Integral Equation Formulation of Macroscopic Quantum Electrodynamics in Dispersive Dielectrics

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We present an integral equation approach for analyzing the macroscopic quantum electrodynamics, in the Heisenberg picture, in finite-size dispersive dielectric objects, in the framework of Hopfield-type models. The approach proposed in this letter, unlike the existing ones, does not require the diagonalization of the Hamiltonian for evaluating the time evolution of the observables. It is particularly attractive because it enables the direct application of consolidated repertory in computational classical electrodynamics to carry out quantum electrodynamics computation in open, dispersive and absorbing environment.

Introduction and summary of the main results.– Motivated by the use of nanophotonic and nanoplasmonic devices in quantum optics and quantum technology applications (e.g., [1], [2]) there is a large interest for macroscopic quantum electrodynamics (e.g., [3]) in open, dispersive, and absorbing environments. Glauber and Lewenstein [4] proposed a model for the quantization of the macroscopic electromagnetic field in the presence of nondispersive and inhomogeneous dielectrics. To take into account dispersion and dissipation two types of approaches have been developed: a) Langevin-noise type approaches based on the introduction of phenomenological noise currents [5–7], which are widely applied in many contexts (e.g., [3] [8] [9]); b) Hopfield type approaches in which the matter is described as an harmonic oscillating bosonic field linearly coupled to the electromagnetic field, which are variations of a model proposed by Hopfield [10]. Huttner and Barnett [11] considered a homogeneous, dispersive, and dissipative medium. The extension to inhomogeneous media was treated in [12–14]. These two approaches are equivalent if in the Langevin noise type approaches the quantized photonic degrees of freedom associated with the fluctuating radiation field are added to the degrees of freedom of the material oscillators (e.g., [15] [16]). In both approaches, the problem is generally solved by diagonalizing the Hamiltonian operator of the entire system, matter, and electromagnetic field. The diagonalization requires either the Green’s function of the dielectric object, or the eigenmodes of the dielectric object, or the solution of Lippmann-Schwinger type equations. Recently, an operative approach to quantum electrodynamics in dispersive dielectric objects based on a polarization-mode expansion has been proposed. The method leads to a system of coupled equations for the coordinates of the mode expansion of the polarization density field operator, which can be efficiently solved for small objects [17].

In this letter we introduce an integral equation formulation of the macroscopic quantum electrodynamics, in the Heisenberg picture, in finite-size dispersive dielectric objects (see Fig. 1), which does not require the diagonalization of the Hamiltonian to evaluate the time evolution of the observables. A Hopfield type model [13], [14] is applied to describe the matter. We consider a linear, isotropic, dispersive, and homogeneous dielectric. \(\chi(\omega)\) denotes the macroscopic susceptibility of the dielectric in the frequency domain, \(V\) the region of the space occupied by the object, \(\partial V\) its boundary, \(n\) the unit vector normal to \(\partial V\) that points outward, and \(V_\infty\) the overall unbounded space. We found that the polarization density field operator in the Heisenberg picture, \(\hat{P}(\mathbf{r}; t)\), is solution of the linear integral-differential equation

\[
h(\omega) \ast \hat{P}(\mathbf{r}; t) - \mathcal{L}\{\hat{P}\}(\mathbf{r}; t) = \hat{F}(\mathbf{r}; t) \quad in \ V \ \text{for} \ 0 \leq t, \quad (1)
\]

where \(\ast\) denotes the time convolution product, \(h(\omega)\) is the inverse Fourier transform of \(\eta(\omega) = 1/\chi(\omega)\), the operator \(\mathcal{L}\) is defined as

\[
\mathcal{L}\{\hat{G}\}(\mathbf{r}; t) \rightarrow \int \frac{d^3 \mathbf{q}}{(2\pi)^3} \hat{G}(\mathbf{q}; t) \hat{A}(\mathbf{q}):(\mathbf{n} \times \hat{A}(\mathbf{q})(\mathbf{n} \times \mathbf{E}(\mathbf{r} - \mathbf{q}) + \mathbf{n} \times \mathbf{E}(\mathbf{r} + \mathbf{q})))
\]

\[
\hat{A}(\mathbf{q})(\mathbf{n} \times \mathbf{E}(\mathbf{r} - \mathbf{q}) + \mathbf{n} \times \mathbf{E}(\mathbf{r} + \mathbf{q})) = \frac{1}{2\pi^2} \int \frac{d^3 \mathbf{q}'}{(2\pi)^3} \hat{A}(\mathbf{q}') \hat{V}(\mathbf{r} - \mathbf{q}' - \mathbf{q})
\]

\[
\hat{V}(\mathbf{r}) = \frac{1}{\chi(\omega)} \mathbf{n} \cdot \nabla \mathbf{E}(\mathbf{r})
\]

\[
\hat{G}(\mathbf{r}; t) := \mathcal{F}^{-1}\{\hat{G}(\omega)\}(\mathbf{r}; t)
\]

\[
\hat{G}(\omega) := \hat{P}(\mathbf{q}; \omega) \mathbf{E}(\mathbf{q})
\]

\[
\mathcal{F}\{\hat{G}(\omega)\}(\mathbf{r}; t) := \int \frac{d^3 \mathbf{q}}{(2\pi)^3} \hat{G}(\mathbf{q}; \omega) \mathbf{E}(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}}
\]

FIG. 1. Sketch of the system under consideration
\[ \mathcal{L}(\hat{\mathbf{P}})(\mathbf{r}; t) = -\mu_0 \frac{\partial}{\partial t} \int_V \frac{\hat{\mathbf{P}}(\mathbf{r}'; t')}{4\pi |\mathbf{r} - \mathbf{r}'|} u(t') d^3r' - \frac{1}{\varepsilon_0} \int_{\partial V} \frac{\hat{\mathbf{P}}(\mathbf{r}'; t') \cdot \mathbf{n}(\mathbf{r}')}{4\pi |\mathbf{r} - \mathbf{r}'|} u(t') d^2r', \] (2)

We use the Coulomb gauge, \( \nabla \cdot \mathbf{A} = 0 \). Therefore, the scalar potential is given by

\[ \phi(\mathbf{r}; t) = \frac{1}{\varepsilon_0} \int_{\partial V} \frac{\mathbf{P}(\mathbf{r}'; t') \cdot \mathbf{n}(\mathbf{r}')}{4\pi |\mathbf{r} - \mathbf{r}'|} q^2 r' \] (9)

where \( \varepsilon_0 \) is the vacuum permittivity. The electric field has two components: the term \(-\mathbf{A}\), which is solenoidal everywhere, namely the transverse component, and the term \(-\nabla \phi\), which is irrotational everywhere, namely the longitudinal component (Appendix A).

In the Coulomb gauge, the degrees of freedom of the entire system are the continuum of matter fields \( \{\mathbf{Y}_\nu\} \) and the vector potential \( \mathbf{A} \). The Lagrangian is the sum of four terms: the matter term \( \mathcal{L}_{\text{mat}} = \mathcal{L}_{\text{mat}}(\mathbf{Y}_\nu, \mathbf{Y}_\nu^\ast) \), the Coulomb term \( \mathcal{L}_{\text{Coul}} = \mathcal{L}_{\text{Coul}}(\mathbf{P}) \), the radiation term \( \mathcal{L}_{\text{rad}} = \mathcal{L}_{\text{rad}}(\mathbf{A}) \), and the matter–radiation interaction term \( \mathcal{L}_{\text{int}} = \mathcal{L}_{\text{int}}(\hat{\mathbf{P}}, \mathbf{A}) \) (e.g., [18, 11, 14, 19]),

\[ \mathcal{L} = \mathcal{L}_{\text{mat}} + \mathcal{L}_{\text{Coul}} + \mathcal{L}_{\text{rad}} + \mathcal{L}_{\text{int}}, \] (10)

where

\[ \mathcal{L}_{\text{mat}} = \int_V d^3r \int_0^\infty d\nu \left( \frac{1}{2} \frac{\mathbf{Y}_\nu^2 - \mathbf{Y}_\nu^2}{2 \nu^2} \right), \] (11a)

\[ \mathcal{L}_{\text{Coul}} = -\int_{\partial V} d^2r \int_{\partial V} d^2r' \frac{P_n(\mathbf{r}; t) P_n(\mathbf{r}'; t)}{8\pi \varepsilon_0 |\mathbf{r} - \mathbf{r}'|}, \] (11b)

\[ \mathcal{L}_{\text{rad}} = \int_{V_{\infty}} d^3r \left[ \frac{\varepsilon_0}{2} \mathbf{A}^2 - \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 \right], \] (11c)

\[ \mathcal{L}_{\text{int}} = \int_V d^3r \hat{\mathbf{P}} \cdot \mathbf{A}, \] (11d)

and \( P_n = \mathbf{P} \cdot \mathbf{n} \) on \( \partial V \).

We now introduce the Hamiltonian in the Coulomb gauge. The momentum \( \mathbf{Q}_\nu(\mathbf{r}; t) \) conjugate to the matter field \( \mathbf{Y}_\nu \) is given by

\[ \mathbf{Q}_\nu = \delta \mathcal{L}/\delta \mathbf{Y}_\nu^\ast = \mathbf{Y}_\nu^\ast + \alpha_\nu \mathbf{A} \text{ in } V, \] (12)

where \( \delta \) denotes the functional derivative in the variable \( \nu \). The momentum \( \mathbf{\Pi}(\mathbf{r}; t) \) conjugate to the vector potential \( \mathbf{A} \) is given by

\[ \mathbf{\Pi} = \delta \mathcal{L}/\delta \mathbf{A}(\mathbf{r}) = \varepsilon_0 \mathbf{A} \text{ in } V_{\infty}, \] (13)

where here \( \delta \) denotes the functional derivative in the variable \( \mathbf{r} \). The Hamiltonian of the entire system \( H \) has three terms: the contribution of the matter \( H_{\text{mat}} = H_{\text{mat}}(\mathbf{Q}_\nu, \mathbf{Y}_\nu, \mathbf{A}) \), the contribution of the Coulomb interaction \( H_{\text{Coul}} = H_{\text{Coul}}(\mathbf{P}) \), and the contribution of the radiation field \( H_{\text{rad}} = H_{\text{rad}}(\mathbf{\Pi}, \mathbf{A}) \),

\[ H = H_{\text{mat}} + H_{\text{Coul}} + H_{\text{rad}}, \] (14)
where

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

\[ H_{\text{Coul}} = \int_{d\nu} d^3 r \int_{d\nu} d^3 r' \frac{P_\nu(r; t) P_\nu(r'; t)}{8\pi\varepsilon_0 |r - r'|}, \tag{15b} \]

\[ H_{\text{rad}} = \int_{V_{\infty}} d^3 r \left[ \frac{1}{2\varepsilon_0} \Pi^2 + \frac{1}{2\mu_0} (\nabla \times A)^2 \right]. \tag{15c} \]

**Quantization.** In the Heisenberg picture, the vector field operators \( \mathbf{Q}_\nu(r; t) \) and \( Y_\nu(r; t) \) correspond to the canonically conjugate matter vector fields \( \mathbf{Q}_\nu(r; t) \) and \( Y_\nu(r; t) \), and the vector field operators \( \mathbf{P}(r; t) \) and \( A(r; t) \) correspond to the canonically conjugate radiation vector fields \( \mathbf{P}(r; t) \) and \( A(r; t) \). They obey the equal time commutation relations:

\[ \left[ \mathbf{Q}_\nu(r; t), \mathbf{Y}_\nu(r'; t) \right] = -i\hbar \mathbf{I} (\nu - \nu') \delta (r - r') \quad r, r' \in V, \]

\[ \left[ \mathbf{P}(r; t), \mathbf{A}(r'; t) \right] = -i\hbar \mathbf{I} (\nu - \nu') \quad r, r' \in V_{\infty}, \]

while all remaining equal-time commutators vanish. We also introduce the vector field operator \( \mathbf{P}(r; t) \) corresponding to the polarization density field \( \mathbf{P} \),

\[ \mathbf{P}(r; t) = \int_0^\infty \alpha_\nu \mathbf{Y}_\nu(r; t) d\nu \quad \text{in } V \quad \text{in } V \]

and the conjugate momentum

\[ \mathbf{Z}(r; t) = \int_0^\infty \alpha_\nu \mathbf{Q}_\nu(r; t) d\nu \quad \text{in } V. \]

They obey the equal time commutation relation

\[ \left[ \mathbf{Z}(r; t), \mathbf{P}(r'; t) \right] = -i\hbar \mathbf{I} (\nu - \nu') \delta (r - r') \quad r, r' \in V, \]

where \( \beta = \int_0^\infty \alpha_\nu^2 d\nu \). The other equal time commutators with \( \mathbf{A} \) and \( \mathbf{P} \) vanish. The polarization current density operator \( \mathbf{J}(r; t) \) is related to the conjugate momentum \( \mathbf{Z} \) by the equation

\[ \mathbf{J} = \mathbf{Z} - \beta \mathbf{A}. \]

The electric field operator \( \mathbf{E}(r; t) \) is given by

\[ \mathbf{E} = -\frac{1}{\varepsilon_0} \mathbf{P} + \mathbf{E}_{\|} \{ \mathbf{P} \} \]

where

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

**Heisenberg equations.** The Heisenberg equations for the canonical variables follow by evaluating the corresponding commutators with the quantized Hamiltonian. The conjugate canonical operators \( Y_\nu \) and \( Q_\nu \), for \( 0 \leq \nu < \infty \), are governed in \( V \) and for \( 0 \leq t \) by the system of equations:

\[ \dot{Y}_\nu = \dot{Q}_\nu - \alpha_\nu \dot{A}, \tag{24a} \]

\[ \dot{Q}_\nu = -\nu^2 Y_\nu + \alpha_\nu E_{\|} \{ \mathbf{P} \}. \tag{24b} \]

They have to be solved with the initial conditions \( Y_\nu(r; t = 0^-) = Y_\nu^{(S)}(r) \) and \( Q_\nu(r; t = 0^-) = Q_\nu^{(S)}(r) \), where with the upper script \( ^{(S)} \) we denote the corresponding operator in the Schrödinger picture. The canonical conjugate operators \( \mathbf{A} \) and \( \Pi \) are governed in \( V_{\infty} \) and for \( 0 \leq t \) by the system of equations

\[ \dot{\mathbf{A}} = \frac{1}{\varepsilon_0} \Pi, \quad \tag{25a} \]

\[ \dot{\Pi} = -\frac{1}{\mu_0} \nabla^2 \mathbf{A} + \mathbf{P}^\perp, \quad \tag{25b} \]

where

\[ \mathbf{P}^\perp = \mathbf{P} - \varepsilon_0 E_{\|} \{ \mathbf{P} \} \quad \text{in } V_{\infty}. \]

Equations (24a) and (24b) have to be solved with the initial conditions \( \mathbf{A}(r; t = 0^-) = \mathbf{A}^{(S)}(r) \) and \( \Pi(r; t = 0^-) = \Pi^{(S)}(r) \). Eliminating the operators \( \mathbf{Q}_\nu \) and \( \Pi \), the system of equations (24a) and (24b) reduce to

\[ \dot{Y}_\nu + \nu^2 Y_\nu = \alpha_\nu \left[ -\dot{A} + E_{\|} \{ \mathbf{P} \} \right] \quad \text{in } V, \]

\[ \dot{A} - \varepsilon_0^2 \nabla^2 \mathbf{A} = \frac{1}{\varepsilon_0} \mathbf{P}^\perp \quad \text{in } V_{\infty}. \]

This system of equations is solved with the initial conditions:

\[ \dot{Y}_\nu(r; t = 0^-) = Y_\nu^{(S)}(r), \] \[ \dot{Y}_\nu(r; t = 0^-) = \dot{Q}_\nu^{(S)}(r) - \alpha_\nu \dot{A}^{(S)}(r) = \dot{Y}_\nu^{(S)}(r), \] \[ \dot{A}(r; t = 0^-) = \mathbf{A}^{(S)}(r), \] \[ \dot{A}(r; t = 0^-) = \frac{1}{\varepsilon_0} \Pi^{(S)}(r). \]

**Constitutive relation.** Solving equation (27a) with the initial conditions (28a) and (28b) and using (18) we obtain the equation governing the polarization density field operator (Appendix C),

\[ \varepsilon_0 \mathbf{E}(r; t) = h_\eta(t) * (\mathbf{P}(r; t) - \mathbf{P}_f(r; t)) \quad \text{in } V \quad 0 \leq t, \tag{29} \]

where

\[ h_\eta(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \eta(\omega) e^{i\omega t} d\omega \] \[ \text{and } \eta(\omega) = 1/\chi(\omega). \] The free polarization density field operator \( \mathbf{P}_f(r; t) \) takes into account the contribution of the initial conditions of the matter field operators and of
the vector potential operator. To represent it, we introduce an orthonormal discrete real basis \( \{ U_m(r) \} \) defined in \( V \): the vector field \( U_m(r) \) is solenoidal in \( V \) but has a normal component to \( \partial V \) different from zero. The operator \( \hat{P}_f \) is given by

\[
\hat{P}_f(r; t) = \hat{P}_f^+(r; t) + \hat{P}_f^-(r; t) - \varepsilon_0 h_X(t) \hat{A}(S_\ast)(r)
\]

where \( h_X(t) \) is the inverse Fourier transform of \( \chi(\omega) \),

\[
\hat{P}_f^+(r; t) = \sum_{m, \nu} U_m(r) \sqrt{\frac{h_\sigma(\nu)}{\nu \pi}} \hat{c}_{m, \nu} e^{-i \omega t},
\]

where \( \hat{g}^\perp \) is the transverse dyadic Green function in free space for the vector potential in the Coulomb gauge. The free vector potential operator \( \hat{A}_f \) takes into account the contribution of the initial conditions. It describes the evolution of the vector potential operator in the absence of dielectric. The transverse dyadic Green function \( \hat{g}^\perp \) is given by

\[
\hat{g}^\perp(r; t) = \hat{g}^\perp(r; t) - \hat{g}^\parallel(r; t),
\]

where \( \hat{g}^\parallel \) is the dyadic Green function in free space for the vector potential in the temporal gauge (in which the scalar potential is chosen equal to zero),

\[
\hat{g}^\parallel(r; t) = \frac{\hat{I} - \hat{r}_u \hat{r}_u}{4\pi r^2} \hat{\delta}(t - r/c_0) + \frac{\hat{r}_u}{4\pi r^3} u(t - r/c_0) \left[ 1 + \frac{c_0}{\hat{r}_u} (t - r/c_0) \right],
\]

and \( \hat{r}_u = \hat{r}/r \).

To represent \( \hat{A}_f \) we use the transverse plane wave modes of free space

\[
\hat{w}_\mu(r) = \frac{1}{(2\pi)^{3/2}} \varepsilon_{s,k} e^{ikr},
\]

\( k \in \mathbb{R}^3 \) is the propagation vector, \( \{ \varepsilon_{s,k} \} \) are the polarization unit vectors with \( \varepsilon_{s,k} = \varepsilon_{s,-k} \) and \( s = 1, 2; \mu \) is a multi-index corresponding to the pair of parameters \( k \) and \( s, \mu = (k, s), M \) represents the set of all possible \( \mu \), and \( \sum_\mu (\cdot) = \sum_{\mu \in M} \int_\mathbb{R}^3 d^3k (\cdot) \). The two polarization vectors are orthogonal among them, \( \varepsilon_{1,k} \cdot \varepsilon_{2,k} = 0 \), and are both transverse to the propagation vector, \( \varepsilon_{1,k} \cdot k = \varepsilon_{2,k} \cdot k = 0 \). The functions \( \{ \hat{w}_\mu \} \) are orthonormal, \( \langle \hat{w}_\mu', \hat{w}_\mu \rangle = \delta_{\mu', \mu} \delta(k - k') \). The free vector potential operator is given by

\[
\hat{A}_f(r; t) = \hat{A}_f^+(r; t) + \hat{A}_f^-(r; t),
\]

where

\[
\hat{A}_f^+(r; t) = -\sum_{\mu} \frac{i}{\omega_\mu} e_{\mu}(r; t) \hat{a}_\mu,
\]

\[
e_{\mu}(r; t) = i \varepsilon_{\mu} \hat{w}_\mu(r) e^{-i \omega_\mu t},
\]

and \( \omega_\mu = c_0 k, \varepsilon_{\mu} = \{ h_\sigma/\sqrt{2 \varepsilon_0 (2\pi)^3} \}^{1/2} \) and \( \hat{A}_f^-(r) = [\hat{A}_f^+(r)]^\dagger \). The operators \( \hat{a}_\mu \) and \( \hat{a}_\mu^\dagger \) are the annihilation and creation operators in the Schrödinger picture for the transverse electromagnetic field mode of the index \( \mu \). They obey the commutation relations \( [\hat{a}_\mu, \hat{a}_\mu^\dagger] = \delta(\mu - \mu') \), while all other commutators vanish.

The electric field operator is given by

\[
\hat{E}(r; t) = -\frac{i}{\varepsilon_0} \frac{\partial}{\partial t} \int_V d^3r' \int_0^\infty dt' \hat{g}^\perp(r - r', t - t') \hat{P}(r'; t') - \frac{1}{\varepsilon_0} \nabla \int_{\partial V} \frac{\hat{P}(r'; t') \cdot \hat{n}(r')}{4\pi |r - r'|} d^2r' + \hat{E}_f
\]

where \( \hat{P}_f \) is the magnetic field operator based on the scalar Green function for the vacuum.
where
\[ \hat{E}_f (r; t) = \hat{E}_f^{(+)} (r; t) + \hat{E}_f^{(-)} (r; t), \] (42)
and \( \hat{E}_f^{(-)} = [\hat{E}_f^{(+)}]^{\dagger}. \) The expression \( (41) \) can be rewritten in such a way the full wave scalar Green function for the vacuum arises, as in classical covariant electrodynamics. We obtain Eq. \( 3 \) (Appendix \( I \)), where \( \hat{E}_f = \hat{E}_f^{+} + \hat{E}_f^{−} \), and
\[ \hat{E}_f^{+} (r; t) = \sum_{\mu} e_{\mu} (r; t) \hat{a}_{\mu}, \] (43)
and \( \hat{E}_f^{−} (r; t) = -\frac{1}{\varepsilon_0} \nabla \int_{\partial V} \frac{\hat{P}(S) (r')} {4\pi |r - r'|} [u(t') - u(t)] d^2r'. \] (44)
The magnetic field operator is given by Eq. \( 4 \) where \( \mathbf{B}_f = \nabla \times \mathbf{A}_f. \)

**Integral Equation for the polarization density field operator.** Combining equations \( 3 \) and \( 29 \) we obtain the integral differential equation \( 1 \) which governs the evolution of the polarization density field operator for \( t \geq 0 \), where
\[ \hat{F}(r; t) = \hat{h}_b (t) \ast \hat{P}(r; t) + \varepsilon_0 \hat{E}_f (r; t) \ast \hat{F}_{ex}(r; t) \] in \( V \), (45)
and
\[ \hat{F}_{ex}(r; t) = \frac{1} {\varepsilon_0^2} \frac{\partial} {\partial t} \int_{\partial V} \frac{\hat{P}(S) (r')} {4\pi |r - r'|} \delta(t') d^3r'. \] (46)
Equation \( 1 \) has to be solved with vanishing initial conditions at \( t = 0^- \) due to the extra impulsive term \( \hat{F}_{ex} \) introduced in the driving term \( \hat{F}(r; t) \). Now, for our convenience we rewrite the expression of the driving field operator \( \hat{F}(r; t) \) in such a way to highlight the dependence on the annihilation and creation operators. We express \( \hat{F}(r; t) \) as
\[ \hat{F}(r; t) = \sum_{\mu} \{ \hat{F}_{\mu}^{(rad)} (r; t) \hat{a}_{\mu} + [\hat{F}_{\mu}^{(rad)} (r; t)]^{\dagger} \hat{a}_{\mu}^{\dagger} \} + \sum_{m, \nu} \{ \hat{F}_{m, \nu}^{mat} (r; t) \hat{c}_{m, \nu} + [\hat{F}_{m, \nu}^{rad} (r; t)]^{\dagger} \hat{c}_{m, \nu}^{\dagger} \} \] (47)
where
\[ \hat{F}_{\mu}^{(rad)} (r; t) = -\frac{1}{\varepsilon_0} \nabla \int_{\partial V} \frac{U_m (r')} {4\pi |r - r'|} \delta(t') d^2r', \] (48)
and
\[ \hat{F}_{m, \nu}^{mat} (r; t) = \sum_{\mu} \frac{\hat{h}_b (t) \ast [\hat{U}_m (r) + \hat{N}_m (r; t)]} {\nu \pi} \] (49)
and
\[ \hat{N}_m (r; t) = \frac{1} {\varepsilon_0^2} \frac{\partial} {\partial t} \int_{\partial V} \frac{U_m (r')} {4\pi |r - r'|} \delta(t') d^2r' \] (50)

Then, the solution of \( 1 \) is given by
\[ \hat{P}(r; t) = \sum_{\mu} \{ \hat{P}_{\mu}^{(rad)} (r; t) \hat{a}_{\mu} + [\hat{P}_{\mu}^{(rad)} (r; t)]^{\dagger} \hat{a}_{\mu}^{\dagger} \} + \sum_{m, \nu} \{ \hat{P}_{m, \nu}^{mat} (r; t) \hat{c}_{m, \nu} + [\hat{P}_{m, \nu}^{(rad)} (r; t)]^{\dagger} \hat{c}_{m, \nu}^{\dagger} \} \] (51)
where the c-vector field \( \hat{P}_{\beta}^{(c)} (r; t) \) is a solution of the c-integral equation.
\[ h_{\eta}(t) \ast \hat{P}_{\beta}^{(c)} (r; t) - \mathcal{L} \{ \hat{P}_{\beta}^{(c)} \} (r; t) = \mathbf{F}_{\beta}^{(c)} (r; t) \] (52)
with the vanishing initial condition at \( t = 0^- \), \( \alpha = \{ r, m \} \) and \( \beta = \{(m, \nu), \mu \} \).

The expression of the linear integral operator \( \mathcal{L} \{ \hat{P}_{\beta}^{(c)} \} (r; t) \) is the same as that given in \( 2 \) The unknown c-vector fields \( \{ \hat{P}_{\beta}^{(c)} \} \) can be determined by solving Eq. \( 52 \) which has the same form of the Volume Integral Equation governing the evolution of the polarization density field induced in the object, excited by the driving c-field \( \mathbf{F}_{\beta}^{(c)} \) \( 20 \). This equation can be numerically solved by using Finite Elements \( 20 \), either by resorting to Marching on Time (MoT) techniques \( 21 \) or by frequency domain analysis and Fourier transform (see Appendix \( F \)).

Equation \( 52 \) can be also transformed into an equivalent Surface Integral Equation \( 22 \), which is associated with a reduced computational burden. The driving c-field \( \mathbf{F}_{\beta}^{(c)} \) depends on the index \( \nu, \mu \) and \( m \). Nevertheless, only few of \( \{ \hat{P}_{\beta}^{(c)} \} \) contribute to the evolution of the statistical functions of any observable (see Appendix \( F \), depending on the initial quantum state of the system.

**Expressions of the electric and magnetic field operators in terms of the c-fields \( \hat{P}_{\beta}^{(c)} \).** It is convenient to express the electric field operator grouping all the terms depending on the annihilation operators \( \hat{a}_{\mu} \) and \( \hat{c}_{m, \nu} \) and all the terms depending on the creation operators \( \hat{a}_{\mu}^{\dagger} \) and \( \hat{c}_{m, \nu}^{\dagger} \). Therefore, we put
\[ \hat{E}(r; t) = \hat{E}^{(c)} (r; t) + \hat{E}^{(-)} (r; t) \] (53)
where \( \hat{E}^{(c)} \) contains only annihilation operators and \( \hat{E}^{(-)} \) contains only creation operators. We note that \( \hat{E}^{(-)} (r; t) = [\hat{E}^{(c)} (r; t)]^{\dagger} \), as we expect. The term \( \hat{E}^{(+)} \), in turn, has two contributions. We express it as follows,
\[ \hat{E}^{(+)} (r; t) = \hat{E}^{rad}_{rad} (r; t) + \hat{E}^{rad}_{mat} (r; t), \] (54)
where
\[ \hat{E}^{rad}_{rad} (r; t) = \sum_{\mu} \hat{E}_{\mu}^{rad} (r; t) \hat{a}_{\mu}, \] (55a)
and
\[ \hat{E}^{rad}_{mat} (r; t) = \sum_{m, \nu} \hat{E}_{m, \nu}^{mat} (r; t) \hat{c}_{m, \nu}. \] (55b)
The c-fields \( \hat{E}_{\mu}^{rad} (r; t) \) and \( \hat{E}_{m, \nu}^{mat} (r; t) \) are given by
\[ E_{\mu}^{(rad)} (r; t) = \varepsilon_{\mu}^{(rad)} (r; t) + e_{\mu} (r; t), \] (56a)
and
\[ E_{m, \nu}^{(mat)} (r; t) = \varepsilon_{m, \nu}^{(mat)} (r; t) + e_{m, \nu} (r; t). \] (56b)
where \( e_\mu \) is given by Eq.\([30]\)
\[
e^{(\alpha)}_\mu (\mathbf{r}; t) = \mathcal{L} \{ p^{(\alpha)}_\mu \}(\mathbf{r}; t),
\]
for \( \alpha = \{ \text{rad}, \text{mat} \} \) and \( \beta = \{ (m, \nu), \mu \} \), and
\[
e_{m,\nu}(\mathbf{r}; t) = -\frac{1}{\varepsilon_0} \nabla \int_{\partial V} \frac{U_{m,\nu}(\mathbf{r}')}{4\pi |\mathbf{r} - \mathbf{r}'|} [u(t') - u(t)]d^2\mathbf{r}'.
\]
Similarly, we proceed to evaluate for magnetic field operator. We express \( \mathbf{B}(\mathbf{r}; t) \) as
\[
\mathbf{B}(\mathbf{r}; t) = \mathbf{B}^{(+)}(\mathbf{r}; t) + \mathbf{B}^{(-)}(\mathbf{r}; t)
\]
where \( \mathbf{B}^{(-)} = [\mathbf{B}^{(+)}]^\dagger \),
\[
\mathbf{B}^{(+)}(\mathbf{r}; t) = \mathbf{B}^{(\text{rad})}_\mu (\mathbf{r}; t) + \mathbf{B}^{(\text{mat})}_\mu (\mathbf{r}; t),
\]
and
\[
\mathbf{B}^{(\text{rad})}_\mu (\mathbf{r}; t) = \sum_\mu \mathbf{B}^{(\text{rad})}_\mu (\mathbf{r}; t) \hat{a}_\mu,
\]
\[
\mathbf{B}^{(\text{mat})}_\mu (\mathbf{r}; t) = \sum_{m,\nu} \mathbf{B}^{(\text{mat})}_{m,\nu}(\mathbf{r}; t) \hat{c}_{m,\nu}.
\]
The c-fields \( \mathbf{B}^{(\text{rad})}_\mu (\mathbf{r}; t) \) and \( \mathbf{B}^{(\text{mat})}_{m,\nu}(\mathbf{r}; t) \) are given by
\[
\mathbf{B}^{(\text{rad})}_\mu (\mathbf{r}; t) = \mathbf{b}^{(\text{rad})}_\mu (\mathbf{r}; t) + \mathbf{b}^{(\mu)}(\mathbf{r}; t),
\]
\[
\mathbf{B}^{(\text{mat})}_{m,\nu}(\mathbf{r}; t) = \mathbf{b}^{(\text{mat})}_{m,\nu}(\mathbf{r}; t),
\]
where \( \mathbf{b}^{(\text{rad})}_\mu = \mathbf{k} \times \mathbf{e}_\mu/\omega_\mu \), and
\[
b^{(\alpha)}_\mu(\mathbf{r}; t) = \mu_0 \nabla \times \int_{V} \frac{\mathbf{p}^{(\alpha)}(\mathbf{r}'; t)}{4\pi |\mathbf{r} - \mathbf{r}'|} u(t') d^3\mathbf{r}'.
\]
for \( \alpha = \{ \text{rad}, \text{mat} \} \) and \( \beta = \{ (m, \nu), \mu \} \).

**Conclusions.** We have developed an integral equation formulation of macroscopic quantum electrodynamics, in the Heisenberg picture, in finite-size dispersive dielectric that enables the direct application of the consolidated repertory in computational classical electrodynamics to evaluate the time evolution of the observables. There is no need to diagonalize the Hamiltonian. The proposed approach can be also applied to inhomogeneous and non isotropic dielectric objects.

**Appendix A: Longitudinal and transverse components**

In general, the longitudinal component \( \mathbf{C}^\|(\mathbf{r}) \) of a regular vector field \( \mathbf{C}(\mathbf{r}) \) defined in \( V_\infty \) is given by the spatial convolution between the longitudinal dyadic delta function (e.g., \([18]\))
\[
\vec{\delta}^\|(\mathbf{r}) = \nabla \nabla (1/4\pi r)
\]
and \( \mathbf{C}(\mathbf{r}) \). Instead, the transverse component \( \mathbf{C}^\perp(\mathbf{r}) \) is given by the by the spatial convolution with the transverse dyadic delta function
\[
\vec{\delta}^\perp(\mathbf{r}) = \vec{1} \delta(\mathbf{r}) - \vec{\delta}^\|(\mathbf{r})
\]
where \( \vec{1} \) is the three-dimensional unit tensor, and \( \delta(\mathbf{r}) \) is the Dirac delta function in the three dimensional space. The vector fields \( \mathbf{C}^\parallel \) and \( \mathbf{C}^\perp \) are orthogonal according to the scalar product \( \langle \mathbf{F}, \mathbf{G} \rangle = \int_{V_\infty} \mathbf{F}^* (\mathbf{r}) \cdot \mathbf{G} (\mathbf{r}) d^3 \mathbf{r} \).

**Appendix B: Solution of equation \( 27a \)**

In this Appendix, we first solve the equation
\[
\ddot{\mathbf{Y}}_\nu + \nu^2 \mathbf{Y}_\nu = \alpha_\nu \mathbf{E} \quad \text{in} \quad V \quad \text{for} \quad \nu \leq \infty \quad \text{(B1)}
\]
with the initial conditions \([28a] \) and \([28b] \) where \( \mathbf{E} = -\mathbf{\dot{A}} + \mathbf{E} \{ \mathbf{P} \} \). Then, we evaluate the relation between the polarization density field operator and the electric field operator. In the Laplace domain Eq.\([51] \) becomes
\[
(s^2 + \nu^2) \hat{\mathbf{Y}}_\nu = \alpha_\nu \hat{\mathbf{E}} + \left[ s \mathbf{\dot{Y}}_\nu^{(S)}(\mathbf{r}) + \mathbf{\ddot{Y}}_\nu^{(S)}(\mathbf{r}) \right] \quad \text{(B2)}
\]
where \( \hat{\mathbf{Y}}_\nu(\mathbf{r}; s) \) is the Laplace transform of \( \mathbf{Y}_\nu(\mathbf{r}; t) \) and \( \hat{\mathbf{E}}(\mathbf{r}; s) \) is the Laplace transform of \( \mathbf{E}(\mathbf{r}; t) \). Therefore, the polarization density field operator is given by
\[
\hat{\mathbf{P}} = \varepsilon_0 \chi(\mathbf{s}) \hat{\mathbf{E}} + \hat{\mathbf{P}}_f \quad \text{in} \quad V \quad \text{(B3)}
\]
where
\[
\chi(s) = \frac{1}{\varepsilon_0} \int_0^\infty dv \frac{\alpha_\nu^2}{s^2 + \nu^2},
\]
and
\[
\hat{\mathbf{P}}_f(\mathbf{r}; s) = \int_0^\infty dv \frac{\alpha_\nu^2}{s^2 + \nu^2} [s \mathbf{\dot{Y}}_\nu^{(S)}(\mathbf{r}) + \mathbf{\ddot{Y}}_\nu^{(S)}(\mathbf{r})]. \quad \text{(B5)}
\]
Due to dissipation the region of convergence of the Laplace transform contains the imaginary axis, therefore, we evaluate \( \chi(s) \) for \( s = i\omega + \epsilon \) where \( \epsilon \downarrow 0 \). By using the relation (e.g., \([23]\))
\[
\frac{1}{x - i\epsilon} = i\pi \delta(x) + \mathcal{P} \frac{1}{x},
\]
where \( \mathcal{P} \) denotes the Cauchy principal value, we obtain for the susceptibility of the object in the frequency domain \( \chi(\omega) = \chi(s = i\omega + \epsilon) \) the following expression
\[
\varepsilon_0 \chi(\omega) = \mathcal{P} \int_0^\infty dv \frac{\alpha_\nu^2}{\nu^2 - \omega^2 - i\pi \alpha_\nu^2/2 \omega}. \quad \text{(B7)}
\]
The coupling coefficient \( \alpha_\nu \) is given by
\[
\alpha_\nu = \frac{2\sigma(\nu)}{\pi} \quad \text{(B8)}
\]
where \( \sigma(\nu) = -\varepsilon_0 \nu \chi_i(\nu) \) and \( \chi_i \) is the imaginary part of \( \chi(\omega) \).

In the time domain, the relation \([53]\) becomes
\[
\hat{\mathbf{P}}(\mathbf{r}; t) = \varepsilon_0 \chi(t) \mathbf{\dot{E}}(\mathbf{r}; t) + \hat{\mathbf{P}}_f(\mathbf{r}; t) \quad \text{in} \quad V \quad \text{(B9)}
\]
where $h_\chi(t)$ is the inverse Fourier transform of the susceptibility of the dielectric $\chi(\omega)$,

$$\hat{P}_f(r; t) = \int_0^\infty d\nu \sqrt{\frac{2\sigma(\nu)}{\pi}} \hat{Y}_\nu^{\text{free}}(r; t), \quad (B10)$$

and

$$\hat{Y}_\nu(r; t) = \hat{Y}_\nu^{(S)}(r) \cos(\nu t) + \frac{1}{\nu} \hat{Y}_\nu^{(S)}(r) \sin(\nu t). \quad (B11)$$

To represent $\hat{P}_f(r; t)$ we introduce the discrete, orthonormal, real basis $\{U_m(r)\}$ defined in $V$: the vector field $U_m(r)$ is solenoidal in $V$ and has a normal component to $\partial V$ different from zero. We denote by $\{\hat{y}_m,\nu\}$ the coordinate operators of the matter field operator $Y_\nu^{(S)}$ and by $\{\hat{q}_m,\nu\}$ the coordinate operators of the conjugate field operator $Q_\nu^{(S)}$, namely $\hat{Y}_\nu^{(S)}(r) = \sum_m U_m(r) \hat{y}_m,\nu$ and $\hat{Q}_\nu^{(S)}(r) = \sum_m U_m(r) \hat{q}_m,\nu$. The coordinate operators obey the commutation relations

$$[\hat{c}_m,\nu, \hat{c}_m',\nu'] = -i\hbar \delta(\nu - \nu') \delta_{m,m'}, \quad (B12)$$

while all other commutators vanish. We now introduce the annihilation $\hat{c}_m,\nu$ and the creation $\hat{c}_m^\dagger,\nu$ operators for the matter field in the Schrödinger picture. We have

$$\hat{c}_m,\nu = \sqrt{\frac{\nu}{2\hbar}} \left( \hat{y}_m,\nu + \frac{i}{\nu} \hat{q}_m^{(S)} \right). \quad (B13)$$

The annihilation and creation operators obey the commutation relations $[\hat{c}_m,\nu, \hat{c}_m',\nu'] = \delta(\mu - \mu') \delta(\nu - \nu')$, $[\hat{c}_m,\nu, \hat{c}_m',\nu'] = 0$, and $[\hat{c}_m^\dagger,\nu, \hat{c}_m'^\dagger,\nu'] = 0$. Therefore, $\hat{P}_f(r; t)$ is represented as

$$\hat{P}_f(r; t) = [\hat{P}_f^{(t)}(r; t) + \hat{P}_f^{(-)}(r; t)] - \varepsilon_0 h_\chi(t) \hat{A}^{(S)}(r) \quad \text{in } V \quad (B14)$$

where

$$\hat{P}_f^{(t)}(r; t) = \sum_{m,\nu} U_m(r) \sqrt{\frac{\sigma(\nu)}{\nu\pi}} \hat{c}_m,\nu e^{-i\nu t}, \quad (B15)$$

$$\hat{P}_f^{(-)} = [\hat{P}_f^{(t)}]^\dagger,$$

and $\sum_{m,\nu} (\cdot) \equiv \sum_m \int_0^\infty d\nu \, (\cdot)$. The known vector field operator $\hat{P}_f$ takes into account the contribution of the initial conditions of the matter field operators and of the vector potential operators to the polarization dynamics. We call it the free polarization density field operator.

### Appendix C: Solution of equation [27b]

In this appendix, we solve the equation

$$\ddot{\hat{A}} - c_0^2 \nabla^2 \hat{A} = \frac{1}{\varepsilon_0} \hat{P} \perp \quad \text{in } V_\infty \quad (C1)$$

with the initial conditions [28c] and [28d] and express the vector potential operator as a function of the polarization density field operator. To solve equation (C1) we represent the vector potential operator in terms of the transverse plane waves in free space [37]

$$\hat{A}(r; t) = \sum_{\mu} \hat{A}_\mu(t) w_\mu(r), \quad (C2)$$

where $\{\hat{A}_\mu(t)\}$ are the coordinate operators of the vector potential. Substituting (C2) into equation (C1) and projecting both sides of equation onto the transverse plane waves we obtain for any $\mu \in M$:

$$\hat{\ddot{A}}_\mu + \omega_\mu^2 \hat{A}_\mu = \frac{1}{\varepsilon_0} \langle w_\mu, \hat{P} \rangle. \quad (C3)$$

We have used the property $\langle w_\mu, \hat{P} \perp \rangle = \langle w_\mu, \hat{P} \rangle$. Solving equation (C3) we obtain

$$\hat{A}_\mu = \frac{1}{\varepsilon_0} g_\mu(t) \ast \langle w_\mu, \hat{P} \rangle + \hat{\alpha}_\mu(t) \quad (C4)$$

where

$$g_\mu(t) = \frac{1}{\omega_\mu} u(t) \sin(\omega_\mu t), \quad (C5)$$

$u(t)$ is the Heaviside function,

$$\hat{\alpha}_\mu(t) = \frac{1}{\varepsilon_0} \langle \hat{w}_\mu, \hat{A}^{(S)} \rangle \cos(\omega_\mu t) + \frac{1}{\varepsilon_0} \langle \hat{w}_\mu, \hat{A}^{(S)} \rangle \sin(\omega_\mu t), \quad (C6)$$

$$\hat{A}^{(S)}_\mu = \langle w_\mu, \hat{A}^{(S)} \rangle \text{ and } \hat{A}^{(S)}_\mu = \langle w_\mu, \hat{A}^{(S)} \rangle. \quad (C7)$$

Therefore, the vector potential operator is given by

$$\hat{A}(r; t) = \frac{1}{\varepsilon_0} \sum_{\mu} [g_\mu(t) \ast \langle w_\mu, \hat{P} \rangle] w_\mu(r) + \hat{A}_f(r; t), \quad (C7)$$

where

$$\hat{A}_f(r; t) = \sum_{\mu} \hat{\alpha}_\mu(t) w_\mu(r). \quad (C8)$$

Since

$$\sum_\mu [g_\mu(t) \ast \langle w_\mu, \hat{P} \rangle] w_\mu(r) = \int_V d^3 r' \left[ \sum_\mu g_\mu(t) \langle w_\mu(r) w_\mu^*(r') \rangle \right] \hat{P}(r'; t), \quad (C9)$$

we obtain from the relation (C7)

$$\hat{A}(r; t) = \mu_0 \int_V d^3 r' \int_0^\infty dt' g^\perp_\mu(r - r'; t - t') \hat{P}(r'; t') + \hat{A}_f(r; t) \quad (C10)$$

where the expression of the dyad $g^\perp_\mu(r - r'; t)\hat{P}(r'; t')$ is

$$g^\perp_\mu(r - r'; t) = c_0^2 \sum_\mu g_\mu(t) \langle w_\mu(r) w_\mu^*(r') \rangle. \quad (C11)$$
The Laplace transform of $\mathcal{G}^\perp (r - r'; s)$ is
\begin{equation}
\mathcal{G}^\perp (r - r'; s) = \sum_\mu \frac{1}{k^2 + s^2/c_0^2} w_\mu (r) w_\mu^*(r').
\end{equation}
(C12)

Using the expression of $w_\mu (r)$ (see [37]) we obtain the following
\begin{equation}
\mathcal{G}^\perp (r - r'; s) = \frac{1}{(2\pi)^3} \int d^3k \mathcal{G}^\perp (k; s) e^{i k \cdot (r-r')}.
\end{equation}
(C13)

where
\begin{equation}
\mathcal{G}^\perp (k; s) = \frac{1}{k^2 + s^2/c_0^2} \left( \frac{1}{k} - \hat{k} \hat{k} \right)
\end{equation}
is the transverse dyadic Green function for the vector potential in the wavenumber domain and in free space. By evaluating the Fourier integral [C13] we obtain (e.g., [24])
\begin{equation}
\mathcal{G}^\perp (r; s) = \mathcal{G} (r; s) - \mathcal{G}^\parallel (r; s),
\end{equation}
(C15)
where
\begin{equation}
\mathcal{G} (r; s) = \frac{e^{-sr/c_0}}{4\pi r} \left[ (\hat{T} - r_u r_u) + \frac{c_0}{sr} (\hat{T} - 3r_u r_u) (1 + \frac{c_0}{sr}) \right].
\end{equation}
(C16)
\begin{equation}
\mathcal{G}^\parallel (r; s) = \frac{c_0^2}{s^2} \left( \frac{1}{4\pi r^3} \nabla \nabla \right). 
\end{equation}
(C17)

The dyad $\mathcal{G} (r; s)$ is the Green function for the vector potential in the temporal gauge, $\mathcal{G}^\perp (r; s)$ is its transverse component and $\mathcal{G}^\parallel (r; s)$ is its longitudinal component. In the time domain, they become
\begin{equation}
\mathcal{g}^\perp (t; s) = \mathcal{g} (t; t) - \mathcal{g}^\parallel (t; t),
\end{equation}
(C18)\begin{equation}
\mathcal{g}^\parallel (t; s) = \mathcal{g}^\parallel (t; t) - \mathcal{g} (t; t),
\end{equation}
where
\begin{equation}
\mathcal{g} (t; t) = \frac{\hat{T} - r_u r_u}{4\pi r^3} \delta (t - r/c_0) +
\frac{c_0}{4\pi r^3} u (t - r/c_0) \left[ 1 + \frac{c_0}{r} (t - r/c_0) \right],
\end{equation}
(C19)\begin{equation}
\mathcal{g}^\parallel (t; t) = \frac{c_0^2}{4\pi r^3} \frac{\hat{T} - r_u r_u}{4\pi r^3} u (t) t.
\end{equation}
(C20)

The operators $\{ \hat{A}_\mu^\parallel \}$ and the operators $\{ \hat{P}_\mu^\parallel \}$ obey the commutation relations
\begin{equation}
[ \hat{P}_\mu^\parallel, \hat{A}_\mu^\parallel ] = -i \delta_{\mu,\nu} \delta (k - k'),
\end{equation}
for any couple $\mu, \mu' \in \mathcal{M}$, while all other commutators vanish. We express them in terms of the annihilation and creation operators $\hat{a}_\mu$ and $\hat{a}_\mu^\dagger$ for the transverse electromagnetic field mode $\mu$, in the Schrödinger picture, defined as:
\begin{equation}
\hat{a}_\mu = \frac{\omega_\mu}{2\xi_\mu} \left( \hat{A}_\mu^\parallel + \frac{i}{\omega_\mu} \hat{P}_\mu^\parallel \right),
\end{equation}
(C22)
where $\xi_\mu = \{ \hbar \omega_\mu/[2\varepsilon_0 (2\pi)^3] \}^{1/2}$. They obey the commutation relation $[\hat{a}_\mu, \hat{a}_\mu^\dagger] = \delta (\mu - \mu'), [\hat{a}_\mu, \hat{a}_\nu] = 0, [\hat{a}_\mu^\dagger, \hat{a}_\nu^\dagger] = 0$. The known vector field operator $\hat{A}_f (r; t)$, then, is rewritten as
\begin{equation}
\hat{A}_f (r; t) = \hat{A}_f^\parallel (r; t) + \hat{A}_f^\perp (r; t),
\end{equation}
(C23)
where $\hat{A}_f^\perp = [\hat{A}_f^\parallel]^\dagger$ and
\begin{equation}
\hat{A}_f^\parallel (r; t) = -\sum_\mu \frac{i}{\mu} \varepsilon_\mu (r; t) \hat{a}_\mu,
\end{equation}
(C24)
\begin{equation}
\varepsilon_\mu (r; t) = 2i \varepsilon_\mu (r) e^{-i\omega_\mu t}.
\end{equation}
(C25)

The known vector field operator $\hat{A}_f$ takes into account the contribution of the initial conditions of the radiation field operators to the evolution of the radiation field. We call it free vector potential field operator.

Appendix D: Covariant integral expression for the electric and magnetic field operators

The expression of the electric field operator is given by (see equation [11])
\begin{equation}
\int_V \frac{\partial}{\partial t} \int_0^\infty d^3 r' \oint_{\partial V} d^2 r'' \mathcal{g}^\perp (r - r'; t - t') \mathbf{P} (r'; t') - \frac{1}{\varepsilon_0} \nabla \oint_{\partial V} d^2 r' \mathbf{E} (r' ; t) - \frac{1}{\varepsilon_0} \nabla \oint_{\partial V} \mathbf{E} (r' ; t) \cdot \mathbf{n} (r')
\end{equation}
(D1)

It can be rewritten in such a way the full wave scalar Green function for the vacuum occurs, as in the classical covariant electrodynamics. We first consider the first term on the right hand side of [D1]. We represent it as
\begin{equation}
\oint_{\partial V} \mathbf{c} (t; r, r') d^3 r'
\end{equation}
(D2)

Its Laplace transform is
\begin{equation}
\mathcal{C} (s; r, r') = -\mu_0 s \mathcal{g}^\perp (r - r'; s) [s \mathbf{P} (r'; s) - \mathbf{P} (s; r')]
\end{equation}
(D3)

where $\mathbf{P} (r; s)$ is the Laplace transform of the polarization density field operator. The dyadic Green function $\mathcal{G} (r; s)$ can be expressed as
\begin{equation}
\mathcal{G} (r; s) = \mathcal{G} (r; s) \mathcal{T} - \frac{c_0^2}{s^2} \nabla \nabla \mathcal{G} (r; s)
\end{equation}
(D4)
where

\[ G(r; s) = \frac{1}{4\pi r} e^{-sr/c_0} \]  

(D5)

is the scalar full wave Green function for the vacuum. Combining (D4), (C15) and (C17) we obtain

\[ \hat{G}^\perp(r; s) = G(r; s) \hat{\mathbf{1}} - \frac{c_0^2}{s^2} \nabla \nabla [G(r; s) - G(r; s = 0)]. \]  

(D6)

Combining now (D6) and (D3) we have

\[
\hat{C}(s; r, r') = -\mu_0 s G(r - r'; s) [s \hat{P}(r'; s) - \hat{P}(r'; t = 0)] + \frac{1}{\varepsilon_0} \nabla \nabla |G(r - r'; s) - G(r - r'; s = 0)| \hat{P}(r'; s) - \frac{1}{\varepsilon_0 s} \nabla \nabla [G(r - r'; s) - G(r - r'; s = 0)] \hat{P}^{(S)}(r'). \quad (D7)
\]

In the time domain, expression (D7) becomes

\[
\hat{\mathbf{e}}(t; r, r') = -\mu_0 \frac{\partial}{\partial t} \int_0^\infty g(r - r'; t - t') \hat{P}(r'; t') dt' + \frac{1}{\varepsilon_0} \nabla \nabla \int_0^\infty [g(r - r'; t - t') - \eta G(r - r'; t - t')] \hat{P}(r'; t') dt' - \frac{1}{\varepsilon_0} \nabla \nabla \left\{ \int_0^\infty [g(r - r'; t - t') - \eta G(r - r'; t - t')] u(t') dt' \right\} \hat{P}^{(S)}(r'),
\]

(D8)

where

\[ g(r; t) = \frac{1}{4\pi r} \delta(t - r/c_0) \]  

(D9)

and

\[ \eta_0(r) = \frac{1}{4\pi r} \delta(t). \]  

(D10)

Using (D8) the expression (D11) is rewritten as

\[
\hat{\mathbf{E}}(r; t) = -\mu_0 \frac{\partial}{\partial t} \int_0^\infty \hat{P}(r'; t') \cdot \mathbf{n}(r') u(t') d^3 r' - \frac{1}{\varepsilon_0} \int_{\partial V} \hat{P}(r'; t') \cdot \mathbf{n}(r') \frac{1}{4\pi |r - r'|} u(t') d^2 r' + \hat{\mathbf{E}}_f
\]

(D11)

The free electric field operator \( \hat{\mathbf{E}}_f \) has two components, the transverse component \( \hat{\mathbf{E}}_{f\perp} \) that only depend on the free vector potential operator, and the longitudinal component \( \hat{\mathbf{E}}_{f\parallel} \) that only depend on the initial value of the free polarization density field operator.

The magnetic field operator is given by

\[
\hat{\mathbf{B}}(r; t) = \mu_0 \nabla \times \int_0^\infty d^3 r' \int_0^\infty dt' \langle r - r'; t - t' \rangle \hat{P}(r', t') + \nabla \times \hat{A}_f(r; t).
\]

(D14)

Appendix E: Integral Equation \[\] in the Laplace and frequency domains

Equation \[\] can be solved in the frequency domain, and then, the solution in the time domain is evaluated by using the inverse Fourier transform. Applying the Laplace transform to both sides of equation \[\] we obtain the integral equation for the Laplace transform of the polar-
where \( \bar{\chi}(s) = \chi(s/i) \),

\[
G(r; s) = \frac{e^{-sr/c_0}}{4\pi r}, \quad \text{(E2)}
\]

and

\[
\mathcal{F}(r; s) = \frac{1}{\chi(s)} \mathcal{P}_f(r; s) + \varepsilon_0 \mathcal{E}_f(r; s) + \mathcal{F}_{ex}(r; s); \quad \text{(E3)}
\]

\( \mathcal{P}_f \) is the Laplace transform of the field operator \( \hat{\mathbf{P}}_f \), \( \mathcal{E}_f \) is the Laplace transform of the field operator \( \hat{\mathbf{E}}_f \), and \( \mathcal{F}_{ex} \) is the Laplace transform of the field operator \( \hat{\mathbf{F}}_{ex} \), defined in Eq. [6]. It is convenient to rewrite the driving field operator \( \mathcal{F}(r; s) \) as

\[
\mathcal{F}(r; s) = \sum_{\mu} \mathcal{F}_{\mu}^{(rad)}(r; s) \hat{a}_\mu + [\mathcal{F}_{\mu}^{(rad)}(r; s*)]^* \hat{a}_\mu^\dagger + \sum_{m, \nu} \mathcal{F}_{m, \nu}^{(mat)}(r; s) \hat{c}_{m, \nu} + [\mathcal{F}_{m, \nu}^{(rad)}(r; s*)]^* \hat{c}_{m, \nu}^\dagger \quad \text{(E4)}
\]

\[
\mathcal{N}_m(r; s) = \frac{s}{c_0} \int_\mathcal{V} \frac{U_m(r')}{4\pi |r - r'|} d^3r' - \frac{1}{\varepsilon_0 s} \nabla \int_{\partial\mathcal{V}} G(r - r'; s) - G(r - r'; s = 0) \hat{\mathcal{P}}^{(S)}(r') d^2r'. \quad \text{(E8)}
\]

It is sufficient to solve equation [E1] for \( s = i\omega + 0^+ \) with \(-\infty < \omega < \infty\). Therefore, equation [E1] reduces to the volume integral equation in the frequency domain

\[
\frac{1}{\chi(\omega)} \mathcal{P}(r; \omega) - \mathcal{L}_\omega \{ \mathcal{P}(r; \omega) \} = \mathcal{F}(r; \omega), \quad \text{(E9)}
\]

where \( \mathcal{P}(r; \omega) = \mathcal{P}(r; s = i\omega + 0^+) \), \( \mathcal{F}(r; \omega) = \mathcal{F}(r; s = i\omega + 0^+) \),

\[
\mathcal{L}_\omega \{ \mathcal{P} \}(r; \omega) = \frac{\omega^2}{c_0^2} \int_\mathcal{V} G(r - r'; \omega) \mathcal{P}(r'; \omega) d^3r' - \nabla \int_{\partial\mathcal{V}} G(r - r'; \omega) \mathcal{P}(r'; \omega) \cdot \mathbf{n}(r') d^2r', \quad \text{(E10)}
\]

\[
G(r; \omega) = \frac{e^{-ikr}}{4\pi r}, \quad \text{(E11)}
\]

where

\[
\mathcal{F}^{(rad)}_{\mu}(r; s) = \varepsilon_0 \left( 1 + \frac{i}{\omega_\mu} \right) \mathcal{E}_\mu(r; s), \quad \text{(E5)}
\]

is the Laplace transform of the field operator \( \hat{\mathbf{F}}^{rad}_{\mu} \) defined in Eq. [E3] and

\[
\mathcal{F}^{(mat)}_{m, \nu}(r; s) = \sqrt{\frac{\hbar a(\nu)}{\nu \pi}} \left\{ \frac{1}{\chi(s)} \frac{1}{s + i\nu} U_m(r) + \mathcal{N}_m(r; s) \right\}, \quad \text{(E7)}
\]

is the Laplace transform of the field operator \( \hat{\mathbf{F}}^{(mat)}_{m, \nu} \) and

\[
\mathcal{F}^{(rad)}_{\mu}(r; \omega) = \frac{1}{\chi(\omega)} \mathcal{P}_\mu(r; \omega) - \mathcal{L}_\omega \{ \mathcal{P}_\mu(r; \omega) \} + \sum_{\alpha} \{ \mathcal{P}_\mu^{(rad)}(r; \omega) \hat{a}_\mu + [\mathcal{P}_\mu^{(rad)}(r; -\omega)]^* \hat{a}_\mu^\dagger \} + \sum_{\alpha} \{ \mathcal{P}_\mu^{(rad)}(r; -\omega) \hat{a}_\alpha + [\mathcal{P}_\mu^{(rad)}(r; \omega)]^* \hat{a}_\alpha^\dagger \} \quad \text{(E12)}
\]

where the c-vector field \( \mathcal{P}^{(a)}_{\beta}(r; \omega) \) is the solution of the c-integral equation

\[
\frac{1}{\chi(\omega)} \mathcal{P}^{(a)}_{\beta}(r; \omega) - \mathcal{L}_\omega \{ \mathcal{P}^{(a)}_{\beta}(r; \omega) \} = \mathcal{F}^{(a)}_{\beta}(r; \omega), \quad \text{(E13)}
\]

\( \alpha = \{ \text{rad, mat} \} \) and \( \beta = \{ (m, \nu), \mu \} \). To deal with the singularities at \( \omega = \pm \omega_\mu \) we use the relation [E6]. The inverse Fourier transform of \( \mathcal{P}^{(a)}_{\beta}(r; \omega) \) gives \( \mathbf{p}^{(a)}_{\beta}(r; t) \).
Appendix F: Statistical functions

The knowledge of the c-functions $p_{H}^{0}(r; t)$ allows to evaluate the statistical functions of any observable of the system, such as the expectation values, the uncertainty, and the correlation functions. In the following, as an example, we consider single counting rate for photoelectric measurements.

The single counting rate $w_I$ is the mean value in the initial state $|\psi_0\rangle$ of the observable (e.g., [18])

$$I(r; t) = \hat{E}^{(-)}(r; t) \cdot \hat{E}^{(+)}(r; t), \quad (F1)$$

which is arranged in the normal order (all the annihilation operators on the right and all the creation operators on the left), therefore, we have

$$w_I(r; t) = \langle \psi_0 | \hat{E}^{(-)}(r; t) \cdot \hat{E}^{(+)}(r; t) | \psi_0 \rangle. \quad (F2)$$

We assume that

$$|\psi_0\rangle = |\gamma_0\rangle \otimes |\phi_0\rangle \quad (F3)$$

where $\otimes$ denotes the tensor product, $|\gamma_0\rangle$ is the initial state of the matter field supposed to be isolated and $|\phi_0\rangle$ is the initial state of the radiation field. The matter field is initially supposed to be at thermodynamic equilibrium with temperature $T_0$. The radiation field, instead, is assumed to be in eigenstates $|n_1, n_2, ..., n_{\mu}, ...\rangle$ of $H_{rad}$, where $n_{\mu}$ is the number of photons in the mode $\mu$. Due to the assumed initial state, the contributions of the mixed terms $\hat{E}_m^{(-)} \cdot \hat{E}_m^{(+)}$ and $\hat{E}_m^{(-)} \cdot \hat{E}_n^{(+)}$ vanish, thus obtaining

$$w_I(r; t) = w_I^{(rad)}(r; t) + w_I^{(mat)}(r; t) \quad (F4)$$

where

$$w_I^{(rad)}(r; t) = \langle \phi_0 | \hat{E}_r^{(-)} \cdot \hat{E}_r^{(+)} | \phi_0 \rangle =$$

$$\sum_{\mu} \sum_{\mu'} [\hat{E}_\mu^{(rad)}(r; t)]^{*} \cdot \hat{E}_{\mu'}^{(rad)}(r; t) \langle \phi_0 | \hat{a}_{\mu}^{\dagger} \hat{a}_{\mu'} | \phi_0 \rangle, \quad (F5)$$

and

$$w_I^{(mat)}(r; t) = \langle \gamma_0 | \hat{E}_m^{(-)} \cdot \hat{E}_m^{(+)} | \gamma_0 \rangle =$$

$$\sum_{m, \nu, m', \nu'} [\hat{E}_{m, \nu}^{(rad)}(r; t)]^{*} \cdot \hat{E}_{m', \nu'}^{(rad)}(r; t) \langle \gamma_0 | \hat{c}_{m, \nu}^{\dagger} \hat{c}_{m', \nu'} | \gamma_0 \rangle. \quad (F6)$$

To further simplify the discussion, we assume that the radiation field is initially in the one-photon state (e.g., [18])

$$|\phi_0\rangle = \sum_{\mu \in M_0} b_\mu |\bar{a}_{\mu}\rangle |0_r\rangle \quad (F7)$$

and the free polarization polarization operator is uniformly distributed in dielectric object. The ket $|0_r\rangle$ is the vacuum state for the radiation field, $b_\mu$ is the mode amplitude and $M_0$ denotes the set of transverse plane wave modes that are initially in the one photon state. The creation operator $\hat{a}_{\mu}$ acting on the vacuum state $|0_r\rangle$ gives a state with one photon $k$. The mode amplitude $b_\mu$ satisfies the normalization condition $\sum_{\mu \in M_0} |b_\mu|^2 = 1$. The contribution of the initial state of the reservoir to the counting rate is given by

$$w_I^{(mat)}(r; t) = \int_0^\infty d\nu \int_0^\infty d\nu' |\hat{E}_\nu^{(mat)}(r; t)|^{*} \cdot \hat{E}_{\nu'}^{(mat)}(r; t) \langle \gamma_0 | \hat{c}_{\nu}^{\dagger} \hat{c}_{\nu'} | \gamma_0 \rangle \quad (F8)$$

where $\hat{E}_\nu^{(mat)}(r; t)$ is the c-electric field generated by a uniform distribution of polarization in the dielectric object,

$$\langle \gamma_0 | \hat{c}_{\nu}^{\dagger} \hat{c}_{\nu'} | \gamma_0 \rangle = \rho_{\nu} \delta(\nu - \nu') \quad (F9)$$

and

$$\rho_{\nu} = \frac{1}{e^{h\nu/k_BT_0} - 1} \quad (F10)$$

(reservoir oscillators with different frequencies are initially uncorrelated). Therefore, we obtain the following

$$w_I^{(mat)}(r; t) = \int_0^\infty d\nu \rho_{\nu} |\hat{E}_\nu^{(mat)}(r; t)|^2. \quad (F11)$$

The contribution of the initial state of the radiation field to the counting rate is given by

$$w_I^{(rad)}(r; t) = \int \sum_{\mu \in M_0} b_\mu |\hat{E}_\mu^{(rad)}(r; t)|^2. \quad (F12)$$

In the considered scenario, only the wave modes with $\mu \in M_0$ contribute to $w_I^{(mat)}$. 


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

[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] R. Mattoo, 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.

[6] 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.

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

[8] 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.

[9] 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.

[10] 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.

[11] 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.

[12] 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.

[13] 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.

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

[15] 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.

[16] 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.

[17] C. Forestiere and G. Miano, “Operative approach to quantum electrodynamics in dispersive dielectric objects based on a polarization-mode expansion,” *Physical Review A*, vol. 106, p. 033701, Sept. 2022. Publisher: American Physical Society.

[18] 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.

[19] 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.

[20] J. G. Van Bladel, *Electromagnetic fields*, vol. 19. John Wiley & Sons, 2007.

[21] S. M. Rao, ed., *Time domain electromagnetics*. Academic Press series in engineering, San Diego: Academic Press, 1999.

[22] R. F. Harrington, “Boundary Integral Formulations for Homogeneous Material Bodies,” *Journal of Electromagnetic Waves and Applications*, vol. 3, no. 1, pp. 1–15, 1969.

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

[24] 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.