the corresponding dispersion relations:

$$Z^L_{KnKn}=\epsilon^2_\infty\frac{\omega^2_L}{\omega^2_P},\quad Z^T_{KnK'n'}=\frac{c^2(K^2-K'^2)}{\omega^2_{Kn}-\omega^2_{K'n'}}.$$

These equations clearly show that for $K=K'$, Z factor does not vanish only if $n=n'$ so we can write the orthogonality relation in the form

$$Z_{K\sigma K'\sigma'}\frac{1}{V}\int d\mathbf{r}\mathbf{A}^{(*)}_{\mathbf{K}'\sigma'}(\mathbf{r})\cdot\mathbf{A}_{\mathbf{K}\sigma}(\mathbf{r})=0,\quad \{\mathbf{K}\sigma\}\neq\{\mathbf{K}'\sigma'\}, \tag{64}$$

where we have introduced the index $\sigma=\{p,n(p)\}$ to shorten the notation. This relation is obviously equivalent to the relation (48), so in agreement with Eq. (49) we can write the orthonormality relation as

$$Z_{K\sigma K'\sigma'}\frac{1}{V}\int d\mathbf{r}\mathbf{A}^*_{\mathbf{K}'\sigma'}(\mathbf{r})\cdot\mathbf{A}_{\mathbf{K}\sigma}(\mathbf{r})=N_K\delta_{\mathbf{K},\mathbf{K}'}\delta_{\sigma,\sigma'}. \tag{65}$$

Now we can put the Hamiltonian H_f (22) in the second-quantized form (54) in the same way as in Sec. III, replacing the general quantum numbers $\{Ks\}$ with $\{\mathbf{K}\sigma\}$.

As we have pointed out in the Introduction, the Hamiltonian H_f is often postulated in the alternative form, well known in classical electrodynamics. We shall here derive that form by calculating H_f part by part. Using the relations (64) and (65) we shall first calculate the term which contains only the electric field,

$$H_{El}=\int d\mathbf{r}\frac{1}{8\pi}\epsilon_\infty \mathbf{E}^2$$

$$=\frac{1}{8\pi c^2}\sum_{\mathbf{K}\sigma}\omega_{K\sigma}^2\epsilon_\infty|C_{K\sigma}|^2(r_{\mathbf{K}\sigma}r_{\mathbf{K}\sigma}^\dagger+r_{\mathbf{K}\sigma}^\dagger r_{\mathbf{K}\sigma}),$$

and then the term which includes only the polarization,

$$H_{Pol}=\int d\mathbf{r}\frac{2\pi}{\omega_P^2}(\dot{\mathbf{P}}^2+\omega_T^2\mathbf{P}^2)$$

$$=\frac{1}{8\pi c^2}\sum_{\mathbf{K}\sigma}\omega_{K\sigma}^2|C_{K\sigma}|^2\omega_P^2\frac{(\omega_{K\sigma}^2+\omega_T^2)}{(\omega_{K\sigma}^2-\omega_T^2)^2}$$

$$\times(r_{\mathbf{K}\sigma}r_{\mathbf{K}\sigma}^\dagger+r_{\mathbf{K}\sigma}^\dagger r_{\mathbf{K}\sigma}).$$

By adding these two terms we find, with the help of Eq. (7),

$$H_E=H_{El}+H_{Pol}$$

$$=\frac{1}{8\pi c^2}\sum_{\mathbf{K}\sigma}\omega_{K\sigma}^2\frac{\partial(\omega_{K\sigma}\epsilon_{K\sigma})}{\partial\omega_{K\sigma}}$$

$$\times|C_{K\sigma}|^2(r_{\mathbf{K}\sigma}r_{\mathbf{K}\sigma}^\dagger+r_{\mathbf{K}\sigma}^\dagger r_{\mathbf{K}\sigma}),$$

which has the same form as H_{El} if we replace

$$\epsilon_\infty\rightarrow\frac{\partial(\omega_{K\sigma}\epsilon_{K\sigma})}{\partial\omega_{K\sigma}}. \tag{66}$$

The magnetic term of H_f also takes a simple form

$$H_B=\int d\mathbf{r}\frac{1}{8\pi}\mathbf{B}^2=\frac{1}{8\pi c^2}\sum_{\mathbf{K}\sigma}\omega_{K\sigma}^2\epsilon_{K\sigma}|C_{K\sigma}|^2$$

$$\times(r_{\mathbf{K}\sigma}r_{\mathbf{K}\sigma}^\dagger+r_{\mathbf{K}\sigma}^\dagger r_{\mathbf{K}\sigma}),$$

so we can write formally the whole Hamiltonian as

$$H_f=H_E+H_B=\int d\mathbf{r}\frac{1}{8\pi}\sum_{\mathbf{K}\sigma}\left[\frac{\partial(\omega_{K\sigma}\epsilon_{K\sigma})}{\partial\omega_{K\sigma}}\mathbf{E}_{\mathbf{K}\sigma}^2+\mathbf{B}_{\mathbf{K}\sigma}^2\right], \tag{67}$$

where only the diagonal parts of $\mathbf{E}^2$ and $\mathbf{B}^2$ contribute to H_f. Let us note that parts of the Hamiltonian (H_{El},H_{Pol},H_B) contain off-diagonal terms which are all canceled when the total Hamiltonian H_f is derived, in agreement with Eq. (54).

The Hamiltonian (67) was obtained by Landau and Lifshitz [13], with polariton operators $r_{\mathbf{K}\sigma}$ as c numbers. In a standard way it can be quantized *a posteriori* by the introduction of the creation and annihilation operators (51) and (52). To derive closer relation with the workers which use that approach [14,15] we shall first calculate, from Eqs. (62) and (64), the normalization constant $C_{K\sigma}$:

$$|C_{K\sigma}|^2=\frac{N_K}{Z_{K\sigma K\sigma}}. \tag{68}$$

For the transverse modes, which are usually analyzed, the simple dispersion relation (63) enables us to introduce the phase v^p and the group v^g velocity of the polaritons in a standard way:

$$v_{K\sigma}^p=\frac{\omega_{K\sigma}}{K},\quad v_{K\sigma}^g=\frac{\partial\omega_{K\sigma}}{\partial K}.$$

These velocities can be easily connected to the weighting factor $Z_{K\sigma K\sigma}$, Eq. (44):

$$\frac{c^2}{v_{K\sigma}^p v_{K\sigma}^g}=\frac{\partial(\omega_{K\sigma}^2\epsilon_{K\sigma})}{\partial\omega_{K\sigma}^2}=Z_{K\sigma K\sigma},$$

$$\frac{v_{K\sigma}^g}{v_{K\sigma}^p}=\frac{\epsilon_{K\sigma}}{Z_{K\sigma K\sigma}}. \tag{69}$$

If we choose the phase of $C_{K\sigma}$ to be $(-i)$, e.g., for the electric field (15), we find

$$\mathbf{E}=\frac{1}{V}\sum_{\mathbf{K}\sigma}\left[\sqrt{\frac{h\omega_{K\sigma}}{\epsilon_{K\sigma}}}\sqrt{\frac{v_{K\sigma}^g}{v_{K\sigma}^p}}a_{\mathbf{K}\sigma}\exp(i\mathbf{K}\cdot\mathbf{r})\mathbf{e}_{\mathbf{K}\sigma}+\text{H.c.}\right]$$

as derived in Ref. [14].

V. DISCUSSION: CONSISTENCY OF THE MODEL

In our approach to the problem of electromagnetic field in the polar medium, we have expanded the Hamiltonian of the system, H_f, Eq. (22), in terms of $\mathbf{A}_{Ks}(\mathbf{r})$ Eq. (25), which are the common eigenmodes of the vector potential $\mathbf{A}_0$ and the polarization $\mathbf{P}$ and therefore they represent the solution of the coupled system. The quantization of the Hamiltonian is then performed by factorizing the expansion coefficient $r_{Ks}(t)$, Eq. (51), into the operator $a_{Ks}(t)$ which satisfies the standard commutation relations (52), and the parameter ζ, Eq. (53), which is chosen to bring the Hamiltonian of the whole system into the diagonal form (54). *A posteriori* we wish to check the validity of the fundamental equal-time commutation relations (ETCR) between the Cartesian components (α,β) of the field $\mathbf{F}$ and the conjugate momentum $\mathbf{\Pi_F}$,

$$[F^\alpha(\mathbf{r},t),\Pi_{\mathbf{F}}^\beta(\mathbf{r}',t)]=i\hbar\,\delta_{\alpha,\beta}\delta(\mathbf{r}-\mathbf{r}'), \tag{70}$$

for the two fields $\mathbf{F}=\{\mathbf{A}_0,\mathbf{P}\}$ involved in our system.

As pointed out in the Introduction, although the present model is well established and based on the standard equations of motion, these equations lead to a real dielectric function which obviously violates the Kramers-Kronig relations. However, the Lorentz-type dielectric function $\epsilon(\mathbf{r},\omega)$, Eq. (5), can be viewed as a limiting case of a correct dielectric function [25] so we expect that the present model is consistent with ETCR.

The ETCR (70) for the fields $\mathbf{A}_0$ and $\mathbf{P}$ can be calculated from Eqs. (33), (34) and (36), (37) respectively:

$$[A_0^\alpha(\mathbf{r},t),\Pi_{\mathbf{A}}^\beta(\mathbf{r}',t)]=i\hbar\,\mathrm{Re}\left\{\frac{1}{V}\sum_{Ks}\frac{1}{N_K}[\epsilon_\infty(\mathbf{r}')\right.$$

$$\left.+\mu 4\pi\chi_K(\mathbf{r}')]A_{Ks}^\alpha(\mathbf{r})A_{Ks}^{\beta*}(\mathbf{r}')\right\}, \tag{71}$$

$$[P^{\alpha}(\mathbf{r},t),\Pi^{\beta}_{\mathbf{P}}(\mathbf{r}',t)]=i\hbar\,\mathrm{Re}\left\{\frac{1}{V}\sum_{Ks}\frac{(4\pi c)^2}{N_K}\frac{1}{\omega_P^2(\mathbf{r}')}\times\left[\mu+\nu\frac{\omega_T^2(\mathbf{r}')}{\omega_K^2}\right]P^{\alpha}_{Ks}(\mathbf{r})P^{\beta *}_{Ks}(\mathbf{r}')\right\}. \tag{72}$$

Note that the eigenmodes $\mathbf{A}_{Ks}$, $\mathbf{P}_{Ks}$ are coupled through the constitutive equation (28), so it could be a problem to satisfy both commutation relations (71) and (72) simultaneously. To clarify that problem we shall first analyze the special cases that include only one field.

(i) *The electromagnetic field*. The equations of motion of the electromagnetic field in the absence of the polarization field follow from Eq. (1), with $\mathbf{P}=\mathbf{0}$. That case was discussed in detail in Sec. IV B, where we have given the appropriate Hamiltonian H_{em}, Eq. (58). The corresponding commutation relations (obviously, with $\mu=0$) follow from Eq. (71):

$$[A_0^{\alpha}(\mathbf{r},t),\Pi^{\beta}_{\mathbf{A}}(\mathbf{r}',t)]=i\hbar\,\mathrm{Re}\left\{\frac{1}{V}\sum_{Ks}\frac{1}{N_K}\epsilon_{\infty}(\mathbf{r}')A^{\alpha}_{Ks}(\mathbf{r})A^{\beta *}_{Ks}(\mathbf{r}')\right\}. \tag{73}$$

Now we can apply the closure relation (61) to the right-hand side of Eq. (73) and it immediately leads to the correct ETCR (70) for the (bare) electromagnetic field.

(ii) *The (nonretarded) polarization field*. If we neglect the influence of the electromagnetic field, we obtain the *nonretarded* limit, discussed in the Appendix. The particle field is expressed in terms of polarization eigenmodes (A13), so we can easily derive the commutation relations between $\mathbf{P}$ and $\mathbf{\Pi_P}$ Eq. (A4):

$$[P^{\alpha}(\mathbf{r},t),\Pi^{\beta}_{\mathbf{P}}(\mathbf{r}',t)]=i\hbar\,\mathrm{Re}\left\{\frac{1}{V}\sum_{Ks}\frac{1}{\omega_P^2(\mathbf{r}')}P^{\alpha}_{Ks}(\mathbf{r})P^{\beta *}_{Ks}(\mathbf{r}')\right\}. \tag{74}$$

The closure relation (A12) then obviously leads to the correct ETCR (70) for the (nonretarded) polarization field.

Note that we can obtain the commutator (74) from Eq. (72) valid for the *retarded* field, if we put $(\mu=1,\nu=0)$ and take the same value for the retarded and nonretarded expansion parameters ζ Eq. (53), and η, Eq. (A15), respectively, which gives

$$N_K=(4\pi c)^2. \tag{75}$$

We can now take the Hamiltonian of the (free) electromagnetic field H_{em}, Eq. (58), and the Hamiltonian of the (nonretarded) polarization field H_{nr}, Eq. (A5), and put them together. The resulting Hamiltonian $H_{em}+H_{nr}$ of such *noninteracting* system can be put exactly in the same form as the (retarded) Hamiltonian H_f Eq. (22), of the *interacting* system, with the simple redefinition of the vector potential (12). It means that the interaction between the fields $\mathbf{A}_0$ and $\mathbf{P}$ enters into the Hamiltonian H_f implicitly through the equations of motion (1) and (2), and this interaction changes the eigenfrequencies of the whole system. In our approach we have expressed the basic fields $\mathbf{A}_0$ and $\mathbf{P}$ through the coupled (polariton) eigenmodes, and in principle one should find a new generalized field and conjugate momenta of *free polaritons* which would satisfy ETCR. It can be easily done in some special cases.

(a) *Longitudinal modes*. In a dielectric j, as discussed in Sec. III, the longitudinal polarization is described as a solution of $\epsilon_K^j=0$, which gives the same frequency ω_L^j, Eq. (31), for all longitudinal modes in that dielectric. The vector potential can be expressed in terms of polarization as

$$\dot{\mathbf{A}}_0(\mathbf{r},t)=\frac{4\pi c}{\epsilon_{\infty}^j}\mathbf{P}(\mathbf{r},t).$$

With this replacement and with $(\mu=1,\nu=0)$, the Lagrangian (17) in the dielectric j takes a simple form

$$L_l=\int_j d\mathbf{r}\frac{2\pi}{\omega_P^{j2}}(\dot{\mathbf{P}}^2-\omega_L^{j2}\mathbf{P}^2),$$

while the momentum (18) conjugate to $\mathbf{A}_0$ vanishes,

$$\mathbf{\Pi_P}=\frac{4\pi}{\omega_P^{j2}}\dot{\mathbf{P}},\quad \mathbf{\Pi_A}=\mathbf{0}. \tag{76}$$

ETCR for the polarization field can be derived by calculating the factor (42): $Z^L_{KK}=(\epsilon_{\infty}^j\omega_L^j/\omega_T^j)^2$. It is now a constant, so from Eq. (49) we can derive the closure relation valid in a dielectric j:

$$\frac{1}{V}\sum_l\frac{(4\pi c)^2}{N_l}\frac{1}{\omega_P^{j2}}P_l^{\alpha}(\mathbf{r})P_l^{\beta *}(\mathbf{r}')=\delta^L_{\alpha,\beta}(\mathbf{r}-\mathbf{r}'), \tag{77}$$

where index l denotes all longitudinal polarization eigenmodes and $\delta^L_{\alpha,\beta}(\mathbf{r}-\mathbf{r}')$ is the longitudinal δ function [17]. The comparison with Eq. (72) then shows that the polarization satisfies ETCR (70).

Note that Eq. (77) takes the nonretarded form (A12) for $N_l=(4\pi c)^2$, Eq. (75). In fact, the longitudinal polarization is not affected by the photon field so one expects that ETCR derived for the nonretardet limit (74), remain preserved.

(b) *Metallic modes*. The modes with the transverse frequency ω_T are the eigenmodes of the free polarization field. Let us assume that all the dielectrics are metals (or inert dielectrics) with $\omega_T=0$. Then the polarization has no restoring force and the dynamics of the system is determined by the photon field. In that case the polarization can be expressed in terms of the vector potential as

$$\dot{\mathbf{P}}(\mathbf{r},t)=-\frac{\omega_P^2(\mathbf{r})}{4\pi c}\mathbf{A}_0(\mathbf{r},t),$$

so with $(\nu=1,\mu=0)$, for the Lagrangian we find,

$$L_t=\int d\mathbf{r}\frac{1}{8\pi}\left\{\frac{1}{c^2}[\epsilon_\infty\dot{\mathbf{A}}_0^2-\omega_P^2\mathbf{A}_0^2]-(\boldsymbol{\nabla}\times\mathbf{A}_0)^2\right\},$$

while the momentum (19) conjugate to $\mathbf{P}$ vanishes,

$$\boldsymbol{\Pi}_\mathbf{A}=\frac{1}{4\pi c^2}\epsilon_\infty\dot{\mathbf{A}}_0,\quad \boldsymbol{\Pi}_\mathbf{P}=\mathbf{0}.$$

The factor $Z_{KK'}$, Eq. (42), becomes the same as in the case $\mathbf{P}=\mathbf{0}$, i.e., $Z_{KK'}=\epsilon_\infty(\mathbf{r})$ Eq. (59), so we can derive the closure relation in the form (61),

$$\frac{1}{V}\sum_{Ks}\frac{1}{N_K}\epsilon_\infty(\mathbf{r})A_{Ks}^\alpha(\mathbf{r})A_{Ks}^{\beta*}(\mathbf{r}')=\delta_{\alpha,\beta}\delta(\mathbf{r}-\mathbf{r}'),\quad (78)$$

where $\delta(\mathbf{r}-\mathbf{r}')$ is a complete δ function, i.e., it includes both the transverse and the longitudinal field. The closure relation (78) is the consequence of the hermiticity of the eigenvalue problem, because for $\omega_T=0$, the eigenvalue $\epsilon_K\omega_K^2$ in Eq. (30) takes a simple form $(\epsilon_\infty\omega_K^2-\omega_P^2)$ determined by only one eigenfrequency ω_K. Combining this result with Eq. (71) we can easily derive ETCR for the vector potential.

(c) *Homogeneous medium.* There are a lot of articles that discuss the problem of the fundamental commutation relations in the simple case of the homogeneous medium. In the standard approach the vector potential and the polarization are expanded in terms of their free eigenmodes (plane waves). The expansion coefficients are then chosen so that the corresponding fields satisfy ETCR (70). Since the eigenfrequencies of the free fields are not a solution of the coupled equations of motion (1) and (2), the Hamiltonian should be written in the form where the interaction between the fields $\mathbf{A}$ and $\mathbf{P}$ is given as an additional term. Then one introduces new operators that oscillate with the eigenfrequencies of the coupled system and from that requirement tries to determine the polariton eigenfrequencies.

In our approach we have briefly analyzed the homogeneous dielectric in Sec. III C and expanded $\mathbf{A}_0$ and $\mathbf{P}$ in terms of plane waves that oscillate with the polariton frequency ω_K.

Let us first analyze the transverse modes. With the $(\mu=1,\nu=0)$ choice and with the help of Eq. (69), we find the following for the commutation relation (71):

$$[A_0^{T\alpha}(\mathbf{r},t),\Pi_\mathbf{A}^{T\beta}(\mathbf{r}',t)]=i\hbar\,\mathrm{Re}\left\{\frac{1}{V}\sum_\mathbf{K}e^{i(\mathbf{r}-\mathbf{r}')\cdot\mathbf{K}}\sum_T e_\mathbf{K}^{T\alpha}e_\mathbf{K}^{T\beta}\sum_n\frac{v_{Kn}^g}{v_{Kn}^p}\right\}.\quad (79)$$

In the case $\omega_T>0$, one can derive the sum rule for the two transverse polariton branches [2,25],

$$\frac{v_{K+}^g}{v_{K+}^p}+\frac{v_{K-}^g}{v_{K-}^p}=1,\quad (80)$$

which transforms Eq. (79) into the following desired form:

$$[A_0^{T\alpha}(\mathbf{r},t),\Pi_\mathbf{A}^{T\beta}(\mathbf{r}',t)]=i\hbar\,\delta_{\alpha,\beta}^\perp(\mathbf{r}-\mathbf{r}').\quad (81)$$

Here

$$\delta_{\alpha,\beta}^\perp(\mathbf{r}-\mathbf{r}')=\frac{1}{V}\sum_\mathbf{K}\sum_T e^{i(\mathbf{r}-\mathbf{r}')\cdot\mathbf{K}}e_\mathbf{K}^{T\alpha}e_\mathbf{K}^{T\beta}$$

represents the transverse δ function $\delta_{\alpha,\beta}^T(\mathbf{r}-\mathbf{r}')$, Eq. (61), in the case of plane waves [17].

In the case $\omega_T=0$ there is only one branch so the sum–rule (80) as well as the commutation relation (81) do not hold. One can restore those equations by introducing an infinitesimally small transverse frequency [15] which would formally give again two transverse branches.

However, for the $\omega_T=0$ case it is more appropriate to take the $(\mu=0,\nu=1)$ choice because (as we have shown in Sec. V B) this immediately leads to the correct ETCR for both the transverse and the longitudinal field.

Let us analyze the longitudinal modes for $\omega_T>0$. In the $\omega_T>0$ case we have obtained correct ETCR for the transverse vector potential (81) using the $(\mu=1,\nu=0)$ choice, but it gives the zero momentum (76) of the longitudinal vector potential. Therefore we are left to calculate ETCR for the longitudinal *polarization* field $\mathbf{P}^L$. Following the results derived in Sec. V A we can take Eqs. (72) and (77) and immediately write

$$[P^{L\alpha}(\mathbf{r},t),\Pi_\mathbf{P}^{L\beta}(\mathbf{r}',t)]=i\hbar\,\delta_{\alpha,\beta}^\parallel(\mathbf{r}-\mathbf{r}'),$$

where

$$\delta_{\alpha,\beta}^\parallel(\mathbf{r}-\mathbf{r}')=\frac{1}{V}\sum_\mathbf{K}e^{i(\mathbf{r}-\mathbf{r}')\cdot\mathbf{K}}e_\mathbf{K}^{L\alpha}e_\mathbf{K}^{L\beta}$$

is the plane-wave representation of the longitudinal δ function $\delta_{\alpha,\beta}^L(\mathbf{r}-\mathbf{r}')$, Eq. (77).

Although in the case $\omega_T>0$ we were not able to express coupled fields by a single operator, we can describe the transverse field by the vector potential $\mathbf{A}_0^T$ and the longitudinal field by the polarization $\mathbf{P}^L$ so that both fields satisfy ETCR (70).

VI. CONCLUSION

In this paper we have developed a quantum-mechanical description of the electromagnetic field propagating in the polar dielectric. We have started with the macroscopic equations of motion for the photon and the matter field, determined by the vector potential $\mathbf{A}_0$ and the polarization $\mathbf{P}$, respectively, together with their coupling. These equations lead to the real dielectric function $\epsilon(\mathbf{r},\omega)$, with quite arbitrary space dependence, while the frequency dependence involves one pole with the well-defined transverse $[\epsilon(\mathbf{r},\omega_T)\rightarrow\infty]$ and longitudinal $[\epsilon(\mathbf{r},\omega_L)\rightarrow 0]$ frequencies. In quan-

tum optics the longitudinal modes were usually neglected, but we have needed them in order to make our retarded theory complete and easily comparable with the nonretarded limit.

All relevant operators are expanded from the beginning in terms of coupled (polariton) eigenmodes whose eigenfrequencies include the interaction between fields. By requiring the standard commutation relations between the creation and annihilation operators for the polaritons, the Hamiltonian of the system is diagonalized regardless of the specific shape of polar dielectrics. This was possible due to the derivation of the orthonormality relation in a general form. However, the corresponding closure relation could not be obtained. From the physical point of view, it becomes important when one tries to derive the equal-time commutation relations between fields ($\mathbf{A}_0$, $\mathbf{P}$) and corresponding conjugate momenta. They are obviously satisfied for the free fields, while in the coupled system one should in principle define a generalized polariton coordinate and the conjugate momentum, which would satisfy the required commutation relations. It could be easily done in some specific cases where one field can be obtained as a time derivative of the other. In our approach we made use of the unambiguity in the momentum definition, e.g., the momentum conjugate to $\mathbf{A}_0$ could be chosen proportional to $\mathbf{D}$ as well as proportional to $\mathbf{E}$. In that way we were able to resolve some difficulties which were present in theories that *a posteriori* analyze the validity of the fundamental commutations relations for the coupled fields.

In fact we were able to reproduce all present results in quantum optics of polar medium that were based on the real macroscopic dielectric function $\epsilon(\mathbf{r},\omega)$. Moreover, we gave a simple expressions for the vector potential and polarization in the system consisting of any number of polar dielectrics. That approach can be efficiently used in the calculation of the quantum optics phenomena in polar media as well as in the interaction of such system with external charges.

APPENDIX: NONRETARDED LIMIT

In many cases it is enough to determine the polarization eigenmodes in the nonretarded limit, e.g., *optical phonons* in ionic crystals or *plasmons* in metals. Formally, one can derive that limit by letting $c\to\infty$ in the retarded Maxwell equations (1). We introduce the (nonretarded) scalar potential Φ in a standard way:

$$\mathbf{E}(\mathbf{r},t)=-\boldsymbol{\nabla}\Phi(\mathbf{r},t). \tag{A1}$$

The gauge, analogous to Eq. (13), that will divide the total potential Φ into the part ϕ determined by the external charge density ρ, Eq. (14), and the part Φ_0 determined by the polarization $\mathbf{P}$, reads as

$$\boldsymbol{\nabla}\cdot[-\epsilon_\infty(\mathbf{r})\boldsymbol{\nabla}\Phi_0(\mathbf{r},t)+4\pi\mathbf{P}(\mathbf{r},t)]=0. \tag{A2}$$

In the absence of external charges we find $\Phi=\Phi_0$, so we can write the constitutive equation (2) as

$$\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2}+\omega_T^2(\mathbf{r})\mathbf{P}(\mathbf{r},t)=\frac{\omega_P^2(\mathbf{r})}{4\pi}[-\boldsymbol{\nabla}\Phi_0(\mathbf{r},t)]. \tag{A3}$$

The equations of motion (A2) and (A3) can be derived from the following nonretarded Lagrangian:

$$L_{nr}=\int d\mathbf{r}\left\{\frac{1}{8\pi}\epsilon_\infty(\boldsymbol{\nabla}\Phi_0)^2+\frac{2\pi}{\omega_P^2}(\dot{\mathbf{P}}^2-\omega_T^2\mathbf{P}^2)-\mathbf{P}\cdot\boldsymbol{\nabla}\Phi_0\right\}.$$

It gives for the generalized momenta,

$$\boldsymbol{\Pi}_\mathbf{P}=\frac{4\pi}{\omega_P^2}\dot{\mathbf{P}},\quad \Pi_\Phi=0, \tag{A4}$$

so the corresponding Hamiltonian is

$$H_{nr}=\int d\mathbf{r}\left\{-\frac{1}{8\pi}\epsilon_\infty(\boldsymbol{\nabla}\Phi_0)^2+\frac{2\pi}{\omega_P^2}(\dot{\mathbf{P}}^2+\omega_T^2\mathbf{P}^2)+\mathbf{P}\cdot\boldsymbol{\nabla}\Phi_0\right\}. \tag{A5}$$

From now on, we shall use the electric field (A1) rather than the scalar potential, so we shall put the Hamiltonian (A5) into the following familiar form:

$$H_{nr}=\int d\mathbf{r}\left\{\frac{2\pi}{\omega_P^2}(\dot{\mathbf{P}}^2+\omega_T^2\mathbf{P}^2)-\frac{1}{2}\mathbf{P}\cdot\mathbf{E}\right\}. \tag{A6}$$

The nonretarded Hamiltonian H_{nr} has been quantized, e.g., for thin dielectric films [26], and here we wish to perform the quantization for dielectrics of any shape, just as we have done in the retarded case. Following the same procedure, we shall first define the polarization eigenmodes $\mathbf{P}_{Ks}$ so as to satisfy Eqs. (A2) and (A3):

$$\boldsymbol{\nabla}\cdot(\epsilon_\infty\mathbf{E}_{Ks}+4\pi\mathbf{P}_{Ks})=0, \tag{A7}$$

$$\mathbf{E}_{Ks}=\chi_K^{-1}\mathbf{P}_{Ks}. \tag{A8}$$

Obviously, the indices (K,s) and the susceptibility χ_K are defined as in Sec. II.

In order to derive orthogonality relation, we now start with the transformation

$$\boldsymbol{\nabla}\cdot[(\epsilon_\infty\mathbf{E}_{Ks}+4\pi\mathbf{P}_{Ks})\Phi^{(*)}_{K's'}-(\epsilon_\infty\mathbf{E}_{K's'}+4\pi\mathbf{P}_{K's'})^{(*)}\Phi_{Ks}]$$
$$=4\pi(\mathbf{P}^{(*)}_{K's'}\cdot\mathbf{E}_{Ks}-\mathbf{P}_{Ks}\cdot\mathbf{E}^{(*)}_{K's'}), \tag{A9}$$

which is valid with and without the complex conjugation $(*)$. The integration of Eq. (A9) over $\mathbf{r}$, with the help of Eq. (A8), gives

$$\int d\mathbf{r}[\chi_K^{-1}(\mathbf{r})-\chi_{K'}^{-1}(\mathbf{r})]\mathbf{P}^{(*)}_{K's'}(\mathbf{r})\cdot\mathbf{P}_{Ks}(\mathbf{r})=0$$

which, using the definition for χ_K, Eq. (4), leads to the general orthogonality relation

$$\int d\mathbf{r}\frac{1}{\omega_P^2(\mathbf{r})}\mathbf{P}^{(*)}_{K's'}(\mathbf{r})\cdot\mathbf{P}_{Ks}(\mathbf{r})=0,\quad K'\neq K. \tag{A10}$$

If we choose the degenerate polarization modes ($K=K'$) as orthogonal, we can write the orthonormality requirement as

$$\frac{1}{V}\int d\mathbf{r}\frac{1}{\omega_P^2(\mathbf{r})}\mathbf{P}_{Ks}^*(\mathbf{r})\cdot\mathbf{P}_{K's'}(\mathbf{r})=\delta_{K,K'}\delta_{s,s'}. \quad \text{(A11)}$$

The closure relation becomes

$$\frac{1}{V}\sum_{Ks}\frac{1}{\omega_P^2(\mathbf{r})}P_{Ks}^{\alpha}(\mathbf{r})P_{Ks}^{\beta *}(\mathbf{r}')=\delta_{\alpha,\beta}\delta(\mathbf{r}-\mathbf{r}'). \quad \text{(A12)}$$

In a common case in which the *dielectric j* is *homogeneous*, Eqs. (A7) and (A8) lead to the longitudinal and transverse modes inside this dielectric, simply determined by the dielectric function ϵ_K^j, Eq. (5). The longitudinal modes ($\epsilon_K^j=0$) have the same frequency $\omega_K=\omega_L^j$ just as in the retarded case, Eq. (31). The transverse modes ($\epsilon_K^j\neq 0$) are divided into the surface modes ($\nabla\cdot\mathbf{E}_{Ks}=0$), whose frequencies generally depend upon the boundaries among dielectrics, and the modes with the frequency $\omega_K=\omega_T^j$, which vanish outside the dielectric *j*. The modes with the frequency ω_T^j have zero electric field ($\mathbf{E}^{Tj}=\mathbf{0}$) and they are obviously the eigenmodes of the free polarization. Note that they were not the proper eigenmodes in the retarded case because of the coupling of transverse polarization with photons.

In order to quantize the Hamiltonian (A6) we shall expand $\mathbf{P}$ and $\mathbf{E}$ in terms of their eigenmodes:

$$\mathbf{P}(\mathbf{r},t)=\frac{1}{\sqrt{V}}\sum_{Ks}[q_{Ks}(t)\mathbf{P}_{Ks}(\mathbf{r})+q_{Ks}^{\dagger}(t)\mathbf{P}_{Ks}^*(\mathbf{r})], \quad \text{(A13)}$$

$$\mathbf{E}(\mathbf{r},t)=\frac{1}{\sqrt{V}}\sum_{Ks}[g_{Ks}(t)\mathbf{E}_{Ks}(\mathbf{r})+q_{Ks}^{\dagger}(t)\mathbf{E}_{Ks}^*(\mathbf{r})],$$

$$q_{Ks}(t)=q_{Ks}\exp(-i\omega_K t).$$

When inserting these expansions into the Hamiltonian (A6), we shall first write the interaction term in the symmetric form, $\mathbf{P}\cdot\mathbf{E}=(\mathbf{P}\cdot\mathbf{E}+\mathbf{E}\cdot\mathbf{P})/2$, and then use Eq. (A8) to replace $\mathbf{E}_{Ks}$ by $\mathbf{P}_{Ks}$. This gives

$$H_{nr}=\frac{1}{V}\int d\mathbf{r}\frac{2\pi}{\omega_P^2}\sum_{Ks}\sum_{K's'}[X_{KsK's'}^{-}(\mathbf{r},t)Y_{KK'}^{-}(\mathbf{r}) + X_{KsK's'}^{+}(\mathbf{r},t)Y_{KK'}^{+}(\mathbf{r})], \quad \text{(A14)}$$

where we have introduced the following abbreviations:

$$X_{KsK's'}^{-}=q_{Ks}q_{K's'}\mathbf{P}_{Ks}\cdot\mathbf{P}_{K's'}+q_{Ks}^{\dagger}q_{K's'}^{\dagger}\mathbf{P}_{Ks}^*\cdot\mathbf{P}_{K's'}^*,$$

$$X_{KsK's'}^{+}=q_{Ks}q_{K's'}^{\dagger}\mathbf{P}_{Ks}\cdot\mathbf{P}_{K's'}^*+q_{Ks}^{\dagger}q_{K's'}\mathbf{P}_{Ks}^*\cdot\mathbf{P}_{K's'},$$

$$Y_{KK'}^{\mp}=\mp\omega_K\omega_{K'}+\omega_T^2-\frac{1}{2}\frac{\omega_P^2}{4\pi}(\chi_K^{-1}+\chi_{K'}^{-1}).$$

After inserting the expressions for the susceptibility χ_K, Eq. (4), we find that $Y_{KK'}^{\mp}$ does not depend upon $\mathbf{r}$:

$$Y_{KK'}^{\mp}=\frac{1}{2}(\omega_K\mp\omega_{K'})^2.$$

Therefore the integration over $\mathbf{r}$ in Eq. (A14) includes only $X_{KsK's'}$ terms with the weighting factor ω_P^{-2}. From the orthogonality relation (A10) we immediately find that only $K'=K$ terms contribute to the Hamiltonian. Since $Y_{KK}^{-}=0$, the orthonormality relation (A11) leads us to

$$H_{nr}=\sum_{Ks}4\pi\omega_K^2(q_{Ks}q_{Ks}^{\dagger}+q_{Ks}^{\dagger}q_{Ks}).$$

As a final step, we introduce the following annihilation a_{Ks} and the creation $a_{Ks}^{\dagger}$ operators:

$$q_{Ks}=\eta_K a_{Ks}, \quad q_{Ks}^{\dagger}=\eta_K^* a_{Ks}^{\dagger},$$

which obey the standard boson commutation relations (52). With the choice

$$\eta_K=\frac{1}{4\pi}\sqrt{\frac{h}{\omega_K}} \quad \text{(A15)}$$

we arrive at the following nonretarded Hamiltonian (A6) in the second-quantized form:

$$H_{nr}=\sum_{Ks}\hbar\omega_K\left(a_{Ks}^{\dagger}a_{Ks}+\frac{1}{2}\right). \quad \text{(A16)}$$

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