# **Unified approach to QED in arbitrary linear media**

Christian Raabe,<sup>1</sup> Stefan Scheel,<sup>2</sup> and Dirk-Gunnar Welsch<sup>1</sup>

1 *Theoretisch-Physikalisches Institut, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, D-07743 Jena, Germany*

2 *Quantum Optics and Laser Science, Blackett Laboratory, Imperial College London, Prince Consort Road,*

*London SW7 2BW, United Kingdom*

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We give a unified approach to macroscopic QED in arbitrary linearly responding media, based on the quite general, nonlocal form of the conductivity tensor as it can be introduced within the framework of linear response theory, and appropriately chosen sets of bosonic variables. The formalism generalizes the quantization schemes that have been developed previously for diverse classes of linear media. In particular, it turns out that the scheme developed for locally responding linear magnetodielectric media can be recovered from the general scheme as a limiting case for weakly spatially dispersive media. With regard to practical applications, we furthermore address the dielectric approximation for the conductivity tensor and the surface impedance method for the calculation of the Green tensor of the macroscopic Maxwell equations, the two central quantities of the theory.

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# **I. INTRODUCTION**

In both classical and quantum electrodynamics, it is often advisable to divide, at least notionally, the matter that interacts with the electromagnetic field into a part that plays the role of a passive background and a remainder, active part that needs to be considered in more detail. By means of suitable coarse-graining and averaging procedures, this leads to the well-known framework of Maxwell's phenomenological equations, where the background—the medium—is treated as a continuum and, quite frequently, by the methods of linear response theory. From this perspective, the characterization of the medium is reduced to the prescription of suitable constitutive relations, i.e., appropriate response functions or susceptibilities.

Depending on the specific kinds of media under consideration, it is under many circumstances sufficiently accurate to work with spatially local response functions, taking into account only (temporal) dispersion and absorption in accordance with causality. For conducting and semiconducting media (not to mention plasmas) as well as superconducting materials, however, the spatially local description can be inadequate due to the existence of almost freely movable charge carriers (conduction electrons, excitons, Cooper pairs) in such media. Hence if one is not willing to restrict one's attention to a crude spatial resolution and/or specific frequency windows, spatial dispersion, i.e., the spatially nonlocal character of the medium response, generally cannot be disregarded for such media. Electrodynamics problems with the inclusion of spatial dispersion have been considered by various authors in different ways, both on the classical and quantum levels; for classical approaches, see, e.g., Refs.  $\left[1-7\right]$  $\left[1-7\right]$  $\left[1-7\right]$ , for quantum ones see, e.g., Refs.  $\left[8,9\right]$  $\left[8,9\right]$  $\left[8,9\right]$  $\left[8,9\right]$ .

A scheme that takes spatial dispersion into account along with dispersion and absorption in sufficiently general terms can also be regarded as an important step towards a satisfactory (quantum) electrodynamics of moving media, which is very much lacking at present. The reason is that a medium, even if it can be assumed to respond spatially locally when it is at rest, will in general appear as responding nonlocally when it is in motion. Given that the polarization of typical Drude-Lorentz-type dielectrics responds to the electric field with a characteristic memory time of the order of  $10^{-9} - 10^{-7}$  $10^{-9} - 10^{-7}$  s [10], already moderate (i.e., nonrelativistic) velocities may lead to the appearance of noticeable spatial nonlocalities. For example, sonoluminescence experiments show that the collapse of a bubble with a typical initial radius of 10–50  $\mu$ m to a final radius of around 1  $\mu$ m occurs on a time scale similar to the characteristic memory time of the response of the surrounding fluid  $\lceil 11 \rceil$  $\lceil 11 \rceil$  $\lceil 11 \rceil$ .

The study of the quantized electromagnetic field in spatially nonlocally responding media and the prospect of elaborating a quantum theory of light in moving media will also open up new ways of investigating quantum effects related to the recently proposed "optical black hole"  $[12,13]$  $[12,13]$  $[12,13]$  $[12,13]$ . So far, the theory has concentrated on purely geometrical optics with some progress being made towards a (scalar) waveoptical description, but a consistent linear-response approach is still lacking.

In this paper, we develop a unified approach to macroscopic QED in linear media, which generalizes previous quantization schemes to a theoretical concept applicable to arbitrary (equilibrium) media that linearly respond to the electromagnetic field. The only basic prerequisite is to have available the conductivity tensor of the medium, which enters the macroscopic Maxwell equations as a complex function of frequency, and, in the general case of spatially dispersive media, in a spatially nonlocal way. In particular, we show that, and how, the quantization schemes proposed for special classes of media turn out to be limiting cases of a quite general, universally valid scheme.

Its practical application requires, for a given conductivity tensor, the determination of the Green tensor of the macroscopic Maxwell equations—a nontrivial task, especially if spatial dispersion must be taken into account. Addressing this problem, we derive, on the basis of the dielectric approximation, general relations that can be advantageously employed to calculate Green tensors using the surface impedance method. To exemplify this, we present exact formulas for a planar, nonmagnetic three-layer system, which show that the properties of the two surfaces are interconnected by "crossing" terms referring to both surfaces.

The paper is organized as follows. In Sec. II, we develop the concept of electromagnetic-field quantization in arbitrary linearly responding media, and in Sec. III, we study in more detail possible choices of appropriate dynamical variables. We then proceed to show in Sec. IV how previously introduced quantization schemes for diverse classes of media can be obtained as special cases. In Sec. V, we address the problem of determining the Green tensor for spatially dispersive systems. We end the paper with some concluding remarks in Sec. VI.

## **II. QUANTIZATION SCHEME**

The effect of any linear, dispersing, and absorbing medium on the electromagnetic field can be described, within the framework of linear response theory, by the relation

<span id="page-1-0"></span>
$$
\underline{\mathbf{j}}(\mathbf{r},\omega) = \int d^3 r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{E}}(\mathbf{r}', \omega) + \underline{\mathbf{j}}_N(\mathbf{r}, \omega), \qquad (1)
$$

where  $\mathbf{j}(\mathbf{r}, \omega)$  and  $\mathbf{E}(\mathbf{r}, \omega)$ , respectively, are the (linearly responding) current density and the electric field in the frequency domain,  $Q(\mathbf{r}, \mathbf{r}', \omega)$  is the complex conductivity ten-sor in the frequency domain [[7,](#page-21-1)[14](#page-21-8)], and  $\mathbf{j}_N(\mathbf{r}, \omega)$  is a Langevin noise source. In order to clearly distinguish quantities such as the electromagnetic fields and the charge and current densities in the time domain from their respective frequency-domain counterparts, the latter are underlined. Other quantities typically given as functions of frequency such as conductivities, permittivities, or permeabilities, are not underlined. Throughout the paper, dot products of vectors (typeset in boldface) and/or second-rank tensors (carrying a two-sided arrow), corresponding to the contraction of "neighboring" indices of the respective Cartesian components, are indicated by a dot between the factors; if no intervening symbol is given, dyadic (or tensor) products are meant, corresponding to juxtaposition of the components without contraction.

According to the Onsager reciprocity theorem  $[7,14]$  $[7,14]$  $[7,14]$  $[7,14]$ , the conductivity tensor should be reciprocal,  $Q_{ij}(\mathbf{r}, \mathbf{r}', \omega)$  $=Q_{ji}(\mathbf{r}', \mathbf{r}, \omega)$ . Except for a translationally invariant (bulk) medium, the spatial arguments **r** and **r**' of  $\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega)$  must be kept as two separate variables in general. We assume that, for chosen  $\omega$ ,  $Q(\mathbf{r}, \mathbf{r}', \omega)$  is the integral kernel of a reasonably well-behaved (integral) operator acting on vector functions in position space. In particular, we assume that  $\hat{Q}(\mathbf{r}, \mathbf{r}', \omega)$  tends (sufficiently rapidly) to zero for  $|\mathbf{r} - \mathbf{r}'|$  $\rightarrow \infty$  and has no strong (i.e., nonintegrable) singularities (specifically, for  $\mathbf{r}' \rightarrow \mathbf{r}$ ). To allow for the spatially nondispersive limit,  $\delta$ -functions and their derivatives must be permitted so that  $Q(\mathbf{r}, \mathbf{r}', \omega)$  may become a (quasi-)local integral kernel. In the remainder of the paper, we will use the superscripts T and  $+$  to indicate transposition and Hermitian conjugation with respect to tensor indices. Since the spatial arguments are not switched by these operations, the operator associated

with an integral kernel  $\vec{A}(\mathbf{r}, \mathbf{r}')$  is Hermitian if  $\vec{A}(\mathbf{r}, \mathbf{r}')$  $=A^+(r', r)$ . In particular, an operator associated with a real kernel is Hermitian if it has the reciprocity property  $\widetilde{A}(\mathbf{r}, \mathbf{r}') = \widetilde{A}^{\mathsf{T}}(\mathbf{r}', \mathbf{r})$ The decomposition  $Q(\mathbf{r},\mathbf{r}',\omega)$  $=$ Re  $Q(\mathbf{r}, \mathbf{r}', \omega) + i \operatorname{Im} Q(\mathbf{r}, \mathbf{r}', \omega)$  of the conductivity tensor is therefore identical with the decomposition of the associated operator into a Hermitian and an anti-Hermitian part,

<span id="page-1-2"></span>
$$
\begin{aligned} \n\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) &= \text{Re } \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \\ \n&= \frac{1}{2} [\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) + \vec{Q}^+(\mathbf{r}', \mathbf{r}, \omega)], \n\end{aligned} \tag{2}
$$

<span id="page-1-3"></span>
$$
\begin{aligned} \n\tilde{\tau}(\mathbf{r}, \mathbf{r}', \omega) &= \operatorname{Im} \tilde{Q}(\mathbf{r}, \mathbf{r}', \omega) \\ \n&= \frac{1}{2i} [\tilde{Q}(\mathbf{r}, \mathbf{r}', \omega) - \tilde{Q}^+(\mathbf{r}', \mathbf{r}, \omega)]. \n\end{aligned} \tag{3}
$$

Since  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is associated with the dissipation of electromagnetic energy (see, e.g., Refs.  $[7,14]$  $[7,14]$  $[7,14]$  $[7,14]$ ), the operator associated with the integral kernel  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is, for real  $\omega$ , a positive definite operator in the case of absorbing media considered throughout this paper.

The conductivity tensor  $\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is the temporal Fourier transform of a response function,

$$
\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)[\equiv 2\pi \vec{Q}(\mathbf{r}, \mathbf{r}', \omega)] = \int dt e^{i\omega t} \vec{Q}(\mathbf{r}, \mathbf{r}', t), \quad (4)
$$

where  $\hat{Q}(\mathbf{r}, \mathbf{r}', t)$  satisfies causality conditions of the type

$$
\ddot{Q}(\mathbf{r}, \mathbf{r}', t) = 0 \quad \text{if } t - \cos \eta |\mathbf{r} - \mathbf{r}'|/c < 0 \tag{5}
$$

for chosen **r** and **r**' and arbitrary directional cosines cos  $\eta$  $(0 \le \cos \eta \le 1)$ . In particular, for cos  $\eta = 0$ , one finds from arguments  $\lceil 14-16 \rceil$  $\lceil 14-16 \rceil$  $\lceil 14-16 \rceil$  similar to those for the case of spatially locally responding media that, for chosen **r** and **r**',  $Q(\mathbf{r}, \mathbf{r}', \omega)$  is analytic in the upper complex  $\omega$  half-plane, fulfills Kramers-Kronig (Hilbert transform) relations, and satisfies the Schwarz reflection principle  $Q^*(\mathbf{r}, \mathbf{r}', \omega)$  $= Q(\mathbf{r}, \mathbf{r}', -\omega^*)$ . Other values of cos  $\eta$  could obviously provide more stringent (spatiotemporal) conditions (see also Ref. [[17](#page-21-10)]) which are, however, not required here.

Let us identify the current density that enters the macroscopic Maxwell equations in the frequency domain with  $\mathbf{j}(\mathbf{r}, \omega)$  as specified in Eq. ([1](#page-1-0)). In this case, the mediumassisted electric field in the frequency domain satisfies the integrodifferential equation

<span id="page-1-1"></span>
$$
\nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \underline{\mathbf{E}}(\mathbf{r}, \omega)
$$

$$
-i\mu_0 \omega \int d^3 r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{E}}(\mathbf{r}', \omega) = i\mu_0 \omega \underline{\mathbf{j}}_N(\mathbf{r}, \omega),
$$
(6)

whose unique solution is

$$
\underline{\mathbf{E}}(\mathbf{r},\omega) = i\mu_0 \omega \int d^3r' \vec{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{j}}_N(\mathbf{r}', \omega), \quad (7)
$$

<span id="page-2-0"></span>with  $G(\mathbf{r}, \mathbf{r}', \omega)$  being the (retarded) Green tensor. It satisfies Eq. ([6](#page-1-1)) with the (tensorial)  $\delta$ -function source,

<span id="page-2-10"></span>
$$
\nabla \times \nabla \times \vec{G}(\mathbf{r}, \mathbf{s}, \omega) - \frac{\omega^2}{c^2} \vec{G}(\mathbf{r}, \mathbf{s}, \omega)
$$

$$
-i\mu_0 \omega \int d^3 r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \vec{G}(\mathbf{r}', \mathbf{s}, \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{s}),
$$
(8)

together with the boundary condition at infinity, and has all the attributes of a (Fourier transformed) causal response function just as  $Q(\mathbf{r}, \mathbf{r}', \omega)$  has them. In particular, it is analytic in the upper  $\omega$  half-plane and the Schwarz reflection principle  $\tilde{G}^*(\mathbf{r}, \mathbf{r}', \omega) = \tilde{G}(\mathbf{r}, \mathbf{r}', -\omega^*)$  is valid. Its basic properties in position space are similar to the ones known from the spatially local theory, in particular, since  $\hat{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is reciprocal, so is  $G(\mathbf{r}, \mathbf{r}', \omega)$ ,  $G(\mathbf{r}, \mathbf{r}', \omega) = G^{\mathsf{T}}(\mathbf{r}', \mathbf{r}, \omega)$ , and, for real  $\omega$ , the generalized integral relation

<span id="page-2-2"></span>
$$
\mu_0 \omega \int d^3 s \int d^3 s' \vec{G}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{\sigma}(\mathbf{s}, \mathbf{s}', \omega) \cdot \vec{G}^*(\mathbf{s}', \mathbf{r}', \omega)
$$
  
= Im  $\vec{G}(\mathbf{r}, \mathbf{r}', \omega)$  (9)

holds (Appendix A).

To quantize the theory, the Langevin noise source  $\underline{\mathbf{j}}_{N}(\mathbf{r},\omega)$ is regarded as an operator  $[\mathbf{j}_N(\mathbf{r}, \omega) \mapsto \hat{\mathbf{j}}_N(\mathbf{r}, \omega)]$  with the commutation relation

$$
\left[\hat{\mathbf{j}}_{N}(\mathbf{r},\omega),\hat{\mathbf{j}}_{N}^{\dagger}(\mathbf{r}',\omega')\right]=\frac{\hbar\omega}{\pi}\delta(\omega-\omega')\tilde{\sigma}(\mathbf{r},\mathbf{r}',\omega). \quad (10)
$$

<span id="page-2-7"></span>The (now operator-valued) equation ([7](#page-2-0)) relates the electric field operator

$$
\hat{\mathbf{E}}(\mathbf{r}) = \int_0^\infty d\omega \hat{\mathbf{E}}(\mathbf{r}, \omega) + \text{H.c.},\tag{11}
$$

and thus all the electromagnetic field operators, to  $\hat{\mathbf{j}}_{N}(\mathbf{r}, \omega)$ and  $\hat{\mathbf{j}}_{N}^{\dagger}(\mathbf{r}, \omega)$ , which may be regarded as the dynamical variables of the overall system consisting of the electromagnetic field and the linear medium (incorporating the reservoir degrees of freedom responsible for absorption). It should be mentioned that, by means of the correspondence

$$
\frac{i}{\varepsilon_0 \omega} \widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega) \leftrightarrow \widetilde{\chi}(\mathbf{r}, \mathbf{r}', \omega), \tag{12}
$$

<span id="page-2-9"></span>where  $\vec{\chi}(\mathbf{r}, \mathbf{r}', \omega)$  is the (nonlocal) dielectric susceptibility tensor, the basic commutation relation  $(10)$  $(10)$  $(10)$  becomes equivalent to the commutation relation derived from a microscopic, linear two-band model of dielectric material  $\lceil 18 \rceil$  $\lceil 18 \rceil$  $\lceil 18 \rceil$ , which has been used to study the quantized electromagnetic field in spatially dispersive dielectrics  $[8,9]$  $[8,9]$  $[8,9]$  $[8,9]$ .

In order to complete the quantization scheme, a Hamiltonian  $\hat{H}$  needs to be introduced [as a functional of  $\hat{\mathbf{j}}_{N}(\mathbf{r}, \omega)$ 

<span id="page-2-1"></span>and  $\hat{\mathbf{j}}_{N}^{\dagger}(\mathbf{r}, \omega)$  so as to generate "free" time evolution according to

$$
[\hat{\mathbf{j}}_{N}(\mathbf{r},\omega),\hat{H}]=\hbar\,\omega\hat{\mathbf{j}}_{N}(\mathbf{r},\omega),\qquad(13)
$$

which constrains the Hamiltonian to the form

<span id="page-2-3"></span>
$$
\hat{H} = \pi \int_0^{\infty} d\omega \int d^3r \int d^3r' \hat{\mathbf{j}}_N^{\dagger}(\mathbf{r}, \omega) \cdot \tilde{\rho}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{j}}_N(\mathbf{r}', \omega), \tag{14}
$$

to within irrelevant *c*-number contributions. A glance at Eqs. ([10](#page-2-0)) and ([13](#page-2-1)) now shows that  $\vec{\rho}(\mathbf{r}, \mathbf{r}', \omega)$  is the integral kernel of the inverse operator of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  (which exists). The validity of the quantization scheme is confirmed by checking that the well-known equaltime commutation relations for the electromagnetic field operators hold, which can be done, in analogy to the spatially local theory (cf. Refs.  $[19-21]$  $[19-21]$  $[19-21]$ ), by properly taking into account the properties of  $G(\mathbf{r}, \mathbf{r}', \omega)$  and  $\hat{Q}(\mathbf{r}, \mathbf{r}', \omega)$  [in particular, Eq.  $(9)$  $(9)$  $(9)$ ].

<span id="page-2-5"></span>The Hamiltonian  $(14)$  $(14)$  $(14)$  may clearly be brought to the diagonal form

$$
\hat{H} = \int_0^\infty d\omega \hbar \omega \int d^3 r \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega)
$$
 (15)

known from the spatially local theory, where  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  is a bosonic field,

$$
[\hat{\mathbf{f}}(\mathbf{r},\omega),\hat{\mathbf{f}}^{\dagger}(\mathbf{r}',\omega')] = \delta(\omega - \omega')\widetilde{I}\delta(\mathbf{r} - \mathbf{r}'),\qquad(16)
$$

<span id="page-2-6"></span><span id="page-2-4"></span>by performing a linear transformation of the variables which we shall assume to be invertible. Writing

$$
\hat{\mathbf{j}}_{\mathcal{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{1/2} \int d^3r' \vec{K}(\mathbf{r}, \mathbf{r}',\omega) \cdot \hat{\mathbf{f}}(\mathbf{r}',\omega), \quad (17)
$$

the diagonalization is achieved and Eqs.  $(10)$  $(10)$  $(10)$  and  $(16)$  $(16)$  $(16)$  are rendered equivalent if we choose the integral kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$  such that, for real  $\omega$ , the integral equation

$$
\int d^3 s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{K}^+(\mathbf{r}', \mathbf{s}, \omega) = \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)
$$
(18)

<span id="page-2-8"></span>holds, which is guaranteed to possess solutions (see Sec. III) since  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is the integral kernel of a positive definite operator.

So far we have considered the "free" medium-assisted electromagnetic field. Its interaction with additional (e.g., atomic) systems can be included in the theory on the basis of the well-known minimal- or multipolar-coupling schemes in the usual way. For instance, let us consider (spinless) pointlike, nonrelativistic, charged particles (charges  $Q_a$ , masses  $m_a$ ) described in terms of positions  $\hat{\mathbf{r}}_a$  and conjugate momenta  $\hat{\mathbf{p}}_a$ , which interact with the medium-assisted field. One can simply add to the Hamiltonian  $(15)$  $(15)$  $(15)$  the term

$$
\hat{H} = \sum_{a} (2m_a)^{-1} [\hat{\mathbf{p}}_a - Q_a \hat{\mathbf{A}} (\hat{\mathbf{r}}_a)]^2 + \hat{W}
$$
(19)

<span id="page-3-0"></span>to obtain the total minimal-coupling Hamiltonian  $\hat{H} + \hat{H}'$ , which [together with commutation relations  $(16)$  $(16)$  $(16)$  and  $[\hat{\mathbf{r}}_a, \hat{\mathbf{p}}_{a'}] = i\hbar \delta_{aa'}\hat{\mathbf{l}}$  may be used to describe the dynamics of the coupled systems. In Eq.  $(19)$  $(19)$  $(19)$ ,  $\hat{A}(r)$  is the vector potential in the Coulomb gauge. Writing

$$
\hat{\mathbf{A}}(\mathbf{r}) = \int_0^\infty d\omega \hat{\mathbf{A}}(\mathbf{r}, \omega) + \text{H.c.},\tag{20}
$$

we may express  $\hat{A}(\mathbf{r}, \omega)$  in terms of the transverse part  $\hat{\mathbf{E}}^{\perp}(\mathbf{r},\omega)$  of  $\hat{\mathbf{E}}(\mathbf{r},\omega)$  as given by Eq. ([7](#page-2-0)),

$$
\hat{\mathbf{\underline{A}}}(\mathbf{r},\omega) = (i\omega)^{-1}\hat{\mathbf{\underline{E}}}^{\perp}(\mathbf{r},\omega). \tag{21}
$$

Taking into account Eq.  $(17)$  $(17)$  $(17)$ , we may eventually express  $\hat{A}(\bf{r})$  in terms of the basic bosonic variables  $\hat{f}(\bf{r},\omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$ . The total Coulomb interaction energy  $\hat{W}$  in Eq. ([19](#page-3-0)) reads

$$
\hat{W} = \frac{1}{2} \sum_{a} Q_a \hat{U}(\hat{\mathbf{r}}_a) + \sum_{a} Q_a \hat{V}(\hat{\mathbf{r}}_a),\tag{22}
$$

where  $\hat{U}(\mathbf{r})$  and  $\hat{V}(\mathbf{r})$  are, respectively, the scalar potentials ascribed to the charged particles and the longitudinal part  $\hat{\mathbf{E}}^{\parallel}(\mathbf{r})$  of  $\hat{\mathbf{E}}(\mathbf{r})$ . Making use of Eq. ([11](#page-2-7)) together with Eqs. ([7](#page-2-0)) and ([17](#page-2-6)), we may express also  $\hat{V}(\mathbf{r})$  in terms of  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$ . For further details, including the multipolar coupling scheme, we refer the reader, e.g., to Ref.  $[22]$  $[22]$  $[22]$ .

# **III. NATURAL VARIABLES AND PROJECTIVE VARIABLES**

Let us now turn to the problem of constructing the integral kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. ([18](#page-2-8)). For this purpose, we consider the eigenvalue problem

<span id="page-3-6"></span>
$$
\int d^3r' \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{F}(\alpha, \mathbf{r}', \omega) = \sigma(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \quad (23)
$$

which, under appropriate regularity assumptions on the conductivity tensor  $\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega)$  such as those listed below Eq. ([1](#page-1-0)), is well-defined. In particular, it features a real (positive) spectrum and a complete set of orthogonal eigensolutions, which we may take to be  $(\delta)$  normalized. Note that the real  $\omega$  plays the role of a parameter here, and  $\alpha$  stands for the collection of (discrete and/or continuous) indices needed to label the eigenfunctions. Adopting a continuum notation, we may write

$$
\int d\alpha \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \quad (24)
$$

$$
\int d^3r \mathbf{F}^*(\alpha, \mathbf{r}, \omega) \cdot \mathbf{F}(\alpha', \mathbf{r}, \omega) = \delta(\alpha - \alpha'), \quad (25)
$$

<span id="page-3-1"></span>
$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \int d\alpha \sigma(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega), \quad (26)
$$

which resembles the expansion of the dielectric susceptibility in the microscopic theory  $[18]$  $[18]$  $[18]$  mentioned above. Substituting Eq. ([26](#page-3-1)) into Eq. ([18](#page-2-8)), we may construct an integral kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$  in the form of

<span id="page-3-3"></span>
$$
\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \int d\alpha \sigma^{1/2}(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega), \tag{27}
$$

where we choose  $\sigma^{1/2}(\alpha,\omega) > 0$  so that the operator associated with  $K(\mathbf{r}, \mathbf{r}', \omega)$  is the positive, Hermitian square-root of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ . Obviously, this solution to Eq.  $(18)$  $(18)$  $(18)$  is not unique, since any other kernel of the form

$$
\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) = \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}(\mathbf{s}, \mathbf{r}', \omega)
$$
(28)

<span id="page-3-7"></span><span id="page-3-2"></span>with  $V(\mathbf{r}, \mathbf{s}, \omega)$  satisfying

$$
\int d^3s \vec{V}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}^+(\mathbf{r}', \mathbf{s}, \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}')
$$
 (29)

also obeys Eq.  $(18)$  $(18)$  $(18)$ . As we are interested in invertible trans-formations ([17](#page-2-6)), the operator corresponding to  $V(\mathbf{r}, \mathbf{s}, \omega)$ should be invertible as well, so that we can replace Eq.  $(29)$  $(29)$  $(29)$ with the stronger unitarity condition

$$
\int d^3s \vec{V}^+(\mathbf{s}, \mathbf{r}, \omega) \cdot \vec{V}(\mathbf{s}, \mathbf{r}', \omega) = \int d^3s \vec{V}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}^+(\mathbf{r}', \mathbf{s}, \omega)
$$

$$
= \vec{I} \delta(\mathbf{r} - \mathbf{r}'). \tag{30}
$$

Without loss of generality (see Appendix B), we can base our further calculations on Eq.  $(27)$  $(27)$  $(27)$ .

Inserting Eq.  $(27)$  $(27)$  $(27)$  into Eq.  $(17)$  $(17)$  $(17)$ , we find that

$$
\hat{\mathbf{j}}_{\mathcal{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{1/2} \int d\alpha \sigma^{1/2}(\alpha,\omega) \mathbf{F}(\alpha,\mathbf{r},\omega) \hat{g}(\alpha,\omega),\tag{31}
$$

<span id="page-3-4"></span>where we have introduced the new variables

$$
\hat{g}(\alpha,\omega) = \int d^3 r \mathbf{F}^*(\alpha, \mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega), \qquad (32)
$$

referred to as the natural variables in the following. Needless to say that they are again of bosonic type,

$$
[\hat{g}(\alpha,\omega), \hat{g}^{\dagger}(\alpha',\omega')] = \delta(\alpha-\alpha')\delta(\omega-\omega').
$$
 (33)

Since the transformation ([32](#page-3-4)) does not mix different  $\omega$  com-ponents, the Hamiltonian ([15](#page-2-5)) is still diagonal when expressed in terms of the natural variables,

$$
\hat{H} = \int_0^\infty d\omega \hbar \omega \int d\alpha \hat{g}^\dagger(\alpha, \omega) \hat{g}(\alpha, \omega), \tag{34}
$$

<span id="page-3-5"></span>and the diagonal expansion of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  reads

as can be easily seen by inverting Eq.  $(32)$  $(32)$  $(32)$ ,

$$
\hat{\mathbf{f}}(\mathbf{r}, \omega) = \int d\alpha \mathbf{F}(\alpha, \mathbf{r}, \omega) \hat{g}(\alpha, \omega),
$$
 (35)

<span id="page-4-0"></span>and combining with Eq.  $(15)$  $(15)$  $(15)$ , on recalling Eq.  $(25)$  $(25)$  $(25)$ .

Let us organize the set of eigenfunctions  $\mathbf{F}(\alpha, \mathbf{r}, \omega)$  into (a discrete number of) subsets labeled by  $\lambda$  ( $\lambda = 1, 2, ..., \Lambda$ ). With the notation  $\alpha \mapsto (\lambda, \beta)$ , Eq. ([35](#page-4-0)) then reads

$$
\hat{\mathbf{f}}(\mathbf{r},\omega) = \sum_{\lambda} \hat{\mathbf{f}}_{\lambda}(\mathbf{r},\omega),
$$
 (36)

<span id="page-4-9"></span>where

$$
\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega) = \int d\beta \mathbf{F}_{\lambda}(\beta, \mathbf{r}, \omega) \hat{g}_{\lambda}(\beta, \omega).
$$
 (37)

<span id="page-4-8"></span>The operators associated with the integral kernels

$$
\vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) = \int d\beta \mathbf{F}_{\lambda}(\beta, \mathbf{r}, \omega) \mathbf{F}_{\lambda}^{*}(\beta, \mathbf{r}', \omega)
$$
(38)

<span id="page-4-10"></span>form a complete set of orthogonal projectors. Obviously, these projectors and the operators associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ and  $K(\mathbf{r}, \mathbf{r}', \omega)$  as given by Eq. ([27](#page-3-3)) are commuting quantities. It is not difficult to see that the variables

$$
\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega) = \int d^3 r' \vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}', \omega)
$$

$$
= \int d\beta \mathbf{F}_{\lambda}(\beta, \mathbf{r}, \omega) \hat{g}_{\lambda}(\beta, \omega), \qquad (39)
$$

<span id="page-4-1"></span>referred to as projective variables in the following, obey the nonbosonic commutation relation

$$
[\hat{\mathbf{f}}_{\lambda}(\mathbf{r},\omega), \hat{\mathbf{f}}_{\lambda'}^{\dagger}(\mathbf{r}',\omega')] = \delta_{\lambda\lambda'} \delta(\omega - \omega') \widetilde{P}_{\lambda}(\mathbf{r},\mathbf{r}',\omega), \quad (40)
$$

<span id="page-4-2"></span>and the Hamiltonian  $(15)$  $(15)$  $(15)$  expressed in terms of the projective variables reads as

$$
\hat{H} = \sum_{\lambda} \int_0^{\infty} d\omega \hbar \omega \int d^3 r \hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega). \tag{41}
$$

<span id="page-4-7"></span>From Eqs.  $(40)$  $(40)$  $(40)$  and  $(41)$  $(41)$  $(41)$  it then follows that

$$
[\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \hat{H}] = \hbar \omega \int d^{3}r' \vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}', \omega)
$$

$$
= \hbar \omega \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega). \tag{42}
$$

<span id="page-4-5"></span>Inserting Eq.  $(35)$  $(35)$  $(35)$  in Eq.  $(17)$  $(17)$  $(17)$ , we obtain

$$
\hat{\mathbf{j}}_{N}(\mathbf{r},\omega) = \sum_{\lambda} \hat{\mathbf{j}}_{N\lambda}(\mathbf{r},\omega),
$$
\n(43)

<span id="page-4-6"></span>where the  $\hat{\mathbf{j}}_{N\lambda}(\mathbf{r}, \omega)$  are given by

$$
\hat{\mathbf{j}}_{N\lambda}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{1/2} \int d^3r' \vec{K}_{\lambda}(\mathbf{r},\mathbf{r}',\omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}',\omega), \quad (44)
$$

<span id="page-4-3"></span>
$$
\vec{K}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) = \int d^3s \vec{P}_{\lambda}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{K}(\mathbf{s}, \mathbf{r}', \omega)
$$

$$
= \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{P}_{\lambda}(\mathbf{s}, \mathbf{r}', \omega). \tag{45}
$$

<span id="page-4-4"></span>Recalling Eq.  $(40)$  $(40)$  $(40)$ , we can easily see that

$$
\left[\hat{\mathbf{j}}_{N\lambda}(\mathbf{r},\omega),\hat{\mathbf{j}}_{N\lambda'}^{\dagger}(\mathbf{r}',\omega')\right] = \frac{\hbar\omega}{\pi}\delta_{\lambda\lambda'}\delta(\omega-\omega')\tilde{\sigma}_{\lambda}(\mathbf{r},\mathbf{r}',\omega),\tag{46}
$$

where  $\vec{\sigma}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega)$  is defined according to Eq. ([45](#page-4-3)) with  $\vec{\sigma}$  in place of K. Summation of Eq. ([46](#page-4-4)) over  $\lambda$  and  $\lambda'$  leads back to Eq.  $(10)$  $(10)$  $(10)$ , so that the two equations are equivalent.

At this stage, we observe that there is the option to base the quantization scheme directly on Eqs.  $(41)$  $(41)$  $(41)$ ,  $(43)$  $(43)$  $(43)$ , and  $(44)$  $(44)$  $(44)$ , regarding the variables  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega)$  as the basic dynamical variables of the theory and assigning to them bosonic commutation relations

$$
[\hat{\mathbf{f}}_{\lambda}(\mathbf{r},\omega), \hat{\mathbf{f}}_{\lambda'}^{\dagger}(\mathbf{r}',\omega')] = \delta_{\lambda\lambda'}\delta(\omega - \omega')\vec{I}\delta(\mathbf{r} - \mathbf{r}') \qquad (47)
$$

<span id="page-4-11"></span>in place of Eq. ([40](#page-4-1)). Note that, in so doing, back reference from the variables  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  to the original variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  is not possible anymore. As can be seen from Eqs. ([44](#page-4-6)) and ([45](#page-4-3)), Eq. ([46](#page-4-4)) is satisfied also when the  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega)$ are considered as bosonic variables, from which it follows [via Eq.  $(43)$  $(43)$  $(43)$ ] that Eq.  $(10)$  $(10)$  $(10)$  also still holds and, as before, this implies that the correct electromagnetic-field commutation relations hold. The second line of Eq. ([42](#page-4-7)) remains of course also true so that the correct time evolution is ensured as well.

Since the state space attributed to the bosonic variables  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r},\omega)$  and  $\hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r},\omega)$  is, in general, different from the state space attributed to the original variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$ [or, equivalently, attributed to  $\hat{g}_{\lambda}(\beta,\omega)$  and  $\hat{g}_{\lambda}^{\dagger}(\beta,\omega)$ ], the allowable states must be restricted by ruling out certain coherent superpositions of states in the sense of a superselection rule. In Appendix C, we show that the condition imposed on the states may be described by means of a set of projectors  $\hat{P}_\lambda$  such that the allowable states  $|\psi\rangle$  can be characterized by

$$
\hat{P}_{\lambda}|\psi\rangle = |\psi\rangle \,\forall \ \lambda,\tag{48}
$$

where the action of the projectors  $\hat{P}_\lambda$  in state space is closely related to the action of the projectors associated with the kernels ([38](#page-4-8)) in position space. As a result, if the total Hamiltonian  $\hat{H}_{\text{tot}}$  composed of the Hamiltonian ([41](#page-4-2)) and possible interaction terms (in the case where additional, active sources are present) commutes with all of the projectors  $\hat{P}_\lambda$ ,

$$
[\hat{P}_{\lambda}, \hat{H}_{\text{tot}}] = 0 \,\forall \lambda, \tag{49}
$$

then allowable states remain allowable in the course of time, and the option of treating the  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega)$  as bosonic variables can be safely exercised. Clearly, all the observables of interest should then also commute with the  $\hat{P}_\lambda$  so that no

with

transition matrix elements between states belonging to different subspaces, i.e., between spaces attributed to different  $\lambda$ values, can ever come into play.

One can also consider decompositions of  $\hat{\mathbf{j}}_N(\mathbf{r}, \omega)$ , where in place of the  $\hat{\mathbf{j}}_{N\lambda}(\mathbf{r}, \omega)$  introduced above other quantities  $\hat{\mathbf{J}}_{N\lambda}(\mathbf{r}, \omega)$  subject to the condition

$$
\sum_{\lambda} \hat{\mathbf{j}}_{N\lambda}(\mathbf{r}, \omega) = \sum_{\lambda} \hat{\mathbf{j}}_{N\lambda}(\mathbf{r}, \omega)
$$
 (50)

<span id="page-5-0"></span>are introduced, whose commutation relations may be quite different from those of the  $\hat{\mathbf{j}}_{N\lambda}(\mathbf{r}, \omega)$ . Obviously, the total noise current density  $\hat{\mathbf{j}}_N(\mathbf{r}, \omega)$  as given by Eq. ([43](#page-4-5)) and the commutation relation  $(10)$  $(10)$  $(10)$  are not changed by such a transformation, briefly referred to as gauge transformation in the following. Moreover, since, with regard to Eq.  $(10)$  $(10)$  $(10)$ , only the sum of the commutators  $\left[\hat{\mathbf{J}}_{N\lambda}(\mathbf{r},\omega), \hat{\mathbf{J}}_{N\lambda'}^{\dagger}(\mathbf{r}',\omega')\right]$  over all  $\lambda$ and  $\lambda'$  is relevant, every chosen set of (algebraically consistent) commutators  $\left[\hat{\mathbf{J}}_{N\lambda}(\mathbf{r},\omega), \hat{\mathbf{J}}_{N\lambda'}^{\dagger}(\mathbf{r}',\omega')\right]$  which leads to Eq. ([10](#page-2-0)) yields, in principle, a consistent quantization scheme in its own right. A "substructure below" Eq. ([10](#page-2-0)) can hence be introduced with some arbitrariness, but since the various available alternatives are not necessarily equivalent to each other, a specific one should not be favored in the absence of good (physical) motivation. In contrast, if the observables of interest—including the Hamiltonian—can be viewed as functionals of  $\hat{\mathbf{j}}_N(\mathbf{r}, \omega)$  [rather than of the individual  $\hat{\mathbf{J}}_{N\lambda}(\mathbf{r}, \omega)$ ], Eqs. ([10](#page-2-0)) and ([14](#page-2-3)) can be regarded, in view of the fluctuation-dissipation theorem(s) (see, e.g., Ref. [[14](#page-21-8)]), as being unique, and hence as an invariable fundament of the theory.

From the above, it may be reasonable to widen the notion of projective variables as follows. If, for a chosen (physically motivated) decomposition of the noise current density, it is possible to (linearly) relate the  $\hat{\mathbf{J}}_{N\lambda}(\mathbf{r}, \omega)$  in Eq. ([50](#page-5-0)) to (new) variables  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  such that, upon considering the latter as bosonic variables, the validity of the basic equations  $(10)$  $(10)$  $(10)$  and  $(13)$  $(13)$  $(13)$  is ensured, then the specific quantization scheme so obtained may be regarded as arising from the general quantization scheme by excluding certain types of (superposition) states from state space, and restricting the dynamics (as well as the allowable observables) accordingly. The  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  may then be seen as projective variables in a wider sense.

#### **IV. DIFFERENT CLASSES OF MEDIA**

We proceed to show that rather different classes of media (usually studied separately) fit into the general quantization scheme developed in Sec. II. The main task to be performed is solving the eigenvalue problem  $(23)$  $(23)$  $(23)$ , which requires knowledge of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  for the specific medium under consideration. In two limiting cases, the exact solution to Eq. ([23](#page-3-6)) can be given straightforwardly, namely, in the case of an inhomogeneous medium without spatial dispersion and in the case of a homogeneous medium that shows spatial dispersion. Let us, therefore, first examine these two cases in detail before considering more general situations.

#### **A. Spatially nondispersive inhomogeneous media**

The complete neglect of spatial dispersion means to regard the medium response, i.e.,  $\hat{Q}(\mathbf{r}, \mathbf{r}', \omega)$ , as being strictly local. If this is assumed, we have

$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \vec{\sigma}(\mathbf{r}, \omega) \,\delta(\mathbf{r} - \mathbf{r}'),\tag{51}
$$

<span id="page-5-2"></span><span id="page-5-1"></span>where  $\vec{\sigma}(\mathbf{r}, \omega)$  can be written in diagonal form as

$$
\vec{\sigma}(\mathbf{r}, \omega) = \sum_{i=1}^{3} \sigma_i(\mathbf{r}, \omega) \mathbf{e}_i(\mathbf{r}, \omega) \mathbf{e}_i^*(\mathbf{r}, \omega),
$$
 (52)

with  $\mathbf{e}_i(\mathbf{r}, \omega)$  (*i*=1,2,3) being orthogonal unit vectors. Hence the eigenvalues  $\sigma(\alpha,\omega)$  and eigenfunctions  $\mathbf{F}(\alpha,\mathbf{r},\omega)$  of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  read as  $[\alpha \mapsto (i, \mathbf{s})]$  $\sigma_i$ (**s**,  $\omega$ ) and

$$
\mathbf{F}_i(\mathbf{s}, \mathbf{r}, \omega) = \mathbf{e}_i(\mathbf{s}, \omega) \, \delta(\mathbf{s} - \mathbf{r}), \tag{53}
$$

respectively. Equation  $(27)$  $(27)$  $(27)$  then becomes

$$
\widetilde{K}(\mathbf{r}, \mathbf{r}', \omega) = \widetilde{K}(\mathbf{r}, \omega) \, \delta(\mathbf{r} - \mathbf{r}'),\tag{54}
$$

where

$$
\vec{K}(\mathbf{r},\omega) = \sum_{i=1}^{3} \sigma_i^{1/2}(\mathbf{r},\omega) \mathbf{e}_i(\mathbf{r},\omega) \mathbf{e}_i^*(\mathbf{r},\omega),
$$
(55)

and Eq.  $(17)$  $(17)$  $(17)$  takes the form

$$
\hat{\mathbf{j}}_{\mathcal{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\,\omega}{\pi}\right)^{1/2} \hat{K}(\mathbf{r},\omega) \cdot \hat{\mathbf{f}}(\mathbf{r},\omega),\tag{56}
$$

which just yields the well-known quantization scheme for a locally responding, possibly anisotropic dielectric material [[19](#page-21-12)[,20](#page-21-15)], upon identifying  $\vec{\sigma}(\mathbf{r}, \omega) = \varepsilon_0 \omega \operatorname{Im} \vec{\chi}(\mathbf{r}, \omega)$ , with  $\vec{\chi}(\mathbf{r}, \omega)$  being the (local) dielectric susceptibility tensor [cf. Eq. ([12](#page-2-9))]. The natural variables  $\hat{g}_i(\mathbf{r}, \omega)$  are here simply the components of  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  along the principal axes of the medium, which may in general vary with position and frequency,

$$
\hat{g}_i(\mathbf{r}, \omega) = \mathbf{e}_i^*(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega),
$$
 (57)

$$
\hat{\mathbf{f}}(\mathbf{r},\omega) = \sum_{i=1}^{3} \mathbf{e}_i(\mathbf{r},\omega) \hat{g}_i(\mathbf{r},\omega).
$$
 (58)

Identifying the index  $\lambda$  introduced in Eq. ([36](#page-4-9)) with *i* and assuming that  $\sigma_i(\mathbf{r}, \omega) \neq \sigma_{i'}(\mathbf{r}, \omega)$  for  $i \neq i'$ , one can define, according to Eq.  $(38)$  $(38)$  $(38)$ , the three projection kernels

$$
\overrightarrow{P}_i(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{e}_i(\mathbf{r}, \omega) \mathbf{e}_i^*(\mathbf{r}', \omega) \delta(\mathbf{r} - \mathbf{r}')
$$
(59)

which, according to Eq.  $(39)$  $(39)$  $(39)$ , give rise to three sets of projective variables,

$$
\hat{\mathbf{f}}_i(\mathbf{r}, \omega) = \mathbf{e}_i(\mathbf{r}, \omega) \hat{g}_i(\mathbf{r}, \omega).
$$
 (60)

As long as the projective variables are not coupled to each other—which is obviously the case for the "free" system governed by the Hamiltonian  $(42)$  $(42)$  $(42)$ —they can be regarded as being of bosonic type. In this case, instead of using the origi-

nal set of bosonic variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$ , one can use three sets of bosonic variables  $\hat{\mathbf{f}}_i(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_i^{\dagger}(\mathbf{r}, \omega)$  associated with the three principal axes of the dielectric medium at each space point.

If two of the three eigenvalues  $\sigma_i(\mathbf{r}, \omega)$  coincide (uniaxial medium), the two corresponding projection kernels  $P_i(\mathbf{r}, \mathbf{r}', \omega)$  should be combined into one projector (projecting on the plane perpendicular to the distinguished axis of the medium), thereby reducing the number of sets of projective variables to two. Clearly, if the three eigenvalues  $\sigma_i(\mathbf{r}, \omega)$  all coincide (isotropic medium), the three projection kernels  $\ddot{P}_i(\mathbf{r}, \mathbf{r}', \omega)$  should be combined to give the unit kernel  $\overline{I}\delta(\mathbf{r}-\mathbf{r}')$ , corresponding to the use of the original variables.

## **B. Spatially dispersive homogeneous media**

In the limiting case of an (infinitely extended) homogeneous medium,  $Q(\mathbf{r}, \mathbf{r}', \omega)$  is translationally invariant, i.e., it is a function of the difference **r**−**r**, and so is then  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ . We may therefore represent it as the spatial Fourier transform

$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{\sigma}(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} , \qquad (61)
$$

<span id="page-6-10"></span><span id="page-6-5"></span>where

$$
\vec{\sigma}(\mathbf{k},\omega) = \sum_{i=1}^{3} \sigma_i(\mathbf{k},\omega) \mathbf{e}_i(\mathbf{k},\omega) \mathbf{e}_i^*(\mathbf{k},\omega),
$$
 (62)

with  $\mathbf{e}_i(\mathbf{k}, \omega)$  (*i*=1,2,3) being orthogonal unit vectors. Consequently, the eigenvalues  $\sigma(\alpha,\omega)$  and eigenfunctions **F**( $\alpha, \mathbf{r}, \omega$ ) of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  are  $[\alpha \mapsto (i, \mathbf{k})] \sigma_i(\mathbf{k}, \omega)$  and

$$
\mathbf{F}_i(\mathbf{k}, \mathbf{r}, \omega) = (2\pi)^{-3/2} e^{i\mathbf{k} \cdot \mathbf{r}} \mathbf{e}_i(\mathbf{k}, \omega),
$$
 (63)

<span id="page-6-0"></span>respectively, and Eq.  $(27)$  $(27)$  $(27)$  reads

$$
\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{K}(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} , \qquad (64)
$$

<span id="page-6-1"></span>where

$$
\vec{K}(\mathbf{k},\omega) = \sum_{i=1}^{3} \sigma_i^{1/2}(\mathbf{k},\omega) \mathbf{e}_i(\mathbf{k},\omega) \mathbf{e}_i^*(\mathbf{k},\omega).
$$
 (65)

The combination of Eqs.  $(17)$  $(17)$  $(17)$ ,  $(64)$  $(64)$  $(64)$ , and  $(65)$  $(65)$  $(65)$  then yields

$$
\hat{\mathbf{j}}_{N}(\mathbf{r}, \omega) = \left(\frac{\hbar \omega}{\pi}\right)^{1/2} \frac{1}{(2\pi)^{3/2}}\n\times \sum_{i=1}^{3} \int d^{3}k e^{i\mathbf{k}\cdot\mathbf{r}} \sigma_{i}^{1/2}(\mathbf{k}, \omega) \mathbf{e}_{i}(\mathbf{k}, \omega) \hat{g}_{i}(\mathbf{k}, \omega),
$$
\n(66)

where the natural variables  $\hat{g}_i(\mathbf{k}, \omega)$  are related to the spatial Fourier components of  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  as

$$
\hat{g}_i(\mathbf{k}, \omega) = \frac{1}{(2\pi)^{3/2}} \int d^3 r e^{-i\mathbf{k} \cdot \mathbf{r}} \mathbf{e}_i^*(\mathbf{k}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega).
$$
 (67)

On the basis of the three unit vectors  $\mathbf{e}_i(\mathbf{k}, \omega)$ , three (different) projection kernels can be introduced,

$$
\vec{P}_i(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} ,
$$
 (68)

provided that  $\sigma_i(\mathbf{k}, \omega) \neq \sigma_{i'}(\mathbf{k}, \omega)$  for  $i \neq i'$ .

Let us consider the particular case of isotropic media without optical activity in more detail. In this situation, the diagonal form of the tensor  $\vec{\sigma}$ (**k**,  $\omega$ ) reads (see Ref. [[7](#page-21-1)])

$$
\vec{\sigma}(\mathbf{k},\omega) = \sigma_{\parallel}(k,\omega)\frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\perp}(k,\omega)\bigg(\vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\bigg),\tag{69}
$$

<span id="page-6-2"></span>i.e.,  $\sigma_1(k,\omega) = \sigma_{\parallel}(k,\omega)$  and  $\sigma_2(k,\omega) = \sigma_3(k,\omega) = \sigma_{\perp}(k,\omega)$  $\neq \sigma_{\parallel}(k,\omega)$ , which implies that  $K(\mathbf{k},\omega)$ , Eq. ([65](#page-6-1)), takes the form

$$
\vec{K}(\mathbf{k},\omega) = \sigma_{\parallel}^{1/2}(k,\omega)\frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\perp}^{1/2}(k,\omega)\bigg(\vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\bigg). \tag{70}
$$

<span id="page-6-8"></span>Thus the well-known longitudinal and transverse tensorial  $\delta$ -functions  $\Delta_{\parallel}(\mathbf{r}-\mathbf{r}')$  and  $\Delta_{\perp}(\mathbf{r}-\mathbf{r}')$ , respectively, can be taken as projection kernels,

$$
\widetilde{P}_{\parallel(\perp)}(\mathbf{r}, \mathbf{r}', \omega) = \widetilde{\Delta}_{\parallel(\perp)}(\mathbf{r} - \mathbf{r}'),\tag{71}
$$

<span id="page-6-9"></span><span id="page-6-3"></span>which may be used to introduce, according to Eq.  $(39)$  $(39)$  $(39)$ , the projective variables

$$
\hat{\mathbf{f}}_{\parallel(\perp)}(\mathbf{r},\omega) = \int d^3s \vec{\Delta}_{\parallel(\perp)}(\mathbf{r}-\mathbf{s}) \cdot \hat{\mathbf{f}}(\mathbf{s},\omega). \tag{72}
$$

#### *1. Unitarily equivalent formulation*

As already pointed out in Sec. II, the integral kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. ([17](#page-2-6)) is not uniquely determined by Eq. ([18](#page-2-8)), since any other kernel  $K'(\mathbf{r}, \mathbf{r}', \omega)$  of the form ([28](#page-3-7)) [together with Eq.  $(30)$  $(30)$  $(30)$ ] is also an allowed kernel. To illustrate this for the isotropic medium under study, we first note that Eq.  $(69)$  $(69)$  $(69)$  may be equivalently rewritten as

$$
\overrightarrow{\sigma}(\mathbf{k},\omega) = \sigma_{\parallel}(k,\omega)\overrightarrow{I} - \mathbf{k} \times \gamma(k,\omega)\overrightarrow{I} \times \mathbf{k},\qquad(73)
$$

<span id="page-6-6"></span><span id="page-6-4"></span>where

$$
\gamma(k,\omega) = [\sigma_{\perp}(k,\omega) - \sigma_{\parallel}(k,\omega)]/k^2. \tag{74}
$$

Since (for real  $\omega$ )  $\sigma_{\parallel}(k,\omega)$  and  $\sigma_{\perp}(k,\omega)$  are both real and positive [in accordance with the requirement that  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ be the integral kernel of a positive definite operator],  $\gamma(k,\omega)$ is real but its sign is not determined by this requirement. However, if  $\gamma(k,\omega)$  is required here and below to be positive throughout, then

$$
\vec{K}'(\mathbf{k}, \omega) = \sigma_{\parallel}^{1/2}(k, \omega)\vec{I} \pm \gamma^{1/2}(k, \omega)\mathbf{k} \times \vec{I}
$$
 (75)

<span id="page-6-7"></span>obeys the equation

$$
\vec{K}'(\mathbf{k}, \omega) \cdot \vec{K}'^+(\mathbf{k}, \omega) = \vec{\sigma}(\mathbf{k}, \omega). \tag{76}
$$

Moreover, it can be shown that  $K'(\mathbf{k}, \omega)$  can be represented in the form

$$
\vec{K}'(\mathbf{k}, \omega) = \vec{K}(\mathbf{k}, \omega) \cdot \vec{V}(\mathbf{k}, \omega), \tag{77}
$$

<span id="page-7-5"></span>with

$$
\vec{V}(\mathbf{k}, \omega) = \frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\parallel}^{1/2}(k, \omega)\sigma_{\perp}^{-1/2}(k, \omega)\left(\vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\right)
$$

$$
\pm \gamma^{1/2}(k, \omega)\sigma_{\perp}^{-1/2}(k, \omega)\mathbf{k} \times \vec{I}
$$
(78)

<span id="page-7-3"></span> $[V^{-1}(\mathbf{k}, \omega) = V^+(\mathbf{k}, \omega)]$ . Hence  $K'(\mathbf{k}, \omega)$  also yields, according to Eq. ([64](#page-6-0)), a valid integral kernel  $K'(\mathbf{r}, \mathbf{r}', \omega)$ ,

$$
\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{K}'(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} , \qquad (79)
$$

<span id="page-7-4"></span>which is related to the integral kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$  according to Eq.  $(28)$  $(28)$  $(28)$ , where

$$
\vec{V}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{V}(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} , \qquad (80)
$$

with the associated operator being unitary. We thus see that the two formulations of the theory based on  $K(\mathbf{r}, \mathbf{r}', \omega)$  and  $\tilde{K}'(\mathbf{r}, \mathbf{r}', \omega)$ , respectively, are unitarily equivalent. Note that  $K'(\mathbf{k}, \omega) \neq K'^{+}(\mathbf{k}, \omega)$ , so that the operator associated with the integral kernel  $K'(\mathbf{r}, \mathbf{r}', \omega)$  is non-Hermitian (as to be expected, see Appendix B). Since the operators associated with  $\hat{K}^{\prime}(\mathbf{r}, \mathbf{r}^{\prime}, \omega)$  [as well as  $\hat{V}(\mathbf{r}, \mathbf{r}^{\prime}, \omega)$ ] and  $\hat{P}_{\parallel(\perp)}(\mathbf{r}, \mathbf{r}^{\prime}, \omega)$  commute, the same projectors may be employed in the two formulations of the theory to introduce projective variables according to Eq.  $(72)$  $(72)$  $(72)$ .

#### *2. Local limit: Magnetodielectric media*

Now let us suppose that  $\sigma_{\parallel}(k,\omega)$  and  $\gamma(k,\omega)$  in Eq. ([73](#page-6-4)) are sufficiently slowly varying functions of *k*, with welldefined and unique long-wavelength limits  $\lim_{k\to 0} \sigma_{\parallel}(k,\omega)$  $= \sigma_{\parallel}(\omega) > 0$  and  $\lim_{k \to 0} \gamma(k, \omega) = \gamma(\omega) > 0$ , so that they may be approximated by these limits under the integral in Eq.  $(61)$  $(61)$  $(61)$  to obtain

<span id="page-7-0"></span>
$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \sigma_{\parallel}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') - \gamma(\omega) \nabla \times [\vec{I} \delta(\mathbf{r} - \mathbf{r}')] \times \nabla'.
$$
\n(81)

It should be pointed out that in the limiting case given by Eq.  $(81)$  $(81)$  $(81)$  the positive definiteness of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  already implies that  $\gamma(\omega)$  must be positive,  $\gamma(\omega) > 0$ ; in the general case as given by Eq. ([61](#page-6-5)) together with Eqs.  $(73)$  $(73)$  $(73)$  and  $(74)$  $(74)$  $(74)$ , the positive definiteness of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  does not automatically restrict  $\gamma(k, \omega)$  to positive values.

In order to see to what type of medium this  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ corresponds, we have to find from Eq.  $(81)$  $(81)$  $(81)$  the full conductivity tensor  $Q(\mathbf{r}, \mathbf{r}', \omega)$ , which is uniquely possible since

Eqs. ([2](#page-1-2)) and ([3](#page-1-3)) are Hilbert transforms of each other (cf. Sec. II). The full conductivity tensor corresponding to Eq.  $(81)$  $(81)$  $(81)$  is thus of the form

<span id="page-7-1"></span>
$$
\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega) = Q^{(1)}(\omega)\widetilde{I}\delta(\mathbf{r} - \mathbf{r}')
$$

$$
-Q^{(2)}(\omega)\nabla \times [\widetilde{I}\delta(\mathbf{r} - \mathbf{r}')] \times \widetilde{\nabla}', \quad (82)
$$

where  $Q^{(1)}(\omega)$  and  $Q^{(2)}(\omega)$  are (Fourier-transformed) response functions, both of which are determined by their respective real parts  $\sigma_{\parallel}(\omega)$  and  $\gamma(\omega)$ . Inserting Eq. ([82](#page-7-1)) into Eq.  $(1)$  $(1)$  $(1)$  and comparing with

<span id="page-7-6"></span>
$$
\hat{\mathbf{j}}(\mathbf{r}, \omega) = -i\varepsilon_0 \omega[\varepsilon(\omega) - 1]\hat{\mathbf{E}}(\mathbf{r}, \omega) \n+ \kappa_0 \nabla \times \{[1 - \kappa(\omega)]\hat{\mathbf{B}}(\mathbf{r}, \omega)\} + \hat{\mathbf{j}}_N(\mathbf{r}, \omega)
$$
(83)

 $[\hat{\mathbf{B}}(\mathbf{r}, \omega) = (i\omega)^{-1} \nabla \times \hat{\mathbf{E}}(\mathbf{r}, \omega)]$ , which is the well-known description of a locally responding (homogeneous) magnetodielectric medium, we can make the identifications

$$
Q^{(1)}(\omega) = -i\varepsilon_0 \omega[\varepsilon(\omega) - 1] \tag{84}
$$

and

$$
Q^{(2)}(\omega) = -i\kappa_0[1 - \kappa(\omega)]/\omega, \qquad (85)
$$

where  $\varepsilon(\omega)$  is the permittivity and  $\mu(\omega) = \kappa^{-1}(\omega)$  the (paramagnetic) permeability of the medium  $(\mu_0 = \kappa_0^{-1})$ . For real  $\omega$ , we thus obtain

$$
\sigma_{\parallel}(\omega) = \varepsilon_0 \omega \operatorname{Im} \varepsilon(\omega) \tag{86}
$$

<span id="page-7-2"></span>and

$$
\gamma(\omega) = -\kappa_0 \operatorname{Im} \kappa(\omega)/\omega. \tag{87}
$$

Note that, because of  $\gamma(\omega) > 0$ , from Eq. ([87](#page-7-2)) it follows that Im  $\kappa(\omega)$  < 0 for  $\omega$  > 0, from which it can be shown that  $\mu(\omega \rightarrow 0) > 1.$ 

At first glance, one might believe (erroneously) that not only paramagnetic  $[\mu(\omega \rightarrow 0) > 1]$  but also diamagnetic  $\left[\mu(\omega) \rightarrow 0\right)$   $\leq$  1] features of a medium (or the combined effect of both) can be consistently described by means of the magnetic permeability  $\mu(\omega)$  which is included, as seen above, in the basic linear-response constitutive relation ([1](#page-1-0)). However, since diamagnetism is basically a nonlinear effect (as the underlying microscopic Hamiltonian is quadratic in the magnetic induction field), it is beyond the scope of linear response theory. If it is desired to include diamagnetic media in the framework of linear electrodynamics nevertheless, one can regard the magnetic field on which the diamagnetic susceptibility depends as being (the mean value of) an externally controlled field independent of the dynamical variables. Note that the Onsager reciprocity theorem needs to be stated in its generalized form in this case, see Refs.  $[7,14]$  $[7,14]$  $[7,14]$  $[7,14]$ . For a more satisfactory account of diamagnetic media, one should, however, resort to a nonlinear response formalism, or to a more microscopic theory.

An obvious solution to Eq. ([18](#page-2-8)) with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  given by Eq.  $(81)$  $(81)$  $(81)$  is provided by

<span id="page-8-0"></span>
$$
\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) = \sigma_{\parallel}^{1/2}(\omega)\vec{I}\delta(\mathbf{r} - \mathbf{r}') \mp i\gamma^{1/2}(\omega)\,\nabla \times \vec{I}\delta(\mathbf{r} - \mathbf{r}'),
$$
\n(88)

which corresponds to the kernel  $(79)$  $(79)$  $(79)$  [together with Eq.  $(75)$  $(75)$  $(75)$ ] when, for an isotropic medium,  $\sigma_{\parallel}(k,\omega)$  and  $\gamma(k,\omega)$  are approximated by  $\sigma_{\parallel}(\omega)$  and  $\gamma(\omega)$ , respectively. The kernel ([79](#page-7-3)) [together with Eq.  $(75)$  $(75)$  $(75)$ ] fits well here since it depends in a particularly simple way on those quantities that we have assumed to approach well-defined limits in the derivation that led to Eq.  $(81)$  $(81)$  $(81)$ , a property which can be attributed to the responsible transformation ([80](#page-7-4)) [together with Eq. ([78](#page-7-5))]. In contrast, the kernel obtained directly from Eq. ([64](#page-6-0)) [together with Eq. ([70](#page-6-8))], by first eliminating  $\sigma_{\perp}(k,\omega)$  by means of Eq. ([74](#page-6-6)) and then approximating  $\sigma_{\parallel}(k,\omega) \mapsto \sigma_{\parallel}(\omega)$  and  $\gamma(k,\omega) \mapsto \gamma(\omega)$ , does not provide an alternative to Eq. ([88](#page-8-0)), as it does not lead to Eq.  $(81)$  $(81)$  $(81)$  when inserted in Eq.  $(18)$  $(18)$  $(18)$ ; it corresponds to a different medium. Correspondingly, the kernel  $V(\mathbf{r}, \mathbf{r}', \omega)$  obtained by expressing Eq. ([80](#page-7-4)) [with Eq. ([78](#page-7-5))] in terms of  $\sigma_{\parallel}(k,\omega)$  and  $\gamma(k,\omega)$  and then approximating them by  $\sigma_{\parallel}(\omega)$  and  $\gamma(\omega)$ , respectively, fails to be unitary.

Substituting for  $K(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. ([17](#page-2-6))  $K'(\mathbf{r}, \mathbf{r}', \omega)$  as given by Eq.  $(88)$  $(88)$  $(88)$  [together with Eqs.  $(86)$  $(86)$  $(86)$  and  $(87)$  $(87)$  $(87)$ ], we may explicitly express the noise current density in terms of the bosonic dynamical variables to obtain

<span id="page-8-9"></span>
$$
\hat{\mathbf{j}}_{\mathcal{N}}(\mathbf{r}, \omega) = \left(\frac{\hbar \varepsilon_0}{\pi}\right)^{1/2} \sqrt{\omega^2 \operatorname{Im} \varepsilon(\omega)} \hat{\mathbf{f}}(\mathbf{r}, \omega)
$$
\n
$$
= i \left(\frac{\hbar \kappa_0}{\pi}\right)^{1/2} \nabla \times \left[\sqrt{-\operatorname{Im} \kappa(\omega)} \hat{\mathbf{f}}(\mathbf{r}, \omega)\right]. \quad (89)
$$

Since the operators associated with the projection kernels  $(71)$  $(71)$  $(71)$  commute with the operators associated with Eqs.  $(88)$  $(88)$  $(88)$ and  $(81)$  $(81)$  $(81)$ , one may introduce the projective variables  $\hat{\mathbf{f}}_{\parallel(\perp)}(\mathbf{r},\omega)$  defined by Eq. ([72](#page-6-3)), which corresponds to a decomposition of  $\hat{\mathbf{j}}_N(\mathbf{r}, \omega)$  into longitudinal and transverse parts,

$$
\hat{\mathbf{j}}_{N}(\mathbf{r},\omega) = \hat{\mathbf{j}}_{N\parallel}(\mathbf{r},\omega) + \hat{\mathbf{j}}_{N\perp}(\mathbf{r},\omega),
$$
\n(90)

<span id="page-8-1"></span>where

$$
\hat{\mathbf{j}}_{\text{N}\parallel}(\mathbf{r},\omega) = \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{1/2} \sqrt{\omega^2 \operatorname{Im}\varepsilon(\omega)} \hat{\mathbf{f}}_{\parallel}(\mathbf{r},\omega),\tag{91}
$$

<span id="page-8-2"></span>
$$
\hat{\mathbf{j}}_{N\perp}(\mathbf{r},\omega) = \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{1/2} \sqrt{\omega^2 \operatorname{Im}\,\varepsilon(\omega)} \hat{\mathbf{f}}_{\perp}(\mathbf{r},\omega)
$$
\n
$$
= i \left(\frac{\hbar\kappa_0}{\pi}\right)^{1/2} \nabla \times \left[\sqrt{-\operatorname{Im}\,\kappa(\omega)} \hat{\mathbf{f}}_{\perp}(\mathbf{r},\omega)\right].
$$
\n(92)

Making use of Eq. ([39](#page-4-10)) and identifying therein the projection kernels  $\tilde{P}_\lambda(\mathbf{r}, \mathbf{r}', \omega)$  with  $\tilde{\Delta}_{\parallel(\perp)}(\mathbf{r} - \mathbf{r}'),$  one may then proceed as described in Sec. III and regard the projective variables  $\hat{\mathbf{f}}_{\parallel}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_{\perp}(\mathbf{r}, \omega)$  as being two independent sets of bosonic variables.

Let us briefly make contact with the quantization scheme described in Refs. [[19](#page-21-12)[,21](#page-21-13)], where  $\hat{\mathbf{j}}_N(\mathbf{r}, \omega)$  is decomposed according to

$$
\hat{\mathbf{j}}_{N}(\mathbf{r},\omega) = \hat{\mathbf{j}}_{Ne}(\mathbf{r},\omega) + \hat{\mathbf{j}}_{Nm}(\mathbf{r},\omega),
$$
\n(93)

<span id="page-8-5"></span><span id="page-8-3"></span>with

$$
\hat{\mathbf{j}}_{Ne}(\mathbf{r},\omega) = \left(\frac{\hbar \varepsilon_0}{\pi}\right)^{1/2} \sqrt{\omega^2 \operatorname{Im} \varepsilon(\omega)} \hat{\mathbf{f}}_e(\mathbf{r},\omega),\tag{94}
$$

<span id="page-8-4"></span>
$$
\hat{\mathbf{j}}_{Nm}(\mathbf{r},\omega) = \mp i \left(\frac{\hbar \kappa_0}{\pi}\right)^{1/2} \nabla \times \left[\sqrt{-\operatorname{Im} \kappa(\omega)} \hat{\mathbf{f}}_m(\mathbf{r},\omega)\right].
$$
\n(95)

The connection between Eqs.  $(90)$  $(90)$  $(90)$ – $(92)$  $(92)$  $(92)$  and Eqs.  $(93)$  $(93)$  $(93)$ – $(95)$  $(95)$  $(95)$  is given by a gauge transformation (cf. Sec. III), which effectively redistributes the first term of Eq.  $(92)$  $(92)$  $(92)$ . It is not difficult to prove that the total noise current as given by Eq.  $(93)$  $(93)$  $(93)$ satisfies the correct commutation relation  $(10)$  $(10)$  $(10)$  [with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  from Eq. ([81](#page-7-0)) together with Eqs. ([86](#page-7-1)) and ([87](#page-7-2))] if  $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$  are regarded as two independent sets of bosonic variables. Since  $\hat{\mathbf{j}}_{Ne}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{j}}_{Nm}(\mathbf{r}, \omega)$  can be linearly related to  $\hat{\mathbf{j}}_{\text{N}\parallel}(\mathbf{r},\omega)$  and  $\hat{\mathbf{j}}_{\text{N}\perp}(\mathbf{r},\omega)$  and thus to  $\hat{\mathbf{j}}_{\text{N}}(\mathbf{r},\omega)$ , the variables  $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$  may be viewed as projective variables in the sense outlined at the end of Sec. III. Since Eq.  $(93)$  $(93)$  $(93)$  [with Eqs.  $(94)$  $(94)$  $(94)$  and  $(95)$  $(95)$  $(95)$ ] is a separation of the noise current into a part attributed to a dielectric polarization and a part attributed to a (paramagnetic) magnetization, the quantization scheme based on Eqs. ([93](#page-8-3))–([95](#page-8-4)), with  $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$ and  $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$  being bosonic variables, may be thought of as following from the general quantization scheme in the case where magnetoelectric crossing effects can be *a priori* excluded from consideration.

## *3. Local limit: Other kinds of media*

The transition to the local limit is not a unique procedure in general. Various kinds of locally responding (homogeneous) media, including nonisotropic ones, may therefore be derived as limiting cases from Eq. ([61](#page-6-5)). To illustrate this, let us represent  $\vec{\sigma}(\mathbf{k}, \omega)$  as given in Eq. ([62](#page-6-10)) in a different orthonormal basis, where the new expansion will be nondiagonal in general,

$$
\vec{\sigma}(\mathbf{k},\omega) = \sum_{i,j=1}^{3} \tilde{\sigma}_{ij}(\mathbf{k},\omega) \tilde{\mathbf{e}}_i(\mathbf{k},\omega) \tilde{\mathbf{e}}_j^*(\mathbf{k},\omega).
$$
(96)

<span id="page-8-7"></span><span id="page-8-6"></span>The new basis vectors  $\tilde{\mathbf{e}}_i(\mathbf{k}, \omega)$  are related to the ones appearing in Eq.  $(62)$  $(62)$  $(62)$  by a unitary transformation,

$$
\widetilde{\mathbf{e}}_i(\mathbf{k}, \omega) = \sum_{k=1}^3 U_{ik}(\mathbf{k}, \omega) \mathbf{e}_k(\mathbf{k}, \omega),
$$
 (97)

$$
U_{ik}(\mathbf{k},\omega) = \widetilde{\mathbf{e}}_i(\mathbf{k},\omega) \cdot \mathbf{e}_k^*(\mathbf{k},\omega).
$$
 (98)

<span id="page-8-8"></span>We may always choose the  $\tilde{\mathbf{e}}_i(\mathbf{k}, \omega)$  so that they are independent of **k**,  $\tilde{\mathbf{e}}_i(\mathbf{k}, \omega) \mapsto \tilde{\mathbf{e}}_i(\omega)$ . If this choice can be made such that the new expansion coefficients,

$$
\tilde{\sigma}_{ij}(\mathbf{k},\omega) = \sum_{k,l=1}^{3} U_{ik}^{*}(\mathbf{k},\omega)\sigma_{kl}(\mathbf{k},\omega)U_{jl}(\mathbf{k},\omega),
$$
 (99)

may be approximately replaced under the **k**-integral according to

$$
\tilde{\sigma}_{ij}(\mathbf{k}, \omega) \mapsto \tilde{\sigma}_{ij}(\mathbf{k} \to 0, \omega) \equiv \tilde{\sigma}_{ij}(\omega) \tag{100}
$$

<span id="page-9-0"></span>when Eq.  $(96)$  $(96)$  $(96)$  is inserted in Eq.  $(61)$  $(61)$  $(61)$ , then in this way the type of locally responding (homogeneous) anisotropic medium defined by Eq.  $(51)$  $(51)$  $(51)$  is recovered. [Equation  $(52)$  $(52)$  $(52)$  is then obtained by diagonalizing  $\tilde{\sigma}_{ij}(\omega)$  by means of yet another (k-independent) unitary transformation.] Similarly, if the ap-proximation ([100](#page-9-0)) is generalized to include further terms of an (assumed) expansion of  $\tilde{\sigma}_{ij}(\mathbf{k}, \omega)$  at  $\mathbf{k} = 0$ , then quasilocal approximations of Eq.  $(61)$  $(61)$  $(61)$  are generated by inserting the truncated expansion into Eq.  $(61)$  $(61)$  $(61)$  and integrating term by term to yield a linear combination of various derivatives of  $\delta$ -functions. In pursuing such approximation procedures whose validity is to be examined in each case and which depends crucially on the choice of the transformations ([97](#page-8-7)) and ([98](#page-8-8)) (i.e., on the choice of the new basis vectors)—it must be kept in mind that any approximate form of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  so derived has to conform to all the general requirements on  $Q(\mathbf{r}, \mathbf{r}', \omega)$ .

In this context, let us address so-called bianisotropic media—the most general kind of locally responding linear media. From the above, such a medium may be viewed as corresponding to a quasilocal approximation of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ [and hence of  $\tilde{Q}(\mathbf{r}, \mathbf{r}', \omega)$ ] that incorporates derivatives of  $\delta$ -functions up to second order. In fact, the constitutive relations, which in classical electrodynamics, for homogeneous bianisotropic media, are typically given in a form such as (see, e.g., Refs. [[7,](#page-21-1)[23](#page-21-16)])

<span id="page-9-3"></span>
$$
\underline{\mathbf{P}}(\mathbf{r},\omega) = \overline{\xi}_{PE}(\omega) \cdot \underline{\mathbf{E}}(\mathbf{r},\omega) + \overline{\xi}_{PB}(\omega) \cdot \underline{\mathbf{B}}(\mathbf{r},\omega), \quad (101)
$$

$$
\underline{\mathbf{M}}(\mathbf{r},\omega) = \overline{\xi}_{ME}(\omega) \cdot \underline{\mathbf{E}}(\mathbf{r},\omega) + \overline{\xi}_{MB}(\omega) \cdot \underline{\mathbf{B}}(\mathbf{r},\omega), \quad (102)
$$

<span id="page-9-2"></span>with  $P(r, \omega)$  and  $M(r, \omega)$ , respectively, being the polarization and magnetization fields in the frequency domain, may be equivalently given in the form of the first term of Eq.  $(1)$  $(1)$  $(1)$ , where the appropriate conductivity tensor reads

<span id="page-9-1"></span>
$$
\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega) = -i\omega \widetilde{\xi}_{PE}(\omega) \delta(\mathbf{r} - \mathbf{r}') + [\widetilde{\xi}_{PB}(\omega) \delta(\mathbf{r} - \mathbf{r}')] \times \widetilde{\nabla}'
$$

$$
+ \nabla \times [\widetilde{\xi}_{ME}(\omega) \delta(\mathbf{r} - \mathbf{r}')]
$$

$$
- (i\omega)^{-1} \nabla \times [\widetilde{\xi}_{MB}(\omega) \delta(\mathbf{r} - \mathbf{r}')] \times \widetilde{\nabla}', \quad (103)
$$

and the polarization and magnetization current densities in the frequency domain,  $-i\omega P(\mathbf{r}, \omega)$  and  $\nabla \times \mathbf{M}(\mathbf{r}, \omega)$ , respectively, are combined into  $\mathbf{j}(\mathbf{r}, \omega)$ . It is not difficult to prove that the validity of the reciprocity property of  $\hat{\varrho}(\mathbf{r}, \mathbf{r}', \omega)$  as given in Eq.  $(103)$  $(103)$  $(103)$  is equivalent to the validity of the transposition relations  $\overline{\xi}_{PE}(\omega) = \overline{\xi}_{PE}^{T}(\omega)$ ,  $\overline{\xi}_{MB}(\omega) = \overline{\xi}_{MB}^{T}(\omega)$ , and  $\widetilde{\xi}_{ME}(\omega) = -\overline{\xi_{PB}^{\mathsf{T}}}(\omega).$ 

If the last (magneticlike) term on the right-hand side of Eq. ([103](#page-9-1)) can be omitted, then one can entirely drop Eq. <span id="page-9-4"></span> $(102)$  $(102)$  $(102)$  and change instead Eq.  $(101)$  $(101)$  $(101)$  so as to read

$$
\mathbf{P}(\mathbf{r}, \omega) = \vec{\xi}_{PE}(\omega) \cdot \mathbf{E}(\mathbf{r}, \omega) + \frac{1}{i\omega} \{\vec{\xi}_{PB}(\omega) \cdot [\nabla \times \mathbf{E}(\mathbf{r}, \omega)] - \nabla \times [\vec{\xi}_{ME}(\omega) \cdot \mathbf{E}(\mathbf{r}, \omega)]\}.
$$
\n(104)

Introducing a third-rank tensor with Cartesian components (antisymmetric in the first two indices)

$$
\zeta_{ijk}(\omega) = (i\omega)^{-1} [\xi_{PBil}(\omega) \epsilon_{jlk} + \xi_{MElj}(\omega) \epsilon_{ilk}] \qquad (105)
$$

 $(\epsilon_{ilk}$ , Levi-Civita permutation symbol; summation over indi-ces occurring twice is understood), we may rewrite Eq. ([104](#page-9-4)) as

$$
P_i(\mathbf{r}, \omega) = \xi_{PEij} E_j(\mathbf{r}, \omega) + \zeta_{ijk}(\omega) \partial_k E_j(\mathbf{r}, \omega), \qquad (106)
$$

which exactly corresponds to the standard form commonly used (see, e.g., Ref.  $[24]$  $[24]$  $[24]$ ).

#### **C. Spatially dispersive inhomogeneous media**

As already mentioned, knowledge of the medium properties, i.e., of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ , is required in order to solve the eigenvalue problem  $(23)$  $(23)$  $(23)$  and to explicitly perform the quantization of the medium-assisted electromagnetic field—a task which, in general, cannot be accomplished in closed form. Nevertheless, to provide some analytical insight into the problem, let us consider media that combine the features of the media considered in Secs. IV A and IV B in an approximate fashion.

## *1. Model*

We assume that the medium permits one to clearly distinguish between the length scales associated with spatial dispersion and inhomogeneity, with the former scale being sufficiently small as compared with the latter one. In this case, the medium can be regarded as having locally the properties of bulk material, and  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  may be approximated as

<span id="page-9-5"></span>
$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{\Omega} \sum_{\mathbf{L}} \sum_{i=1}^{3} \sum_{\mathbf{k}} \sigma_{i\mathbf{L}\mathbf{k}}(\omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \times \theta_{\mathbf{L}}(\mathbf{r}) \theta_{\mathbf{L}}(\mathbf{r}') \mathbf{e}_{i\mathbf{L}\mathbf{k}}(\omega) \mathbf{e}_{i\mathbf{L}\mathbf{k}}^{*}(\omega), \quad (107)
$$

from which the eigenfunctions of the associated operator are seen to be

$$
\mathbf{F}_{i\mathbf{L}\mathbf{k}}(\mathbf{r},\omega) = \Omega^{-1/2} \theta_{\mathbf{L}}(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}} \mathbf{e}_{i\mathbf{L}\mathbf{k}}(\omega). \quad (108)
$$

<span id="page-9-7"></span>Here the medium is thought of as being divided into unit cells of volume  $\Omega$  which form a Bravais-type lattice, the cutoff function  $\theta_{\text{L}}(\textbf{r})$  is unity if **r** is in the cell of lattice vector **L** and zero otherwise,  $e_{iLk}(\omega)$  are, for chosen **L**, **k**, and  $\omega$ , a triplet  $(i=1,2,3)$  of orthogonal unit vectors, and the wave vector **k** runs over the reciprocal lattice. Note that, for each cell **L**,

$$
\vec{\sigma}_{Lk}(\omega) = \sum_{i=1}^{3} \sigma_{iLk}(\omega) \mathbf{e}_{iLk}(\omega) \mathbf{e}_{iLk}^{*}(\omega)
$$
(109)

<span id="page-9-6"></span>corresponds to the diagonal form in the  $(\mathbf{k}, \omega)$  domain of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  for bulk material [cf. Eq. ([62](#page-6-10))].

The main features of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  as given in Eq. ([107](#page-9-5)) can be summarized as follows. (i)  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is zero whenever **r** and **r**' are not in the same cell, so that  $\Omega^{1/3}$  determines the length scale on which spatial dispersion is at most observed. (ii) The dependence on **L** of  $\vec{\sigma}_{Lk}(\omega)$  [Eq. ([109](#page-9-6))] for an inhomogeneous medium introduces an **L**-dependence into Eq.  $(107)$  $(107)$  $(107)$  which should be sufficiently weak, so that noticeable violations of the translational invariance of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  may occur only on a length scale that is large compared with  $\Omega^{1/3}$ . Needless to say, the main features do not essentially change if  $\theta_{\text{L}}(\textbf{r})$  is replaced with another—but qualitatively similar cutoff function.

Let us denote by  $L(r)$  the particular lattice vector whose cell contains the point **r**, so that  $L(r)$  plays the role of a coarse-grained position variable. With the notations  $\theta_{\mathbf{L}(\mathbf{r})}(\mathbf{r}') \mapsto \theta(\mathbf{L}(\mathbf{r}), \mathbf{r}')$  and  $\overrightarrow{\sigma}_{\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega) \mapsto \overrightarrow{\sigma}_{\mathbf{k}}(\mathbf{L}(\mathbf{r}), \omega)$ , Eq.  $(107)$  $(107)$  $(107)$  together with Eq.  $(109)$  $(109)$  $(109)$  can be rewritten as

$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \theta(\mathbf{L}(\mathbf{r}'), \mathbf{r}) \vec{\sigma}(\mathbf{L}(\mathbf{r}'), \mathbf{r} - \mathbf{r}', \omega), \qquad (110)
$$

<span id="page-10-1"></span><span id="page-10-0"></span>with

$$
\vec{\sigma}(\mathbf{L}(\mathbf{r}'), \mathbf{r} - \mathbf{r}', \omega) = \frac{1}{\Omega} \sum_{\mathbf{k}} \vec{\sigma}_{\mathbf{k}}(\mathbf{L}(\mathbf{r}'), \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}.
$$
\n(111)

Note that for arbitrary (continuous) values s, the function  $\theta$ (s, r) can be regarded as being symmetric. Using Eq.  $(108)$  $(108)$  $(108)$ , we find that Eq.  $(27)$  $(27)$  $(27)$  takes the form

$$
\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\theta(\mathbf{L}(\mathbf{r}'), \mathbf{r})}{\Omega} \sum_{\mathbf{k}} \vec{K}_{\mathbf{k}}(\mathbf{L}(\mathbf{r}'), \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')},\tag{112}
$$

where

$$
\vec{K}_{\mathbf{k}}(\mathbf{L}(\mathbf{r}),\omega) = \sum_{i=1}^{3} \sigma_{i\mathbf{k}}^{1/2}(\mathbf{L}(\mathbf{r}),\omega) \mathbf{e}_{i\mathbf{k}}(\mathbf{L}(\mathbf{r}),\omega) \mathbf{e}_{i\mathbf{k}}^{*}(\mathbf{L}(\mathbf{r}),\omega),
$$
\n(113)

 $[\sigma_{iL(r)k}(\omega) \mapsto \sigma_{i k}(L(r), \omega), e_{iL(r)k}(\omega) \mapsto e_{i k}(L(r), \omega)].$ 

It can be shown that Eq.  $(110)$  $(110)$  $(110)$  [with Eq.  $(111)$  $(111)$  $(111)$ ] indeed contains (and, in a sense, interpolates) the two limiting cases studied in Secs. IV A and IV B. For the proof, we observe that in the case of negligible spatial dispersion, the cell size can be shrunk to zero,  $\Omega \rightarrow 0$ , so that the lattice vectors take on continuous values,  $L(\mathbf{r}) \rightarrow \mathbf{r}$ . As the lattice becomes finer and finer, the reciprocal lattice becomes more and more coarse, and, for **r** and **r**' unequal but in the same cell, all the points of the reciprocal lattice with  $\mathbf{k} \neq 0$  give rise to rapidly oscillating terms in Eq. ([111](#page-10-1)). In the limit  $\Omega \rightarrow 0$ , these terms oscillate infinitely rapidly and average to zero (when applying the operator associated with Eq.  $(110)$  $(110)$  $(110)$  [with Eq.  $(111)$  $(111)$  $(111)$ ] to any reasonable function), so that they may be set equal to zero. Taking also into account that  $\theta(L(\mathbf{r}'), \mathbf{r})/\Omega \rightarrow \delta(\mathbf{r}')$  $-\mathbf{r}'$ ) in this limit, we see that Eq. ([110](#page-10-0)) [with Eq. ([111](#page-10-1))]

indeed approaches Eq.  $(51)$  $(51)$  $(51)$  for vanishing spatial dispersion [note the correspondences  $\sigma_{ik=0}(\mathbf{L}(\mathbf{r}) = \mathbf{r}, \omega) = \sigma_i(\mathbf{r}, \omega)$  and  $\mathbf{e}_{i\mathbf{k}=0}(\mathbf{L}(\mathbf{r})\mathbf{=} \mathbf{r},\omega) \mathbf{=} \mathbf{e}_i(\mathbf{r},\omega)$ .

On the other hand, in the limiting case of an infinitely extended homogeneous medium, there is no **L**-dependence of the medium properties so that we are free to increase the cell size indefinitely,  $\Omega \rightarrow \infty$ . Consequently, we may let  $\theta(L(\mathbf{r}'), \mathbf{r}) \rightarrow 1$  in Eq. ([110](#page-10-0)) and  $\vec{\sigma}_{k}(L(\mathbf{r}), \omega) \rightarrow \vec{\sigma}(\mathbf{k}, \omega)$ ,  $\Omega^{-1}\Sigma_{\mathbf{k}} \rightarrow (2\pi)^{-3} \int d^3k$  in Eq. ([111](#page-10-1)), which reveals that Eq.  $(110)$  $(110)$  $(110)$  [with Eq.  $(111)$  $(111)$  $(111)$ ] approaches Eq.  $(61)$  $(61)$  $(61)$  as expected.

#### *2. Magnetodielectric media*

To quantize the electromagnetic field in a inhomogeneous magnetodielectric medium specified in terms of  $\varepsilon(\mathbf{r}, \omega)$  and  $\kappa(\mathbf{r}, \omega) = \mu^{-1}(\mathbf{r}, \omega)$ , let us consider a medium that is both sufficiently weakly inhomogeneous and sufficiently weakly spatially dispersive, so that  $\Omega$  in Eq. ([111](#page-10-1)) can be chosen on a scale intermediate between the scales of spatial dispersion and inhomogeneity. We may then approximately let  $L(r)$  be a continuous variable,  $L(r) \rightarrow r$  in Eq. ([110](#page-10-0)), and yet, at the same time, approximately treat the k-sum in Eq. ([111](#page-10-1)) as an integral, so that Eq.  $(110)$  $(110)$  $(110)$  [with Eq.  $(111)$  $(111)$  $(111)$ ] approximates to  $[\theta(\mathbf{L}(\mathbf{r}'), \mathbf{r}) \rightarrow \theta(\mathbf{r}', \mathbf{r})]$ 

<span id="page-10-2"></span>
$$
\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\theta(\mathbf{r}', \mathbf{r})}{(2\pi)^3} \int d^3k \vec{\sigma}(\mathbf{r}', \mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}.
$$
 (114)

For a medium that is locally of the type described by Eq.  $(73)$  $(73)$  $(73)$ , we may set

$$
\overrightarrow{\sigma}(\mathbf{r}, \mathbf{k}, \omega) = \sigma_{\parallel}(\mathbf{r}, k, \omega)\overrightarrow{I} - \mathbf{k} \times \gamma(\mathbf{r}, k, \omega)\overrightarrow{I} \times \mathbf{k}, \quad (115)
$$

where

$$
\gamma(\mathbf{r},k,\omega) = [\sigma_{\perp}(\mathbf{r},k,\omega) - \sigma_{\parallel}(\mathbf{r},k,\omega)]/k^2 > 0. \quad (116)
$$

Assuming that in the **k**-integral in Eq. ([114](#page-10-2)),  $\sigma_{\parallel}(\mathbf{r}, k, \omega)$  and  $\gamma(\mathbf{r}, k, \omega)$  may be approximated, respectively, by welldefined (and unique) long-wavelength limits  $\sigma_{\parallel}(\mathbf{r}, \omega)$  $=\lim_{k\to 0} \sigma_{\parallel}(\mathbf{r}, k, \omega)$  and  $\gamma(\mathbf{r}, \omega) = \lim_{k\to 0} \gamma(\mathbf{r}, k, \omega)$ , the cutoff function  $\theta(\mathbf{r}', \mathbf{r})$  has—due to the rapid oscillations of the exponential for large  $|\mathbf{r}-\mathbf{r}'|$ —no effect with regard to an application of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  from Eq. ([114](#page-10-2))] and can be dropped, and we obtain, as a generalization of Eq.  $(81)$  $(81)$  $(81)$ ,

<span id="page-10-3"></span>
$$
\begin{aligned} \n\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}(\mathbf{r}', \omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ \n&- \nabla \times \left[ \gamma(\mathbf{r}', \omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \right] \times \tilde{\nabla}'. \quad (117) \n\end{aligned}
$$

With the identifications

$$
\sigma_{\parallel}(\mathbf{r}, \omega) = \varepsilon_0 \omega \operatorname{Im} \varepsilon(\mathbf{r}, \omega), \qquad (118)
$$

$$
\gamma(\mathbf{r}, \omega) = -\kappa_0 \operatorname{Im} \kappa(\mathbf{r}, \omega) / \omega \tag{119}
$$

[cf. Eqs.  $(86)$  $(86)$  $(86)$  and  $(87)$  $(87)$  $(87)$ ], Eqs.  $(82)$  $(82)$  $(82)$  and  $(83)$  $(83)$  $(83)$  generalize to

<span id="page-11-1"></span>
$$
\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega) = -i\varepsilon_0 \omega \left[\varepsilon(\mathbf{r}', \omega) - 1\right] \widetilde{I} \delta(\mathbf{r} - \mathbf{r}')
$$

$$
-\frac{1}{i\omega} \nabla \times \left\{\kappa_0 \left[1 - \kappa(\mathbf{r}', \omega)\right] \widetilde{I} \delta(\mathbf{r} - \mathbf{r}')\right\} \times \widetilde{\nabla}'
$$
(120)

and

$$
\hat{\mathbf{j}}(\mathbf{r}, \omega) = -ie_0\omega[\varepsilon(\mathbf{r}, \omega) - 1]\hat{\mathbf{E}}(\mathbf{r}, \omega) \n+ \kappa_0 \nabla \times \{[1 - \kappa(\mathbf{r}, \omega)]\hat{\mathbf{B}}(\mathbf{r}, \omega)\} + \mathbf{j}_N(\mathbf{r}, \omega),
$$
\n(121)

respectively.

Unfortunately, Eq. ([88](#page-8-0)) does not generalize to

<span id="page-11-0"></span>
$$
\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) = \sigma_{\parallel}^{1/2}(\mathbf{r}', \omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}')
$$
  
\n
$$
\equiv i \gamma^{1/2}(\mathbf{r}', \omega) \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}') \quad \text{(wrong)},
$$
\n(122)

as could have been suspected. Indeed, straightforward calculation shows that, for spatially varying permittivity and permeability, the kernel  $(122)$  $(122)$  $(122)$  does not solve Eq.  $(18)$  $(18)$  $(18)$  [with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  as given in Eq. ([117](#page-10-3))], which implies that  $\hat{\mathbf{j}}_{N}(\mathbf{r}, \omega)$ cannot be related to the variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  as in Eq. ([89](#page-8-9)), with  $\varepsilon(\omega)$  and  $\kappa(\omega)$  being simply replaced with their inhomogeneous counterparts  $\varepsilon(\mathbf{r}, \omega)$  and  $\kappa(\mathbf{r}, \omega)$ , respectively. In order to obtain an explicit expression for the kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$ required in Eq.  $(17)$  $(17)$  $(17)$ , one has instead to return to Eq.  $(18)$  $(18)$  $(18)$  and solve it with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  from Eq. ([117](#page-10-3))—a problem that is, however, very difficult to solve in general. Although this does not at all limit the practical applicability of the theory [since all one typically has to know about  $K(\mathbf{r}, \mathbf{r}', \omega)$  is that it satisfies its defining equation  $(18)$  $(18)$  $(18)$  for the chosen conductivity  $(120)$  $(120)$  $(120)$ , it may be useful to have at hand at least an approximate form for weak inhomogeneity, such as (Appen- $\text{dix } D$ 

<span id="page-11-3"></span>
$$
\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \sigma_{\parallel}^{1/2}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \mp i \gamma^{1/2}(\omega) \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}')
$$
  
+ 
$$
\frac{1}{2} [\sigma_{\parallel}(\mathbf{r}, \omega) - \sigma_{\parallel}(\omega)] \vec{M}_0(\mathbf{r}, \mathbf{r}', \omega)
$$
  
+ 
$$
\frac{1}{2} \nabla \times \{ [\gamma(\mathbf{r}, \omega) - \gamma(\omega)] \nabla \times \vec{M}_0(\mathbf{r}, \mathbf{r}', \omega) \},
$$
(123)

<span id="page-11-4"></span>where

$$
\vec{M}_0(\mathbf{r}, \mathbf{r}', \omega) = \sigma_{\parallel}^{-1/2}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}')
$$
  
\n
$$
\pm i \gamma^{-1/2}(\omega) \nabla \times m_0(\mathbf{r}, \mathbf{r}', \omega) \vec{I}
$$
  
\n
$$
+ \sigma_{\parallel}^{-1/2}(\omega) \nabla \times m_0(\mathbf{r}, \mathbf{r}', \omega) \vec{I} \times \tilde{\nabla}',
$$
\n(124)

with  $m_0(\mathbf{r}, \mathbf{r}', \omega) = -(4\pi |\mathbf{r} - \mathbf{r}'|)^{-1} e^{-|\mathbf{r} - \mathbf{r}'|/\alpha(\omega)}$  $, \alpha(\omega)$  $=[\gamma(\omega)/\sigma_{\parallel}(\omega)]^{1/2}$  > 0. If the lack of exact knowledge of

 $K(\mathbf{r}, \mathbf{r}', \omega)$  really happens to be an obstacle in an application, one can alternatively resort to the approach on the basis of Eqs. ([93](#page-8-3))–([95](#page-8-4)) by simply replacing therein  $\varepsilon(\omega)$  and  $\kappa(\omega)$  by  $\varepsilon(\mathbf{r}, \omega)$  and  $\kappa(\mathbf{r}, \omega)$ , respectively.

# **V. GREEN TENSOR CONSTRUCTION FOR SPATIALLY DISPERSIVE BODIES**

Practical application of the quantization scheme requires the solution of the classical problem of the determination of the Green tensor  $G(\mathbf{r}, \mathbf{r}', \omega)$  for a given conductivity tensor  $Q(\mathbf{r}, \mathbf{r}', \omega)$ . Typically, one has to deal with systems of bodies each of which can be regarded as being homogeneous in its respective interior region. The physical surfaces of the bodies, including the boundary surfaces between adjacent bodies, may be said to be the particular space regions where the material properties differ significantly from the intrabody (bulk-material) properties. Physical surfaces are therefore not mathematical ones but more or less fuzzy boundary layers. For not too small bodies, however, they usually contain only a small fraction of the overall material, so that they may often be approximately replaced with (sharp) mathematical surfaces, with the idealization that the intrabody bulkmaterial properties hold immediately beyond them.

## **A. Dielectric approximation**

The application of the point of view just outlined to (systems of internally homogeneous) spatially dispersive bodies is commonly referred to as the dielectric approximation, for which it is obviously required that the characteristic length scale of spatial dispersion is small in comparison with the typical linear extensions of the bodies. Hence making use of the same notation as employed in Eq.  $(110)$  $(110)$  $(110)$ , we assume the conductivity tensor of a system of spatially dispersing bodies, in the dielectric approximation, to be of the form

$$
\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega) = \theta(\mathbf{L}(\mathbf{r}'), \mathbf{r}) \widetilde{Q}(\mathbf{L}(\mathbf{r}'), \mathbf{r} - \mathbf{r}', \omega), \qquad (125)
$$

<span id="page-11-2"></span>with the vector **L** labeling now the bodies in place of the lattice cells in Sec. IV C 1;  $L(r')$  singles out the particular body that contains the position **r**', and the quantities  $Q(\mathbf{L}, \mathbf{r} - \mathbf{r}', \omega)$  are the bulk-material conductivity tensors ascribed to the various bodies. Regions outside all actual bodies, if any, are formally viewed as bodies in this notation.) Note that  $Q(\mathbf{r}, \mathbf{r}', \omega)$  as given by Eq. ([125](#page-11-2)) satisfies the reciprocity condition. Corrections to the dielectric approximation in the form of surface currents or, equivalently, boundary conditions might be required for systems such as needleshaped bodies, thin films, or the like, where the dielectric approximation can be insufficient. However, as such corrections cannot be convincingly justified without detailed (model) assumptions about the specific nature of the physical surfaces, we do not consider them in the following.

#### **B. Integral equations and the surface impedance method**

The dielectric approximation renders it possible to formulate integral equations from which the Green tensor may then be derived. Here we outline the surface impedance method which, in connection with its application to the calculation of Casimir forces, has recently given rise to controversial discussions (see, e.g., Refs.  $[25-28]$  $[25-28]$  $[25-28]$ ) concerning the range of validity and the approximations involved in this method.

To begin with, let us consider a spatially dispersive material described by a conductivity tensor  $\widetilde{Q}(\mathbf{r}, \mathbf{r}', \omega)$  [not yet approximated by a form like Eq.  $(125)$  $(125)$  $(125)$ ], and let *V* denote some space region of interest. Further, let  $\tilde{G}_{aux}(\mathbf{r}, \mathbf{r}', \omega)$  be the Green tensor for an auxiliary problem to be specified yet. Then, if  $\underline{\mathbf{j}}(\mathbf{r}, \omega)$  is an arbitrarily chosen current density in the frequency domain, given inside and/or outside *V*, and if  $\mathbf{E}(\mathbf{r},\omega)$  and  $\mathbf{B}(\mathbf{r},\omega) = (i\omega)^{-1}\nabla \times \mathbf{E}(\mathbf{r},\omega)$ , respectively, are the (classical, frequency-domain) electric and induction fields associated with this current density, the identity

<span id="page-12-1"></span>
$$
\int_{V} d^{3}r \mathbf{E}(\mathbf{r}, \omega) \cdot \nabla \times \vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)
$$
\n
$$
+ i\varepsilon_{0}\omega \int_{V} d^{3}r \mathbf{E}(\mathbf{r}, \omega) \cdot \left[ \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) - (i\varepsilon_{0}\omega)^{-1} \int_{V} d^{3}s \vec{Q}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{s}, \mathbf{r}', \omega) \right]
$$
\n
$$
= \int_{V} d^{3}r \mathbf{j}_{V}(\mathbf{r}, \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)
$$
\n
$$
+ \int_{\partial V} da(\mathbf{r}) [\mathbf{E}(\mathbf{r}, \omega) \times \mathbf{e}_{n}(\mathbf{r})] \cdot \vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)
$$
\n
$$
+ \mu_{0}^{-1} \int_{\partial V} da(\mathbf{r}) [\mathbf{B}(\mathbf{r}, \omega) \times \mathbf{e}_{n}(\mathbf{r})] \cdot \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)
$$
\n(126)

(being a Green-type formula) holds, as can be proven correct by partial integration and employing the reciprocity property of  $Q(\mathbf{r}, \mathbf{s}, \omega)$ . Here,  $da(\mathbf{r})$  and  $\mathbf{e}_n(\mathbf{r})$  denote the absolute value and the unit vector of the surface element at **r** on the surface  $\partial V$  of *V*. Further,

<span id="page-12-0"></span>
$$
\underline{\mathbf{j}}_V(\mathbf{r}, \omega) = (i\mu_0 \omega)^{-1} \left[ \nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \underline{\mathbf{E}}(\mathbf{r}, \omega) - i\mu_0 \omega \int_V d^3r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{E}}(\mathbf{r}', \omega) \right]
$$
(127)

<span id="page-12-6"></span>and

$$
\widetilde{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) = (i\mu_0 \omega)^{-1} \nabla \times \widetilde{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega).
$$
 (128)

Note that  $\mathbf{j}_V(\mathbf{r}, \omega)$  does not agree with  $\mathbf{j}(\mathbf{r}, \omega)$ , in general, as the  $\mathbf{r}'$ -integral in Eq. ([127](#page-12-0)) extends only over *V*.

Let us now assume that  $\hat{Q}(\mathbf{r}, \mathbf{r}', \omega)$  can be treated in the dielectric approximation according to Eq.  $(125)$  $(125)$  $(125)$ , at least for the medium in *V*. In this case,  $\mathbf{j}_V(\mathbf{r}, \omega)$  agrees with  $\mathbf{j}(\mathbf{r}, \omega)$ inside *V*, and  $\hat{Q}(\mathbf{r}, \mathbf{s}, \omega)$  in Eq. ([126](#page-12-1)) can be replaced with the bulk-medium conductivity tensor  $\tilde{Q}(\mathbf{L}_V, \mathbf{r}-\mathbf{s}, \omega)$  attributed to

the medium in  $V$  ( $L_V$  is the vector labeling region  $V$ ). Hence if  $G_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  is required, at least for  $\mathbf{r} \in V$ , to obey the equation

<span id="page-12-2"></span>
$$
\nabla \times \nabla \times \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)
$$

$$
- i\mu_0 \omega \int_V d^3s \vec{Q}(\mathbf{L}_V, \mathbf{r} - \mathbf{s}, \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'),
$$
(129)

<span id="page-12-3"></span>then Eq.  $(126)$  $(126)$  $(126)$  simplifies to

$$
(i\mu_0\omega)^{-1}\mathbf{E}(\mathbf{r}',\omega)\theta(\mathbf{L}_V,\mathbf{r}')
$$
  
\n
$$
= (i\mu_0\omega)^{-1}\mathbf{E}^{(\text{in})}(\mathbf{r}',\omega)
$$
  
\n
$$
+ \int_{\partial V} da(\mathbf{r})[\mathbf{E}(\mathbf{r},\omega) \times \mathbf{e}_n(\mathbf{r})] \cdot \vec{\Gamma}_{\text{aux}}(\mathbf{r},\mathbf{r}',\omega)
$$
  
\n
$$
+ \mu_0^{-1} \int_{\partial V} da(\mathbf{r})[\mathbf{B}(\mathbf{r},\omega) \times \mathbf{e}_n(\mathbf{r})] \cdot \vec{G}_{\text{aux}}(\mathbf{r},\mathbf{r}',\omega),
$$
\n(130)

<span id="page-12-5"></span>where

$$
\mathbf{E}^{(\text{in})}(\mathbf{r}, \omega) = i\mu_0 \omega \int_V d^3r' \mathbf{j}(\mathbf{r}', \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{r}', \mathbf{r}, \omega)
$$

$$
= i\mu_0 \omega \int_V d^3r' \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{j}(\mathbf{r}', \omega). \quad (131)
$$

Since the characteristic length of spatial dispersion is assumed to be sufficiently small as compared with the linear extensions of *V*, we may extend, with little error, the *s*-integral in Eq. ([129](#page-12-2)) to the whole space and, therefore, we may identify  $\ddot{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. ([130](#page-12-3)) with the corresponding bulk-medium Green tensor as given in Appendix E. Note that Eq. ([130](#page-12-3)) may be viewed as a statement of Huyghens' principle (for  $r'$  inside  $V$ ) and of the extinction theorem (for  $\mathbf{r}'$  outside *V*), see Refs.  $[23,29]$  $[23,29]$  $[23,29]$  $[23,29]$ , which have been wellknown as a suitable starting point for field calculations on the basis of integral equation methods, see, in particular, Refs.  $\left[3-5\right]$  $\left[3-5\right]$  $\left[3-5\right]$  where the Wiener-Hopf technique has been used to construct solutions for a particular functional form of  $\ddot{Q}$ **L**<sub>*V*</sub>, **r**−**s**, $\omega$ ).

The surface impedance method (see, e.g., Ref. [[30](#page-21-23)]) consists of the assumption that a linear relation between the tangential field components of  $\mathbf{E}(\mathbf{r}, \omega)$  and  $\mathbf{B}(\mathbf{r}, \omega)$  exists on the surface  $\partial V$ ,

<span id="page-12-4"></span>
$$
\mathbf{e}_n(\mathbf{r}) \times \mathbf{E}(\mathbf{r}, \omega) = \mu_0^{-1} \int_{\partial V} da(\mathbf{r}') [\tilde{Z}(\mathbf{r}, \mathbf{r}', \omega) \times \mathbf{e}_n(\mathbf{r}')] \cdot [\mathbf{B}(\mathbf{r}', \omega) \times \mathbf{e}_n(\mathbf{r}')] \tag{132}
$$

 $(\mathbf{r} \text{ on } \partial V)$ , where the tensor  $\mathbf{Z}(\mathbf{r}, \mathbf{r}', \omega)$  is the dyadic surface impedance. Equivalently, one may consider the inverted form

<span id="page-13-0"></span>
$$
\mu_0^{-1} \underline{\mathbf{B}}(\mathbf{r}, \omega) \times \mathbf{e}_n(\mathbf{r}) = \int_{\partial V} da(\mathbf{r}') [\underline{\mathbf{E}}(\mathbf{r}', \omega) \times \mathbf{e}_n(\mathbf{r}')] \newline \cdot \vec{Y}(\mathbf{r}', \mathbf{r}, \omega) \cdot \vec{I}_n(\mathbf{r}), \tag{133}
$$

where  $\tilde{I}_n(\mathbf{r}) = \tilde{I} - \mathbf{e}_n(\mathbf{r}) \mathbf{e}_n(\mathbf{r})$  is a tangential projector, and  $Y(\mathbf{r}, \mathbf{r}', \omega)$  may be referred to as the surface admittance. Once the relation  $(132)$  $(132)$  $(132)$  [or Eq.  $(133)$  $(133)$  $(133)$ ] has been adopted, one may convert Eq. ([130](#page-12-3)) (with  $\mathbf{r}' \in V$ ) into an integral equation for  $\mathbf{B}(\mathbf{r}, \omega)$  [or  $\mathbf{E}(\mathbf{r}, \omega)$ ] inside *V*, which can then be solved in terms of  $\underline{\mathbf{E}}^{(\text{in})}(\mathbf{r}, \omega)$  and the surface impedance, without specifying the medium properties outside *V*.

At first glance, the method has some advantageous features so that it has been enjoying a reputation in the literature. One of these features is that the necessity to know the medium properties everywhere outside *V* is replaced, so to speak, by the necessity to know the surface impedance for  $\partial V$ , which is a great reduction at first glance. Another one is that the application of continuity conditions may be sidestepped to some extent. Both of these points are to be qualified, however, and must be seen in the context of the following remarks.

First, since the current density  $\mathbf{j}(\mathbf{r}, \omega)$  outside *V* does not contribute to Eq. ([131](#page-12-5)), the solution for  $\mathbf{E}(\mathbf{r}, \omega)$  can be unique only if  $\mathbf{j}(\mathbf{r}, \omega)$  is located completely inside *V*; solutions to the homogeneous integral equation can then be excluded from consideration. The surface impedance method applied to the volume *V* can thus yield, for a given surface impedance, the Green tensor  $G(\mathbf{r}, \mathbf{r}', \omega)$  only for both **r** and **r**' located in *V* (which may be sufficient for many applications, e.g., in the calculation of dispersion forces). Second, Eqs. ([132](#page-12-4)) and ([133](#page-13-0)) implicitly demand that the tangential components of the electric and the induction field uniquely exist on the surface  $\partial V$  (i.e., they should be continuous across the surface), or else ambiguities were encountered. Quasilocal approximations of the conductivity tensor should hence be excluded if they contain contributions more singular than  $\delta$ -functions.

Provided that Eq.  $(132)$  $(132)$  $(132)$  [or Eq.  $(133)$  $(133)$  $(133)$ ] holds, the method requires knowledge of  $Z(\mathbf{r}, \mathbf{r}', \omega)$  [or  $Y(\mathbf{r}, \mathbf{r}', \omega)$ ], which plays the role of an external input. If  $\overrightarrow{Z}(\mathbf{r}, \mathbf{r}', \omega)$  [or  $Y(\mathbf{r}, \mathbf{r}', \omega)$  is not known from the very beginning, one can try to determine it *a posteriori* from the solution found, by appropriately specifying the effect of the medium on the electromagnetic field outside the space region under consideration as well. We proceed with an example where this line can be pursued explicitly, on the basis of the dielectric approximation.

#### **C. Example: Nonmagnetic, planar systems**

Let *V* denote the slablike region between two parallel planes  $z=0$  and  $z=d$ , and let  $\mathbf{j}(\mathbf{r},\omega)$  be located completely inside *V*. In order to apply the surface impedance method to the region  $V$ , we assume that the medium in  $V$  is nonmagnetic and can be treated in the dielectric approximation. Introducing Fourier transforms according to  $[\mathbf{r} = (\boldsymbol{\rho}, z)]$ 

$$
\mathbf{E}(\mathbf{r}, \omega) = \int d^2q e^{i\mathbf{q} \cdot \boldsymbol{p}} \mathbf{E}(z, \mathbf{q}, \omega)
$$
 (134)

and taking into account the lateral translational invariance of the system, we may write Eq.  $(133)$  $(133)$  $(133)$  in the Fourier domain as

<span id="page-13-1"></span>
$$
\mu_0^{-1} \mathbf{B}(z, \mathbf{q}, \omega) \times \mathbf{e}_z = (2\pi)^2 \{ [\mathbf{E}(d, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \widetilde{Y}(d, z, -\mathbf{q}, \omega) - [\mathbf{E}(0, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \widetilde{Y}(0, z, -\mathbf{q}, \omega) \} \cdot \widetilde{I}_z
$$
\n(135)

 $[I_z = I - e_z e_z]$ , where *z* takes on the two values *z*=0 and *z*=*d* (the two terms enter with opposite signs because of the opposite surface normals on the two sides of  $\partial V$ ). Similarly, the Fourier transformed version of Eq. ([130](#page-12-3)) reads

<span id="page-13-2"></span>
$$
\mathbf{E}(z', \mathbf{q}, \omega) \theta(\mathbf{L}_V, z') - \mathbf{E}^{(\text{in})}(z', \mathbf{q}, \omega)
$$
\n
$$
= i\mu_0 \omega(2\pi)^2 \{ [\mathbf{E}(d, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{\Gamma}_{\text{aux}}(d, z', -\mathbf{q}, \omega) \right.
$$
\n
$$
- [\mathbf{E}(0, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{\Gamma}_{\text{aux}}(0, z', -\mathbf{q}, \omega) \right.
$$
\n
$$
+ \mu_0^{-1} [\mathbf{B}(d, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{G}_{\text{aux}}(d, z', -\mathbf{q}, \omega) \left. - \mu_0^{-1} [\mathbf{B}(0, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{G}_{\text{aux}}(0, z', -\mathbf{q}, \omega) \right]. \tag{136}
$$

<span id="page-13-3"></span>Note that  $G_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  and  $\Gamma_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  are translationally invariant because they refer to bulk material. By means of Eq. ([135](#page-13-1)), Eq. ([136](#page-13-2)) takes the form

$$
\mathbf{E}(z', \mathbf{q}, \omega) \theta(\mathbf{L}_V, z') - \mathbf{E}^{(\text{in})}(z', \mathbf{q}, \omega)
$$
  
= 
$$
\mathbf{E}(d, \mathbf{q}, \omega) \cdot \vec{I}_z \cdot \vec{R}(d, z', \mathbf{q}, \omega)
$$
  
- 
$$
\mathbf{E}(0, \mathbf{q}, \omega) \cdot \vec{I}_z \cdot \vec{R}(0, z', \mathbf{q}, \omega),
$$
 (137)

with  $(\zeta=0, \zeta=d)$ 

$$
\overrightarrow{R}(\zeta, z', \mathbf{q}, \omega) = i\mu_0 \omega (2\pi)^2 \mathbf{e}_z \times [\overrightarrow{\Gamma}_1(\zeta, z', -\mathbf{q}, \omega) \n+ (2\pi)^2 \overrightarrow{Y}(\zeta, d, -\mathbf{q}, \omega) \cdot \overrightarrow{I}_z \cdot \overrightarrow{G}_1(d, z', -\mathbf{q}, \omega) \n- (2\pi)^2 \overrightarrow{Y}(\zeta, 0, -\mathbf{q}, \omega) \cdot \overrightarrow{I}_z \cdot \overrightarrow{G}_1(0, z', -\mathbf{q}, \omega)]
$$
\n(138)

[note that  $\mathbf{e}_z \cdot R(\zeta, z', \mathbf{q}, \omega) = 0$ ]. From Eq.  $(137)$  $(137)$  $(137)$  it follows that (Appendix F)

<span id="page-13-4"></span>
$$
\mathbf{E}(0, \mathbf{q}, \omega) \cdot \vec{I}_z = [\mathbf{E}^{(\text{in})}(0 + , \mathbf{q}, \omega) \cdot \vec{C}^{\sharp} \n- \mathbf{E}^{(\text{in})}(d - , \mathbf{q}, \omega) \cdot \vec{D}^{\sharp}] \cdot [\vec{A} \cdot \vec{C}^{\sharp} - \vec{B} \cdot \vec{D}^{\sharp}]^{\sharp},
$$
\n(139)

<span id="page-13-5"></span>
$$
\mathbf{E}(d, \mathbf{q}, \omega) \cdot \vec{I}_z = [\mathbf{E}^{(\text{in})}(0 + , \mathbf{q}, \omega) \cdot \vec{A}^* - \mathbf{E}^{(\text{in})}(d - , \mathbf{q}, \omega) \cdot \vec{B}^*] \cdot [\vec{C} \cdot \vec{A}^* - \vec{D} \cdot \vec{B}^*]^*,
$$
\n(140)

<span id="page-13-6"></span>where

$$
\vec{A} = \vec{A}(\mathbf{q}, \omega) = \vec{I}_z \cdot [\vec{I} + \vec{R}(0, 0 + \mathbf{q}, \omega)] \cdot \vec{I}_z, \qquad (141)
$$

$$
\vec{B} = \vec{B}(\mathbf{q}, \omega) = \vec{I}_z \cdot \vec{R}(0, d - \mathbf{q}, \omega) \cdot \vec{I}_z, \tag{142}
$$

$$
\vec{C} = \vec{C}(\mathbf{q}, \omega) = -\vec{I}_z \cdot \vec{R}(d, 0 + \mathbf{q}, \omega) \cdot \vec{I}_z, \tag{143}
$$

$$
\vec{D} = \vec{D}(\mathbf{q}, \omega) = \vec{I}_z \cdot [\vec{I} - \vec{R}(d, d - \mathbf{q}, \omega)] \cdot \vec{I}_z, \qquad (144)
$$

<span id="page-14-0"></span>and the superscript  $\sharp$  denotes a pseudoinversion operation with respect to the  $I_z$ -space, as explained in Appendix F. The solution to Eq.  $(137)$  $(137)$  $(137)$  can now be readily obtained by substituting Eqs.  $(139)$  $(139)$  $(139)$  and  $(140)$  $(140)$  $(140)$  back in the right-hand side of Eq. ([137](#page-13-3)). Specifying in Eq. ([131](#page-12-5)) the source of  $\mathbf{E}^{(\text{in})}(\mathbf{r}, \omega)$  so as to correspond to a point dipole situated in *V*,  $\mathbf{j}(\mathbf{r}, \omega)$  $=(i\mu_0\omega)^{-1}\mathbf{p}\delta(\mathbf{r}-\mathbf{r}_p)$ , from the corresponding position-space solution  $\underline{\mathbf{E}}(\mathbf{r}, \omega) \theta(\mathbf{L}_V, \mathbf{r})$ , one can then read off, according to  $\mathbf{E}(\mathbf{r}, \omega) \theta(\mathbf{L}_V, \mathbf{r}) = \mathbf{p} \cdot \mathbf{G}(\mathbf{r}_p, \mathbf{r}, \omega) \theta(\mathbf{L}_V, \mathbf{r}) \theta(\mathbf{L}_V, \mathbf{r}_p)$ , the (interesting part of the) Green tensor expressed in terms of the surface admittance, i.e., in terms of the quantities  $\ddot{Y}(z, z', \mathbf{q}, \omega)$  (*z*=0,*d*; *z'*=0,*d*).

In order to illustrate the calculation of the admittance, let us assume that the (nonmagnetic) media to the left and right of  $z=0$  and  $z=d$ , respectively, are (in the sense of the dielectric approximation) homogeneous half-spaces. Using the indices 0, 1, and 2 to distinguish the left  $(z < 0)$  half-space, the volume  $V(0 \le z \le d)$ , and the right half-space  $(z > d)$ , respectively, and associating with each of the three regions the corresponding bulk-medium Green tensor  $G_j(\mathbf{r}, \mathbf{r}', \omega)$  (Appendix E) and the associated auxiliary tensor  $\Gamma_j(\mathbf{r}, \mathbf{r}', \omega)$  defined according to Eq.  $(128)$  $(128)$  $(128)$ , one can derive the admittance by applying the continuity conditions. Introducing the abbreviations

$$
\vec{M}(\mathbf{q},\omega) = -\vec{I}_z \cdot \vec{G}_1(0,d-, \mathbf{q}, \omega) \cdot \vec{I}_z, \tag{145}
$$

<span id="page-14-3"></span>
$$
\widetilde{N}(\mathbf{q},\omega) = -\widetilde{I}_z \cdot [\widetilde{G}_1(0,0+, \mathbf{q},\omega) + \widetilde{G}_0(0,0-, \mathbf{q},\omega)] \cdot \widetilde{I}_z,
$$
\n(146)

$$
\vec{P}(\mathbf{q},\omega) = \vec{I}_z \cdot [\vec{G}_1(d,d-, \mathbf{q},\omega) + \vec{G}_2(0,0+, \mathbf{q},\omega)] \cdot \vec{I}_z,
$$
\n(147)

$$
\vec{S}(\mathbf{q}, \omega) = \vec{I}_z \cdot \vec{G}_1(d, 0 + \mathbf{q}, \omega) \cdot \vec{I}_z, \tag{148}
$$

$$
\vec{T}(\mathbf{q}, \omega) = \vec{I}_z \cdot \vec{\Gamma}_1(0, d - \mathbf{q}, \omega) \cdot \vec{I}_z,\tag{149}
$$

$$
\vec{U}(\mathbf{q},\omega) = \vec{I}_z \cdot [\vec{\Gamma}_1(0,0+, \mathbf{q}, \omega) + \vec{\Gamma}_0(0,0-, \mathbf{q}, \omega)] \cdot \vec{I}_z,
$$
\n(150)

$$
\vec{V}(\mathbf{q},\omega) = -\vec{I}_z \cdot \left[\vec{\Gamma}_1(d,d-\mathbf{q},\omega) + \vec{\Gamma}_2(0,0+\mathbf{q},\omega)\right] \cdot \vec{I}_z,\tag{151}
$$

$$
\widetilde{W}(\mathbf{q},\omega) = -\overrightarrow{I}_z \cdot \overrightarrow{\Gamma}_1(d,0+\mathbf{q},\omega) \cdot \overrightarrow{I}_z,\tag{152}
$$

<span id="page-14-4"></span>one eventually finds (Appendix G) the  $Y(z, z', q, \omega)$  ( $z=0, d;$  $z' = 0, d$  to be (for notational convenience, the arguments **q**,  $\omega$  are suppressed)

<span id="page-14-1"></span>
$$
\overrightarrow{Y}(0,0) = \frac{1}{(2\pi)^2} [\overrightarrow{U} \cdot \overrightarrow{S}^{\sharp} - \overrightarrow{T} \cdot \overrightarrow{P}^{\sharp}] \cdot [\overrightarrow{M} \cdot \overrightarrow{P}^{\sharp} - \overrightarrow{N} \cdot \overrightarrow{S}^{\sharp}]^{\sharp},
$$
\n(153)

$$
\overrightarrow{Y}(0,d) = \frac{1}{(2\pi)^2} [\overrightarrow{U} \cdot \overrightarrow{N}^{\sharp} - \overrightarrow{T} \cdot \overrightarrow{M}^{\sharp}] \cdot [\overrightarrow{P} \cdot \overrightarrow{M}^{\sharp} - \overrightarrow{S} \cdot \overrightarrow{N}^{\sharp}]^{\sharp},
$$
\n(154)

$$
\widetilde{Y}(d,0) = \frac{1}{(2\pi)^2} [\widetilde{V} \cdot \widetilde{P}^* - \widetilde{W} \cdot \widetilde{S}^*] \cdot [\widetilde{M} \cdot \widetilde{P}^* - \widetilde{N} \cdot \widetilde{S}^*]^*,
$$
\n(155)

<span id="page-14-2"></span>
$$
\overleftrightarrow{Y}(d,d) = \frac{1}{(2\pi)^2} [\overrightarrow{V} \cdot \overrightarrow{M}^* - \overrightarrow{W} \cdot \overrightarrow{N}^*] \cdot [\overrightarrow{P} \cdot \overrightarrow{M}^* - \overrightarrow{S} \cdot \overrightarrow{N}^*]^*,
$$
\n(156)

which are, quite naturally, far more complicated than the simple Kliever-Fuchs expressions mentioned in Appendix E.

The surface impedance-based technique as outlined above may be analogously applied to planar multilayer structures composed of nonlocally responding media given the dielectric approximation). A typical problem of methods such as transfer matrix methods (see, e.g., Ref. [[29](#page-21-20)]) or (closely related) techniques based on recursive calculation of generalized reflection and/or transmission coefficients (see, e.g., Refs. [[23,](#page-21-16)[31,](#page-21-24)[32](#page-21-25)]), which are widely used for treating multilayer structures, is that straightforward generalization beyond the most frequently considered case of locally responding dielectric or magnetodielectric media is not easily viable (see, however, Ref.  $[33]$  $[33]$  $[33]$  for an interesting extension). The advantage of surface impedance techniques may be viewed, in this regard, as resulting from the fact that the starting point is an integral-equation formulation of the propagation problem, which applies to nonlocally responding media in nearly the same way as to locally responding ones (given the dielectric approximation). By contrast, the generalization of approaches directly based on Maxwell's differential equations are complicated by the qualitative change of the nature of these equations to integrodifferential equations as soon as nonlocally responding media are brought into play.

## **VI. SUMMARY AND CONCLUDING REMARKS**

We have developed a general quantization scheme for the macroscopic electromagnetic field in arbitrary linearly responding media, which offers a unified approach to QED in linear media. Describing the medium response by a nonlocal conductivity tensor, any of the possible electromagnetic features of a linear medium in equilibrium is covered by the scheme, in particular, spatial dispersion. Central quantities of the scheme are the noise current that is intimately connected with the absorption necessarily observed in any linear medium in equilibrium, the bosonic dynamical variables associated with the noise current, and the Green tensor of the phenomenological Maxwell equations, in which the medium

properties enter via the conductivity tensor. Inclusion in the theory of additional atomic sources that interact with the medium-assisted electromagnetic field may then be straightforwardly performed along standard lines, despite the fact that the "free" field already incorporates the interaction with some background material.

From a careful analysis of the dynamical variables and quasi--local limiting forms of the nonlocal conductivity tensor, we have shown how quantization schemes previously developed for locally responding media can be recovered as special applications of the general quantization scheme. In particular, a locally responding magnetodielectric medium can be viewed as a special quasilocal limiting case of an isotropic, spatially dispersive medium without optical activity, where the (local) dielectric permittivity and magnetic permeability are just two contributions to one and the same quasilocal conductivity tensor. As a result, application of the general quantization scheme shows that the electromagnetic field in such a medium can be quantized by using a single set of bosonic variables.

Generally, the use of a single set of bosonic variables means that the noise current which enters the macroscopic Maxwell equations is not divided into parts (associated, e.g., with a polarization and a magnetization) regarded as representing independent degrees of freedom, but is rather treated as an entity. This may be particularly advantageous for future studies of (quantum) electrodynamics in moving media, simplifying the discussion of transformations to different frames of reference. However, the theory also admits, by appropriate projection, the use of several independent sets of bosonic variables, which in fact corresponds to the neglect of certain kinds of interactions in the sense of superselection rules.

Since exact solutions of Maxwell's equations are not available in closed form in general, even more so if spatial dispersion is taken into account, one has to resort to approximation methods to obtain explicit expressions for the Green tensor, the latter being one of the cornerstones of the theory. To assist in such intentions we have considered in some detail the dielectric approximation for the conductivity tensor, which consists of approximating the conductivity tensor of a system of spatially dispersive bodies by joining together bulk-medium conductivity tensors (which are routinely handled in reciprocal space). Although the information relevant to physical surface regions is lost in this way, the dielectric approximation has been a key tool to render tractable electromagnetic propagation problems in spatially dispersive media. Accepting the dielectric approximation, the problem of finding the Green tensor becomes then solvable via integral-equation and surface-impedance techniques.

As already mentioned, diamagnetic media are not covered by the quantization scheme developed in this paper—a scheme that exhausts the possibilities offered by the linearresponse framework. Furthermore, it should be pointed out that the scheme also does not automatically apply to linearly amplifying media. Although both types of media do not really fit into the linear-response framework, they may be forced into it, but not without reservations and alterations of the whole scheme. Clearly, the concept of linear amplification has a range of applicability very much smaller than that of linear dissipation.

Concluding, this work provides the most general quantization scheme for the electromagnetic field in linearly responding, absorbing materials to date, from which previously given schemes can be recovered as limiting cases. It will serve as a foundation for investigations of surface plasmon effects involving strong spatial dispersion, and as a starting point for the investigation of moving media.

*Note added in proof.* Recently, a paper by Suttorp [[35](#page-21-27)] has appeared where a (Huttner-Barnett-like) model of the electromagnetic field linearly interacting with a spatially dispersive medium modeled in terms of harmonic oscillators is given. The results obtained from this model are in line with our theory, and may be regarded as providing additional support for it from a more microscopic point of view.

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## **APPENDIX A: DERIVATION OF EQ. [\(9\)](#page-2-2)**

<span id="page-15-0"></span>The linear integrodifferential equation  $(8)$  $(8)$  $(8)$  can be represented as

$$
\int d^3s \vec{H}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{G}(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \quad (A1)
$$

<span id="page-15-4"></span>where the integral kernel

$$
\vec{H}(\mathbf{r}, \mathbf{r}', \omega) = \nabla \times \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}') - \frac{\omega^2}{c^2} \vec{I} \delta(\mathbf{r} - \mathbf{r}')
$$

$$
-i\mu_0 \omega \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \tag{A2}
$$

is reciprocal,

$$
\vec{H}(\mathbf{r}, \mathbf{r}', \omega) = \vec{H}^{\mathsf{T}}(\mathbf{r}', \mathbf{r}, \omega),\tag{A3}
$$

<span id="page-15-2"></span>since  $Q(\mathbf{r}, \mathbf{r}', \omega)$  is reciprocal. Hence the transposed equation of Eq.  $(A1)$  $(A1)$  $(A1)$  takes the form

$$
\int d^3 s \vec{G}^\mathsf{T}(\mathbf{s}, \mathbf{r}, \omega) \cdot \vec{H}(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'). \tag{A4}
$$

Multiplying from the right with  $G(\mathbf{r}', \mathbf{s}', \omega)$ , integrating over **r'**, and using Eq. ([A1](#page-15-0)), one can see that the Green tensor is also reciprocal,

$$
\tilde{G}(\mathbf{r}, \mathbf{r}', \omega) = \tilde{G}^{\mathsf{T}}(\mathbf{r}', \mathbf{r}, \omega). \tag{A5}
$$

<span id="page-15-3"></span><span id="page-15-1"></span>Because of Eq.  $(A5)$  $(A5)$  $(A5)$ , the complex conjugate of Eq.  $(A4)$  $(A4)$  $(A4)$ reads

$$
\int d^3s \vec{G}^*(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{H}^*(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'). \tag{A6}
$$

Taking the dot product of Eq.  $(A1)$  $(A1)$  $(A1)$  from the left with  $G^*(s', r, \omega)$  and integrating over **r**, taking the dot product of Eq. ([A6](#page-15-3)) from the right with  $G(\mathbf{r}', \mathbf{s}', \omega)$  and integrating over <span id="page-16-1"></span>**r'**, and subtracting the two resulting equations, one derives

Im 
$$
\vec{G}(\mathbf{r}, \mathbf{r}', \omega) = -\int d^3s \int d^3s' \vec{G}(\mathbf{r}, \mathbf{s}, \omega)
$$
  
 
$$
\cdot \left[ \text{Im } \vec{H}(\mathbf{s}, \mathbf{s}', \omega) \right] \cdot \vec{G}^*(\mathbf{s}', \mathbf{r}', \omega). \quad (A7)
$$

From Eq.  $(A2)$  $(A2)$  $(A2)$  it is seen that

<span id="page-16-0"></span>Im 
$$
\vec{H}(\mathbf{r}, \mathbf{r}', \omega) = -\frac{\text{Im } \omega^2}{c^2} \vec{l} \delta(\mathbf{r} - \mathbf{r}') - \mu_0 \text{Re}[\omega \vec{Q}(\mathbf{r}, \mathbf{r}', \omega)].
$$
  
(A8)

Insertion of Eq.  $(A8)$  $(A8)$  $(A8)$  into Eq.  $(A7)$  $(A7)$  $(A7)$  and restriction to real frequencies leads, upon recalling Eq.  $(2)$  $(2)$  $(2)$ , to Eq.  $(9)$  $(9)$  $(9)$ .

# **APPENDIX B: NONUNIQUENESS OF THE KERNEL**  $K(\mathbf{r},\mathbf{r}',\omega)$

<span id="page-16-2"></span>The transition from  $K(\mathbf{r}, \mathbf{r}', \omega)$  to  $K'(\mathbf{r}, \mathbf{r}', \omega)$  according to Eq.  $(28)$  $(28)$  $(28)$  can be reinterpreted as a redefinition of the dynamical variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$  according to

$$
\hat{\mathbf{f}}(\mathbf{r}, \omega) = \int d^3 r' \, \vec{V}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}'(\mathbf{r}', \omega), \quad (\text{B1})
$$

$$
\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega) = \int d^{3}r' \vec{V}^{*}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}^{\prime \dagger}(\mathbf{r}', \omega). \tag{B2}
$$

<span id="page-16-3"></span>Inserting Eq.  $(B1)$  $(B1)$  $(B1)$  into Eq.  $(17)$  $(17)$  $(17)$  yields

$$
\hat{\mathbf{j}}_{\mathcal{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{1/2} \int d^3r' \vec{K}'(\mathbf{r},\mathbf{r}',\omega) \cdot \hat{\mathbf{f}}'(\mathbf{r}',\omega), \quad (B3)
$$

where  $K'(\mathbf{r}, \mathbf{r}', \omega)$  is just given by Eq. ([28](#page-3-7)). With regard to the transformations  $(B1)$  $(B1)$  $(B1)$  and  $(B2)$  $(B2)$  $(B2)$ , the significance of replac-ing Eq. ([29](#page-3-2)) with Eq. ([30](#page-3-1)) is that the variables  $\hat{\mathbf{f}}'(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^{\prime\dagger}(\mathbf{r},\omega)$  are uniquely expressible in terms of the  $\hat{\mathbf{f}}(\mathbf{r},\omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$ , and so are on an equal footing with them—the unitary operator associated with  $V(\mathbf{r}, \mathbf{r}', \omega)$  uniquely maps a set of bosonic variables onto a fully equivalent set of bosonic variables. Hence  $V(\mathbf{r}, \mathbf{r}', \omega)$  may be thought of as being included in the chosen set of dynamical variables. In this sense, it is sufficient to base the calculations in Sec. III on the Hermitian operator associated with the integral kernel  $K(\mathbf{r}, \mathbf{r}', \omega)$  as defined by Eq. ([27](#page-3-3)).

It is worth noting that the operator associated with  $K'(\mathbf{r}, \mathbf{r}', \omega)$  as defined by Eq. ([28](#page-3-7)) is non-Hermitian whenever  $V(\mathbf{r}, \mathbf{r}', \omega)$  is nontrivial. To see this, let us conversely assume that the operator associated with  $K'(\mathbf{r}, \mathbf{r}', \omega)$  is Hermitian,

<span id="page-16-4"></span>
$$
\int d^3 s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}(\mathbf{s}, \mathbf{r}', \omega) = \int d^3 s \vec{V}^+(\mathbf{s}, \mathbf{r}, \omega) \cdot \vec{K}(\mathbf{s}, \mathbf{r}', \omega).
$$
\n(B4)

Applying from the left the operator associated with  $\hat{V}$  and from the right the operator associated with  $\dot{V}^+$  and recalling Eq.  $(29)$  $(29)$  $(29)$ , one sees that

<span id="page-16-5"></span>
$$
\int d^3s \vec{V}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{K}(\mathbf{s}, \mathbf{r}', \omega) = \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}^+(\mathbf{r}', \mathbf{s}, \omega).
$$
\n(B5)

Applying the operator associated with  $\tilde{K}$  from the left to Eq.  $(B4)$  $(B4)$  $(B4)$  and from the right to Eq.  $(B5)$  $(B5)$  $(B5)$  and comparing the results, one finds that the operators associated with  $\hat{V}$  and  $\hat{\sigma}$ commute [recall Eq.  $(18)$  $(18)$  $(18)$ ], so that the operator associated with  $\hat{V}$  maps each (possibly degenerate) eigenspace of the operator associated with  $\vec{\sigma}$  onto itself. Specifically, this implies that the operator associated with  $\hat{V}$  commutes with the spectral projectors of the operator associated with  $\vec{\sigma}$  [and, therefore, also with the projectors ([38](#page-4-8))]. Since the spectral projectors of the operators associated with  $\vec{\sigma}$  and  $\vec{K}$  are the same [cf. Eqs.  $(26)$  $(26)$  $(26)$  and  $(27)$  $(27)$  $(27)$ ], the operators associated with *V* and  $\hat{K}$  also commute; but then, since the operator associated with  $\hat{K}$  is invertible (being a positive operator), Eq. ([B4](#page-16-4)) [or Eq.  $(B5)$  $(B5)$  $(B5)$  shows that the operator associated with  $\tilde{V}$  is Hermitian, i.e.,

$$
\vec{V}(\mathbf{r}, \mathbf{r}', \omega) = \vec{V}^+(\mathbf{r}', \mathbf{r}, \omega).
$$
 (B6)

Since the operator associated with  $\hat{V}$  is also unitary, in the diagonal expansion

$$
\vec{V}(\mathbf{r}, \mathbf{r}', \omega) = \int d\alpha v(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega) \quad (B7)
$$

one must have  $v(\alpha, \omega) = \pm 1$  for each  $\alpha$ , which means that the Hermitian operator associated with  $K'(\mathbf{r}, \mathbf{r}', \omega)$  can differ from the operator associated with  $K(\mathbf{r}, \mathbf{r}', \omega)$  only by the trivial type of unitary transformation that merely replaces some of the basis functions  $\mathbf{F}(\alpha, \mathbf{r}, \omega)$  with  $-\mathbf{F}(\alpha, \mathbf{r}, \omega)$ . Conversely, this shows that any (in this sense) nontrivial  $V(\mathbf{r}, \mathbf{r}', \omega)$  necessarily yields a non-Hermitian  $K'(\mathbf{r}, \mathbf{r}', \omega)$ .

# **APPENDIX C: REDUCED STATE SPACE AND SUPERSELECTION RULE**

Let us consider the state space spanned by the Fock states associated with  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega)$  so that an arbitrary, normalizable state  $|\phi\rangle$  in this space can be represented in the form

<span id="page-17-0"></span>
$$
|\phi\rangle = |0\rangle\langle 0|\phi\rangle + \sum_{k_1=1}^3 \int_0^\infty d\omega_1 \int d^3r_1 \phi_{k_1}(\mathbf{r}_1, \omega_1) |1_{k_1}(\mathbf{r}_1, \omega_1)\rangle
$$
  
+ 
$$
\sum_{k_1,k_2=1}^3 \int_0^\infty d\omega_1 \int_0^\infty d\omega_2 \int d^3r_1 \int d^3r_2
$$
  
× 
$$
\phi_{k_1k_2}(\mathbf{r}_1, \omega_1, \mathbf{r}_2, \omega_2) |1_{k_1}(\mathbf{r}_1, \omega_1), 1_{k_2}(\mathbf{r}_2, \omega_2)\rangle + \dots,
$$
 (C1)

where

$$
\hat{f}_k(\mathbf{r}, \omega)|0\rangle = 0, \tag{C2}
$$

$$
\hat{f}_{k}^{\dagger}(\mathbf{r},\omega)|0\rangle = |1_{k}(\mathbf{r},\omega)\rangle, \tag{C3}
$$

<span id="page-17-5"></span>
$$
\hat{f}_{k_N}^{\dagger}(\mathbf{r}_N, \omega_N) \cdots \hat{f}_{k_1}^{\dagger}(\mathbf{r}_1, \omega_1) |0\rangle = |1_{k_1}(\mathbf{r}_1, \omega_1), \dots, 1_{k_N}(\mathbf{r}_N, \omega_N)\rangle.
$$
\n(C4)

The normalization of  $\ket{\phi}$  can be obtained by using the formula (which can be viewed as a special case of the Bloch-De Dominicis theorem [[14](#page-21-8)])

$$
\langle 0|\hat{f}_{k_M}(\mathbf{r}_M, \omega_M) \cdots \hat{f}_{k_1}(\mathbf{r}_1, \omega_1)\hat{f}_{k'_1}^{\dagger}(\mathbf{r}'_1, \omega'_1) \cdots \hat{f}_{k'_N}^{\dagger}(\mathbf{r}'_N, \omega'_N)|0\rangle
$$
  

$$
= \delta_{MN} \sum_{\pi \in S_N} \prod_{l=1}^N \delta_{k_l, k'_{\pi(l)}} \delta(\mathbf{r}_l - \mathbf{r}'_{\pi(l)}) \delta(\omega_l - \omega'_{\pi(l)}) \qquad (C5)
$$

 $(\langle 0 | 0 \rangle = 1; S_N$  is the group of permutations of *N* objects).

In order to construct a reduced state space in which the operators  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega)$  defined by Eq. ([39](#page-4-10)) behave like bosonic operators, let us first introduce states  $|0\rangle$ <sub> $\lambda$ </sub>, according to

$$
\hat{f}_{\lambda i}(\mathbf{r}, \omega)|0\rangle_{\lambda} = 0, \tag{C6}
$$

$$
\hat{f}_{\lambda i}(\mathbf{r}, \omega) |0\rangle_{\lambda'} = |0\rangle_{\lambda'} \hat{f}_{\lambda i}(\mathbf{r}, \omega) \quad (\lambda \neq \lambda') \tag{C7}
$$

<span id="page-17-4"></span> $\left(\sqrt{0}\right|0\rangle_{\lambda}=1$ , such that

$$
|0\rangle = \bigotimes_{\lambda=1}^{\Lambda} |0\rangle_{\lambda}.
$$
 (C8)

Now let us introduce, for each  $\lambda$ , an orthogonal projector  $\hat{P}_\lambda$ as the sum of orthogonal projectors  $\hat{P}^{(i)}_{\lambda}$  $\binom{N}{\lambda}$ ,

$$
\hat{P}_{\lambda} = \sum_{N=0}^{\infty} \hat{P}_{\lambda}^{(N)},
$$
\n(C9)

$$
\hat{P}_{\lambda}^{(N)\dagger} = \hat{P}_{\lambda}^{(N)},\tag{C10}
$$

$$
\hat{P}_{\lambda}^{(N)} \hat{P}_{\lambda}^{(N')} = \delta_{NN'} \hat{P}_{\lambda}^{(N)} \tag{C11}
$$

<span id="page-17-2"></span><span id="page-17-1"></span>and specify  $\hat{P}^{(l)}_{\lambda}$  $N^{(N)}$  in such a way that, when applied to a quantum state of the form  $(C1)$  $(C1)$  $(C1)$ , it picks out the  $(N+1)$ <sup>th</sup> term on the right-hand side of Eq.  $(C1)$  $(C1)$  $(C1)$  and incorporates *N* positionspace projection kernels belonging to the chosen value of  $\lambda$ ,

$$
\hat{P}_{\lambda}^{(0)} = |0\rangle_{\lambda} \langle 0|_{\lambda}, \qquad (C12)
$$

<span id="page-17-6"></span><span id="page-17-3"></span>
$$
\hat{P}_{\lambda}^{(N)} = \frac{1}{N!} \sum_{k_1} \int_0^{\infty} d\omega_1 \int d^3 r_1 \sum_{k_2} \int_0^{\infty} d\omega_2
$$
\n
$$
\times \int d^3 r_2 \cdots \sum_{k_N} \int_0^{\infty} d\omega_N \int d^3 r_N \hat{f}_{\lambda k_1}^{\dagger}(\mathbf{r}_1, \omega_1)
$$
\n
$$
\times \hat{f}_{\lambda k_2}^{\dagger}(\mathbf{r}_2, \omega_2) \cdots \hat{f}_{\lambda k_N}^{\dagger}(\mathbf{r}_N, \omega_N) \hat{P}_{\lambda}^{(0)} \hat{f}_{\lambda k_N}(\mathbf{r}_N, \omega_N)
$$
\n
$$
\times \hat{f}_{\lambda k_{N-1}}(\mathbf{r}_{N-1}, \omega_{N-1}) \cdots \hat{f}_{\lambda k_1}(\mathbf{r}_1, \omega_1) \qquad (C13)
$$

 $(N=1,2,...)$ . It is not difficult to prove that Eqs. ([C10](#page-17-1)) and ([C11](#page-17-2)) are fulfilled, where the latter equation fixes the normalization factor  $1/N!$  in Eq. ([C13](#page-17-3)) and that, in view of Eqs.  $(C8)$  $(C8)$  $(C8)$  and  $(C11)$  $(C11)$  $(C11)$ , the commutation relation

$$
\left[\hat{P}_{\lambda}^{(N)}, \hat{P}_{\lambda'}^{(N')}\right] = 0\tag{C14}
$$

holds.

We may now define a reduced state space that contains only those (normalizable) vectors that have the separable form

$$
|\phi\rangle^{(\text{red})} = \bigotimes_{\lambda=1}^{\Lambda} |\phi\rangle_{\lambda},
$$
 (C15)

$$
\hat{P}_{\lambda}|\phi\rangle_{\lambda} = |\phi\rangle_{\lambda},\tag{C16}
$$

with each vector  $|\phi\rangle$  being, by construction, a superposition of vectors

$$
|N\rangle_{\lambda} = \sum_{k_1} \int_0^{\infty} d\omega_1 \int d^3 r_1 \cdots \sum_{k_N} \int_0^{\infty} d\omega_N \int d^3 r_N
$$
  
 
$$
\times C_{\lambda k_1 \cdots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)
$$
  
 
$$
\times |1_{\lambda k_1}(\mathbf{r}_1, \omega_1), \dots, 1_{\lambda k_N}(\mathbf{r}_N, \omega_N)\rangle, \qquad (C17)
$$

<span id="page-17-7"></span>where, in analogy to Eq.  $(C4)$  $(C4)$  $(C4)$ ,

$$
\begin{aligned} \n\left| 1_{\lambda k_1}(\mathbf{r}_1, \omega_1), \dots, 1_{\lambda k_N}(\mathbf{r}_N, \omega_N) \right\rangle \\ \n&= \hat{f}_{\lambda k_N}^{\dagger}(\mathbf{r}_N, \omega_N) \cdots \hat{f}_{\lambda k_1}^{\dagger}(\mathbf{r}_1, \omega_1) |0\rangle_{\lambda}. \n\end{aligned} \n\tag{C18}
$$

The important feature of these states is that the result of performing the integrations in Eq.  $(C17)$  $(C17)$  $(C17)$  is not changed if the wave function  $C_{\lambda k_1 \cdots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)$  is replaced according to

$$
C_{\lambda k_1 \cdots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)
$$
  
\n
$$
\mapsto \int d^3 r'_1 \cdots \int d^3 r'_N(\vec{P}_\lambda)_{k_1 k'_1}(\mathbf{r}_1, \mathbf{r}'_1, \omega_1) \cdots
$$
  
\n
$$
\times (\vec{P}_\lambda)_{k_N k'_N}(\mathbf{r}_N, \mathbf{r}'_N, \omega_N)
$$
  
\n
$$
\times C_{\lambda k'_1 \cdots \lambda k'_N}(\mathbf{r}'_1, \omega_1, \dots, \mathbf{r}'_N, \omega_N).
$$
 (C19)

It is also not changed if  $C_{\lambda k_1 \cdots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)$  is symmetrized with respect to the labels 1, ... ,*N*. Wave functions that can be reduced to the same standardized wave function by these operations are thus fully equivalent representatives of the same vector. Without loss of generality, one can thus adopt the convention to employ only such standardized wave functions.

<span id="page-18-0"></span>The commutation relation  $(40)$  $(40)$  $(40)$  implies that

$$
e^{\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)} \hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega') e^{-\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)}
$$
  
=  $\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'}(\vec{P}_{\lambda})_{k k'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')$  (C20)

with  $\epsilon$  being a parameter. As Eq. ([C20](#page-18-0)) is a similarity transformation, it generalizes to

$$
e^{\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)} F[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega')] e^{-\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)}
$$
  
=  $F[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'}(\vec{P}_{\lambda})_{k k'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')],$  (C21)

where  $F = F[\hat{f}^{\dagger}_{\lambda' k'}(\mathbf{r}', \omega')]$  is any well-behaved functional of  $\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega')$ . Comparison of the terms of first order in  $\epsilon$  on both sides yields

<span id="page-18-1"></span>
$$
\left[\hat{f}_{\lambda k}(\mathbf{r}, \omega), F[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega')] \right]
$$
\n
$$
= \left\{ \frac{\partial}{\partial \epsilon} F[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'}(\tilde{F}_{\lambda})_{k k'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')] \right\}_{\epsilon=0} . \quad (C22)
$$

Let us consider the particular functional  $F_N[\hat{f}_{\lambda'k'}^{\dagger}(\mathbf{r}', \omega')]$  appearing in Eqs.  $(C17)$  $(C17)$  $(C17)$  and  $(C18)$  $(C18)$  $(C18)$ ,

$$
F_N[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega')] = \sum_{k_1} \int_0^{\infty} d\omega_1 \int d^3 r_1 \cdots \sum_{k_N} \int_0^{\infty} d\omega_N \int d^3 r_N
$$
  
×  $C_{\lambda k_1 \dots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)$   
×  $\hat{f}_{\lambda k_N}^{\dagger}(\mathbf{r}_N, \omega_N) \cdots \hat{f}_{\lambda k_1}^{\dagger}(\mathbf{r}_1, \omega_1).$  (C23)

If the convention to use only standardized wave functions is adopted, one may write

$$
F_N[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'}(\tilde{P}_{\lambda})_{k k'}(\mathbf{r}, \mathbf{r}', \omega') \delta(\omega - \omega')]
$$
  
=  $F_N[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'} \delta_{k k'} \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega')],$  (C24)

which means that the right-hand side of Eq.  $(C22)$  $(C22)$  $(C22)$  may be evaluated, for this functional, just as an ordinary functional derivative, i.e.,

<span id="page-18-2"></span>
$$
\left[\hat{f}_{\lambda k}(\mathbf{r},\omega), F_N[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}',\omega')] \right] = \frac{\delta F_N[\hat{f}_{\lambda' k'}^{\dagger}(\mathbf{r}',\omega')] }{\delta \hat{f}_{\lambda k}^{\dagger}(\mathbf{r},\omega)};
$$
\n(C25)

but since, due to the definition of the reduced state space, only commutators of the type  $(C25)$  $(C25)$  $(C25)$  (for all *N*) are required,

and since Eq.  $(C25)$  $(C25)$  $(C25)$  can be obtained from Eq.  $(47)$  $(47)$  $(47)$  in the same way that Eq.  $(C22)$  $(C22)$  $(C22)$  has been obtained from Eq.  $(40)$  $(40)$  $(40)$ , Eq. ([47](#page-4-11)) is generally valid for the reduced state space.

#### **APPENDIX D: DERIVATION OF EQ. [\(123\)](#page-11-3)**

For notational convenience, let us write here the integral equation  $(18)$  $(18)$  $(18)$  in the compact operator form

$$
\mathcal{K}\mathcal{K}^{\dagger}=\sigma,\tag{D1}
$$

<span id="page-18-3"></span>with  $K$  and  $\sigma$  being, respectively, the operators associated (for chosen  $\omega$ ) with the integral kernels  $K(\mathbf{r}, \mathbf{r}', \omega)$  and  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ . Accordingly, Eqs. ([28](#page-3-7)) and ([30](#page-3-1)) read  $K' = KV$ and  $V^{\dagger}V = VV^{\dagger} = I$ , respectively (*I* is the unit operator). Assuming that the (Hermitian and positive) operator  $\sigma$  takes the form

$$
\sigma = \sigma_0 + \epsilon \sigma_1, \tag{D2}
$$

<span id="page-18-4"></span>with  $\epsilon$  being a small, real parameter, we may try to find a solution to Eq.  $(D1)$  $(D1)$  $(D1)$  by the perturbative ansatz

$$
\mathcal{K}' = \mathcal{K}'_0 + \epsilon \mathcal{K}'_1 + \cdots, \tag{D3}
$$

<span id="page-18-5"></span>where  $\mathcal{K}'_0$  is a solution to Eq. ([D1](#page-18-3)) for  $\epsilon = 0$ . Substituting Eqs.  $(D2)$  $(D2)$  $(D2)$  and  $(D3)$  $(D3)$  $(D3)$  into Eq.  $(D1)$  $(D1)$  $(D1)$ , we see that the first-order correction  $K_1'$  obeys the equation

$$
\mathcal{K}_0' \mathcal{K}_1'^\dagger + \mathcal{K}_1' \mathcal{K}_0'^\dagger = \sigma_1,\tag{D4}
$$

<span id="page-18-6"></span>which determines the Hermitian part of  $K_0'K_1'^\dagger$  (recall that  $\sigma_1$ ) is Hermitian), whereas the anti-Hermitian part is left undetermined. The solution to Eq.  $(D4)$  $(D4)$  $(D4)$  may therefore be written as

$$
\mathcal{K}'_1 = \frac{1}{2}(\sigma_1 + \mathcal{A})\mathcal{M}_0,\tag{D5}
$$

where  $\mathcal{M}_0 = (\mathcal{K}_0^{\dagger})^{-1}$ , and  $\mathcal{A} = -\mathcal{A}^{\dagger}$  is an arbitrary anti-Hermitian operator, which may be simply set to zero. Note that the freedom to choose  $A$  corresponds to the freedom to choose V. We hence obtain to first order in  $\sigma-\sigma_0$ 

$$
\mathcal{K}' = \mathcal{K}'_0 + \frac{1}{2}(\sigma - \sigma_0)\mathcal{M}_0.
$$
 (D6)

<span id="page-18-7"></span>If  $\sigma_0$ ,  $\mathcal{K}'_0$ , and  $\sigma$  are identified with the operators associated with the kernels  $(81)$  $(81)$  $(81)$ ,  $(88)$  $(88)$  $(88)$ , and  $(117)$  $(117)$  $(117)$  respectively, Eq.  $(D6)$  $(D6)$  $(D6)$ is just the operator equivalent of Eq.  $(123)$  $\left[\ddot{K}^{\prime}(\mathbf{r},\mathbf{r}^{\prime},\omega) \leftrightarrow \ddot{K}(\mathbf{r},\mathbf{r}^{\prime},\omega)\right].$ 

To calculate explicitly the integral kernel  $\dot{M}_0(\mathbf{r}, \mathbf{r}', \omega)$  associated with  $\mathcal{M}_0$ , we consider the Fourier representation

$$
\vec{K}'_0(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{K}'_0(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \tag{D7}
$$

where, according to Eq.  $(75)$  $(75)$  $(75)$ ,

$$
\vec{K}'_0(\mathbf{k}, \omega) = \sigma_{\parallel}^{1/2}(\omega) [\vec{I} \pm \alpha(\omega) \mathbf{k} \times \vec{I}] \tag{D8}
$$

 $[\alpha(\omega) = [\gamma(\omega)/\sigma_{\parallel}(\omega)]^{1/2} > 0]$ , which shows that the kernel  $\widetilde{M}_0(\mathbf{r}, \mathbf{r}', \omega)$  can be given by the Fourier integral

<span id="page-19-0"></span>
$$
\vec{M}_0(\mathbf{r}, \mathbf{r}', \omega) = \sigma_{\parallel}^{-1/2}(\omega) \times \int \frac{d^3k}{(2\pi)^3} [\vec{I} \mp \alpha(\omega) \mathbf{k} \times \vec{I}]^{-1} e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} ,
$$
\n(D9)

<span id="page-19-1"></span>and it is not difficult to prove that

$$
[\vec{I} \pm \alpha(\omega) \mathbf{k} \times \vec{I}]^{-1} = \vec{I} \mp \alpha(\omega) \frac{\mathbf{k} \times \vec{I}}{1 + \alpha(\omega)^2 k^2} + \alpha(\omega)^2 \frac{\mathbf{k} \times \vec{I} \times \mathbf{k}}{1 + \alpha(\omega)^2 k^2}.
$$
 (D10)

Introducing the function

$$
m_0(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \frac{e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}}{\alpha(\omega)^{-2} + k^2}
$$
  
=  $-(4\pi |\mathbf{r} - \mathbf{r}'|)^{-1} e^{-|\mathbf{r} - \mathbf{r}'|/\alpha(\omega)},$  (D11)

we may rewrite Eq.  $(D9)$  $(D9)$  $(D9)$  [with Eq.  $(D10)$  $(D10)$  $(D10)$ ] to obtain Eq. ([124](#page-11-4)). Note that the (Yukawa-type) function  $m_0(\mathbf{r}, \mathbf{r}', \omega)$  satisfies the equation

$$
[-\Delta + \alpha(\omega)^{-2}]m_0(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}')
$$
 (D12)

together with the boundary condition  $m_0(\mathbf{r}, \mathbf{r}', \omega) \rightarrow 0$  for  $|\mathbf{r}-\mathbf{r}'|$  → ∞.

## **APPENDIX E: BULK-MEDIUM GREEN TENSOR AND KLIEVER-FUCHS IMPEDANCE**

For (translationally invariant) bulk material,

$$
\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) = \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \vec{Q}(\mathbf{k}, \omega),
$$
(E1)

<span id="page-19-3"></span>the solution to Eq.  $(8)$  $(8)$  $(8)$  has the form

$$
\vec{G}^{(0)}(\mathbf{r}, \mathbf{r}', \omega) = \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \vec{G}^{(0)}(\mathbf{k}, \omega), \quad \text{(E2)}
$$

where  $\vec{G}^{(0)}(\mathbf{k}, \omega)$  is the solution to a simple 3  $\times$  3 matrix equation. In particular, for an isotropic medium without optical activity,

$$
\widetilde{Q}(\mathbf{k},\omega) = Q_{\parallel}(k,\omega)\frac{\mathbf{k}\mathbf{k}}{k^2} + Q_{\perp}(k,\omega)\left(\widetilde{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\right),\qquad(E3)
$$

<span id="page-19-2"></span>one finds that

$$
\widetilde{G}^{(0)}(\mathbf{k},\omega) = \frac{\widetilde{I} - \mathbf{k}\mathbf{k}/k^2}{D_{\perp}(k,\omega)} - \frac{\mathbf{k}\mathbf{k}/k^2}{D_{\parallel}(k,\omega)},
$$
(E4)

<span id="page-19-5"></span>where

$$
D_{\perp}(k,\omega) = k^2 - \omega^2/c^2 - i\mu_0 \omega Q_{\perp}(k,\omega),
$$
 (E5)

$$
D_{\parallel}(k,\omega) = \omega^2/c^2 + i\mu_0 \omega Q_{\parallel}(k,\omega),
$$
 (E6)

<span id="page-19-6"></span>and

 $Q_{\parallel(\perp)}(k,\omega) = -i\varepsilon_0 \omega \left[\varepsilon_{\parallel(\perp)}(k,\omega) - 1\right]$  (E7)  $(E7)$  in "dielectric" notation. In the general case where

$$
\widetilde{Q}(\mathbf{k},\omega) = Q_{\parallel}(\mathbf{k},\omega)\frac{\mathbf{k}\mathbf{k}}{k^2} + \left(\widetilde{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\right) \cdot \widetilde{Q}_{\perp}(\mathbf{k},\omega) \cdot \left(\widetilde{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\right),\tag{E8}
$$

 $D_{\parallel}(k,\omega)$  changes to  $D_{\parallel}(\mathbf{k},\omega)$ ,  $D_{\perp}(k,\omega)$  changes to the tensor

$$
\vec{D}_{\perp}(\mathbf{k},\omega) = k^2 - \omega^2/c^2 - i\mu_0 \omega \vec{Q}_{\perp}(\mathbf{k},\omega),
$$
 (E9)

and the first term on the right-hand side of Eq.  $(E4)$  $(E4)$  $(E4)$  has to be replaced according to  $(I_k = I - \mathbf{k} \mathbf{k}/k^2)$ 

$$
\frac{\vec{I} - \mathbf{k}\mathbf{k}/k^2}{D_{\perp}(k,\omega)} \mapsto \vec{I}_k \cdot [\vec{I}_k \cdot \vec{D}_{\perp}(\mathbf{k},\omega) \cdot \vec{I}_k]^{-1} \cdot \vec{I}_k. \tag{E10}
$$

It may be convenient—particularly with regard to systems that are translationally invariant only in a particular plane, say the *xy* plane—to rewrite Eq. ([E2](#page-19-3)) as  $[r=(\rho, z), \mathbf{k}]$  $=$ **q**, $\beta$ <sup>)</sup>

$$
\vec{G}^{(0)}(\mathbf{r}, \mathbf{r}', \omega) = \int d^2q e^{i\mathbf{q} \cdot (\boldsymbol{\rho} - \boldsymbol{\rho}')}\vec{G}^{(0)}(z, z', \mathbf{q}, \omega), \quad \text{(E11)}
$$

<span id="page-19-7"></span><span id="page-19-4"></span>where

$$
\vec{G}^{(0)}(z,z',\mathbf{q},\omega) = \int \frac{d\beta}{(2\pi)^3} e^{i\beta(z-z')} \vec{G}^{(0)}(\mathbf{k},\omega). \quad \text{(E12)}
$$

The analytical properties of the integrand in Eq.  $(E12)$  $(E12)$  $(E12)$  with respect to  $\beta$  depend on the specific decay to zero of  $\ddot{Q}$ (**r**  $-$  **r**', ω) for  $|\mathbf{r} - \mathbf{r}'| \rightarrow \infty$ . For sufficiently rapid decay, Eq.  $(E12)$  $(E12)$  $(E12)$  admits an evaluation by contour integration in the complex  $\beta$  plane, which will be governed—focusing again on isotropic media without optical activity—by the solutions  $\beta = \beta_{\nu}^{\perp,\parallel}(q,\omega)$  of the two dispersion equations  $D_{\perp,\parallel}(k,\omega) = 0$ . In contrast to the case of spatially nondispersive material, these equations are transcendental with respect to  $\beta$  rather than polynomial, so that nothing can be said about the number of their solutions in general. Specifically, if there are more than two functions  $\beta^{\perp}_{\nu}(q,\omega)$  [and/or one or more functions  $\beta_{\nu}^{\parallel}(q,\omega)$ ], the medium is said to support "additional" (inhomogeneous plane) waves.

We close this appendix with the following (perhaps not well-known) observation. Inserting Eq. ([E4](#page-19-2)) in Eq. ([E12](#page-19-4)) and setting  $z-z' = 0$ ±, and making use of the decompositions

$$
\widetilde{I} - \mathbf{k} \mathbf{k}/k^2 = \mathbf{e}_s(\mathbf{q}) \mathbf{e}_s(\mathbf{q}) + \mathbf{e}_p(\mathbf{k}) \mathbf{e}_p(\mathbf{k}),
$$
 (E13)

$$
\overrightarrow{I}_z = \mathbf{e}_s(\mathbf{q})\mathbf{e}_s(\mathbf{q}) + \mathbf{q}\mathbf{q}/q^2, \tag{E14}
$$

with  $\mathbf{e}_s(\mathbf{q}) = \mathbf{q} \times \mathbf{e}_z/q$  and  $\mathbf{e}_p(\mathbf{k}) = \mathbf{k} \times \mathbf{q} \times \mathbf{e}_z/kq$  being polarization unit vectors, one can show that

$$
\overrightarrow{I}_z \cdot \overrightarrow{G}^{(0)}(0 \pm , 0, \mathbf{q}, \omega) \cdot \overrightarrow{I}_z = (i\mu_0 \omega)^{-1} [Z_s(q, \omega) \mathbf{e}_s(\mathbf{q}) \mathbf{e}_s(\mathbf{q})
$$

$$
+ Z_p(q, \omega) \mathbf{q} \mathbf{q} / q^2], \qquad (E15)
$$

where

$$
Z_s(q,\omega) = i\mu_0 \omega \int_{-\infty}^{\infty} \frac{d\beta}{(2\pi)^3} \frac{e^{i\beta 0 \pm}}{D_{\perp}(k,\omega)},
$$
 (E16)

<span id="page-20-1"></span><span id="page-20-0"></span>
$$
Z_p(q,\omega) = i\mu_0 \omega \int_{-\infty}^{\infty} \frac{d\beta}{(2\pi)^3} \frac{e^{i\beta 0 \pm}}{k^2} \left[ \frac{\beta^2}{D_{\perp}(k,\omega)} - \frac{q^2}{D_{\parallel}(k,\omega)} \right].
$$
\n(E17)

With Eqs.  $(E5)$  $(E5)$  $(E5)$ – $(E7)$  $(E7)$  $(E7)$ ,  $(E16)$  $(E16)$  $(E16)$ , and  $(E17)$  $(E17)$  $(E17)$  are recognized (up to a trivial factor) as the surface impedance expressions first derived by Kliever and Fuchs  $[34]$  $[34]$  $[34]$  for a spatially dispersive half-space by assuming specular electron reflection (see also Ref. [[26](#page-21-29)]).

## **APPENDIX F: DERIVATION OF EQS. [\(139\)](#page-13-4) AND [\(140\)](#page-13-5)**

From inspection of Eq.  $(137)$  $(137)$  $(137)$  it is seen that only the tangential components of the electric field, which are assumed to be continuous at  $z' = 0$  and  $z' = d$ , contribute to the righthand side of this equation. We therefore evaluate Eq.  $(137)$  $(137)$  $(137)$  at  $z' = 0$ + and  $z' = d$ – and take the tangential  $(\tilde{I}_z)$  component thereof to obtain the linear equations (the arguments **q** and  $\omega$ are kept fixed in this appendix and are suppressed for notational convenience)

<span id="page-20-2"></span>
$$
\begin{split} \mathbf{\underline{E}}(0+) \cdot \vec{I}_z \cdot [\vec{I} + \vec{R}(0,0+)] \cdot \vec{I}_z - \mathbf{\underline{E}}(d-) \cdot \vec{I}_z \cdot \vec{R}(d,0+) \cdot \vec{I}_z \\ = \mathbf{\underline{E}}^{(\text{in})}(0+) \cdot \vec{I}_z \end{split} \tag{F1}
$$

<span id="page-20-3"></span>and

$$
\mathbf{E}(0+) \cdot \vec{I}_z \cdot \vec{R}(0,d-) \cdot \vec{I}_z + \mathbf{E}(d-) \cdot \vec{I}_z \cdot [\vec{I} - \vec{R}(d,d-) ] \cdot \vec{I}_z
$$
\n
$$
= \mathbf{E}^{(\text{in})}(d-) \cdot \vec{I}_z, \tag{F2}
$$

respectively, which are to be solved for the tangential components  $\mathbf{E}(0+) \cdot I_z$  and  $\mathbf{E}(d-) \cdot I_z$ . To represent the solution in a compact form, we note that one may introduce for a general *A* satisfying  $I_z \cdot A \cdot I_z = A$  a pseudoinverse  $A^{\#}$  with respect to the  $\tilde{I}_z$ -space by the conditions  $\tilde{A} \cdot \tilde{A}^* = \tilde{I}_z = \tilde{A}^* \cdot \tilde{A}$  and  $I_z \cdot A^{\#} \cdot I_z = A^{\#}$ . This pseudoinverse, which may also be denoted, informally, by  $\tilde{I}_z \cdot (\tilde{I}_z \cdot \tilde{A} \cdot \tilde{I}_z)^{-1} \cdot \tilde{I}_z$ , is unique whenever it exists, which is the case if and only if the eigenspace of *A* corresponding to the eigenvalue zero contains no further linearly independent vectors in addition to  $\mathbf{e}_z$  (i.e., if  $\tilde{I}_z \cdot \tilde{A} \cdot \tilde{I}_z$  is invertible when considered as a mapping taking only the smaller  $\hat{I}_z$ -space into itself). It is then straightforward to see that the block-matrix formula

<span id="page-20-6"></span>
$$
\begin{pmatrix}\n\vec{A} & \vec{B} \\
\vec{C} & \vec{D}\n\end{pmatrix}\n\cdot\n\begin{pmatrix}\n\vec{C}^* \cdot [\vec{A} \cdot \vec{C}^* - \vec{B} \cdot \vec{D}^*]^* & \vec{A}^* \cdot [\vec{C} \cdot \vec{A}^* - \vec{D} \cdot \vec{B}^*]^* \\
\vec{C} & \vec{D}\n\end{pmatrix}
$$
\n
$$
= \begin{pmatrix}\n\vec{I}_z & 0 \\
0 & \vec{I}_z\n\end{pmatrix}
$$
\n(F3)

is generally valid whenever the requisite pseudoinverses exist, so that the solution to Eqs.  $(F1)$  $(F1)$  $(F1)$  and  $(F2)$  $(F2)$  $(F2)$  can be written in the form

$$
\mathbf{E}(0+) \cdot \vec{I}_z = [\mathbf{E}^{(\text{in})}(0+) \cdot \vec{C}^{\sharp} - \mathbf{E}^{(\text{in})}(d-) \cdot \vec{D}^{\sharp}] \cdot [\vec{A} \cdot \vec{C}^{\sharp} - \vec{B} \cdot \vec{D}^{\sharp}]^{\sharp},
$$
\n(F4)

$$
\mathbf{E}(d-)\cdot\vec{I}_z = [\mathbf{E}^{(\text{in})}(0+)\cdot\vec{A}^{\sharp} - \mathbf{E}^{(\text{in})}(d-)\cdot\vec{B}^{\sharp}] \cdot [\vec{C}\cdot\vec{A}^{\sharp} - \vec{D}\cdot\vec{B}^{\sharp}]^{\sharp}
$$
\n(F5)

together with Eqs.  $(141)$  $(141)$  $(141)$ – $(144)$  $(144)$  $(144)$ . Recalling again the continuity of the tangential component of the electric field, we are left with Eqs.  $(139)$  $(139)$  $(139)$  and  $(140)$  $(140)$  $(140)$ . It should be noted that the above inversion procedure fails at particular values of the (suppressed) arguments  $q$  and  $\omega$ , because of singularities. However, as we are dealing with a lossy system, such singularities—corresponding to guided waves—may appear only when Im  $\omega < 0$  (for real values of **q**).

## **APPENDIX G: DERIVATION OF EQS. [\(153\)](#page-14-1)–[\(156\)](#page-14-2)**

Let us attribute to the three regions  $j=0$  ( $z<0$ ),  $j=1$  $(0 \lt z \lt d)$ ,  $j=2$   $(z \gt d)$  bulk-medium conductivities  $Q_j(\mathbf{r}-\mathbf{r}', \omega)$ , which combine to the overall conductivity ten-sor in the sense of Eq. ([125](#page-11-2))  $[j \leftrightarrow L]$ . For each of these regions, we construct the translationally invariant bulkmedium Green tensor  $G_j(\mathbf{r}, \mathbf{r}', \omega)$  and, according to Eq. ([128](#page-12-6)), the associated auxiliary tensor  $\Gamma_j(\mathbf{r}, \mathbf{r}', \omega)$  in terms of their Fourier components  $\tilde{G}_j(z, z', \mathbf{q}, \omega)$  and  $\tilde{\Gamma}_j(z, z', \mathbf{q}, \omega)$ [defined according to Eqs.  $(E11)$  $(E11)$  $(E11)$  and  $(E12)$  $(E12)$  $(E12)$ ]. The three regions are thus described on an equal footing so that for the field in each region, an equation similar to Eq.  $(136)$  $(136)$  $(136)$  holds. Evaluating the tangential components of these three equations (which together determine the field in all of space) and using the continuity conditions at  $z=0$  and  $z=d$ , one obtains two sets of equations for the tangential boundary values,

<span id="page-20-4"></span>
$$
\{[\mathbf{E}(d) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(d, d-) + [\mu_0^{-1} \mathbf{B}(d) \times \mathbf{e}_z] \cdot \vec{G}_1(d, d-) - [\mathbf{E}(0) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(0, d-) - [\mu_0^{-1} \mathbf{B}(0) \times \mathbf{e}_z] \cdot \vec{G}_1(0, d-) \} \cdot \vec{I}_z
$$
  
= -\{[\mathbf{E}(d) \times \mathbf{e}\_z] \cdot \vec{\Gamma}\_2(0, 0+) - [\mu\_0^{-1} \mathbf{B}(d) \times \mathbf{e}\_z] \cdot \vec{\Gamma}\_2(0, 0+) \} \cdot \vec{I}\_z \qquad (G1)

and

<span id="page-20-5"></span>
$$
\{[\mathbf{E}(d) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(d, 0+) + [\mu_0^{-1} \mathbf{B}(d) \times \mathbf{e}_z] \cdot \vec{G}_1(d, 0+) - [\mathbf{E}(0) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(0, 0+) - [\mu_0^{-1} \mathbf{B}(0) \times \mathbf{e}_z] \cdot \vec{G}_1(0, 0+) \} \cdot \vec{I}_z
$$
  
= 
$$
\{[\mathbf{E}(0) \times \mathbf{e}_z] \cdot \vec{\Gamma}_0(0, 0-) - [\mu_0^{-1} \mathbf{B}(0) \times \mathbf{e}_z] \cdot \vec{G}_0(0, 0-) \} \cdot \vec{I}_z,
$$
 (G2)

where, for notational convenience, the arguments  $q$  and  $\omega$  of **E** and **B**, and the arguments  $-q$  and  $\omega$  of  $\tilde{G}_j$  and  $\tilde{\Gamma}_j$  have been suppressed. Solving these linear relations for  $\mathbf{B}(0)$  $\times$  **e**<sub>*z*</sub> and **B**(*d*)  $\times$  **e**<sub>*z*</sub> in terms of **E**(0)  $\times$  **e**<sub>*z*</sub> and **E**(*d*)  $\times$  **e**<sub>*z*</sub>, we need only compare the result with Eq. ([135](#page-13-1)) to verify Eqs.  $(153)$  $(153)$  $(153)$ - $(156)$  $(156)$  $(156)$ . This can be conveniently done by representing Eqs.  $(G1)$  $(G1)$  $(G1)$  and  $(G2)$  $(G2)$  $(G2)$  in the form

<span id="page-21-30"></span>
$$
\mu^{-1} \left( \frac{\mathbf{B}(0) \times \mathbf{e}_z}{\mathbf{B}(d) \times \mathbf{e}_z} \right)^{\mathsf{T}} \cdot \left( \begin{array}{cc} \vec{M} & \vec{N} \\ \vec{P} & \vec{S} \end{array} \right) = \left( \begin{array}{cc} \mathbf{E}(0) \times \mathbf{e}_z \\ \mathbf{E}(d) \times \mathbf{e}_z \end{array} \right)^{\mathsf{T}} \cdot \left( \begin{array}{cc} \vec{T} & \vec{U} \\ \vec{V} & \vec{W} \end{array} \right), \tag{G3}
$$

where  $\dot{M}$ ,  $\dot{N}$ ,  $\dot{P}$ ,  $\ddot{S}$ ,  $\ddot{T}$ ,  $\ddot{U}$ ,  $\ddot{V}$ , and  $\ddot{W}$  are given in Eqs.  $(145)$  $(145)$  $(145)$ – $(152)$  $(152)$  $(152)$ . Rewriting Eq.  $(135)$  $(135)$  $(135)$   $(z=0,d)$  in an analogous form and applying to Eq.  $(G3)$  $(G3)$  $(G3)$  the inversion formula  $(F3)$  $(F3)$  $(F3)$ , one obtains

$$
\begin{pmatrix} -\vec{Y}(0,0) & -\vec{Y}(0,d) \\ \vec{Y}(d,0) & \vec{Y}(d,d) \end{pmatrix} = \frac{1}{(2\pi)^2} \begin{pmatrix} \vec{T} & \vec{U} \\ \vec{V} & \vec{W} \end{pmatrix} \cdot \begin{pmatrix} \vec{P}^* \cdot [\vec{M} \cdot \vec{P}^* - \vec{N} \cdot \vec{S}^*]^* & \vec{M}^* \cdot [\vec{P} \cdot \vec{M}^* - \vec{S} \cdot \vec{N}^*]^* \\ \vec{S}^* \cdot [\vec{N} \cdot \vec{S}^* - \vec{M} \cdot \vec{P}^*]^* & \vec{N}^* \cdot [\vec{S} \cdot \vec{N}^* - \vec{P} \cdot \vec{M}^*]^* \end{pmatrix},
$$
(G4)

which immediately leads to Eqs.  $(153)$  $(153)$  $(153)$ – $(156)$  $(156)$  $(156)$ .

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