Journal of the Optical Society of America B

OPTICAL PHYSICS

Langevin noise approach for lossy media and the lossless limit

George W. Hanson,[1,*] Frieder Lindel,[2] and Stefan Yoshi Buhmann[2,3]

[1]Department of Electrical Engineering, University of Wisconsin-Milwaukee, 3200 N. Cramer St., Milwaukee, Wisconsin 53211, USA
[2]Institut für Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany
[3]e-mail: stefan.buhmann@physik.uni-freiburg.de
*Corresponding author: george@uwm.edu

Received 4 August 2020; revised 9 December 2020; accepted 18 December 2020; posted 6 January 2021 (Doc. ID 404295); published 0 MONTH 0000

The Langevin noise approach for quantization of macroscopic electromagnetics for three-dimensional, inhomogeneous environments is compared with normal-mode quantization. Recent works on the applicability of the method are discussed, and several examples are provided showing that for closed systems the Langevin noise approach reduces to the usual cavity mode expansion method when loss is eliminated.

https://doi.org/10.1364/JOSAB.404295

1. INTRODUCTION

Methods for the study of the quantum properties of light, and the interaction of quantized light and atoms and other multi-leveled systems, were initially developed for vacuum. The observation of Purcell in 1946 that the spontaneous emission rate of an atom was dependent on the atom's environment [1] was a motivating factor for the study of how cavity materials affect quantized light. The incorporation of simplified models of materials (lossless, dispersionless dielectrics, perfect metals) is accommodated in quantum models in a fairly straightforward manner [2]. However, the Kramers–Kronig relations [3] require that absorption is always accomplished by dispersion, and vice versa. Whereas in classical electromagnetics dispersion and absorption are easily accounted for, in macroscopic quantum models this is not the case, since a naive implementation of absorption causes the commutators to vanish at long times, violating the Heisenberg uncertainty principle.

Motivated by the fluctuation–dissipation theorem [4–18], macroscopic quantum electrodynamics (QED), as the quantum version of classical macroscopic electrodynamics, is a phenomenological dipolar, fully quantum,macroscopic theory developed to accommodate lossy, dispersive materials, and open environments. It has been widely applied to a variety of problems since it is expressed in terms of the Green function, and allows for very general media, including anisotropic, nonreciprocal, and nonlocal materials [16,19–22]. For inhomogeneous, complex-shaped regions, the Green function can be computed numerically [23]. In [24], the phenomenological assumptions are derived from a canonical, path-integral formulation; this approach was later extended to moving media [25]. The equivalence of the approach with an alternative based on auxiliary fields [26] was demonstrated explicitly [27]. A critical assessment is provided in [28] (see also [29,30]), where a comparison with a generalized Huttner–Barnett approach [4] (canonical quantization of a bath of oscillators, based on [31]) is discussed. Canonical quantization for rather general media is discussed in [32–34]. Dissipation and dielectric models are also discussed in a wide range of other works (see, e.g., [35,36]).

In [28], the practical equivalence of the Langevin noise approach (LNA) and Huttner–Barnett descriptions is shown. More precisely, it is shown that in an open system, the material oscillator degrees of freedom included in the standard LNA must be augmented by quantized photonic degrees of freedom associated with fluctuating fields coming from infinity and scattered by the inhomogeneities of the medium. If space is considered to consist of a uniform background having some small absorption, the free fields coming from infinity are absorbed, and the standard LNA applies. However, it is often of interest to model finite regions of space having nonabsorbing materials. In [28], a scheme is developed considering a finite region of space (which may be vacuum), surrounded by a weakly absorbing/dispersive medium ε_{inf} that extends to infinity, and fluctuating polarization currents in ε_{inf} generate the missing free fields, in which case the Huttner–Barnett and LNAs are shown to be equivalent.

Nevertheless, questions about the validity of the LNA remain [37,38], particularly, concerning various limiting procedures such as assuming the material region of interest shrinks to zero, or the limit of a lossless material is taken. In this work, we compare the LNA with the standard cavity normal-mode approach, which we refer to as normal-mode QED (NMQED) in the following, which is valid for media characterized by Hermitian permittivity tensors (lossless, and, therefore, nondispersive).

Although it is known that the LNA recovers various quantities correctly, such as the atomic spontaneous decay rate, here we show for several explicit examples that the LNA results in exactly the same formulation (final equations) as the NMQED, although the former allows for much more general materials than the latter. Several possible geometries may be envisioned: (1) finite-size, PEC-wall cavities (i.e., closed systems) containing lossless inhomogeneous media; (2) same as (1) but for lossy, dispersive media; (3) large-cavity limit cavities containing lossless inhomogeneous media; (4) same as (3) but for lossy inhomogeneous media; (5) open systems, which admit loss even when the materials themselves are lossless. Cases (3) and (4) are actually subsets of (1) and (2); in the former, plane wave eigenfunctions are used, whereas in the latter, more general cavity eigenfunctions are used. For (1), NMQED is standard, often with homogeneous media (e.g., vacuum). The LNA does not apply to Case (1) directly, but can be applied to Case (2), the lossy version. Here, we show that the LNA recovers exactly the NMQED equations for several problems considered in the lossless limit [i.e., as Case (2) reduces to Case (1)]. For Case (3), the NMQED is often used for homogeneous environments (utilizing discrete plane wave mode functions to represent the actual mode continuum). Again, the LNA cannot be applied directly to Case (3), although it applies to Case (4) and again recovers the NMQED result in the lossless limit. In fact, the resulting equations from the LNA, e.g., the density operator or population evolution, are easily converted to the NMQED (and, sometimes, vice versa) using a simple Green function relation. For the study of nonabsorbing materials, we point out the need to retain dissipation in the LNA model until the final steps of the calculation, at which point the lossless limit can be taken. Similarly, if, say, the medium inhomogeneities vanish (e.g., the structure of interest, such as a metal resonator, shrinks to zero size), that limit must be taken at the end of the development. The lossless limiting case in the LNA has also been examined in [6], and recently in [39].

Open systems, Case (5), cannot be modeled using cavity normal modes, but it can be modeled using LNA (in the references cited above, it is inherently a system-bath approach). For open systems, a quasinormal mode quantization (also based on a Langevin noise model) is a useful and natural approach for arbitrarily lossy open system modes [40], and it implements a formulation akin to the standard modal approach, but for open lossy systems. An advantage of quasinormal modes beyond the LNA is to explore nonlinear quantum optics at the system level, where it is no longer valid to treat the medium as a bath, e.g., [41,42].

2. BASIC RELATIONS

We first consider an environment/reservoir such as a three-dimensional (3D) cavity $\Omega \subseteq \mathbb{R}^3$ with closed surface Σ, having a uniform background material characterized by $\varepsilon_{\text{bulk}}$ and containing a region $\Omega_1 \subseteq \Omega$ inhomogeneously filled with material characterized by relative permittivity tensor $\varepsilon_1(\mathbf{r}, \omega)$ [assumptions about $\varepsilon_1(\mathbf{r}, \omega)$ will be described in each subsection below]. The permittivity for all $\mathbf{r} \in \Omega$ is $\varepsilon(\mathbf{r}, \omega)$. We will assume the magnetic permeability is the unity tensor, although

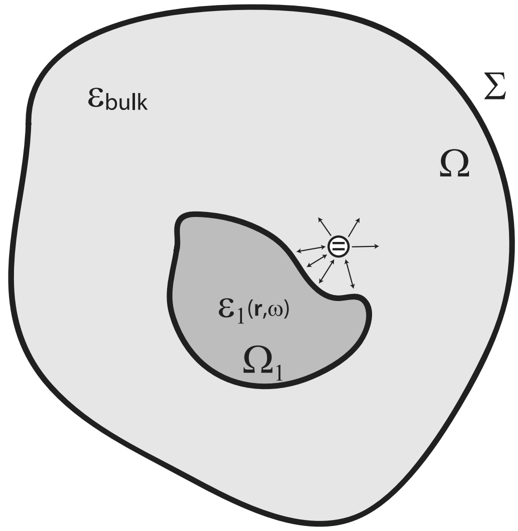

Fig. 1. Two-level system in the vicinity of an inhomogeneous region $\Omega_1 \subseteq \Omega \subseteq \mathbb{R}^3$.

including a permeability response does not change the presented conclusions. As the notation indicates, we can allow $\Omega_1 = \Omega$, and Ω can be finite (e.g., a closed system with surface Σ perfectly conducting), or in the large-cavity limit. The geometry is depicted in Fig. 1, including a two-level system located somewhere within Ω. We compare two formulations.

A. Normal-Mode QED Approach

NMQED is the usual textbook [43–47] and research [48,49] approach for (i) closed empty cavities, where $\boldsymbol{\varepsilon}(\mathbf{r}, \omega) = \mathbf{I}$, with $\mathbf{I}$ the identity operator; (ii) closed cavities filled with lossless, dispersionless media, where $\boldsymbol{\varepsilon}(\mathbf{r})$ is a real-valued, symmetric tensor; and (iii) closed cavities homogeneously filled with lossy media. For the first two cases, classical mode functions $\mathbf{E}_{\mathbf{k}}(\mathbf{r}) = \mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}})$ can be defined that satisfy [50,51]

$$\nabla \times \nabla \times \mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}}) = \frac{\omega_{\mathbf{k}}^2}{c^2} \boldsymbol{\varepsilon}(\mathbf{r}) \cdot \mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}}), \tag{1}$$

subject to boundary conditions on the cavity walls, $\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}})|_{\mathbf{r}_{\text{wall}}} = \mathbf{0}$, $\hat{\mathbf{n}}$ being the unit normal vector to the wall, with eigenfunction orthogonality [50],

$$\int \mathbf{E}_{\mathbf{k}}^*(\mathbf{r}, \omega_{\mathbf{k}}) \cdot \boldsymbol{\varepsilon}(\mathbf{r}) \cdot \mathbf{E}_{\mathbf{k}'}(\mathbf{r}, \omega_{\mathbf{k}'}) \mathrm{d}^3\mathbf{r} = \delta_{\mathbf{k}\mathbf{k}'}. \tag{2}$$

Under the restriction of a Hermitian permittivity tensor, and defining a subspace of differentiable vector functions dense in the Hilbert space of Lebesgue integrable vector functions $\mathbf{L}^2$, the operator $L_E : \mathbf{L}^2(\Omega)^3 \to \mathbf{L}^2(\Omega)^3$, $L_E \mathbf{x} \equiv \nabla \times \nabla \times \mathbf{x} - \frac{\omega_{\mathbf{k}}^2}{c^2} \varepsilon(\mathbf{r}) \cdot \mathbf{x}$, with boundary condition $B(\mathbf{x}) = \mathbf{n} \times \mathbf{x}|_\Gamma = 0$ or $B(\mathbf{x}) = \mathbf{n} \times \nabla \times \mathbf{x}|_\Gamma = 0$ is self-adjoint (SA) and negative-definite, and the modes form an orthonormal, complete set in the Hilbert space of square-integrable functions [50],

$$\mathbf{I}\delta(\mathbf{r} - \mathbf{r}') = \sum\nolimits_{\mathbf{k}} \mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}}) \mathbf{E}_{\mathbf{k}}^*(\mathbf{r}', \omega_{\mathbf{k}}) \cdot \varepsilon(\mathbf{r}'). \tag{3}$$

The electric field operator in the Schrödinger picture is

$$\hat{\mathbf{E}}(\mathbf{r})^{\mathrm{NMQED}} = \sum_{\mathbf{k}} \hat{\mathbf{E}}_{\mathbf{k}}(\mathbf{r}) + \mathrm{H.c.}, \tag{4}$$

where

$$\hat{\mathbf{E}}_{\mathbf{k}}(\mathbf{r}) = i\sqrt{\frac{\hbar\omega_{\mathbf{k}}}{2\varepsilon_0}}\hat{a}_{\mathbf{k}}\mathbf{E}_{\mathbf{k}}(\mathbf{r}), \tag{5}$$

and where $\hat{a}_{\mathbf{k}}$, $\hat{a}^{\dagger}_{\mathbf{k}}$ are annihilation and creation operators that satisfy

$$\left[\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}'}\right] = \left[\hat{a}^{\dagger}_{\mathbf{k}}, \hat{a}^{\dagger}_{\mathbf{k}'}\right] = 0, \ \left[\hat{a}_{\mathbf{k}}, \hat{a}^{\dagger}_{\mathbf{k}'}\right] = \delta_{\mathbf{k}\mathbf{k}'}. \tag{6}$$

In the Heisenberg picture, Eq. (6) becomes equal-time commutators.

The free-field Hamiltonian is (dropping the zero-point energy)

$$\hat{H}^{\mathrm{NMQED}} = \sum_{\mathbf{k}s} \hbar\omega_{\mathbf{k}}\hat{a}^{\dagger}_{\mathbf{k}s}\hat{a}_{\mathbf{k}s}, \tag{7}$$

and eigenfunctions of the Hamiltonian are the multimode number (Fock) states,

$$|n_1\rangle|n_2\rangle|n_3\ldots\rangle \equiv |n_1, n_2, n_3\ldots\rangle = |\{n_j\}\rangle, \tag{8}$$

which can be obtained from the ground state as

$$|n_1\rangle|n_2\rangle\ldots n_i,\ldots = \frac{(\hat{a}^{\dagger}_1)^{n_1}}{\sqrt{n_1!}}\cdots\frac{(\hat{a}^{\dagger}_i)^{n_i}}{\sqrt{n_i!}}\cdots|0\rangle. \tag{9}$$

For the special case of an optically large vacuum cavity, the cavity mode functions become

$$\mathbf{E}_{\mathbf{k}s}(\mathbf{r}) \to \frac{\mathbf{e}_{\mathbf{k}s}}{\sqrt{V}}e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{10}$$

which satisfy periodic boundary conditions (Ω is assumed to be the union of boxes of volume V), where s indicates spin (polarization), with $\mathbf{e}_{\mathbf{k}s}$ being an orthonormal set of polarization functions such that $\mathbf{e}_{\mathbf{k}s}\cdot\mathbf{e}_{\mathbf{k}'s'} = \delta_{\mathbf{k}\mathbf{k}'}\delta_{ss'}$, and satisfy the transversality condition $\mathbf{k}\cdot\mathbf{e}_{\mathbf{k}s} = 0$. The polarization vectors form a right-handed coordinate system, $\mathbf{e}_{\mathbf{k}1}\times\mathbf{e}_{\mathbf{k}2} = \mathbf{k}/|\mathbf{k}|$. In Eq. (10), V is a quantization volume such that

$$\int_V \mathbf{E}^*_{\mathbf{k}s}(\mathbf{r})\cdot\mathbf{E}_{\mathbf{k}'s'}(\mathbf{r})\mathrm{d}^3\mathbf{r} = \delta_{\mathbf{k}\mathbf{k}'}\delta_{ss'}. \tag{11}$$

Note, however, that this is not an open system (truly infinite space), which inherently allows dissipation (photons going to infinity and never coming back). Mathematically, the difference between a large cavity and a true open system is that for the latter, modes must satisfy the Sommerfeld radiation condition, which renders the operator L_E to be non-SA; the Sommerfeld radiation condition is an outgoing wave condition, and the adjoint condition is an inward-traveling wave.

Finally, for Case (iii), a cavity homogeneously filled with lossy media, rather than L_E, the operator $L\mathbf{x} \equiv \nabla\times\nabla\times\mathbf{x}$ can be defined such that eigenfunctions of L satisfy the boundary condition $B(\mathbf{x}) = \mathbf{0}$, and the resulting operator is SA. The cavity must be homogeneously filled; material inhomogeneities in piecewise constant media would necessitate boundary conditions B such that $B \neq B^*$, rendering the problem non-SA.

In the usual NMQED, the photonic Green function is not explicitly needed, although it implicitly arises in, e.g., atom–atom coupling terms. However, to make connection with the LNA, it is important to connect the mode functions $\mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}})$ with the Green tensor, which is defined by

$$\nabla\times\nabla\times\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2_{\mathbf{k}}}{c^2}\boldsymbol{\varepsilon}(\mathbf{r})\cdot\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{I}\delta(\mathbf{r}-\mathbf{r}') \tag{12}$$

and satisfies $\mathbf{G}(\mathbf{r}, \mathbf{r}')^{\mathrm{T}} = \mathbf{G}(\mathbf{r}', \mathbf{r})$. The Green tensor can be expanded as

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mathbf{k}} c^2\frac{\mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}})\mathbf{E}^*_{\mathbf{k}}(\mathbf{r}', \omega_{\mathbf{k}})}{\omega^2_{\mathbf{k}} - \omega^2}. \tag{13}$$

Equation (13) formally encompasses the case of transverse modes, forming a transverse Green function, or could include longitudinal modes as well. It should be emphasized that Eq. (13) is only valid for closed cavities and the three cases discussed, although the Green tensor concept itself extends to dispersive and lossy inhomogeneous media. For certain spatial positions, a quasinormal mode expansion of the Green function is also possible [40,41].

An important expression relating the Green function and modal summation is obtained by integrating Eq. (13) with respect to frequency and using the Sokhotski–Plemelj (SP) identity

$$\lim_{\epsilon\to 0^+}\frac{1}{x\pm i\epsilon} = \mathrm{PV}\left(\frac{1}{x}\right) \mp i\pi\delta(x), \tag{14}$$

leading to

$$\frac{1}{\pi}\int_0^\infty \mathrm{d}\omega\frac{\omega^2}{c^2}\mathrm{Im}\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mathbf{k}}\frac{\omega_{\mathbf{k}}}{2}\mathbf{E}_{\mathbf{k}}(\mathbf{r}, \omega_{\mathbf{k}})\mathbf{E}^*_{\mathbf{k}}(\mathbf{r}', \omega_{\mathbf{k}}). \tag{15}$$

This is the key relationship that allows converting between the LNA and NMQED, and it will be needed in the following. Since the case $\mathbf{r} = \mathbf{r}'$ is often needed in field-atom interactions, it is worth noting that in the event of material loss at point $\mathbf{r}$ ($\mathrm{Im}(\varepsilon(\mathbf{r}, \omega_\lambda)) > 0$), $\mathrm{Im}\mathbf{G}(\mathbf{r}, \mathbf{r}, \omega) \to \infty$, which is not seen with the transverse Green function/transverse mode expansion.

B. Langevin Noise Approach

The LNA is developed in detail in [6–18], and here we merely use the main results as needed. We now allow a dispersive absorbing (complex-valued) permittivity, with causality requiring $\varepsilon(\mathbf{r}, -\omega) = \varepsilon^*(\mathbf{r}, \omega^*)$. For the Green function, $\mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{G}(\mathbf{r}, \mathbf{r}', -\omega^*)$, and we impose the condition

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \to \mathbf{0} \text{ for } |\mathbf{r}-\mathbf{r}'| \to \infty, \tag{16}$$

associated with some material absorption. This is an often-overlooked requirement, which is discussed further in Section 5.

The electric field operator in the Schrödinger picture is

$$\hat{\mathbf{E}}(\mathbf{r})^{\mathrm{LNA}} = \int_0^\infty \mathrm{d}\omega_\lambda\, \hat{\mathbf{E}}(\mathbf{r}, \omega_\lambda) + \mathrm{H.c.}, \tag{17}$$

where ω_λ is a continuum modal frequency (not a Fourier transform frequency), with

$$\hat{\mathbf{E}}(\mathbf{r},\omega_\lambda)=i\sqrt{\frac{\hbar}{\pi\varepsilon_0}\frac{\omega_\lambda^2}{c^2}}\int \mathrm{d}^3\mathbf{r}'\mathbf{G}(\mathbf{r},\mathbf{r}',\omega_\lambda)\cdot\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}',\omega_\lambda))}\cdot\hat{\mathbf{f}}(\mathbf{r}',\omega_\lambda), \quad (18)$$

where $\hat{\mathbf{f}}, \hat{\mathbf{f}}^\dagger$ are canonically conjugate field variables, which are continuum bosonic operator-valued vectors of the combined matter-field system that satisfy

$$\left[\hat{f}_k(\mathbf{r},\omega),\hat{f}^\dagger_{k'}(\mathbf{r}',\omega')\right]=\delta_{kk'}\delta(\omega-\omega')\delta(\mathbf{r}-\mathbf{r}'), \quad (19)$$

$$\left[\hat{f}_k(\mathbf{r},\omega),\hat{f}_{k'}(\mathbf{r}',\omega')\right]=\left[\hat{f}^\dagger_k(\mathbf{r},\omega),\hat{f}^\dagger_{k'}(\mathbf{r}',\omega')\right]=0. \quad (20)$$

Comparing the two approaches, $\int_0^\infty \mathrm{d}\omega_\lambda\,\hat{\mathbf{f}}(\mathbf{r},\omega_\lambda)$ is seen to be the continuous analog of $\sum_{\mathbf{k},s}\mathbf{E}_\mathbf{k}(\mathbf{r})\hat{a}_{\mathbf{k}s}$. It is worth noting that $\mathrm{Im}(\varepsilon(\mathbf{r},\omega_\lambda))$ is a positive semi-definite matrix in the anisotropic case [16], so that its square root is well-defined.

More complicated environments, including nonlocal and nonreciprocal media, have also been considered [16,19–22]. The conclusions described below hold for generally lossy, inhomogeneous, nonreciprocal media.

The free field-matter Hamiltonian is

$$\hat{H}^{\mathrm{LNA}}=\int_0^\infty \mathrm{d}\omega\int \mathrm{d}\mathbf{r}\,\hbar\omega\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega), \quad (21)$$

which is analogous to Eq. (7). Energy eigenstates of the free Hamiltonian are compositions of $|n_i(\mathbf{r},\omega_\lambda)\rangle$ (analogous to $|\{n\}\rangle$ in the cavity mode case), which indicates that the λ^{th} field mode of the nonuniform continuum is populated with n quanta, and that it is vector-valued with field component in the i^{th} direction. As a trivial example, the one-quanta states are obtained from the ground state as

$$|1_i(\mathbf{r},\omega_\lambda)\rangle=\hat{f}^\dagger_i(\mathbf{r},\omega_\lambda)|\{0\}\rangle. \quad (22)$$

An important relation in developing LNA formulations is the "magic formula" [7],

$$\frac{\omega^2}{c^2}\int \mathrm{d}^3\mathrm{r}'\mathrm{Im}\left(\varepsilon(\mathbf{r}',\omega)\right)\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{G}^\dagger(\mathbf{r}_0,\mathbf{r}',\omega)=\mathrm{Im}\mathbf{G}(\mathbf{r},\mathbf{r}_0,\omega), \quad (23)$$

generalized for tensor permittivity as [16,21]

$$\frac{\omega^2}{c^2}\int \mathrm{d}^3\mathrm{r}''\mathbf{G}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{T}(\mathbf{r}'',\omega)\cdot\mathbf{T}^\dagger(\mathbf{r}'',\omega)\cdot\mathbf{G}^\dagger(\mathbf{r}',\mathbf{r}'',\omega)=\left(\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)-\mathbf{G}^\dagger(\mathbf{r}',\mathbf{r},\omega)\right)/2i, \quad (24)$$

where $\mathbf{T}(\mathbf{r},\omega)=\sqrt{\mathrm{Im}\boldsymbol{\varepsilon}(\mathbf{r},\omega)}$ (and valid for nonreciprocal media using $\mathbf{T}(\mathbf{r},\omega)\cdot\mathbf{T}^\dagger(\mathbf{r},\omega)=\frac{1}{2i}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)-\boldsymbol{\varepsilon}^\dagger(\mathbf{r},\omega)]$). The above integrals generally do not need to be evaluated explicitly, but they are used in the derivation of system equations; their use removes $\mathrm{Im}(\varepsilon(\mathbf{r}',\omega))$ from the resulting equations, allowing the lossless limit to be subsequently taken.

Furthermore, the correlation relation can be shown to be [16]

$$\langle 0|\mathbf{E}(\mathbf{r},\omega)\mathbf{E}^\dagger(\mathbf{r},\omega')|0\rangle=\frac{\hbar k_0^2}{2\varepsilon_0^2}N\mathrm{Im}(\mathbf{G}(\mathbf{r},\mathbf{r},\omega))\delta(\omega-\omega'),$$

where $N(\omega,T)=2/(\exp(\hbar\omega/k_BT)-1)$ for negative frequencies and $N(\omega,T)=1+2/(\exp(\hbar\omega/k_BT)-1)$ for positive frequencies, where k_B is Boltzmann's constant.

Conversion to the time-domain is achieved by changing to the Heisenberg picture, where operators $\hat{A}$ transform as $\hat{A}_H(t)=e^{i\hat{H}_{\mathrm{Sch}}t/\hbar}\hat{A}_{\mathrm{Sch}}e^{-i\hat{H}_{\mathrm{Sch}}t/\hbar}$, leading to

$$\hat{\mathbf{E}}(\mathbf{r},t)=\int_0^\infty \mathrm{d}\omega_\lambda i\sqrt{\frac{\hbar}{\pi\varepsilon_0}\frac{\omega_\lambda^2}{c^2}}\times\int \mathbf{G}(\mathbf{r},\mathbf{r}',\omega_\lambda)\cdot\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}',\omega_\lambda))}\cdot\hat{\mathbf{f}}(\mathbf{r}',\omega_\lambda,t)\mathrm{d}^3\mathbf{r}'+\mathrm{H.c.} \quad (25)$$

In summary, to compare the two methods, the NMQED is the standard method ubiquitous in quantum optics. It is a natural and convenient method to study cavity-QED (e.g., Jaynes–Cummings models), nonclassical light, and many-quanta correlations. It puts the system background (e.g., cavity) on a similar footing as the system (e.g., an atom), both being modes/harmonic oscillators. The LNA is a system-bath approach that focuses attention on the system (e.g., the atom), while rigorously accounting for the system environment, the latter being relegated to the status of a bath. Although NMQED can be complimented by system-bath decay operators that approximately account for the non-Hermitian (outgoing and incoming) nature of the cavity modes in real systems, the commutation rules assumed are formally only valid for $Q\to\infty$, a restriction not needed for the LNA. In the LNA, there is often some confusion about the integration limits and the limit $\mathrm{Im}(\varepsilon(\mathbf{r},\omega))\to 0$, discussed further in the following.

3. EXAMPLE I: EXCITED ATOM INTRODUCED INTO A STRUCTURED RESERVOIR—NON-MARKOVIAN WEISSKOPF–WIGNER ANALYSIS

As a first example, in this section, we consider introducing an excited-state atom at $\mathbf{r}=\mathbf{r}_0$, $t=0$ into a structured reservoir [22], comparing the NMQED and LNAs in the context of 3D quantization in the limit $\mathrm{Im}(\varepsilon(\mathbf{r},\omega))\to 0$.

The multipolar-coupling, Coulomb gauge Hamiltonian operator is

$$H=H^{\mathrm{NMQED/LNA}}+\hbar\omega_0\hat{\sigma}_+\sigma_--\hat{\boldsymbol{p}}\cdot\hat{\mathbf{E}}(\mathbf{r}_0), \quad (26)$$

where $\hat{\sigma}_\pm$ represents the canonically conjugate two-level atomic operators ($\hat{\sigma}_+=|e\rangle\langle g|$, $\hat{\sigma}_-=|g\rangle\langle e|=\hat{\sigma}_+^\dagger$, with $|e\rangle$ and $|g\rangle$ being the excited and ground atomic states, respectively), and $\hat{\boldsymbol{p}}=(\hat{\sigma}_++\hat{\sigma}_-)\gamma$ is the dipole operator, where γ is the dipole operator matrix-element, assumed real-valued. The first term in each case is the free Hamiltonian for the field modes (field-matter modes for the LNA), the second term is the free Hamiltonian for the dipole, and the last term is the interaction term.

The equation of motion is

$$\frac{d}{dt}|\psi\rangle = -\frac{i}{\hbar}H|\psi\rangle, \tag{27}$$

and in each case the atom-field product states are

$$|\psi(t)\rangle^{\mathrm{NMQED}} = c_e(t)|e,0\rangle + \sum_\lambda c_\lambda(t)|g,1_\lambda\rangle, \tag{28}$$

$$|\psi(t)\rangle^{\mathrm{LNA}} = c_e(t)|e,0\rangle + \int \mathrm{d}^3\mathbf{r}\int_0^\infty \mathrm{d}\omega_\lambda c_{gi}(\mathbf{r},\omega_\lambda,t)|g,1_i(\mathbf{r},\omega_\lambda)\rangle, \tag{29}$$

where $|e,0\rangle \equiv |e\rangle\otimes|\{0\}\rangle$ and $|g,1_i(\mathbf{r},\omega_\lambda)\rangle \equiv |g\rangle\otimes|\{1_i(\mathbf{r},\omega_\lambda)\}\rangle$. The interaction Hamiltonian $\hat{\boldsymbol{p}}\cdot\hat{\mathbf{E}}(\mathbf{r}_0)\sim(\hat{\sigma}_+ + \hat{\sigma}_-)(\hat{\boldsymbol{f}} + \hat{\boldsymbol{f}}^\dagger)$ acting on the initial state $|e,0\rangle$ leads to an infinite-dimensional Hilbert space of the set of states $A = \{|e,0\rangle, |g,1\rangle, |e,2\rangle, |g,3\rangle, |e,4\rangle, \ldots\}$, where the $n>1$ photons could be in the same or different field modes. Here, we truncate the space to consist of $\{|e,0\rangle, |g,1\rangle\}$, which is equivalent to a rotating wave approximation even when using the full interaction Hamiltonian.

For the NMQED, plugging $|\psi(t)\rangle^{\mathrm{NMQED}}$ into the equation of motion and defining

$$g_{\mathbf{k}} = \gamma\cdot i\sqrt{\frac{\hbar\omega_{\mathbf{k}}}{2\varepsilon_0}}\mathbf{E}_{\mathbf{k}}(\mathbf{r}_0), \tag{30}$$

multiplying by $\langle e,0|$ and $\langle g,1_{\lambda'}|$, and discarding higher-order terms like $\hat{a}^\dagger_{\mathbf{k}s}(0)|g,1_\lambda\rangle \sim |g,2_\lambda\rangle$ leads to [45]

$$\frac{d}{dt}c_e = -ic_e\omega_0 + \frac{i}{\hbar}\sum_\lambda g_\lambda c_\lambda, \tag{31}$$

$$\frac{d}{dt}c_\lambda = \frac{i}{\hbar}c_e g^*_\lambda - i\omega_\lambda c_\lambda. \tag{32}$$

Defining slowly varying amplitudes $c_{\mathrm{es}}(t) = c_e(t)e^{i\omega_0 t}$ and $c_{\lambda s}(t) = c_\lambda(t)e^{i\omega_\lambda t}$, where ω_0 is the energy level transition frequency, we have

$$c_{\lambda s}(t) = \frac{i}{\hbar}g^*_\lambda\int_0^t \mathrm{d}t' c_{\mathrm{es}}(t')e^{i(\omega_\lambda-\omega_0)t'}, \tag{33}$$

and so the population is obtained by solving the Volterra integral equation of the second kind

$$\frac{dc_{\mathrm{es}}(t)}{dt} = \int_0^t D(t,t')c_{\mathrm{es}}(t')\mathrm{d}t', \tag{34}$$

with the kernel

$$D^{\mathrm{NMQED}}(t,t') = -\frac{1}{\hbar^2}\sum_\lambda |g_\lambda|^2 e^{-i(\omega_\lambda-\omega_0)(t-t')}. \tag{35}$$

The Volterra integral equation has been widely utilized in quantum optics (see, e.g., [52–54]) and can accommodate non-Markovian processes. The procedure for numerically solving the Volterra integral equation is shown in [22,55]. The initial-value condition $c_{eo}(0) = 1$ is assumed, representing an initially excited atom.

Repeating the same procedure for the LNA (details are in [22]) leads to Eq. (34), where [8,10]

$$D^{\mathrm{LNA}}(t,t') = -\frac{1}{\hbar\pi\varepsilon_0}\int_0^\infty \mathrm{d}\omega_\lambda \frac{\omega_\lambda^2}{c^2}\gamma\cdot\mathrm{Im}\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_\lambda)\cdot\gamma \times e^{-i(\omega_\lambda-\omega_0)(t-t')} \doteqdot D^{\mathrm{NMQED}}(t,t'), \tag{36}$$

using Eqs. (23), (30), and (15), where $\doteqdot$ indicates equality in the lossless limit of the LNA formulation [i.e., when Eq. (15) holds]. The term $\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}',\omega_\lambda))}$ does not appear in the expression for D^{LNA}. Since the LNA can accommodate generally lossy, dispersive media, the LNA approach exactly recovers the NMQED as a special case. There is no need to explicitly take the limit as $\mathrm{Im}\{\varepsilon(\mathbf{r},\omega)\}\to\mathbf{0}$, as one merely computes the Green function assuming lossless media. This is discussed further in Section 5. The LNA also applies to open systems, where the Green function accounts for the infinite space. The vacuum limit is obtained merely by using the vacuum Green function.

To recover the familiar Markov result, setting $c_{\mathrm{es}}(t') = c_{\mathrm{es}}(t)$ and using the SP identity $\int_0^\infty e^{\pm i(\omega-\omega_0)\tau}\mathrm{d}\tau = \pi\delta(\omega-\omega_0)\pm i\,\mathrm{PV}\left(\frac{1}{\omega-\omega_0}\right)$, Eq. (34) can be solved as

$$c_{\mathrm{es}}(t) = c_{\mathrm{es}}(0)\,e^{-\Gamma\frac{1}{2}t}e^{i\delta t}, \tag{37}$$

and the probability of excited-state occupation is $P(t) = |c_{\mathrm{es}}(t)|^2 = |c_{\mathrm{es}}(0)|^2 e^{-\Gamma t}$. In Eq. (37),

$$\Gamma = 2\frac{\pi}{\hbar\pi\varepsilon_0}\frac{\omega_0^2}{c^2}\gamma\cdot\mathrm{Im}\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_\lambda)\cdot\gamma, \tag{38}$$

$$\delta = \frac{1}{\hbar\pi\varepsilon_0}\mathrm{PV}\int_0^\infty \mathrm{d}\omega_\lambda\frac{\omega_\lambda^2}{c^2}\frac{\gamma\cdot\mathrm{Im}\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_\lambda)\cdot\gamma}{(\omega_\lambda-\omega_0)}, \tag{39}$$

where Γ is the usual decay rate [56], and for vacuum, $\Gamma^{\mathrm{vac}} = \gamma^2\omega_0^3/\pi\varepsilon_0\hbar c^3$. Note that here we start with the Green function and obtain the normal-mode result, whereas in [51] they start with the normal modes and obtain the Green function (albeit for the lossy case).

4. EXAMPLE II: DRIVEN ATOM IN A STRUCTURED RESERVOIR—DENSITY OPERATOR ANALYSIS

As a second example, we consider an atom in a structured reservoir under the action of an external pump. The derivation follows the familiar route [57], and, for the LNA, details are available in [21]. The resulting Schrödinger picture master equation (ME) is, under the Born and Markov approximations,

$$\begin{aligned}\frac{d}{dt}\rho(t) = &-\frac{i}{\hbar}\left[\hat{H}_{\mathrm{S}},\rho(t)\right] - \int_0^t \mathrm{d}\tau\,\Big(J^{n+1}_{ph}(\tau)\hat{\sigma}_+\hat{\sigma}_-(-\tau)\rho(t)\\ &- J^{n+1}_{ph}(\tau)\hat{\sigma}_-(-\tau)\rho(t)\hat{\sigma}_+ - J^{n+1}_{ph}(-\tau)\hat{\sigma}_-\rho(t)\hat{\sigma}_+(-\tau)\\ &+ J^{n+1}_{ph}(-\tau)\rho(t)\hat{\sigma}_+(-\tau)\hat{\sigma}_- - J^{n}_{ph}(-\tau)\hat{\sigma}_-\hat{\sigma}_+(-\tau)\rho(t)\\ &+ J^{n}_{ph}(-\tau)\hat{\sigma}_+(-\tau)\rho(t)\hat{\sigma}_- + J^{n}_{ph}(\tau)\hat{\sigma}_+\rho(t)\hat{\sigma}_-(-\tau)\\ &- J^{n}_{ph}(\tau)\rho(t)\hat{\sigma}_-(-\tau)\hat{\sigma}_+,\end{aligned} \tag{40}$$

where $H_S = \hbar(\omega_d - \omega_L)\hat{\sigma}^+\hat{\sigma}^- + \frac{\hbar\Omega}{2}(\sigma^+ + \sigma^-)$. For the NMQED,

$$J_{ph}^{n+1}(\tau) = \sum_{\mathbf{k}} J_{\mathbf{k}}(\bar{n}(\omega_{\mathbf{k}}) + 1)e^{-i(\omega_{\mathbf{k}}-\omega_L)\tau}, \quad \textbf{(41)}$$

$$J_{ph}^{n}(\tau) = \sum_{\mathbf{k}} J_{\mathbf{k}}\bar{n}(\omega_{\mathbf{k}})e^{-i(\omega_{\mathbf{k}}-\omega_L)\tau}, \quad \textbf{(42)}$$

$$J_{\mathbf{k}} = \frac{\omega_{\mathbf{k}}}{2\hbar\varepsilon_0}\gamma \cdot \mathbf{E}_{\mathbf{k}}(\mathbf{r})\mathbf{E}_{\mathbf{k}}^*(\mathbf{r}) \cdot \gamma, \quad \textbf{(43)}$$

and for the LNA,

$$J_{ph}^{n+1}(\tau) = \int_0^\infty \mathrm{d}\omega J_{ph}(\omega)\,(\bar{n}(\omega) + 1)\,e^{-i(\omega-\omega_L)\tau}, \quad \textbf{(44)}$$

$$J_{ph}^{n}(\tau) = \int_0^\infty \mathrm{d}\omega J_{ph}(\omega)\bar{n}(\omega)e^{-i(\omega-\omega_L)\tau}, \quad \textbf{(45)}$$

$$J_{ph}(\omega) = \frac{\omega^2}{c^2}\frac{\gamma \cdot \mathrm{Im}\,(\mathbf{G}\,(\mathbf{r}, \mathbf{r}, \omega)) \cdot \gamma}{\pi\hbar\varepsilon_0}, \quad \textbf{(46)}$$

and, where $\bar{n}$ is the average number of thermal photons, $\bar{n} = (e^{\frac{\hbar\omega}{k_B T}} - 1)^{-1}$.

Using Eq. (15), it is easy to show that

$$\sum_{\mathbf{k}} J_{\mathbf{k}}e^{-i\omega_{\mathbf{k}}\tau} = \int_0^\infty \mathrm{d}\omega J(\omega)e^{-i\omega\tau}, \quad \textbf{(47)}$$

and, thus,

$$\frac{d\rho(t)^{\mathrm{LNA}}}{dt} \doteq \frac{d\rho(t)^{\mathrm{NMQED}}}{dt}, \quad \textbf{(48)}$$

and the system evolution is the same for both approaches.

As a special case, if we set $\bar{n} = 0$ and turn off the pump, $H_S = \hbar\omega_d\hat{\sigma}^+\hat{\sigma}^-$, in which case $\hat{\sigma}_\mp(-\tau) = \hat{\sigma}_\mp e^{\pm i\omega_d\tau}$, we obtain the familiar ME for a single atom interacting with its environment,

$$\frac{d}{dt}\rho = -i(\omega_d - \Delta_d)[\hat{\sigma}^+\hat{\sigma}^-, \rho(t)]$$
$$+\frac{\gamma(\omega_d)}{2}(2\hat{\sigma}_-\rho(t)\hat{\sigma}_+ - \hat{\sigma}_+\hat{\sigma}_-\rho(t) - \rho(t)\hat{\sigma}_+\hat{\sigma}_-), \quad \textbf{(49)}$$

where we used the SP identity and where $\gamma(\omega_d) = 2\pi J(\omega_d)$, $\Delta_d = \frac{1}{\hbar^2}\,\mathrm{PV}\int_0^\infty \mathrm{d}\omega J(\omega)/(\omega - \omega_d)$. The ME for a multi-atom system, allowing for, e.g., the study of entanglement, is also the same for the NMQED and LNAs.

5. COMMENTS ON THE CONNECTION BETWEEN NORMAL-MODE QED AND LANGEVIN NOISE APPROACHES, AND VALIDITY OF THE LANGEVIN NOISE APPROACH

NMQED is well-founded mathematically, based on canonical quantization and completeness of the eigenfunctions of SA operators [58,59]. Much of quantum optics is based on electric field operators of the forms of Eqs. (4) and (5) using plane wave eigenfunctions [Eq. (10)] (including microscopic models). As more complicated environments have been considered, the eigenfunctions based on Eq. (1) have been used. However, all of the aforementioned eigenfunctions only form complete sets in limited settings (closed cavities, usually lossless, dispersionless materials), where material parameters are represented by Hermitian (SA) tensors. Note that completeness is important, not only for Eq. (15), but also for validity of Eqs. (4) and (5), which are also eigenfunction expansions.

Two comments are important: (1) Some level of loss must be maintained in the system when using Eqs. (17) and (18); it is impermissible to let $\mathrm{Im}(\varepsilon(\mathbf{r}, \omega_\lambda)) \to 0$ until after that term drops out from the formulation, typically after using Eq. (23) or Eq. (24). One cannot take this limit in Eqs. (17) and (18). (2) If in Fig. 1 $\varepsilon_{\mathrm{bulk}}$ is lossless, then it is also impermissible to let the size of the region of interest shrink to zero to implement the vacuum limit (i.e., $\Omega_1 \to 0$ in Fig. 1), until after using Eq. (23) or Eq. (24), after which the Green function is merely the vacuum Green function for the cavity or open space (if $\varepsilon_{\mathrm{bulk}}$ is lossy, than one can allow the limit $\Omega_1 \to 0$ at the onset). In the presented examples, using Eq. (15), the LNA reduces to the NMQED result for closed cavities; alternatively, using Eq. (15), the NMQED result can be generalized to involve the Green function, allowing cavities with lossy, dispersive materials to be considered, and even open geometries. However, this is not a general result (i.e., this does not universally hold).

In a practical sense, lossless materials do not exist, aside from vacuum. Therefore, it is not unreasonable to consider space to be filled with a background medium having perhaps $\mathrm{Re}(\varepsilon) \simeq 1$ and $\mathrm{Im}(\varepsilon) > 0$, into which the actual structure of interest is placed, as depicted in Fig. 1. The Green function accounts for the entire permittivity $\varepsilon(\mathbf{r}, \omega)$, including the background, and after $\mathrm{Im}(\varepsilon(\mathbf{r}, \omega))$ is removed from the formulation using Eqs. (23) and (24) and only the Green function remains, one can consider lossless materials.

A. Lossless Limit of the "Magic Formula": Eq. (23)

The connection between the NMQED and the LNA is established by virtue of the conversion formula [Eq. (15)]—showing that NMQED is a special case of the LNA in the lossless limit. However, the explicit presence of the factor $\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}, \omega))}$ in the field expansion [Eq. (18)] indicates that this limit has to be understood in a strict sense as a mathematical limiting procedure, where $\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}, \omega))} \to 0$ while $\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}, \omega))} > 0$. In fact, the presence of $\sqrt{\mathrm{Im}(\varepsilon(\mathbf{r}, \omega))}$ in the field explansion is an artifact of normalizing the bosonic canonically conjugate field variables and is avoided if one instead works with the noise polarization.

In either case, after evaluating operator dynamics or taking quantum expectation values, one typically arrives at the left-hand side of the integral relation [Eq. (23)]. The right-hand side of this formula is obviously finite in the above-defined lossless limit $\mathrm{Im}\epsilon(\mathbf{r}, \omega) \to 0+$. At first glance, the left-hand side seems to vanish in this limit due to the presence of the factor $\mathrm{Im}(\varepsilon(\mathbf{r}', \omega))$. However, this conclusion is premature as a careful evaluation of the spatial integral will reveal a factor canceling $\mathrm{Im}(\varepsilon(\mathbf{r}', \omega))$, so that the limit may be taken to give the same result as the right-hand side of the equation.

To illustrate this, consider the case of a bulk medium with permittivity $\varepsilon(\omega) = \varepsilon_{\mathrm{R}}(\omega) + \mathrm{i}\delta$ with ε_{R} real. The respective Green tensor is given by

$$\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)=-\frac{1}{3k^2}\boldsymbol{\delta}(\boldsymbol{\rho})-\frac{\mathrm{e}^{\mathrm{i}k\rho}}{4\pi k^2\rho^3}$$
$$\times\{[1-\mathrm{i}k\rho-(k\rho)^2]\mathbf{I}-[3-3\mathrm{i}k\rho-(k\rho)^2]\boldsymbol{e}_\rho\boldsymbol{e}_\rho\}, \tag{50}$$

with $\boldsymbol{\rho}=\mathbf{r}-\mathbf{r}'$, $\rho=|\boldsymbol{\rho}|$, $\boldsymbol{e}_\rho=\boldsymbol{\rho}/\rho$, and $k=\sqrt{\varepsilon(\omega)}\omega/c$ such that $\mathrm{Im}k>0$. In the limit $\mathbf{r}_0\to\mathbf{r}$, $\mathbf{r}_0\neq\mathbf{r}$, we hence have

$$\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{G}^{(0)\dagger}(\mathbf{r}_0,\mathbf{r}',\omega)\propto\mathrm{e}^{-2\delta\rho}. \tag{51}$$

To leading order in δ, this implies

$$\int\mathrm{d}^3\mathrm{r}'\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{G}^{(0)\dagger}(\mathbf{r}_0,\mathbf{r}',\omega)=\mathrm{O}(1/\delta), \tag{52}$$

so that

$$\int\mathrm{d}^3\mathrm{r}'\mathrm{Im}\varepsilon(\omega)\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{G}^{(0)\dagger}(\mathbf{r}_0,\mathbf{r}',\omega) \tag{53}$$

remains finite in the limit $\mathrm{Im}\varepsilon(\omega)\equiv\delta\to0+$. In Appendix A, we explicitly demonstrate the validity of the integral relation [Eq. (23)] in the lossless limit for the more general case of arbitrary $\mathbf{r}$, $\mathbf{r}_0$.

An alternative way to establish contact with the nonabsorbing case was suggested in [28]. Here, the region of interest is surrounded by a strictly lossless region $\mathrm{Im}\varepsilon(\omega)=0$ at infinity (or sufficiently far, respectively). It was shown that under such conditions the integral relation [Eq. (23)] has an additional term,

$$\frac{\omega^2}{c^2}\int_\Omega\mathrm{d}^3\mathbf{r}'\mathrm{Im}\left(\varepsilon\left(\mathbf{r}',\omega\right)\right)\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{G}^\dagger(\mathbf{r}_0,\mathbf{r}',\omega)$$
$$+\oint_\Sigma d^2\mathbf{r}'\mathbf{F}\left(\mathbf{r}',\mathbf{r},\mathbf{r}_0\right)=\mathrm{Im}\mathbf{G}(\mathbf{r},\mathbf{r}_0,\omega), \tag{54}$$

where

$$\oint_\Sigma\mathrm{d}^2\mathbf{r}'\mathbf{F}\left(\mathbf{r}',\mathbf{r},\mathbf{r}_0\right)=\frac{\omega}{c}\sqrt{\varepsilon_{\mathrm{bulk}}}\oint_\Sigma\mathrm{d}^2\mathbf{r}'\mathbf{G}^{\mathrm{T}}\left(\mathbf{r}',\mathbf{r}\right)$$
$$\cdot\mathbf{R}\times\mathbf{R}\times\mathbf{G}^*\left(\mathbf{r}',\mathbf{r}_0\right), \tag{55}$$

and Σ is the bounding surface that is far from the system in question. In the event of an absorbing (perhaps limitingly low-loss) background medium $\varepsilon_{\mathrm{bulk}}$, the Green tensor vanishes on Σ, and the surface contribution vanishes accordingly. This is commensurate with the requirement $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\to\mathbf{0}$ for $|\mathbf{r}-\mathbf{r}'|\to\infty$. Thus, one must retain material absorption of the background environment if Eq. (23) or Eq. (24) is to be used, to ensure that no boundry contribution arises. Physically, one could argue that the assumption of a background environment without at least some small amount of absorption is generally a fiction anyway, aside from perhaps evacuated superconducting chambers. Alternatively, in [28], it is shown that by implementing the developed scheme of replacing the missing free incident field with polarization currents at infinity with a lossless interior region, to bring the LNA into accordance with the Huttner–Barnett result, and by including the boundary term one recovers the usual LNA.

B. Commutation Relations of Normal-Mode Operators in the Lossless Limit

Even in the lossy case where $\mathrm{Im}[\epsilon(\omega)]>0$ one can formally define normal-mode creation and annihilation operators via Eqs. (4) and (5). These operators can be expressed in terms of the polaritonic creation and annihilation operators $\hat{\mathbf{f}}^{(\dagger)}$ via the Green's function but will in general not obey bosonic commutation relations. In the lossless limit, however, the known commutation relations from NMQED are reproduced as is shown in the following. This shows that the algebra of these operators in the lossless limit is equivalent in the normal-mode and the Langevin noise quantization schemes, underpinning the equivalence of the two approaches in the lossless limit.

By inverting Eqs. (4) and (5) and using Eq. (10), one obtains the normal-mode creation and annihilation operators in terms of the positive ($\hat{\boldsymbol{E}}^{(+)}$) and negative ($\hat{\boldsymbol{E}}^{(-)}$) frequency components of the electric field operator,

$$\hat{a}_{\mathbf{k}s}=-\mathrm{i}\sqrt{\frac{\epsilon_0}{4\pi^3\hbar\omega_{\mathbf{k}}}}\int\mathrm{d}^3r\,\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\mathbf{e}_{\mathbf{k}s}\cdot\hat{\boldsymbol{E}}^{(+)}(\mathbf{r}). \tag{56}$$

Here we have used $\hat{\boldsymbol{E}}^{(+)\dagger}=\hat{\boldsymbol{E}}^{(-)}$. Inserting the expression for the electric field operator in the LNA by means of Eq. (18) we find the normal-mode annihilation operators in terms of the polaritonic ones,

$$\hat{a}_{\mathbf{k}s}=\sqrt{\frac{1}{4\pi^4\omega_{\mathbf{k}}}}\int_0^\infty\mathrm{d}\omega_\lambda\frac{\omega_\lambda^2}{c^2}\int\mathrm{d}^3r\int\mathrm{d}^3r'\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}$$
$$\times\mathbf{e}_{\mathbf{k}s}\cdot\mathbf{G}\left(\mathbf{r},\mathbf{r}',\omega_\lambda\right)\cdot\sqrt{\mathrm{Im}\left(\varepsilon\left(\mathbf{r}',\omega_\lambda\right)\right)}\cdot\hat{\mathbf{f}}\left(\mathbf{r}',\omega_\lambda\right). \tag{57}$$

Note in general that the normal-mode creation and annihilation operators formally defined here via Eq. (56) are different from the photon creation and annihilation operators in a general, lossy optical environment. The latter have been shown to be linear combinations of creation *and* annihilation operators of the polaritonic field operators [4,60]. Since we are here only interested in showing that the correct commutation relations are reproduced in the lossless case, we stick with the formal definition of the normal-mode creation and annihilation operators in Eq. (56) although their physical interpretation in the lossy case is not straightforward.

Using the commutation relations of the polaritonic creation and annihilation operators in Eqs. (19) and (20) as well as the "magic formula" [Eq. (23)], one can obtain the commutation relations of the normal-mode operators in case of lossy material: $[\hat{a}_{\mathbf{k}s},\hat{a}_{\mathbf{k}'s'}]=[\hat{a}^\dagger_{\mathbf{k}s},\hat{a}^\dagger_{\mathbf{k}'s'}]=0$ and

$$[\hat{a}_{\mathbf{k}s},\hat{a}^\dagger_{\mathbf{k}'s'}]=\frac{\epsilon_0\mu_0}{8\pi^4\sqrt{\omega_{\mathbf{k}}\omega_{\mathbf{k}'}}}\int\mathrm{d}^3r\int\mathrm{d}^3r'\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\mathrm{e}^{-\mathrm{i}\mathbf{k}'\cdot\mathbf{r}'}$$
$$\times\mathbf{e}_{\mathbf{k}s}\cdot\int_0^\infty\mathrm{d}\omega_\lambda\omega_\lambda^2\mathrm{Im}[\mathbf{G}(\mathbf{r},\mathbf{r}',\omega_\lambda)]\cdot\mathbf{e}^*_{\mathbf{k}'s'}. \tag{58}$$

For $\mathrm{Im}[\epsilon(\omega)]>0$, Eq. (58) will in general not be given by $\delta_{\mathbf{k}\mathbf{k}'}\delta_{ss'}$ as it is the case in NMQED, Eq. (6). This can be seen, e.g., from the fact that the exponentials in the bulk Green function [cf. Eq. (50)] are not purely oscillating but have a decaying factor $e^{-\mathrm{Im}(\sqrt{\epsilon})\omega\rho/c}$.

Nevertheless, in the lossless limit, the commutation relations as found in Eq. (6) using NMQED are reproduced. This can be seen from Eq. (58) by evaluating the frequency integral using Eq. (15) such that

$$[\hat{a}_{\mathbf{k}s}, \hat{a}^{\dagger}_{\mathbf{k}'s'}] = \frac{1}{16\pi^3\sqrt{\omega_{\mathbf{k}}\omega_{\mathbf{k}'}}}\sum_{s''}\sum_{\mathbf{k}''}\omega_{\mathbf{k}''}\int \mathrm{d}^3r \int \mathrm{d}^3r' \times \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\mathbf{e}_{\mathbf{k}s}\cdot\hat{\boldsymbol{E}}_{\mathbf{k}''s''}(\mathbf{r},\omega_{\mathbf{k}''})\mathrm{e}^{-\mathrm{i}\mathbf{k}'\cdot\mathbf{r}'}\mathbf{e}^*_{\mathbf{k}'s'}\cdot\hat{\boldsymbol{E}}^*_{\mathbf{k}''s''}(\mathbf{r}',\omega_{\mathbf{k}''}). \tag{59}$$

In a last step, we use Eqs. (10) and (11) to carry out the spatial integrals and obtain

$$[\hat{a}_{\mathbf{k}s}, \hat{a}^{\dagger}_{\mathbf{k}'s'}] = \delta_{\mathbf{k}\mathbf{k}'}\delta_{ss'}, \tag{60}$$

as desired.

6. CONCLUSION

The LNA for quantization of macroscopic electromagnetics for 3D, inhomogeneous environments has been compared with the usual normal-mode quantization in quantum optics. The conditions of validity of the normal-mode expansion were discussed, and it was shown using several examples that the LNA reduces exactly to the normal-mode expansion formulation in the lossless limit. Conditions on applying the LNA to finite structures were also discussed.

APPENDIX A: LOSSLESS LIMIT FOR THE BULK CASE

In this appendix, we explicitly show that the "magic formula" in Eq. (23) holds also in the limit of lossless media for the case of a single bulk dielectric material described by $\epsilon(\mathbf{r},\omega) = \epsilon(\omega)$. This means we show that

$$\lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\frac{\omega^2}{c^2}\int \mathrm{d}^3\mathrm{r}'\mathrm{Im}\left(\varepsilon\left(\mathbf{r}',\omega\right)\right) \times \mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{G}^{(0)*}(\mathbf{r}',\mathbf{r}_0,\omega) = \lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\mathrm{Im}\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}_0,\omega). \tag{A1}$$

Here, $\mathbf{G}^{(0)}$ is the bulk Green tensor. Note that compared to Eq. (23), we have already used that the Green's tensor for bulk isotropic dielectric material obeys Onsager reciprocity, i.e., $G^{(0)}_{ij}(\mathbf{r},\mathbf{r}') = G^{(0)}_{ji}(\mathbf{r}',\mathbf{r})$. We will show that Eq. (A1) holds by using the bulk Green tensor $\mathbf{G}^{(0)}$ in its $(2+1)$-dimensional decomposition [17],

$$\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}_0,\omega) = -\frac{1}{k^2}\delta^3(\mathbf{r}-\mathbf{r}_0)\boldsymbol{e}_z\boldsymbol{e}_z + \frac{i}{8\pi^2}\int \mathrm{d}^2k_\parallel \frac{\mathrm{e}^{i\boldsymbol{k}_\parallel\cdot(\mathbf{r}-\mathbf{r}_0)}}{k^\perp} \times \sum_{\sigma=s,p}[\boldsymbol{e}_{\sigma+}\boldsymbol{e}_{\sigma+}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0) + \boldsymbol{e}_{\sigma-}\boldsymbol{e}_{\sigma-}\mathrm{e}^{-ik^\perp(z-z_0)}\theta(z_0-z)]. \tag{A2}$$

Here, $k^\perp = \sqrt{k^2 - k_\parallel^2}$ and $k = \sqrt{\epsilon(\omega)}\omega/c$, and we have defined the polarization vectors,

$$\boldsymbol{e}_{p\pm} = \frac{1}{k_\parallel}\begin{pmatrix} k_y \\ -k_x \\ 0 \end{pmatrix}, \quad \boldsymbol{e}_{p\pm} = \frac{1}{k}\begin{pmatrix} k^\perp k_x/k_\parallel \\ k^\perp k_y/k_\parallel \\ k_\parallel \end{pmatrix}. \tag{A3}$$

Inserting the first term of the Green's tensor in Eq. (A2) into the left-hand side of Eq. (A1), one obtains

$$\lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\frac{1}{|k|^4}\mathrm{Im}[\epsilon(\omega)]\frac{\omega^2}{c^2}\int \mathrm{d}^3r'\delta^3(\mathbf{r}-\mathbf{r}')\delta^3(\mathbf{r}'-\mathbf{r}_0) = \lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\frac{1}{|k|^4}\mathrm{Im}[\epsilon(\omega)]\frac{\omega^2}{c^2}\delta^3(\mathbf{r}-\mathbf{r}_0) = 0. \tag{A4}$$

For the terms of the left-hand side of Eq. (A1) consisting of the product of a first and a second term of the bulk Green's tensor in Eq. (A2), one finds

$$\lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\mathrm{Im}[\epsilon(\omega)]\frac{\omega^2}{c^2}\frac{\mathrm{i}}{8\pi^2|k|^2}\int \mathrm{d}^2k_\parallel \mathrm{e}^{\mathrm{i}\boldsymbol{k}_\parallel(\mathbf{r}-\mathbf{r}_0)}k_\parallel \times\left\{\theta(z-z_0)\left[\frac{\boldsymbol{e}_z\boldsymbol{e}^*_{p-}\mathrm{e}^{-\mathrm{i}k^{\perp *}(z-z_0)}}{kk^{\perp *}} - \boldsymbol{e}_{p+}\boldsymbol{e}_z\frac{\mathrm{e}^{\mathrm{i}k^\perp(z-z_0)}}{k^*k^\perp}\right] + \theta(z_0-z)\left[\frac{\boldsymbol{e}_z\boldsymbol{e}^*_{p+}\mathrm{e}^{\mathrm{i}k^{\perp *}(z-z_0)}}{kk^{\perp *}} - \boldsymbol{e}_{p-}\boldsymbol{e}_z\frac{\mathrm{e}^{-\mathrm{i}k^\perp(z-z_0)}}{k^*k^\perp}\right]\right\}. \tag{A5}$$

This term again vanishes in the limit of $\mathrm{Im}[\epsilon(\omega)]\to 0$.

Hence, we are left with the terms stemming from the second term of the Green's tensor in Eq. (A2) only. Inserting the second and third rows of Eq. (A2) into the left-hand side of Eq. (A1), one finds

$$\lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\mathrm{Im}[\epsilon(\omega)]\frac{\omega^2}{c^2}\frac{1}{16\pi^2}\int_{-\infty}^{\infty}\mathrm{d}z'\int \mathrm{d}^2k_\parallel\frac{\mathrm{e}^{\mathrm{i}\boldsymbol{k}_\parallel\cdot(\mathbf{r}-\mathbf{r}_0)}}{|k^\perp|^2} \times\sum_\sigma\left[\boldsymbol{e}_{\sigma+}\boldsymbol{e}_{\sigma+}\mathrm{e}^{ik^\perp(z-z')}\theta(z-z') + \boldsymbol{e}_{\sigma-}\boldsymbol{e}_{\sigma-}\mathrm{e}^{-ik^\perp(z-z')}\theta(z'-z)\right] \cdot\left[\boldsymbol{e}^*_{\sigma-}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^{\perp *}(z'-z_0)}\theta(z'-z_0) + \boldsymbol{e}^*_{\sigma+}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{ik^{\perp *}(z'-z_0)}\theta(z_0-z')\right]. \tag{A6}$$

Here, we carried out the $\mathbf{r}'_\parallel$ integral leading to a factor $\delta^2(\boldsymbol{k}_\parallel + \boldsymbol{k}'_\parallel)$, which in turn has been used to perform the $k'_\parallel$ integral. Finally, we also used

$$\boldsymbol{e}_{\sigma\pm}(-k_\parallel)\boldsymbol{e}_{\sigma\pm}(-k_\parallel) = \boldsymbol{e}_{\sigma\mp}(k_\parallel)\boldsymbol{e}_{\sigma\mp}(k_\parallel), \tag{A7}$$

$$\boldsymbol{e}_{\sigma\pm}\cdot\boldsymbol{e}^*_{\sigma'\pm} = \delta_{\sigma\sigma'}, \tag{A8}$$

$$\boldsymbol{e}_{\sigma\pm}\cdot\boldsymbol{e}^*_{\sigma'\mp} \propto \delta_{\sigma\sigma'}. \tag{A9}$$

The remaining z' integral can be carried out straightforwardly, and some lengthy algebra shows that Eq. (A6) can be further reduced to

$$\lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\mathrm{Im}[\epsilon(\omega)]\frac{\omega^2}{c^2}\frac{1}{16\pi^2}\int \mathrm{d}^2k_\parallel \frac{\mathrm{e}^{\mathrm{i}\boldsymbol{k}_\parallel\cdot(\mathbf{r}-\mathbf{r}_0)}}{|k^\perp|^2}\sum_\sigma\Bigg\{\frac{1}{2\mathrm{Im}[k^\perp]}\Big[\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^\perp(z-z_0)}\theta(z_0-z)$$
$$+\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^{\perp*}(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{+ik^{\perp*}(z-z_0)}\theta(z_0-z)\Big]-\frac{\mathrm{i}\boldsymbol{e}_{\sigma-}\cdot\boldsymbol{e}^*_{\sigma+}}{2\mathrm{Re}[k^\perp]}\Big[\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{ik^\perp(z_0-z)}\theta(z_0-z)$$
$$-\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^{\perp*}(z-z_0)}\theta(z-z_0)-\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{-ik^{\perp*}(z_0-z)}\theta(z_0-z)\Big]\Bigg\}. \tag{A10}$$

To derive Eq. (A10), we also used $\boldsymbol{e}_{\sigma-}\cdot\boldsymbol{e}^*_{\sigma+}=\boldsymbol{e}_{\sigma+}\cdot\boldsymbol{e}^*_{\sigma-}$. Next, we rewrite

$$\frac{1}{\mathrm{Im}[k^\perp]}=\frac{|k^\perp|^2}{\mathrm{Im}[k^\perp]\mathrm{Re}[k^\perp]}\mathrm{Re}\left[\frac{1}{k^\perp}\right]=\frac{2|k^\perp|^2c^2}{\mathrm{Im}[\epsilon(\omega)]\omega^2}\mathrm{Re}\left[\frac{1}{k^\perp}\right], \tag{A11}$$

$$\frac{1}{\mathrm{Re}[k^\perp]}=-\frac{|k^\perp|^2}{\mathrm{Re}[k^\perp]\mathrm{Im}[k^\perp]}\mathrm{Im}\left[\frac{1}{k^\perp}\right]=-\frac{2|k^\perp|^2c^2}{\mathrm{Im}[\epsilon(\omega)]\omega^2}\mathrm{Im}\left[\frac{1}{k^\perp}\right], \tag{A12}$$

in order to find that Eq. (A10) is equivalent to

$$\lim_{\epsilon(\omega)=1+\mathrm{i}\delta\to 1}\frac{1}{8\pi^2}\int \mathrm{d}^2k_\parallel \mathrm{e}^{\mathrm{i}\boldsymbol{k}_\parallel\cdot(\mathbf{r}-\mathbf{r}_0)}\times\sum_\sigma\Bigg\{\mathrm{Re}\left[\frac{1}{k^\perp}\right]\frac{1}{2}\Big[\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^\perp(z-z_0)}\theta(z_0-z)$$
$$+\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^{\perp*}(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{+ik^{\perp*}(z-z_0)}\theta(z_0-z)\Big]-\boldsymbol{e}_{\sigma-}\cdot\boldsymbol{e}^*_{\sigma+}\mathrm{Im}\left[\frac{1}{k^\perp}\right]\frac{1}{2\mathrm{i}}\Big[\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{ik^\perp(z_0-z)}\theta(z_0-z)$$
$$-\boldsymbol{e}_{\sigma+}\boldsymbol{e}^*_{\sigma-}\mathrm{e}^{-ik^{\perp*}(z-z_0)}\theta(z-z_0)-\boldsymbol{e}_{\sigma-}\boldsymbol{e}^*_{\sigma+}\mathrm{e}^{-ik^{\perp*}(z_0-z)}\theta(z_0-z)\Big]\Bigg\}. \tag{A13}$$

This was the crucial step, since the factor $\mathrm{Im}[\epsilon(\omega)]$ was canceled, meaning that now we are ready to take the limit $\mathrm{Im}[\epsilon(\omega)]\to 0$. In this limit, we find that $k\in\mathbb{R}$, which also leads to the fact that $k^\perp=\sqrt{k^2-k_\parallel^2}$ is either real or purely imaginary depending on whether $k_\parallel<k$ or $k_\parallel>k$, respectively. This way, we find that in the second and third row of Eq. (A13), we can use

$$\boldsymbol{e}^*_{\sigma\pm}=\boldsymbol{e}_{\sigma\pm}\quad \text{if}\quad k^\perp,k\in\mathbb{R}, \tag{A14}$$

whereas in the third and fourth row of Eq. (A14), we have

$$\boldsymbol{e}^*_{\sigma\pm}=\boldsymbol{e}_{\sigma\mp}\quad \text{if}\quad k^\perp\in\mathrm{i}\mathbb{R},\ k\in\mathbb{R}. \tag{A15}$$

Since $\boldsymbol{e}_{\sigma\pm}\cdot\boldsymbol{e}_{\sigma'\pm}=\delta_{\sigma\sigma'}$, and using Eq. (A7) again, we finally find that Eq. (A13) can be further reduced to

$$\frac{1}{8\pi^2}\int \mathrm{d}^2k_\parallel \mathrm{e}^{i\boldsymbol{k}_\parallel\cdot(\mathbf{r}-\mathbf{r}_0)}\sum_{\sigma=s,p}\Bigg\{\mathrm{Re}\left[\frac{1}{k^\perp}\right]\frac{1}{2}[\boldsymbol{e}_{\sigma+}\boldsymbol{e}_{\sigma+}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma-}\boldsymbol{e}_{\sigma-}\mathrm{e}^{-ik^\perp(z-z_0)}\theta(z_0-z)+\mathrm{c.c.}(k_\parallel\to -k_\parallel)]$$
$$-\mathrm{Im}\left[\frac{1}{k^\perp}\right]\frac{1}{2i}[\boldsymbol{e}_{\sigma+}\boldsymbol{e}_{\sigma+}\mathrm{e}^{ik^\perp(z-z_0)}\theta(z-z_0)+\boldsymbol{e}_{\sigma-}\boldsymbol{e}_{\sigma-}\mathrm{e}^{-ik^\perp(z-z_0)}\theta(z_0-z)-\mathrm{c.c.}(k_\parallel\to -k_\parallel)]\Bigg\}. \tag{A16}$$

Here, $\mathrm{c.c.}(k_\parallel\to -k_\parallel)$ denotes adding the complex conjugate of the preceding term, which has also been subject to the replacement $k_\parallel\to -k_\parallel$. Equation (A16) is equivalent to $\lim_{\mathrm{Im}[\epsilon(\omega)]\to 0+}\mathrm{Im}\mathbf{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)$ [cf. Eq. (A2)] as desired.

Funding. German Research Foundation (BU 1803/3-1); Studienstiftung des deutschen Volkes.

Acknowledgment. The authors gratefully acknowledge discussions with Stephen Hughes. F. L. thanks the Studienstiftung des deutschen Volkes for financial support.

Disclosures. The authors declare no conflicts of interest.

REFERENCES AND NOTE

1. E. M. Purcell, H. C. Torrey, and R. V. Pound, "Resonance absorption by nuclear magnetic moments in a solid," Phys. Rev. **69**, 37–39 (1946).
2. R. J. Glauber and M. Lewenstein, "Quantum optics of dielectric media," Phys. Rev. A **43**, 467–491 (1991).
3. J. D. Jackson, *Classical Electrodynamics*, 3rd ed. (Wiley, 1999).
4. B. Huttner and S. M. Barnett, "Quantization of the electromagnetic field in dielectrics," Phys. Rev. A **46**, 4306–4322 (1992).
5. R. Matloob, R. Loudon, S. M. Barnett, and J. Jeffers, "Electromagnetic field quantization in absorbing dielectrics," Phys. Rev. A **52**, 4823–4838 (1995).
6. T. Gruner and D.-G. Welsch, "Green-function approach to the radiation-field quantization for homogeneous and inhomogeneous Kramers-Kronig dielectrics," Phys. Rev. A **53**, 1818–1829 (1996).
7. H. T. Dung, L. Knöll, and D.-G. Welsch, "Three-dimensional quantization of the electromagnetic field in dispersive and absorbing inhomogeneous dielectrics," Phys. Rev. A **57**, 3931–3942 (1998).
8. H. T. Dung, L. Knöll, and D.-G. Welsch, "Spontaneous decay in the presence of dispersing and absorbing bodies: general theory and application to a spherical cavity," Phys. Rev. A **62**, 053804 (2000).
9. M. Wubs and L. G. Suttorp, "Transient QED effects in absorbing dielectrics," Phys. Rev. A **63**, 043809 (2001).
10. L. Knöll, S. Scheel, and D.-G. Welsch, "QED in dispersing and absorbing media," in *Coherence and Statistics of Photons and Atoms*, J. Perina, ed. (Wiley-VCH, 2001).
11. O. Di Stefano, S. Savasta, and R. Girlanda, "Microscopic calculation of noise current operators for electromagnetic field quantization in absorbing material systems," J. Opt. B **3**, 288–292 (2001).
12. L. G. Suttorp and M. Wubs, "Field quantization in inhomogeneous absorptive dielectrics," Phys. Rev. A **70**, 013816 (2004).
13. L. G. Suttorp and A. J. van Wonderen, "Fano diagonalization of a polariton model for an inhomogeneous absorptive dielectric," Europhys. Lett. **67**, 766 (2004).
14. R. Matloob, "Electromagnetic field quantization in a linear isotropic dielectric," Phys. Rev. A **69**, 052110 (2004).
15. R. Matloob, "Electromagnetic field quantization in a linear isotropic permeable dielectric medium," Phys. Rev. A **70**, 022108 (2004).
16. S. Y. Buhmann, D. T. Butcher, and S. Scheel, "Macroscopic quantum electrodynamics in nonlocal and nonreciprocal media," New J. Phys. **14**, 083034 (2012).
17. S. Y. Buhmann, *Dispersion Forces I*, Springer Tracts in Modern Physics (2012), Vol. **247**.
18. S. Y. Buhmann, *Dispersion Forces II*, Springer Tracts in Modern Physics (2012), Vol. **248**.
19. C. Raabe, S. Scheel, and D.-G. Welsch, "Unified approach to QED in arbitrary linear media," Phys. Rev. A **75**, 053813 (2007).
20. S. A. R. Horsley and T. G. Philbin, "Canonical quantization of electromagnetism in spatially dispersive media," New J. Phys. **16**, 013030 (2014).
21. S. A. Hassani Gangaraj, G. W. Hanson, and M. Antezza, "Robust entanglement with three-dimensional nonreciprocal photonic topological insulators," Phys. Rev. A **95**, 063807 (2017).
22. G. W. Hanson, S. A. Hassani Gangaraj, M. G. Silveirinha, M. Antezza, and F. Monticone, "Non-Markovian transient Casimir-Polder force and population dynamics on excited and ground state atoms: weak and strong coupling regimes in generally non-reciprocal environments," Phys. Rev. A **99**, 042508 (2019).
23. C. P. Van Vlack, "Dyadic Green functions and their applications in classical and quantum nanophotonics," Ph.D. Dissertation (Queen's University, 2012).
24. T. G. Philbin, "Canonical quantization of macroscopic electromagnetism," New J. Phys. **12**, 123008 (2010).
25. S. A. R. Horsley, "Canonical quantization of the electromagnetic field interacting with a moving dielectric medium," Phys. Rev. A **86**, 023830 (2012).
26. A. Tip, "Linear absorptive dielectrics," Phys. Rev. A **57**, 4818 (1998).
27. A. Tip, L. Knöll, S. Scheel, and D.-G. Welsch, "Equivalence of Langevin and auxiliary-field quantization methods for absorbing dielectrics," Phys. Rev. A **63**, 043806 (2001).
28. A. Drezet, "Equivalence between the Hamiltonian and Langevin noise descriptions of plasmon polaritons in a dispersive and lossy inhomogeneous medium," Phys. Rev. A **96**, 033849 (2017).
29. A. Drezet, "Dual-Lagrangian description adapted to quantum optics in dispersive and dissipative dielectric media," Phys. Rev. A **94**, 053826 (2016).
30. A. Drezet, "Quantizing polaritons in inhomogeneous dissipative systems," Phys. Rev. A **95**, 023831 (2017).
31. J. J. Hopfield, "Theory of the contribution of excitons to the complex dielectric constant of crystals," Phys. Rev. **112**, 1555–1567 (1958).
32. M. Amooshahi, "Canonical quantization of electromagnetic field in an anisotropic polarizable and magnetizable medium," J. Math. Phys. **50**, 062301 (2009).
33. F. Kheirandish and M. Amooshahi, "Electromagnetic field quantization in a linear polarizable and magnetizable medium," Phys. Rev. A **74**, 042102 (2006).
34. M. Amooshahi and F. Kheirandish, "Electromagnetic field quantization in a magnetodielectric medium with external charges," Phys. Rev. A **76**, 062103 (2007).
35. W. E. I. Sha, A. Y. Liu, and W. C. Chew, "Dissipative quantum electromagnetics," IEEE J. Multiscale Multiphys. Comput. Technol. **3**, 198–213 (2018).
36. O. Di Stefano, S. Savasta, and R. Girlanda, "Mode expansion and photon operators in dispersive and absorbing dielectrics," J. Mod. Opt. **48**, 67–84 (2001).
37. V. Dorier, J. Lampart, S. Guérin, and H. R. Jauslin, "Canonical quantization for quantum plasmonics with finite nanostructures," Phys. Rev. A **100**, 042111 (2019).
38. V. Dorier, S. Guérin, and H. R. Jauslin, "Critical review of quantum plasmonic models for finite-size media," arXiv:1911.03134v1 (2019).
39. R. Trivedi, K. Fischer, S. Fan, and J. Vuckovic, "Few-particle scattering from localized quantum systems in spatially structured bosonic baths," arXiv:2009.06860v1 (2020).
40. 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," Phys. Rev. Lett. **122**, 213901 (2019).
41. S. Hughes, S. Franke, C. Gustin, M. K. Dezfouli, A. Knorr, and M. Richter, "Theory and limits of on-demand single-photon sources using plasmonic resonators: a quantized quasinormal mode approach," ACS Photon. **6**, 2168–2180 (2019).
42. A. I. Fernández-Domínguez, S. I. Bozhevolnyi, and N. A. Mortensen, "Plasmon-enhanced generation of nonclassical light," ACS Photon. **5**, 3447–3451 (2018).
43. P. W. Milonni, *An Introduction to Quantum Optics and Quantum Fluctuations* (Oxford University, 2019).
44. P. Milonni, *The Quantum Vacuum* (Academic, 1994).
45. J. Garrison and R. Chiao, *Quantum Optics* (Oxford University, 2008).
46. C. Gerry and P. Knight, *Introductory Quantum Optics* (Cambridge University, 2006).
47. G. Grynberg, A. Aspect, and C. Fabrw, *Introduction to Quantum Optics* (Cambridge University, 2010).
48. Z.-Y. Li, L.-L. Lin, and Z.-Q. Zhang, "Spontaneous emission from photonic crystals: full vectorial calculations," Phys. Rev. Lett. **84**, 4341–4344 (2000).
49. K. W. Chan, C. K. Law, and J. H. Eberly, "Quantum entanglement in photon-atom scattering," Phys. Rev. A **68**, 022110 (2003).
50. J. Van Bladel, "Field expansions in cavities containing gyrotropic media," IRE Trans. Microwave Theory Tech. **10**, 9–13 (1962).
51. M. Wubs, L. G. Suttorp, and A. Lagendijk, "Multiple-scattering approach to interatomic interactions and superradiance in inhomogeneous dielectrics," Phys. Rev. A **70**, 053823 (2004).
52. P. R. Berman and G. W. Ford, "Spontaneous decay, unitarity, and the Weisskopf–Wigner approximation," Adv. At. Mol. Opt. Phys. **59**, Chap. 5, 175–221 (2010).
53. P. R. Berman and G. W. Ford, "Spectrum in spontaneous emission: beyond the Weisskopf-Wigner approximation," Phys. Rev. A **82**, 023818 (2010).
54. J. F. Leandro and F. L. Semião, "Entanglement in Weisskopf–Wigner theory of atomic decay in free space," Opt. Commun. **282**, 4736–4740 (2009).

55. W. H. Press, S. A. Teukolsky, W. T. Vetterling, and B. P. Flannery, *Numerical Recipes* (Cambridge University, 2007).
56. L. Novotny and B. Hecht, *Principles of Nano-Optics*, 2nd Ed. (Cambridge University, 2012).
57. H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University, 2007).
58. From a strict mathematical perspective, self-adjointness is actually not enough, but SA together with having a compact inverse does guarantee the validity of eigenfunction expansions [59]. This condition is satisfied by typical differential equations arising in electromagnetics and quantum mechanical problems, which have compact integral inverse operators.
59. G. W. Hanson and A. B. Yakovlev, *Operator Theory for Electromagnetics* (Springer-Verlag, 2002).
60. S. De Liberato, "Virtual photons in the ground state of a dissipative system," Nat. Commun. **8**, 1–6 (2017).