Quantizing polaritons in inhomogeneous dissipative systems

Aurélien Drezet
Université Grenoble Alpes, CNRS, Institut Néel, F-38000 Grenoble, France
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In this article we provide a general analysis of canonical quantization for polaritons in dispersive and dissipative electromagnetic inhomogeneous media. We compare several approaches based either on the Huttner-Barnett model [B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992)] or the Green function, Langevin-noise method [T. Gruner and D.-G. Welsch, Phys. Rev. A **53**, 1818 (1996)] which includes only material oscillators as fundamental variables. We show that in order to preserve unitarity, causality, and time symmetry, one must necessarily include with an equal footing both electromagnetic modes and material fluctuations in the evolution equations. This becomes particularly relevant for all nanophotonics and plasmonics problems involving spatially localized antennas or devices.

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I. INTRODUCTION

Tremendous progress has been realized in the last decades concerning the theoretical foundation of quantum optics in dielectric media. While the historical approach proposed by Jauch and Watson [1] was already based on the standard canonical quantization formalism for fields, it neglected dispersion and dissipation which are intrinsic properties of any causal dielectric media satisfying Kramers-Krönig relations. Since then, several important studies were devoted to the extension of the method to inhomogeneous and artificially structured media which are central issues in modern microphotonics and nanophotonics [2–4]. Furthermore, theoretical approaches adapted to transparent but dispersive media with negligible losses have been also developed based on different techniques such as the slowly varying envelope approximation [5] or the quasimodal expansion method which is valid near resonance for polaritons [6–9]. More recently, losses were included in the theory by adding phenomenologically some optical dissipation channels in the light propagation path [10,11]. Such a method was successfully used for the modeling of Casimir forces in dissipative media [12] and surface plasmon polaritons [13–15].

Moreover, the most fundamental progress was probably done when Huttner and Barnett, and others [16–22], proposed a self-consistent canonical quantization procedure for a homogeneous and causal dielectric medium by coupling photonic degrees of freedom with mechanical oscillator variables acting as thermal baths. The method, based on the pioneer works by Fano and Hopfield [23,24] (see also Ref. [25]), was subsequently extended to several inhomogeneous systems including anisotropic and magnetic properties [26–32]. In parallel to these theoretical works based on the standard canonical quantization method, a different and powerful axis of research appeared after the work by Gruner and Welsch [33,34] (see also Ref. [35]) based on the quantum Langevin-noise approach used in cavity QED (i.e., quantum electrodynamics) [36]. The method is also known as the Green tensor method [34] since it relies on efficient Green dyadic techniques used nowadays in nanophotonics and plasmonics [37,38]. This “Langevin-noise” approach, which actually extends earlier “semiclassical” researches based on the fluctuation-dissipation theorem by Lifshitz and many others in the context of Casimir and optical forces [39–50], was successfully applied in the recent years to many issues concerning photonics [51–60] and nanoplasmonics where dissipation can not be neglected [61–69]. In this context, the relationship between the Huttner-Barnett approach on the one side and the Langevin-noise method on the other side has attracted much attention in the last years, and several works attempted to demonstrate the validity of the Langevin-noise method from a rigorous Hamiltonian perspective which is more in a agreement with the canonical Huttner-Barnett approach [26–32,44].

The aim of this work is to revisit these derivations of the equivalence between the Langevin-noise and Hamiltonian methods and to show that some unphysical assumptions actually limit the domain of validity of the previous attempts. More precisely, as we will show in this work, the analysis and derivations always included some hypothesis concerning causality and boundary conditions which actually lead to circularity in the deductions and are not applicable to the most general inhomogeneous systems used in nano-optics. Specifically, these derivations, like the fluctuation-dissipation reasoning in Lifshitz and Rytov works [39–41,43], give too much emphasis on the material origin of quantum fluctuations for explaining macroscopic quantum electrodynamics in continuous media. However, as it was already pointed out in the 1970s [43,70–73], one must include with an equal footing both field and matter fluctuations in a self-consistent QED Hamiltonian in order to preserve rigorously unitarity and causality [43,74]. While this does not impact too much the homogeneous medium case considered by Huttner and Barnett [17], it is crucial to analyze further the inhomogeneous medium problem in order to give a rigorous foundation to the Gruner and Welsch theory [34] based on fluctuating currents. This is the central issue tackled in this work.

The layout of this paper is as follows: In Sec. II, we review the Lagrangian method developed in our previous work [75] based on an alternative dual formalism for describing the Huttner-Barnett model. In this section, we summarize the essential elements of the general Lagrangian and Hamiltonian models necessary for this study. In particular, we present the fundamental issue about the correct definition of Hamiltonian which will be discussed at length in this article. In Sec. III, we provide a quantitative discussion of the Huttner-Barnett model for a homogeneous dielectric medium. We discuss a modal expansion into plane waves and separate explicitly the electromagnetic field into classical eigenmodes and noise-related Langevin modes. We show that both contributions are necessary for preserving unitarity and time symmetry. We consider limit cases such as the ideal Hopfield-Fano polaritons [23,24] without dissipation and the weakly dissipative polariton modes considered by Milonni and others [6–9]. We discuss the physical interpretation of the Hamiltonian of the whole system and interpret the various contributions with respect to the Langevin-noise method and to the lossless Hopfield-Fano limit. In Sec. IV, we generalize our analysis to the inhomogeneous medium case by using a Green dyadic formalism in both the frequency and time domains. We demonstrate that in general it is necessary to keep both pure photonic and material fluctuations to preserve the unitarity and time symmetry of the quantum evolution. We conclude with a discussion about the physical meaning of the Hamiltonian in presence of inhomogeneities and interpret the various terms associated with photonic and material modes.

II. HUTTNER-BARNETT MODEL AND THE DUAL LAGRANGIAN FORMALISM

In Ref. [75] we developed a Lagrangian formalism adapted to QED in dielectric media without magnetic properties. Here, we will use this model to derive our approach, but a standard treatment based on the minimal coupling scheme [76] or the Power-Zienau [77] transformation would lead to similar results. We start with the dual Lagrangian density:

$$\mathcal{L} = \frac{\mathbf{B}^2 - \mathbf{D}^2}{2} + \mathbf{F}\cdot\nabla\times\mathbf{P} - \frac{\mathbf{P}^2}{2} + \mathcal{L}_M, \tag{2.1}$$

where $\mathbf{B}(\mathbf{x},t)$ and $\mathbf{D}(\mathbf{x},t)$ are the magnetic and displacement fields, respectively. In this formalism, the usual magnetic potential $\mathbf{A}$, defined such as $\mathbf{B} = \nabla\times\mathbf{A}$, is replaced by the dual electric potential $\mathbf{F}$ [in the "Coulomb" gauge $\nabla\cdot\mathbf{F}(\mathbf{x},t) = 0$] defined by

$$\mathbf{B}(\mathbf{x},t) = \frac{1}{c}\partial_t\mathbf{F}(\mathbf{x},t), \quad \mathbf{D}(\mathbf{x},t) = \nabla\times\mathbf{F}(\mathbf{x},t), \tag{2.2}$$

implying

$$\nabla\times\mathbf{B}(\mathbf{x},t) = \frac{1}{c}\partial_t\mathbf{D}(\mathbf{x},t),\ \nabla\cdot\mathbf{D}(\mathbf{x},t) = 0. \tag{2.3}$$

The material part $\mathcal{L}_M$ of the Lagrangian density in Eq. (2.1) reads as

$$\mathcal{L}_M = \int_0^{+\infty} d\omega \frac{(\partial_t\mathbf{X}_\omega)^2 - \omega^2\mathbf{X}_\omega^2}{2} \tag{2.4}$$

with $\mathbf{X}_\omega(\mathbf{x},t)$ the material oscillator fields describing the Huttner-Barnett bath coupled to the electromagnetic field. The coupling depends on the polarization density which is defined by

$$\mathbf{P}(\mathbf{x},t) = \int_0^{+\infty} d\omega\sqrt{\frac{2\sigma_\omega(\mathbf{x})}{\pi}}\mathbf{X}_\omega(\mathbf{x},t), \tag{2.5}$$

where the coupling function $\sigma_\omega(\mathbf{x}) \geqslant 0$ defines the conductivity of the medium at the harmonic pulsation ω. From Eq. (2.1) and Euler-Lagrange equations we deduce the dynamical laws for the electromagnetic field

$$\nabla\times\mathbf{E}(\mathbf{x},t) = -\frac{1}{c}\partial_t\mathbf{B}(\mathbf{x},t),\ \nabla\cdot\mathbf{B}(\mathbf{x},t) = 0 \tag{2.6}$$

with the electric field $\mathbf{E}(\mathbf{x},t) = \mathbf{D}(\mathbf{x},t) - \mathbf{P}(\mathbf{x},t)$. Similarly for the material oscillators we have

$$\partial_t^2\mathbf{X}_\omega(\mathbf{x},t) + \omega^2\mathbf{X}_\omega(\mathbf{x},t) = \sqrt{\frac{2\sigma_\omega(\mathbf{x})}{\pi}}\mathbf{E}(\mathbf{x},t). \tag{2.7}$$

We point out that the Lagrangian density in Eq. (2.1) includes a term $-\frac{\mathbf{P}^2}{2}$ which is necessary for the derivation of the dynamical laws for the material fields $\mathbf{X}_\omega$ [75]. Furthermore, to complete the QED canonical quantization procedure of the material field we introduce the lowering $\mathbf{f}_\omega(\mathbf{x},t)$ and rising $\mathbf{f}_\omega^\dagger(\mathbf{x},t)$ operators for the bosonic material field from the relation $\mathbf{f}_\omega(\mathbf{x},t) = \frac{i\partial_t\mathbf{X}_\omega(\mathbf{x},t)+\omega\mathbf{X}_\omega(\mathbf{x},t)}{\sqrt{2\hbar\omega}}$. As explained in Ref. [75] by using the equal-time commutation relations between the canonical variables $\mathbf{X}_\omega(\mathbf{x},t)$ and $\partial_t\mathbf{X}_\omega(\mathbf{x},t)$, we deduce the fundamental rules

$$[\mathbf{f}_\omega(\mathbf{x},t),\mathbf{f}_{\omega'}^\dagger(\mathbf{x}',t)] = \delta(\omega-\omega')\delta^3(\mathbf{x}-\mathbf{x}')\mathbf{I} \tag{2.8}$$

(with $\mathbf{I} = \hat{\mathbf{x}}\otimes\hat{\mathbf{x}} + \hat{\mathbf{y}}\otimes\hat{\mathbf{y}} + \hat{\mathbf{z}}\otimes\hat{\mathbf{z}}$ the unit dyad) and $[\mathbf{f}_\omega(\mathbf{x},t),\mathbf{f}_{\omega'}(\mathbf{x}',t)] = [\mathbf{f}_\omega^\dagger(\mathbf{x},t),\mathbf{f}_{\omega'}^\dagger(\mathbf{x}',t)] = 0$ allowing a clear interpretation of $\mathbf{f}_\omega(\mathbf{x},t)$ and $\mathbf{f}_\omega^\dagger(\mathbf{x},t)$ as lowering and rising operators for the bosonic states associated with the matter oscillators.

Moreover, Eqs. (2.5) and (2.7) can be formally integrated, leading to

$$\mathbf{P}(\mathbf{x},t) = \mathbf{P}^{(0)}(\mathbf{x},t) + \int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}(\mathbf{x},t-\tau), \tag{2.9}$$

where t_0 is an initial time and where $\mathbf{P}^{(0)}(\mathbf{x},t)$ is a fluctuating dipole density distribution defined by

$$\begin{aligned}\mathbf{P}^{(0)}(\mathbf{x},t) &= \int_0^{+\infty} d\omega\sqrt{\frac{2\sigma_\omega(\mathbf{x})}{\pi}}\mathbf{X}_\omega^{(0)}(\mathbf{x},t)\\ &= \int_0^{+\infty} d\omega\sqrt{\frac{\hbar\sigma_\omega(\mathbf{x})}{\pi\omega}}\left[\mathbf{f}_\omega^{(0)}(\mathbf{x},t) + \mathbf{f}_\omega^{\dagger(0)}(\mathbf{x},t)\right]\end{aligned} \tag{2.10}$$

with $\mathbf{X}_\omega^{(0)}(\mathbf{x},t) = \cos[\omega(t-t_0)]\mathbf{X}_\omega(\mathbf{x},t_0) + \sin[\omega(t-t_0)]\partial_t\mathbf{X}_\omega(\mathbf{x},t_0)/\omega$ and where by definition $\mathbf{f}_\omega^{(0)}(\mathbf{x},t) = \mathbf{f}_\omega(\mathbf{x},t_0)e^{-i\omega(t-t_0)}$. We therefore have

$$\begin{aligned}\mathbf{D}(\mathbf{x},t) &= \mathbf{E}(\mathbf{x},t) + \mathbf{P}(\mathbf{x},t)\\ &= \mathbf{P}^{(0)}(\mathbf{x},t) + \mathbf{E}(\mathbf{x},t) + \int_0^{t-t_0} d\tau\,\chi(\mathbf{x},\tau)\mathbf{E}(\mathbf{x},t-\tau),\end{aligned} \tag{2.11}$$

which is reminiscent of the general linear response theory used in thermodynamics [78]. We point out that the term $\int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}(\mathbf{x},t-\tau)$ can be seen as an induced dipole density. However, as we will show in the next section, the electric field itself is decomposed into a purely fluctuating term $\mathbf{E}^{(0)}(\mathbf{x},t)$ and a scattered field $\mathbf{E}^{(s)}(\mathbf{x},t)$ which depends on the density $\mathbf{P}^{(0)}$. Therefore, the contribution $\int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}(\mathbf{x},t-\tau)$ to $\mathbf{P}$ is also decomposed into a pure photon-fluctuation term $\int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}^{(0)}(\mathbf{x},t-\tau)$ and an induced term $\int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}^{(s)}(\mathbf{x},t-\tau)$ related to material fluctuations $\mathbf{P}^{(0)}$.

Importantly, the linear susceptibility $\chi(\mathbf{x},\tau)$ which is defined by

$$\chi(\mathbf{x},\tau)=\int_0^{+\infty}d\omega\frac{2\sigma_\omega(\mathbf{x})}{\pi}\frac{\sin\omega\tau}{\omega}\Theta(\tau) \quad (2.12)$$

characterizes completely the dispersive and dissipative dielectric medium. We can show that the permittivity $\widetilde{\varepsilon}(\mathbf{x},\omega)=1+\int_0^{+\infty}d\tau\,\chi(\mathbf{x},\tau)e^{i\omega\tau}$ is an analytical function in the upper part of the complex plane $\omega=\omega'+i\omega''$, i.e., $\omega''>0$, provided $\chi(\mathbf{x},\tau)$ is finite for any time $\tau\geqslant 0$. From this we deduce the symmetry $\widetilde{\varepsilon}(\mathbf{x},-\omega)^*=\widetilde{\varepsilon}(\mathbf{x},\omega^*)$ and it is possible to derive the general Kramers-Krönig relations existing between the real part $\mathrm{Re}[\widetilde{\varepsilon}(\mathbf{x},\omega)]\equiv\widetilde{\varepsilon}'(\mathbf{x},\omega)$ and the imaginary part $\mathrm{Im}[\widetilde{\varepsilon}(\mathbf{x},\omega)]\equiv\widetilde{\varepsilon}''(\mathbf{x},\omega)$ of the dielectric permittivity. Therefore, the Huttner-Barnett model characterized by the conductivity $\sigma_\omega(\mathbf{x})$ is fully causal and can be applied to describe any inhomogeneous dielectric media in the linear regime.

The central issue of this paper concerns the definition of the Hamiltonian $H(t)$ in the Huttner-Barnett model. We remind that in Ref. [75] we derived the result

$$H(t)=\int d^3\mathbf{x}\frac{:\mathbf{B}^2+\mathbf{E}^2:}{2}+H_M \quad (2.13)$$

with $H_M(t)=\int d^3\mathbf{x}\int_0^{+\infty}d\omega\frac{:(\partial_t\mathbf{X}_\omega)^2+\omega^2\mathbf{X}_\omega^2:}{2}$ where $:[\ldots]:$ means, as usually, a normally ordered product for removing the infinite zero-point energy. Inserting the definition for $\mathbf{f}_\omega(\mathbf{x},t)$ obtained earlier we get for the material part

$$H_M(t)=\int d^3\mathbf{x}\int_0^{+\infty}d\omega\hbar\omega\mathbf{f}_\omega^\dagger(\mathbf{x},t)\mathbf{f}_\omega(\mathbf{x},t), \quad (2.14)$$

which has the standard structure for oscillators (i.e., without the infinite zero-point energy).

However, Huttner and Barnett [17] after diagonalizing their Hamiltonian found that the total evolution is described in the homogeneous medium case by

$$H_M^{(0)}(t)=\int d^3\mathbf{x}\int_0^{+\infty}d\omega\hbar\omega\mathbf{f}_\omega^{\dagger(0)}(\mathbf{x},t)\mathbf{f}_\omega^{(0)}(\mathbf{x},t). \quad (2.15)$$

While as we will see this is actually a correct description "for all practical purposes" in a homogeneous dissipative medium for large class of physical boundary conditions, this is in general not acceptable in order to preserve time symmetry and unitarity in the full Hilbert space for interacting matter and light. The general method based on Langevin forces and noises avoided quite generally mentioning that difficult point. We emphasize that while the conclusions presented in Refs. [33–35] are accepted by more or less all authors on the subject [51–59,61–69], there have been some few dissident views (see Refs. [79,80]) claiming that in the context of an input-output formalism, the Langevin-noise formalism is not complete unless we consider as well fluctuations of the free-photon modes (see also the replies with an opposite perspective in Refs. [81,82]). In this work, we will generalize and give a rigorous QED-like Hamiltonian foundation to the prescriptions of Refs. [79,80] and we will show that it is actually necessary to include a full description of photonic and material quantum excitations in order to preserve unitarity. In order to appreciate this fact further, we will first consider the problem associated with quantization of the electromagnetic field.

III. QUANTIZATION OF ELECTROMAGNETIC WAVES IN A HOMOGENEOUS DIELECTRIC MEDIUM

A. General modal expansion

We first introduce the paradigmatic homogeneous medium case considered initially by Huttner and Barnett [17], i.e., with $\chi(\mathbf{x},\tau)=\chi(\tau)$. We start with Faraday's law $\nabla\times\mathbf{E}(\mathbf{x},t)=-\frac{1}{c}\partial_t\mathbf{B}(\mathbf{x},t)$ rewritten according to Eq. (2.11) as

$$\nabla\times\mathbf{D}(\mathbf{x},t)=-\frac{1}{c}\partial_t\mathbf{B}(\mathbf{x},t)+\nabla\times\mathbf{P}^{(0)}(\mathbf{x},t)-\frac{1}{c}\int_0^{t-t_0}d\tau\,\chi(\tau)\partial_{t-\tau}\mathbf{B}(\mathbf{x},t-\tau). \quad (3.1)$$

Inserting Eq. (2.3) and using the Coulomb (transverse) gauge condition we get

$$\frac{1}{c^2}\partial_t^2\mathbf{F}(\mathbf{x},t)-\nabla^2\mathbf{F}(\mathbf{x},t)-\nabla\times\mathbf{P}^{(0)}(\mathbf{x},t)+\frac{1}{c^2}\int_0^{t-t_0}d\tau\,\chi(\tau)\partial_{t-\tau}^2\mathbf{F}(\mathbf{x},t-\tau)=0. \quad (3.2)$$

We use the modal expansion method developed in Ref. [75] and write

$$\mathbf{F}(\mathbf{x},t)=\sum_{\alpha,j}q_{\alpha,j}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) \quad (3.3)$$

with α a generic label for the wave vector $\mathbf{k}_\alpha$, $\Phi_\alpha(\mathbf{x})=e^{i\mathbf{k}_\alpha\cdot\mathbf{x}}/\sqrt{V}$ (here we consider as it is usually done the periodical "box" Born–von Karman expansion in the rectangular box of volume V), $j=1$ or 2, labels the two transverse polarization states with unit vectors $\hat{\boldsymbol{\epsilon}}_{\alpha,1}=\mathbf{k}_\alpha\times\hat{\mathbf{z}}/|\mathbf{k}_\alpha\times\hat{\mathbf{z}}|$ and $\hat{\boldsymbol{\epsilon}}_{\alpha,2}=\hat{\mathbf{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,1}$ (conventions and details are given in Appendix A of Ref. [75]). Inserting Eq. (3.3) into Eq. (3.2), we obtain the dynamical equation

$$\ddot{q}_{\alpha,j}(t)+\int_0^{t-t_0}d\tau\,\chi(\tau)\ddot{q}_{\alpha,j}(t-\tau)+\omega_\alpha^2q_{\alpha,j}(t)=S_{\alpha,j}^{(0)}(t) \quad (3.4)$$

with the time-dependent source term

$$S_{\alpha,j}^{(0)}(t)=c^2\int d^3\mathbf{x}\,\nabla\times\mathbf{P}^{(0)}(\mathbf{x},t)\cdot\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha^*(\mathbf{x}). \quad (3.5)$$

To solve this equation, we use the Laplace transform of the fields which is defined below.

We are interested in the evolution for $t\geqslant t_0$ of a field $A(t)$ for which Fourier transform is not necessarily well mathematically defined since the field is not going to zero fast enough for $t\to+\infty$ (e.g., a fluctuating current or field). The method followed here is to consider the forward Laplace transform of the different evolution equations (such an approach was also used by Suttorp by mixing both forward and backward Laplace transforms [27]). To deal with this problem, we first change the time t variable in $t'=t-t_0$ and define $A'(t')=A(t)$. We define the (forward) Laplace transform of

$A'(t')$ as

$$\overline{A'}(p)=\int_0^{+\infty}dt'e^{-pt'}A'(t')=\int_{t_0}^{+\infty}dt\,e^{-p(t-t_0)}A(t) \quad (3.6)$$

with $p=\gamma-i\omega$ (ω a real number and $\gamma\geqslant 0$). The presence of the term $e^{-\gamma t'}$ ensures the convergence. We will not here introduce the backward Laplace transform $\int_{-\infty}^{t_0}dt\,e^{+p(t-t_0)}A(t)=\int_{-t_0}^{+\infty}du\,e^{-p(u+t_0)}A(-u)$ since the time t_0 is arbitrary and can be sent into the remote past if needed.

As it is well known, the (forward) Laplace transform is connected to the usual Fourier transform since we have

$$\overline{A'}(\gamma-i\omega)=2\pi\underline{\widetilde{A}}(\omega)e^{-i\omega t_0}, \quad (3.7)$$

where $\underline{\widetilde{A}}(\omega)=\int_{-\infty}^{+\infty}\frac{dt}{2\pi}\underline{A}(t)e^{i\omega t}$ is the Fourier transform of $\underline{A}(t)=A(t)\Theta(t-t_0)e^{-\gamma(t-t_0)}$ with respect to t.

Now, for the specific problem considered here we obtain the separation $q_{\alpha,j}(t)=q^{(s)}_{\alpha,j}(t)+q^{(0)}_{\alpha,j}(t)$. The (0) contribution corresponds to what classically we call a sum of eigenmodes supported by the medium (i.e., with $\mathbf{P}^{(0)}=0$) while the (s) term is the fluctuating field generated by the Langevin source $\mathbf{P}^{(0)}(t)$. More explicitly, we have for the source term

$$q^{(s)}_{\alpha,j}(t)=\int_0^{t-t_0}d\tau\,H_\alpha(\tau)S^{(0)}_{\alpha,j}(t-\tau), \quad (3.8)$$

where we use Eq. (3.5). The propagator function $H_\alpha(\tau)$ is expressed as a Bromwich contour:

$$H_\alpha(\tau)=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\frac{e^{p\tau}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2}, \quad (3.9)$$

where $\bar{\chi}(p)$ is defined as $\overline{\chi}(\mathbf{x},p)=\int_0^{+\infty}d\tau\,e^{-p\tau}\chi(\mathbf{x},\tau)$. We remind that a Bromwich integral by definition will vanish for $\tau<0$ so that on the left side we actually mean $\theta(\tau)H_\alpha(\tau)$.

The function $H_\alpha(\tau)$ has some remarkable properties which should be emphasized here. We first introduce the "zeros" of $\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2$, i.e., the set of roots $\Omega^{(\pm)}_{\alpha,m}$ solutions of $\Omega^{(\pm)}_{\alpha,m}\sqrt{\tilde{\varepsilon}(\Omega^{(\pm)}_{\alpha,m})}\pm\omega_\alpha=0$. From the causal properties of $\tilde{\varepsilon}$ we have $\tilde{\varepsilon}(-\omega^*)=\tilde{\varepsilon}(\omega)^*$ and therefore we deduce $\Omega^{(\pm)\,*}_{\alpha,m}=-\Omega^{(\mp)}_{\alpha,m}$ implying that the "+" and "−" roots are not independent. The important fact is that the roots are located in the lower complex plane associated with a negative imaginary part of the frequency [17] (this is proven in Appendix A). Now, as shown in Appendix B the integral in Eq. (3.9) can be computed by contour integration in the complex plane after closing the contour with a semicircle in the lower plane and using the Cauchy residue theorem. We get

$$H_\alpha(\tau)=\sum_m\frac{-1}{2i\omega_\alpha}\frac{e^{-i\Omega^{(-)}_{\alpha,m}\tau}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}+\text{c.c.} \quad (3.10)$$

We also get $H_\alpha(\tau)=0$ for $\tau\leqslant 0$ (after integration in the upper plane and considering in detail the case $\tau=0$). Clearly, the function $H_\alpha(\tau)$ is here expanded into a sum of modes which define the polaritons of the problem (this is shown explicitly in the next subsection). Here, the normal-mode frequency $\Omega^{(-)}_{\alpha,m}$ consists of complex numbers ensuring the damped nature of the waves in the future direction. We emphasize that in our knowledge this kind of formula has never been discussed before. The expansion in Eq. (3.10) is, however, rigorous and generalizes the quasimodal approximations used in the weak dissipation regime and discussed in Sec. III F.

Similarly, the source-free term $q^{(0)}_{\alpha,j}(t)$ reads as

$$q^{(0)}_{\alpha,j}(t)=U_\alpha(t-t_0)\dot{q}_{\alpha,j}(t_0)+\dot{U}_\alpha(t-t_0)q_{\alpha,j}(t_0) \quad (3.11)$$

with the new propagator function

$$U_\alpha(\tau)=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\frac{[1+\bar{\chi}(p)]e^{p\tau}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2}=\sum_m\frac{-\tilde{\varepsilon}(\Omega^{(-)}_{\alpha,m})}{2i\omega_\alpha}\frac{e^{-i\Omega^{(-)}_{\alpha,m}\tau}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}+\text{c.c.} \quad (3.12)$$

Like for H_α we get $U_\alpha(\tau)=0$ for $\tau\leqslant 0$. Additionally, the boundary condition at $t=t_0$ [i.e., $q^{(0)}_{\alpha,j}(t_0)=q_{\alpha,j}(t_0)$] imposes $\frac{d}{d\tau}U_\alpha(\tau)|_{\tau=0}=1$ (see Appendix B).

B. Classical eigenmodes

The electromagnetic field can be calculated using the expansion (3.8) or (3.11). First, the mathematical and physical structure of the free field is seen by using the modal expansion

$$\begin{aligned}\mathbf{F}^{(0)}(\mathbf{x},t)&=\sum_{\alpha,j}[U_\alpha(t-t_0)\dot{q}_{\alpha,j}(t_0)+\dot{U}_\alpha(t-t_0)q_{\alpha,j}(t_0)]\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})\\&=\sum_{\alpha,j,m}\frac{-\tilde{\varepsilon}(\Omega^{(-)}_{\alpha,m})}{2i\omega_\alpha}\frac{e^{-i\Omega^{(-)}_{\alpha,m}(t-t_0)}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}[\dot{q}_{\alpha,j}(t_0)-i\Omega^{(-)}_{\alpha,m}q_{\alpha,j}(t_0)]\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})+\text{H.c.},\end{aligned} \quad (3.13)$$

where we used the symmetries of the modal expansion [83] together with $\Omega^{(-)}_{\alpha,m}=\Omega^{(-)}_{-\alpha,m}$. From this we directly obtain

$$\begin{aligned}\mathbf{D}^{(0)}(\mathbf{x},t)&=i\sum_{\alpha,j}\frac{\omega_\alpha}{c}[U_\alpha(t-t_0)\dot{q}_{\alpha,j}(t_0)+\dot{U}_\alpha(t-t_0)q_{\alpha,j}(t_0)]\hat{\boldsymbol{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})\\&=\sum_{\alpha,j,m}\frac{-\tilde{\varepsilon}(\Omega^{(-)}_{\alpha,m})}{2c}\frac{e^{-i\Omega^{(-)}_{\alpha,m}(t-t_0)}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}[\dot{q}_{\alpha,j}(t_0)-i\Omega^{(-)}_{\alpha,m}q_{\alpha,j}(t_0)]\hat{\boldsymbol{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})+\text{H.c.}\end{aligned} \quad (3.14)$$

and similarly for the magnetic field

$$\begin{aligned}\mathbf{B}^{(0)}(\mathbf{x},t)&=\frac{1}{c}\sum_{\alpha,j}[\dot{U}_\alpha(t-t_0)\dot{q}_{\alpha,j}(t_0)+\ddot{U}_\alpha(t-t_0)q_{\alpha,j}(t_0)]\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})\\&=\sum_{\alpha,j,m}\frac{\Omega^{(-)}_{\alpha,m}\tilde{\varepsilon}(\Omega^{(-)}_{\alpha,m})}{2c\omega_\alpha}\frac{e^{-i\Omega^{(-)}_{\alpha,m}(t-t_0)}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}[\dot{q}_{\alpha,j}(t_0)-i\Omega^{(-)}_{\alpha,m}q_{\alpha,j}(t_0)]\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})+\text{H.c.}\end{aligned} \quad (3.15)$$

The (transverse) electric field associated with these free solutions can also be obtained from the definition $\mathbf{D}^{(0)}(\mathbf{x},t)=\mathbf{E}^{(0)}(\mathbf{x},t)+\int_0^{t-t_0}d\tau\,\chi(\tau)\mathbf{E}^{(0)}(\mathbf{x},t-\tau)$. We have thus $\overline{\mathbf{D}'}^{(0)}(\mathbf{x},p) = [1+\bar{\chi}(p)]\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)$ and considering the Laplace transform in Eqs. (3.11) and (B4) we can express the electric field as a function of $H_\alpha(\tau)$, i.e.,

$$\begin{aligned}\mathbf{E}^{(0)}(\mathbf{x},t) &= i\sum_{\alpha,j}\frac{\omega_\alpha}{c}[H_\alpha(t-t_0)\dot{q}_{\alpha,j}(t_0)\\ &\quad + \dot{H}_\alpha(t-t_0)q_{\alpha,j}(t_0)]\hat{\boldsymbol{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})\\ &= \sum_{\alpha,j,m}\frac{-1}{2c}\frac{e^{-i\Omega^{(-)}_{\alpha,m}(t-t_0)}}{\frac{\partial[\omega\sqrt{\bar{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}[\dot{q}_{\alpha,j}(t_0)\\ &\quad - i\Omega^{(-)}_{\alpha,m}q_{\alpha,j}(t_0)]\hat{\boldsymbol{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{H.c.}\end{aligned} \tag{3.16}$$

From Eqs. (3.15) and (3.16) we see that $\nabla\times\mathbf{E}^{(0)}(\mathbf{x},t) = -\partial_t\mathbf{B}^{(0)}(\mathbf{x},t)/c$ in agreement with Maxwell's equation for a free field (the other Maxwell's equations are also automatically fulfilled by definition).

Importantly, in the vacuum case where $\chi(\tau)\to 0$ we have $\Omega^{(-)}_{\alpha,m}\to\omega_\alpha$ and we find that the vacuum fields are given by

$$\begin{aligned}\mathbf{F}^{(v)}(\mathbf{x},t) &= \sum_{\alpha,j} ic\sqrt{\frac{\hbar}{2\omega_\alpha}}c^{(v)}_{\alpha,j}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{H.c.},\\ \mathbf{D}^{(v)}(\mathbf{x},t) &= \sum_{\alpha,j} -\sqrt{\frac{\hbar\omega_\alpha}{2}}c^{(v)}_{\alpha,j}(t)\hat{\mathbf{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{H.c.},\\ \mathbf{B}^{(v)}(\mathbf{x},t) &= \sum_{\alpha,j}\sqrt{\frac{\hbar\omega_\alpha}{2}}c^{(v)}_{\alpha,j}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{H.c.}\end{aligned} \tag{3.17}$$

with $c^{(v)}_{\alpha,j}(t) = c_{\alpha,j}(t_0)e^{-i\omega_\alpha(t-t_0)}$ as expected. In the general case, however, causality imposes that the imaginary part of $\Omega^{(-)}_{\alpha,m}$ is negative. Therefore, the optical modes labeled by α, j, and m are damped in time [the only exception being of course the vacuum case where the only contribution to the field arises from the source-free term (0) since the (s) terms vanishes together with $\mathbf{P}^{(0)}$]. As a consequence, if $t-t_0\to+\infty$, the free terms vanish asymptotically. In particular, if $t_0\to-\infty$ (corresponding to initial conditions fixed in the infinite remote past), we can omit for all practical purposes the contribution of $\mathbf{F}^{(0)}$, $\mathbf{D}^{(0)}$, and $\mathbf{B}^{(0)}$ to the field observed at any finite time t (unless we are in the vacuum). This is indeed what was implicitly done by Huttner and Barnet [17] and Gruner and Welsch [34] and that is why for all calculational needs they completely omitted the discussion of the $\mathbf{F}^{(0)}$, $\mathbf{D}^{(0)}$, and $\mathbf{B}^{(0)}$ fields. However, for preserving the unitarity of the full evolution one must necessarily include both (0) and (s) terms. While this problem is apparently only technical, we will see in the following its importance for inhomogeneous systems.

C. Fluctuating Langevin modes

The previous discussion concerning the omission of the (0) source-free terms is very important since it explains the mechanism at work in the Huttner-Barnet model [17]. To clarify that point further, we now express the scattered field (s) using a Green tensor formalism (in Appendix C we introduce alternative descriptions based on vectorial and scalar potentials). We first observe that from $\mathbf{D}^{(s)}(\mathbf{x},t) = \nabla\times\mathbf{F}^{(s)}(\mathbf{x},t)$ we obtain after integration by parts

$$\begin{aligned}\mathbf{D}^{(s)}(\mathbf{x},t) &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\int d^3\mathbf{x}'\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip)\\ &\quad\cdot\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)e^{p(t-t_0)}\end{aligned} \tag{3.18}$$

with the dyadic propagator [33,34,37,38]

$$\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip) = \sum_{\alpha,j}\frac{\omega_\alpha^2\Phi_\alpha(\mathbf{x})\Phi^*_\alpha(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\otimes\hat{\boldsymbol{\epsilon}}_{\alpha,j}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2}. \tag{3.19}$$

The meaning of the tensor $\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip)$, which depends on χ, becomes more clear if we introduce the Green tensor $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)$ solution of

$$\begin{aligned}&\nabla\times\nabla\times\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) + \frac{p^2}{c^2}[1+\bar{\chi}(p)]\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)\\ &\quad = \mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}') = \boldsymbol{\delta}(\mathbf{x}-\mathbf{x}').\end{aligned} \tag{3.20}$$

We observe that if we separate the tensor into a transverse and longitudinal part, we get $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) = \mathbf{G}_{\chi,\perp}(\mathbf{x},\mathbf{x}',ip) + \mathbf{G}_{\chi,\parallel}(\mathbf{x},\mathbf{x}',ip)$ with for the transverse dyadic Green function

$$\mathbf{G}_{\chi,\perp}(\mathbf{x},\mathbf{x}',ip) = \sum_{\alpha,j}\frac{c^2\Phi_\alpha(\mathbf{x})\Phi^*_\alpha(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\otimes\hat{\boldsymbol{\epsilon}}_{\alpha,j}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2} \tag{3.21}$$

and for the longitudinal part

$$\frac{p^2}{c^2}[1+\bar{\chi}(p)]\mathbf{G}_{\chi,\parallel}(\mathbf{x},\mathbf{x}',ip) = \boldsymbol{\delta}_\parallel(\mathbf{x}-\mathbf{x}') \tag{3.22}$$

with the unit longitudinal dyadic distribution $\boldsymbol{\delta}_\parallel(\mathbf{x}-\mathbf{x}') = \sum_\alpha\hat{\mathbf{k}}_\alpha\otimes\hat{\mathbf{k}}_\alpha\Phi^*_\alpha(\mathbf{x}')\Phi_\alpha(\mathbf{x})$. We have also the important relations between the tensors $\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip)$ and $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)$:

$$\begin{aligned}\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip) &= -\frac{p^2}{c^2}[1+\bar{\chi}(p)]\mathbf{G}_{\chi,\perp}(\mathbf{x},\mathbf{x}',ip) + \boldsymbol{\delta}_\perp(\mathbf{x}-\mathbf{x}')\\ &= -\frac{p^2}{c^2}[1+\bar{\chi}(p)]\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) + \boldsymbol{\delta}(\mathbf{x}-\mathbf{x}')\\ &= \nabla\times\nabla\times\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip).\end{aligned} \tag{3.23}$$

Now, if we write $\overline{\mathbf{D}'}(\mathbf{x},p) = \overline{\mathbf{E}'}(\mathbf{x},p) + \overline{\mathbf{P}'}(\mathbf{x},p) = [1+\bar{\chi}(p)]\overline{\mathbf{E}'}(\mathbf{x},p) + \overline{\mathbf{P}'}^{(0)}(\mathbf{x},p)$ and introduce the relation $\overline{\mathbf{E}'}(\mathbf{x},p) = \overline{\mathbf{E}'}^{(0)}(\mathbf{x},p) + \overline{\mathbf{E}'}^{(s)}(\mathbf{x},p)$ with $\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)$ the free mode given by Eq. (3.16) and $\overline{\mathbf{E}'}^{(s)}(\mathbf{x},p)$ the source field, induced by $\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)$, which is given by

$$\overline{\mathbf{E}'}^{(s)}(\mathbf{x},p) = -\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p), \tag{3.24}$$

we get $\overline{\mathbf{D}'}^{(s)}(\mathbf{x},p) = [1+\bar{\chi}(p)]\overline{\mathbf{E}'}^{(s)}(\mathbf{x},p) + \overline{\mathbf{P}'}^{(0)}(\mathbf{x},p)$, i.e., Eq. (3.18) as it could be checked directly after comparing Eq. (3.19) with Eqs. (3.21) and (3.22). Other important relations between the dyadic formalism and scalar Green function are given in Appendix D.

Importantly, we can write all scattered fields as a function of the rising and lowering operators $\mathbf{f}^{\dagger(0)}_\omega(\mathbf{x},t)$, $\mathbf{f}^{(0)}_\omega(\mathbf{x},t)$. In order to give explicit expressions, we use the Fourier transform notations (i.e., with $p=\gamma-i\omega$ with $\gamma\to 0^+$) and the relation [75]

$$\widetilde{\mathbf{P}}^{(0)}(\mathbf{x},\omega) = \int_0^{+\infty} d\omega' \sqrt{\frac{\hbar\sigma_{\omega'}(\mathbf{x})}{\pi\omega'}} [\mathbf{f}^{(0)}_{\omega'}(\mathbf{x},t_0)e^{i\omega' t_0}\delta(\omega-\omega') + \mathbf{f}^{\dagger(0)}_{\omega'}(\mathbf{x},t_0)e^{-i\omega' t_0}\delta(\omega+\omega')]. \quad (3.25)$$

We obtain

$$\mathbf{D}^{(s)}(\mathbf{x},t) = \sum_{\alpha,j}\int_0^{+\infty} d\omega \frac{\omega_\alpha^2 \Phi_\alpha(\mathbf{x})\hat{\boldsymbol{\epsilon}}_{\alpha,j}}{\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}} f^{(0)}_{\omega,\alpha,j}(t) + \text{H.c.}, \quad (3.26)$$

$$\mathbf{E}^{(s)}_{\perp}(\mathbf{x},t) = \sum_{\alpha,j}\int_0^{+\infty} d\omega \frac{\omega^2 \Phi_\alpha(\mathbf{x})\hat{\boldsymbol{\epsilon}}_{\alpha,j}}{\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}} f^{(0)}_{\omega,\alpha,j}(t) + \text{H.c.}, \quad (3.27)$$

where $f^{(0)}_{\omega,\alpha,j}(t)$ is a lowering operator associated with the transverse fluctuating field and defined as

$$f^{(0)}_{\omega,\alpha,j}(t) = \int d^3\mathbf{x}'\Phi^*_\alpha(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\cdot \mathbf{f}^{(0)}_\omega(\mathbf{x},t) \quad (3.28)$$

such that from Eq. (2.8) we get the mode commutator

$$\left[f^{(0)}_{\omega,\alpha,j}(t), f^{\dagger(0)}_{\omega',\beta,k}(t)\right] = \delta_{\alpha,\beta}\delta_{j,k}\delta(\omega-\omega') \quad (3.29)$$

and the harmonic time evolution $f^{(0)}_{\omega,\alpha,j}(t) = f^{(0)}_{\omega,\alpha,j}(t_0)e^{-i\omega_\alpha(t-t_0)}$ if we impose the initial condition $f^{(0)}_{\omega,\alpha,j}(t_0) = f_{\omega,\alpha,j}(t_0) = \int d^3\mathbf{x}'\Phi^*_\alpha(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\cdot\mathbf{f}_\omega(\mathbf{x},t_0)$.

For the longitudinal electric field we deduce from Eqs. (3.22) and (3.24) $\overline{\mathbf{E}'}^{(s)}_{\parallel}(\mathbf{x},p) = -\frac{\overline{\mathbf{P}'}^{(0)}_{\parallel}(\mathbf{x},p)}{[1+\bar{\chi}(p)]}$ [in agreement with the definition $\overline{\mathbf{D}'}_{\parallel}(\mathbf{x},p)=0$] and, therefore,

$$\mathbf{E}^{(s)}_{\parallel}(\mathbf{x},t) = -\sum_{\alpha}\int_0^{+\infty} d\omega \frac{\Phi_\alpha(\mathbf{x})\hat{\boldsymbol{k}}_\alpha}{\tilde{\varepsilon}(\omega)}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}} f^{(0)}_{\omega,\alpha,\parallel}(t) + \text{H.c.} \quad (3.30)$$

We have $f^{(0)}_{\omega,\alpha,\parallel}(t) = \int d^3\mathbf{x}'\Phi^*_\alpha(\mathbf{x}')\hat{\boldsymbol{k}}_\alpha\cdot\mathbf{f}^{(0)}_\omega(\mathbf{x},t)$ and the commutator $[f^{(0)}_{\omega,\alpha,\parallel}(t), f^{\dagger(0)}_{\omega',\beta,\parallel}(t)] = \delta_{\alpha,\beta}\delta_{j,k}\delta(\omega-\omega')$ and the time evolution $f^{(0)}_{\omega,\alpha,\parallel}(t) = f^{(0)}_{\omega,\alpha,\parallel}(t_0)e^{-i\omega_\alpha(t-t_0)}$ with similar initial condition as for the transverse field. Regrouping these definitions we have obviously

$$\mathbf{f}^{(0)}_\omega(\mathbf{x},t) = \sum_\alpha \hat{\boldsymbol{k}}_\alpha\Phi_\alpha(\mathbf{x})f^{(0)}_{\omega,\alpha,\parallel}(t) + \sum_{\alpha,j}\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})f^{(0)}_{\omega,\alpha,j}(t). \quad (3.31)$$

Furthermore, we can easily show that we have also $\overline{\mathbf{B}'}^{(s)}(\mathbf{x},p) = \int d^3\mathbf{x}'\frac{p}{c}\nabla\times\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)$ leading to

$$\mathbf{B}^{(s)}(\mathbf{x},t) = \sum_{\alpha,j}\int_0^{+\infty} d\omega \frac{\omega c\mathbf{k}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2} \times\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}} f^{(0)}_{\omega,\alpha,j}(t) + \text{H.c.} \quad (3.32)$$

The description of the scattered field (s) given here corresponds exactly to what Huttner and Barnett [17] called the quantized field obtained after generalizing the diagonalization procedure of Fano and Hopfield [23,24]. Here, we justify these modes by using the Laplace transform method and by taking the limit $t_0 \to -\infty$ explicitly. This means that we neglect the contribution of the (0) transverse field which is infinitely damped at time t. Importantly, $\mathbf{P}^{(0)}(\mathbf{x},t)$ does not vanish since the time evolution of $\mathbf{f}^{(0)}_\omega(\mathbf{x},t)$ in Eq. (2.10) is harmonic.

D. A discussion on causality and time symmetry

It is important to further comment about causality and on the structure of the total field as a sum of (0) and (s) modes. The (0) (classical polariton) modes are indeed exponentially damped in the future direction meaning that a privileged temporal direction apparently holds in this model. This would mean that we somehow break the time symmetry of the problem. However, since the evolution equations are fundamentally time symmetric, this should clearly not be possible. Similarly, propagators such as $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)$ are also spatially damped at large distance [see Eq. (C1)] since we have terms like $\sim\frac{e^{i\omega\sqrt{\tilde{\varepsilon}(\omega)}|\mathbf{x}-\mathbf{x}'|/c}}{4\pi|\mathbf{x}-\mathbf{x}'|}$. This also seems to imply a privileged time direction and would lead to a kind of paradox. However, we should remind that only the sum $(0)+(s)$ has a physical meaning and this sum must preserve time symmetry. Indeed, we remind that time reversal applied to electrodynamics implies that if $\mathbf{E}(t)$, $\mathbf{B}(t)$, and $\mathbf{X}_\omega(t)$ are a solution of the coupled set of equations given in Sec. II, then the time-reversed solutions [84] $\mathbf{E}_T(t)=\mathbf{E}(-t)$, $\mathbf{B}_T(t)=-\mathbf{B}(-t)$, and $\mathbf{X}_{\omega,T}(t)=\mathbf{X}_\omega(-t)$ is also defining a solution of the same equations [we have also $\mathbf{F}_T(t)=\mathbf{F}(-t)$ and $\mathbf{P}_T(t)=\mathbf{P}(-t)$]. Now, considering the dipole density evolution we get from Eq. (2.9) after some manipulations

$$\mathbf{P}_T(\mathbf{x},t) = \mathbf{P}^{(0)}_T(\mathbf{x},t) + \int_{t+t_0}^0 \chi(\mathbf{x},-\tau)d\tau\mathbf{E}_T(\mathbf{x},t-\tau), \quad (3.33)$$

where $\mathbf{P}^{(0)}_T$ is defined as $\mathbf{P}^{(0)}$ [see the definition (2.10)] but with $\mathbf{X}^{(0)}_\omega(\mathbf{x},t) = \cos[\omega(t-t_0)]\mathbf{X}_\omega(\mathbf{x},t_0) + \frac{\sin[\omega(t-t_0)]}{\omega}\partial_t\mathbf{X}_\omega(\mathbf{x},t_0)$ replaced by $\mathbf{X}^{(0)}_{\omega,T}(\mathbf{x},t) = \cos[\omega(t+t_0)]\mathbf{X}_{\omega,T}(\mathbf{x},-t_0) + \frac{\sin[\omega(t+t_0)]}{\omega}\partial_t\mathbf{X}_{\omega,T}(\mathbf{x},-t_0)$. The presence of the time $-t_0$ everywhere has a clear meaning. Indeed, from special relativity choosing a time reference frame with $t'=-t$ (passive time-reversal transformation) implies that the causal evolution of $\mathbf{P}$ defined for $t\geqslant t_0$ will become an anticausal evolution defined for $t'\leqslant t'_0=-t_0$. Going back to the active time-reversal transformation (3.33) we see that the new linear susceptibility $\chi_T(\mathbf{x},\tau)=\chi(\mathbf{x},-\tau)$ is given by

$$\chi(\mathbf{x},-\tau) = -\int_0^{+\infty} d\omega\frac{2\sigma_\omega(\mathbf{x})}{\pi}\frac{\sin\omega\tau}{\omega}\Theta(-\tau) \quad (3.34)$$

[$\Theta(-\tau)$ is explicitly written in order to emphasize the anticausal structure]. However, since we have $\widetilde{\chi_T}(\omega)=\int_{-\infty}^{+\infty}\frac{dt}{2\pi}e^{i\omega t}\chi_T(t)=\int_{-\infty}^{+\infty}\frac{dt}{2\pi}e^{i\omega t}\chi(-t)$ and $\chi(-t)=\chi^*(-t)$ we have $\widetilde{\chi_T}(\omega)=\int_{-\infty}^{+\infty}\frac{dt}{2\pi}e^{i\omega t}\chi^*(-t)=\int_{-\infty}^{+\infty}\frac{du}{2\pi}e^{-i\omega u}\chi^*(u)=\widetilde{\chi}^*(\omega)$. This means that the new permittivity (with poles in the upper half complex frequency space) is now associated with growing anticausal modes since $\widetilde{\varepsilon_T}''(\omega)<0$. The (active) time-reversed evolution is defined for $t<-t_0$ so that we indeed get modes decaying into the past direction while growing into the future direction. This becomes even more clear for the time-reversal evolution of the electromagnetic field given by the separation in (0) and (s) modes. The time reversal applied on the (0) field such as $\mathbf{E}_T^{(0)}(t)$ corresponding to classical polaritons is now involving frequency $-\Omega_{\alpha,m}^{(-)}$ instead of $\Omega_{\alpha,m}^{(-)}$. This leads to reversed temporal evolution such as $e^{i\Omega_{\alpha,m}^{(-)}(t+t_0)}$ and $e^{-i\Omega_{\alpha,m}^{(-)*}(t+t_0)}$ associated with growing waves in the future direction (since the new poles are now in the upper half complex frequency space). More generally, we have shown (see Appendix C) that the full evolution of either (0) or (s) fields is completely defined by the knowledge of the Green function and propagators such as

$$\Delta_\chi(\tau,|\mathbf{x}-\mathbf{x}'|) = c^2 \sum_\alpha H_\alpha(\tau)\Phi_\alpha^*(\mathbf{x}')\Phi_\alpha(\mathbf{x}) \quad (3.35)$$

[see Eq. (C4)] with the causal function $H_\alpha(\tau)$ given by the Bromwich-Fourier integral $\int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{e^{-i\omega\tau}}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}$. Now, like for the susceptibility $\chi_T(t)$, time reversal leads to a new propagator $\Delta_{\chi,T}(\tau,|\mathbf{x}-\mathbf{x}'|) = \Delta_\chi(-\tau,|\mathbf{x}-\mathbf{x}'|)$ and therefore by a reasoning equivalent to the previous one to

$$\Delta_{\chi,T}(\tau,|\mathbf{x}-\mathbf{x}'|) = c^2 \sum_\alpha H_\alpha^*(-\tau)\Phi_\alpha^*(\mathbf{x}')\Phi_\alpha(\mathbf{x}) \quad (3.36)$$

involving the complex conjugate $H_\alpha^*(-\tau)$ with a Fourier expansion which again involves the anticausal permittivity $\tilde{\varepsilon}^*(\omega)$. This naturally leads to growing waves in the future direction and to Green function spatially growing as $\sim \frac{e^{i\omega\sqrt{\tilde{\varepsilon}*(\omega)}|\mathbf{x}-\mathbf{x}'|/c}}{4\pi|\mathbf{x}-\mathbf{x}'|}$. The full equivalence between the two representations of the total field $(s)+(0)$ is completed if first we observe that the initial conditions at t_0 [i.e., $X_\omega(t_0)$, $\mathbf{E}(t_0)$, etc.] are now replaced by "final" boundary condition at $t_f = -t_0$ [i.e., $X_{\omega,T}(-t_0)$, $\mathbf{E}_T(-t_0)$, etc.]. Second, since the $-t_0$ value is arbitrary, we can send it into the remote future if we want and we will have thus an evolution expressed in terms of growing modes for all times $t \leqslant t_f$. The value of the field at such a boundary is of course arbitrary, so that if we want the two representations can describe the same field if for the final field at t_f we take the field evolving from t_0 using the usual causal evolution from past to future. This equivalence is of course reminiscent from the dual representations obtained using either retarded or advanced waves [85–87]. Indeed, the total electric field $\mathbf{E}$ can always be separated into $\mathbf{E}_{\text{in}} + \mathbf{E}_{ret}$ or equivalently $\mathbf{E}_{out} + \mathbf{E}_{adv}$ where ret and adv label the retarded and advanced source fields, respectively, and in and out label the homogeneous "free" fields coming from the past and from the future, respectively. This implies that only a specific choice of boundary conditions in the past or future can lead to a completely causal evolution and, therefore, time symmetry is not broken in the evolution equations but only through a specification of the boundary conditions. In other words, in agreement with the famous Loschmidt and Poincaré objections to Boltzmann, strictly deriving time irreversibility from an intrinsically time-reversible dynamics is obviously impossible without additional postulates. This point was fully recognized already by Boltzmann long ago and was the basis for his statistical interpretation of the second law of thermodynamics [87].

E. Fano-Hopfield diagonalization procedure

In order to further understand the implication of the Huttner-Barnett model [17], we should discuss how the full Hamiltonian finally reads in this formalism. This is central since the Langevin-noise equations developed by Gruner and Welsch [34] use only the $H_M^{(0)}(t)$ Hamiltonian. In the Huttner-Barnett model, the full dynamics is determined by the complete knowledge of the dipole density $\mathbf{P}^{(0)}(t)$ so that the results of Gruner and Welsch [34] should be in principle justifiable. However, if we are not careful, this will ultimately break unitary and time symmetry. In order to understand the physical mechanism at work, one should first clarify the relation existing between the Huttner-Barnett model, using $\mathbf{P}^{(0)}(t)$ as a fundamental field, and the historical Hopfield-Fano approach [23,24] for defining discrete polariton modes as normal coordinate solutions of the full Hamiltonian.

We remind that the historical method for dealing with polariton is indeed based on the pioneer work by Fano and Hopfield [23,24] for diagonalizing the full Hamiltonian. The procedure is actually reminiscent of the classical problem of finding normal coordinates and normal (real-valued) eigenfrequencies associated with free vibrations of a system of linearly coupled harmonic oscillators. The classical diagonalization method [88] relies on the resolution of secular equations already studied by Laplace. Actually, the first "semiclassical" treatment made by Born and Huang [25] is mathematically correct and equivalent to the one made later by Fano and Hopfield even though the relation between those formalisms is somehow hidden behind the mathematical symbols. The full strategy for finding normal coordinates and frequencies becomes clear if we use Fourier transforms of the various fields for the problem under study. Indeed, a Fourier expansion of a field component $A(t) = \int_{-\infty}^{+\infty} d\Omega\, \tilde{A}(\Omega)e^{-i\Omega}$ will obviously define the needed harmonic expansion. In the problems considered by Born, Huang, Fano, and Hopfield, the Fourier spectrum $\tilde{A}(\Omega)$ is a sum of Dirac distributions $\delta(\Omega \mp \Omega_n)$ peaked on the real-valued eigenvibrations Ω_n. This specific situation needs a complete discussion since the Hopfield-Fano model [23,24] leads to an exact diagonalization of the full Hamiltonian $H(t)$. This will in turn make clear some fundamental relations with the Laplace transform formalism used in the previous subsections.

We start with the Fourier transformed equations

$$\nabla\times\nabla\times\tilde{\mathbf{E}}(\Omega) - \frac{\Omega^2}{c^2}\tilde{\mathbf{E}}(\Omega) = \frac{\Omega^2}{c^2}\tilde{\mathbf{P}}(\Omega), \quad (3.37)$$

$$(\omega^2-\Omega^2)\tilde{\mathbf{X}}_\omega(\Omega) = \sqrt{\frac{2\sigma_\omega}{\pi}}\tilde{\mathbf{E}}(\Omega), \quad (3.38)$$

and

$$\int_0^{+\infty} d\omega \sqrt{\frac{2\sigma_\omega}{\pi}}\tilde{\mathbf{X}}_\omega(\Omega) = \tilde{\mathbf{P}}(\Omega). \quad (3.39)$$

With the constraint $\tilde{\mathbf{E}}(\Omega) = \tilde{\mathbf{E}}(-\Omega)^*$, $\tilde{\mathbf{X}}_\omega(\Omega) = \tilde{\mathbf{X}}_\omega(-\Omega)^*$ coming from the real-valued nature of the fields. From Eq. (3.38), we get using the properties of distributions

$$\tilde{\mathbf{X}}_\omega(\Omega) = P\left[\frac{1}{\omega^2-\Omega^2}\right]\sqrt{\frac{2\sigma_\omega}{\pi}}\tilde{\mathbf{E}}(\Omega) + \tilde{\mathbf{X}}_\omega^{(\text{sym})}(\Omega) \quad (3.40)$$

with

$$\widetilde{\mathbf{X}_\omega}^{(\mathrm{sym})}(\Omega) = \sqrt{\frac{\hbar}{2\omega}}\left[f_\omega^{(\mathrm{sym})}\delta(\omega-\Omega) + f_\omega^{(\mathrm{sym})*}\delta(\omega+\Omega)\right], \tag{3.41}$$

and where we used the reality constraint and introduced constants of motions $f_\omega^{(\mathrm{sym})}$, $f_\omega^{(\mathrm{sym})*}$ which will become annihilation and creation operators in the second quantized formalism. The principal value can be conveniently written in different ways [89], and this fact leads to two different equivalent representations of Eq. (3.40):

$$\begin{aligned}\widetilde{\mathbf{X}_\omega}(\Omega) &= \frac{1}{\omega^2-(\Omega+i0^+)^2}\sqrt{\frac{2\sigma_\omega}{\pi}}\widetilde{\mathbf{E}}(\Omega) + \widetilde{\mathbf{X}_\omega}^{(in)}(\Omega)\\ &= \frac{1}{\omega^2-(\Omega-i0^+)^2}\sqrt{\frac{2\sigma_\omega}{\pi}}\widetilde{\mathbf{E}}(\Omega) + \widetilde{\mathbf{X}_\omega}^{(out)}(\Omega),\end{aligned} \tag{3.42}$$

where $\widetilde{\mathbf{X}_\omega}^{(in)}(\Omega)$ and $\widetilde{\mathbf{X}_\omega}^{(out)}(\Omega)$ can also be written like Eq. (3.41), i.e., respectively as $\sqrt{\frac{\hbar}{2\omega}}[f_\omega^{(in)}\delta(\omega-\Omega) + f_\omega^{(in)*}\delta(\omega+\Omega)]$ or $\sqrt{\frac{\hbar}{2\omega}}[f_\omega^{(out)}\delta(\omega-\Omega) + f_\omega^{(out)*}\delta(\omega+\Omega)]$. This discussion is reminiscent of the different representation given in Sec. III D involving retarded, advanced, or time-symmetric modes. Of course, the usual causal representation is (in) which is obtained from the definition of $f_\omega^{(0)}(t)$ given in Sec. III at the limit $t_0 \to -\infty$. However, all the descriptions are rigorously equivalent. It is also easy to show that we have $f_\omega^{(\mathrm{sym})} = \frac{f_\omega^{(in)}+f_\omega^{(out)}}{2}$ which justifies why we called this field symmetrical. It corresponds to a representation of the problem mixing boundary conditions in the future and the past in a symmetrical way like it was used, for instance, by Feynman and Wheeler in their description of electrodynamics [85] as discussed in Sec. III D.

Now, after inserting the causal representation of Eq. (3.42) in (3.39) and then into (E3) we get

$$\nabla\times\nabla\times\widetilde{\mathbf{E}}(\Omega) - \frac{\Omega^2}{c^2}\widetilde{\varepsilon}(\Omega)\widetilde{\mathbf{E}}(\Omega) = \frac{\Omega^2}{c^2}\widetilde{\mathbf{P}}^{(in)}(\Omega) \tag{3.43}$$

with $\widetilde{\mathbf{P}}^{(in)}(\Omega) = \int_0^{+\infty} d\omega\sqrt{\frac{2\sigma_\omega}{\pi}}\widetilde{\mathbf{X}_\omega}^{(in)}(\Omega)$ and

$$\widetilde{\varepsilon}(\omega) = 1 + \int_0^{+\infty} d\tau\,\chi(\tau)e^{i\omega\tau}. \tag{3.44}$$

This causal permittivity $\widetilde{\varepsilon}(\Omega)$ being given by the Huttner-Barnett model [17,75], the secular equations $\omega_\alpha^2 = \Omega^2\widetilde{\varepsilon}(\Omega)$ for transverse modes have no root in the upper complex frequency half-plane and in particular on the real frequency axis (the longitudinal term is discussed below). This means that, unlike Eq. (3.40), Eq. (3.43) has in general no Dirac term corresponding to independent eigenmodes. The electric field is thus represented by a source term

$$\widetilde{\mathbf{E}}(\mathbf{x},\Omega) = \frac{\Omega^2}{c^2}\int d^3\mathbf{x}'\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',\Omega)\cdot\widetilde{\mathbf{P}}^{(in)}(\mathbf{x}',\Omega) \tag{3.45}$$

obtained like in the previous subsection using a causal Green function. The absence of free normal modes for the electric field is of course reminiscent from the rapid decay of the free modes (0) when $t_0 \to -\infty$ as discussed before. The representation given here does not distinguish between transverse and longitudinal fields but this should naturally occur since we have the constraint $\nabla\cdot\widetilde{\mathbf{E}}(\Omega) = -\nabla\cdot\widetilde{\mathbf{P}}(\Omega)$ which implies $\widetilde{\mathbf{E}}_\parallel(\Omega) = -\widetilde{\mathbf{P}}_\parallel(\Omega)$. Together with Eq. (E3), we thus get

$$\widetilde{\mathbf{E}}_\parallel(\Omega) = -\widetilde{\mathbf{P}}_\parallel(\Omega) = -\frac{\widetilde{\mathbf{P}}_\parallel^{(in)}(\Omega)}{\widetilde{\varepsilon}(\Omega)}. \tag{3.46}$$

Equation (3.46) is actually reminiscent of the charge screening by $\widetilde{\varepsilon}(\Omega)$. Here, we used the fact that $\widetilde{\varepsilon}(\Omega)$ has no root on the real axis, otherwise, the imaginary part of the permittivity should vanish and the medium would be lossless at the frequency Ω, a fact which is prohibited by physical consideration about irreversibility [90]. This reasoning is rigorously not valid at $\Omega = 0$ since the imaginary part of the permittivity is an odd function on the real axis. But then in general to have a root at $\Omega = 0$ it would require that the real part of the permittivity vanishes as well and this is not allowed from the usual permittivity model [see Eq. (A5)] which makes therefore this possibility very improbable.

The previous reasoning is clearly formally equivalent to the ones obtained in the previous subsection, and in both cases the field $\widetilde{\mathbf{P}}^{(in)}(\mathbf{x}',\Omega)$ or $\mathbf{P}^{(0)}(t)$ completely determines the electromagnetic evolution. Still, there are exceptions for instance in the Drude-Lorentz model with $\widetilde{\varepsilon}(\Omega) = 1 + \frac{\omega_p^2}{\omega_0^2-(\Omega+i0^+)^2}$ which forms the basis for the Hopfield [24] polariton model. This model is rigorously not completely lossless since we have $\widetilde{\varepsilon}(\Omega) = 1 + P[\frac{\omega_p^2}{\omega_0^2-\Omega^2}] + \frac{i\pi\omega_p^2}{2\omega_0}[\delta(\Omega-\omega_0) - \delta(\Omega+\omega_0)]$ corresponding to a singular absorption peak.

Moreover, in this Hopfield model [24], which is a limit case of the Huttner-Barnett model [17,91], we get the exact evolution equation

$$\partial_t^2\mathbf{P} + \omega_0^2\mathbf{P} = \omega_p^2\mathbf{E} \tag{3.47}$$

which in the case of the longitudinal modes leads to solving the secular equation $(\omega_0^2-\Omega^2)\widetilde{\mathbf{P}}_\parallel(\Omega) = -\omega_p^2\widetilde{\mathbf{P}}_\parallel(\Omega)$. It has the solution

$$\widetilde{\mathbf{P}}_\parallel(\Omega) = \boldsymbol{\beta}\delta(\Omega-\omega_L) + \boldsymbol{\beta}^*\delta(\Omega+\omega_L), \tag{3.48}$$

where $\omega_L = \sqrt{\omega_0^2+\omega_p^2}$ is the longitudinal plasmon frequency. However, relating this result to Eq. (3.46) requires careful calculations since $\widetilde{\varepsilon}(\Omega)$ is here a highly singular distribution. Indeed, applying Eq. (3.46) will lead to find solutions of $\widetilde{\varepsilon}(\Omega)\widetilde{\mathbf{P}}_\parallel(\Omega) = \widetilde{\mathbf{P}}_\parallel^{(in)}(\Omega)$ with $\widetilde{\mathbf{P}}_\parallel^{(in)}(\Omega) = \boldsymbol{\alpha}\delta(\Omega-\omega_0) + \boldsymbol{\alpha}^*\delta(\Omega+\omega_0)$ with $\boldsymbol{\alpha}(\mathbf{x})$ a longitudinal vector field. This singular (in) field at $\Omega = \pm\omega_0$ seems to imply that polaritons are resonant at such frequency in contradiction with the result leading to $\Omega = \pm\omega_L$ for such polariton modes. Now, due to the presence of absorption peaks at $\Omega = \pm\omega_0$ we have near this singular point $\boldsymbol{\alpha} = \frac{i\pi\omega_p^2}{2\omega_0}\widetilde{\mathbf{P}}_\parallel(\omega_0)$ where $\widetilde{\mathbf{P}}_\parallel(\omega_0)$ is supposed to be regular [92]. Moreover, outside this narrow absorption band, the medium is effectively lossless and instead of Eq. (3.46) we have $(1+\frac{\omega_p^2}{\omega_0^2-\Omega^2})\widetilde{\mathbf{P}}_\parallel(\Omega) = 0$ which has the singular solution $\widetilde{\mathbf{P}}_\parallel(\Omega) = \boldsymbol{\beta}\delta(\Omega-\omega_L) + \boldsymbol{\beta}^*\delta(\Omega+\omega_L)$ with $\boldsymbol{\beta}(\mathbf{x})$ a longitudinal vector field. Importantly, from the hypothesis of regularity at $\pm\omega_0$ we have $\widetilde{\mathbf{P}}_\parallel(\omega_0) = 0$ and therefore $\boldsymbol{\alpha} = 0$ which means that $\widetilde{\mathbf{P}}_{\parallel}^{(in)}(\Omega) = 0$ everywhere. The Lorentz-Drude model leads therefore to genuine longitudinal polariton eigenfrequencies $\pm\omega_L$ solutions of $\widetilde{\varepsilon}(\omega_L) = 0$. We emphasize that the same result could be obtained using the Laplace transform method. We have indeed $\overline{\mathbf{E}'}_{\parallel}^{(s)}(\mathbf{x},p) = -\frac{\overline{\mathbf{P}'}_{\parallel}^{(0)}(\mathbf{x},p)}{1+\omega_p^2/(p^2+\omega_0^2)}$ with $\overline{\mathbf{P}'}_{\parallel}^{(0)}(\mathbf{x},p) = \frac{p\mathbf{P}_{\parallel}(\mathbf{x},t_0)+\partial_{t_0}\mathbf{P}_{\parallel}(\mathbf{x},t_0)}{\omega_0^2+p^2}$. Therefore, we can rewrite the longitudinal scattered field as $\overline{\mathbf{E}'}_{\parallel}^{(s)}(\mathbf{x},p) = -\frac{p\mathbf{P}_{\parallel}(\mathbf{x},t_0)+\partial_{t_0}\mathbf{P}_{\parallel}(\mathbf{x},t_0)}{\omega_L^2+p^2}$. Moreover, since there is no longitudinal (0) electric field and since $\mathbf{P}_{\parallel} = -\mathbf{E}_{\parallel}$ we have $\mathbf{P}_{\parallel}(t) = \cos[\omega_L(t-t_0)]\mathbf{P}_{\parallel}(t_0) + \sin[\omega_L(t-t_0)]\partial_{t_0}\mathbf{P}_{\parallel}(t_0)$ which shows that the genuine longitudinal polariton oscillates at the frequency ω_L as expected.

The transverse polariton modes of the Hopfield model are obtained in a similar way from Eq. (3.43) with $\widetilde{\mathbf{P}}_{\perp}^{(in)}(\Omega) = \boldsymbol{\gamma}\delta(\Omega-\omega_0) + \boldsymbol{\gamma}^*\delta(\Omega+\omega_0)$ with $\boldsymbol{\gamma}(\mathbf{x})$ a transverse vector field. As explained in Appendix G, solving the problem with a plane-wave expansion labeled by α and j leads again to a secular equation $\omega_\alpha^2 - \Omega_{\alpha,\pm}2\widetilde{\varepsilon}(\Omega_{\alpha,\pm}) = 0$ for the two transverse modes ($\pm$) giving a quartic dispersion relation

$$\omega_\alpha^2\omega_0^2 - \Omega_{\alpha,\pm}^2(\omega_\alpha^2 + \omega_L^2) + \Omega_{\alpha,\pm}^4 = 0 \tag{3.49}$$

with two usual Hopfield solutions

$$\Omega_{\pm} = \frac{\sqrt{\omega_\alpha^2 + \omega_L^2 \pm \sqrt{(\omega_\alpha^2+\omega_L^2)^2 - 4\omega_\alpha^2\omega_0^2}}}{\sqrt{2}}. \tag{3.50}$$

The two-mode fields have now the structure

$$\widetilde{\mathbf{E}}(\mathbf{x},\Omega) = \sum_{\alpha,j,\pm} \widetilde{E}_{\alpha,j,\pm}(\Omega)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) \tag{3.51}$$

with $\widetilde{E}_{\alpha,j,\pm}(\Omega) = \phi_{\alpha,j,\pm}\delta(\Omega-\Omega_{\alpha,\pm}) + \eta_j\phi^*_{-\alpha,j,\pm}\delta(\Omega+\Omega_{\alpha,\pm})$ with $\phi_{\alpha,j,\pm}$ an amplitude coefficient for the mode (see Appendix E and [83] for a derivation).

One of the most important issues in the context of the Hopfield model concerns the Hamiltonian. Indeed, in this model the full Hamiltonian (2.13) $H(t) = \int d^3\mathbf{x}\frac{\mathbf{B}^2+\mathbf{E}^2}{2} + H_M$ reads as

$$H(t) = \int d^3\mathbf{x}\left[\frac{\mathbf{B}^2+\mathbf{E}^2}{2} + \frac{(\partial_t\mathbf{P})^2 + \omega_0^2\mathbf{P}^2}{2\omega_p^2}\right]. \tag{3.52}$$

If we isolate first the longitudinal term we get

$$\begin{aligned} H_{\parallel}(t) &= \int d^3\mathbf{x}\left[\frac{\mathbf{P}_{\parallel}^2}{2} + \frac{(\partial_t\mathbf{P}_{\parallel})^2 + \omega_0^2\mathbf{P}_{\parallel}^2}{2\omega_p^2}\right] \\ &= \int d^3\mathbf{x}\left[\frac{(\partial_t\mathbf{P}_{\parallel})^2 + \omega_L^2\mathbf{P}_{\parallel}^2}{2\omega_p^2}\right] = 2\frac{\omega_L^2}{\omega_p^2}\int d^3\mathbf{x}\,\boldsymbol{\beta}^*\boldsymbol{\beta}. \end{aligned} \tag{3.53}$$

We can of course introduce a Fourier transform of the dipole field as $\mathbf{P}_{\parallel} = \sum_\alpha P_\alpha(\Omega)\hat{\boldsymbol{k}}_\alpha\Phi_\alpha(\mathbf{x})$ and polariton field amplitudes $\beta_\alpha = \int d^3\mathbf{x}\,\boldsymbol{\beta}(\mathbf{x})\cdot\hat{\boldsymbol{k}}_\alpha\Phi_\alpha(\mathbf{x})$. We thus have $H_{\parallel}(t) = 2\frac{\omega_L^2}{\omega_p^2}\sum_\alpha\beta_\alpha^*\beta_\alpha$. This expression of the Hamiltonian is standard for normal coordinates expansion in linearly coupled harmonic oscillators.

Furthermore, using commutators like Eq. (2.8), one can deduce $[\mathbf{P}(\mathbf{x},t),\partial_t\mathbf{P}(\mathbf{x}',t)] = i\hbar\delta^3(\mathbf{x}'-\mathbf{x})\mathbf{I}$ and other similar ones. In the Fourier space, we thus obtain $[P_\alpha(t),\dot{P}_\beta(t)] = i\hbar\delta_{\alpha,\beta}$ which leads after straightforward transformation to the commutators $[f_{\alpha,\parallel}(t),f_{\beta,\parallel}^\dagger(t)] = \delta_{\alpha,\beta}$, $[f_{\alpha,\parallel}(t),f_{\beta,\parallel}(t)] = [f_{\alpha,\parallel}^\dagger(t),f_{\beta,\parallel}^\dagger(t)] = 0$ with $\beta_\alpha = \omega_p\sqrt{\frac{\hbar}{2\omega_L}}f_{\alpha,\parallel}$ [the time dependence in the Heisenberg picture means $f_{||,\beta}(t) = f_{||,\beta}e^{-i\omega_L t}$]. This naturally leads to the Hopfield-Fano Hamiltonian expansion for longitudinal polaritons:

$$H_{\parallel}(t) = \hbar\omega_L\sum_\alpha f_{\alpha,\parallel}^\dagger(t)f_{\alpha,\parallel}(t). \tag{3.54}$$

A similar analysis can be handled for the transverse polariton modes, but the calculation is a bit longer (see Appendix E). To summarize this calculation in few words: Using a Fourier expansion of the different primary transverse field operators in Eq. (3.52) we get after some manipulations the Hopfield-Fano expansion [23,24]

$$\begin{aligned} H_{\perp}(t) &= 2\sum_{\alpha,j,\pm}\left[1 + \frac{\omega_0^2}{\omega_p^2}\left(\frac{\omega_\alpha^2}{\Omega_{\alpha,\pm}^2} - 1\right)^2\right]\phi_{\alpha,j,\pm}^\dagger\phi_{\alpha,j,\pm} \\ &= \sum_{\alpha,j,\pm}\hbar\Omega_{\alpha,\pm}\alpha f_{\alpha,j,\pm}^\dagger(t)f_{\alpha,j\pm}(t) \end{aligned} \tag{3.55}$$

with the operator $f_{\alpha,j,\pm}(t) = f_{\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t}$ obeying the usual commutation rules for rising and lowering operators. Again, this result is expected in a modal expansion using normal coordinates and again the same result could be alternatively obtained using the Laplace transform method. To summarize, the approach developed previously using the Laplace transform formalism agrees with the normal coordinate methods based on the Fourier expansion in the frequency domain. Both approaches lead to the conclusion that for a homogeneous medium, the various electromagnetic and material fields are completely determined by the knowledge of the matter oscillating dipole density $\mathbf{P}^{(in)}(t)$ (Fourier's method) or $\mathbf{P}^{(0)}(t)$ (Laplace's method). In the limit $t_0 \to -\infty$, both approaches are equivalent and there is no contribution of the free field in a homogeneous dissipative medium (the residual $\mathbf{E}^{(0)}$, $\mathbf{B}^{(0)}$ is exponentially damped in the regime $t_0 \to -\infty$). We also showed that if losses in the Huttner-Barnett model are sharply confined in the frequency domain, we can find exact polaritonic modes which agree with the historical method developed by Fano and Hopfield [23,24]. These modes fully diagonalize the Hamiltonian $H(t)$. While we focused our study on the particular Drude-Lorentz model, the result is actually generalizable [93] to homogeneous media with conductivity $\sigma(\Omega) = \sum_n\frac{\pi\omega_{p,n}^2}{2}[\delta(\Omega-\omega_{0,n}) + \delta(\Omega+\omega_{0,n})]$ which lead to a permittivity

$$\widetilde{\varepsilon}(\Omega) = 1 + \sum_n P\left[\frac{\omega_{p,n}^2}{\omega_{0,n}^2 - \Omega^2}\right] + i\frac{\sigma(\Omega)}{\Omega}. \tag{3.56}$$

In particular, the Hamiltonian can in these special cases be written as a sum of harmonic oscillator terms corresponding to the different longitudinal and transversal polariton modes. We thus write $H_{\parallel} = \sum_{m,\alpha}\hbar\Omega_{\alpha,m}f_{\alpha,m,\parallel}^\dagger f_{\alpha,m,\parallel}$ and $H_{\perp} = \sum_{m',\alpha,j}\hbar\Omega'_{\alpha,m'}f_{\alpha,j,m'}^\dagger f_{\alpha,j,m'}$ where m and m' label the discrete longitudinal and transverse polariton modes. However, for a more general Huttner-Barnett model where the permittivity is only constrained by Kramers-Kronig relations, such a simple interpretation is not possible and the Hamiltonian is not fully diagonalized. The additional physical requirement [90] imposing that the imaginary part of the permittivity should be rigorously positive valued, i.e., $\widetilde{\varepsilon}''(\Omega) > 0$, also prohibits these exceptional cases which should therefore only appear as ideal limits with loss confined in infinitely narrow absorption bands. However, as we will show in the next subsection, the lossless idealization represents a good approximation for a quite general class of medium with weak dissipation. The previous results of Hopfield and Fano have still a physical meaning and are, for example, used with success for the description of planar cavity polaritons [30,94–99].

F. Approximately transparent medium case: Milonni's approach

It is particularly relevant to consider what happens in the Huttner-Barnett approach if we relax a bit the demanding constraints of the original Hopfield-Fano model based on Eq. (3.56). For this we consider a medium with low loss such as the medium can be considered with a good approximation as transparent in a given spectral band where the field is supposed to be limited. This approach was introduced by Milonni [7] and is based on the Hamiltonian obtained long ago by Brillouin [100] and later by Landau and Lifschitz [40,44,90] for dispersive but slowly absorbing media. The main idea is to replace the electromagnetic energy density $(\mathbf{E}^2+\mathbf{B}^2)/2$ in the full Hamiltonian by a term like $[\frac{d\omega_c\tilde{\varepsilon}(\omega_c)}{d\omega_c}\mathbf{E}^2+\mathbf{B}^2]/2$ where $\tilde{\varepsilon}(\omega_c)$ is the approximately real-valued permittivity of the field at the central pulsation ω_c with which the wave packet propagates. Since this approach has been successfully applied to quantize polaritons [6,8,9,44] or surface plasmons [101,102], it is particularly interesting to justify it in the context of the more rigorous Huttner-Barnett approach developed here. In the mean time, this will justify the use of Hopfield-Fano approach as an effective method applicable for the low-loss regime which is a good assumption in most dielectric (excluding metals supporting lossy plasmon modes).

From Poynting's theorem, it is usual in macroscopic electromagnetism to isolate the work density $W_e = \mathbf{E}\cdot\partial_t\mathbf{D}$ such as the energy conservation reads as

$$\partial_t u = W_e + \partial_t\left(\frac{\mathbf{B}^2}{2}\right) = -\nabla\cdot(c\mathbf{E}\times\mathbf{B}). \quad (3.57)$$

By direct integration we thus get the usual formula for the time derivative of the total energy $H(t)=\int d^3\mathbf{x}\,u(\mathbf{x},t)$ such as

$$\frac{d}{dt}H(t) = \frac{d}{dt}\left(\int d^3\mathbf{x}\frac{\mathbf{B}^2}{2}\right) + \int d^3\mathbf{x}\,\mathbf{E}\cdot\partial_t\mathbf{D} = \frac{d}{dt}\left(\int d^3\mathbf{x}\frac{\mathbf{B}^2}{2}\right) + \int d^3\mathbf{x}\,\mathbf{E}_\perp\cdot\partial_t\mathbf{D} \quad (3.58)$$

which cancels if the fields decay sufficiently at spatial infinity (assumption which will be done in the following) as it can be proven after using the Poynting vector divergence and Stokes theorem. We now consider a temporal integration window δt to compute the average derivative

$$\frac{\int_{\delta t}dt'\int d^3\mathbf{x}\,\mathbf{E}_\perp(t')\cdot\partial_t\mathbf{D}(t')}{\delta t} + \frac{\delta}{\delta t}\left(\int d^3\mathbf{x}\frac{\mathbf{B}^2}{2}\right) \simeq 0, \quad (3.59)$$

where $\delta\int d^3\mathbf{x}\frac{\mathbf{B}^2}{2}$ means the magnetic energy variation during the time δt and $\int_{\delta t}dt'[\ldots] = \int_t^{t+\delta t}dt'[\ldots]$ is an integration domain from an initial time t to a final time $t+\delta t$. The next step is to Fourier expand the field in the frequency domain and we write $\mathbf{E}_\perp = \mathbf{E}_\perp^{(+)} + \mathbf{E}_\perp^{(-)}$ where the positive frequency part of the field is defined as $\mathbf{E}_\perp^{(+)}(t) = \int_0^{+\infty}d\omega\,\widetilde{\mathbf{E}_\perp}(\omega)e^{-i\omega t}$ {the negative frequency part is then $\mathbf{E}_\perp^{(-)}(t) = [\mathbf{E}_\perp^{(+)}(t)]^\dagger$}. We use similar notation for the displacement field and we introduce the Fourier field $\widetilde{\mathbf{D}}(\omega)$. In order to achieve the integration (3.59), the temporal window will be supposed sufficiently large compared to the typical period $2\pi/\omega_c$ of the light pulse. This allows us to simplify the calculation and most contributions cancel out during the integration [44,90,100]. Additionally, to perform the calculation we assume that we have $\widetilde{\mathbf{D}}(\omega) = \widetilde{\varepsilon}(\omega)\widetilde{\mathbf{E}_\perp}(\omega)$. This is a usual formula in classical physics where the term $\mathbf{P}^{(0)}$ is supposed equal to zero, but here we are dealing with a quantized theory and we can not omit this term. Furthermore, we showed that in the Huttner-Barnett model when $t_0\to-\infty$ only the (s) contributions discussed in Sec. III C remain [17]. Assuming therefore this regime, the transverse fields $\mathbf{D}^{(s)}$ and $\mathbf{E}^{(s)}$ are fully expressed as a function of operators $f^{(0)}_{\omega,\alpha,j}(t)$. Equations (3.26) and (3.27) allow us to define the Fourier fields

$$\widetilde{\mathbf{D}}^{(s)}(\mathbf{x},\omega) = \sum_{\alpha,j}\frac{\omega_\alpha^2\Phi_\alpha(\mathbf{x})\hat{\boldsymbol{\epsilon}}_{\alpha,j}}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}}f^{(0)}_{\omega,\alpha,j}(t_0)e^{i\omega_\alpha t_0}, \quad (3.60)$$

$$\widetilde{\mathbf{E}}^{(s)}_\perp(\mathbf{x},\omega) = \sum_{\alpha,j}\frac{\omega^2\Phi_\alpha(\mathbf{x})\hat{\boldsymbol{\epsilon}}_{\alpha,j}}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}}f^{(0)}_{\omega,\alpha,j}(t_0)e^{i\omega_\alpha t_0} \quad (3.61)$$

for $\omega>0$. [For $\omega<0$ we have $\widetilde{\mathbf{D}}^{(s)}(\mathbf{x},\omega) = \widetilde{\mathbf{D}}^{(s)\dagger}(\mathbf{x},-\omega)$ where $\widetilde{\mathbf{D}}^{(s)}(\mathbf{x},-\omega)$ is given by Eq. (3.60) at the positive frequency $-\omega$. Similar symmetries and properties hold for the electric and magnetic fields.] We thus see that the relation $\widetilde{\mathbf{D}}(\omega)\simeq\widetilde{\varepsilon}(\omega)\widetilde{\mathbf{E}_\perp}(\omega)$ is approximately fulfilled if we consider only frequencies near a resonance at $\omega_\alpha^2 = \tilde{\varepsilon}(\omega)\omega^2$ [the spectral distribution in Eq. (3.60) is thus extremely peaked since losses are weak]. This dispersion relation occurs for transverse polariton modes $\omega\simeq\Omega_{\alpha,m}$ (neglecting the imaginary part), and if the wave packet of spectral extension $\delta\omega\sim 1/\delta t$ is centered on such a wavelength, we can replace with a good approximation ω_α^2 by $\tilde{\varepsilon}(\omega)\omega^2$ in the numerator of Eq. (3.60) leading thus to $\widetilde{\mathbf{D}}(\omega)\simeq\widetilde{\varepsilon}(\omega)\widetilde{\mathbf{E}_\perp}(\omega)$. After this assumption, the calculation can be done like in classical textbooks [90,100] and Eq. (3.59) becomes (this usual calculation will not be repeated here)

$$\frac{\delta}{\delta t}H(t) = \frac{\delta}{\delta t}\left\{\int d^3\mathbf{x}\left[\frac{d\omega_c\tilde{\varepsilon}(\omega_c)}{d\omega_c}\mathbf{E}_\perp^{(-)}\mathbf{E}_\perp^{(+)} + \frac{\mathbf{B}^2}{2}\right]\right\} = 0, \quad (3.62)$$

where ω_c denotes now the transverse polariton frequency $\omega_c \simeq \mathrm{Re}[\Omega_{\alpha,m}]$ for the homogeneous medium considered here. In this formula, the imaginary part of $\tilde{\varepsilon}(\omega_c)$ is systematically neglected in agreement with the reasoning discussed for instance in Ref. [90]. Alternatively, Eq. (3.62) could be rewritten using the time average $\langle \mathbf{B}^2\rangle \simeq 2\mathbf{E}_\perp^{(-)}\mathbf{E}_\perp^{(+)}$ in order to get the classical Brillouin formula for the electric energy density in the medium but this will not be useful here. Moreover, we introduce the mode operators

$$E^{(+)}_{\alpha,j,m}(t) = \int_{\delta\omega_{\alpha,m}} d\omega \frac{\omega^2}{\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}} f^{(0)}_{\omega,\alpha,j}(t), \tag{3.63}$$

where $\delta\omega_{\alpha,m}$ is a frequency window centered on the polariton pulsation $\omega_c \simeq \mathrm{Re}[\Omega_{\alpha,m}] := \Omega'_{\alpha,m}$. Now, if we suppose that the electromagnetic field is given by a sum of such transverse modes (without overlap of the frequency domains $\delta\omega_{\alpha,m}$), then Eq. (3.62) reads as

$$\frac{\delta}{\delta t}H(t) = \frac{\delta}{\delta t}\left(\sum_{\alpha,j,m}\left\{\frac{d[\Omega'_{\alpha,m}\tilde{\varepsilon}(\Omega'_{\alpha,m})]}{d\Omega'_{\alpha,m}} + \frac{\omega_\alpha^2}{{\Omega'_{\alpha,m}}^2}\right\}E^{(-)}_{\alpha,j,m}E^{(+)}_{\alpha,j,m}\right) = 0, \tag{3.64}$$

where the contribution $\frac{\omega_\alpha^2}{{\Omega'_{\alpha,m}}^2}$ arises from a modal expansion of the magnetic field and from using the resonance condition in the numerator of Eq. (3.32) (which involves $\omega\omega_\alpha \simeq \omega^2\omega_\alpha/\Omega_{\alpha,m}$).

What is also fundamental here is that we have the commutators (the derivation in the complex plane in given in Appendix F)

$$[E^{(+)}_{\alpha,j,m}(t), E^{(-)}_{\beta,l,n}(t)] = \delta_{\alpha,\beta}\delta_{j,l}\delta_{m,n}\frac{\hbar\Omega_{\alpha,m}}{2}\frac{d\Omega^2_{\alpha,m}}{d\omega_\alpha^2} \tag{3.65}$$

and $[E^{(+)}_{\alpha,j,m}(t), E^{(+)}_{\beta,l,n}(t)] = [E^{(-)}_{\alpha,j,m}(t), E^{(-)}_{\beta,l,n}(t)] = 0$. These relations imply the existence of effective rising and lowering operators $f^{(+)}_{\alpha,j,m}(t)$ for polaritons defined by $E^{(+)}_{\alpha,j,m}(t) = \sqrt{(\frac{\hbar\Omega_{\alpha,m}}{2}\frac{d\Omega^2_{\alpha,m}}{d\omega_\alpha^2})}f_{\alpha,j,m}(t)$.

These relations were phenomenologically obtained by Milonni [7,9] after quantizing Brillouin's energy formula. Here, we justify this result from the ground using the Huttner-Barnett formalism. Importantly, after defining the optical index of the polariton mode $n_{\alpha,m} \simeq \sqrt{\tilde{\varepsilon}(\Omega'_{\alpha,m})}$, we can rewrite $\frac{d[\Omega'_{\alpha,m}\tilde{\varepsilon}(\Omega'_{\alpha,m})]}{d\Omega'_{\alpha,m}} + \frac{\omega_\alpha^2}{{\Omega'_{\alpha,m}}^2}$ as $2n_{\alpha,m}c/v_g(\Omega'_{\alpha,m})$ where $v_g(\Omega'_{\alpha,m})$ is the group velocity of the mode defined by $d\Omega'_{\alpha,m}/dk_\alpha$. This allows us to rewrite the operators as $E^{(+)}_{\alpha,j,m}(t) = \sqrt{[\frac{\hbar\Omega_{\alpha,m}}{2}\frac{v_g(\Omega'_{\alpha,m})}{n_{\alpha,m}c}]}f_{\alpha,j,m}(t)$ [since $\frac{v_g(\Omega'_{\alpha,m})}{n_{\alpha,m}c} = \frac{d\Omega^2_{\alpha,m}}{d\omega_\alpha^2}$] and finally to have

$$\frac{\delta}{\delta t}H(t) = \frac{\delta}{\delta t}\left(\sum_{\alpha,j,m}\hbar\Omega_{\alpha,m}f^\dagger_{\alpha,j,m}f_{\alpha,j,m}\right) = 0. \tag{3.66}$$

The total energy is thus defined as $H(t) = \sum_{\alpha,j,m}\hbar\Omega_{\alpha,m}f^\dagger_{\alpha,j,m}f_{\alpha,j,m}$ which is a constant of motion defined up to an arbitrary additive constant. This formula involves only the transverse modes so that actually it gives the energy $H_\perp(t)$ associated with the transverse polariton modes in weekly dissipative medium and represents a generalization of Hopfield-Fano results as an effective but approximative model.

Few remarks are here necessary. First, the model proposed here relies on the assumption that the field is a sum of wave packets spectrally nonoverlapping. This hypothesis, which was also made by Garrison and Chio [9], was then called the "quasimultimonochromatic" approximation. This assumption is certainly not necessary since Milonni's model includes as a limit the rigorous Hopfield-Fano model [23,24] which does not rely on such an assumption. In order to justify further Milonni's approach [7] and relax the hypothesis made, it is enough to observe first that in Eq. (3.60) the approximation $\widetilde{\mathbf{D}}(\omega) \simeq \tilde{\varepsilon}(\omega)\widetilde{\mathbf{E}}_\perp(\omega)$ is quite robust even if the field is spectrally very broad. Indeed, since losses are here supposed to be very weak, the resonance will practically cancel out if ω differs significantly of a value where the condition $\omega_\alpha^2 = \tilde{\varepsilon}(\omega)\omega^2$ occurs. Second, if we insert formally Eqs. (3.60) and (3.61) with the previous assumption into Eq. (3.62), then instead of the term $\frac{\delta}{\delta t}[\int d^3\mathbf{x}\frac{d\omega_c\tilde{\varepsilon}(\omega_c)}{d\omega_c}\mathbf{E}_\perp^{(-)}\mathbf{E}_\perp^{(+)}]$ in Eq. (3.62) we get a term $\frac{\delta}{\delta t}[\int_0^{+\infty}d\omega\int_0^{+\infty}d\omega'\int d^3\mathbf{x}\frac{d\omega\tilde{\varepsilon}^*(\omega)}{d\omega}\widetilde{\mathbf{E}_\perp}(\omega)\widetilde{\mathbf{E}_\perp}^*(\omega')e^{i(\omega'-\omega)t}]$. This contribution is in general more complicated because $\frac{d\omega\tilde{\varepsilon}^*(\omega)}{d\omega}$ depends on ω. However, using explicitly Eqs. (3.60) and (3.61) and especially the Fourier expansion in plane waves, we see that for the specific fields considered here Eq. (3.64) still holds. This means that we can again introduce polariton operators $E^{(+)}_{\beta,l,n}(t)$ and $E^{(-)}_{\beta,l,n}(t)$ defined by Eq. (3.63). As previously, these operators depend on a frequency window $\delta\omega_{\alpha,m}$ and here these are introduced quite formally for taking into account the fact that the resonance $\frac{1}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}$ is extremely peaked near the different polariton frequencies $\Omega_{\alpha,m}$. A product like $\frac{1}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}\frac{1}{\omega_\alpha^2-\tilde{\varepsilon}^*(\omega')\omega'^2}$ occurring in the integration will thus not contribute unless the frequencies ω' and ω are in a given window $\delta\omega_{\alpha,m}$. Equation (3.64) thus results as a very good practical approximation.

Remarkably, as observed in Eq. (3.65) (and explained in Appendix E), the commutator does not depend explicitly on the size of these windows (which are only supposed to be small compared to the separation between the different mode frequencies and large enough to include the resonance peaks as explained in Appendix E). Therefore, this allows us to renormalize these operators as before by introducing the same rising and lowering polariton operators $f_{\alpha,j,m}$ such as Eq. (3.66) and $H_\perp(t) = \sum_{\alpha,j,m}\hbar\Omega_{\alpha,m}f^\dagger_{\alpha,j,m}f_{\alpha,j,m}$ hold identically. We thus have completed the justification of the Milonni's approach for dielectric medium with weak absorption [7].

Another remark concerns the longitudinal electric field which was omitted here since it does not play an active role in pulse propagation through the medium. We have indeed $\int d^3\mathbf{x}\,\mathbf{E}\cdot\partial_t\mathbf{D} = \int d^3\mathbf{x}\,\mathbf{E}_\perp\cdot\partial_t\mathbf{D}$ so that the reasoning was only done on the transverse modes. However, this was not necessary and one could have kept the longitudinal electric field all along the reasoning. Since the transverse part is a constant as we showed before, this should be the case for the longitudinal part as well since $H(t) - H_\perp(t)$ is also an integral of motion. Now, a reasoning similar to the previous one for transverse waves will lead to the Brillouin formula for the longitudinal electric energy:

$$\frac{\delta}{\delta t}H_{\parallel}(t) = \frac{\delta}{\delta t}\int d^3\mathbf{x}\int d^3\mathbf{x}\,\mathbf{E}_{\parallel}\cdot\partial_t\mathbf{D}$$
$$\simeq \frac{\delta}{\delta t}\left\{\sum_{\alpha,m}\frac{d[\Omega'_{\alpha,m}\tilde{\varepsilon}(\Omega'_{\alpha,m})]}{d\Omega'_{\alpha,m}}E^{(-)}_{\alpha,m,\parallel}E^{(+)}_{\alpha,m,\parallel}\right\} = 0, \tag{3.67}$$

where the longitudinal polariton modes are defined by

$$E^{(+)}_{\alpha,m}(t) = \int_{\delta\omega_{\alpha,m}} d\omega\frac{-1}{\tilde{\varepsilon}(\omega)}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}}f^{(0)}_{\omega,\alpha,\parallel}(t). \tag{3.68}$$

In this formalism, the longitudinal polariton frequencies $\Omega'_{\alpha,m}$ are the solutions of $\tilde{\varepsilon}'(\Omega'_{\alpha,m}) \simeq 0$ (where losses are again supposed to be weak). The commutator can be defined using a method equivalent to Eq. (3.65) and we get

$$[E^{(+)}_{\alpha,m,\parallel}(t),E^{(-)}_{\beta,n,\parallel}(t)] = \delta_{\alpha,\beta}\delta_{m,n}\frac{\hbar}{|M_{\alpha,m}|} \tag{3.69}$$

with $M_{\alpha,m} = \frac{d\tilde{\varepsilon}'(\omega)}{d\omega}|_{\Omega'_{\alpha,m}}$. After defining the lowering polariton operator as $E^{(+)}_{\alpha,m,\parallel}(t) = f_{\alpha,m,\parallel}(t)\sqrt{(\frac{\hbar}{|M_{\alpha,m}|})}$ we thus obtain

$$\frac{\delta}{\delta t}H_{\parallel}(t) \simeq \frac{\delta}{\delta t}\left(\sum_{\alpha,m}\hbar\Omega'_{\alpha,m}f^{\dagger}_{\alpha,m,\parallel}f_{\alpha,m,\parallel}\right) = 0 \tag{3.70}$$

as it should be. Milonni's approach [7] leads therefore to an effective justification of longitudinal polaritons as well and this includes the Hopfield-Fano [24] model as a limiting case when losses are vanishing outside infinitely narrow absorption bands.

A final important remark should be done since it concerns the general significance of the scattered field (s) in the lossless limit. Indeed, we see from Eq. (3.63) that the transverse mode operators $E^{(+)}_{\alpha,j,m}(t)$ and $E^{(+)}_{\alpha,j,m}(t)$ rigorously vanish in the limit $\sigma_\omega \to 0$ [this is not true for longitudinal operators (3.68) which are physically linked to bound and Coulombian fields]. In agreement with Sec. III C, we thus conclude that in the vacuum limit one should consider the (0) fields as the only surviving contribution. However, we also see that for all practical needs, if the losses are weak but not equal to zero, then by imposing $t_0 \to -\infty$ the (0) terms should cancel and only will survive a scattered term which will formally looks as a free photon in a bulk medium with optical index $n_\omega \simeq \sqrt{\varepsilon_\omega}$. Therefore, we justify the formal canonical quantization procedure used by Milonni and others [6–9,44,101,102] which reduces to the historical quantization methods in the (quasi-)nondispersive limit [1,3]. However, this can only be considered as an approximation and therefore the original claim presented in Ref. [33] that the scattered field (s) is sufficient for justifying the exact limit $\sigma_\omega \to 0$ without the (0) term was actually unfounded. As we will see, this will become especially relevant when we will generalize the Langevin-noise approach to an inhomogeneous medium.

G. Energy conservation puzzle and the interpretation of the Hamiltonian for a homogeneous dielectric medium

The central issue in this work is to interpret the physical meaning of quantized polariton modes in the general Huttner-Barnett framework of Sec. II and this will go far beyond the limiting Hopfield-Fano [23,24] or Milonni approaches [7] which are valid in restricted conditions when losses and/or dispersion are weak enough. For the present purpose we will focus on the homogeneous medium case (the most general inhomogeneous medium case is analyzed in the next section). It is fundamental to compare the mode structure of Eqs. (3.26), (3.27), (3.30), and (3.32) on the one side and the mode structure of Eqs. (3.14), (3.15), and (3.16) on the other side which are associated, respectively, with the free modes "(0)" and the scattered modes "(s)". The (0) modes are the eigenstates of the classical propagation problem when we can cancel the fluctuating term $\mathbf{P}^{(0)}$. This is, however, not allowed in QED since we are now considering operators in the Hilbert space and one cannot omit these terms without breaking unitarity. Inversely, the scattered modes are the modes which were considered by Huttner and Barnett [17]. Moreover, only these (s) modes survive here if the initial time t_0 is sent into the remote past, i.e., if $t_0 \to -\infty$. For all operational needs, it is therefore justified to omit altogether the (0) mode contribution in the homogeneous medium case. Clearly, this was the choice made by Gruner and Welsch [34] and later by more or less all authors working on the subject (see, however, Refs. [79,80]) which accepted this rule even for nonhomogeneous media. If we accept this axiom, then the Hamiltonian of the problem seems to reduce to $H^{(0)}_M$, i.e., in the homogeneous medium case, to

$$H^{(0)}_M(t) = \int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}^{\dagger(0)}_{\omega}(\mathbf{x},t)\mathbf{f}^{(0)}_{\omega}(\mathbf{x},t)$$
$$= \sum_{\alpha,j}\int_0^{+\infty} d\omega\hbar\omega f^{\dagger(0)}_{\omega,\alpha,j}(t),f^{(0)}_{\omega,\alpha,j}(t)$$
$$+\sum_{\alpha}\int_0^{+\infty} d\omega\hbar\omega f^{\dagger(0)}_{\omega,\alpha,\parallel}(t),f^{(0)}_{\omega,\alpha,\parallel}(t) \tag{3.71}$$

which depends only on the fluctuating operators $f^{(0)}_{\omega,\alpha,\parallel}(t)$, $f^{\dagger(0)}_{\omega,\alpha,\parallel}(t)$ in agreement with the Langevin force and noise approach of Gruner and Welsch [34].

Now, there is apparently a paradox: the complete Hamiltonian of the system is in agreement with Eq. (2.13) given by

$$H(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t)^2 + \mathbf{E}(\mathbf{x},t)^2}{2} :$$
$$+\int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}^{\dagger}_{\omega}(\mathbf{x},t)\mathbf{f}_{\omega}(\mathbf{x},t). \tag{3.72}$$

Can we show that $H(t)$ is actually equivalent to $H^{(0)}_M(t)$? This is indeed the case for all practical purposes at least for the homogeneous medium case treated by Huttner and Barnett [17] that we analyzed in details before. To see that, remember that $H(t)$ is actually a constant of motion, therefore, we should have $H(t) = H(t_0)$. This equality reads as also $H(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2+\mathbf{E}(\mathbf{x},t_0)^2}{2} : +H_M(t_0)$. Now, the central point here is to use the boundary condition at time

t_0 which implies $\mathbf{f}_\omega(\mathbf{x},t_0) = \mathbf{f}_\omega^{(0)}(\mathbf{x},t_0)$. Furthermore, since the time evolution of $\mathbf{f}_\omega^{(0)}$ is harmonic we have $\mathbf{f}_\omega^{\dagger(0)}(\mathbf{x},t)\mathbf{f}_\omega^{(0)}(\mathbf{x},t) = \mathbf{f}_\omega^{\dagger(0)}(\mathbf{x},t_0)\mathbf{f}_\omega^{(0)}(\mathbf{x},t_0)$. Altogether, these relations imply

$$H_M(t_0) = H_M^{(0)}(t_0) = H_M^{(0)}(t) \tag{3.73}$$

so that we have

$$H(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2 + \mathbf{E}(\mathbf{x},t_0)^2}{2} : + H_M^{(0)}(t). \tag{3.74}$$

In other words, we get a description in which the Fock number states associated with the fluctuating operators $\mathbf{f}_\omega^{(0)}(\mathbf{x},t)$ or, equivalently, $f_{\omega,\alpha,j}^{(0)}(t)$, $f_{\omega,\alpha,\|}^{(0)}(t)$ diagonalize not the full Hamiltonian but only a part that we noted $H_M^{(0)}(t)$. However, this is not problematic since the remaining term

$$H_{\text{rem}}(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2 + \mathbf{E}(\mathbf{x},t_0)^2}{2} : \tag{3.75}$$

is also clearly by definition a constant of motion since it only depends on fields at time t_0. How can we interpret this constant of motion? We can clearly rewrite it as $H_{\text{rem}}(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2 + [\mathbf{D}(\mathbf{x},t_0) - \mathbf{P}(\mathbf{x},t_0)]^2}{2} :$. Therefore, in the $\mathbf{F}$ potential vector formalism defined in Sec. II, this constant depends on both $c_{\alpha,j}(t_0)$ and $c_{\alpha,j}^\dagger(t_0)$, i.e., lowering and rising operators associated with the transverse $\mathbf{D}$ and $\mathbf{B}$ fields, and it also depends on the operators $\mathbf{f}_\omega^\dagger(\mathbf{x},t_0)$, $\mathbf{f}_\omega(\mathbf{x},t_0)$ which are associated with the fluctuating dipole distribution at the initial time t_0. Furthermore, since we have also $\mathbf{D}^{(s)}(t_0) = 0$, $\mathbf{B}^{(s)}(t_0) = 0$, $\mathbf{E}^{(s)}(t_0) = -\mathbf{P}^{(0)}(t_0)$ and since $\mathbf{D}(t_0) = \mathbf{D}^{(0)}(t_0) = \mathbf{E}^{(0)}(t_0)$, $\mathbf{B}(t_0) = \mathbf{B}^{(0)}(t_0)$ we can alternatively write

$$H_{\text{rem}}(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}^{(0)}(\mathbf{x},t_0)^2}{2} + \frac{[\mathbf{D}^{(0)}(\mathbf{x},t_0) - \mathbf{P}^{(0)}(\mathbf{x},t_0)]^2}{2} : . \tag{3.76}$$

This means that the remaining term $H_{\text{rem}}(t)$ depends on the knowledge of $\mathbf{D}^{(0)}(t_0)$ and $\mathbf{B}^{(0)}(t_0)$ electromagnetic free field. Since in the limit $t_0 \to -\infty$ these (0) electromagnetic terms vanish at any finite time t, this would justify to consider $H_{\text{rem}}(t)$ as an inoperative constant. However, we have also the contribution of $\mathbf{P}^{(0)}(t_0)$ which plays a fundamental role in the determination of $\mathbf{D}^{(s)}(t)$, $\mathbf{B}^{(s)}(t)$, and $\mathbf{E}^{(s)}(t)$ at the finite time t. Therefore, while $H_{\text{rem}}(t)$ is a constant of motion, it nevertheless contains quantities which will affect the evolution of the surviving (s) fields at time t. It is for this reason that we can say that for "all practical purposes only" $H_{\text{rem}}(t)$ is unnecessary and that $H_M^{(0)}(t)$ is sufficient for describing the energy problem.

There are, however, many remarks to be done here concerning this analysis. First, while the Hamiltonian $H_M^{(0)}(t)$ gives a good view of the energy up to an additive inoperative constant, it is the full Hamiltonian which is necessary for deriving the equations of motion from Hamilton's equations or equivalently from Heisenberg's evolution like $i\hbar\frac{d}{dt}A(t) = [A(t),H(t)]$. It is also only with $H(t)$ that time symmetry is fully preserved. In particular, do not forget that in deriving the Fano-Hopfield [23,24] formalism we introduced [see Eq. (3.43)] the evolution $\nabla \times \nabla \times \widetilde{\mathbf{E}}(\Omega) - \frac{\Omega^2}{c^2}\widetilde{\varepsilon}(\Omega)\widetilde{\mathbf{E}}(\Omega) = \frac{\Omega^2}{c^2}\widetilde{\mathbf{P}}^{(in)}(\Omega)$ which depends on the causal "in" field $\widetilde{\mathbf{P}}^{(in)}(\Omega)$ and on the causal permittivity $\widetilde{\varepsilon}(\Omega)$. Since the knowledge of $\mathbf{P}^{(in)}(t)$ is equivalent to $\mathbf{P}^{(0)}(t)$ in the limit $t_0 \to -\infty$, the (forward) Laplace transform method is leading to the same result that the usual Fano method and this fits as well with the Gruner and Welsch Langevin's equations [34]. However, instead of Eq. (3.43) we could equivalently use the anticausal equation

$$\nabla \times \nabla \times \widetilde{\mathbf{E}}(\Omega) - \frac{\Omega^2}{c^2}\widetilde{\varepsilon}^*(\Omega)\widetilde{\mathbf{E}}(\Omega) = \frac{\Omega^2}{c^2}\widetilde{\mathbf{P}}^{(out)}(\Omega), \tag{3.77}$$

where $\widetilde{\mathbf{P}}^{(out)}(\Omega)$ replaces $\widetilde{\mathbf{P}}^{(out)}(\Omega)$ and where the causal permittivity $\widetilde{\varepsilon}(\Omega)$ becomes now $\widetilde{\varepsilon}^*(\Omega)$ which is associated with amplification instead of the usual dissipation. The Green integral equation now becomes

$$\widetilde{\mathbf{E}}(\mathbf{x},\Omega) = \frac{\Omega^2}{c^2}\int d^3\mathbf{x}'\mathbf{G}_{\chi_T}(\mathbf{x},\mathbf{x}',\Omega) \cdot \widetilde{\mathbf{P}}^{(out)}(\mathbf{x}',\Omega), \tag{3.78}$$

where $\chi_T(t)$ replaces $\chi(t)$ [see Eq. (3.34)] and is associated with the anticausal dynamics which is connected to the time-reversed evolution. Both formalisms developed with either "in" or "out" fields are completely equivalent, but this shows that we have the freedom to express the scattered field in terms of $\mathbf{f}_\omega^{(in)}(\mathbf{x},t)$ and $\mathbf{f}_\omega^{(in)\dagger}(\mathbf{x},t)$ or in terms of $\mathbf{f}_\omega^{(out)}(\mathbf{x},t)$ and $\mathbf{f}_\omega^{(out)\dagger}(\mathbf{x},t)$. Since there are in general no fully propagative "free" electromagnetic modes (if we exclude the lossless medium limit considered by Hopfield and Fano [23,24]), then the surviving fields at finite time t obtained either with $t_0 \to -\infty$ or $t_f \to +\infty$ correspond to decaying or growing waves in agreement with the results discussed for the Laplace transform methods. The full Hamiltonian $H(t)$ is thus expressed equivalently either as

$$H(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2 + \mathbf{E}(\mathbf{x},t_0)^2}{2} : + \int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{(in)\dagger}(\mathbf{x},t)\mathbf{f}_\omega^{(in)}(\mathbf{x},t) \tag{3.79}$$

[with $\mathbf{f}_\omega^{(in)}(\mathbf{x},t) := \mathbf{f}_\omega^{(0)}(\mathbf{x},t) = \mathbf{f}_\omega(\mathbf{x},t_0)e^{-i\omega(t-t_0)}$ the fluctuating field defined in Sec. II] or as

$$H(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_f)^2 + \mathbf{E}(\mathbf{x},t_f)^2}{2} : + \int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{(out)\dagger}(\mathbf{x},t)\mathbf{f}_\omega^{(out)}(\mathbf{x},t) \tag{3.80}$$

with $\mathbf{f}_\omega^{(out)}(\mathbf{x},t) = \mathbf{f}_\omega(\mathbf{x},t_f)e^{-i\omega(t-t_f)}$ the fluctuating field using using a final boundary condition at time t_f. In the limits $t_0 \to -\infty$, $t_f \to +\infty$, this leads to

$$H(t) = H_{in,\text{rem}}(t) + \int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{(in)\dagger}(\mathbf{x},t)\mathbf{f}_\omega^{(in)}(\mathbf{x},t) \tag{3.81}$$

or

$$H(t) = H_{out,\text{rem}}(t) + \int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{(out)\dagger}(\mathbf{x},t)\mathbf{f}_\omega^{(out)}(\mathbf{x},t), \tag{3.82}$$

where the remaining terms [see Eq. (3.75)] $H_{in,\mathrm{rem}}(t)$ and $H_{out,\mathrm{rem}}(t)$ are two different integrals of motion. Therefore, it shows that the representation chosen by Gruner and Welsch in Ref. [34] is not univocal and that one could reformulate all the theory in terms of $\widetilde{\mathbf{P}}^{(out)}(\Omega)$ instead of $\widetilde{\mathbf{P}}^{(in)}(\Omega)$ to respect time symmetry. Furthermore, we emphasize that while the two Hamiltonians $\int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{(0)\dagger}(\mathbf{x},t)\mathbf{f}_\omega^{(0)}(\mathbf{x},t)$ and $\int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{(f)\dagger}(\mathbf{x},t)\mathbf{f}_\omega^{(f)}(\mathbf{x},t)$ have formally the same mathematical structure, they are not associated with the same physical electromagnetic fields since Eqs. (3.45) and (3.78) correspond, respectively, to decaying and growing radiated fields. Causality therefore requires to make a choice between two different representations. It is only the choice on a boundary condition in the remote past or future together with thermodynamical considerations which allow us to favor the decaying regime given by Eq. (3.45).

A different but related point to be discussed here concerns the Hopfield-Fano limit [23,24] for which the general conductivity like $\sigma(\Omega) = \sum_n \frac{\pi\omega_{p,n}^2}{2}[\delta(\Omega-\omega_{0,n})+\delta(\Omega+\omega_0)]$ leads to quasilossless permittivity [see Eq. (3.56)]. In this regime, we found that an exact diagonalization procedure can be handled out leading to genuine transverse and longitudinal polaritons. These exceptions also fit with the time-symmetry considerations discussed previously since the quasiabsence of absorption makes the problem much more time symmetrical than in the cases where the only viable representations involve either $\widetilde{\mathbf{P}}^{(out)}(\Omega)$ or $\widetilde{\mathbf{P}}^{(in)}(\Omega)$. Of course, the Hopfield-Fano model is an idealization which, in the context of the Huttner-Barnett framework, gets a clear physical interpretation only as an approximation for low-loss media, as explained in Sec. III G.

This leads us to a new problem, which is certainly the most important in this work: Considering the vacuum limit $\chi(t)\to 0$ discussed after Eq. (3.16) we saw that only the (0) electromagnetic modes survive in this regime and that the (s) modes are killed together with $\mathbf{P}^{(0)}(t)\to 0$. These vacuum modes are completely decoupled from the undamped mechanical oscillator motion $\mathbf{X}_\omega(t)=\mathbf{X}_\omega^{(0)}(t)$. In this limit, time symmetry is of course respected and we see that the full set of eigenmodes diagonalizing the Hamiltonian corresponds to the uncoupled free photons and free mechanical oscillator motions. Another way to see that is to use again the Fourier formalism instead of the Laplace transform method. From Eq. (3.43) we see that in the vacuum limit it is not the scattered field defined by Eq. (3.45) which survives [since $\mathbf{P}^{(0)}(t)\to 0$] but an additional term corresponding to free-space photon modes. While this should be clear after our discussion this point has tremendous consequences if we want to generalize properly the Huttner-Barnett approach to an inhomogeneous medium. In such a medium, the permittivity $\widetilde{\varepsilon}(\mathbf{x},\Omega)$ is position dependent. Now, generally speaking, in nanophotonics we consider problems where a dissipative object like a metal particle is confined in a finite region of space surrounded by vacuum. The susceptibility $\chi(\mathbf{x},\tau)$ therefore vanishes outside the object and we expect electromagnetic vacuum modes associated with free-space photon to play a important role in the final analysis. This should contrast with the Huttner-Barnett case for homogeneous medium which supposes an unphysical infinite dissipative medium supporting bulk polaritons or plasmons. The inhomogeneous polariton case will be treated in the next section.

IV. QUANTIZING POLARITONS IN AN INHOMOGENEOUS MEDIUM

A. The Green dyadic problem in the time domain

In order to deal with the most general situation of polaritons in inhomogeneous media, we need first to consider the formal separation between source fields and free-space photon mode. The separation used here is clearly different from the one developed in the previous section since we now consider on the one side as source term the total polarization $\mathbf{P}(\mathbf{x},t)$ [see Eq. (2.9)] which includes both the fluctuating term $\mathbf{P}^{(0)}(\mathbf{x},t)$ but also the induced polarization $\int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}(\mathbf{x},t-\tau)$ and on the other side as source-free terms some general photon mode solutions of Maxwell equations in vacuum. In order not to get confused with the previous notations, we now label (v) the vacuum modes and (d) the modes induced by the total dipolar distribution $\mathbf{P}(\mathbf{x},t)$.

We start with the second-order dynamical equation

$$\frac{1}{c^2}\partial_t^2\mathbf{F}(\mathbf{x},t)-\boldsymbol{\nabla}^2\mathbf{F}(\mathbf{x},t)-\boldsymbol{\nabla}\times\mathbf{P}(\mathbf{x},t)=0 \tag{4.1}$$

which can be solved using the method developed for Eq. (3.1). Indeed, by imposing $\chi=0$ in Eq. (3.1) and by replacing $\mathbf{P}^{(0)}$ by $\mathbf{P}$, $\mathbf{F}^{(0)}$ by $\mathbf{F}^{(v)}$, $\mathbf{F}^{(s)}$ by $\mathbf{F}^{(d)}$, and so on in the calculations of Sec. III we can easily obtain the formalism needed. Consider first the vacuum fields $\mathbf{F}^{(v)}$, $\mathbf{D}^{(v)}$, and $\mathbf{B}^{(v)}$. From Eqs. (3.14) and (3.15) in the limit $\chi(\tau)\to 0$ we get a plane-wave modal expansion for the free-photon field which we write in analogy with Eq. (3.17) as

$$\begin{aligned}
\mathbf{F}^{(v)}(\mathbf{x},t)&=\sum_{\alpha,j} ic\sqrt{\frac{\hbar}{2\omega_\alpha}}c_{\alpha,j}^{(v)}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})+\text{c.c.},\\
\mathbf{D}^{(v)}(\mathbf{x},t)&=\sum_{\alpha,j}-\sqrt{\frac{\hbar\omega_\alpha}{2}}c_{\alpha,j}^{(v)}(t)\hat{\mathbf{k}}_\alpha\times\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})+\text{c.c.},\\
\mathbf{B}^{(v)}(\mathbf{x},t)&=\sum_{\alpha,j}\sqrt{\frac{\hbar\omega_\alpha}{2}}c_{\alpha,j}^{(v)}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})+\text{c.c.}
\end{aligned} \tag{4.2}$$

with the modal expansion coefficients $c_{\alpha,j}^{(v)}(t)=c_{\alpha,j}(t_0)e^{-i\omega_\alpha(t-t_0)}$ showing the harmonic structure of the fields. Of course, this transverse vacuum field satisfies Maxwell's equations without source terms and in particular $\boldsymbol{\nabla}\times\mathbf{E}^{(v)}(\mathbf{x},t)=-\partial_t\mathbf{B}^{(v)}(\mathbf{x},t)/c$ with $\mathbf{E}^{(v)}(\mathbf{x},t)=\mathbf{D}^{(v)}(\mathbf{x},t)$.

We now study the scattered fields $\mathbf{E}^{(d)}$, $\mathbf{D}^{(d)}$, and $\mathbf{B}^{(d)}$. Like for the calculations presented in Sec. III for the homogeneous medium case, it appears convenient to use the Green dyadic formalism which is well adapted for nanophotonics studies, in particular for numerical computation of fields in complex dielectric environment where no obvious spatial symmetry is visible. The details are given in Appendix G. Here, we only need the expansion for the scattered electric field $\mathbf{E}^{(d)}=\mathbf{E}-\mathbf{E}^{(v)}=\mathbf{D}^{(d)}-\mathbf{P}$:

$$\begin{aligned}
\mathbf{E}^{(d)}(\mathbf{x},t)=&-\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p(t-t_0)}\\
&\times\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}(\mathbf{x}',p),
\end{aligned} \tag{4.3}$$

where we used the dyadic Green function for vacuum $\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)$ which is the solution of Eq. (3.20) for $\chi(\tau)=0$ [see Appendixes D and G and, in particular, Eqs. (D1) and (G3)]. In the time domain this can be written [see the derivation of Eq. (G5) in Appendix G] as

$$\mathbf{E}^{(d)}(\mathbf{x},t) = \int_0^{t-t_0} d\tau \int d^3\mathbf{x}'\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') \cdot \mathbf{P}(\mathbf{x}',t-\tau) - \mathbf{P}(\mathbf{x},t), \tag{4.4}$$

where $\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')$ is the inverse Laplace transform of $\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip)$ [see Eq. (D7)].

We emphasize here once again the fundamental role played by the boundary conditions at t_0. What we showed (see also Appendix G) is that at the initial time t_0 we have $\mathbf{D}^{(d)}(\mathbf{x},t_0)=0$ and thus $\mathbf{D}^{(v)}(\mathbf{x},t_0)=\mathbf{D}(\mathbf{x},t_0)$ which means, by definition of our vacuum modes, $\mathbf{E}^{(v)}(\mathbf{x},t_0)=\mathbf{D}(\mathbf{x},t_0)$. In other words, the electric field associated with vacuum modes equals the total displacement fields at the initial time. This is interesting since it also implies $\mathbf{E}(\mathbf{x},t_0)=\mathbf{E}^{(v)}(\mathbf{x},t_0)-\mathbf{P}(\mathbf{x},t_0)$ [which means $\mathbf{E}^{(d)}(\mathbf{x},t_0)=-\mathbf{P}(\mathbf{x},t_0)$]. This can be written after separation into transverse and longitudinal parts as

$$\begin{aligned}\mathbf{E}_\perp(\mathbf{x},t_0) &= \mathbf{E}^{(v)}(\mathbf{x},t_0)-\mathbf{P}_\perp(\mathbf{x},t_0)\\ &= \mathbf{D}^{(v)}(\mathbf{x},t_0)-\mathbf{P}_\perp(\mathbf{x},t_0),\\ \text{i.e., } \mathbf{E}_\perp^{(d)}(\mathbf{x},t_0) &= -\mathbf{P}_\perp(\mathbf{x},t_0)\end{aligned} \tag{4.5}$$

for the transverse (solenoidal or divergence-free) components and

$$\mathbf{E}_\parallel(\mathbf{x},t_0) = -\mathbf{P}_\parallel(\mathbf{x},t_0) \tag{4.6}$$

for the longitudinal (irrotational or curl-free) components. Equation (4.6) is well known in QED since it rigorously agrees with the definition of the longitudinal field obtained in usual Coulomb gauge using the **A** potential instead of **F**.

B. Separation between fluctuating and induced current: Macroscopic versus microscopic description

Until now, we did not specify the form of the dipole density $\mathbf{P}(\mathbf{x},t)$. The separation between source and free terms for the field was therefore analyzed from a microscopic perspective where the diffracted fields $\mathbf{E}^{(d)}(\mathbf{x},t)$ and $\mathbf{B}^{(d)}(\mathbf{x},t)$ were generated by the full microscopic current. In order to generalize the description given in Sec. III for the homogeneous medium, we will now use the separation (2.9) of $\mathbf{P}(\mathbf{x},t)$ into a fluctuating term $\mathbf{P}^{(0)}(\mathbf{x},t)$ and an induced contribution $\int_0^{t-t_0}\chi(\mathbf{x},\tau)d\tau\,\mathbf{E}(\mathbf{x},t-\tau)$ of essentially classical origin. Using the Laplace transform we get $\overline{\mathbf{P}'}(\mathbf{x},p)=\overline{\mathbf{P}'}^{(0)}(\mathbf{x},p)+\overline{\chi}(\mathbf{x},p)\overline{\mathbf{E}'}(\mathbf{x},p)$. Now, from the previous section we have therefore for the Laplace transform of the electric field the following Lippman-Schwinger integral equation:

$$\overline{\mathbf{E}'}(\mathbf{x},p) = \overline{\mathbf{D}'}^{(v)}(\mathbf{x},p) - \int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip) \cdot[\bar{\chi}(\mathbf{x}',p)\overline{\mathbf{E}'}(\mathbf{x}',p)+\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)]. \tag{4.7}$$

In order to get a meaningful separation of the total field, we here define

$$\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p) = \overline{\mathbf{D}'}^{(v)}(\mathbf{x},p) - \int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip) \cdot\bar{\chi}(\mathbf{x}',p)\overline{\mathbf{E}'}^{(0)}(\mathbf{x}',p) \tag{4.8}$$

and

$$\mathbf{G}_\chi(\mathbf{x},\mathbf{x}'',ip) = \mathbf{G}_v(\mathbf{x},\mathbf{x}'',ip) - \int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip) \cdot\bar{\chi}(\mathbf{x}',p)\mathbf{G}_\chi(\mathbf{x}',\mathbf{x}'',ip). \tag{4.9}$$

We have clearly

$$\nabla\times\nabla\times\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)+\frac{p^2}{c^2}[1+\bar{\chi}(\mathbf{x}',p)]\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) = \boldsymbol{\delta}(\mathbf{x}-\mathbf{x}') \tag{4.10}$$

and

$$\nabla\times\nabla\times\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)+\frac{p^2}{c^2}[1+\bar{\chi}(\mathbf{x}',p)]\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)=0. \tag{4.11}$$

This allows us to interpret $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)$ as the Green function of the inhomogeneous dielectric medium while $\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)$ is a free solution of Maxwell's equation in the dielectric medium in absence of the fluctuating source $\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)$. By direct replacement of Eqs. (4.8) and (4.9) into (4.7), one gets

$$\overline{\mathbf{E}'}(\mathbf{x},p)=\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)-\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p), \tag{4.12}$$

meaning that the total field can be seen as the sum of the free solution $\overline{\mathbf{E}'}^{(0)}(\mathbf{x},p)$ and of scattering contribution $\overline{\mathbf{E}'}^{(s)}(\mathbf{x},p)$ induced by the fluctuating source $\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)$. In Appendix H, we give the calculations for the electric field in the time domain by using the inverse Laplace transforms of $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)$. Here, the important point is that the boundary conditions at t_0 together with the field equations determine the full evolution and we have necessarily

$$\begin{aligned}&\mathbf{E}^{(0)}(\mathbf{x},t_0)=\mathbf{D}^{(0)}(\mathbf{x},t_0)=\mathbf{D}^{(v)}(\mathbf{x},t_0),\\ &\mathbf{D}^{(s)}(\mathbf{x},t_0)=\mathbf{E}^{(s)}(\mathbf{x},t_0)+\mathbf{P}^{(0)}(\mathbf{x},t_0)=0,\\ &\mathbf{B}^{(s)}(\mathbf{x},t_0)=0,\\ &\mathbf{B}^{(0)}(\mathbf{x},t_0)=\mathbf{B}^{(v)}(\mathbf{x},t_0)=\mathbf{B}(\mathbf{x},t_0).\end{aligned} \tag{4.13}$$

We point out that the description of the longitudinal field should be treated independently in this formalism since at any time t we have the constraint $\mathbf{E}_\parallel(\mathbf{x},t)=-\mathbf{P}_\parallel(\mathbf{x},t)$ which shows that fluctuating current and field are not independent. More precisely, if we insert the constraint $\mathbf{E}_\parallel(\mathbf{x},t)=-\mathbf{P}_\parallel(\mathbf{x},t)$ into the Lagrangian formalism developed in Sec. II, we get an effective Lagrangian density for the longitudinal field which reads as

$$\mathcal{L}_\parallel(\mathbf{x},t)=\int_0^{+\infty}d\omega\frac{[\partial_t\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)]^2-\omega^2\mathbf{X}_{\omega,\parallel}^2(\mathbf{x},t)}{2}-\frac{\mathbf{P}_\parallel^2(\mathbf{x},t)}{2}. \tag{4.14}$$

From Eq. (4.14) we get the Euler-Lagrange equation

$$\partial_t^2\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)+\omega^2\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)=-\sqrt{\frac{2\sigma_\omega(\mathbf{x})}{\pi}}\mathbf{P}_\parallel(\mathbf{x},t), \quad (4.15)$$

with $\mathbf{P}_\parallel(\mathbf{x},t)=\int_0^{+\infty}d\omega\sqrt{\frac{2\sigma_\omega(\mathbf{x})}{\pi}}\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)$ and which agrees with Eq. (2.7) if the constraint $\mathbf{E}_\parallel(\mathbf{x},t)=-\mathbf{P}_\parallel(\mathbf{x},t)$ is used. Now, the formal solution of Eq. (4.15) is obtained from Eq. (4.16):

$$\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)=\mathbf{X}^{(0)}_{\omega,\parallel}(\mathbf{x},t)-\sqrt{\frac{2\sigma_\omega(\mathbf{x})}{\pi}}\times\int_0^{t-t_0}d\tau\frac{\sin\omega\tau}{\omega}\mathbf{P}_\parallel(\mathbf{x},t-\tau) \quad (4.16)$$

and allows for a separation between a fluctuating term $\mathbf{X}^{(0)}_{\omega,\parallel}(\mathbf{x},t)$ and a source term $\mathbf{X}^{(s)}_{\omega,\parallel}(\mathbf{x},t)$. From this we naturally deduce

$$\mathbf{P}_\parallel(\mathbf{x},t)=\mathbf{P}^{(0)}_\parallel(\mathbf{x},t)-\int_0^{t-t_0}d\tau\,\chi(\mathbf{x},\tau)\mathbf{P}_\parallel(\mathbf{x},t-\tau). \quad (4.17)$$

Integral equations (4.16) or (4.17) could in principle be solved iteratively in order to find expressions $\mathbf{P}_\parallel$ and $\mathbf{X}_{\omega,\parallel}$ which are linear functionals of $\mathbf{P}^{(0)}_\parallel$ and $\mathbf{X}^{(0)}_{\omega,\parallel}$. Alternatively, this can be done self-consistently using the Laplace transform of Eq. (4.17) which reads as $\overline{\mathbf{P}'}_\parallel(\mathbf{x},p)=\overline{\mathbf{P}'}^{(0)}_\parallel(\mathbf{x},p)-\bar{\chi}(\mathbf{x},p)\overline{\mathbf{P}'}_\parallel(\mathbf{x},p)$ and leads to

$$\overline{\mathbf{P}'}_\parallel(\mathbf{x},p)=-\frac{\overline{\mathbf{P}'}^{(0)}_\parallel(\mathbf{x},p)}{1+\bar{\chi}(\mathbf{x},p)}. \quad (4.18)$$

In the time domain, we have thus

$$\mathbf{P}_\parallel(\mathbf{x},t)=\mathbf{P}^{(0)}_\parallel(\mathbf{x},t)-\int_0^{t-t_0}\chi_{\text{eff}}(\mathbf{x},\tau)\mathbf{P}^{(0)}_\parallel(\mathbf{x},t-\tau) \quad (4.19)$$

with the effective susceptibility defined as

$$\chi_{\text{eff}}(\mathbf{x},\tau)=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\frac{e^{p\tau}\bar{\chi}(\mathbf{x},p)}{1+\bar{\chi}(\mathbf{x},p)}. \quad (4.20)$$

We have equivalently for the effective susceptibility $\chi_{\text{eff}}(\mathbf{x},\tau)=\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}e^{-i\omega\tau}\frac{\tilde{\varepsilon}(\mathbf{x},\omega)-1}{\tilde{\varepsilon}(\mathbf{x},\omega)}$. After closing the contour in the complex plane we get $\chi_{\text{eff}}(\mathbf{x},\tau)=i\sum_m\frac{1}{\frac{\partial\tilde{\varepsilon}(\mathbf{x},\omega)}{\partial\omega}|_{\Omega_m}}e^{-i\Omega_m\tau}+$ c.c. where the sum is taken over the longitudinal modes solutions of $\tilde{\varepsilon}(\mathbf{x},\Omega_m)=0$ (with $\Omega''_m<0$ and $\Omega'_m>0$ by definition). The frequencies considered here are in general spatially dependent since the medium is inhomogeneous and is therefore very often difficult to find. In the limit of the homogeneous lossless medium, we obtain the Hopfield-Fano [23,24] model.

We emphasize that the present description of the polariton field contrast with the integral solution of Eq. (3.43) $\nabla\times\nabla\times\widetilde{\mathbf{E}}(\Omega)-\frac{\Omega^2}{c^2}\tilde{\varepsilon}(\Omega)\widetilde{\mathbf{E}}(\Omega)=\frac{\Omega^2}{c^2}\widetilde{\mathbf{P}}^{(in)}(\Omega)$ which was obtained for the homogeneous medium

$$\widetilde{\mathbf{E}}(\mathbf{x},\Omega)=\frac{\Omega^2}{c^2}\int d^3\mathbf{x}'\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',\Omega)\cdot\widetilde{\mathbf{P}}^{(in)}(\mathbf{x}',\Omega) \quad (4.21)$$

and which included only a scattering contribution (s) due to the cancellation of the (0) term for $t_0\to-\infty$. Here, we can not neglect or cancel the (0) mode solutions since in general the medium is not necessarily lossy at spatial infinity. This will be in particular the case for all scattering problems involving a localized system such as a metal or dielectric antenna supporting plasmon-polariton localized modes. However, mostly all studies, inspired by the success of the Huttner-Barnett model [17] for the homogeneous lossy medium, and following the Langevin-noise method proposed Gruner and Welsch [27–29,32,34], neglected or often even completely omitted the contribution of the (0) modes. Still, these (0) modes are crucial for preserving the unitarity of the full matter field dynamics and can not be rigorously omitted. Only in those cases where absorption is present at infinity can we omit the (0) modes.

To clarify this point further, consider a medium made of a spatially homogeneous background susceptibility $\bar{\chi}_1(p)$ and of a localized susceptibility $\bar{\chi}_2(\mathbf{x},p)$ such as $\bar{\chi}(\mathbf{x},p)\to 0$ at spatial infinity. The electromagnetic field propagating into the medium with total permittivity $\bar{\chi}(\mathbf{x},p)=\bar{\chi}_1(p)+\bar{\chi}_2(\mathbf{x},p)$ can be thus formally developed using the Lippman-Schwinger equation as

$$\begin{aligned}\overline{\mathbf{E}'}(\mathbf{x},p)&=\overline{\mathbf{E}'}^{(v)}(\mathbf{x},p)-\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot[\bar{\chi}(\mathbf{x}',p)\overline{\mathbf{E}'}(\mathbf{x},p)+\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)]\\&=\overline{\mathbf{E}'}^{(1)}(\mathbf{x},p)-\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}',ip)\cdot[\bar{\chi}_2(\mathbf{x}',p)\overline{\mathbf{E}'}(\mathbf{x},p)+\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)],\end{aligned} \quad (4.22)$$

where we have defined a background free field

$$\overline{\mathbf{E}'}^{(1)}(\mathbf{x},p)=\overline{\mathbf{D}'}^{(v)}(\mathbf{x},p)-\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\bar{\chi}_1(\mathbf{x}',p)\overline{\mathbf{E}'}^{(1)}(\mathbf{x}',p) \quad (4.23)$$

and a Green dyadic tensor for the background medium

$$\mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}'',ip)=\mathbf{G}_v(\mathbf{x},\mathbf{x}'',ip)-\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\bar{\chi}_1(\mathbf{x}',p)\mathbf{G}_{\chi_1}(\mathbf{x}',\mathbf{x}'',ip). \quad (4.24)$$

We have naturally $\nabla\times\nabla\times\mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}',ip)+\frac{p^2}{c^2}[1+\bar{\chi}_1(\mathbf{x}',p)]\mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}',ip)=\boldsymbol{\delta}(\mathbf{x}-\mathbf{x}')$ and similarly $\nabla\times\nabla\times\overline{\mathbf{E}'}^{(1)}(\mathbf{x},p)+\frac{p^2}{c^2}[1+\bar{\chi}_1(\mathbf{x}',p)]\overline{\mathbf{E}'}^{(1)}(\mathbf{x},p)=0$.

Importantly, in the time domain we can write using Appendix H

$$\begin{aligned}\mathbf{E}(\mathbf{x},t)=\mathbf{E}^{(1)}(\mathbf{x},t)&-\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{\partial_\tau^2\mathbf{U}_{\chi_1}(\tau,\mathbf{x},\mathbf{x}')}{c^2}\\&\times\left[\mathbf{P}^{(0)}(\mathbf{x}',t-\tau)+\int_0^{t-\tau}d\tau'\chi_2(\mathbf{x}',\tau')\mathbf{E}(\mathbf{x}',t-\tau-\tau')\right]\\&-\mathbf{P}^{(0)}(\mathbf{x},t)+\int_0^t d\tau\,\chi_2(\mathbf{x},\tau)\mathbf{E}(\mathbf{x},t-\tau),\end{aligned} \quad (4.25)$$

where $\mathbf{U}_\chi$ is the dyadic propagator defined in Eq. (H3). We can check that we have $\mathbf{E}(\mathbf{x},t_0)=\mathbf{E}^{(1)}(\mathbf{x},t_0)-\mathbf{P}^{(0)}(\mathbf{x},t_0)$. Moreover, since from Eq. (4.23) (written in the time domain) we have $\mathbf{E}^{(1)}(\mathbf{x},t_0)=\mathbf{D}^{(v)}(\mathbf{x},t_0)$ and since $\mathbf{P}^{(0)}(\mathbf{x},t_0)=\mathbf{P}(\mathbf{x},t_0)$, we deduce that at the initial time t_0, $\mathbf{D}(\mathbf{x},t_0)=\mathbf{E}(\mathbf{x},t_0)+$

$\mathbf{P}(\mathbf{x},t_0) = \mathbf{E}^{(1)}(\mathbf{x},t_0) = \mathbf{D}^{(v)}(\mathbf{x},t_0)$ as it should be to agree with the general formalism presented in Sec. IV A.

Now, since the background dissipative medium is not spatially bound, the $\mathbf{E}^{(1)}(\mathbf{x},t)$ field associated with damped modes will vanish if $t_0 \to -\infty$ as explained before. We could therefore be tempted [27–29,32,34] to eliminate from the start $\mathbf{E}^{(1)}(\mathbf{x},t)$ and thus get in the Laplace transform language the effective formula

$$\overline{\mathbf{E}'}(\mathbf{x},p) = -\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}',ip) \cdot [\bar{\chi}_2(\mathbf{x}',p)\overline{\mathbf{E}'}(\mathbf{x},p) + \overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p)] = -\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_{\chi}(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p) \quad (4.26)$$

with the total Green dyadic function

$$\mathbf{G}_{\chi}(\mathbf{x},\mathbf{x}'',ip) = \mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}'',ip) - \int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_{\chi_1}(\mathbf{x},\mathbf{x}',ip) \cdot \bar{\chi}_2(\mathbf{x}',p)\mathbf{G}_{\chi}(\mathbf{x}',\mathbf{x}'',ip). \quad (4.27)$$

obeying to Eq. (4.10) with the total permittivity $\bar{\chi}(\mathbf{x},p) = \bar{\chi}_1(p) + \bar{\chi}_2(\mathbf{x},p)$. It is straightforward to check that $\mathbf{G}_{\chi}(\mathbf{x},\mathbf{x}'',ip)$ satisfies also Eq. (4.9) so that it is the same Green function.

However, removing $\mathbf{E}^{(1)}(\mathbf{x},t)$ from the start in Eq. (4.25) would mean that the boundary conditions at the initial time t_0 have been obliviated since we should now necessarily have $\mathbf{D}(\mathbf{x},t_0) = \mathbf{E}(\mathbf{x},t_0) + \mathbf{P}(\mathbf{x},t_0) = \mathbf{E}^{(1)}(\mathbf{x},t_0) = 0$. This corresponds to a very specific boundary condition which is certainly allowed in classical physics [where we can set $c_{\alpha,j}(t_0) = 0$] but which in the quantum formalism means that we break the unitarity of the evolution. To say it differently, it means that in the Langevin-noise formalism [27–29,32,34], the photon field $\mathbf{F}$ is not anymore an independent canonical contribution to the evolution since all electromagnetic fields are induced by the material part. The Green formalism presented by Gruner and Welsch [34], and abundantly used since [51–69], represents therefore an alternative theory which rigorously speaking is not equivalent, contrary to the claim in Refs. [27–29,32], to the Lagrangian formalism discussed in Sec. II for the general Huttner-Barnett model [17]. Our analysis, as already explained in the Introduction, agrees with the general studies made in the 1970s and 1980s in QED [43,70–74] since one must include with an equal footing both the field and matter fluctuations in a self-consistent QED in order to preserve rigorously unitarity and causality.

C. Discussions concerning unitarity and Hamiltonians

Two issues are important to emphasize here. First, observe that in the limit where the background susceptibility $\chi_1(\mathbf{x},\tau)$ vanishes, then the term $\mathbf{E}^{(1)}(\mathbf{x},t) = \mathbf{D}^{(v)}(\mathbf{x},t)$ in general does not cancel at any time, and therefore the coupling to photonic modes can not be omitted even in practice from the evolution at finite time t. This is is particularly important in nanophotonics where an incident exiting photon field interact with a localized nanoantenna. It is therefore crucial to analyze further the impact of our findings on the quantum dynamics of polaritons in presence of sources such as quantum fluorescent emitters. This will the subject of a subsequent article.

The second issue concerns the Hamiltonian definition in the formalism. Indeed, the definition of the full system Hamiltonian $H(t)$ was previously given for the homogeneous medium case in Sec. III G. We showed [see Eq. (3.74)] that $H(t)$ is given by

$$H(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2 + \mathbf{E}(\mathbf{x},t_0)^2}{2} : + H_M^{(0)}(t), \quad (4.28)$$

where $H_M^{(0)}(t)$ is the material Hamiltonian defined in Eq. (3.71) and which depends only on the free mode operators $\mathbf{f}_\omega^{\dagger(0)}(\mathbf{x},t)$, $\mathbf{f}_\omega^{(0)}(\mathbf{x},t)$. This $H_M^{(0)}(t) = \int d^3\mathbf{x} \int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_\omega^{\dagger(0)}(\mathbf{x},t)\mathbf{f}_\omega^{(0)}(\mathbf{x},t)$ is the Hamiltonian considered by the noise-Langevin approach and the remaining term [see Eq. (3.75)] $H_{\text{rem}}(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2+\mathbf{E}(\mathbf{x},t_0)^2}{2} :$ is an additional constant of motion. This constant proved to be irrelevant for all practical purposes since the only surviving electromagnetic fields (i.e., if $t_0 \to -\infty$) are the induced (s) modes which are generated by the fluctuating dipole density $\mathbf{P}^{(0)}(\mathbf{x},t)$ (see, however, the different remarks concerning time symmetry at the end of Sec. III G). Now, for the inhomogeneous problem the complete reasoning leading to Eq. (4.28) is still rigorously valid. The main difference being that in general the constant of motion $H_{\text{rem}}(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2+\mathbf{E}(\mathbf{x},t_0)^2}{2} :$ is not irrelevant at all since the (0) electromagnetic modes are not in general vanishing even if $t_0 \to -\infty$.

In order to clarify this point, we should now physically interpret the term $H_{\text{rem}}(t)$. We first start with the less relevant term in optics: the longitudinal polariton. Indeed, the longitudinal field is here decoupled from the rest and evolves independently using the Lagrangian density $\mathcal{L}_\parallel(\mathbf{x},t)$ defined in Eq. (4.14) (this should not be necessarily true if the polaritons are coupled to external sources such as fluorescent emitters). We thus get the following Hamiltonian:

$$H_\parallel(t) = : \int d^3\mathbf{x}\left\{\int_0^{+\infty} d\omega\frac{[\partial_t\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)]^2}{2} + \frac{\omega^2\mathbf{X}_{\omega,\parallel}^2(\mathbf{x},t)}{2} + \frac{\mathbf{P}_\parallel^2(\mathbf{x},t)}{2}\right\} : . \quad (4.29)$$

$H_\parallel(t)$ is a constant of motion and can used (with the Hamiltonian formalism) to deduce the evolution equation [see Eq. (4.15)] and the solution Eq. (4.16). Since $H_\parallel(t)$ is a constant of motion, we have $H_\parallel(t) = H_\parallel(t_0)$ and from the form of the solution we obtain the equivalent formula

$$H_\parallel(t) = \int d^3\mathbf{x}\left[\int_0^{+\infty} d\omega\hbar\omega\mathbf{f}_{\omega,\parallel}^{\dagger(0)}(\mathbf{x},t)\mathbf{f}_{\omega,\parallel}^{(0)}(\mathbf{x},t) + \frac{:\mathbf{P}_\parallel^{(0)}(\mathbf{x},t_0)^2:}{2}\right], \quad (4.30)$$

where we have clearly by definition

$$H_{\text{rem},\parallel}(t) = \int d^3\mathbf{x}\frac{:\mathbf{P}_\parallel^{(0)}(\mathbf{x},t_0)^2:}{2} \quad (4.31)$$

and thus $H_\parallel(t) = H^{(0)}_{M,\parallel}(t) + H_{\text{rem},\parallel}(t)$ [with $H^{(0)}_{M,\parallel}(t) = \int d^3\mathbf{x} \int_0^{+\infty} d\omega\hbar\omega \mathbf{f}^{\dagger(0)}_{\omega,\parallel}(\mathbf{x},t)\mathbf{f}^{(0)}_{\omega,\parallel}(\mathbf{x},t)$]. While $H_{\text{rem},\parallel}(t)$ is a constant of motion, it is not irrelevant here since the equivalence of Eqs. (4.29) and (4.30) leads to the complete solution (4.16) for $\mathbf{X}_{\omega,\parallel}(\mathbf{x},t)$. Oppositely, taking $H^{(0)}_{M,\parallel}(t)$ and omitting $H_{\text{rem},\parallel}(t)$ would lead to the free solution $\mathbf{X}^{(0)}_{\omega,\parallel}(\mathbf{x},t)$ in contradiction with the dynamical law. This again stresses the importance of keeping all contributions in the evolution and Hamiltonian.

In order to analyze the transverse field Hamiltonian, we should comment further on the difference between the description using the $\mathbf{F}$ potential used in this work and the most traditional treatment using the $\mathbf{A}$ potential (in the Coulomb gauge). Indeed, by analogy with the separation between (d) and (v) modes discussed in Sec. VI A we can using the $\mathbf{A}$ potential vector representation get a separation between free-space modes $\mathbf{A}^{(v')}(\mathbf{x},t)$ and source field $\mathbf{A}^{(d')}(\mathbf{x},t)$. Here, we label these modes by an additional prime for reasons which will become clear below. First, the source field contribution (d') is given by

$$\mathbf{A}^{(d')}(\mathbf{x},t) = \frac{1}{c}\int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \Delta_v(\tau,|\mathbf{x}-\mathbf{x}'|)\cdot \mathbf{J}_\perp(\mathbf{x}',t-\tau) \tag{4.32}$$

and with by definition $\mathbf{J}_\perp(\mathbf{x},t) = \partial_t \mathbf{P}_\perp(\mathbf{x},t)$. Importantly, we have $\mathbf{A}^{(d')}(\mathbf{x},t_0) = 0$ meaning also $\mathbf{A}^{(v')}(\mathbf{x},t_0) = \mathbf{A}(\mathbf{x},t_0)$. The free-space modes (v') are easily obtained using a plane-wave expansion as

$$\mathbf{A}^{(v')}(\mathbf{x},t) = \sum_{\alpha,j} -ic\sqrt{\frac{\hbar}{2\omega_\alpha}} a^{(v)}_{\alpha,j}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{c.c.}, \tag{4.33}$$

where $a^{(v)}_{\alpha,j}(t) = a_{\alpha,j}(t_0)e^{-i\omega_\alpha(t-t_0)}$. Using the $\mathbf{A}$ potential, we therefore get for the free electromagnetic fields

$$\mathbf{B}^{(v')}(\mathbf{x},t) = \sum_{\alpha,j}\sqrt{\frac{\hbar\omega_\alpha}{2}} a^{(v)}_{\alpha,j}(t)\hat{\mathbf{k}}_\alpha \times \hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{c.c.},$$
$$\mathbf{E}^{(v')}_\perp(\mathbf{x},t) = \sum_{\alpha,j}\sqrt{\frac{\hbar\omega_\alpha}{2}} a^{(v)}_{\alpha,j}(t)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) + \text{c.c.} \tag{4.34}$$

These transverse fields satisfy Maxwell's equation in vacuum like the free fields given by Eq. (4.2) do as well. However, it should now be clear that these two sets of free fields given either by Eq. (4.34) or (4.2) are not equivalent. To see that, we must express the source field $\mathbf{E}^{(d')}(\mathbf{x},t)$ and $\mathbf{B}^{(d')}(\mathbf{x},t)$. The details are given in Appendix I. The importance is that while $\mathbf{E}_\parallel(\mathbf{x},t_0) = -\mathbf{P}_\parallel(\mathbf{x},t_0)$, which is Eq. (4.6), the transverse field $\mathbf{E}^{(d')}_\perp$ obtained in this representation is not identical to $\mathbf{E}^{(d)}_\perp$. In particular, this transverse field vanishes at t_0:

$$\mathbf{E}^{(d')}_\perp(\mathbf{x},t_0) = 0 \tag{4.35}$$

and therefore at this initial time it differs by an amount $-\mathbf{P}_\perp(\mathbf{x},t_0)$ from the scattered field given by Eq. (4.5). We thus get $\mathbf{E}(\mathbf{x},t_0) = \mathbf{E}^{(v')}_\perp(\mathbf{x},t_0) - \mathbf{P}_\parallel \mathbf{x},t_0)$. Importantly, by comparing Eqs. (4.5) and (4.35) we obtain a relation for the free-space modes in the two representations using either the $\mathbf{F}$ or $\mathbf{A}$ potential vectors:

$$\mathbf{E}^{(v')}_\perp(\mathbf{x},t_0) = \mathbf{E}^{(v)}_\perp(\mathbf{x},t_0) - \mathbf{P}_\perp(\mathbf{x},t_0). \tag{4.36}$$

This is reminiscent from the relation $\mathbf{E}_\perp(\mathbf{x},t_0) = \mathbf{D}(\mathbf{x},t_0) - \mathbf{P}_\perp(\mathbf{x},t_0)$. It shows that while the two fields $\mathbf{E}^{(v')}_\perp$ and $\mathbf{E}^{(v)}_\perp$ are solutions of the same Maxwell's equations in vacuum, they are not defined by the same initial conditions. We must therefore be extremely careful when dealing with the modes in order not to get confused with the solutions chosen. We also mention that the scattered magnetic field $\mathbf{B}^{(d')}$ given in Appendix I [see Eq. (I7)] and using the $\mathbf{A}$ potential also differs from the scatted magnetic field obtained using the $\mathbf{F}$ potential. Moreover, a comparison with the formulas obtained in Sec. VI A shows that $\mathbf{B}^{(d')}(\mathbf{x},t)$ differs from $\mathbf{B}^{(d)}(\mathbf{x},t)$ but that at time t_0 both vanish so that $\mathbf{B}^{(d')}(\mathbf{x},t_0) = \mathbf{B}^{(d)}(\mathbf{x},t_0) = 0$. It implies that $\mathbf{B}^{(v')}(\mathbf{x},t_0) = \mathbf{B}(\mathbf{x},t_0) = \mathbf{B}^{(v)}(\mathbf{x},t_0)$ so that while $\mathbf{B}^{(v')}(\mathbf{x},t)$ differs from $\mathbf{B}^{(v)}(\mathbf{x},t)$ for $t > t_0$, they become equal at the initial time t_0. Again, this stresses the difference between the representations based either on $\mathbf{F}$ or $\mathbf{A}$.

Now, this description using $\mathbf{A}$ leads to a clear interpretation of $H_{\text{rem},\perp}(t) = \int d^3\mathbf{x} : \frac{\mathbf{B}(\mathbf{x},t_0)^2 + \mathbf{E}_\perp(\mathbf{x},t_0)^2}{2} :$. Indeed, at time t_0 only the (v') solution survives for the transverse part of the field. Importantly, the set of free-space solutions (v') actually depends on lowering and rising operators $a^{(v')}_{\alpha,j}(t)$, $a^{\dagger(v')}_{\alpha,j}(t)$ defined such that $[a^{(v')}_{\alpha,j}(t), a^{\dagger(v')}_{\beta,k}(t)] = \delta_{\alpha,\beta}\delta_{j,k}$ and $a^{(v')}_{\alpha,j}(t) = a^{(v')}_{\alpha,j}(t_0)e^{-i\omega_\alpha(t-t_0)}$ with $a^{(v')}_{\alpha,j}(t_0) = a_{\alpha,j}(t_0)$. Therefore, by a reasoning similar to the one leading to Eq. (3.74) we deduce

$$H_\perp(t) = \int d^3\mathbf{x}\int_0^{+\infty} d\omega\hbar\omega \mathbf{f}^{\dagger(0)}_{\omega,\perp}(\mathbf{x},t)\mathbf{f}^{(0)}_{\omega,\perp}(\mathbf{x},t) + \sum_{\alpha,j}\hbar\omega_\alpha a^{\dagger(v')}_{\alpha,j}(t)a^{(v')}_{\alpha,j}(t). \tag{4.37}$$

We clearly here get a physical interpretation of the remaining term $H_{\text{rem},\perp}(t)$ as an energy sum over the transverse photon modes propagating in free space. These free photons are calculated using the $\mathbf{A}$ potential vector. From Eq. (4.36) we know that these modes differ in general from those in the $\mathbf{F}$ potential vector since $\mathbf{E}^{(v')}_\perp(\mathbf{x},t_0)$ is not identical to $\mathbf{E}^{(v)}_\perp(\mathbf{x},t_0)$ unless the polarization density $\mathbf{P}_\perp(\mathbf{x},t_0)$ cancels (which is the case in vacuum).

Now, in classical physics the meaning of expansion (4.37) is clear: it corresponds to a diagonalization of the Hamiltonian in terms of normal coordinates, i.e., like for classical mechanics [88], and similarly to the Huang, Fano, Hopfield procedure for polaritons [23–25]. In QED, the problem is different since, as explained in details in Ref. [75], fields like $a_{\alpha,j}(t)$ and $\mathbf{f}_{\omega,\perp}(\mathbf{x},t)$ (and their Hermitian conjugate variables) do not commute, unlike it is for $c_{\alpha,j}(t)$ and $\mathbf{f}_{\omega,\perp}(\mathbf{x},t)$. It is thus not possible to find common eigenstates of the $A^{(v')}$ operators for photons (in the usual representation) and for $X^{(0)}_\omega$ associated with the material fluctuations. This is not true for the representation using $F^{(v)}$ and $X^{(0)}_\omega$ operators, but now the full Hamiltonian is not fully diagonalized as seen from Eq. (4.28). Only if one neglects the remaining term $H_{\text{rem}}(t)$, like it was done in Refs. [27–29,32,34], can we diagonalize the Hamiltonian. However, then we get the troubles concerning unitarity, causality, and time symmetry discussed along this paper.

V. GENERAL CONCLUSION AND PERSPECTIVES

The general formalism discussed in this article using the **F** potential provides a natural way for dealing with QED in dispersive and dissipative media. It is based on a canonical quantization procedure generalizing the early work of Huttner and Barnett [16–22] for polaritons in homogeneous media. The method is unambiguous as far as we conserve all terms associated with free photons $\mathbf{F}^{(0)}$ and material fluctuations $\mathbf{P}^{(0)}$ for describing the quantum evolution. In particular, in order to preserve the full unitarity and the time symmetry of the coupled system of equations we have to include in the evolution terms associated with fluctuating electromagnetic modes $\mathbf{E}^{(0)}, \mathbf{B}^{(0)}$ which have a classical interpretation as polariton eigenmodes and can not in general be omitted if the medium is spatially localized in vacuum. We also discussed an alternative representation based on the potential **A** instead of **F**. At the end, both representations are clearly equivalent and could be used for generalizing the present theory to other linear media including tensorial anisotropy, magnetic properties, and constitutive equations coupling **E** and **B** (magneto and/or electric media). Moreover, the most important finding of this article concerns the comparisons between the generalized Huttner-Barnett approach, advocated here, which involves both photonic and material-independent degrees of freedom, and the Langevin-noise method proposed initially by Gruner and Welsch [34] which involves only the material degrees of freedom associated with fluctuating currents. We showed that rigorously speaking the Langevin-noise method is not equivalent to the full Hamiltonian QED evolution coupling photonic and material fields. Only in the regime where the dissipation of the bulk surrounding medium is nonvanishing at spatial infinity could we, i.e., for all practical purposes, identify the two theories. However, even with such assumptions, the Langevin-noise model is breaking time symmetry since it considers only decaying modes while the full Hamiltonian theory used in our work accepts also growing waves associated with antithermodynamic processes. We claim that this is crucial in nanophotonics and/or plasmonics where quantum emitters, spatially localized, are coupled to photonic and material modes available in the complex environment, e.g., near nanoantennas in vacuum (i.e., in a spatial domain where losses are vanishing at infinity). Since most studies consider the interaction between molecules or quantum dots and plasmon and/or polaritons using the Langevin-noise approach, we think that it is urgent to clarify and clean up the problem by analyzing the coupling regime using the full Hamiltonian evolution advocated in this work. Finally, we suggest that this work could impact the interpretation and discussion of pure QED effects such as the Casimir force or the Lamb shift which are strongly impacted by polariton and plasmon modes. All this will be the subject for future works and, therefore, the present detailed analysis is expected to play an important role in nanophotonics and plasmonics for both the classical and quantum regimes.

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APPENDIX A: ABSENCE OF ZEROS IN THE UPPER HALF-PLANE

The relation

$$\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2 = 0 \tag{A1}$$

admits zeros $\Omega_{\alpha,m}^{(\pm)}$ as postulated in the text. Writing $\omega = \omega' + i\omega''$ one of such zero and $\tilde{\varepsilon}(\omega) = \varepsilon' + i\varepsilon''$ the condition (A1) means

$$(\omega'^2 - \omega''^2)\varepsilon' - 2\omega'\omega''\varepsilon'' = \omega_\alpha^2,$$
$$(\omega'^2 - \omega''^2)\varepsilon'' + 2\omega'\omega''\varepsilon' = 0 \tag{A2}$$

from which we deduce after eliminating $(\omega'^2 - \omega''^2)$

$$2\omega'\omega''\frac{\varepsilon'^2 + \varepsilon''^2}{\varepsilon''} = -\omega_\alpha^2. \tag{A3}$$

Therefore, a necessary but not sufficient condition for having zeros is that if $\omega'\omega'' > 0$ for such a zero, then $\varepsilon'' < 0$ while if $\omega'\omega'' < 0$, then $\varepsilon'' > 0$. Actually, we also see from Eq. (A2) that the zeros are allowed to be located along the real or imaginary axis of the complex ω plane if $\varepsilon'' = 0$ along these axes. This is in general not possible for a large class of permittivity function. Consider, for example, the quite general causal permittivity, i.e., satisfying the Kramers-Krönig relation, defined by

$$\tilde{\varepsilon}(\omega) = 1 + \sum_n \frac{f_n}{\omega_n^2 - (\omega + i\gamma_n)^2} \tag{A4}$$

with $f_n, \gamma_n > 0$. Then, we have also

$$\tilde{\varepsilon}(\omega' + i\omega'') = 1 + \sum_n f_n \frac{\omega_n^2 - \omega'^2 + (\omega'' + \gamma_n)^2 + 2i\omega'(\gamma_n + \omega'')}{\left[\omega_n^2 - \omega'^2 + (\omega'' + \gamma_n)^2\right]^2 + 4\omega'^2(\gamma_n + \omega'')^2}. \tag{A5}$$

Clearly, $\varepsilon'' > 0$ if $\omega' > 0$ and $\omega'' \geqslant 0$ in contradiction with the necessary condition for zeros' existence mentioned before. This reasoning is valid in one quarter plane but now, if $\omega' + i\omega''$ is a zero, $-\omega' + i\omega''$ is also a zero. Therefore, the absence of a zero in the quarter plane $\omega' > 0, \omega'' \geqslant 0$ implies the absence of zero in the second quarter plane $\omega' < 0, \omega'' \geqslant 0$ and therefore Eq. (A1) does not admit any zero in the upper half-plane for a very usual permittivity like Eq. (A4). Actually, the case $\omega' = 0$ should be handled separately. We find from Eq. (A5) that for such value, $\varepsilon'' = 0$. This is acceptable in order to have a zero existence in agreement with Eq. (A2). However, from Eq. (A2) we find also that if a zero exists along the axis $\omega' = 0$, then we should have as well $\varepsilon' = -\omega_\alpha^2/\omega''^2 < 0$. This is in contradiction with Eq. (A5) which implies $\varepsilon' > 0$. This completes the proof for the permittivity given by Eq. (A4).

The question concerning the generality of the proof is, however, still open. Huttner and Barnett mentioned the existence of such a proof in the Landau and Lifschitz textbook [90] but it is relevant to detail the missing proof here. In order to get the complete result, we will use a method used by

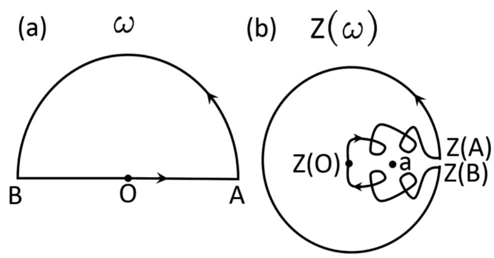

FIG. 1. Integration path deformation leading to the proof that Eq. (A1) has no solution in the upper frequency half-plane (see Refs. [90,103]).

Landau and Lifshitz (see Ref. [103], p. 380). In the complex ω plane, we define $Z(\omega)=\tilde{\varepsilon}(\omega)\omega^2$. From the properties of $\tilde{\varepsilon}(\omega)$ we deduce $Z(-\omega^*)=Z(\omega)^*$. This implies that Z is real along the imaginary axis and that $Z'(\omega')=Z'(-\omega')$ and $Z''(\omega')=-Z''(-\omega')$ along the real axis. Furthermore, due to causality we have $\tilde{\varepsilon}''(\omega')>0$ and therefore $Z''(\omega')>0$ if $\omega'>0$ along the real axis. Now, we consider the closed contour integral (see Fig. 1)

$$\oint_C \frac{d\omega}{2i\pi}\frac{\frac{dZ(\omega)}{d\omega}}{Z(\omega)-a} \tag{A6}$$

along C made of the real axis and of the semicircle C^+ of infinite radius $R\to+\infty$ in the upper half-plane. However, a is a real number and since Z is not real along the real axis there is no pole along C unless a is infinite or null. Therefore, since Z is analytical in the upper half-plane, Eq. (A6) gives the numbers of zeros of $Z(\omega)-a=0$ in this half-space.

We then rewrite Eq. (A6) as an integral in the complex Z plane:

$$\oint_{C'} \frac{dZ}{2i\pi}\frac{1}{Z-a}, \tag{A7}$$

where C' is the image of C along the mapping $\omega\mapsto Z(\omega)$. In particular, the origin O is mapped on itself while the semicircle of radius R is mapped onto the circle of radius R^2. The half real axis OA corresponding to $\omega>0$ is mapped onto a complex curve located in the upper half-plane of the complex Z space (since $Z''>0$ along this half line). Similarly, the second half axis OB is mapped in the lower half-plane. As shown on the figure, if $0<a<+\infty$, then the contour integral omits the point $Z=a$ and there is no pole involved in the integral which therefore vanishes. We thus deduce that Eq. (A1) has no solution in the upper half plane in the ω space, which is the proof needed.

APPENDIX B: POLAR EXPANSION OF A CAUSAL GREEN FUNCTION

The calculation of $H_\alpha(\tau)$ defined by the Bromwich integral (3.9) for $\tau>0$

$$\begin{aligned} H_\alpha(\tau) &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\frac{e^{p\tau}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2} \\ &= \int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}\frac{e^{-i\omega\tau}}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)(\omega+i\eta)^2}e^{+\eta\tau} \end{aligned} \tag{B1}$$

can be handled after closing the contour integral in the lower plane. However, since $\frac{1}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}$ is not analytical in such lower plane, we must include the poles $\Omega^{(\pm)}_{\alpha,m}$ (all located in the lower plane, see Appendix A), i.e., the residues, in the integral. We use the separation

$$\frac{1}{\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2}=\frac{1}{2\omega_\alpha}\left[\frac{1}{\omega_\alpha-\omega\sqrt{\tilde{\varepsilon}(\omega)}}+\frac{1}{\omega_\alpha+\omega\sqrt{\tilde{\varepsilon}(\omega)}}\right] \tag{B2}$$

and express it as a function of $\omega=\Omega^{(\pm)}_{\alpha,m}+\rho e^{i\varphi}$ near each pole (i.e., in the limit $\rho\to 0$). We get

$$\frac{1}{\omega_\alpha\pm\omega\sqrt{\tilde{\varepsilon}(\omega)}}\approx\pm\frac{1}{\rho e^{i\varphi}\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(\pm)}_{\alpha,m}}}. \tag{B3}$$

From the condition $\Omega^{(\pm)*}_{\alpha,m}=-\Omega^{(\mp)}_{\alpha,m}$ and the equality $\{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\}^*=\frac{\partial[-\omega^*\sqrt{\tilde{\varepsilon}(-\omega^*)}]}{\partial-\omega^*}=\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}|_{-\omega^*}$ we then get for $\tau>0$ after integration in the lower plane Eq. (3.10) and $H_\alpha(\tau)=0$ for $\tau<0$ (after integration in the upper plane where no pole is present). The value at $\tau=0$ deserves some careful analysis. Indeed, if $\tau=0$ the integration along the semicircle does not vanish exponentially with it radius R and if we choose to integrate in the upper half-plane (where there is no pole) we get $\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}\frac{1}{\omega_\alpha^2-\omega^2\tilde{\varepsilon}(\omega)}=-\int_0^\pi\frac{id\varphi R\,e^{i\varphi}}{2\pi}\frac{1}{\omega^2-R^2e^{2i\varphi}}=O(1/R)$ if $R\to+\infty$ since $\tilde{\varepsilon}(R\,e^{i\varphi})=1$ in this limit in the upper half plane. Therefore, we have indeed $H_\alpha(0)=0$ and the function is continuous at $\tau=0$. Of course, the null value for negative time t' has no meaning since the Laplace transform is only interested in the evolution for positive time.

This leads to the sum rule

$$\sum_m\frac{1}{\omega_\alpha}\mathrm{Im}\left\{\frac{1}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}\right\}=0. \tag{B4}$$

The free term $q^{(0)}_{\alpha,j}(t)$ is defined as

$$\begin{aligned} q^{(0)}_{\alpha,j}(t) &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\frac{[1+\bar{\chi}(p)][pq'_{\alpha,j}(0)+\dot{q}'_{\alpha,j}(0)]e^{p(t-t_0)}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2} \\ &= U_\alpha(t-t_0)\dot{q}_{\alpha,j}(t_0)+\dot{U}_\alpha(t-t_0)q_{\alpha,j}(t_0) \end{aligned} \tag{B5}$$

with

$$\begin{aligned} U_\alpha(\tau) &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}\frac{[1+\bar{\chi}(p)]e^{p\tau}}{\omega_\alpha^2+[1+\bar{\chi}(p)]p^2} \\ &= \sum_m\frac{-\tilde{\varepsilon}(\Omega^{(-)}_{\alpha,m})}{2i\omega_\alpha}\frac{e^{-i\Omega^{(-)}_{\alpha,m}\tau}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}+\text{c.c.} \end{aligned} \tag{B6}$$

Like for H_α we get $U_\alpha(0)=0$. The last line of Eq. (B5) is therefore justified from the fact that the Laplace transform of $\frac{d}{d\tau}U_\alpha(\tau)$ is $p\overline{U_\alpha}(p)-U_\alpha(0)=p\overline{U_\alpha}(p)$. Now, the boundary condition at $t=t_0$ imposes $\frac{d}{d\tau}U_\alpha(\tau)|_{\tau=0}=1$. Therefore, from Eq. (C1) we deduce the second sum rule:

$$\sum_m\frac{1}{\omega_\alpha}\mathrm{Re}\left\{\frac{\tilde{\varepsilon}(\Omega^{(-)}_{\alpha,m})\Omega^{(-)}_{\alpha,m}}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\big|_{\Omega^{(-)}_{\alpha,m}}}\right\}=1. \tag{B7}$$

The value at $\tau=0$ is not defined univocally since $\frac{d}{d\tau}U_\alpha(\tau)$ defined through the Bromwich integral of $U_\alpha(\tau)$ is discontinuous. We point out that considering a direct integration at $\tau = 0$ could lead to contradictions since the integration along $C^{\pm}$ does not vanish. If we choose to integrate in the upper half-plane (where there is no pole), we get

$$\int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{-i\omega\tilde{\varepsilon}(\omega)}{\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2} = -\int_0^{\pi} \frac{d\varphi R^2 e^{2i\varphi}}{2\pi} \frac{1}{\omega^2 - R^2 e^{2i\varphi}} = 1/2 \quad \text{(B8)}$$

if $R \to +\infty$ since $\tilde{\varepsilon}(R\,e^{i\varphi}) = 1$ in this limit in the upper half-plane. We would get $\frac{d}{d\tau}U_\alpha(\tau)|_{\tau=0} = 1/2$ (a similar calculation could be done in the lower space including poles and residues and we would obtain once again $1 - 1/2 = 1/2$). Here, we considered carefully the limit to prevent us from such a contradiction.

APPENDIX C: THE SOURCE FIELD: A VECTORIAL AND SCALAR POTENTIAL DISCUSSION

The source field can be written as $\mathbf{F}^{(s)} = \sum_{\alpha,j} q^{(s)}_{\alpha,j}\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha$. After some algebras, we get

$$\begin{aligned}\mathbf{F}^{(s)}(\mathbf{x},t) &= \int_{\gamma-i\infty}^{\gamma+i\infty} \frac{idp}{2\pi} e^{p(t-t_0)} \int d^3\mathbf{x}' G_\chi(|\mathbf{x}-\mathbf{x}'|,ip) \\ &\quad \cdot \nabla' \times \overline{\mathbf{P}'}^{(0)}(\mathbf{x}',p) \\ &= \int_{-\infty}^{+\infty} d\omega\, e^{-i\omega t} \int d^3\mathbf{x}' G_\chi(|\mathbf{x}-\mathbf{x}'|,\omega + i0^+) \\ &\quad \cdot \nabla' \times \underline{\widetilde{\mathbf{P}}}^{(0)}(\mathbf{x}',\omega),\end{aligned} \quad \text{(C1)}$$

where we introduced the Green function

$$\begin{aligned}G_\chi(|\mathbf{x}-\mathbf{x}'|,ip) &= c^2 \sum_\alpha \frac{\Phi_\alpha(\mathbf{x})\Phi^*_\alpha(\mathbf{x}')}{\omega_\alpha^2 + [1+\bar{\chi}(p)]p^2} \\ &= c^2 \int \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}}{c^2k^2 + [1+\bar{\chi}(p)]p^2} \\ &= \frac{e^{-p\sqrt{1+\bar{\chi}(p)}|\mathbf{x}-\mathbf{x}'|/c}}{4\pi|\mathbf{x}-\mathbf{x}'|}\end{aligned} \quad \text{(C2)}$$

computed by contour integration in the complex plane and solution of $\nabla^2 G_\chi(|\mathbf{x}-\mathbf{x}'|,ip) - [1+\bar{\chi}(p)]\frac{p^2}{c^2}G_\chi(|\mathbf{x}-\mathbf{x}'|,ip) = \delta^3(\mathbf{x}-\mathbf{x}')$. Of course, along the real axis $\gamma \to 0^+$ we get $G_\chi(|\mathbf{x}-\mathbf{x}'|,\omega + i0^+) = \frac{e^{i\omega\sqrt{\tilde{\varepsilon}(\omega)}|\mathbf{x}-\mathbf{x}'|/c}}{4\pi|\mathbf{x}-\mathbf{x}'|}$ which is the usual Green function for a homogeneous medium. We can also write this field without introducing ω by using the Green propagator $\Delta_\chi(\tau,R) = \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}G_\chi(R,ip)$ which leads to

$$\begin{aligned}\mathbf{F}^{(s)}(\mathbf{x},t) &= \int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \Delta_\chi(\tau,|\mathbf{x}-\mathbf{x}'|) \\ &\quad \cdot \nabla' \times \mathbf{P}^{(0)}(\mathbf{x}',t-\tau).\end{aligned} \quad \text{(C3)}$$

We have also

$$\begin{aligned}\Delta_\chi(t-t',|\mathbf{x}-\mathbf{x}'|) &= c^2 \sum_\alpha H_\alpha(t-t')\Phi^*_\alpha(\mathbf{x}')\Phi_\alpha(\mathbf{x}) \\ &= c^2 \sum_{\alpha,m} \frac{-1}{2i\omega_\alpha} \frac{e^{-i\Omega^{(-)}_{\alpha,m}(t-t')}\Phi^*_\alpha(\mathbf{x}')\Phi_\alpha(\mathbf{x})}{\frac{\partial[\omega\sqrt{\tilde{\varepsilon}(\omega)}]}{\partial\omega}\Big|_{\Omega^{(-)}_{\alpha,m}}} + \text{c.c.}\end{aligned} \quad \text{(C4)}$$

which represent the generalization of retarded propagator expansion for a lossy and dispersive medium. The role of causality is here crucial since the modes are always damped when the time is growing in the future direction as expected from pure thermodynamical considerations. This means in particular that $\Delta_\chi(t,|\mathbf{x}-\mathbf{x}'|)$ tends to vanish exponentially as t goes to infinity. Importantly, in the vacuum limit $[\chi(\tau) \to 0]$ we get naturally

$$\begin{aligned}\Delta_v(\tau,R) &= c^2 \sum_\alpha \frac{\sin\omega_\alpha\tau}{\omega_\alpha}\Phi^*_\alpha(\mathbf{x}')\Phi_\alpha(\mathbf{x}) \\ &= \frac{\delta(\tau - R/c)}{4\pi R}\end{aligned} \quad \text{(C5)}$$

and in the limit $t_0 \to -\infty$ we obtain the retarded potential

$$\mathbf{F}^{(s)}(\mathbf{x},t) = \int d^3\mathbf{x}' \frac{\nabla' \times \mathbf{P}^{(0)}(\mathbf{x}',t - |\mathbf{x}-\mathbf{x}'|/c)}{4\pi|\mathbf{x}-\mathbf{x}'|}. \quad \text{(C6)}$$

However, in the vacuum limit we have also $\mathbf{P}^{(0)}(\mathbf{x},t) \to 0$, therefore, $\mathbf{F}^{(s)}(\mathbf{x},t)$ actually vanishes and we get in this limit $\mathbf{F}(\mathbf{x},t) = \mathbf{F}^{(0)}(\mathbf{x},t)$ as it should be. Now, from Eq. (C3) and from the field definition we easily get the integral formulas for $\mathbf{D}^{(s)}(\mathbf{x},t)$ and $\mathbf{B}^{(s)}(\mathbf{x},t)$:

$$\begin{aligned}\mathbf{D}^{(s)}(\mathbf{x},t) &= \nabla \times \nabla \times \int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \Delta_\chi(\tau,|\mathbf{x}-\mathbf{x}'|) \\ &\quad \cdot \mathbf{P}^{(0)}(\mathbf{x}',t-\tau), \\ \mathbf{B}^{(s)}(\mathbf{x},t) &= \nabla \times \int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \frac{1}{c}\Delta_\chi(\tau,|\mathbf{x}-\mathbf{x}'|) \\ &\quad \cdot \partial_{t-\tau}\mathbf{P}^{(0)}(\mathbf{x}',t-\tau) \\ &\quad + \nabla \times \int d^3\mathbf{x}' \frac{1}{c}\Delta_\chi(t-t_0,|\mathbf{x}-\mathbf{x}'|) \cdot \mathbf{P}^{(0)}(\mathbf{x}',t_0).\end{aligned} \quad \text{(C7)}$$

These equations have a clear interpretation in terms of generalized Hertz potentials. In particular, taking the limit $t_0 \to -\infty$ and using the properties of convolutions, together with the fact that $\Delta_\chi(t-t_0,|\mathbf{x}-\mathbf{x}'|) \to 0$, we get

$$\begin{aligned}\mathbf{D}^{(s)}(\mathbf{x},t) &= \nabla \times \nabla \times \boldsymbol{\Xi}(\mathbf{x},t), \\ \mathbf{B}^{(s)}(\mathbf{x},t) &= \nabla \times \frac{1}{c}\partial_t \boldsymbol{\Xi}(\mathbf{x},t)\end{aligned} \quad \text{(C8)}$$

with

$$\boldsymbol{\Xi}(\mathbf{x},t) = \int_{-\infty}^{+\infty} dt' \int d^3\mathbf{x}' \Delta_\chi(t-t',|\mathbf{x}-\mathbf{x}'|)\mathbf{P}^{(0)}(\mathbf{x}',t'). \quad \text{(C9)}$$

APPENDIX D: COMPLEMENT CONCERNING THE GREEN DYADIC TENSOR IN A HOMOGENEOUS MEDIUM

By rewriting $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)$ in Eq. (3.20) we get after some rearrangements

$$\begin{aligned}\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) &= G_\chi(\mathbf{x},\mathbf{x}',ip)\mathbf{I} - \frac{c^2}{p^2[1+\bar{\chi}(p)]}\nabla\otimes\nabla G_\chi(\mathbf{x},\mathbf{x}',ip)\\ &= -\frac{c^2\nabla\times\nabla\times[G_\chi(\mathbf{x},\mathbf{x}',ip)\mathbf{I}]}{p^2[1+\bar{\chi}(p)]}\end{aligned} \tag{D1}$$

which involves the scalar Green function defined in Eq. (C2). From

$$\begin{aligned}\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip) &= \nabla\times\nabla\times\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)\\ &= -\frac{p^2}{c^2}[1+\bar{\chi}(p)]\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)+\mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}')\end{aligned} \tag{D2}$$

we have also

$$\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip) = \nabla\times\nabla\times[G_\chi(\mathbf{x},\mathbf{x}',ip)\mathbf{I}]+\mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}'). \tag{D3}$$

These formulas must be taken carefully since they are not actually valid at the source location, i.e., if $\mathbf{x}\to\mathbf{x}'$ due to the bad convergence of the series defining the dyadic Green function. After regularization, we can obtain the result

$$\begin{aligned}\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) &= \text{P.V.}\left\{G_\chi(\mathbf{x},\mathbf{x}',ip)\mathbf{I} - \frac{c^2}{p^2[1+\bar{\chi}(p)]}\nabla\otimes\nabla G_\chi(\mathbf{x},\mathbf{x}',ip)\right\}\\ &\quad + \frac{c^2}{p^2[1+\bar{\chi}(p)]}\mathbf{L}\delta^3(\mathbf{x}-\mathbf{x}')\end{aligned} \tag{D4}$$

and

$$\begin{aligned}\mathbf{S}_\chi(\mathbf{x},\mathbf{x}',ip) &= (\mathbf{I}-\mathbf{L})\delta^3(\mathbf{x}-\mathbf{x}')\\ &\quad +\text{P.V.}\{\nabla\times\nabla\times[G_\chi(\mathbf{x},\mathbf{x}',ip)\mathbf{I}]\}\end{aligned} \tag{D5}$$

with $\mathbf{L}$ a dyadic term depending on the way we define the principal value operator (P.V.) [38,104]:

$$\mathbf{L} = \oint_{(\Sigma)}\frac{\mathbf{n}\otimes\mathbf{R}}{4\pi R^2}dS. \tag{D6}$$

For a small exclusion spherical volume surrounding the point $\mathbf{x}'$ we get $\mathbf{L}=\mathbf{I}/3$, i.e., the depolarization field predicted by the Clausius-Mosotti formula [84].

In the particular case $\chi=0$ we write $\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip)=\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)$ and similarly for other Green functions. We also consider the time evolution which in vacuum relies on the propagators

$$\begin{aligned}\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip),\\ \mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}') &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip).\end{aligned} \tag{D7}$$

Explicit calculations lead to

$$\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') = \sum_{\alpha,j}\omega_\alpha^2\frac{\sin(\omega_\alpha\tau)}{\omega_\alpha}\Phi_\alpha(\mathbf{x})\Phi_\alpha^*(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\otimes\hat{\boldsymbol{\epsilon}}_{\alpha,j} \tag{D8}$$

and similarly for the transverse part of $\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')$:

$$\mathbf{U}_{v,\perp}(\tau,\mathbf{x},\mathbf{x}') = \sum_{\alpha,j}c^2\frac{\sin(\omega_\alpha\tau)}{\omega_\alpha}\Phi_\alpha(\mathbf{x})\Phi_\alpha^*(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\otimes\hat{\boldsymbol{\epsilon}}_{\alpha,j}, \tag{D9}$$

while for the longitudinal part we get

$$\mathbf{U}_{v,\parallel}(\tau,\mathbf{x},\mathbf{x}') = c^2\tau\sum_\alpha\Phi_\alpha(\mathbf{x})\Phi_\alpha^*(\mathbf{x}')\hat{\boldsymbol{k}}_\alpha\otimes\hat{\boldsymbol{k}}_\alpha. \tag{D10}$$

We deduce automatically the boundary conditions $\mathbf{U}_{v,\perp}(0,\mathbf{x},\mathbf{x}')=\mathbf{U}_{v,\parallel}(0,\mathbf{x},\mathbf{x}')=\mathbf{Q}_v(0,\mathbf{x},\mathbf{x}')=0$. We also obtain

$$\partial_\tau\mathbf{U}_{v,\perp}(\tau,\mathbf{x},\mathbf{x}') = \sum_{\alpha,j}c^2\cos(\omega_\alpha\tau)\Phi_\alpha(\mathbf{x})\Phi_\alpha^*(\mathbf{x}')\hat{\boldsymbol{\epsilon}}_{\alpha,j}\otimes\hat{\boldsymbol{\epsilon}}_{\alpha,j} \tag{D11}$$

and

$$\partial_\tau\mathbf{U}_{v,\parallel}(\tau,\mathbf{x},\mathbf{x}') = c^2\sum_\alpha\Phi_\alpha(\mathbf{x})\Phi_\alpha^*(\mathbf{x}')\hat{\boldsymbol{k}}_\alpha\otimes\hat{\boldsymbol{k}}_\alpha \tag{D12}$$

from which we obtain the boundary condition $\partial_\tau\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')_{\tau=0}=c^2\mathbf{I}\sum_\alpha\Phi_\alpha(\mathbf{x})\Phi_\alpha^*(\mathbf{x}')=c^2\mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}')$. We thus obtain

$$\begin{aligned}\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') &= \nabla\times\nabla\times\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')\\ &= -\frac{\partial_\tau^2\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')}{c^2}.\end{aligned} \tag{D13}$$

Finally, from Eq. (D4) we find explicitly for the time-dependent $\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')$ field

$$\begin{aligned}\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') &= (\mathbf{I}-\mathbf{L})\delta^3(\mathbf{x}-\mathbf{x}')\delta(\tau)\\ &\quad +\text{P.V.}\{\nabla\times\nabla\times[\Delta_v(\tau,\mathbf{x},\mathbf{x}')\mathbf{I}]\}.\end{aligned} \tag{D14}$$

Some important remarks should be done here concerning the use of inverse Laplace transforms at time $\tau=0$. Indeed, rigorously speaking the Bromwich integral in Eqs. (D7) vanishes for $\tau<0$. Therefore, it is better to write

$$\begin{aligned}\theta(\tau)\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip),\\ \theta(\tau)\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}') &= \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip).\end{aligned} \tag{D15}$$

This implies that a Heaviside function $\Theta(\tau)$ (defined by $\Theta=1$ for $\tau\geqslant 0$ and $\Theta=0$ for $\tau<0$) should be included on both sides of Eqs. (D8), (D9), and (D10). An important observation is that with this definition, $\theta(\tau)\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')$ or $\theta(\tau)\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')$ are causal since they vanish for negative time. These functions are therefore retarded propagator. This is not the case for Eqs. (D8), (D9) and (D10) without the $\Theta(\tau)$ function on both sides. Now, this is important for time derivatives since we have by definition of the Bromwich integral $\theta(\tau)\partial_\tau\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}[p\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)-\mathbf{U}_v(0,\mathbf{x},\mathbf{x}')]$ and similarly

$\theta(\tau)\partial_\tau^2\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}') = \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}[p^2\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip) - p\mathbf{U}_v(0,\mathbf{x},\mathbf{x}') - \dot{\mathbf{U}}_v(0,\mathbf{x},\mathbf{x}')]$. Using the boundary conditions at $\tau = 0$ for $\mathbf{U}_v$ we can rederive consistently Eqs. (D11), (D12) and (D14) which are now understood with Heaviside functions $\theta(\tau)$ on both sides of all equations. In particular, we have $\theta(\tau)[\nabla\times\nabla\times\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}') + \frac{1}{c^2}\partial_\tau^2\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')] = 0$. However, if we define the retarded Green function as $\mathbf{U}_{ret,v}(\tau,\mathbf{x},\mathbf{x}')\theta(\tau)\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')$ and consider directly the second-order time derivative of $\mathbf{U}_{ret,v}$ we get instead $-\frac{1}{c^2}\partial_\tau^2\mathbf{U}_{ret,v}(\tau,\mathbf{x},\mathbf{x}') = \Theta(\tau)[\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}') - \mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}')\delta(\tau)]$ and thus

$$\nabla\times\nabla\times\mathbf{U}_{ret,v}(\tau,\mathbf{x},\mathbf{x}') + \frac{1}{c^2}\partial_\tau^2\mathbf{U}_{ret,v}(\tau,\mathbf{x},\mathbf{x}') = \mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}')\delta(\tau) \tag{D16}$$

with a singular term in the equation [we removed the $\theta(\tau)$ function in the right-hand term since $\delta(\tau)\Theta(\tau) = \delta(\tau)\Theta(0) = \delta(\tau)$]. The choice between $\mathbf{U}_{ret,v}$ or $\mathbf{U}_v$ is unambiguous and results obtained are therefore equivalent with both definitions.

APPENDIX E: TRANSVERSE POLARITONS IN THE HOPFIELD MODEL: A CONSISTENCY CHECK

From Eq. (3.38) we deduce in the Hopfield model

$$\left(\omega_0^2 - \Omega^2\right)\widetilde{\mathbf{P}}(\Omega) = \omega_p^2\widetilde{\mathbf{E}}(\Omega). \tag{E1}$$

This leads to the solution

$$\widetilde{\mathbf{P}}(\Omega) = \frac{\omega_p^2}{\left[\omega_0^2 - (\Omega + i0^+)^2\right]}\widetilde{\mathbf{E}}(\Omega) + \widetilde{\mathbf{P}}(\Omega)^{(in)} \tag{E2}$$

and therefore to

$$\nabla\times\nabla\times\widetilde{\mathbf{E}}(\Omega) - \frac{\Omega^2}{c^2}\widetilde{\varepsilon}(\Omega)\widetilde{\mathbf{E}}(\Omega) = \frac{\Omega^2}{c^2}\widetilde{\mathbf{P}}(\Omega)^{(in)}, \tag{E3}$$

where the permittivity is given by the lossless Lorentz-Drude formula $\widetilde{\varepsilon}(\Omega) = 1 + \frac{\omega_p^2}{[\omega_0^2-(\Omega+i0^+)^2]}$. For the transverse fields, we expand the different fields as

$$\widetilde{\mathbf{E}}_\perp(\mathbf{x},\Omega) = \sum_{\alpha,j}\widetilde{E}_{\alpha,j}(\Omega)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}),$$
$$\widetilde{\mathbf{P}}_\perp(\mathbf{x},\Omega) = \sum_{\alpha,j}\widetilde{P}_{\alpha,j}(\Omega)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) \tag{E4}$$

with $\widetilde{E}_{\alpha,j}(\Omega)^* = \eta_j\widetilde{E}_{-\alpha,j}(-\Omega)$ and $\widetilde{P}_{\alpha,j}(\Omega)^* = \eta_j\widetilde{P}_{-\alpha,j}(-\Omega)$. Now, we write $\widetilde{\mathbf{P}}_\perp^{(in)}(\Omega) = \boldsymbol{\gamma}\delta(\Omega-\omega_0) + \boldsymbol{\gamma}^*\delta(\Omega+\omega_0)$ with $\boldsymbol{\gamma}(\mathbf{x})$ a transverse vector field. We thus have $\widetilde{P}_{\alpha,j}(\Omega)^{(in)} = \gamma_{\alpha,j}\delta(\Omega-\omega_0) + \eta_j\gamma^*_{-\alpha,j}\delta(\Omega+\omega_0)$ with $\gamma_{\alpha,j} = \int d^3\mathbf{x}\,\boldsymbol{\gamma}(\mathbf{x})\cdot\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x})^*$. We thus get the equation

$$\left[\omega_\alpha^2 - \Omega^2\widetilde{\varepsilon}(\Omega)\right]\widetilde{E}_{\alpha,j}(\Omega) = \Omega^2\widetilde{P}_{\alpha,j}(\Omega)^{(in)}. \tag{E5}$$

Near the singularities ω_0 we get

$$\left[\omega_\alpha^2 - \omega_0^2 - i\frac{\pi\omega_p^2\omega_0}{2}\delta(\Omega-\omega_0)\right]\widetilde{E}_{\alpha,j}(\Omega) = \omega_0^2\gamma_{\alpha,j}\delta(\Omega-\omega_0) \tag{E6}$$

and therefore supposing the regularity (as for the longitudinal case) we have $-i\frac{\pi\omega_p^2}{2\omega_0}\widetilde{E}_{\alpha,j}(\omega_0) = \gamma_{\alpha,j}$. The secular equation $[\omega_\alpha^2 - \Omega^2\widetilde{\varepsilon}(\Omega)]\widetilde{E}_{\alpha,j,\pm}(\Omega) = 0$ associated with the transverse modes can be easily solved and this indeed leads to

$$\widetilde{\mathbf{E}}_\perp(\mathbf{x},\Omega) = \sum_{\alpha,j,\pm}\widetilde{E}_{\alpha,j,\pm}(\Omega)\hat{\boldsymbol{\epsilon}}_{\alpha,j}\Phi_\alpha(\mathbf{x}) \tag{E7}$$

with $\widetilde{E}_{\alpha,j,\pm}(\mathbf{x},\Omega) = \phi_{\alpha,j,\pm}\delta(\Omega-\Omega_{\alpha,\pm}) + \eta_j\phi^*_{-\alpha,j,\pm}\delta(\Omega+\Omega_{\alpha,\pm})$ with $\phi_{\alpha,j,\pm}$ an amplitude coefficient for the transverse polariton mode. Importantly, we again get from the regularity condition $\widetilde{E}_{\alpha,j}(\omega_0) = 0$ and thus $\gamma_{\alpha,j} = 0$ like for the longitudinal mode. This implies $\widetilde{\mathbf{P}}(\Omega)^{(in)} = 0$ and from Eq. (E2) we get $\widetilde{\mathbf{P}}_\perp(\Omega) = \frac{\omega_p^2}{[\omega_0^2-(\Omega+i0^+)^2]}\widetilde{\mathbf{E}}_\perp(\Omega)$. At the end of the day, we obtain the following field amplitudes:

$$E_{\alpha,j,\pm}(t) = \phi_{\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t} + \eta_j\phi_{-\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t},$$
$$B_{\alpha,j,\pm}(t) = \frac{\omega_\alpha}{\Omega_{\alpha,\pm}}(\eta_j\phi_{\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t} - \phi_{-\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t}) \tag{E8}$$

and

$$\frac{P_{\alpha,j,\pm}(t)}{\omega_p^2} = \frac{(\phi_{\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t} + \eta_j\phi_{-\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t})}{\omega_0^2 - \Omega_{\alpha,\pm}^2},$$
$$\frac{i\dot{P}_{\alpha,j,\pm}(t)}{\omega_p^2\Omega_{\alpha,\pm}} = \frac{(\phi_{\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t} - \eta_j\phi_{-\alpha,j,\pm}e^{-i\Omega_{\alpha,\pm}t})}{\omega_0^2 - \Omega_{\alpha,\pm}^2} \tag{E9}$$

[using definitions similar to Eq. (E7)]. These define the Hopfield transformation between the old variables $E_{\alpha,j,\pm}(t)$, $B_{\alpha,j,\pm}(t)$, $P_{\alpha,j,\pm}(t)$, $\dot{P}_{\alpha,j,\pm}(t)$ and the new polaritonic variables representing the normal coordinates of the problem $\phi_{\alpha,j,\pm}$ and $\phi_{-\alpha,j,\pm}$. Up to a normalization, this is equivalent to the work by Hopfield.

APPENDIX F: MILONNI'S MODEL AND RISING AND LOWERING POLARITON OPERATORS

We start with the amplitude for the transverse polariton field

$$E^{(+)}_{\alpha,j,m}(t) = \int_{\delta\omega_{\alpha,m}}d\omega\frac{\omega^2}{\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}}f^{(0)}_{\omega,\alpha,j}(t), \tag{F1}$$

where $\delta\omega_{\alpha,m}$ is a frequency window centered on the polariton pulsation $\omega_c \simeq \mathrm{Re}[\Omega_{\alpha,m}] := \Omega'_{\alpha,m}$. This field is equivalently written as

$$E^{(+)}_{\alpha,j,m}(t) = \int_0^{+\infty}\frac{d\omega\,F_{\alpha,m}(\omega)\omega^2}{\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}}f^{(0)}_{\omega,\alpha,j}(t), \tag{F2}$$

where $F_{\alpha,m}(\omega)$ is a window function such as $F_{\alpha,m}(\omega) \simeq 1$ if ω belongs to the interval $\delta\omega_{\alpha,m}$ and $F_{\alpha,m}(\omega) \simeq 0$ otherwise. From the commuting properties of $f^{(0)}_{\omega,\alpha,j}(t)$ [see Eqs. (2.8) and (3.29)] we deduce the commutator

$$[E^{(+)}_{\alpha,j,m}(t), E^{(-)}_{\beta,l,n}(t)] = \frac{\hbar}{\pi}\delta_{\alpha,\beta}\delta_{j,l}\int_0^{+\infty}\frac{d\omega\,\omega^4\tilde{\varepsilon}''(\omega)}{\left|\omega_\alpha^2 - \tilde{\varepsilon}(\omega)\omega^2\right|^2}F_{\alpha,m}(\omega)F_{\beta,n}(\omega) \tag{F3}$$

with $F_{\alpha,m}(\omega)F_{\beta,n}(\omega)\simeq\delta_{n,m}F_{\alpha,m}(\omega)$. Now, consider the integral

$$I=\int_0^{+\infty}d\omega\frac{\omega^4\tilde{\varepsilon}''(\omega)}{\left|\omega_\alpha^2-\tilde{\varepsilon}(\omega)\omega^2\right|^2}F_{\alpha,m}(\omega)$$
$$=\int_0^{+\infty}d\omega\frac{\omega^4\tilde{\varepsilon}''(\omega)}{\left[\omega_\alpha^2-\tilde{\varepsilon}'(\omega)\omega^2\right]^2+\omega^4\tilde{\varepsilon}''(\omega)^2}F_{\alpha,m}(\omega).\quad\text{(F4)}$$

Since for weak losses the integrand is extremely peaked on the value $\Omega_{\alpha,m}$, we can write I as

$$I\simeq\int_{-\infty}^{+\infty}d\omega\frac{\Omega_{\alpha,m}^4\tilde{\varepsilon}''(\Omega_{\alpha,m})}{\left[\omega_\alpha^2-\tilde{\varepsilon}'(\omega)\omega^2\right]^2+\Omega_{\alpha,m}^4\tilde{\varepsilon}''(\Omega_{\alpha,m})^2}.\quad\text{(F5)}$$

We use the approximation $\omega_\alpha^2-\tilde{\varepsilon}'(\omega)\omega^2\simeq\omega_\alpha^2-\tilde{\varepsilon}'(\Omega_{\alpha,m})\Omega_{\alpha,m}^2-(\omega-\Omega_{\alpha,m})\frac{d[\tilde{\varepsilon}'(\omega)\omega^2]}{d\omega}|_{\Omega_{\alpha,m}}=-(\omega-\Omega_{\alpha,m})\frac{d[\tilde{\varepsilon}'(\omega)\omega^2]}{d\omega}|_{\Omega_{\alpha,m}}$ which is valid near the pole where the condition $\omega_\alpha^2\simeq\tilde{\varepsilon}'(\Omega_{\alpha,m})\Omega_{\alpha,m}^2$ approximately holds for transverse polaritons. We thus get

$$I=\frac{1}{\tilde{\varepsilon}''(\Omega_{\alpha,m})}\int_{-\infty}^{+\infty}du\frac{\gamma^2}{u^2+\gamma^2}$$
$$=\frac{\pi\Omega_{\alpha,m}^2}{\frac{d[\tilde{\varepsilon}'(\omega)\omega^2]}{d\omega}|_{\Omega_{\alpha,m}}}=\frac{\pi\Omega_{\alpha,m}}{2}\frac{d\Omega_{\alpha,m}^2}{d\omega_\alpha^2}\quad\text{(F6)}$$

with $\gamma^2=\{\frac{\Omega_{\alpha,m}^2\tilde{\varepsilon}''(\Omega_{\alpha,m})}{\frac{d[\tilde{\varepsilon}'(\omega)\omega^2]}{d\omega}|_{\Omega_{\alpha,m}}}\}^2$ and $u=\omega--\Omega_{\alpha,m}$ and where we used the integral $\int_{-\infty}^{+\infty}du\frac{\gamma^2}{u^2+\gamma^2}=\pi\gamma$ which is easily calculated in the complex plane. From this we finally obtain the commutator of Eq. (3.65). We point out that the result does not explicitly depend on the extension of the frequency windows $\delta\omega_{\alpha,m}$ if losses and dispersion are weak enough to have $\gamma\ll\delta\omega_{\alpha,m}$.

For the longitudinal polariton field, we have a similar calculation. Starting with the definition

$$E_{\alpha,m}^{(+)}(t)=\int_0^{+\infty}d\omega\,F_{\alpha,m}(\omega)\frac{-1}{\tilde{\varepsilon}(\omega)}\sqrt{\frac{\hbar\sigma_\omega}{\pi\omega}}f_{\omega,\alpha,\parallel}^{(0)}(t),\quad\text{(F7)}$$

we can calculate the commutator $[E_{\alpha,m,\parallel}^{(+)}(t),E_{\beta,n,\parallel}^{(-)}(t)]$. We have

$$[E_{\alpha,m,\parallel}^{(+)}(t),E_{\beta,n,\parallel}^{(-)}(t)]$$
$$=\frac{\hbar}{\pi}\delta_{\alpha,\beta}\int_0^{+\infty}d\omega\frac{\tilde{\varepsilon}''(\omega)}{|\tilde{\varepsilon}(\omega)|^2}F_{\alpha,m}(\omega)F_{\beta,n}(\omega)\quad\text{(F8)}$$

with again $F_{\alpha,m}(\omega)F_{\beta,n}(\omega)\simeq\delta_{n,m}F_{\alpha,m}(\omega)$. We have to evaluate the integral

$$I=\int_0^{+\infty}d\omega\frac{\tilde{\varepsilon}''(\omega)}{|\tilde{\varepsilon}(\omega)|^2}F_{\alpha,m}(\omega)$$
$$=\int_0^{+\infty}d\omega\frac{\tilde{\varepsilon}''(\omega)}{[\tilde{\varepsilon}'(\omega)]^2+\tilde{\varepsilon}''(\omega)^2}F_{\alpha,m}(\omega),\quad\text{(F9)}$$

which like for the transverse modes in the limit of weak losses and dispersion leads after straightforward calculations to

$$I\simeq\frac{\pi}{\left|\frac{d\tilde{\varepsilon}'(\omega)}{d\omega}|_{\Omega'_{\alpha,m}}\right|}.\quad\text{(F10)}$$

From this we deduce the commutator given in Eq. (3.69).

APPENDIX G: DERIVING THE TOTAL SCATTERED FIELD USING THE GREEN DYADIC FORMALISM

Starting with dyadic formalism, we get for the (d) electric field

$$\mathbf{D}^{(d)}(\mathbf{x},t)=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p(t-t_0)}\int d^3\mathbf{x}'\cdot\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}(\mathbf{x}',p).\quad\text{(G1)}$$

Equivalently, by using the inverse Laplace transform (see Appendix D for details) $\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip)$, we obtain in the time domain

$$\mathbf{D}^{(d)}(\mathbf{x},t)=\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')\cdot\mathbf{P}(\mathbf{x}',t-\tau).\quad\text{(G2)}$$

Like before (see Appendix D), we should rigorously introduce a multiplication by the Heaviside function $\Theta(t-t_0)$ on the left-hand term (this will be omitted in the following since by definition we are only interested in the evolution for $t\geqslant t_0$). This analysis implies $\mathbf{D}^{(d)}(\mathbf{x},t_0)=0$ and therefore we have $\mathbf{D}(\mathbf{x},t_0)=\mathbf{D}^{(v)}(\mathbf{x},t_0)$.

Moreover, most studies consider instead of the propagator $\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip)$ the dyadic Green function $\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)$ which is a solution of

$$\nabla\times\nabla\times\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)+\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)$$
$$=\boldsymbol{\delta}(\mathbf{x}-\mathbf{x}')=\mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}')\quad\text{(G3)}$$

and which is actually connected to $\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip)$ by $\mathbf{S}_v(\mathbf{x},\mathbf{x}',ip)=\mathbf{I}\delta^3(\mathbf{x}-\mathbf{x}')-\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)$. This dyadic Green function is very convenient since practical calculations very often involve not the displacement field $\mathbf{D}$, but the electric field $\mathbf{E}=\mathbf{D}-\mathbf{P}$. We obtain

$$\mathbf{E}(\mathbf{x},t)=\mathbf{E}^{(v)}(\mathbf{x},t)-\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p(t-t_0)}$$
$$\times\int d^3\mathbf{x}'\frac{p^2}{c^2}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}(\mathbf{x}',p).\quad\text{(G4)}$$

Alternatively, in the time domain we have for the scattered field $\mathbf{E}^{(d)}(\mathbf{x},t)=\mathbf{E}(\mathbf{x},t)-\mathbf{E}^{(v)}(\mathbf{x},t)$

$$\mathbf{E}^{(d)}(\mathbf{x},t)=\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\mathbf{Q}_v(\tau,\mathbf{x},\mathbf{x}')\cdot\mathbf{P}(\mathbf{x}',t-\tau)-\mathbf{P}(\mathbf{x},t).\quad\text{(G5)}$$

This can be rewritten by using the propagator $\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)$ together with Eq. (D13) as

$$\mathbf{E}^{(d)}(\mathbf{x},t)=-\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{\partial_\tau^2\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')}{c^2}$$
$$\cdot\mathbf{P}(\mathbf{x}',t-\tau)-\mathbf{P}(\mathbf{x},t)\quad\text{(G6)}$$

or again if we want to introduce the retarded Green function [see Eq. (D16)] as

$$\mathbf{E}^{(d)}(\mathbf{x},t)=-\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{\partial_\tau^2\mathbf{U}_{ret,v}(\tau,\mathbf{x},\mathbf{x}')}{c^2}\cdot\mathbf{P}(\mathbf{x}',t-\tau).\quad\text{(G7)}$$

Finally, we can also use the dyadic formalism to represent the magnetic field $\mathbf{B}^{(d)}(\mathbf{x},t)$. Starting from Eq. (2.6) which yields $\nabla\times\overline{\mathbf{E}'}(\mathbf{x},p)=-\frac{p}{c}\overline{\mathbf{B}'}(\mathbf{x},p)+\frac{\mathbf{B}(\mathbf{x},t_0)}{c}$ and therefore

$$\overline{\mathbf{B}'}(\mathbf{x},p)=\overline{\mathbf{B}'}^{(v)}(\mathbf{x},p)+\int d^3\mathbf{x}'\frac{p}{c}\nabla\times\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\overline{\mathbf{P}'}(\mathbf{x}',p), \quad \text{(G8)}$$

where we introduced the definition $\overline{\mathbf{B}'}^{(v)}(\mathbf{x},p)=\frac{\nabla\times\overline{\mathbf{D}'}^{(v)}(\mathbf{x},p)}{-p/c}+\frac{\mathbf{B}(\mathbf{x},t_0)}{p}$ [here we used the condition $\mathbf{B}(\mathbf{x},t_0)=\mathbf{B}^{(v)}(\mathbf{x},t_0)$]. In the time domain, we thus directly obtain

$$\mathbf{B}^{(d)}(\mathbf{x},t)=\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{1}{c}\nabla\times\partial_\tau\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')\cdot\mathbf{P}(\mathbf{x}',t-\tau) \quad \text{(G9)}$$

which yields $\mathbf{B}^{(d)}(\mathbf{x},t_0)=0$ as expected.

APPENDIX H: EVOLUTION OF THE ELECTROMAGNETIC FIELD IN THE TIME DOMAIN

In the time domain we get for the electric field discussed in Sec. IV B

$$\mathbf{E}^{(0)}(\mathbf{x},t)=\mathbf{D}^{(v)}(\mathbf{x},t)-\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{\partial_\tau^2\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')}{c^2}\times\int_0^{t-\tau-t_0}d\tau'\chi(\mathbf{x}',\tau')\mathbf{E}^{(0)}(\mathbf{x}',t-\tau-\tau')-\int_0^{t-t_0}\chi(\mathbf{x},\tau')d\tau'\mathbf{E}^{(0)}(\mathbf{x},t-\tau') \quad \text{(H1)}$$

and for the magnetic field

$$\mathbf{B}^{(0)}(\mathbf{x},t)=\mathbf{B}^{(v)}(\mathbf{x},t)+\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{1}{c}\nabla\times\partial_\tau\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')\times\int_0^{t-\tau-t_0}d\tau'\chi(\mathbf{x}',\tau')\mathbf{E}^{(0)}(\mathbf{x}',t-\tau-\tau'), \quad \text{(H2)}$$

which are completed by the constitutive relation $\mathbf{D}^{(0)}(\mathbf{x},t)=\int_0^{t-t_0}d\tau\,\chi(\mathbf{x}',\tau)\mathbf{E}^{(0)}(\mathbf{x},t-\tau)+\mathbf{E}^{(0)}(\mathbf{x},t)$. Similarly, we obtain for the new propagator

$$\mathbf{U}_\chi(\tau,\mathbf{x},\mathbf{x}')=\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p\tau}\mathbf{G}_\chi(\mathbf{x},\mathbf{x}',ip) \quad \text{(H3)}$$

the integral formula

$$\mathbf{U}_\chi(t,\mathbf{x},\mathbf{x}'')=\mathbf{U}_v(t,\mathbf{x},\mathbf{x}'')-\int_0^{t}d\tau\int d^3\mathbf{x}'\frac{\partial_\tau^2\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')}{c^2}\times\int_0^{t-\tau}\chi(\mathbf{x}',\tau')d\tau'\mathbf{U}_\chi(t-\tau-\tau',\mathbf{x}',\mathbf{x}'')-\int_0^{t}d\tau'\chi(\mathbf{x},\tau')\mathbf{U}_\chi(t-\tau',\mathbf{x},\mathbf{x}'') \quad \text{(H4)}$$

We have the important properties $\mathbf{U}_\chi(t=0,\mathbf{x},\mathbf{x}'')=0$, $\partial_t\mathbf{U}_\chi(t,\mathbf{x},\mathbf{x}')|_{t=0}=c^2\delta^3(\mathbf{x}-\mathbf{x}')\mathbf{I}$. The total electromagnetic field in the time domain is thus expressed as

$$\mathbf{E}(\mathbf{x},t)=\mathbf{E}^{(0)}(\mathbf{x},t)-\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{\partial_\tau^2\mathbf{U}_\chi(\tau,\mathbf{x},\mathbf{x}')}{c^2}\cdot\mathbf{P}^{(0)}(\mathbf{x}',t-\tau)-\mathbf{P}^{(0)}(\mathbf{x},t), \quad \text{(H5)}$$

$$\mathbf{B}(\mathbf{x},t)=\mathbf{B}^{(0)}(\mathbf{x},t)+\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\frac{1}{c}\nabla\times\partial_\tau\mathbf{U}_\chi(\tau,\mathbf{x},\mathbf{x}')\cdot\mathbf{P}^{(0)}(\mathbf{x}',t-\tau) \quad \text{(H6)}$$

with $\mathbf{E}=\mathbf{E}^{(s)}+\mathbf{E}^{(0)}$, $\mathbf{B}=\mathbf{B}^{(s)}+\mathbf{B}^{(0)}$. Finally, the constitutive relation for the scattered displacement field $\mathbf{D}^{(s)}=\mathbf{D}-\mathbf{D}^{(0)}$ reads as $\mathbf{D}^{(s)}(\mathbf{x},t)=\int_0^{t-t_0}d\tau\,\chi(\mathbf{x}',\tau)\mathbf{E}^{(s)}(\mathbf{x},t-\tau)+\mathbf{E}^{(s)}(\mathbf{x},t)+\mathbf{P}^{(0)}(\mathbf{x},t)$.

APPENDIX I: DESCRIPTION OF THE SCATTERED FIELD USING THE STANDARD COULOMB GAUGE

In the usual Coulomb gauge representation, the longitudinal contribution $\mathbf{E}_\parallel^{(d')}(\mathbf{x},t)$ is given by the usual instantaneous Coulomb field

$$\mathbf{E}_\parallel^{(d')}(\mathbf{x},t)=-\nabla[V(\mathbf{x},t)]=\nabla\left[\int d^3\mathbf{x}'\frac{\nabla'\cdot\mathbf{P}(\mathbf{x}',t)}{4\pi|\mathbf{x}-\mathbf{x}'|}\right]=-\mathbf{P}_\parallel(\mathbf{x},t). \quad \text{(I1)}$$

Since there is no other longitudinal contribution, this leads to $\mathbf{E}_\parallel(\mathbf{x},t_0)=-\mathbf{P}_\parallel(\mathbf{x},t_0)$ which is Eq. (4.6). As mentioned already, this is the usual result. However, the most important term here is the transverse source field $\mathbf{E}_\perp^{(d')}(\mathbf{x},t)=-\frac{1}{c}\partial\mathbf{A}^{(d')}(\mathbf{x},t)$. We get for this term

$$\mathbf{E}_\perp^{(d')}(\mathbf{x},t)=-\frac{1}{c^2}\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\Delta_v(\tau,|\mathbf{x}-\mathbf{x}'|)\cdot\partial_{t-\tau}^2\mathbf{P}_\perp(\mathbf{x}',t-\tau)-\frac{1}{c^2}\int d^3\mathbf{x}'\Delta_v(t-t_0,|\mathbf{x}-\mathbf{x}'|)\cdot\partial_{t_0}\mathbf{P}_\perp(\mathbf{x}',t_0), \quad \text{(I2)}$$

which is also written as

$$\mathbf{E}_\perp^{(d')}(\mathbf{x},t)=-\frac{1}{c^2}\int_0^{t-t_0}d\tau\int d^3\mathbf{x}'\mathbf{U}_{v,\perp}(\tau,\mathbf{x},\mathbf{x}')\cdot\partial_{t-\tau}^2\mathbf{P}(\mathbf{x}',t-\tau)-\frac{1}{c^2}\int d^3\mathbf{x}'\mathbf{U}_{v,\perp}(t-t_0,\mathbf{x},\mathbf{x}'|)\cdot\partial_{t_0}\mathbf{P}(\mathbf{x}',t_0). \quad \text{(I3)}$$

Equivalently, we have in the frequency domain

$$\mathbf{E}_\perp^{(d')}(\mathbf{x},t)=-\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p(t-t_0)}\int d^3\mathbf{x}'\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\left[\frac{p^2}{c^2}\overline{\mathbf{P}'}(\mathbf{x}',p)-p\mathbf{P}_\perp(\mathbf{x}',t_0)\right]. \quad \text{(I4)}$$

The scattered magnetic field $\mathbf{B}^{(d')}$ is given by

$$\mathbf{B}^{(d')}(\mathbf{x},t) = \nabla \times \int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \frac{1}{c}\Delta_v(\tau,|\mathbf{x}-\mathbf{x}'|)\cdot\partial_{t-\tau}\mathbf{P}_\perp(\mathbf{x}',t-\tau)$$
$$= \int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \frac{1}{c}\nabla\times\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')\cdot\partial_{t-\tau}\mathbf{P}(\mathbf{x}',t-\tau) \tag{I5}$$

or, equivalently, by

$$\mathbf{B}^{(d')}(\mathbf{x},t) = \int_0^{t-t_0} d\tau \int d^3\mathbf{x}' \frac{1}{c}\nabla\times\partial_\tau\mathbf{U}_v(\tau,\mathbf{x},\mathbf{x}')\cdot\mathbf{P}(\mathbf{x}',t-\tau) - \int d^3\mathbf{x}'\frac{1}{c}\nabla\times\mathbf{U}_v(t-t_0,\mathbf{x},\mathbf{x}')\cdot\mathbf{P}(\mathbf{x}',t_0). \tag{I6}$$

In the frequency domain, this gives

$$\mathbf{B}^{(d')}(\mathbf{x},t) = \int_{\gamma-i\infty}^{\gamma+i\infty}\frac{idp}{2\pi}e^{p(t-t_0)}\int d^3\mathbf{x}'\nabla\times\mathbf{G}_v(\mathbf{x},\mathbf{x}',ip)\cdot\left[\frac{p}{c}\overline{\mathbf{P}'}(\mathbf{x}',p) - \frac{\mathbf{P}(\mathbf{x}',t_0)}{c}\right]. \tag{I7}$$

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[92] More precisely, we have near the singularity ω_0 the condition $\boldsymbol{\alpha}\delta(\Omega-\omega_0) = \frac{i\pi\omega_p^2}{2\omega_0}\delta(\Omega-\omega_0)\widetilde{\mathbf{P}}_\parallel(\omega_0) + \widetilde{\mathbf{P}}_\parallel(\Omega)$. If we suppose $\widetilde{\mathbf{P}}_\parallel(\omega_0)$ to be regular, we get the result discussed in the text. If $\widetilde{\mathbf{P}}_\parallel(\omega_0)$ is singular, then $\frac{i\pi\omega_p^2}{2\omega_0}\delta(\Omega-\omega_0)\widetilde{\mathbf{P}}_\parallel(\omega_0)$ would be even more singular and this would contradict the equation. Therefore, regularity at $\pm\omega_0$ must be imposed.
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