Quantization of Macroscopic Electrodynamics in Absorptive
Dielectrics of Finite Size in Unbounded Space

Carlo Forestiere

Department of Electrical Engineering and Information Technology,
Università degli Studi di Napoli Federico II,
via Claudio 21, Napoli, 80125, Italy

Giovanni Miano
Department of Electrical Engineering and Information Technology,
Università degli Studi di Napoli Federico II,
via Claudio 21, Napoli, 80125, Italy
Abstract

An operative approach to the quantization of macroscopic electrodynamics for absorptive dielectrics of finite size in unbounded space is presented. It is based on the combination of a Hopfield-type model for the polarization of the dielectric, and an expansion of the polarization density field in terms of the static longitudinal and transverse modes of the dielectric object, which only depend on its shape and size. To account for the dissipation of the matter, the polarization is described through a continuum of oscillating harmonic fields that are linearly coupled the electromagnetic field. The Coulomb gauge is applied, the Coulomb electric field is expressed in terms of the polarization field, and the radiation field is expanded in terms of transverse plane waves in free space. The Heisenberg equations for the coordinate operators of the polarization density field are obtained in a closed form. The initial state of the radiation and matter comes into play through the driving operators. Even if the equations for the longitudinal and transverse coordinate operators of the polarization are coupled due to the interaction of the polarization with the radiation, few longitudinal and transverse modes are needed for dielectrics with size of the same order of the characteristic wavelength \( \lambda_c = \min_{\omega} \{ c_0 / |\omega \sqrt{\chi(\omega)}| \} \) where \( \chi(\omega) \) is the macroscopic susceptibility of the dielectric in the frequency domain. Eventually, the polarization density field operator and the electric field operator are expressed in terms of the driving field operators by the same relations of the classical electrodynamics.

I. INTRODUCTION

In the last twenty years, there has been a large interest in quantum electrodynamics phenomena in metallic and dielectric structures of finite size motivated by the prospect of using plasmonic and photonic devices for quantum optics and quantum technology applications (e.g., [1], [2]). While the problem of quantization of the macroscopic electromagnetic field in nondispersive and homogeneous dielectrics has been successfully studied since the work of Jauch and Watson [3], for dispersive and finite size dielectrics in unbounded space the problem is significantly more difficult.

Glauber and Lewenstein [4] have proposed two quantization schemes for the electromagnetic field in non dispersive and non homogeneous dielectrics in unbounded space, both based on the generalized Coulomb gauge \( \nabla \cdot [\epsilon(r)A] = 0 \). In the first scheme, they expand
the electromagnetic field in terms of the full wave eigenmodes of the dielectric object, which is a continuum set in unbounded space. In the second scheme, they expand the electromagnetic field in terms of a continuum basis of functions based on plane waves that satisfy the generalized Coulomb gauge. They also discuss the relation between the two quantization schemes in the framework of the electromagnetic scattering theory.

To deal with dispersive dielectrics there is a need to introduce dynamical variables that represent the degrees of freedom of the matter. The established approaches are mainly based on either the Hopfield microscopic model \([5]\), or the Langevin-noise approach \([6, 7]\); see, also, the review in ref. \([8]\).

Hopfield represented the polarization field of a homogeneous dielectric as a harmonic oscillating bosonic field linearly coupled to the electromagnetic field \([5]\). He quantized the electromagnetic field by applying the Coulomb gauge. This model was firstly introduced by Fano \([9]\), who explained it in terms of an atomic medium. It also applies to the oscillations of free electrons in metals.

Huttner and Barnett \([10]\) extended the Hopfield model to account for the losses of the matter by coupling the polarization field to the electromagnetic field and to a continuum of reservoir fields. They used the Hamiltonian in the Coulomb gauge, applied the standard canonical quantization method to the entire system and, upon assuming the homogeneity of the medium, diagonalized the Hamiltonian with the Fano method. For inhomogeneous media the diagonalization of the Hamiltonian requires the classical dyadic Green function for the electric field in the presence of the dielectric object, as shown in \([11]\).

In the Huttner-Barnett model the diagonalization of the matter Hamiltonian (polarization field + reservoir field) yields to a set of dressed continuum fields that are coupled to the electromagnetic field. This fact suggested that absorptive dielectrics, satisfying the Kramers-König relations, can be equivalently described by only one continuum set of harmonic oscillating field directly coupled to the electromagnetic field \([12, 13]\).

In Ref. \([12]\), following \([4]\), the electromagnetic field is expressed in terms of the full wave eigenmodes of a non-dispersive reference dielectric body, then the Hamiltonian in the fundamental fields \(D\) and \(B\) is quantized and diagonalized by the Fano method. In Ref. \([13]\) the Hamiltonian based on the Coulomb gauge is quantized and it is diagonalized by the Fano method. In this case, the diagonalization also requires the dyadic Green function for the electric field in the presence of the dielectric object, as in \([11]\).
The Hopfield-type models have been applied to nonhomogeneous, dispersive and lossless dielectrics in combination with the Power-Zienau-Wooley Lagrangian (e.g., [14], [15]). In [14] the diagonalization of the Hamiltonian by the Fano method leads to the solution of the classical electromagnetic scattering problem for nonhomogeneous dielectrics in the frequency domain. In Ref. [15] the diagonalization of the Hamiltonian is brought back to the numerical solution of the Lippmann-Schwinger equation, which is equivalent to the solution of a classical electromagnetic scattering problem.

The Hopfield model has been applied to quantize the plasmons in the full-retarded regime in metal particles, by expanding the current density field in terms of the electrostatic modes of the particle [16]. A canonical quantization with numerical mode decomposition for diagonalizing the Hamiltonian has been recently proposed [17].

The Langevin-noise approach is based on the addition of phenomenological fluctuating currents to deal with the problem of dissipation and dispersion [6, 18, 19]. Inhomogeneous media are dealt with by means of the classical dyadic Green function for the electric field in the presence of the dielectric object to express the electromagnetic field in terms of the noise current operator [7, 20]. The Langevin based scheme is extensively used in many contexts (e.g., [21, 22]).

The Hopfield-type approach and the Langevin noise approach are equivalent if, in the Langevin noise approach, the quantized photonic degrees of freedom associated with the fluctuating radiation fields are added to the degrees of freedom of the material oscillators [23, 24]. From an operational perspective, all these schemes require the full wave solution of a classical electromagnetic scattering problem in unbounded space for the diagonalization of the Hamiltonian: or the computation of wave eigenmodes of the dielectric object, or the computation of the Green’s function in the presence of the dielectric object, or the solution of three dimensional Lippmann-Schwinger type equations.

In this paper, we propose an operative approach to the quantization of the macroscopic electromagnetic field in absorptive dielectrics of finite size in unbounded space that uses a new modal expansion recently proposed for classical electromagnetic scattering problems [25]. Our approach is based on a Hopfield-type model [5] but it does not rely on an explicit diagonalization of the Hamiltonian. Our strategy is as it follows. As in Ref. [12] and [13], we first describe the polarization of the dielectric as a “matter field” of continuum harmonic oscillators linearly coupled to the electromagnetic field. Then, we determine the Hamiltonian...
of the entire system in the Coulomb gauge. At this point, instead of diagonalizing the
Hamiltonian, by following our approach to the classical electromagnetic scattering:

a) we expand the matter field and, hence, the polarization field, in terms of the static
longitudinal eigenmodes \{\textbf{U}_m^\parallel(\mathbf{r})\} and the static transverse eigenmodes \{\textbf{U}_m^\perp(\mathbf{r})\} of the
dielectric object (see Appendix A), which only depend on the shape and size of the dielectric
object. These modes are a discrete basis for the solenoidal vector fields defined in the
bounded region occupied by the dielectric.

b) we express the irrotational component of the electric field (Coulomb electric field) in
terms of the longitudinal coordinates of the polarization field;

c) we expand the solenoidal component of the electromagnetic field (radiation field) in
terms of the transverse plane waves in free space;

d) we quantize in a standard fashion the coordinates of the matter field, the radiation
field, and the corresponding conjugate momenta, by enforcing the canonical commutation
relations between the field operators and their conjugate momenta;

e) we determine the Heisenberg equations of motion for the coordinate operators of the
matter and radiation field operators;

f) we eliminate the coordinate operators of the radiation field and determine the equations
of motion for the longitudinal and transverse coordinate operators of the polarization field.

In the Laplace domain they are:

\[
\hat{P}_m^\parallel + \frac{\bar{\chi}(s)}{\kappa_m^\parallel} \hat{P}_m^\parallel + \bar{\chi}(s) \sum_{m'} s \left[ S_{mm'}^\parallel(s) \hat{P}_{m'}^\parallel + S_{mm'}^\perp(s) \hat{P}_{m'}^\perp \right] = \hat{D}_m^\parallel(s),
\]

\[
\hat{P}_m^\perp + \left( \frac{s}{c_0} \right)^2 \frac{\bar{\chi}(s)}{\kappa_m^\perp} \hat{P}_m^\perp + \bar{\chi}(s) \sum_{m'} s \left[ S_{mm'}^\perp(s) \hat{P}_{m'}^\parallel + \delta S_{mm'}^\perp(s) \hat{P}_{m'}^\perp \right] = \hat{D}_m^\perp(s),
\]

where: \{\hat{P}_m^\parallel(s)\} and \{\hat{P}_m^\perp(s)\} are the Laplace transform of the longitudinal and transverse
coordinate operators of the polarization; \bar{\chi}(s) is the macroscopic susceptibility of the dielec-
tric in the Laplace domain; \kappa_m^\parallel is the eigenvalue corresponding to the longitudinal mode
\textbf{U}_m^\parallel(\mathbf{r}), which is a positive dimensionless quantity (see Appendix A); \kappa_m^\perp is the eigenvalue
corresponding to the transverse mode \textbf{U}_m^\perp(\mathbf{r}), which is a positive quantity homogeneous to
the reciprocal of the square of length, and it is positive (see Appendix A); \(c_0\) is the light
velocity in vacuum;

\[
S_{mm'}^{ab}(s) = \frac{1}{c_0} \int_V d^3 \mathbf{r} \int_V d^3 \mathbf{r}' \textbf{U}_m^a(\mathbf{r}) \textbf{G}_{\perp}(\mathbf{r} - \mathbf{r'}; s) \textbf{U}_m'^b(\mathbf{r}')
\]
where $a, b = \|, \perp$, $\overrightarrow{G}^\perp(r - r'; s)$ is the transverse dyadic full wave Green function for the vacuum; $\delta S_{\perp\perp}^{\perp\perp}(s)$ has the same expression of $S_{\perp\perp}^{\perp\perp}(s)$ but the static term $\overrightarrow{G}^\perp(r - r'; s = 0)$ is subtracted to $\overrightarrow{G}^\perp(r - r'; s)$; $\{\hat{D}^\parallel_m\}$ and $\{\hat{D}^\perp_m\}$ are the driving coordinate operators taking into account the contributions of: the incident electromagnetic field; the initial state of the radiation field; the radiation field generated by the initial state of the polarization field; the noise due to the dielectric medium fluctuations;

The second term on the left hand side of the equation governing $\hat{P}^\parallel_m$ is connected to the electroquasistatic (plasmon) oscillations of the medium, while the same term in the equation governing $\hat{P}^\perp_m$ is connected to the magnetoquasistatic oscillations of the medium. The natural frequencies of these oscillations are determined by the susceptibility $\chi(s)$ and the eigenvalues $\{\kappa_m^\parallel\}$, $\{\kappa_m^\perp\}$. The eigenvalues $\{\kappa_m^\parallel\}$ only depend on the shape of the object, while the eigenvalues $\{\kappa_m^\parallel\}$ also depend on its size: $\{\kappa_m^\parallel\}$ scales as $1/a^2$ as the size of the object $a$ varies, where $a$ is the radius of the smallest sphere that encloses the dielectric.

The coefficients $\{sS_{\perp\perp}^{a\perp\perp}\}$ and $\{s\delta S_{\perp\perp}^{\perp\perp}\}$ describe the coupling between the longitudinal and the transverse coordinate operators due to the interaction of the polarization with the radiation. They account for the exchange of the electromagnetic energy between the modes $U^a_m$ and $U^b_{m'}$, which is a non conservative process due to the radiated energy to infinity. The amplitudes of $sS_{\perp\perp}^{a\perp\perp}$ and $s\delta S_{\perp\perp}^{\perp\perp}$ tend to zero as $s \to 0$ and tend to finite limits as $|s| \to \infty$. For fixed values of $s$, the amplitude of $sS_{\perp\perp}^{a\perp\perp}$ scales as the dimensionless parameter $\gamma^2$ as the size of the particle $a$ varies, where $\gamma = |s|a/c_0$; the amplitude of $s\delta S_{\perp\perp}^{\perp\perp}$ instead scales as $\gamma^4$.

As expected, the equations of motion for the coordinate operators of the polarization have the same structure of the corresponding equations of the classical electrodynamics (see in [25]). Therefore, the representation of the polarization field operator in terms of the static modes of the dielectric object leads to the same advantages of the classical regime. In particular, the longitudinal and transverse modes of the dielectric object are the natural modes of the polarization in the small size limit $a \ll \lambda_c$ where $\lambda_c = \min_\omega \{c_0/|\omega\sqrt{\chi(\omega)}|\}$ is a “characteristic wavelength” accounting for the strength of the coupling between the matter and the electromagnetic field, i.e., $|\chi|\gamma^2 \ll 1$. Indeed, the longitudinal modes diagonalize the contribution to the Hamiltonian of the electroquasistatic (Coulomb) interaction energy, while the transverse modes diagonalize the magnetostatic interaction energy between the transverse modes. Therefore, only a small number of longitudinal and transverse modes of
the dielectric object are sufficient to accurately describe the response of the polarization even when $a \sim \lambda_c$ as in the classical electromagnetic scattering \[25\]. For example, for Drude-Lorentz dielectric functions we have $\lambda_c = c_0/\omega_p$ where $\omega_p$ is the plasma frequency of the medium.

g) we express the coordinate operators of the polarization as

$$\begin{pmatrix} \hat{P}_\parallel(s) \\ \hat{P}_\perp(s) \end{pmatrix} = H(s) \begin{pmatrix} \hat{P}_\parallel(0) \\ \hat{P}_\perp(0) \end{pmatrix},$$

where $\hat{P}_\parallel$ and $\hat{P}_\perp$ are, respectively, the column vectors of the longitudinal and transverse coordinate operators of the polarization, $\hat{D}_\parallel$ and $\hat{D}_\perp$ are the column vectors of the driving coordinate operators, and $H$ is the transfer matrix function of the corresponding classical electrodynamics problem, which has been extensively studied in Ref. \[25\];

h) we eventually express in the Laplace domain the solenoidal component of the electric field operator $\hat{E}_s(r; s)$ and the irrotational component $\hat{E}_c(r; s)$ as function of the polarization field operator $\hat{P}(r; s)$,

$$\hat{E}_s(r; s) = -\mu_0 \int_V \frac{G\parallel(r - r'; s)}{r - r'} [s\hat{P}(r'; s) - \hat{P}(r'; 0)] d^3r' + \hat{E}_s^{\text{vac}}(r; s),$$

$$\hat{E}_c(r; s) = -\frac{1}{4\pi \epsilon_0} \nabla \int_{\partial V} \hat{P}(r'; t) \cdot \hat{n}(r') d^2r',$$

where $\hat{P}(r; 0)$ is the polarization density field operator in the Schrödinger picture, and $\hat{E}_s^{\text{vac}}(r; s)$ is the contribution to $\hat{E}_s$ of the initial state of the radiation field in the absence of the dielectric; $\mu_0$ denotes the vacuum permeability and $\epsilon_0$ the vacuum permittivity.

The paper is organized as it follows. In Sec. \[\text{III}\] we introduce the Hamiltonian formulation of the classical electrodynamic problem in the Coulomb gauge for an absorptive dielectric of finite size. In Sec. \[\text{III}\] we first quantize the matter and radiation field, and the corresponding conjugate momenta. Then, we represent the matter fields in terms of the static longitudinal and transverse modes of the dielectric body, and the radiation field in terms of the transverse plane wave in free space. In Sec. \[\text{IV}\] we express the Hamiltonian operator of the system in terms of the coordinate operators and conjugate momentum operators of the matter and radiation fields. In Sec. \[\text{V}\] we first derive the Heisenberg equations of motion for the coordinate operators and and conjugate momentum operators, and then reduce the full set of equations to a system of differential - integral equations of convolution type for the
coordinate operators of the matter field. In Sec. VI we give the equations governing the evolution of the coordinates of the polarization field operator in the Laplace domain, we solve them, and we express the polarization field operator in terms of the driving field operator of the system. In Sec. VII we give the expression of the electric field operator in terms of the polarization field operator. We discuss the main results in Sec. VIII.

II. CLASSICAL FIELD EQUATIONS

We consider an absorptive and homogeneous dielectric of finite size. We denote with $V$ the region occupied by the dielectric, $\partial V$ its boundary, $\mathbf{n}$ the (unit vector) normal to $\partial V$ pointing outward, $a$ the radius of the smallest sphere that contains $V$ and $V_\infty$ the entire unbounded space. An electric field $\mathbf{E}_{\text{inc}}(\mathbf{r};t)$, which is solenoidal in $V$, is applied for $t > 0$.

The electric polarization describes the macroscopic state of the dielectric. Due to the homogeneity of the dielectric, the polarization density field $\mathbf{P}(\mathbf{r};t)$ is solenoidal in $V$ but its normal component to $\partial V$ is different from zero. Therefore, a surface polarization charge lies on the dielectric surface with surface density given by $\mathbf{P} \cdot \mathbf{n}$. We denote with $\mathbf{E}(\mathbf{r};t)$ the electric field and with $\mathbf{B}(\mathbf{r};t)$ the magnetic field generated by the induced polarization density field (i.e., the induced electromagnetic field).

The macroscopic electromagnetic response of the dielectric is described for $t \geq 0$ by the equation

$$
\mathbf{P}(\mathbf{r};t) = \begin{cases} 
\epsilon_0 \int_0^\infty h_\chi(t-\tau)[\mathbf{E}(\mathbf{r};\tau) + \mathbf{E}_{\text{inc}}(\mathbf{r};\tau)]d\tau + \mathbf{P}_0(\mathbf{r};t) & \text{in } V, \\
0 & \text{in } V_\infty \setminus V,
\end{cases}
$$

(1)

where $\epsilon_0$ is the vacuum permittivity, $h_\chi(t)$ is the inverse Fourier transform of the susceptibility of the dielectric $\chi(\omega)$,

$$
h_\chi(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \chi(\omega)e^{i\omega t}d\omega,
$$

(2)

and $\mathbf{P}_0(\mathbf{r};t)$ is the free evolution of the polarization density field (in the absence of the interaction with the electromagnetic field), taking into account the contribution of the initial state of the polarization.

The real part of the susceptibility $\chi_r(\omega)$ is an even function of $\omega$, whereas the imaginary part $\chi_i(\omega)$ is an odd function, so that $\chi(-\omega) = \chi^*(\omega)$. Since the dielectric is dissipative,
\( \chi_i(\omega) \) is negative for \( \omega > 0 \). The causality implies that \( h_\chi(t) = 0 \) for \( t < 0 \), and in the frequency domain it implies that \( \chi(\omega) \) obey the Kramers–Kronig relations \((-\infty < \omega < +\infty)\)

\[
\chi_r(\omega) = \frac{2}{\pi} \mathcal{P} \int_{0}^{\infty} \frac{\omega' \chi_i(\omega')}{\omega'^2 - \omega^2} d\omega',
\]

\[
\chi_i(\omega) = -\frac{2\omega}{\pi} \mathcal{P} \int_{0}^{\infty} \frac{\chi_r(\omega')}{\omega'^2 - \omega^2} d\omega',
\]

where \( \mathcal{P} \) denotes the Cauchy principal value.

To study the electrodynamics of absorptive dielectrics through a Hamiltonian formulation we model the medium as a continuum set of harmonic oscillators with natural frequency \( \nu \), where \( 0 < \nu < \infty \). Each harmonic oscillator is described by the vector field \( Y_\nu(\mathbf{r}; t) \) defined in \( V \); throughout the paper we denote the continuum set \( \{Y_\nu(\mathbf{r}; t)\} \) as “matter field”. In the region \( V \), the polarization density field is expressed in terms of the matter field as (e.g., [12], [13])

\[
P = \int_{0}^{\infty} \alpha_\nu Y_\nu d\nu
\]

where \( \alpha_\nu \) is the coupling parameter between the matter field and the electromagnetic field. The choice

\[
\alpha_\omega = \sqrt{\frac{2}{\pi} \sigma(\omega)}
\]

returns the constitutive relation \( \mathbb{1} \) in \( V \) (see Appendix \( \mathbb{2} \)). For absorptive dielectrics \( \sigma(\omega) \) is positive for \( \omega > 0 \). The quantity \( \alpha_\nu^2 / \epsilon_0 \) is homogeneous with the frequency, and the quantity \( \sigma(\omega) \) is homogeneous with the electrical conductivity.

The free evolution term \( P_0(\mathbf{r}; t) \) is given by

\[
P_0(\mathbf{r}; t) = \int_{0}^{\infty} \sqrt{\frac{2\sigma(\omega)}{\pi}} Y_{\omega}^{(0)}(\mathbf{r}; t) d\omega
\]

where

\[
Y_{\omega}^{(0)}(\mathbf{r}; t) = Y_\omega(\mathbf{r}; 0) \cos(\omega t) + \frac{1}{\omega} \dot{Y}_\omega(\mathbf{r}; 0) \sin(\omega t),
\]

\( Y_\omega(\mathbf{r}; 0) \) is the initial value of \( Y_\omega(\mathbf{r}; t) \) and \( \dot{Y}_\omega(\mathbf{r}; 0) \) is the initial value of \( \dot{Y}_\omega(\mathbf{r}; t) \).
A. Lagrangian in the Coulomb gauge

Throughout this manuscript, we use the scalar product

$$\langle \mathbf{F}, \mathbf{G} \rangle_W = \int_W \mathbf{F}^* (\mathbf{r}) \cdot \mathbf{G} (\mathbf{r}) \, d^3 \mathbf{r},$$

(10)

and the norm \( \| \mathbf{F} \|_W = \sqrt{\langle \mathbf{F}, \mathbf{F} \rangle_W} \). The scalar product is defined in \( V \) if the domain is not explicitly indicated.

It is convenient to represent the electric field \( \mathbf{E} \) in \( V_\infty \) as

$$\mathbf{E} = \mathbf{E}_s + \mathbf{E}_c$$

(11)

where \( \mathbf{E}_s (\mathbf{r}; t) \) is the solenoidal component in \( V_\infty \) (radiation field), and \( \mathbf{E}_c (\mathbf{r}; t) \) is the irrotational component (Coulomb field). This is the Helmholtz decomposition for vector fields defined in \( V_\infty \). The vector fields \( \mathbf{E}_s \) and \( \mathbf{E}_c \) are orthogonal according to the scalar product

$$\langle \mathbf{E}_s, \mathbf{E}_c \rangle_{V_\infty}.$$

We introduce the vector potential \( \mathbf{A} (\mathbf{r}; t) \) in the Coulomb gauge,

$$\nabla \cdot \mathbf{A} = 0 \quad \text{in} \ V_\infty,$$

(12)

to represent the solenoidal components of the electromagnetic field. We have

$$\mathbf{E}_s = -\dot{\mathbf{A}},$$

(13)

$$\mathbf{B} = \nabla \times \mathbf{A},$$

(14)

where dot denotes the partial derivative with respect to time.

The Coulomb electric field is given by

$$\mathbf{E}_c (\mathbf{r}; t) = -\frac{1}{4 \pi \varepsilon_0} \nabla \int_{\partial V} \frac{P_n (\mathbf{r}'; t)}{|\mathbf{r} - \mathbf{r}'|} (d^2 \mathbf{r}') \quad \text{in} \ V_\infty,$$

(15)

where \( P_n = \mathbf{P} \cdot \mathbf{n} \) on \( \partial V \). The field \( \mathbf{E}_c \) solenoidal in \( V \) and \( V_\infty \setminus V \) but its normal component to \( \partial V \) is discontinuous due to the surface polarization charge of the dielectric.

The degrees of freedom of the entire system are the matter field \( \{ \mathbf{Y}_\nu (\mathbf{r}; t) \} \) and the vector potential \( \mathbf{A} (\mathbf{r}; t) \) (\( \nu \) and \( \mathbf{r} \) are the labels of the degrees of freedom). The Lagrangian in the Coulomb gauge is the sum of four terms: the matter term \( \mathcal{L}_{\text{mat}} \), the Coulomb term \( \mathcal{L}_{\text{Cou}} \), the radiation term \( \mathcal{L}_{\text{rad}} \), and the interaction term between matter and radiation \( \mathcal{L}_{\text{int}} \) (e.g., [26], [10], [13], [25]),

$$\mathcal{L} = \mathcal{L}_{\text{mat}} + \mathcal{L}_{\text{Cou}} + \mathcal{L}_{\text{rad}} + \mathcal{L}_{\text{int}},$$

(16)
where

\[ L_{\text{mat}}(Y_\nu, \dot{Y}_\nu) = \int_V d^3r \int_0^\infty d\nu \left( \frac{1}{2} \dot{Y}_\nu^2 - \frac{\nu^2}{2} Y_\nu^2 \right), \]

\[ L_{\text{Cou}}(Y_\nu) = -\int_{\partial V} d^2r \int_{\partial V} d^2r' \frac{1}{2\epsilon_0} \frac{P_n(r; t)P_n(r'; t)}{4\pi|\mathbf{r} - \mathbf{r}'|}, \]

\[ L_{\text{rad}}(\mathbf{A}, \dot{\mathbf{A}}) = \int_{V_\infty} d^3r \left[ \frac{\epsilon_0}{2} \dot{\mathbf{A}}^2 - \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 \right], \]

\[ L_{\text{int}}(\dot{Y}_\nu, \mathbf{A}) = \int_V \dot{\mathbf{P}} \cdot (\mathbf{A} + \mathbf{A}_{\text{inc}}) d^3r, \]

\[ \mathbf{P} \cdot \mathbf{n} \text{ and } \mathbf{P} \text{ are functions of } Y_\nu \text{ (see relation 5)}; \]

\[ \mathbf{A}_{\text{inc}}(\mathbf{r}; t) = -\int_0^t \mathbf{E}_{\text{inc}}(\mathbf{r}; \tau) d\tau + \mathbf{A}_{\text{inc}}(\mathbf{r}; t = 0) \]

is the incident vector potential.

\[ \mathbf{B. \ Canonical \ variables \ and \ Hamiltonian} \]

We now introduce the conjugate momenta of matter and radiation fields in the Coulomb gauge. The momentum conjugate \( Q_\nu(\mathbf{r}; t) \) to the matter field \( Y_\nu(\mathbf{r}; t) \) is given by

\[ Q_\nu = \dot{Y}_\nu + \alpha_\nu(\mathbf{A} + \mathbf{A}_{\text{inc}}) \text{ in } V, \]

and the momentum conjugate to the vector potential is given by

\[ \Pi = \epsilon_0 \dot{\mathbf{A}} \text{ in } V_\infty. \]

The Hamiltonian has three terms: the contribution of the matter \( H_{\text{mat}} \), the contribution of the Coulomb interaction \( H_{\text{Cou}} \) and the contribution of the radiation field \( H_{\text{rad}} \),

\[ H = H_{\text{mat}} + H_{\text{Cou}} + H_{\text{rad}}, \]

where

\[ H_{\text{mat}}(Q_\nu, Y_\nu, \mathbf{A}) = \int_V d^3r \int_0^\infty d\nu \left\{ \frac{1}{2} [Q_\nu - \alpha_\nu(\mathbf{A} + \mathbf{A}_{\text{inc}})]^2 + \frac{\nu^2}{2} Y_\nu^2 \right\}, \]

\[ H_{\text{Cou}}(Y_\nu) = \int_{\partial V} d^2r \int_{\partial V} d^2r' \frac{1}{2\epsilon_0} \frac{P_n(r; t)P_n(r'; t)}{4\pi|\mathbf{r} - \mathbf{r}'|}, \]

\[ H_{\text{rad}}(\Pi, \mathbf{A}) = \int_{V_\infty} d^3r \left[ \frac{1}{2\epsilon_0} \Pi^2 + \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 \right]. \]

The Hamilton’s equations for the matter field and the conjugate momentum are in \( V \), for \( 0 < \nu < \infty \),

\[ \dot{Y}_\nu = Q_\nu - \alpha_\nu(\mathbf{A} + \mathbf{A}_{\text{inc}}), \]

\[ \dot{Q}_\nu = -\nu^2 Y_\nu + \alpha_\nu \mathbf{E}_c \{Y_\nu\} \].

11
The Hamilton’s equations for the radiation field and the conjugate momentum are in $V_\infty$

\[
\dot{\mathbf{A}} = \frac{1}{\varepsilon_0} \Pi , \quad (23a)
\]
\[
\dot{\Pi} = -\frac{1}{\mu_0} \nabla^2 \mathbf{A} + \left( \dot{\mathbf{P}} + \varepsilon_0 \mathbf{E}_c \{ Y_\nu \} \right) . \quad (23b)
\]

We recall that the Coulomb field $\mathbf{E}_c$ is expressed as function of the normal component of $\mathbf{P}$ on $\partial V$ through relation (15); $\mathbf{P}$, in turn, is expressed as function of $Y_\nu$ through relation (5).

The second term on the left hand side of Eq. 23b (between round brackets) is the solenoidal component of the current density field in $V_\infty$. From equations 22a-23b we obtain

\[
\ddot{Y}_\nu + \nu^2 Y_\nu = \alpha_\nu \left[ -(\dot{\mathbf{A}} + \dot{\mathbf{A}}_{\text{inc}}) + \mathbf{E}_c \{ Y_\nu \} \right] \text{ in } V , \quad (24)
\]
\[
\dddot{\mathbf{A}} - \varepsilon_0^2 \nabla^2 \mathbf{A} = \frac{1}{\varepsilon_0} \left( \dot{\mathbf{P}} + \varepsilon_0 \mathbf{E}_c \{ Y_\nu \} \right) \text{ in } V_\infty . \quad (25)
\]

In Appendix B we derive the constitutive relation (1) with $\alpha_\nu$ given by (7) by solving Eq. 24.

III. QUANTIZATION AND EXPANSION OF THE VECTOR FIELD OPERATORS

In this section, we first quantize the matter and radiation fields in a standard fashion (e.g., [10, 13, 26]) by enforcing the canonical commutation relations between the field operators and their conjugate momenta. Then, we introduce the bases for representing them. We assume that the incident electromagnetic field is a classical field.

A. Quantization

The vector field operators $\hat{\mathbf{Q}}_\nu (\mathbf{r})$ and $\hat{Y}_\nu (\mathbf{r})$ correspond to the canonically conjugate matter vector fields $\mathbf{Q}_\nu$ and $Y_\nu$; the vector field operators $\hat{\Pi} (\mathbf{r})$ and $\hat{\mathbf{A}} (\mathbf{r})$ correspond to the radiation vector fields $\Pi$ and $\mathbf{A}$. These are the fundamental vector field operators of the problem. They obey the commutation relations:

\[
\left[ \hat{\mathbf{Q}}_\nu (\mathbf{r}), \hat{Y}_{\nu'} (\mathbf{r}') \right] = -i\hbar \hat{\mathbf{I}} \delta (\nu - \nu') \delta (\mathbf{r} - \mathbf{r}') \quad \mathbf{r}, \mathbf{r}' \in V , \quad (26)
\]
\[
\left[ \hat{\Pi} (\mathbf{r}), \hat{\mathbf{A}} (\mathbf{r}') \right] = -i\hbar \delta^\perp (\mathbf{r} - \mathbf{r}') \quad \mathbf{r}, \mathbf{r}' \in V_\infty , \quad (27)
\]

while all other commutators vanish; here $\hat{\mathbf{I}}$ is the three-dimensional unit tensor, $\delta^\perp (\mathbf{r}) = \hat{\mathbf{I}} \delta (\mathbf{r}) - \delta^\parallel (\mathbf{r})$ and $\delta^\parallel (\mathbf{r}) = \nabla \nabla (1/4\pi r)$ (e.g., [20]).
We also introduce the vector field operator $\hat{P}(\mathbf{r})$ corresponding to the polarization density field $\mathbf{P}$,

$$\hat{P}(\mathbf{r}) = \int_0^\infty \alpha_\nu \hat{Y}_\nu(\mathbf{r}) d\nu, \quad (28)$$

and the vector field operator

$$\hat{Z}(\mathbf{r}) = \int_0^\infty \alpha_\nu \hat{Q}_\nu(\mathbf{r}) d\nu. \quad (29)$$

They obey the commutation relation

$$\left[ \hat{Z}(\mathbf{r}), \hat{P}(\mathbf{r}') \right] = -i\hbar a_0 \mathbf{1} \delta(\mathbf{r} - \mathbf{r}') \quad \mathbf{r}, \mathbf{r}' \in V \quad (30)$$

where

$$a_0 = \int_0^\infty \alpha_\nu^2 d\nu. \quad (31)$$

The operator $\hat{Z}(\mathbf{r})$ is canonically conjugate to $\hat{P}(\mathbf{r})$.

\textbf{B. Expansion of the matter operators}

We represent the matter field operators, which are defined in $V$, by applying the Helmholtz decomposition for vector fields defined in a bounded domain. The vector field $\hat{Y}_\nu$, for any $\nu$ is expressed as

$$\hat{Y}_\nu = \hat{Y}^\parallel_\nu + \hat{Y}^\perp_\nu, \quad (32)$$

where $\hat{Y}^\parallel_\nu$ is the longitudinal component of $\hat{Y}_\nu$, that is curl free and solenoidal in $V$ and its normal component to $\partial V$ is equal to $\hat{Y}^\parallel_\nu \cdot \mathbf{n}$; $\hat{Y}^\perp_\nu$ is the transverse component of $\hat{Y}_\nu$ that is solenoidal in $V$ and its normal component on $\partial V$ is equal to zero, while its curl is different from zero. This decomposition is unique. The vector fields $\hat{Y}^\parallel_\nu$ and $\hat{Y}^\perp_\nu$ are orthogonal according to the scalar product $\langle \hat{Y}^\parallel_\nu, \hat{Y}^\perp_\nu \rangle$. We represent the vector field operators $\{\hat{Q}_\nu\}$ in the same way.

By following [25], we now expand the longitudinal components of $\hat{Y}_\nu$ and $\hat{Q}_\nu$ in terms of the static longitudinal modes of the dielectric body $\{U^\parallel_m(\mathbf{r})\}$, and the transverse components in terms of the static transverse modes of the body $\{U^\perp_m(\mathbf{r})\}$. The longitudinal modes $\{U^\parallel_m(\mathbf{r})\}$ are the eigenfunctions of the electrostatic integral operator defined in Eq[A1] of the Appendix [A]. The transverse modes $\{U^\perp_m(\mathbf{r})\}$ are the eigenfunctions of the magnetostatic
integral operator defined in Eq. A2 of the Appendix A. Both the integral operators have a
discrete spectrum. The longitudinal modes and transverse modes are orthonormal according
to the scalar product $\langle \mathbf{F}, \mathbf{G} \rangle$; they are also mutually orthogonal. The set of modes $\{ \mathbf{U}_m^\parallel (\mathbf{r}) \}$
is a base for the space of longitudinal vector fields defined on $\mathcal{V}$, and the set of modes
$\{ \mathbf{U}_m^\perp (\mathbf{r}) \}$ is a base for the space of transverse vector fields defined on $\mathcal{V}$. These modes
satisfy the closure relation
\[
\sum_{m=1,2,...} \left[ \mathbf{U}_m^\parallel (\mathbf{r}) \otimes \mathbf{U}_m^\parallel (\mathbf{r}') + \mathbf{U}_m^\perp (\mathbf{r}) \otimes \mathbf{U}_m^\perp (\mathbf{r}') \right] = \hat{\mathbf{I}} \delta(\mathbf{r} - \mathbf{r}') \quad \mathbf{r}, \mathbf{r}' \in \mathcal{V}. 
\] (33)

Both sets of modes are dimensionless quantities.

The vector field operators $\hat{\mathbf{Q}}_\nu^a$ and $\hat{\mathbf{Y}}_\nu^a$, with $a = \parallel, \perp$, are represented as:
\[
\hat{\mathbf{Y}}_\nu^a(\mathbf{r}) = \sum_{m=1,2,...} \hat{y}_\nu^a U_m^a(\mathbf{r}), 
\] (34)
\[
\hat{\mathbf{Q}}_\nu^a(\mathbf{r}) = \sum_{m=1,2,...} \hat{q}_\nu^a U_m^a(\mathbf{r}), 
\] (35)

where $\{ \hat{y}_\nu^a \}$ is the set of the coordinate operators of $\hat{\mathbf{Y}}_\nu^a$; $\{ \hat{q}_\nu^a \}$ is the set of the coordinate
operators of $\hat{\mathbf{Q}}_\nu^a$. Since the static longitudinal and transverse modes are real functions, the
coordinate operators are Hermitian. They obey the commutation relations
\[
[\hat{q}_{\nu,m}', \hat{y}_{\nu,m}] = -i\hbar \delta'(\nu' - \nu) \delta_{m'm} \delta_{ab}, 
\] (36)
for $m, m' = 1, 2, 3, \ldots$ and for $a, b = \parallel, \perp$, while all other commutators vanish. The operator
$\hat{q}_m^a$ is canonically conjugate to the operator $\hat{y}_m^a$.

In the region $\mathcal{V}$, the polarization field operator $\hat{\mathbf{P}}$ and its conjugate momentum $\hat{\mathbf{Z}}$ are
expressed as
\[
\hat{\mathbf{P}}(\mathbf{r}) = \sum_{m=1,2,...} \left[ \hat{p}_m^\parallel \mathbf{U}_m^\parallel (\mathbf{r}) + \hat{p}_m^\perp \mathbf{U}_m^\perp (\mathbf{r}) \right], 
\] (37)
\[
\hat{\mathbf{Z}}(\mathbf{r}) = \sum_{m=1,2,...} \left[ \hat{z}_m^\parallel \mathbf{U}_m^\parallel (\mathbf{r}) + \hat{z}_m^\perp \mathbf{U}_m^\perp (\mathbf{r}) \right], 
\] (38)

where
\[
\hat{p}_m^a = \int_0^\infty \alpha_\nu \hat{y}_{\nu,m}^a d\nu, 
\] (39)
\[
\hat{z}_m^a = \int_0^\infty \alpha_\nu \hat{q}_{\nu,m}^a d\nu. 
\] (40)
and \( a = \|, \perp \). The coordinate operators \( \hat{p}_m^a \) and \( \hat{z}_m^a \) obey the commutation relations

\[
[\hat{z}_m^a, \hat{p}_m^b] = -i\hbar a_0 \delta_{m'm} \delta_{ab} \quad (41)
\]

for \( m, m' = 1, 2, 3, \ldots \) and \( a, b = \|, \perp \). The operator \( \hat{z}_m^a \) is canonically conjugate to the operator \( \hat{p}_m^a \).

C. Expansion of the radiation field operators

We use the transverse plane wave modes

\[
w_{\mu}(r) = \frac{1}{(2\pi)^{3/2} \epsilon_{s,k}} e^{i k \cdot r} \quad (42)
\]

to represent the radiation field operators \( \hat{A}(r) \) and \( \hat{\Pi}(r) \); \( k \in \mathbb{R}^3 \) is the propagation vector, \( \{\epsilon_{s,k}\} \) are the polarization unit vectors with \( \epsilon_{s,k} = \epsilon_{s,-k} \) and \( s = 1, 2; \mu \) is a multi-index corresponding to the pair of parameters \( k \) and \( s, \mu = (k, s) \), \( \mathcal{M} \) represents the set of all possible \( \mu \), and \( \sum_\mu (\cdot) \) denotes \( \sum_{\mu \in \mathcal{M}} \int_{\mathbb{R}^3} d^3k \ (\cdot) \). The two polarization vectors are orthogonal among them, \( \epsilon_{1,k} \cdot \epsilon_{2,k} = 0 \), and are both transverse to the propagation vector, \( \epsilon_{1,k} \cdot k = \epsilon_{2,k} \cdot k = 0 \). The functions \( \{w_\mu\} \) are orthonormal:

\[
\langle w_{\mu'}, w_\mu \rangle = \delta_{s',s} \delta(k - k') \quad (43)
\]

These modes are also dimensionless quantities.

We represent \( \hat{A}(r) \) and \( \hat{\Pi}(r) \) as:

\[
\hat{A}(r) = \sum_{\mu \in \mathcal{M}} \hat{A}_\mu w_\mu(r), \quad (44)
\]
\[
\hat{\Pi}(r) = \sum_{\mu \in \mathcal{M}} \hat{\Pi}_\mu w_\mu(r), \quad (45)
\]

where \( \{\hat{A}_\mu\} \) is the set of the coordinate operators of \( \hat{A} \) and \( \{\hat{\Pi}_\mu\} \) is the set of the coordinate operators of \( \hat{\Pi} \). Since \( \hat{A} \) and \( \hat{\Pi} \) are Hermitian, and the modes \( \{w_\mu\} \) are complex with \( w_\mu^* = w_{-\mu} \), we have \( \hat{A}_\mu^\dagger = \hat{A}_{-\mu} \) and \( \hat{\Pi}_\mu^\dagger = \hat{\Pi}_{-\mu} \). The operators \( \{\hat{A}_\mu\} \) and \( \{\hat{\Pi}_\mu\} \) obey the commutation relations

\[
[\hat{\Pi}_{\mu'}, \hat{A}_\mu^\dagger] = -i\hbar \delta(\mu' - \mu), \quad (46)
\]
for any couple \( \mu, \mu' \in \mathcal{M} \), while all other commutators vanish. The operator \( \hat{\Pi}_\mu \) is canonically conjugate to the operator \( \hat{A}_\mu^\dagger \). The coordinate operators of the radiation fields commutate with the coordinate operators of the matter and bath fields.

We represent the solenoidal vector potential describing the incident classical field in the same way. We indicate with \( \{ A_{\mu}^{\text{inc}} \} \) the coordinates of this field.

**IV. MODAL EXPANSION OF THE HAMILTONIAN OPERATOR**

The Hamiltonian operator is given by

\[
\hat{H} = \hat{H}_{\text{Cou}} + \hat{H}_{\text{rad}} + \hat{H}_{\text{mat}}
\]

where \( \hat{H}_{\text{mat}} \), \( \hat{H}_{\text{Cou}} \) and \( \hat{H}_{\text{rad}} \) are obtained from 21a-21c by substituting each physical variable with the corresponding operator. Now we express them in terms of the expansions 34, 35, 44 and 45.

**A. Coulomb energy**

Only the longitudinal component of the matter field contributes to the Coulomb interaction energy \( \hat{H}_{\text{Cou}} \). We have

\[
\hat{H}_{\text{Cou}} = \sum_{m=1,2,...} \int_0^\infty d\nu \int_0^\infty d\nu' \frac{\alpha_\nu \alpha_{\nu'}}{2\varepsilon_0 \kappa_m^\|} \hat{y}_{\nu,m} \hat{y}_{\nu',m}^\dagger,
\]

where \( \kappa_m^\| \) is the eigenvalue associated to \( U_m^\| \) (see Eq. A1 of the Appendix A). By using 39 we obtain

\[
\hat{H}_{\text{Cou}} = \sum_{m=1,2,...} \frac{1}{2\varepsilon_0 \kappa_m} \hat{p}_m^2.
\]

The static longitudinal modes of the dielectric body diagonalize the Coulomb interaction energy [16].

**B. Radiation energy**

The expression of \( \hat{H}_{\text{rad}} \) in terms of the canonically conjugate coordinate operators of the radiation field is

\[
\hat{H}_{\text{rad}} = \sum_{\mu \in \mathcal{M}} \left( \frac{1}{2\varepsilon_0} \hat{\Pi}_\mu^\dagger \hat{\Pi}_\mu + \frac{\varepsilon_0 \omega_\mu^2}{2} \hat{A}_\mu^\dagger \hat{A}_\mu \right).
\]
where

$$\omega_\mu^2 = c_0^2 k_\mu^2.$$  \hspace{1cm} (51)

The transverse plane wave modes diagonalize $\hat{H}_{\text{rad}}$.

C. Matter energy

The matter term $\hat{H}_{\text{mat}}$ has three contributions:

$$\hat{H}_{\text{mat}} = \hat{H}_{\text{mat}}' + \hat{H}_{\text{mat}}'' + \hat{H}_{\text{mat}}'''$$  \hspace{1cm} (52)

where

$$\hat{H}_{\text{mat}}' = \sum_{m=1,2,\ldots} \int_0^\infty d\nu \left[ \frac{1}{2} (\hat{q}_{\nu,m}^2 + \hat{q}^\perp_{\nu,m}) + \frac{\nu^2}{2} (\hat{y}_{\nu,m}^2 + \hat{y}^\perp_{\nu,m}) \right],$$

$$\hat{H}_{\text{mat}}'' = -\sum_{m=1,2,\ldots} \int_0^\infty d\nu \alpha_\nu \left( \hat{q}_{\nu,m} R_{\nu,m}^|| + \hat{q}^\perp_{\nu,m} R_{\nu,m}^\perp \right) \left( \hat{A}_\mu + A^\text{inc}_\mu \right),$$

$$\hat{H}_{\text{mat}}''' = \int_0^\infty \frac{\alpha^2_\nu}{2} d\nu \sum_{\mu',\mu \in \mathcal{M}} W_{\mu',\mu} (\hat{A}_{\mu'} + A_{\mu'}^\text{inc})^\dagger (\hat{A}_{\mu} + A_{\mu}^\text{inc}),$$

and

$$R_{m\mu}^a = \langle U_m^a, w_\mu \rangle,$$  \hspace{1cm} (56)

$$W_{\mu',\mu} = \langle w_{\mu'}, w_\mu \rangle,$$  \hspace{1cm} (57)

$$A_{\mu}^\text{inc}(t) = \langle w_\mu, A^\text{inc} \rangle V_\infty,$$  \hspace{1cm} (58)

with $a = ||, \perp$. The longitudinal and transverse modes diagonalize the first term, $\hat{H}_{\text{mat}}'$. The second term, $\hat{H}_{\text{mat}}''$, takes into account the interaction between matter and radiation fields. The third term, $\hat{H}_{\text{mat}}'''$, is the diamagnetic contribution to the energy of the matter.

The terms $\hat{H}_{\text{Cou}}, \hat{H}_{\text{rad}}$ and $\hat{H}_{\text{mat}}'$ are diagonal because of the expansion bases we have used, whereas $\hat{H}_{\text{mat}}''$ and $\hat{H}_{\text{mat}}'''$ are not diagonal. As we shall see, we introduce a formulation that allows us to eliminate the diamagnetic term without making any approximation. Therefore, we only have to address the difficulties arising from the nondiagonal term $\hat{H}_{\text{mat}}''$. The use of the static longitudinal and transverse modes of the dielectric object allows us to overcome these difficulties. In particular, as in the classical regime, a limited set of static longitudinal and transverse modes are needed for objects with size of the order of the characteristic wavelength accounting for the strength of the coupling between the matter and electromagnetic field.
V. HEISENBERG PICTURE

In this section, we first formulate the equations of motion for the coordinate operators of the matter field and the radiation field, in the Heisenberg picture. Then, we eliminate the coordinate operators of the radiation field and derive the equation of motion for the coordinate operators of the matter field.

A. Heisenberg’s equations for the matter and radiation fields

The Heisenberg’s equation for the generic time invariant operator $\hat{u}$ of the system is

$$\dot{\hat{u}} = \frac{1}{i\hbar} [\hat{u}, \hat{H}]$$

where the Hamiltonian is given by [47].

1. Matter

The equations governing the time evolution of $\hat{y}_\nu,m$ and $\hat{q}_\nu,m$, with $m = 1, 2, 3 \ldots$ and $0 < \nu < \infty$, are

$$\dot{\hat{y}}_\nu,m = \hat{\mathcal{A}}_\nu,m - \alpha_\nu \sum_{\mu \in \mathcal{M}} \mathcal{R}_{\mu m} (\hat{A}_\mu + \hat{A}^{inc}_\mu),$$

$$\dot{\hat{q}}_\nu,m = -\nu^2 \hat{y}_\nu,m - \frac{\alpha_\nu}{\epsilon_0 \kappa_m} \int_0^\infty \alpha_\nu \hat{y}_\nu,m d\nu.$$  

(60a)

(60b)

The equations governing the time evolution of $\hat{y}_\nu,m$ and $\hat{q}_\nu,m$ are

$$\dot{\hat{y}}_\nu,m = \hat{\mathcal{A}}_\nu,m - \alpha_\nu \sum_{\mu \in \mathcal{M}} \mathcal{R}_{\mu m} (\hat{A}_\mu + \hat{A}^{inc}_\mu),$$

$$\dot{\hat{q}}_\nu,m = -\nu^2 \hat{y}_\nu,m.$$  

(61a)

(61b)

By combining equations 60a,61b we eliminate the conjugate momenta $\hat{q}_\nu,m$ and $\hat{q}_\nu,m$, and obtain

$$\dot{\hat{y}}_\nu,m + \nu^2 \hat{y}_\nu,m + \frac{\alpha_\nu}{\epsilon_0 \kappa_m} \int_0^\infty \alpha_\nu \hat{y}_\nu,m d\nu = -\alpha_\nu \sum_{\mu \in \mathcal{M}} \mathcal{R}_{\mu m} (\hat{A}_\mu + \hat{A}^{inc}_\mu),$$

$$\dot{\hat{y}}_\nu,m + \nu^2 \hat{y}_\nu,m = -\alpha_\nu \sum_{\mu \in \mathcal{M}} \mathcal{R}_{\mu m} (\hat{A}_\mu + \hat{A}^{inc}_\mu).$$  

(62)

(63)

Once $\{\hat{y}_\nu,m\}$, $\{\hat{y}_\nu,m\}$ and $\{\hat{A}_\mu\}$ have been evaluated, equations 60a and 61a allow to calculate the conjugate momenta $\{\hat{q}_\nu,m\}$ and $\{\hat{q}_\nu,m\}$. 

18
2. Radiation

The equations governing the time evolution of \( \hat{A}_\mu \) and \( \hat{\Pi}_\mu \), with \( \mu \) belonging to \( \mathcal{M} \), are

\[
\dot{\hat{A}}_\mu = \frac{1}{\varepsilon_0} \hat{\Pi}_\mu, \\
\dot{\hat{\Pi}}_\mu = -\varepsilon_0 \omega^2_\mu \hat{A}_\mu + \sum_{m=1,2,...}^{a=||,\perp} \int_0^\infty d\nu \alpha_\nu R^a_\mu m \hat{y}^a_\nu m - \int_0^\infty \alpha^2_\nu d\nu \sum_{\mu' \in \mathcal{M}} W^\mu_\mu' (\hat{A}_{\mu'} + A^\text{inc}_{\mu'})
\]

(64a)

(64b)

where

\[
R^a_\mu m = \langle w_\mu, U^a_m \rangle = (R^a_\mu m)^*. 
\]

(65)

By combining equations 60a, 61a, 64a, 64b and by using the closure relation 33 we eliminate the conjugate momenta \( \hat{\Pi}_\mu, \hat{q}^\parallel_\nu m \) and \( \hat{q}^\perp_\nu m \), and obtain

\[
\ddot{\hat{A}}_\mu + \omega^2_\mu \hat{A}_\mu = \frac{1}{\varepsilon_0} \sum_{m=1,2,...}^{a=||,\perp} \int_0^\infty d\nu \alpha_\nu R^a_\mu m \dot{\hat{y}}^a_\nu m. 
\]

(66)

Note that in this equation the coupling terms associated to \( W^\mu_\mu' \) have disappeared without any approximation. This is a mere consequence of the fact that we have eliminated the conjugate momenta by substitution. Once the operators \( \{ \hat{A}_\mu \} \) have been evaluated, equation 64a allow to calculate the conjugate momenta operators \( \{ \hat{\Pi}_\mu \} \).

B. Equations of Motion for the Matter Coordinate Operators

We now derive the equations governing the dynamics of the coordinate operators of the matter field in the time domain, then, we rewrite them in the Laplace domain.

1. Time domain

To eliminate the operators \( \{ \hat{A}_\mu \} \) into the systems of equations 62 and 63 we need to express them as function of the operators \( \{ \hat{y}^\parallel_m \} \) and \( \{ \hat{y}^\perp_m \} \). By solving equation 66 we obtain

\[
\dot{\hat{A}}_\mu = \frac{1}{\varepsilon_0} \sum_{m=1,2,...}^{a=||,\perp} R^a_\mu m \int_0^\infty d\nu \alpha_\nu w^\omega_\mu (t) * \dot{\hat{y}}^a_\nu m (t) - \dot{\hat{E}}_\mu, 
\]

(67)

where \( u_1(t) * u_2(t) \) denotes the convolution integral of \( u_1(t) \) and \( u_2(t) \),

\[
w^\omega_\mu (t) = \theta (t) \cos(\omega_\mu t),
\]

(68)
\( \theta(t) \) is the Heaviside function,

\[
\hat{E}_\mu(t) = \omega_\mu \hat{A}_\mu(0) \sin(\omega_\mu t) - \frac{1}{\varepsilon_0} \hat{\Pi}_\mu(0) \cos(\omega_\mu t),
\]

(69)

\( \hat{A}_\mu(0) \) and \( \hat{\Pi}_\mu(0) \) are, respectively, the operators \( \hat{A}_\mu \) and \( \hat{\Pi}_\mu \) in the Schrödinger picture. The operator \( \hat{E}_\mu \) is the coordinate operator of the contribution to \( \hat{E}_s \) due to the initial conditions of the radiation field in the absence of the dielectric body.

By substituting expressions 67 into the differential equations 62 and 63, we obtain the system of integro-differential equations of convolution type for the coordinate operators of the matter field, with \( m = 1, 2, 3, \ldots \), and \( 0 < \nu < \infty \),

\[
\dddot{y}_\parallel^{m} + \nu^2 \ddot{y}_\parallel^{m} + \frac{\alpha_\nu}{\varepsilon_0} \int_0^\infty \alpha_\nu \ddot{y}_\parallel^{m} d\nu + \frac{\alpha_\nu}{\varepsilon_0} \sum_{m'=1,2,\ldots} \int_0^\infty d\nu \alpha_\nu s^{\parallel a}_{mmm'}(t) \ast \dddot{y}_\parallel^{m'}(t) = \alpha_\nu (c_\parallel^{m} + \dot{f}_\parallel^{m}),
\]

(70)

\[
\dddot{y}_\perp^{m} + \nu^2 \ddot{y}_\perp^{m} + \frac{\alpha_\nu}{\varepsilon_0} \sum_{m'=1,2,\ldots} \int_0^\infty d\nu \alpha_\nu s^{\perp a}_{mmm'}(t) \ast \dddot{y}_\perp^{m'}(t) = \alpha_\nu (c_\perp^{m} + \dot{f}_\perp^{m}),
\]

(71)

where \( a = ||, \perp \),

\[
s^{ab}_{mmm'}(t) = \sum_{\mu \in M} \langle U_m^a, w_\mu \rangle \langle w_\mu, U_{m'}^b \rangle w_\omega(t),
\]

(72)

\[
c_\parallel^m(t) = -\langle U_m, \hat{A}_{\text{inc}} \rangle,
\]

(73)

\[
c_\perp^m(t) = \sum_{\mu \in M} R_{\mu m}^{a} \hat{E}_\mu(t).
\]

(74)

The c-functions \( c_\parallel^m(t) \) and \( c_\perp^m(t) \) take into account the action of the incident classical field, and the operators \( \dot{f}_\parallel^m(t) \) and \( \dot{f}_\perp^m(t) \) take into account the contribution due to the initial conditions of the radiation field operators. The kernel \( s^{ab}_{mmm'}(t) \) in the convolution integrals can be expressed as (see Appendix C)

\[
s^{ab}_{mmm'}(t) = \frac{1}{c_0^2} \int_V d^3r \int_V d^3r' U_m^a(r) \hat{\overleftarrow{g}}^{a\perp}(r-r';t) U_{m'}^b(r')
\]

(75)

where \( \hat{\overleftarrow{g}}^{a\perp}(r;t) \) is the transverse Green function in free space for the electric field, and the dot indicates the partial derivative with respect to time; the expression of \( \hat{\overleftarrow{g}}^{a\perp}(r;t) \) and \( \hat{\overleftarrow{g}}^{\perp} \) are given, respectively, by \( \text{C10} \) and \( \text{C11} \). The convolution integrals describe the energy
exchange between the longitudinal and transverse coordinate operators of the matter field that is mediated by the radiation field. This is a nonconservative process due to the energy radiated toward infinity.

Equations 70 and 71 have to be solved with the initial conditions \( \hat{y}_{\nu,m}^a(t=0) = \hat{y}_{\nu,m,0}^a \) and \( \dot{\hat{y}}_{\nu,m}(t=0) = \dot{\hat{y}}_{\nu,m,0} \), with \( a = \parallel, \perp \), where \( \hat{y}_{\nu,m}(0) \) and \( \dot{\hat{y}}_{\nu,m}(0) \) are, respectively, the operators \( \hat{y}_{\nu,m}^a \) and \( \dot{\hat{y}}_{\nu,m}^a \) in the Schrödinger picture.

2. Laplace domain

Let us indicate with \( U(s) \) the (unilateral) Laplace transform of a generic function \( u(t) \), 

\[ U(s) = \mathcal{L}\{u(t)\} = \int_0^\infty u(t) e^{-st} dt. \]

In our problem, the region of convergence includes the imaginary axis because of the matter and radiation losses.

Equations 70 and 71 become in the Laplace domain

\[
\begin{align*}
(s^2 + \nu^2) \hat{Y}_{\nu,m}^\parallel + \frac{\alpha_\nu}{\epsilon_0 \kappa_m} \int_0^\infty \alpha_\nu \hat{Y}_{\nu,m}^\parallel d\nu + \frac{\alpha_\nu}{\epsilon_0} \sum_{m'=1,2,...} \int_0^\infty d\nu \alpha_\nu s S_{mm'}^a \hat{Y}_{\nu,m'}^a = \hat{Y}_{\nu,m}^\parallel, \\
(s^2 + \nu^2) \hat{Y}_{\nu,m}^\perp + \frac{\alpha_\nu}{\epsilon_0} \sum_{m'=1,2,...} \int_0^\infty d\nu \alpha_\nu s S_{mm'}^a \hat{Y}_{\nu,m'}^a = \hat{Y}_{\nu,m}^\perp,
\end{align*}
\]

where: \( \hat{Y}_{\nu,m}(s) \) and \( \hat{Y}_{\nu,m}^\perp(s) \) are the Laplace transform of \( \hat{y}_{\nu,m}^\parallel(t) \) and \( \hat{y}_{\nu,m}^\perp(t) \), respectively; 
\( S_{mm'}^a(s) \) is the Laplace transform of \( s_{mm'}^a(t) \),

\[
S_{mm'}^a(s) = \frac{1}{\epsilon_0} \int_V d^3r \int_V d^3r' \mathbf{U}_m^a(r) \mathbf{G}_\perp(r - r'; s) \mathbf{U}_{m'}^b(r');
\]

\( \mathbf{G}_\perp(r; s) \) is the transverse dyadic Green function for the electric field in the Laplace domain for the free space, whose expression is given by \( \mathbf{C}_6 \). \( \hat{V}_{\nu,m}^a(s) \) is a known operator given by

\[
\hat{V}_{\nu,m}^a = \alpha_\nu (\hat{C}_m^a + \hat{F}_m^a) + \hat{I}_{\nu,m}^a;
\]

\( \hat{C}_m^a(s) \) is the Laplace transform of \( c_{m}^a(t) \), \( \hat{F}_m^a \) is the Laplace transform of \( f_{m}^a \),

\[
\hat{I}_{\nu,m}^a(s) = [s \hat{y}_{\nu,m}^a(0) + \dot{\hat{y}}_{\nu,m}^a(0)] + \frac{\alpha_\nu}{\epsilon_0} \sum_{m'=1,2,...} \sum_{b=\parallel,\perp} \int_0^\infty d\nu \alpha_\nu S_{mm'}^{ab}(s) \hat{y}_{\nu,m'}^{b}(0).
\]

and \( \hat{y}_{\nu,m}^{b}(0) \) are the coordinate operators of the matter field in the Schrödinger picture.

The term \( \hat{L}_{\nu,m}(s) \) takes into account the contribution due to the initial conditions of the
coordinate operators of the matter field. It appears in two different ways: directly, through
the first two terms (in square brackets), and, indirectly, through the last term, where the
effect of the initial condition is mediated by the radiation.

From both the physical and computational point of view it is convenient to express the
transverse dyadic Green function as
\[ \mathbf{G}_\perp(r,s) = \mathbf{g}_\perp(r) + \mathbf{G}^\perp_d(r,s) \] (Appendix C) where
\[ \mathbf{g}_\perp(r) \] is the static transverse dyadic Green function in free space, which diverges as \( 1/r \) for
\( r \to 0 \) and \( \mathbf{G}^\perp_d(r,s) \) is the dynamic part, which is a regular function of \( r \). The extraction of
the singularity \( 1/r \) allows to adopt effective numerical schemes for the computation of the
coefficients \( S_{nm'}^{ab} \). Furthermore, from the definition of the static transverse modes of the
dielectric body (Appendix B) we obtain
\[ S_{nm'}^{\perp\perp}(s) = \frac{s}{c^2_0 \kappa_m^\perp} + \delta S_{nm'}^{\perp\perp}(s), \quad (81) \]
where \( \kappa_m^\perp \) is the eigenvalue associated to the transverse mode \( U_m^\perp(r) \) and
\[ \delta S_{nm'}^{\perp\perp}(s) = \frac{1}{c^2_0} \int_V d^3r \int_V d^3r' U_m^a(r) s \mathbf{G}^\perp_d(r - r'; s) U_{m'}^b(r'). \quad (82) \]
Equation (81) is a consequence of the orthogonality of the static transverse modes of the body.

VI. POLARIZATION FIELD OPERATOR

In this section, we first present the equations governing the dynamics of the polarization
coordinate operators in the Heisenberg picture, in the Laplace domain, and then, we give
the expressions for the polarization density field operator.

A. Governing equations

First, by multiplying both sides of Eqs. (76) and (77) by \( \alpha_{\nu}/(s^2 + \nu^2) \) and, then, by integrating
with respect to \( \nu \) over \((0, \infty)\) we obtain, with \( m = 1, 2, 3 \ldots \),
\[ \left( \frac{1}{\chi} + \frac{1}{\kappa_m^\parallel} \right) \dot{\hat{P}}_m^\parallel + \sum_{m'} s \left( S_{nm'}^{\parallel\parallel} \hat{P}_m^\parallel + S_{nm'}^{\parallel\perp} \hat{P}_m^\perp \right) = \frac{1}{\chi} \hat{D}_m^\parallel, \quad (83) \]
\[ \left( \frac{1}{\chi} + \frac{s^2}{c^2_0 \kappa_m^\parallel} \right) \dot{\hat{P}}_m^\perp + \sum_{m'} s \left( S_{nm'}^{\perp\parallel} \hat{P}_m^\parallel + \delta S_{nm'}^{\perp\perp} \hat{P}_m^\perp \right) = \frac{1}{\chi} \hat{D}_m^\perp, \quad (84) \]
where \( \hat{P}_m^\parallel \) and \( \hat{P}_m^\perp \) are the Laplace transform of the polarization coordinate operators,
\[ \tilde{\chi}(s) = \frac{1}{\epsilon_0} \int_0^\infty \frac{\alpha_n^2}{(s^2 + \nu^2)} d\nu, \quad (85) \]
\[
\hat{D}^a_m = \tilde{\chi} [C^a_m + \hat{F}^a_m + \sum_{m' \neq m, b=\|,\perp} S^{ab}_{mm'} \hat{p}^b_m(0)] + \hat{F}^a_m(0),
\]
(86)

\[
\hat{P}^a_m(0) = \int_0^\infty \frac{\alpha_{\nu} s^2 + \nu^2}{s^2 + \nu^2} \left[ s \hat{y}^a_{\nu,m}(0) + \dot{\hat{y}}^a_{\nu,m}(0) \right] d\nu,
\]
(87)

and \( \hat{p}^b_m(0) \) are the coordinate operators of the polarization in the Schrödinger picture. The function \( \tilde{\chi}(s) \) is the susceptibility of the dielectric in the Laplace domain, namely, the Laplace transform of the dielectric impulse response \( h_\chi(t) \) (see Appendix B). The term \( \hat{P}^a_m(0) \) takes into account the contribution of the free evolution of the polarization in the absence of interaction with both the components of the electromagnetic field (the solenoidal and irrotational components).

Equations 83 and 84 govern the evolution of the coordinate operators of the polarization in the Laplace domain and have the same algebraic structure of the classical regime [25]. The coefficients \( \{s_{SS}^{ab}_{mm'}\} \) and the susceptibility are c-functions, the unknowns and the driving terms are operators. The second term on the left hand side of Eq. 83 which governs \( \hat{P}^\|_m \), is responsible for the electroquasistatic (plasmon) oscillations of the medium, while the second term on the right hand side of Eq. 84 which governs \( \hat{P}^\perp_m \), is responsible for the magnetoquasistatic oscillations of the medium. The eigenvalues \( \{\kappa^\|_m\} \) only depend on the shape of the object, while the eigenvalues \( \{\kappa^\perp_m\} \) also depend on its size: \( \{\kappa^\|_m\} \) scales as \( 1/a^2 \) as the size of the object \( a \) varies, where \( a \) is the radius of the smallest sphere that encloses the dielectric object.

The coefficients \( \{s_{SS}^{ab}_{mm'}\} \) and \( \{s_{\delta S}^{\perp\perp}_{mm'}\} \) describe the coupling between the longitudinal and the transverse coordinate operators due to the interaction of the polarization with the radiation field. The amplitudes of \( s_{SS}^{ab}_{mm'} \) and \( s_{\delta S}^{\perp\perp}_{mm'} \) tend to zero as \( s \to 0 \) and tend to finite limits as \( |s| \to \infty \). In Appendix D we show that, for fixed values of \( s \), the amplitude of \( s_{SS}^{ab}_{mm'} \) scales as the dimensionless parameter \( \gamma^2 \) as the size of the particle \( a \) varies, where \( \gamma = |s|a/c_0 \) and \( a \) is the radius of the smallest sphere enclosing the dielectric. The amplitude of \( s_{\delta S}^{\perp\perp}_{mm'} \) instead scales as \( \gamma^4 \).

The dimensionless parameter \( \gamma \) allows to discriminate the regime in which the effects of the coupling coefficients are negligible from the one in which their role is important. Indeed,
for $|\chi|\gamma^2 \ll 1$ we can disregard the terms \{sS_{mm}^{ab}, \hat{P}_m^a\} in Eqs.\[\text{83 and 84}\] and obtain

$$
\hat{P}_m^\parallel \approx \frac{\kappa_m^\parallel}{\kappa_m^\parallel + \chi(s)} \hat{D}_m^\parallel, \quad (88)
$$

$$
\hat{P}_m^\perp \approx \frac{c_0^2 \kappa_m^\perp}{c_0^2 \kappa_m^\perp + s^2 \chi(s)} \hat{D}_m^\perp, \quad (89)
$$

The constraint $|\chi|\gamma^2 \ll 1$ is certainly satisfied in the small size limit $a \ll \lambda_c$ where $\lambda_c = \min(\omega_0/|\omega \sqrt{|\chi(\omega)|}|)$ is a ”characteristic wavelength” accounting for the strength of the coupling between the matter and electromagnetic field. For example, for the Drude-Lorentz dielectric function, we obtain $\lambda_c = c_0/\omega_p$ where $\omega_p$ is the plasma frequency of the medium. Indeed, in the small size limit the longitudinal modes diagonalize the contribution to the Hamiltonian of the electroquasistatic (Coulomb) interaction energy, while the transverse modes diagonalize the magnetostatic interaction energy between the transverse modes. As in classical electrodynamics, only a few static longitudinal and transverse modes are needed to properly describe the response of a dielectric object even when its size $a$ is larger than the characteristic wave-length $\lambda_c$.

### B. Transfer function

We now rewrite Eqs.\[\text{83 and 84}\] by using the matrix notation. We have

$$
M \begin{bmatrix} \hat{P}_m^\parallel \\
\hat{P}_m^\perp \end{bmatrix} = \begin{bmatrix} \hat{D}_m^\parallel \\
\hat{D}_m^\perp \end{bmatrix}, \quad (90)
$$

where: $\hat{P}^\parallel = \begin{bmatrix} \hat{P}_1^\parallel, \hat{P}_2^\parallel, \ldots \end{bmatrix}^\top$ is the column vector of the longitudinal coordinate operators of the polarization, $\hat{P}^\perp = \begin{bmatrix} \hat{P}_1^\perp, \hat{P}_2^\perp, \ldots \end{bmatrix}^\top$ is the column vector of the transverse coordinate operators; $M$ is the block matrix

$$
M = \begin{bmatrix} M^{\parallel\parallel} & M^{\parallel\perp} \\
M^{\perp\parallel} & M^{\perp\perp} \end{bmatrix}; \quad (91)
$$

the elements of the blocks $M^{\parallel\parallel}, M^{\parallel\perp}, M^{\perp\parallel}, M^{\perp\perp}$ are given by

$$
M_{mnm'}^{\parallel\parallel}(s) = \begin{cases} 1 + \frac{\chi(s)}{\kappa_m^{\parallel}} + \frac{s\chi(s)S_{mm'}^{\parallel\parallel}(s)}{\kappa_m^{\parallel}} & m = m' \\
s\chi(s)S_{mm'}^{\parallel\perp}(s) & m \neq m' \end{cases} \quad (92)
$$
\[
M_{mm'}^{\perp\perp}(s) = \begin{cases} 
1 + \bar{\chi}(s) \frac{s^2}{c_0^m c_m} + s\bar{\chi}(s) \delta S_{mm}^{\perp\perp}(s) & m = m' \\
\bar{\chi}(s) \delta S_{mm'}^{\perp\perp}(s) & m \neq m'
\end{cases} \tag{93}
\]

\[
M_{mn}^{\perp\perp}(s) = s\bar{\chi}(s) S_{mn}^{\perp\perp}(s), \quad M_{mn}^{\parallel\parallel}(s) = s\bar{\chi}(s) S_{mn}^{\parallel\parallel}(s); \tag{94}
\]

\[\hat{D}^\parallel = \left[ \hat{D}_1^\parallel, \hat{D}_2^\parallel, \ldots \right]^T \text{ and } \hat{D}^\perp = \left[ \hat{D}_1^\perp, \hat{D}_2^\perp, \ldots \right]^T\]
are column vectors describing the driving coordinate operators of the polarization. The elements of the matrix \(M\) are \(c\)-functions, while the elements of the vectors \(P^\parallel, P^\perp, D^\parallel\) and \(D^\perp\) are operators.

In the Heisenberg picture, in the Laplace domain, the coordinate operators of the polarization field operator are given by

\[
\begin{bmatrix} \hat{P}^\parallel(s) \\ \hat{P}^\perp(s) \end{bmatrix} = \begin{bmatrix} H(s) \\ (s^{1/2}) \end{bmatrix} \begin{bmatrix} \hat{D}^\parallel(s) \\ \hat{D}^\perp(s) \end{bmatrix} \tag{95}
\]

where \(H(s) = M^{-1}(s)\) is the transfer function of the classical electrodynamics problem, which has been extensively studied in Ref. [25].

The driving coordinate operators can be expressed as

\[
\begin{bmatrix} \hat{D}^\parallel(s) \\ \hat{D}^\perp(s) \end{bmatrix} = \begin{bmatrix} (U^\parallel, \hat{\mathbf{D}}) \\ (U^\perp, \hat{\mathbf{D}}) \end{bmatrix} \tag{96}
\]

where \(U^\parallel = \left[ U_1^\parallel, U_2^\parallel, \ldots \right]^T, U^\perp = \left[ U_1^\perp, U_2^\perp, \ldots \right]^T\), the driving vector field operator \(\hat{\mathbf{F}}(\mathbf{r}; s)\) has four terms,

\[
\hat{\mathbf{D}} = \mathbf{D}^{inc}(\mathbf{r}; s) + \hat{\mathbf{D}}^{vac}(\mathbf{r}; s) + \hat{\mathbf{D}}^{pol}(\mathbf{r}; s) + \hat{\mathbf{D}}^{noise}(\mathbf{r}; s), \tag{97}
\]

with

\[
\mathbf{D}^{inc}(\mathbf{r}; s) = -\epsilon_0 \bar{\chi}(s) \mathcal{L} \left\{ A^{inc} \right\}, \tag{98a}
\]

\[
\hat{\mathbf{D}}^{vac}(\mathbf{r}; s) = \epsilon_0 \bar{\chi}(s) \hat{\mathbf{E}}^{vac}_s(\mathbf{r}; s), \tag{98b}
\]

\[
\hat{\mathbf{D}}^{pol}(\mathbf{r}; s) = \frac{\chi(s)}{c_0^2} \int_V s \hat{G}^\perp(\mathbf{r} - \mathbf{r}'; s) \hat{\mathbf{P}}(\mathbf{r}'; 0)d^3 \mathbf{r}', \tag{98c}
\]

\[
\hat{\mathbf{D}}^{noise}(\mathbf{r}; s) = \int_0^\infty \frac{\alpha_v}{\nu^{2+\nu}} \left[ s \hat{\mathbf{Y}}_v(\mathbf{r}; 0) + \hat{\mathbf{Y}}_v(\mathbf{r}; 0) \right] d\nu, \tag{98d}
\]

and \(\hat{\mathbf{E}}^{vac}_s\) is the Laplace transform of

\[
\hat{\mathbf{E}}^{vac}_s(\mathbf{r}; t) = \sum_{\mu \in \mathcal{M}} \hat{\mathbf{E}}_{\mu}(t) w_\mu(\mathbf{r}). \tag{99}
\]
\( \hat{E}_{s}^{\text{vac}} \) is the contribution to the solenoidal electric field operator due to the initial state of the radiation field in the absence of the dielectric. The Laplace transform of the polarization density field operator \( \hat{\mathbf{P}}(\mathbf{r}; t) \), which we indicate with \( \hat{\mathbf{P}}(\mathbf{r}; s) \), is expressed in terms of the driving vector field operator \( \hat{\mathbf{D}}(\mathbf{r}; s) \) by relation

\[
\hat{\mathbf{P}}(\mathbf{r}; s) = \mathbf{U}^\dagger(\mathbf{r}) \mathcal{H}(s) \langle \mathbf{U}(\mathbf{r}'), \hat{\mathbf{D}}(\mathbf{r}'; s) \rangle
\]

(100)

where \( \mathbf{U} = [\mathbf{U}^\|, \mathbf{U}^\perp]^\top \).

The expression in the time domain becomes

\[
\hat{\mathbf{P}}(\mathbf{r}; t) = \mathbf{U}^\dagger(\mathbf{r}) h(t) * \langle \mathbf{U}(\mathbf{r}'), \hat{\mathbf{D}}(\mathbf{r}'; t) \rangle
\]

(101)

where \( h(t) = \mathcal{L}^{-1}\{\mathcal{H}(s)\} \) is the impulse response,

\[
\hat{\mathbf{D}} = \hat{\mathbf{D}}^{\text{inc}}(\mathbf{r}; t) + \hat{\mathbf{D}}^{\text{vac}}(\mathbf{r}; t) + \hat{\mathbf{D}}^{\text{pol}}(\mathbf{r}; t) + \hat{\mathbf{D}}^{\text{noise}}(\mathbf{r}; t),
\]

(102)

with

\[
\hat{\mathbf{D}}^{\text{inc}}(\mathbf{r}; t) = -\epsilon_0 h_\mathbf{x}(t) * \hat{\mathbf{A}}^{\text{inc}}(\mathbf{r}; t),
\]

(103a)

\[
\hat{\mathbf{D}}^{\text{vac}}(\mathbf{r}; t) = \epsilon_0 h_\mathbf{x}(t) * \hat{\mathbf{E}}^{\text{vac}}(\mathbf{r}; t),
\]

(103b)

\[
\hat{\mathbf{D}}^{\text{pol}}(\mathbf{r}; t) = \frac{1}{c_0} h_\mathbf{x}(t) * \int_V \frac{\mathbf{g}^\perp}{c_0^2} (\mathbf{r} - \mathbf{r}'; t) \hat{\mathbf{P}}(\mathbf{r}'; 0) d^3\mathbf{r}',
\]

(103c)

and

\[
\hat{\mathbf{D}}^{\text{noise}}(\mathbf{r}; t) = \hat{\mathbf{P}}_0(\mathbf{r}; t)
\]

(103d)

\[
\hat{\mathbf{P}}_0(\mathbf{r}; t) = \int_0^\infty \sqrt{\frac{2\sigma(\omega)}{\pi}} \hat{\mathbf{Y}}^{(0)}_\omega(\mathbf{r}; t) d\omega,
\]

(104)

\[
\hat{\mathbf{Y}}^{(0)}_\omega(\mathbf{r}; t) = \hat{\mathbf{Y}}_\omega(\mathbf{r}; 0) cos(\omega t) + \frac{1}{\omega} \hat{\mathbf{Y}}_\omega(\mathbf{r}; 0) sin(\omega t);
\]

(105)

\( \hat{\mathbf{Y}}_\omega(\mathbf{r}; 0) \) is the operator \( \hat{\mathbf{Y}}_\omega(\mathbf{r}) \), \( \hat{\mathbf{Y}}_\omega(\mathbf{r}; 0) \) is the operator and \( \hat{\mathbf{Y}}_\omega(\mathbf{r}) \) in the Schrödinger picture.

**VII. ELECTRIC FIELD OPERATOR**

In this section, we give the expressions for the electric field operator in terms of the polarization field operator obtained in the previous section.

The electric field operator \( \hat{\mathbf{E}} \) has two contributions: the solenoidal component \( \hat{\mathbf{E}}_s \) in \( V_\infty \), and the irrotational component \( \hat{\mathbf{E}}_c \) in \( V_\infty \),

\[
\hat{\mathbf{E}} = \hat{\mathbf{E}}_s + \hat{\mathbf{E}}_c.
\]

(106)
A. Solenoidal component

In the time domain, the solenoidal component of electric field operator is given by

\[ \hat{E}_s = -\dot{\hat{A}}, \]  

(107)

hence

\[ \hat{E}_s(r; t) = -\sum_{\mu \in \mathcal{M}} \hat{A}_\mu(t) w_\mu(r). \]  

(108)

By using Eqs. 39 and 67 from Eq. 108 we obtain

\[ \hat{E}_s(r; t) = -\frac{1}{\varepsilon_0} \sum_{\mu \in \mathcal{M}} \sum_{m=1,2,...} \frac{R_{\mu m}^a}{a=\|,\perp} \omega_\mu(t) \ast \hat{p}_m^a(t) w_\mu(r) + \hat{E}^{\text{vac}}_s(r; t). \]  

(109)

In the Laplace domain this relation becomes

\[ \hat{E}_s(r; s) = -\frac{1}{\varepsilon_0} \sum_{\mu \in \mathcal{M}} \sum_{m=1,2,...} \frac{R_{\mu m}^a}{a=\|,\perp} W_\mu(s) [s \hat{P}_m^a(s) - \hat{p}_m^a(0)] w_\mu(r) + \hat{E}^{\text{vac}}_s(r; s), \]  

(110)

where \( \hat{E}_s(r; s) \) is the Laplace transform of \( \hat{E}_s \), \( W_\mu(s) \) is the Laplace transform of \( w_\mu(t) \), and \( \{\hat{p}_m^a(0)\} \), with \( a=\|,\perp \), are the coordinate operators of the polarization in the Schrödinger picture. By using C3 we obtain from 110

\[ \hat{E}_s(r; s) = -\mu_0 \int_V s \mathbf{G}^\perp \mathbf{r} \cdot \hat{P}(r'; s) [s \hat{P}(r'; s) - \hat{P}(r'; 0)] d^3 r' + \hat{E}^{\text{vac}}_s(r; s) \]  

(111)

where \( \hat{P}(r'; s) \) is the Laplace transform of the polarization field operator and \( \hat{P}(r; 0) \) is the polarization field operator in the Schrödinger picture. In time domain, relation 111 becomes

\[ \hat{E}_s(s; t) = -\mu_0 \int_V d^3 r' \int_0^\infty d\tau \mathbf{G}^\perp \mathbf{r} \cdot \hat{P}(r'; t - \tau) \hat{P}(r'; \tau) + \hat{E}_s^{\text{vac}}(r; t). \]  

(112)

B. Irrotational component

In the time domain, the Coulomb field operator is given by:

\[ \hat{E}_c(s) = -\frac{1}{4\pi \varepsilon_0} \nabla \int_{\partial V} \frac{\hat{P}(r'; t) \cdot \hat{n}(r')}{|\mathbf{r} - \mathbf{r}'|} d^2 r'. \]  

(113)

Inside the dielectric, by using Eq. A1 we obtain

\[ \hat{E}_c(r; t) = -\frac{1}{\varepsilon_0} \sum_{m=1,2,...} \frac{1}{K_m} \hat{p}_m^\parallel(t) U_m^\parallel(r). \]  

(114)
It is convenient to move the gradient operator under the integral sign in \[113\]. For \( \mathbf{r} \not\in \partial V \) we obtain
\[
\hat{\mathbf{E}}_c(\mathbf{r}; t) = \frac{1}{4\pi \varepsilon_0} \int_{\partial V} \frac{(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3} \hat{\mathbf{P}}(\mathbf{r}'; t) \cdot \hat{n}(\mathbf{r}') d^3 \mathbf{r}'. \tag{115}
\]

When \( \mathbf{r} \in \partial V \) an additional additive term for the normal component is needed, it is given by
\[
\delta \hat{\mathbf{E}}_c(\mathbf{r}; t) \cdot \hat{n}(\mathbf{r}) \bigg|_{\partial V^\pm} = \mp \frac{1}{2\varepsilon_0} \hat{\mathbf{P}}(\mathbf{r}; t) \cdot \hat{n}(\mathbf{r}) \bigg|_{\partial V} \tag{116}
\]
where \( \partial V^\pm \) are the external and integral page of the surface \( \partial V \).

### C. Total field

The total electric field operator \( \hat{\mathbf{E}} \) can be expressed as function of the polarization field operator \( \hat{\mathbf{P}} \) by using the (total) dyadic Green function for the electric field in free space. To show this, we first need to express the Coulomb field operator as a volume integral of the polarization field operator. By using the Gauss theorem, from Eq. \[113\] we obtain
\[
\hat{\mathbf{E}}_c(\mathbf{r}; t) = \frac{1}{4\pi \varepsilon_0} \nabla \int_V \nabla \left( \frac{1}{|\mathbf{r} - \mathbf{r}'|} \right) \cdot \hat{\mathbf{P}}(\mathbf{r}'; t) d^3 \mathbf{r}'. \tag{117}
\]

For \( \mathbf{r} \not\in V \) we can move the gradient operator on the left hand side under the integral sign
\[
\hat{\mathbf{E}}_c(\mathbf{r}; t) = \frac{1}{\varepsilon_0} \int_V \nabla \nabla \left( \frac{1}{4\pi r} \right) \cdot \hat{\mathbf{P}}(\mathbf{r}'; t) d^3 \mathbf{r}'. \tag{118}
\]

For \( \mathbf{r} \in V \), the singularity in \( 1/|\mathbf{r} - \mathbf{r}'| \) yields an additional term (e.g., \[27\]),
\[
\hat{\mathbf{E}}_c(\mathbf{r}; t) = -\frac{1}{\varepsilon_0} \left[ \frac{1}{3} \hat{\mathbf{P}}(\mathbf{r}; t) - \lim_{\delta \to 0} \int_{V_\delta} \nabla \nabla g_0(\mathbf{r} - \mathbf{r}') \hat{\mathbf{P}}(\mathbf{r}'; t) d^3 \mathbf{r}' \right], \tag{119}
\]
where \( V_\delta \) is a sphere of radius \( \delta \) centered at \( \mathbf{r} \), and \( g_0(\mathbf{r}) = 1/4\pi r \); explicitly,
\[
\nabla \nabla \frac{1}{4\pi r} = \frac{1}{4\pi r^3} (3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}} - \hat{\mathbf{T}}). \tag{120}
\]

The expression between the square brackets on the right hand side of Eq. \[119\] is the irrotational component of the polarization density field in \( V_\infty \).

By summing in the Laplace domain the solenoidal term \( \hat{\mathbf{E}}_s \) and the irrotational term \( \hat{\mathbf{E}}_c \) we obtain for \( \mathbf{r} \not\in V \)
\[
\hat{\mathbf{E}}(\mathbf{r}; s) = -\mu_0 \int_V s \hat{G}(\mathbf{r} - \mathbf{r}'; s) [s \hat{\mathbf{P}}(\mathbf{r}'; s) - \hat{\mathbf{P}}(\mathbf{r}'; 0)] d^3 \mathbf{r}' + \hat{\mathbf{E}}_0 \tag{121}
\]
where \( \vec{G} \) is the (total) dyadic Green function for the electric field in the Laplace domain and in free space,

\[
\vec{G} (\mathbf{r}; s) = \frac{1}{4\pi e^{sr/c_0}} \left[ (\hat{I} - \hat{\mathbf{r}} \otimes \hat{\mathbf{r}}) + (\hat{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}) \frac{c_0^2}{sr} \left( \frac{1}{c_0} + \frac{1}{sr} \right) \right]
\]

and

\[
\hat{\mathbf{E}}_0(\mathbf{r}; s) = \hat{\mathbf{E}}_{\text{vac}}(\mathbf{r}; s) + \frac{1}{s} \hat{\mathbf{E}}_c(\mathbf{r}; t = 0).
\]

The term \( \hat{\mathbf{E}}_0(\mathbf{r}; s) \) takes into account the contribution of the initial conditions for the solenoidal electromagnetic field operators and for the polarization field operator (through the Coulomb field). For \( \mathbf{r} \in \mathcal{V} \) expression (121) becomes

\[
\hat{\mathbf{E}}(\mathbf{r}; s) = \frac{1}{3s\varepsilon_0} [s\hat{\mathbf{P}}(\mathbf{r}; s) - \hat{\mathbf{P}}(\mathbf{r}; 0)] - \mu_0 \lim_{\delta \to 0} \int_{\mathcal{V} - \mathcal{V}_0} s\vec{G}(\mathbf{r} - \mathbf{r}'; s) [s\hat{\mathbf{P}}(\mathbf{r}'; s) - \hat{\mathbf{P}}(\mathbf{r}'; 0)] d^3\mathbf{r}' + \hat{\mathbf{E}}_0.
\]

**VIII. DISCUSSION**

We have introduced a novel operative approach for modeling in the Heisenberg picture the quantum electrodynamics of absorptive dielectrics of finite size in unbounded space. It is based on a Hopfield-type model in which the polarization is described through a matter field consisting of a continuum of harmonic oscillators linearly coupled to the electromagnetic field.

We use the Hamiltonian in the Coulomb gauge. We represent the polarization field of the dielectric in terms of the static longitudinal and transverse modes of the dielectric object, which are a basis for the solenoidal vector fields defined on the region occupied by the dielectric and only depend on the geometry of the dielectric object. We express the irrotational component of the electric field in terms of the coordinates of the polarization field. We represent the solenoidal component of the electromagnetic field in terms of the transverse plane wave modes of the free space.

We determine the equation of motion for the longitudinal and transverse coordinate operators of the polarization field, which are coupled. The coupling is due to the interaction of the polarization with the solenoidal component of the electromagnetic field, which we describe through the full-wave transverse dyadic Green function for the electric field in free space. As expected, the equations of motion have the same structure of the equations obtained in the classical regime [25].
The longitudinal modes diagonalize the Coulomb interaction energy, and the transverse modes diagonalize the magnetic interaction energy. We leave the other interaction energy terms not diagonalized. Since in the limit of small size dielectrics the longitudinal and transverse modes are the natural modes of polarization, for objects with sizes of the same order \( \lambda_c = \min \omega / \omega \sqrt{|\chi(\omega)|} \) only few modes are needed, and a numerical diagonalization is straightforward, as indicated by the classical electrodynamics problem \[25\].

In the Laplace domain, the initial state of the radiation and matter come into play on the right hand side of the system of Eqs. \[83\]-\[84\] through several terms: the noise operators of the matter \( \{ \hat{N}_m^\parallel \}, \{ \hat{N}_m^\perp \} \), the initial state of the solenoidal component of the electromagnetic field \( \{ \hat{D}_m^\parallel \}, \{ \hat{D}_m^\perp \} \), and the initial state of the polarization, \( \{ \sum_{m'=1,2,\ldots} S_{mm'}^b \hat{P}_{mb}(0) \}, \{ \sum_{m'=1,2,\ldots} S_{mm'}^b \hat{P}_{mb}(0) \} \).

The dynamics of the polarization field operator is modeled as a linear system by considering, as output variables, the coordinate of the polarization and, as input variables, the coordinates of the driving vector field operator, which also take into account an incident classical electric field. The evolution of the system is governed by the same transfer function \( H(s) \) of the classical electrodynamics, which only depends on the classical susceptibility of the dielectric and its geometry. Eventually, we also give the expression of the electric field operator in terms of the polarization field operator through the dyadic Green function for the electric field in vacuum.

We have proposed a general method for the quantization of macroscopic electrodynamics in absorptive dielectrics of finite size in unbounded space, which leads to operative schemes that can be implemented by using the standard tool of computational electromagnetics.
Appendix A: Longitudinal and transverse static modes of the dielectric body

Following Ref. [25], the static longitudinal (electrostatic) modes of the dielectric domain are solutions of the eigenvalue problem [28], [29],

\[
\nabla \oint_{\partial V} \frac{U_m^\parallel (r') \cdot \hat{n} (r')} {4\pi |r - r'|} d^2 r' = \frac{1}{\kappa_m^\parallel} U_m^\parallel (r) \quad \text{in } V, \tag{A1}
\]

where \(\kappa_m^\parallel\) is the eigenvalue associated to the eigenmode \(U_m^\parallel (r)\), and \(m = 1, 2, 3 \ldots\); \(\kappa_m^\parallel\) is a dimensionless quantity. The eigenvalues are discrete, real, positive, and equal or greater than two \((\kappa_m^\parallel \geq 2)\). The eigenmodes and the eigenvalues only depend on the shape of the body \(V\), they do not depend on its size. The solution of the problem \(A1\) can be obtained using the method outlined in Refs. [30, 31].

Following Ref. [25], we exploit the static transverse (magnetostatic) modes of the particle to represent the transverse components of the matter and bath vector fields. They are solutions of the eigenvalue problem [32, 33]

\[
\int_V \frac{U_m^\perp (r') \cdot \hat{n} (r')} {4\pi |r - r'|} d^3 r' = \frac{1}{\kappa_m^\perp} U_m^\perp (r) \quad \text{in } V, \tag{A2}
\]

with

\[
U_m^\perp (r) \cdot \hat{n} (r) = 0 \quad \text{on } \partial V, \tag{A3}
\]

where \(\kappa_m^\perp\) is the eigenvalue associated to the eigenmode \(U_m^\perp\), and \(m = 1, 2, 3 \ldots\); \(1/\kappa_m^\perp\) is homogeneous with the square of length. Equation \(A2\) with the constraint \(A3\) holds in weak form in the functional space equipped with the inner product \(\langle F, G \rangle\), and constituted by the vector fields that are solenoidal in \(V\) and have zero normal component to \(\partial V\). The eigenvalues are discrete, real and positive. The problem \(A2\) can be solved by using standard tools of computational electromagnetism as outlined in Ref. [33].

Appendix B: Dielectric susceptibility

The polarization density field is expressed in terms of the matter fields \(\{Y_\nu\}\) through Eq. [5] where the coupling coefficient \(\alpha_\nu\) is given by [7] In this Appendix we show that the polarization density field verifies Eq. [1]

The time evolution of the matter field is governed by the equation

\[
\ddot{Y}_\nu + \nu^2 Y_\nu = \alpha_\nu (E + E_{inc}) \quad \text{in } V \tag{B1}
\]
where \( 0 < \nu < \infty \). In the Laplace domain it becomes

\[
(s^2 + \nu^2) \hat{Y} = \alpha_\nu (\hat{E} + \hat{E}_{\text{inc}}) + [sY_\nu(\mathbf{r}; 0) + \dot{Y}_\nu(\mathbf{r}; 0)]
\]

(B2)

where \( \hat{Y}(\mathbf{r}; s) \) is the Laplace transform of \( Y_\nu(\mathbf{r}; t) \), \( \hat{E}(\mathbf{r}; s) \) is the Laplace transform of \( E(\mathbf{r}; t) \), \( \hat{E}_{\text{inc}}(\mathbf{r}; s) \) is the Laplace transform of \( E_{\text{inc}}(\mathbf{r}; t) \); \( Y_\nu(\mathbf{r}; 0) \) and \( \dot{Y}_\nu(\mathbf{r}; 0) \) are the initial conditions for \( Y_\nu \) and \( \dot{Y}_\nu \), respectively. Therefore, in the Laplace domain the polarization field density is given by

\[
\hat{P} = \varepsilon_0 \tilde{\chi}(s)(\hat{E} + \hat{E}_{\text{inc}}) + \hat{P}_0
\]

(B3)

where

\[
\tilde{\chi}(s) = \frac{1}{\varepsilon_0} \int_0^\infty \frac{\alpha_\nu^2}{s^2 + \nu^2} d\nu \quad \text{(B4)}
\]

and

\[
\hat{P}_0 = \int_0^\infty \frac{\alpha_\nu}{s^2 + \nu^2} [sY_\nu(\mathbf{r}; 0) + \dot{Y}_\nu(\mathbf{r}; 0)] d\nu.
\]

(B5)

The region of convergence of the Laplace transform contains the imaginary axis, therefore we evaluate \( \tilde{\chi}(s) \) for \( s = i\omega + \epsilon \) where \( \epsilon \downarrow 0 \). By using the relation (e.g., \([34]\))

\[
\frac{1}{x - i\epsilon} = i\pi \delta(x) + \mathcal{P} \frac{1}{x},
\]

(B6)

where \( \mathcal{P} \) denotes the Cauchy principal value, we obtain for \( \chi(\omega) \equiv \tilde{\chi}(s = i\omega + \epsilon) \) the following expression

\[
\chi(\omega) = \mathcal{P} \int_0^\infty \frac{\alpha_\nu^2}{\nu^2 - \omega^2} d\nu - i \frac{\pi}{2} \frac{\alpha_\nu}{\omega},
\]

(B7)

hence

\[
\alpha_\omega = \sqrt{-2 \pi \omega \chi_i(\omega)}
\]

(B8)

where \( \chi_i \) is the imaginary part of \( \chi(\omega) \).

In the time domain we have in \( V \)

\[
P(\mathbf{r}; t) = \varepsilon_0 \int_0^\infty h_\chi(t - \tau)[E(\mathbf{r}; \tau) + E_{\text{inc}}(\mathbf{r}; \tau)] d\tau + P_0(\mathbf{r}; t)
\]

(B9)

where \( h_\chi(t) \) is the inverse Fourier transform of the susceptibility of the dielectric \( \chi(\omega) \),

\[
h_\chi(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \chi(\omega)e^{i\omega t} d\omega,
\]

(B10)

and \( P_0(\mathbf{r}; t) \) is the free evolution term taking into account the initial state of the matter field, namely,

\[
P_0(\mathbf{r}; t) = \int_0^\infty \sqrt{\frac{2\sigma(\omega)}{\pi}} Y_\omega^{(0)}(\mathbf{r}; t) d\omega
\]

(B11)
where
\[
\mathbf{Y}^{(0)}_\omega (\mathbf{r}; t) = \mathbf{Y}_\omega (\mathbf{r}; 0) \cos(\omega t) + \frac{1}{\omega} \dot{\mathbf{Y}}_\omega (\mathbf{r}; 0) \sin(\omega t),
\]  
(B12)

\(\mathbf{Y}_\omega (\mathbf{r}; 0)\) is the initial value of \(\mathbf{Y}_\omega (\mathbf{r}; t)\) and \(\dot{\mathbf{Y}}_\omega (\mathbf{r}; 0)\) is the initial value of \(\dot{\mathbf{Y}}_\omega (\mathbf{r}; t)\).

**Appendix C: Expression of the kernel \(s_{mnm'}^{ab}(t)\) and transverse dyadic Green function in free space**

In this Appendix we evaluate the kernel \(s_{mnm'}^{ab}(t)\), whose expression is given by [72] and introduce the transverse dyadic Green function for the electric field in free space.

The Laplace transform of the kernel \(s_{mnm'}^{ab}(t)\) is
\[
S_{mnm'}^{ab}(s) = \sum_{\mu \in \mathcal{M}} \frac{s}{s^2 + c_0^2 k^2} \langle U^a_m, w_\mu \rangle \langle w^*_\mu, U^b_{m'} \rangle.
\]  
(C1)

We rewrite it as
\[
S_{mnm'}^{ab}(s) = \frac{s}{c_0^2} \int d^3 \mathbf{r} \int d^3 \mathbf{r}' U^a_m (\mathbf{r}) \mathbf{G}^\perp (\mathbf{r} - \mathbf{r}'; s) U^b_{m'} (\mathbf{r}'),
\]  
(C2)

where \(\mathbf{G}^\perp (\mathbf{r} - \mathbf{r}'; s)\) is the dyad
\[
\mathbf{G}^\perp (\mathbf{r} - \mathbf{r}'; s) = \sum_{\mu \in \mathcal{M}} \frac{1}{k^2 + s^2 / c_0^2} w_\mu (\mathbf{r}) \otimes w^*_\mu (\mathbf{r}').
\]  
(C3)

By using the expression of \(w_q (\mathbf{r})\) (see [12]) we obtain
\[
\mathbf{G}^\perp (\mathbf{r}; s) = \frac{1}{(2\pi)^3} \int \mathbf{G}^\perp (\mathbf{k}; s) e^{i \mathbf{k} \cdot \mathbf{r}} d^3 \mathbf{k}
\]  
(C4)

where
\[
\mathbf{G}^\perp (\mathbf{k}; s) = \frac{1}{k^2 + s^2 / c_0^2} \left( \mathbf{I} - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}} \right)
\]  
(C5)

is the transverse dyadic Green function for the electric field in the wavenumber domain, in free space. By evaluating the Fourier integral [C3] we obtain (e.g., [35])
\[
\mathbf{G}^\perp (\mathbf{r}; s) = \frac{\mathbf{I} - \hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi r} e^{-sr/c_0} + c_0 \frac{\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi r^2} \left[ \frac{1}{s} e^{-sr/c_0} - \frac{c_0}{s^2 r} (1 - e^{-sr/c_0}) \right].
\]  
(C6)

This is the expression in the Laplace domain of the transverse dyadic Green function for the electric field in free space.

It is useful to express \(\mathbf{G}^\perp (\mathbf{r}; s)\) as
\[
\mathbf{G}^\perp (\mathbf{r}; s) = \mathbf{G}^\perp_0 (\mathbf{r}) + \mathbf{G}^\perp_d (\mathbf{r}; s),
\]  
(C7)
where
\[ \vec{g}^{\perp}_0 (r) = \vec{G}^{\perp}(r; s = 0) = (\vec{I} + \hat{r} \otimes \hat{r}) \frac{1}{8\pi r} \] (C8)
and
\[ \vec{G}^{\perp}_d (r; s) = \vec{G}^{\perp}(r; s) - \vec{g}^{\perp}_0 (r). \] (C9)

The term \( \vec{g}^{\perp}_0 \) is the static transverse dyadic Green function for the free space, which diverges as \( 1/r \) for \( r \to 0 \). The dynamic part \( \vec{G}^{\perp}_d \), which tends to zero as \( s \) for \( s \to 0 \), is a regular function of \( r \).

In the time domain, the transverse dyadic Green function in free space is given by
\[ \vec{g}^{\perp} (r; t) = \frac{(\vec{I} - \hat{r} \otimes \hat{r})}{4\pi r} \delta(t - r/c_0) + c_0 \frac{(\vec{I} - 3\hat{r} \otimes \hat{r})}{4\pi r^2} \left[ \theta(t - r/c_0) + \frac{c_0}{r} \theta(t - r/c_0)(t - r/c_0) - \frac{c_0}{r} \theta(t) \right], \] (C10)
and its partial derivative with respect to the time is given by
\[ \dot{\vec{g}}^{\perp} (r; t) = \frac{(\vec{I} - \hat{r} \otimes \hat{r})}{4\pi r} \delta^{(2)}(t - r/c_0) + c_0 \frac{(\vec{I} - 3\hat{r} \otimes \hat{r})}{4\pi r^2} \left[ \delta(t - r/c_0) + \frac{c_0}{r} \theta(t - r/c_0) - \frac{c_0}{r} \theta(t) \right]. \] (C11)

In conclusion, the expression of \( s_{ab}^{mm'}(t) \) is
\[ s_{ab}^{mm'}(t) = \frac{1}{c_0^2} \int_V d^3 r \int_V d^3 r' U_m^a(r) \vec{g}^{\perp}(r - r'; t) U_{m'}^b(r'). \] (C12)

**Appendix D: Small size limit**

We now analyze the behavior of the coefficients \( s_{mm'}^{ab}(s) \) in the small size limit, i.e., \( \gamma = (a|s|/c_0) << 1 \). We have:
\[ s_{mm'}^\parallel\parallel (s) = \gamma^2 \Sigma_{mm'}^\parallel + \mathcal{O}(\gamma^4), \] (D1)
\[ s_{mm'}^\parallel\perp (s) = \gamma^2 \Sigma_{mm'}^\perp + \mathcal{O}(\gamma^5), \] (D2)
\[ s_{mm'}^\perp\perp (s) = \frac{\gamma^2}{k_m^2} \delta_{mm'} + \gamma^4 \Sigma_{mm'}^{\perp\perp} + \mathcal{O}(\gamma^4), \] (D3)
where

\[ \Sigma_{\parallel \parallel}^{\parallel \parallel} = \frac{1}{4\pi a^2} \int_V d^3r \int_V d^3r' \frac{U^\parallel_{m}(r) \cdot U^\parallel_{m'}(r')}{|r-r'|}, \quad \text{(D5)} \]

\[ \Sigma_{\parallel \perp}^{\parallel \perp} = \frac{1}{4\pi a^2} \int_V d^3r \int_V d^3r' \frac{U^\parallel_{m}(r) \cdot U^\perp_{m'}(r')}{|r-r'|}, \quad \text{(D6)} \]

The quantities \( \Sigma_{\parallel \parallel}^{\parallel \parallel}, \Sigma_{\parallel \perp}^{\parallel \perp}, \Sigma_{\perp \perp}^{\perp \perp} \), and \( 1/(a^2 \kappa_p^\perp) \) do not depend on the size of the dielectric object \( a \) and on the complex variable \( s \), they only depend on the particle shape. Equations [D1],[D3] have been obtained by using the identities

\[
\int_V d^3r \int_V d^3r' \frac{(r-r') \otimes (r-r')}{|r-r'|^3} U^b_{p'}(r') = \\
\quad \oint_S d^2r \oint_S d^2r' (U^a_p(r) \cdot \hat{n}) \frac{|r-r'|}{|r-r'|} (U^b_{p'}(r') \cdot \hat{n}') + \\
\quad \int_V d^3r \int_V d^3r' \frac{U^a_p(r) \cdot U^b_{p'}(r')}{|r-r'|}. \quad \text{(D7)}
\]

\[
\int_V d^3r \int_V d^3r' \frac{(r-r') \otimes (r-r')}{|r-r'|^3} U^b_{p'}(r') = \\
\quad -\oint_S d^2r \oint_S d^2r' (U^a_p(r) \cdot \hat{n}) |r-r'| \frac{|r-r'|^2}{|r-r'|} (U^b_{p'}(r') \cdot \hat{n}') + \\
\quad -\frac{1}{2} \int_V d^3r \int_V d^3r' |r-r'| U^a_p(r) \cdot U^b_{p'}(r'). \quad \text{(D8)}
\]

and the asymptotic expression of the transverse dyadic Green function

\[
\mathbf{G}^\perp(r; s) = \mathbf{g}_0^\perp(r) - \frac{2}{12\pi r} \left( \frac{s r}{c_0} \right) + \frac{\left( \mathbf{T}^\mathbf{r} - \hat{r} \otimes \hat{r} \right)}{32\pi r} \left( \frac{s r}{c_0} \right)^2 + O \left( \frac{s r}{c_0} \right)^3 \quad \text{(D9)}
\]

where \( \mathbf{g}_0^\perp(r) \) is given by [CS]

[1] M. S. Tame, K. R. McEnery, S. K. Ozdemir, J. Lee, S. A. Maier, and M. S. Kim, “Quantum plasmonics,” Nature Physics, vol. 9, pp. 329–340, June 2013.

[2] F. Flamini, N. Spagnolo, and F. Sciarrino, “Photonic quantum information processing: a review,” vol. 82, p. 016001, Nov. 2018. Publisher: IOP Publishing.
[3] J. M. Jauch and K. M. Watson, “Phenomenological Quantum-Electrodynamics,” *Physical Review*, vol. 74, pp. 950–957, Oct. 1948. Publisher: American Physical Society.

[4] R. J. Glauber and M. Lewenstein, “Quantum optics of dielectric media,” *Physical Review A*, vol. 43, pp. 467–491, Jan. 1991. Publisher: American Physical Society.

[5] J. J. Hopfield, “Theory of the Contribution of Excitons to the Complex Dielectric Constant of Crystals,” *Physical Review*, vol. 112, pp. 1555–1567, Dec. 1958. Publisher: American Physical Society.

[6] R. Matloob, R. Loudon, S. M. Barnett, and J. Jeffers, “Electromagnetic field quantization in absorbing dielectrics,” *Physical Review A*, vol. 52, pp. 4823–4838, Dec. 1995. Publisher: American Physical Society.

[7] T. Gruner and D.-G. Welsch, “Green-function approach to the radiation-field quantization for homogeneous and inhomogeneous Kramers-Kronig dielectrics,” *Physical Review A*, vol. 53, pp. 1818–1829, Mar. 1996. Publisher: American Physical Society.

[8] S. Scheel and S. Buhmann, “Macroscopic quantum electrodynamics - Concepts and applications,” *Acta Physica Slovaca. Reviews and Tutorials*, vol. 58, Oct. 2008.

[9] U. Fano, “Atomic Theory of Electromagnetic Interactions in Dense Materials,” *Physical Review*, vol. 103, pp. 1202–1218, Sept. 1956. Publisher: American Physical Society.

[10] B. Huttner and S. M. Barnett, “Quantization of the electromagnetic field in dielectrics,” *Physical Review A*, vol. 46, pp. 4306–4322, Oct. 1992. Publisher: American Physical Society.

[11] L. G. Suttorp and M. Wubs, “Field quantization in inhomogeneous absorptive dielectrics,” *Physical Review A*, vol. 70, p. 013816, July 2004. Publisher: American Physical Society.

[12] N. A. R. Bhat and J. E. Sipe, “Hamiltonian treatment of the electromagnetic field in dispersive and absorptive structured media,” *Physical Review A*, vol. 73, p. 063808, June 2006. Publisher: American Physical Society.

[13] T. G. Philbin, “Canonical quantization of macroscopic electromagnetism,” *New Journal of Physics*, vol. 12, p. 123008, Dec. 2010.

[14] C. R. Gubbin, S. A. Maier, and S. De Liberato, “Real-space Hopfield diagonalization of inhomogeneous dispersive media,” *Physical Review B*, vol. 94, p. 205301, Nov. 2016. Publisher: American Physical Society.

[15] V. Dorier, J. Lampart, S. Guérin, and H. R. Jauslin, “Canonical quantization for quantum plasmonics with finite nanostructures,” *Physical Review A*, vol. 100, p. 042111, Oct. 2019.
C. Forestiere, G. Miano, M. Pascale, and R. Tricarico, “Quantum theory of radiative decay rate and frequency shift of surface plasmon modes,” *Physical Review A*, vol. 102, p. 043704, Oct. 2020. Publisher: American Physical Society.

D.-Y. Na, J. Zhu, and W. C. Chew, “Diagonalization of the Hamiltonian for finite-sized dispersive media: Canonical quantization with numerical mode decomposition,” *Physical Review A*, vol. 103, p. 063707, June 2021. Publisher: American Physical Society.

T. Gruner and D.-G. Welsch, “Correlation of radiation-field ground-state fluctuations in a dispersive and lossy dielectric,” *Physical Review A*, vol. 51, pp. 3246–3256, Apr. 1995. Publisher: American Physical Society.

W. Vogel and D.-G. Welsch, *Quantum Optics*. John Wiley and Sons, 3rd ed., 2006.

H. T. Dung, L. Knöll, and D.-G. Welsch, “Three-dimensional quantization of the electromagnetic field in dispersive and absorbing inhomogeneous dielectrics,” *Physical Review A*, vol. 57, pp. 3931–3942, May 1998. Publisher: American Physical Society.

S. Franke, S. Hughes, M. K. Dezfouli, P. T. Kristensen, K. Busch, A. Knorr, and M. Richter, “Quantization of Quasinormal Modes for Open Cavities and Plasmonic Cavity Quantum Electrodynamics,” *Physical Review Letters*, vol. 122, p. 213901, May 2019. Publisher: American Physical Society.

G. W. Hanson, F. Lindel, S. Y. Buhmann, and S. Y. Buhmann, “Langevin noise approach for lossy media and the lossless limit,” *JOSA B*, vol. 38, pp. 758–768, Mar. 2021. Publisher: Optical Society of America.

A. Drezet, “Equivalence between the Hamiltonian and Langevin noise descriptions of plasmon polaritons in a dispersive and lossy inhomogeneous medium,” *Physical Review A*, vol. 96, p. 033849, Sept. 2017. Publisher: American Physical Society.

V. Dorier, S. Guérin, and H.-R. Jauslin, “Critical review of quantum plasmonic models for finite-size media,” *Nanophotonics*, vol. 9, pp. 3899–3907, Sept. 2020. Publisher: De Gruyter Section: Nanophotonics.

C. Forestiere and G. Miano, “Time-domain formulation of electromagnetic scattering based on a polarization-mode expansion and the principle of least action,” *Physical Review A*, vol. 104, p. 013512, July 2021. Publisher: American Physical Society.
[26] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, “Photons and Atoms-Introduction to Quantum Electrodynamics,” *Photons and Atoms-Introduction to Quantum Electrodynamics, by Claude Cohen-Tannoudji, Jacques Dupont-Roc, Gilbert Grynberg*, pp. 486. ISBN 0-471-18433-0. Wiley-VCH, February 1997., p. 486, 1997.

[27] J. G. Van Bladel, *Singular Electromagnetic Fields and Sources*. Wiley, Jan. 1996. Google-Books-ID: QB_iwAEACAAJ.

[28] D. R. Fredkin and I. D. Mayergoyz, “Resonant Behavior of Dielectric Objects (Electrostatic Resonances),” *Physical Review Letters*, vol. 91, p. 253902, Dec. 2003. Publisher: American Physical Society.

[29] C. Forestiere, G. Miano, and G. Rubinacci, “Resonance frequency and radiative Q-factor of plasmonic and dielectric modes of small objects,” *Phys. Rev. Research*, vol. 2, p. 043176, Nov. 2020. Publisher: American Physical Society.

[30] I. D. Mayergoyz, D. R. Fredkin, and Z. Zhang, “Electrostatic (plasmon) resonances in nanoparticles,” *Phys. Rev. B*, vol. 72, p. 155412, Oct. 2005. Publisher: American Physical Society.

[31] I. D. Mayergoyz, *Plasmon Resonances In Nanoparticles*. World Scientific, Dec. 2012. Google-Books-ID: Ny27CgAAQBAJ.

[32] C. Forestiere, G. Gravina, G. Miano, M. Pascale, and R. Tricarico, “Electromagnetic modes and resonances of two-dimensional bodies,” *Physical Review B*, vol. 99, p. 155423, Apr. 2019. Publisher: American Physical Society.

[33] C. Forestiere, G. Miano, G. Rubinacci, M. Pascale, A. Tamburrino, R. Tricarico, and S. Ventre, “Magnetoquasistatic resonances of small dielectric objects,” *Phys. Rev. Research*, vol. 2, p. 013158, Feb. 2020. Publisher: American Physical Society.

[34] W. Heitler, *The Quantum Theory of Radiation*. Dover Publications, 1984. Google-Books-ID: 8jVRAAAAAMAAJ.

[35] H. F. Arnoldus, “Transverse and longitudinal components of the optical self-, near-, middle- and far-field,” *Journal of Modern Optics*, vol. 50, pp. 755–770, Apr. 2003.