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Field quantization in inhomogeneous anisotropic dielectrics with spatio-temporal dispersion

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Abstract

A quantum damped-polariton model is constructed for an inhomogeneous anisotropic linear dielectric with arbitrary dispersion in space and time. The model Hamiltonian is completely diagonalized by determining the creation and annihilation operators for the fundamental polariton modes as specific linear combinations of the basic dynamical variables. Explicit expressions are derived for the time-dependent operators describing the electromagnetic field, the dielectric polarization and the noise term in the latter. It is shown how to identify bath variables that generate the dissipative dynamics of the medium.

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1. Introduction

Quantization of the electromagnetic field in a linear dielectric medium is a nontrivial task for various reasons. First of all, since the response of a dielectric to external fields is frequency-dependent in general, temporal dispersion should be taken into account. The well-known Kramers–Kronig relation implies that dispersion is necessarily accompanied by dissipation, so that the quantization procedure has to describe an electromagnetic field that is subject to damping. Furthermore, since the transverse and the longitudinal parts of the electromagnetic field play a different role in the dynamics, the quantization scheme should treat these parts separately. For inhomogeneous and spatially dispersive media this leads to complications in the quantization procedure, which further increase in the presence of anisotropy.

When the losses in a specific range of frequencies are small, temporal dispersion can be neglected. Field quantization in an inhomogeneous isotropic dielectric medium without spatio-temporal dispersion has been accomplished by employing a generalized transverse gauge, which depends on the dielectric constant [1–6].

A phenomenological scheme for field quantization in lossy dielectrics has been formulated on the basis of the fluctuation–dissipation theorem [7–10]. By adding a fluctuating noise term to the Maxwell equations and postulating specific commutation relations for the operator associated with the noise, one arrives at a quantization procedure that has been quite successful in describing the electromagnetic field in lossy dielectrics. An equivalent description in terms of auxiliary fields has been given as well [11, 12], while a related formalism has been presented recently [13]. However, all of these quantization schemes have the drawback that the precise physical nature of the noise term is not obvious, since its connection to the basic dynamical variables of the system is left unspecified. As a consequence, the status of the commutation relations for the noise operator is that of a postulate.

A justification of the above phenomenological quantization scheme has been sought by adopting a suitable model for lossy dielectrics. To that end use has been made of an extended version of the Hopfield polariton model [14] in which damping effects are accounted for by adding a dynamical coupling to a bath environment. Huttner and Barnett [15, 16] were the first to employ such a damped-polariton model in order to achieve field quantization for a lossy dielectric. Their treatment, which is confined to a spatially homogeneous medium, yields an explicit expression for the noise term as a linear combination of the canonical variables of the model. In a later development, an alternative formulation of the quantization procedure in terms of path integrals has been given [17], while Laplace transformations have been used to simplify the original formalism [18]. More recently, the effects of spatial inhomogeneities in the medium have been incorporated by solving an inhomogeneous version of the damped-polariton model [19–21].

In this way a full understanding of the phenomenological quantization scheme has been reached, at least for those dielectrics that can be represented by the damped-polariton models mentioned above. The latter proviso implies a limitation in various ways. First, one would like to include in a general model not only the effects of spatial inhomogeneity, but also those of spatial dispersion. Furthermore, it would be desirable to incorporate the consequences of spatial anisotropy, so that the theory encompasses crystalline media as well. Finally, while treating temporal dispersion and the associated damping, we would like to refrain from introducing a bath environment in the Hamiltonian from the start. Instead, we wish to formulate the Hamiltonian in terms of a full set of material variables, from which the dielectric polarization emerges by a suitable projection. In this way we will be able to account for any temporal dispersion that is compatible with a few fundamental principles like causality and net dielectric loss. For a homogeneous isotropic dielectric without spatial dispersion such an approach has been suggested before [16, 22].

Recently, several attempts have been made to remove some of the limitations that are inherent to the earlier treatments. In [23] the effects of spatial dispersion are considered in a path-integral formalism for a model that is a generalization of that of the original Huttner–Barnett approach. The discussion is confined to homogeneous dielectrics and to leading orders in the wavenumber, so that an analysis of the effects of arbitrary spatial dispersion in an inhomogeneous medium is out of reach. In [24] crystalline media have been discussed in the framework of a damped-polariton model with an anisotropic tensorial bath coupling. A complete diagonalization of the model along the lines of [15, 16] turned out to face difficulties due to the tensorial complexity, so that the full dynamics of the model is not presented. Both spatial dispersion and anisotropy are incorporated in the quantization scheme discussed in [25]. Use is made of a Langevin approach in which a damping term of a specific form is introduced. The commutation relations for the noise operator are postulated, as in the phenomenological quantization scheme. Finally, several treatments have appeared in which a dielectric model is formulated while avoiding the explicit introduction of a bath [26, 27]. However, a complete expression for the noise polarization operator in terms of the basic dynamical variables of the model is not presented in these papers. A direct proof of the algebraic properties of the latter operator is not furnished either.

In the present paper, we shall show how the damped-polariton model can be generalized in such a way that all of the above restrictions are removed. As we shall see, our general model describes the quantization and the time evolution of the electromagnetic field in an inhomogeneous anisotropic lossy dielectric with arbitrary spatio-temporal dispersion. A crucial step in arriving at our goals will be the complete diagonalization of the Hamiltonian. It will lead to explicit expressions for the operators describing the electromagnetic field and the dielectric polarization, and for the noise contribution contained in the latter. In this way the commutation relations for the noise operator will be derived rigorously from our general model, instead of being postulated along the lines of the phenomenological scheme. Finally, we shall make contact with previous treatments by showing how to construct a bath that generates damping phenomena in the dynamical evolution of the model.

2. Model Hamiltonian

In this section, we shall construct the general form of the Hamiltonian for a polariton model describing an anisotropic inhomogeneous dispersive dielectric. The result, which we shall obtain by starting from a few general principles, will contain several coefficients that can be chosen at will. As we shall see in a subsequent section, these coefficients can be adjusted in such a way that the susceptibility gets the appropriate form for any causal lossy dielectric that we would like to describe.

The Hamiltonian of the electromagnetic field is taken to have the standard form

$$H_{\rm f} = \int {\rm d}\mathbf{r} \left\{ \frac{1}{2\varepsilon_0} [\mathbf{\Pi}(\mathbf{r})]^2 + \frac{1}{2\mu_0} [\nabla \wedge \mathbf{A}(\mathbf{r})]^2 \right\} \tag{1}$$

with the Hermitian vector potential $\mathbf{A}(\mathbf{r})$ and its associated Hermitian canonical momentum $\mathbf{\Pi}(\mathbf{r})$. We use the Coulomb gauge $\nabla \cdot \mathbf{A} = 0$. In this gauge both $\mathbf{\Pi}$ and $\mathbf{A}$ are transverse. The canonical commutation relations read

$$[\mathbf{\Pi}(\mathbf{r}), \mathbf{A}(\mathbf{r}')] = -{\rm i}\hbar\delta_{\rm T}(\mathbf{r}-\mathbf{r}'), \qquad [\mathbf{\Pi}(\mathbf{r}), \mathbf{\Pi}(\mathbf{r}')] = 0, \qquad [\mathbf{A}(\mathbf{r}), \mathbf{A}(\mathbf{r}')] = 0 \tag{2}$$

where the transverse delta function is defined as $\boldsymbol{\delta}_{\rm T}(\mathbf{r}) = \mathsf{I}\,\delta(\mathbf{r}) + \nabla\nabla(4\pi r)^{-1}$, with I the unit tensor.

The Hamiltonian of the dielectric material medium is supposed to have the general form

$$H_{\rm m} = \hbar \int {\rm d}\mathbf{r} \int_0^\infty {\rm d}\omega\, \omega \mathbf{C}_{\rm m}^\dagger(\mathbf{r},\omega) \cdot \mathbf{C}_{\rm m}(\mathbf{r},\omega) \tag{3}$$

with the standard commutation relations for the creation and annihilation operators:

$$\left[\mathbf{C}_{\rm m}(\mathbf{r},\omega), \mathbf{C}_{\rm m}^\dagger(\mathbf{r}',\omega')\right] = \mathsf{I}\,\delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'), \qquad [\mathbf{C}_{\rm m}(\mathbf{r},\omega), \mathbf{C}_{\rm m}(\mathbf{r}',\omega')] = 0. \tag{4}$$

The medium operators commute with the field operators.

The material creation and annihilation operators are assumed to form a complete set describing all material degrees of freedom. Hence, any material dynamical variable, for instance the dielectric polarization density, can be expressed in terms of these operators. For a linear dielectric medium, the Hermitian polarization density is a linear combination of the medium operators, which has the general form

$$\mathbf{P}(\mathbf{r}) = -{\rm i}\hbar \int {\rm d}\mathbf{r}' \int_0^\infty {\rm d}\omega' \mathbf{C}_{\rm m}(\mathbf{r}',\omega') \cdot \mathsf{T}(\mathbf{r}',\mathbf{r},\omega') + {\rm h.c.} \tag{5}$$

The complex tensorial coefficient T appearing in this expression will be determined later on, when the dielectric susceptibility is properly identified. On a par with $\mathbf{P}$ we define its associated canonical momentum density $\mathbf{W}$, again as a linear combination of the medium operators

$$\mathbf{W}(\mathbf{r}) = -\int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega' \omega' \mathbf{C}_\mathrm{m}(\mathbf{r}', \omega') \cdot \mathsf{S}(\mathbf{r}', \mathbf{r}, \omega') + \mathrm{h.c.} \tag{6}$$

with a new complex tensorial coefficient S that is closely related to T, as we shall see below. For future convenience we inserted a factor ω' in the integrand and a minus sign in front of the integral.

As $\mathbf{W}$ and $\mathbf{P}$ are a canonical pair, they must satisfy the standard commutation relations:

$$[\mathbf{W}(\mathbf{r}), \mathbf{P}(\mathbf{r}')] = -\mathrm{i}\hbar \mathsf{I}\, \delta(\mathbf{r} - \mathbf{r}'), \qquad [\mathbf{W}(\mathbf{r}), \mathbf{W}(\mathbf{r}')] = 0, \qquad [\mathbf{P}(\mathbf{r}), \mathbf{P}(\mathbf{r}')] = 0. \tag{7}$$

Hence, the coefficients S and T have to fulfil the requirements:

$$\int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega'' \tilde{\mathsf{T}}(\mathbf{r}'', \mathbf{r}, \omega'') \cdot \mathsf{T}^*(\mathbf{r}'', \mathbf{r}', \omega'') - \mathrm{c.c.} = 0 \tag{8}$$

$$\int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega'' \omega'' \tilde{\mathsf{S}}(\mathbf{r}'', \mathbf{r}, \omega'') \cdot \mathsf{T}^*(\mathbf{r}'', \mathbf{r}', \omega'') + \mathrm{c.c.} = \mathsf{I}\, \delta(\mathbf{r} - \mathbf{r}') \tag{9}$$

$$\int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega'' \omega''^2 \tilde{\mathsf{S}}(\mathbf{r}'', \mathbf{r}, \omega'') \cdot \mathsf{S}^*(\mathbf{r}'', \mathbf{r}', \omega'') - \mathrm{c.c.} = 0 \tag{10}$$

where the tilde denotes the transpose of a tensor and the asterisk the complex conjugate.

Furthermore, the Hamiltonian should contain terms describing the interaction between the field and the medium. Two contributions can be distinguished: a transverse part and a longitudinal part. In a minimal-coupling scheme, which we shall adopt here, the transverse part is a bilinear expression involving the transverse vector potential $\mathbf{A}$ and the canonical momentum density $\mathbf{W}$. To ensure compatibility with Maxwell's equations an expression quadratic in $\mathbf{A}$ should be present as well, as we shall see in the following. For dielectrics with spatial dispersion both expressions are non-local. The general form of the transverse contribution to the interaction Hamiltonian is

$$H_\mathrm{i} = -\hbar \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}'\, \mathbf{W}(\mathbf{r}) \cdot \mathsf{F}_1(\mathbf{r}, \mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') + \frac{1}{2}\hbar \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' \mathbf{A}(\mathbf{r}) \cdot \mathsf{F}_2(\mathbf{r}, \mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') \tag{11}$$

with real tensorial coefficients F_1 and F_2 that will be fixed in due time. In view of the form of the second term we may take F_2 to be symmetric upon interchanging both its spatial variables and its indices.

The longitudinal contribution to the interaction Hamiltonian is given by the electrostatic energy involving the polarization density, which reads

$$H_\mathrm{es} = \frac{1}{2\varepsilon_0} \int \mathrm{d}\mathbf{r} \{[\mathbf{P}(\mathbf{r})]_\mathrm{L}\}^2 = \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' \frac{\nabla \cdot \mathbf{P}(\mathbf{r}) \nabla' \cdot \mathbf{P}(\mathbf{r}')}{8\pi\varepsilon_0 |\mathbf{r} - \mathbf{r}'|}. \tag{12}$$

Here the subscript L denotes the longitudinal part of the polarization density, which is defined as $[\mathbf{P}(\mathbf{r})]_L = -\nabla \int \mathrm{d}\mathbf{r}'\, \mathbf{P}(\mathbf{r}') \cdot \nabla(4\pi|\mathbf{r} - \mathbf{r}'|)^{-1}$. Furthermore, ∇' is the spatial derivative acting on a function of $\mathbf{r}'$.

The total Hamiltonian $H = H_\mathrm{f} + H_\mathrm{m} + H_\mathrm{i} + H_\mathrm{es}$ is given by the sum of (1), (3), (11) and (12). It depends on the tensorial coefficients $\mathsf{F}_1, \mathsf{F}_2$ and implicitly on T and S through $\mathbf{P}$ and $\mathbf{W}$. All of these coefficients can as yet be chosen at will, as long as the identities (8)–(10) are satisfied. To derive constraints on these coefficients we turn to the equations of motion.

The Heisenberg equations of motion that follow from the total Hamiltonian are

$$\dot{\mathbf{A}}(\mathbf{r},t)=\frac{1}{\varepsilon_0}\mathbf{\Pi}(\mathbf{r},t) \tag{13}$$

$$\dot{\mathbf{\Pi}}(\mathbf{r},t)=\frac{1}{\mu_0}\Delta\mathbf{A}(\mathbf{r},t)+\hbar\int \mathrm{d}\mathbf{r}'\,\mathbf{W}(\mathbf{r}',t)\cdot[\mathsf{F}_1(\mathbf{r}',\mathbf{r})]_{\mathrm{T}}-\hbar\int \mathrm{d}\mathbf{r}'[\mathsf{F}_2(\mathbf{r},\mathbf{r}')]_{\mathrm{T}}\cdot\mathbf{A}(\mathbf{r}',t) \tag{14}$$

$$\dot{\mathbf{C}}_{\mathrm{m}}(\mathbf{r},\omega,t)=-\mathrm{i}\omega\mathbf{C}_{\mathrm{m}}(\mathbf{r},\omega,t)-\mathrm{i}\omega\int \mathrm{d}\mathbf{r}'\int \mathrm{d}\mathbf{r}''\mathsf{S}^*(\mathbf{r},\mathbf{r}',\omega)\cdot\mathsf{F}_1(\mathbf{r}',\mathbf{r}'')\cdot\mathbf{A}(\mathbf{r}'',t)$$
$$+\frac{1}{\varepsilon_0}\int \mathrm{d}\mathbf{r}'\,\mathsf{T}^*(\mathbf{r},\mathbf{r}',\omega)\cdot[\mathbf{P}(\mathbf{r}',t)]_{\mathrm{L}'} \tag{15}$$

where all operators now depend on time. The subscript L' denotes the longitudinal part with respect to $\mathbf{r}'$. The time derivative of the polarization density follows by combining (5) and (15):

$$\dot{\mathbf{P}}(\mathbf{r},t)=-\hbar\int \mathrm{d}\mathbf{r}'\int_0^\infty \mathrm{d}\omega'\omega'\mathbf{C}_{\mathrm{m}}(\mathbf{r}',\omega',t)\cdot\mathsf{T}(\mathbf{r}',\mathbf{r},\omega')-\hbar\int \mathrm{d}\mathbf{r}'\int \mathrm{d}\mathbf{r}''$$
$$\times\int \mathrm{d}\mathbf{r}'''\int_0^\infty \mathrm{d}\omega'\omega'\tilde{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega')\cdot\mathsf{S}^*(\mathbf{r}',\mathbf{r}'',\omega')\cdot\mathsf{F}_1(\mathbf{r}'',\mathbf{r}''')\cdot\mathbf{A}(\mathbf{r}''',t)$$
$$-\mathrm{i}\frac{\hbar}{\varepsilon_0}\int \mathrm{d}\mathbf{r}'\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega'\tilde{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega')\cdot\mathsf{T}^*(\mathbf{r}',\mathbf{r}'',\omega')\cdot[\mathbf{P}(\mathbf{r}'',t)]_{\mathrm{L}''}+\mathrm{h.c.} \tag{16}$$

where h.c. denotes the Hermitian conjugate of all preceding terms. Upon using (9) one finds that the second term (together with its Hermitian conjugate) equals $-\hbar\int \mathrm{d}\mathbf{r}'\mathsf{F}_1(\mathbf{r},\mathbf{r}')\cdot\mathbf{A}(\mathbf{r}',t)$. Furthermore, the last term (again together with its Hermitian conjugate) vanishes on account of (8).

Eliminating $\mathbf{\Pi}$ from (13) and (14) we find an inhomogeneous wave equation for the vector potential

$$\Delta\mathbf{A}(\mathbf{r},t)-\frac{1}{c^2}\ddot{\mathbf{A}}(\mathbf{r},t)=-\mu_0\hbar\int \mathrm{d}\mathbf{r}'\mathbf{W}(\mathbf{r}',t)\cdot[\mathsf{F}_1(\mathbf{r}',\mathbf{r})]_{\mathrm{T}}+\mu_0\hbar\int \mathrm{d}\mathbf{r}'[\mathsf{F}_2(\mathbf{r},\mathbf{r}')]_{\mathrm{T}}\cdot\mathbf{A}(\mathbf{r}',t) \tag{17}$$

where the first term at the right-hand side can be expressed in terms of the medium operators by substituting (6). According to the Maxwell equations the vector-potential source term, which is given by the right-hand side of (17), should equal $-\mu_0[\dot{\mathbf{P}}(\mathbf{r},t)]_{\mathrm{T}}$. Hence, comparison with (16) leads to the identity

$$-\hbar\int \mathrm{d}\mathbf{r}'\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega'\omega'\{\mathbf{C}_{\mathrm{m}}(\mathbf{r}',\omega',t)\cdot\mathsf{S}(\mathbf{r}',\mathbf{r}'',\omega')+\mathbf{C}_{\mathrm{m}}^\dagger(\mathbf{r}',\omega',t)\cdot\mathsf{S}^*(\mathbf{r}',\mathbf{r}'',\omega')\}$$
$$\cdot[\mathsf{F}_1(\mathbf{r}'',\mathbf{r})]_{\mathrm{T}}-\hbar\int \mathrm{d}\mathbf{r}'[\mathsf{F}_2(\mathbf{r},\mathbf{r}']_{\mathrm{T}}\cdot\mathbf{A}(\mathbf{r}',t)$$
$$=-\hbar\int \mathrm{d}\mathbf{r}'\int_0^\infty \mathrm{d}\omega'\omega'\{\mathbf{C}_{\mathrm{m}}(\mathbf{r}',\omega',t)\cdot[\mathsf{T}(\mathbf{r}',\mathbf{r},\omega')]_{\mathrm{T}}$$
$$+\mathbf{C}_{\mathrm{m}}^\dagger(\mathbf{r}',\omega',t)\cdot[\mathsf{T}^*(\mathbf{r}',\mathbf{r},\omega')]_{\mathrm{T}}\}-\hbar\int \mathrm{d}\mathbf{r}'[\mathsf{F}_1(\mathbf{r},\mathbf{r}')]_{\mathrm{T}}\cdot\mathbf{A}(\mathbf{r}',t). \tag{18}$$

Upon equating the coefficient of the vector potential we arrive at the relation $[\mathsf{F}_1(\mathbf{r},\mathbf{r}')]_{\mathrm{TT}'}=[\mathsf{F}_2(\mathbf{r},\mathbf{r}')]_{\mathrm{TT}'}$, which connects the transverse parts of the tensors F_1 and F_2. A second relation, namely $[\mathsf{T}(\mathbf{r},\mathbf{r}',\omega)]_{\mathrm{T}'}=\int \mathrm{d}\mathbf{r}''\mathsf{S}(\mathbf{r},\mathbf{r}'',\omega)\cdot[\mathsf{F}_1(\mathbf{r}'',\mathbf{r}')]_{\mathrm{T}'}$, follows by equating the coefficient of $\mathbf{C}_{\mathrm{m}}$. Combining these two relations with (8)–(10) we thus have found that the tensors T,

S, F_1 and F_2 have to satisfy five conditions. Apart from these constraints the coefficients can be chosen freely while constructing our model Hamiltonian. We shall use this freedom to impose instead of the relations following from (18) two somewhat stronger conditions that result upon including the longitudinal parts:

$$\mathsf{F}_1(\mathbf{r},\mathbf{r}') = \mathsf{F}_2(\mathbf{r},\mathbf{r}') \tag{19}$$

$$\mathsf{T}(\mathbf{r},\mathbf{r}',\omega) = \int \mathrm{d}\mathbf{r}''\,\mathsf{S}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathsf{F}_1(\mathbf{r}'',\mathbf{r}'). \tag{20}$$

As the first of these equalities shows, the bilinear coupling of $\mathbf{W}$ with $\mathbf{A}$ and the quadratic vector-potential term in (11) have to occur simultaneously. This is a well-known consequence of the minimal-coupling scheme. In view of (19) we shall omit the subscripts of F_i in the following. Furthermore, we shall use (20) to eliminate S from the formalism altogether.

Summarizing the above results, we have obtained the following Hamiltonian for a linear inhomogeneous anisotropic dispersive dielectric interacting with the electromagnetic field:

$$\begin{aligned}H = &\int \mathrm{d}\mathbf{r}\left\{\frac{1}{2\varepsilon_0}[\mathbf{\Pi}(\mathbf{r})]^2 + \frac{1}{2\mu_0}[\nabla\wedge\mathbf{A}(\mathbf{r})]^2\right\} + \hbar\int \mathrm{d}\mathbf{r}\int_0^\infty \mathrm{d}\omega\,\omega\mathbf{C}_{\mathrm{m}}^\dagger(\mathbf{r},\omega)\cdot\mathbf{C}_{\mathrm{m}}(\mathbf{r},\omega)\\ &+\hbar\int \mathrm{d}\mathbf{r}\int \mathrm{d}\mathbf{r}'\int_0^\infty \mathrm{d}\omega\,\omega\left[\mathbf{C}_{\mathrm{m}}(\mathbf{r},\omega)\cdot\mathsf{T}(\mathbf{r},\mathbf{r}',\omega) + \mathbf{C}_{\mathrm{m}}^\dagger(\mathbf{r},\omega)\cdot\mathsf{T}^*(\mathbf{r},\mathbf{r}',\omega)\right]\cdot\mathbf{A}(\mathbf{r}')\\ &+\frac{1}{2}\hbar\int \mathrm{d}\mathbf{r}\int \mathrm{d}\mathbf{r}'\mathbf{A}(\mathbf{r})\cdot\mathsf{F}(\mathbf{r},\mathbf{r}')\cdot\mathbf{A}(\mathbf{r}') + \frac{1}{2\varepsilon_0}\int \mathrm{d}\mathbf{r}\,\{[\mathbf{P}(\mathbf{r})]_{\mathrm{L}}\}^2.\end{aligned} \tag{21}$$

The complex tensorial coefficient T can be chosen freely. It has to satisfy two constraints, the first of which has been written already in (8). The second one follows by substituting (20) in (10):

$$\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''\,\omega''^2\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r},\omega'')\cdot\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'') - \mathrm{c.c.} = 0. \tag{22}$$

Finally, the insertion of (20) in (9) leads to the following equality:

$$\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''\,\omega''\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r},\omega'')\cdot\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'') + \mathrm{c.c.} = \mathsf{F}(\mathbf{r},\mathbf{r}'). \tag{23}$$

This relation defines the real tensor F in terms of T. It shows that $\mathsf{F}(\mathbf{r},\mathbf{r}')$ satisfies the symmetry property $\tilde{\mathsf{F}}(\mathbf{r},\mathbf{r}') = \mathsf{F}(\mathbf{r}',\mathbf{r})$, as we know already from the way F_2 occurs in (11). As an integral kernel the tensor $\mathsf{F}(\mathbf{r},\mathbf{r}')$ is positive-definite. This is established by taking the scalar products of (23) with real vectors $\mathbf{v}(\mathbf{r})$ and $\mathbf{v}(\mathbf{r}')$, and integrating over $\mathbf{r}$ and $\mathbf{r}'$. The result is positive for any choice of $\mathbf{v}$. As a consequence, the inverse of F is well defined.

The polarization density is given by (5), while the canonical momentum density reads according to (6) with (20):

$$\mathbf{W}(\mathbf{r}) = -\int \mathrm{d}\mathbf{r}'\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega'\,\omega'\mathbf{C}_{\mathrm{m}}(\mathbf{r}',\omega')\cdot\mathsf{T}(\mathbf{r}',\mathbf{r}'',\omega')\cdot\mathsf{F}^{-1}(\mathbf{r}'',\mathbf{r}) + \mathrm{h.c.} \tag{24}$$

where the right-hand side contains the inverse of F.

The Hamiltonian (21) has been constructed by starting from general forms for its parts H_{f}, H_{m}, H_{i} and H_{es} and requiring consistency with Maxwell's equations. It may be related to a Lagrange formalism, as is shown in appendix A.

In the following, we shall investigate the dynamics of the model defined by (21). As the Hamiltonian is quadratic in the dynamical variables it is possible to accomplish a complete diagonalization. This will be the subject of the next section.

3. Diagonalization of the Hamiltonian

We wish to find a diagonal representation of the Hamiltonian (21) in the form

$$H = \hbar \int \mathrm{d}\mathbf{r} \int_0^\infty \mathrm{d}\omega\, \omega\, \mathbf{C}^\dagger(\mathbf{r}, \omega) \cdot \mathbf{C}(\mathbf{r}, \omega). \tag{25}$$

The creation and annihilation operators satisfy the standard commutation relations of the form (4). They are linear combinations of the dynamical variables in (21):

$$\mathbf{C}(\mathbf{r}, \omega) = \int \mathrm{d}\mathbf{r}' \left\{ \mathbf{f}_1(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{A}(\mathbf{r}') + \mathbf{f}_2(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{\Pi}(\mathbf{r}') \right.$$
$$\left. + \int_0^\infty \mathrm{d}\omega' \left[\mathbf{f}_3(\mathbf{r}, \mathbf{r}', \omega, \omega') \cdot \mathbf{C}_\mathrm{m}(\mathbf{r}', \omega') + \mathbf{f}_4(\mathbf{r}, \mathbf{r}', \omega, \omega') \cdot \mathbf{C}_\mathrm{m}^\dagger(\mathbf{r}', \omega') \right] \right\} \tag{26}$$

with as-yet unknown tensorial coefficients $\mathbf{f}_i$, the first two of which are taken to be transverse in their second argument. To determine $\mathbf{f}_i$ we use Fano's method [28]: we evaluate the commutator $[\mathbf{C}(\mathbf{r}, \omega), H]$ and equate the result to $\hbar\omega\mathbf{C}(\mathbf{r}, \omega)$. Comparing the contributions involving the various canonical operators we arrive at the four equations

$$\frac{\mathrm{i}}{\varepsilon_0} \mathbf{f}_1(\mathbf{r}, \mathbf{r}', \omega) = \omega \mathbf{f}_2(\mathbf{r}, \mathbf{r}', \omega) \tag{27}$$

$$\frac{\mathrm{i}}{\mu_0} \Delta' \mathbf{f}_2(\mathbf{r}, \mathbf{r}', \omega) - \mathrm{i}\hbar \int \mathrm{d}\mathbf{r}'' \mathbf{f}_2(\mathbf{r}, \mathbf{r}'', \omega) \cdot [\mathsf{F}(\mathbf{r}'', \mathbf{r}')]_{\mathrm{T}'}$$
$$+ \int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega'' \omega'' \{ \mathbf{f}_3(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot [\mathsf{T}^*(\mathbf{r}'', \mathbf{r}', \omega'')]_{\mathrm{T}'}$$
$$- \mathbf{f}_4(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot [\mathsf{T}(\mathbf{r}'', \mathbf{r}', \omega'')]_{\mathrm{T}'} \} = \omega \mathbf{f}_1(\mathbf{r}, \mathbf{r}', \omega) \tag{28}$$

$$-\mathrm{i}\hbar\omega' \int \mathrm{d}\mathbf{r}'' \mathbf{f}_2(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathsf{T}}(\mathbf{r}', \mathbf{r}'', \omega') + \omega' \mathbf{f}_3(\mathbf{r}, \mathbf{r}', \omega, \omega')$$
$$+ \frac{\hbar}{\varepsilon_0} \int \mathrm{d}\mathbf{r}'' \int \mathrm{d}\mathbf{r}''' \int_0^\infty \mathrm{d}\omega'' \{ \mathbf{f}_3(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot [\mathsf{T}^*(\mathbf{r}'', \mathbf{r}''', \omega'')]_{\mathrm{L}'''}$$
$$+ \mathbf{f}_4(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot [\mathsf{T}(\mathbf{r}'', \mathbf{r}''', \omega'')]_{\mathrm{L}'''} \} \cdot \tilde{\mathsf{T}}(\mathbf{r}', \mathbf{r}''', \omega') = \omega \mathbf{f}_3(\mathbf{r}, \mathbf{r}', \omega, \omega') \tag{29}$$

$$-\mathrm{i}\hbar\omega' \int \mathrm{d}\mathbf{r}'' \mathbf{f}_2(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathsf{T}}^*(\mathbf{r}', \mathbf{r}'', \omega') - \omega' \mathbf{f}_4(\mathbf{r}, \mathbf{r}', \omega, \omega')$$
$$- \frac{\hbar}{\varepsilon_0} \int \mathrm{d}\mathbf{r}'' \int \mathrm{d}\mathbf{r}''' \int_0^\infty \mathrm{d}\omega'' \{ \mathbf{f}_3(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot [\mathsf{T}^*(\mathbf{r}'', \mathbf{r}''', \omega'')]_{\mathrm{L}'''}$$
$$+ \mathbf{f}_4(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot [\mathsf{T}(\mathbf{r}'', \mathbf{r}''', \omega'')]_{\mathrm{L}'''} \} \cdot \tilde{\mathsf{T}}^*(\mathbf{r}', \mathbf{r}''', \omega') = \omega \mathbf{f}_4(\mathbf{r}, \mathbf{r}', \omega, \omega'). \tag{30}$$

The solution of these equations can be obtained by a method that is a generalization of that used in our earlier work [19]. The details are given in appendices B and C. The results are

$$\mathbf{f}_1(\mathbf{r}, \mathbf{r}', \omega) = \frac{\omega^2}{c^2} \int \mathrm{d}\mathbf{r}'' \mathsf{T}^*(\mathbf{r}, \mathbf{r}'', \omega) \cdot [\mathsf{G}(\mathbf{r}'', \mathbf{r}', \omega - \mathrm{i}0)]_{\mathrm{T}'} \tag{31}$$

$$\mathbf{f}_2(\mathbf{r}, \mathbf{r}', \omega) = \mathrm{i}\mu_0\omega \int \mathrm{d}\mathbf{r}'' \mathsf{T}^*(\mathbf{r}, \mathbf{r}'', \omega) \cdot [\mathsf{G}(\mathbf{r}'', \mathbf{r}', \omega - \mathrm{i}0)]_{\mathrm{T}'} \tag{32}$$

$$\mathbf{f}_3(\mathbf{r}, \mathbf{r}', \omega, \omega') = \mathsf{I}\,\delta(\mathbf{r} - \mathbf{r}')\delta(\omega - \omega') - \mu_0\hbar\omega \int \mathrm{d}\mathbf{r}'' \int \mathrm{d}\mathbf{r}''' \mathsf{T}^*(\mathbf{r}, \mathbf{r}'', \omega)$$
$$\cdot [\mathsf{G}(\mathbf{r}'', \mathbf{r}''', \omega - \mathrm{i}0)]_{\mathrm{T}'''} \cdot \tilde{\mathsf{T}}(\mathbf{r}', \mathbf{r}''', \omega')$$

$$+\mu_0\hbar\frac{\omega^2}{\omega-\omega'-\mathrm{i}0}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}''',\omega') \tag{33}$$

$$\mathbf{f}_4(\mathbf{r},\mathbf{r}',\omega,\omega')=\mu_0\hbar\omega\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot[\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)]_{\mathrm{T}'''}\cdot\tilde{\mathbf{T}}^*(\mathbf{r}',\mathbf{r}''',\omega')$$
$$-\mu_0\hbar\frac{\omega^2}{\omega+\omega'}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\cdot\tilde{\mathbf{T}}^*(\mathbf{r}',\mathbf{r}''',\omega'). \tag{34}$$

The Green function $\mathbf{G}(\mathbf{r},\mathbf{r}',z)$ occurring in these expressions is defined as the solution of the differential equation:

$$-[\mathbf{G}(\mathbf{r},\mathbf{r}',z)\times\overleftarrow{\nabla}']\times\overleftarrow{\nabla}'+\frac{z^2}{c^2}\mathbf{G}(\mathbf{r},\mathbf{r}',z)+\frac{z^2}{c^2}\int \mathrm{d}\mathbf{r}''\mathbf{G}(\mathbf{r},\mathbf{r}'',z)\cdot\boldsymbol{\chi}(\mathbf{r}'',\mathbf{r}',z)=\mathbf{I}\,\delta(\mathbf{r}-\mathbf{r}'). \tag{35}$$

The spatial derivative operator ∇' acts to the left on the argument $\mathbf{r}'$ of $\mathbf{G}(\mathbf{r},\mathbf{r}',z)$. According to this inhomogeneous wave equation the Green function determines the propagation of waves through a medium that is characterized by a tensor $\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',z)$. The latter plays the role of a non-local anisotropic susceptibility, as will become clear in the next section. It is defined in terms of $\mathbf{T}$ and its complex conjugate as

$$\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',z)\equiv\frac{\hbar}{\varepsilon_0}\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega\left[\frac{1}{\omega-z}\tilde{\mathbf{T}}(\mathbf{r}'',\mathbf{r},\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega)+\frac{1}{\omega+z}\tilde{\mathbf{T}}^*(\mathbf{r}'',\mathbf{r},\omega)\cdot\mathbf{T}(\mathbf{r}'',\mathbf{r}',\omega)\right] \tag{36}$$

with the frequency argument z either in the upper half or the lower half of the complex z-plane, which has got a cut along the real axis. Likewise, the Green function in (35) is defined in the complex z-plane with a cut along the real axis. Both the susceptibility and the Green function are discontinuous across the cut.

We have succeeded now in finding the diagonal representation of the Hamiltonian of our model. The diagonalizing operators are given by (26), with coefficients that are listed in (31)–(34).

4. Field, polarization and susceptibility

Once we have the diagonal representation of the Hamiltonian at our disposal, we can determine the full time evolution of the dynamical variables. In the following we will derive the time dependence of the vector potential, the electric field and the polarization. As we shall need a few properties of the tensors $\boldsymbol{\chi}$ and $\mathbf{G}$, we shall discuss these first.

From its definition (36) it follows that the tensor $\boldsymbol{\chi}$ satisfies the symmetry relations

$$\tilde{\boldsymbol{\chi}}(\mathbf{r},\mathbf{r}',z)=\boldsymbol{\chi}(\mathbf{r}',\mathbf{r},-z) \tag{37}$$

and

$$\boldsymbol{\chi}^*(\mathbf{r},\mathbf{r}',z)=\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',-z^*) \tag{38}$$

so that $\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',z)$ is real on the imaginary axis. The discontinuity across the cut along the real axis is given by

$$\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)-\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',\omega-\mathrm{i}0)=\frac{2\pi\mathrm{i}\hbar}{\varepsilon_0}\int \mathrm{d}\mathbf{r}''\,\tilde{\mathbf{T}}(\mathbf{r}'',\mathbf{r},\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega) \tag{39}$$

for positive ω and by

$$\chi(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)-\chi(\mathbf{r},\mathbf{r}',\omega-\mathrm{i}0)=-\frac{2\pi\mathrm{i}\hbar}{\varepsilon_0}\int\mathrm{d}\mathbf{r}''\,\tilde{\mathsf{T}}^*(\mathbf{r}'',\mathbf{r},-\omega)\cdot\mathsf{T}(\mathbf{r}'',\mathbf{r}',-\omega)\tag{40}$$

for negative ω. Hence, we may write (36) as

$$\chi(\mathbf{r},\mathbf{r}',z)=\frac{1}{2\pi\mathrm{i}}\int_{-\infty}^{\infty}\mathrm{d}\omega\frac{1}{\omega-z}[\chi(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)-\chi(\mathbf{r},\mathbf{r}',\omega-\mathrm{i}0)]\tag{41}$$

which is the well-known Kramers–Kronig relation for the Fourier transform of a causal function. The identities (8), (23) and (22) can be rewritten in terms of the discontinuity across the cut:

$$\int_{-\infty}^{\infty}\mathrm{d}\omega[\chi(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)-\chi(\mathbf{r},\mathbf{r}',\omega-\mathrm{i}0)]=0\tag{42}$$

$$\int_{-\infty}^{\infty}\mathrm{d}\omega\,\omega[\chi(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)-\chi(\mathbf{r},\mathbf{r}',\omega-\mathrm{i}0)]=\frac{2\pi\mathrm{i}\hbar}{\varepsilon_0}\mathsf{F}(\mathbf{r},\mathbf{r}')\tag{43}$$

$$\int_{-\infty}^{\infty}\mathrm{d}\omega\,\omega^2[\chi(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)-\chi(\mathbf{r},\mathbf{r}',\omega-\mathrm{i}0)]=0.\tag{44}$$

Incidentally, we remark that for large $|z|$ the asymptotic behaviour of χ follows from (41) with (42)–(44) as

$$\chi(\mathbf{r},\mathbf{r}',z)\simeq-\frac{\hbar}{\varepsilon_0}\mathsf{F}(\mathbf{r},\mathbf{r}')\frac{1}{z^2}+\mathcal{O}\left(\frac{1}{z^4}\right).\tag{45}$$

The above symmetry properties can be used to prove analogous symmetry relations for the Green function G. By taking the complex conjugate of (35) and using (38) one derives

$$\mathsf{G}^*(\mathbf{r},\mathbf{r}',z)=\mathsf{G}(\mathbf{r},\mathbf{r}',-z^*).\tag{46}$$

The adjoint equation of (35) reads

$$-\nabla\times[\nabla\times\mathsf{G}(\mathbf{r},\mathbf{r}',z)]+\frac{z^2}{c^2}\mathsf{G}(\mathbf{r},\mathbf{r}',z)+\frac{z^2}{c^2}\int\mathrm{d}\mathbf{r}''\,\chi(\mathbf{r},\mathbf{r}'',z)\cdot\mathsf{G}(\mathbf{r}'',\mathbf{r}',z)=\mathsf{I}\,\delta(\mathbf{r}-\mathbf{r}')\tag{47}$$

as follows from (35) after multiplication by $\mathsf{G}(\mathbf{r}',\mathbf{r}'',z)$, integration over $\mathbf{r}'$ and a partial integration. Comparing this differential equation to that obtained by taking the transpose of (35) and interchanging the position variables one finds with the use of (37) the reciprocity relation:

$$\tilde{\mathsf{G}}(\mathbf{r},\mathbf{r}',z)=\mathsf{G}(\mathbf{r}',\mathbf{r},-z).\tag{48}$$

Having obtained the relevant physical properties of the tensors χ and G, we return to a discussion of the time dependence of the dynamical variables. Inverting (26) by means of the canonical commutation relations we get

$$\mathbf{A}(\mathbf{r})=\mathrm{i}\hbar\int\mathrm{d}\mathbf{r}'\int_0^{\infty}\mathrm{d}\omega\,\tilde{\mathbf{f}}_2^*(\mathbf{r}',\mathbf{r},\omega)\cdot\mathbf{C}(\mathbf{r}',\omega)+\mathrm{h.c.}\tag{49}$$

Substitution of (32) yields with the help of (46) and (48)

$$\mathbf{A}(\mathbf{r},t)=\mu_0\hbar\int\mathrm{d}\mathbf{r}'\int\mathrm{d}\mathbf{r}''\int_0^{\infty}\mathrm{d}\omega\,\omega[\mathsf{G}(\mathbf{r},\mathbf{r}',\omega+\mathrm{i}0)]_{\mathrm{T}}\cdot\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r}',\omega)\cdot\mathbf{C}(\mathbf{r}'',\omega)\,\mathrm{e}^{-\mathrm{i}\omega t}+\mathrm{h.c.}\tag{50}$$

where we accounted for the time dependence of the diagonalizing operator $\mathbf{C}(\mathbf{r}'',\omega)$.

By differentiation with respect to space and time we can easily determine the time evolution of the electromagnetic fields. Taking the curl of (50) we get

$$\mathbf{B}(\mathbf{r},t) = \mu_0\hbar \int d\mathbf{r}' \int d\mathbf{r}'' \int_0^\infty d\omega\, \omega \nabla \times \mathsf{G}(\mathbf{r},\mathbf{r}',\omega+i0)\cdot\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r}',\omega)\cdot\mathbf{C}(\mathbf{r}'',\omega)\,e^{-i\omega t} + \text{h.c.} \tag{51}$$

Furthermore, the transverse part of the electric field follows from (50) by differentiation with respect to t:

$$[\mathbf{E}(\mathbf{r},t)]_T = i\mu_0\hbar \int d\mathbf{r}' \int d\mathbf{r}'' \int_0^\infty d\omega\, \omega^2 [\mathsf{G}(\mathbf{r},\mathbf{r}',\omega+i0)]_T \cdot\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r}',\omega)\cdot\mathbf{C}(\mathbf{r}'',\omega)\,e^{-i\omega t} + \text{h.c.} \tag{52}$$

To determine the longitudinal part of the electric field we first have to derive an expression for the polarization.

The medium operator $\mathbf{C}_m(\mathbf{r},\omega)$ is a linear combination of the diagonalizing operator and its Hermitian conjugate:

$$\mathbf{C}_m(\mathbf{r},\omega) = \int d\mathbf{r}' \int_0^\infty d\omega' [\tilde{\mathbf{f}}_3^*(\mathbf{r}',\mathbf{r},\omega',\omega)\cdot\mathbf{C}(\mathbf{r}',\omega') - \tilde{\mathbf{f}}_4(\mathbf{r}',\mathbf{r},\omega',\omega)\cdot\mathbf{C}^\dagger(\mathbf{r}',\omega')] \tag{53}$$

as follows by taking the inverse of (26). Substituting (33)–(34) and inserting the result in (5) we get after some algebra

$$\mathbf{P}(\mathbf{r},t) = \frac{i\hbar}{c^2} \int d\mathbf{r}' \int d\mathbf{r}'' \int d\mathbf{r}''' \int_0^\infty d\omega\, \omega^2 \chi(\mathbf{r},\mathbf{r}',\omega+i0)\cdot\mathsf{G}(\mathbf{r}',\mathbf{r}'',\omega+i0)\cdot\tilde{\mathsf{T}}(\mathbf{r}''',\mathbf{r}'',\omega) \cdot\mathbf{C}(\mathbf{r}''',\omega)\,e^{-i\omega t} - i\hbar \int d\mathbf{r}' \int_0^\infty d\omega\, \tilde{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega)\cdot\mathbf{C}(\mathbf{r}',\omega)\,e^{-i\omega t} + \text{h.c.} \tag{54}$$

where we have employed (8) and (36). The longitudinal part of this expression can be rewritten with the use of (47) as:

$$[\mathbf{P}(\mathbf{r},t)]_L = -\frac{i\hbar}{c^2} \int d\mathbf{r}' \int d\mathbf{r}'' \int_0^\infty d\omega\, \omega^2 [\mathsf{G}(\mathbf{r},\mathbf{r}',\omega+i0)]_L \cdot\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r}',\omega)\cdot\mathbf{C}(\mathbf{r}'',\omega)\,e^{-i\omega t} + \text{h.c.} \tag{55}$$

From the Maxwell equation $\nabla\cdot(\varepsilon_0\mathbf{E}+\mathbf{P}) = 0$ it follows that the left-hand side is proportional to the longitudinal part $[\mathbf{E}(\mathbf{r},t)]_L$ of the electric field. The ensuing expression for the latter is analogous to (52), so that we arrive at the following result for the complete electric field:

$$\mathbf{E}(\mathbf{r},t) = i\mu_0\hbar \int d\mathbf{r}' \int d\mathbf{r}'' \int_0^\infty d\omega\, \omega^2 \mathsf{G}(\mathbf{r},\mathbf{r}',\omega+i0)\cdot\tilde{\mathsf{T}}(\mathbf{r}'',\mathbf{r}',\omega)\cdot\mathbf{C}(\mathbf{r}'',\omega)\,e^{-i\omega t} + \text{h.c.} \tag{56}$$

Inspection of (54) shows that the polarization consists of two terms. The first term is proportional to the electric field, at least in Fourier space and after taking a spatial convolution integral. The proportionality factor is $\chi(\mathbf{r},\mathbf{r}',\omega)$, which plays the role of a susceptibility tensor, as we anticipated in the previous section. The second term in (54) is not related to the electric field. It represents a noise polarization density $\mathbf{P}_n(\mathbf{r},t)$ defined as

$$\mathbf{P}_n(\mathbf{r},t) = -i\hbar \int d\mathbf{r}' \int_0^\infty d\omega\, \tilde{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega)\cdot\mathbf{C}(\mathbf{r}',\omega)\,e^{-i\omega t} + \text{h.c.} \tag{57}$$

that has to be present so as to yield a quantization scheme in which the validity of the canonical commutation relations in the presence of dissipation is guaranteed. Introducing the Fourier transform $\mathbf{P}_n(\mathbf{r},\omega)$ via

$$\mathbf{P}_n(\mathbf{r},t) = \int_0^\infty d\omega\, \mathbf{P}_n(\mathbf{r},\omega)\,e^{-i\omega t} + \text{h.c.} \tag{58}$$

and its counterparts $\mathbf{E}(\mathbf{r}, \omega)$ and $\mathbf{P}(\mathbf{r}, \omega)$, we get from (54) with (56):

$$\mathbf{P}(\mathbf{r}, \omega) = \int \mathrm{d}\mathbf{r}' \chi(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0) \cdot \mathbf{E}(\mathbf{r}', \omega) + \mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega). \tag{59}$$

The Fourier-transformed noise polarization density is proportional to the diagonalizing operator:

$$\mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega) = -\mathrm{i}\hbar \int \mathrm{d}\mathbf{r}' \, \tilde{\mathsf{T}}(\mathbf{r}', \mathbf{r}, \omega) \cdot \mathbf{C}(\mathbf{r}', \omega), \tag{60}$$

as follows from (57) and (58). As we have got now an explicit expression for $\mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega)$ we can derive its commutation relation. By employing (39) we obtain

$$\left[\mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega), \mathbf{P}_{\mathrm{n}}^{\dagger}(\mathbf{r}', \omega')\right] = -\frac{\mathrm{i}\hbar\varepsilon_0}{2\pi}[\chi(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0) - \chi(\mathbf{r}, \mathbf{r}', \omega - \mathrm{i}0)]\delta(\omega - \omega'). \tag{61}$$

This commutation relation is a generalization of that postulated in the phenomenological quantization scheme for isotropic dielectrics without spatial dispersion [7–10]. In the present approach, we have been able to prove its validity.

Both the fields and the polarization density can be rewritten in terms of $\mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega)$. We get from (51), (54) and (56) upon eliminating $\mathbf{C}(\mathbf{r}, \omega)$ in favour of $\mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega)$:

$$\mathbf{E}(\mathbf{r}, t) = -\mu_0 \int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega\, \omega^2 \mathsf{G}(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0) \cdot \mathbf{P}_{\mathrm{n}}(\mathbf{r}', \omega)\, \mathrm{e}^{-\mathrm{i}\omega t} + \text{h.c.} \tag{62}$$

$$\mathbf{B}(\mathbf{r}, t) = \mathrm{i}\mu_0 \int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega\, \omega \nabla \times \mathsf{G}(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0) \cdot \mathbf{P}_{\mathrm{n}}(\mathbf{r}', \omega)\, \mathrm{e}^{-\mathrm{i}\omega t} + \text{h.c.} \tag{63}$$

$$\begin{aligned}\mathbf{P}(\mathbf{r}, t) = &-\frac{1}{c^2} \int \mathrm{d}\mathbf{r}' \int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega\, \omega^2 \chi(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0) \cdot \mathsf{G}(\mathbf{r}', \mathbf{r}'', \omega + \mathrm{i}0) \cdot \mathbf{P}_{\mathrm{n}}(\mathbf{r}'', \omega)\, \mathrm{e}^{-\mathrm{i}\omega t} \\ &+ \int_0^\infty \mathrm{d}\omega\, \mathbf{P}_{\mathrm{n}}(\mathbf{r}, \omega) \mathrm{e}^{-\mathrm{i}\omega t} + \text{h.c.}\end{aligned} \tag{64}$$

By adding (62) and (64) we get an expression for the dielectric displacement $\mathbf{D}(\mathbf{r}, t)$. Upon using (47) we may write it as

$$\mathbf{D}(\mathbf{r}, t) = -\int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega \nabla \times [\nabla \times \mathsf{G}(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0)] \cdot \mathbf{P}_{\mathrm{n}}(\mathbf{r}', \omega) \mathrm{e}^{-\mathrm{i}\omega t} + \text{h.c.} \tag{65}$$

Clearly, the dielectric displacement is purely transverse. Comparing with (63) we find that Maxwell's equation $\nabla \times \mathbf{B}(\mathbf{r}, t) = \mu_0 \partial \mathbf{D}(\mathbf{r}, t)/\partial t$ is satisfied.

It is instructive to return to the time-dependent representation of the linear constitutive relation (59):

$$\mathbf{P}(\mathbf{r}, t) = \int \mathrm{d}\mathbf{r}' \int_{-\infty}^{t} \mathrm{d}t' \chi(\mathbf{r}, \mathbf{r}', t - t') \cdot \mathbf{E}(\mathbf{r}', t') + \mathbf{P}_{\mathrm{n}}(\mathbf{r}, t) \tag{66}$$

with the time-dependent susceptibility tensor defined by writing

$$\chi(\mathbf{r}, \mathbf{r}', \omega + \mathrm{i}0) = \int_0^\infty \mathrm{d}t\, \chi(\mathbf{r}, \mathbf{r}', t)\, \mathrm{e}^{\mathrm{i}\omega t}. \tag{67}$$

The convolution integral in the first term of (66), which expresses the causal response of the medium, depends on the electric field at all times t' preceding t and at all positions $\mathbf{r}'$, whereas the second contribution is the noise term, which in classical theory does not appear. Sometimes [26] a different splitting of the various contributions to the polarization density is proposed, by writing an equation of the general form of (66) in which the response term covers only a limited range of values of t', for instance $t' \in [0, t]$ for $t > 0$. In such a formulation the convolution integral does not represent the full causal response of the medium, so that part of the response is hidden in the second term. As a consequence, the latter is no longer a pure noise term, so that it cannot be omitted in the classical version of the theory.

The above expressions for the fields and the polarization density in terms of the Fourier-transformed noise polarization density satisfying the commutation relations (61) are the central results in the present formalism for field quantization in inhomogeneous anisotropic dielectric media with spatio-temporal dispersion. Although we are describing dissipative media, it has not been necessary to explicitly introduce a bath, as is commonly done in the context of damped-polariton treatments [15, 16, 19, 20]. In the following section, we shall show how a bath may be identified in the present model.

5. Bath degrees of freedom

In the Hamiltonian (21) the dielectric medium is described by the operators $\mathbf{C}_\mathrm{m}(\mathbf{r},\omega)$ and $\mathbf{C}^\dagger_\mathrm{m}(\mathbf{r},\omega)$. The polarization density $\mathbf{P}(\mathbf{r})$ and its canonical conjugate $\mathbf{W}(\mathbf{r})$ are given in (5) and (24) as suitable linear combinations of the medium operators $\mathbf{C}_\mathrm{m}$ and $\mathbf{C}^\dagger_\mathrm{m}$. Since the latter depend on the continuous variable ω, they describe many more degrees of freedom than $\mathbf{P}$ and $\mathbf{W}$. The extra degrees of freedom can be taken together to define a so-called bath, which is independent of $\mathbf{P}$ and $\mathbf{W}$. Although the name might suggest otherwise, the bath as introduced in this way is part of the medium itself, and not some external environment. Its role is to account for the dissipative effects in the dispersive medium, which may arise for instance through a leak of energy by heat production. In the following we shall identify the operators associated with the bath. Subsequently, we shall show how the Hamiltonian can be rewritten so as to give an explicit description of the coupling between the polarization and the bath. In this way, we will be able to compare our model to its counterparts in previous papers [15, 16, 19, 20].

The bath will be described by operators $\mathbf{C}_\mathrm{b}(\mathbf{r},\omega)$ and $\mathbf{C}^\dagger_\mathrm{b}(\mathbf{r},\omega)$ satisfying the usual commutation relations. These bath operators are linear combinations of the medium operators:

$$\mathbf{C}_\mathrm{b}(\mathbf{r},\omega) = \int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega' \big[\mathsf{H}_1(\mathbf{r},\mathbf{r}',\omega,\omega')\cdot\mathbf{C}_\mathrm{m}(\mathbf{r}',\omega') + \mathsf{H}_2(\mathbf{r},\mathbf{r}',\omega,\omega')\cdot\mathbf{C}^\dagger_\mathrm{m}(\mathbf{r}',\omega')\big] \tag{68}$$

with tensor coefficients H_i that will be determined presently. Since the bath variables are by definition independent of both $\mathbf{P}(\mathbf{r}')$ and $\mathbf{W}(\mathbf{r}')$ for all $\mathbf{r}'$, they have to commute with the latter. With the use of (5) and (24) we get from these commutation relations the following conditions:

$$\int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega'' [\mathsf{H}_1(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'') + \mathsf{H}_2(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega'')] = 0 \tag{69}$$

$$\int \mathrm{d}\mathbf{r}'' \int_0^\infty \omega''\omega'' [\mathsf{H}_1(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'') - \mathsf{H}_2(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega'')] = 0. \tag{70}$$

To determine H_i we start from the following *Ansatz*:

$$\mathsf{H}_1(\mathbf{r},\mathbf{r}',\omega,\omega') = \int \mathrm{d}\mathbf{r}'' \left[\delta(\omega-\omega')\mathsf{h}_1(\mathbf{r},\mathbf{r}'',\omega) + \frac{1}{\omega-\omega'+\mathrm{i}0}\mathsf{h}_2(\mathbf{r},\mathbf{r}'',\omega)\right]\cdot\tilde{\mathsf{T}}(\mathbf{r}',\mathbf{r}'',\omega') \tag{71}$$

$$\mathsf{H}_2(\mathbf{r},\mathbf{r}',\omega,\omega') = -\int \mathrm{d}\mathbf{r}'' \frac{1}{\omega+\omega'}\mathsf{h}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\tilde{\mathsf{T}}^*(\mathbf{r}',\mathbf{r}'',\omega') \tag{72}$$

with new tensor coefficients h_i. Substituting these expressions in (69) and (70) and using (36) and (39), we find that both of these conditions are simultaneously satisfied when h_1 and h_2 are related as

$$\int d\mathbf{r}''\, \mathsf{h}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\chi(\mathbf{r}'',\mathbf{r}',\omega+\mathrm{i}0) = \frac{1}{2\pi\mathrm{i}}\int d\mathbf{r}''\mathsf{h}_1(\mathbf{r},\mathbf{r}'',\omega)\cdot[\chi(\mathbf{r}'',\mathbf{r}',\omega+\mathrm{i}0) - \chi(\mathbf{r}'',\mathbf{r}',\omega-\mathrm{i}0)]. \qquad (73)$$

Hence, we are left with a single independent coefficient. It can be determined by imposing the standard commutation relation of the form (4) for $\mathbf{C}_\mathrm{b}(\mathbf{r},\omega)$ and $\mathbf{C}_\mathrm{b}^\dagger(\mathbf{r}',\omega')$. Using (68) with (71) and (72) we arrive at the condition:

$$\frac{1}{2\mathrm{i}}\int d\mathbf{r}''\int d\mathbf{r}'''\mathsf{h}_1(\mathbf{r},\mathbf{r}'',\omega)\cdot[\chi(\mathbf{r}'',\mathbf{r}''',\omega+\mathrm{i}0)-\chi(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)]\cdot\tilde{\mathsf{h}}_1^*(\mathbf{r}',\mathbf{r}''',\omega) = \frac{\pi\hbar}{\varepsilon_0}\mathsf{I}\,\delta(\mathbf{r}-\mathbf{r}'). \qquad (74)$$

The vanishing of the commutator of $\mathbf{C}_\mathrm{b}(\mathbf{r},\omega)$ with $\mathbf{C}_\mathrm{b}(\mathbf{r}',\omega')$ is warranted on the strength of (73).

In view of (39) a solution of (74) is

$$\mathsf{h}_1(\mathbf{r},\mathbf{r}',\omega) = \tilde{\mathsf{T}}^{-1}(\mathbf{r}',\mathbf{r},\omega) \qquad (75)$$

and hence, on account of (73)

$$\mathsf{h}_2(\mathbf{r},\mathbf{r}',\omega) = \frac{\hbar}{\varepsilon_0}\int d\mathbf{r}''\,\mathsf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot\chi^{-1}(\mathbf{r}'',\mathbf{r}',\omega+\mathrm{i}0). \qquad (76)$$

It should be noted that the coefficients h_i are determined up to a unitary transformation. This freedom, which is available to H_i as well, corresponds to a natural arbitrariness in the choice of the bath operators themselves.

As the bath operators have been identified now, we can rewrite the Hamiltonian so as to clarify their role in the dynamics of our model. To that end we have to eliminate the medium operators $\mathbf{C}_\mathrm{m}$ in favour of the bath operators $\mathbf{C}_\mathrm{b}$. Employing (5), (24) and (68) we can write the medium operators as

$$\mathbf{C}_\mathrm{m}(\mathbf{r},\omega) = \int d\mathbf{r}'\,\mathsf{T}^*(\mathbf{r},\mathbf{r}',\omega)\cdot\left[\frac{\mathrm{i}}{\hbar}\omega\int d\mathbf{r}''\mathsf{F}^{-1}(\mathbf{r}',\mathbf{r}'')\cdot\mathbf{P}(\mathbf{r}'')-\mathbf{W}(\mathbf{r}')\right] + \int d\mathbf{r}'\int_0^\infty d\omega'\big[\tilde{\mathsf{H}}_1^*(\mathbf{r}',\mathbf{r},\omega',\omega)\cdot\mathbf{C}_\mathrm{b}(\mathbf{r}',\omega')-\tilde{\mathsf{H}}_2(\mathbf{r}',\mathbf{r},\omega',\omega)\cdot\mathbf{C}_\mathrm{b}^\dagger(\mathbf{r}',\omega')\big]. \qquad (77)$$

With the use of this expression the contributions involving the medium operators in (21) can be rewritten. In this way, we arrive at the following alternative form for the Hamiltonian of our model:

$$H = \int d\mathbf{r}\left\{\frac{1}{2\varepsilon_0}[\mathbf{\Pi}(\mathbf{r})]^2+\frac{1}{2\mu_0}[\nabla\wedge\mathbf{A}(\mathbf{r})]^2\right\}+\hbar\int d\mathbf{r}\int_0^\infty d\omega\,\omega\,\mathbf{C}_\mathrm{b}^\dagger(\mathbf{r},\omega)\cdot\mathbf{C}_\mathrm{b}(\mathbf{r},\omega)$$
$$+\frac{\varepsilon_0}{2\pi\mathrm{i}\hbar^2}\int d\mathbf{r}\int d\mathbf{r}'\int d\mathbf{r}''\int d\mathbf{r}'''\,\mathbf{P}(\mathbf{r})\cdot\mathsf{F}^{-1}(\mathbf{r},\mathbf{r}')$$
$$\cdot\left\{\int_0^\infty d\omega\,\omega^3[\chi(\mathbf{r}',\mathbf{r}'',\omega+\mathrm{i}0)-\chi(\mathbf{r}',\mathbf{r}'',\omega-\mathrm{i}0)]\right\}\cdot\mathsf{F}^{-1}(\mathbf{r}'',\mathbf{r}''')\cdot\mathbf{P}(\mathbf{r}''')$$
$$+\frac{1}{2\varepsilon_0}\int d\mathbf{r}\,\{[\mathbf{P}(\mathbf{r})]_\mathrm{L}\}^2+\frac{1}{2}\hbar\int d\mathbf{r}\int d\mathbf{r}'\,\mathbf{W}(\mathbf{r})\cdot\mathsf{F}(\mathbf{r},\mathbf{r}')\cdot\mathbf{W}(\mathbf{r}')$$

$$-\hbar\int \mathrm{d}\mathbf{r}\int \mathrm{d}\mathbf{r}'\,\mathbf{W}(\mathbf{r})\cdot\mathsf{F}(\mathbf{r},\mathbf{r}')\cdot\mathbf{A}(\mathbf{r}') + \frac{1}{2}\hbar\int \mathrm{d}\mathbf{r}\int \mathrm{d}\mathbf{r}'\mathbf{A}(\mathbf{r})\cdot\mathsf{F}(\mathbf{r},\mathbf{r}')\cdot\mathbf{A}(\mathbf{r}')$$
$$-\frac{\mathrm{i}\hbar}{\varepsilon_0}\int \mathrm{d}\mathbf{r}\int \mathrm{d}\mathbf{r}'\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega\big[\mathbf{C}_\mathrm{b}^\dagger(\mathbf{r},\omega)\cdot\mathsf{T}^*(\mathbf{r},\mathbf{r}',\omega)$$
$$\cdot\,\chi^{-1}(\mathbf{r}',\mathbf{r}'',\omega+\mathrm{i}0)\cdot\mathbf{P}(\mathbf{r}'') - \mathrm{h.c.}\big]. \tag{78}$$

As before, the tensor coefficients T and F are related by (23), while the susceptibility χ follows from T via (36), which implies (39).

We are now in a position to compare the present model with that discussed in previous papers [15, 16, 19, 20]. The Hamiltonian (78) takes account of anisotropy and spatial dispersion. To make contact with the earlier treatments these features should be left out. In those circumstances both the susceptibility χ and the tensor coefficients T, F are isotropic and local, so that one has for instance:

$$\chi(\mathbf{r},\mathbf{r}',z) = \chi(\mathbf{r},z)\mathbf{I}\delta(\mathbf{r}-\mathbf{r}'). \tag{79}$$

Furthermore, the dielectric medium of the present model has got an arbitrary temporal dispersion: the frequency dependence of the susceptibility is governed by that of T, as is obvious from (36). In our previous treatment of the inhomogeneous damped-polariton model [19, 20] the scalar susceptibility satisfied a sum rule to the effect that the integral $\int_0^\infty \mathrm{d}\omega\,\omega^3[\chi(\mathbf{r},\omega+\mathrm{i}0)-\chi(\mathbf{r},\omega-\mathrm{i}0)]$, a generalization of which occurs in the third term of (78), is proportional to the square of an effective frequency $\tilde{\omega}_0(\mathbf{r})$. The latter parameter already figured in the original Hamiltonian in [15, 16], albeit in a space-independent form. Finally, in our earlier work we followed the notation in [15, 16] by representing the bath operators $\mathbf{C}_\mathrm{b}(\mathbf{r},\omega)$ and their Hermitian conjugates by equivalent position and momentum operators $\mathbf{Y}_\omega(\mathbf{r})$ and $\mathbf{Q}_\omega(\mathbf{r})$. Implementing this alternative notation here as well, one shows that (78) indeed reduces to the Hamiltonian in [19, 20] for the special case of an isotropic spatially-nondispersive medium.

6. Conclusion

The Hamiltonian model that we have considered in this paper is a suitable tool to underpin the quantum formalism for a general linear dielectric medium and of the electromagnetic field propagating through such a medium. By solving our model we have succeeded in giving a justification of the postulates on which the phenomenological quantization scheme for electrodynamics in dielectric media is usually based.

Our model incorporates many features to warrant the generality of the description. Apart from allowing for inhomogeneities and anisotropies of the medium it has the virtue of accommodating a quite general spatio-temporal dispersion. In fact, the susceptibility tensor of the dielectric medium has been identified in (36), which implies the Kramers–Kronig relation (41). According to that relation the susceptibility in the complex frequency plane is determined by the discontinuity across the cut along the real axis. All anisotropic inhomogeneous linear media with spatio-temporal dispersion that respond causally to an external electric field are characterized by a susceptibility tensor $\chi(\mathbf{r},\mathbf{r}',\omega)$ of the form (41). Under the assumption that the dielectric medium is lossy without net gain, the discontinuity of $\chi(\mathbf{r},\mathbf{r}',\omega)$ for $\omega > 0$ is a positive-definite integral kernel. Hence, one may use (39) to introduce a tensor $\mathsf{T}(\mathbf{r},\mathbf{r}',\omega)$, which is uniquely defined up to a unitary transformation. Subsequently, one can construct the model Hamiltonian (21) and proceed with its diagonalization. In conclusion, our formalism applies to all anisotropic lossy dielectric media with a spatio-temporal dispersion that is compatible with the fundamental principles of causality and positive-definiteness of the dissipative energy loss. Incidentally, it may be remarked that amplifying dielectric media, which have been treated in the context of the phenomenological quantization scheme as well [10, 29], are not covered by the present damped-polariton model. To describe media with a sustained gain, e.g. a laser above threshold, one has to incorporate a driving mechanism in the Hamiltonian, which accounts for the ongoing input of energy that is indispensable for a stationary gain.

As we have shown, the time evolution of the dynamical variables for field and matter can be determined completely by deriving the operators that diagonalize the Hamiltonian. The diagonalizing operators are closely related to the noise part of the polarization density, which plays an important role in the phenomenological quantization scheme. The proof of the commutation properties of the noise polarization density follows from its relation to the diagonalizing operators.

In setting up our model Hamiltonian we have avoided to introduce a bath environment from the beginning. The subsequent formalism could be developed without ever discussing such a bath. Nevertheless, one may be interested in an analysis of the complete set of degrees of freedom of the dielectric medium in our model. If that analysis is carried out, one finds, as we have seen above, that specific combinations of medium variables can be associated with what may be called a bath. The coupling of the polarization to this bath can be held responsible for the dissipative losses that characterize a dispersive dielectric.

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Appendix A. Lagrangian formulation

In this appendix we shall show how the Hamiltonian (21) can be related to a Lagrange formalism. We start by postulating the following Lagrangian for an anisotropic linear dielectric with spatio-temporal dispersion that interacts with the electromagnetic field:

$$L = \int \mathrm{d}\mathbf{r} \left\{ \frac{1}{2}\varepsilon_0[\dot{\mathbf{A}}(\mathbf{r})]^2 - \frac{1}{2\mu_0}[\nabla \wedge \mathbf{A}(\mathbf{r})]^2 \right\} + \frac{1}{2}\int \mathrm{d}\mathbf{r} \int_0^\infty \mathrm{d}\omega \{[\dot{\mathbf{Q}}_{\mathrm{m}}(\mathbf{r},\omega)]^2 - \omega^2[\mathbf{Q}_{\mathrm{m}}(\mathbf{r},\omega)]^2\} + \int \mathrm{d}\mathbf{r}\, \dot{\mathbf{P}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) - \frac{1}{2\varepsilon_0}\int \mathrm{d}\mathbf{r} \{[\mathbf{P}(\mathbf{r})]_{\mathrm{L}}\}^2. \tag{A.1}$$

Here $\mathbf{A}(\mathbf{r})$ is the transverse vector potential and $\mathbf{Q}_{\mathrm{m}}(\mathbf{r},\omega)$ are material coordinates depending on position and frequency. The polarization density $\mathbf{P}(\mathbf{r})$ is taken to be an anisotropic and non-local linear combination of these material coordinates of the form

$$\mathbf{P}(\mathbf{r}) = \int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega'\, \mathbf{Q}_{\mathrm{m}}(\mathbf{r}',\omega') \cdot \mathsf{T}_0(\mathbf{r}',\mathbf{r},\omega') \tag{A.2}$$

with a real tensor coefficient $\mathsf{T}_0(\mathbf{r},\mathbf{r}',\omega)$. One easily verifies that the Lagrangian equations have the form

$$\Delta \mathbf{A}(\mathbf{r},t) - \frac{1}{c^2}\ddot{\mathbf{A}}(\mathbf{r},t) = -\mu_0[\dot{\mathbf{P}}(\mathbf{r},t)]_{\mathrm{T}} \tag{A.3}$$

$$\ddot{\mathbf{Q}}_{\mathrm{m}}(\mathbf{r},\omega,t) + \omega^2\mathbf{Q}_{\mathrm{m}}(\mathbf{r},\omega,t) = \int \mathrm{d}\mathbf{r}\, \mathsf{T}_0(\mathbf{r},\mathbf{r}',\omega) \cdot \mathbf{E}(\mathbf{r}',t) \tag{A.4}$$

with the electric field given as $\mathbf{E}(\mathbf{r},t) = -\dot{\mathbf{A}}(\mathbf{r},t) - (1/\varepsilon_0)[\mathbf{P}(\mathbf{r},t)]_{\mathrm{L}}$. The first Lagrangian differential equation is consistent with Maxwell's equation, as it should. The second

Lagrangian equation shows that the material coordinates are harmonic variables that are driven by the electric field in an anisotropic and non-local way.

Introducing the momenta $\mathbf{\Pi}(\mathbf{r})$ and $\mathbf{P}_{\mathrm{m}}(\mathbf{r}, \omega)$ associated with $\mathbf{A}$ and $\mathbf{Q}_{\mathrm{m}}$ as

$$\mathbf{\Pi}(\mathbf{r}) = \varepsilon_0 \dot{\mathbf{A}}(\mathbf{r}) \tag{A.5}$$

$$\mathbf{P}_{\mathrm{m}}(\mathbf{r}, \omega) = \dot{\mathbf{Q}}_{\mathrm{m}}(\mathbf{r}, \omega) + \int \mathrm{d}\mathbf{r}' \, \mathsf{T}_0(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{A}(\mathbf{r}') \tag{A.6}$$

we obtain the Hamiltonian corresponding to (A.1) in the standard fashion. The result is

$$\begin{aligned} H = \int \mathrm{d}\mathbf{r} \left\{ \frac{1}{2\varepsilon_0} [\mathbf{\Pi}(\mathbf{r})]^2 + \frac{1}{2\mu_0} [\nabla \wedge \mathbf{A}(\mathbf{r})]^2 \right\} + \frac{1}{2} \int \mathrm{d}\mathbf{r} \int_0^\infty \mathrm{d}\omega \{ [\mathbf{P}_{\mathrm{m}}(\mathbf{r}, \omega)]^2 + \omega^2 [\mathbf{Q}_{\mathrm{m}}(\mathbf{r}, \omega)]^2 \} \\ - \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' \int_0^\infty \mathrm{d}\omega \, \mathbf{P}_{\mathrm{m}}(\mathbf{r}, \omega) \cdot \mathsf{T}_0(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{A}(\mathbf{r}') \\ + \frac{1}{2} \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' \int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega \, \mathbf{A}(\mathbf{r}) \cdot \tilde{\mathsf{T}}_0(\mathbf{r}', \mathbf{r}, \omega) \cdot \mathsf{T}_0(\mathbf{r}', \mathbf{r}'', \omega) \cdot \mathbf{A}(\mathbf{r}'') \\ + \frac{1}{2\varepsilon_0} \int \mathrm{d}\mathbf{r} \, \{ [\mathbf{P}(\mathbf{r})]_{\mathrm{L}} \}^2. \end{aligned} \tag{A.7}$$

As a final step we wish to rewrite the Hamiltonian in terms of material creation and annihilation operators $\mathbf{C}_{\mathrm{m}}^\dagger$ and $\mathbf{C}_{\mathrm{m}}$ as used in (21) of the main text. We introduce the latter by writing

$$\mathbf{P}_{\mathrm{m}}(\mathbf{r}, \omega) = \left(\frac{\hbar\omega}{2} \right)^{1/2} \int \mathrm{d}\mathbf{r}' \big[\mathbf{C}_{\mathrm{m}}(\mathbf{r}', \omega) \cdot \mathsf{U}(\mathbf{r}', \mathbf{r}, \omega) + \mathbf{C}_{\mathrm{m}}^\dagger(\mathbf{r}', \omega) \cdot \mathsf{U}^*(\mathbf{r}', \mathbf{r}, \omega) \big] \tag{A.8}$$

$$\mathbf{Q}_{\mathrm{m}}(\mathbf{r}, \omega) = \mathrm{i} \left(\frac{\hbar}{2\omega} \right)^{1/2} \int \mathrm{d}\mathbf{r}' \big[\mathbf{C}_{\mathrm{m}}(\mathbf{r}', \omega) \cdot \mathsf{U}(\mathbf{r}', \mathbf{r}, \omega) - \mathbf{C}_{\mathrm{m}}^\dagger(\mathbf{r}', \omega) \cdot \mathsf{U}^*(\mathbf{r}', \mathbf{r}, \omega) \big] \tag{A.9}$$

with tensorial coefficients U that satisfy the unitarity condition

$$\int \mathrm{d}\mathbf{r}'' \tilde{\mathsf{U}}(\mathbf{r}'', \mathbf{r}, \omega) \cdot \mathsf{U}^*(\mathbf{r}'', \mathbf{r}', \omega) = \mathsf{I} \, \delta(\mathbf{r} - \mathbf{r}'). \tag{A.10}$$

Substituting (A.8) and (A.9) in (A.7) we recover (21) of the main text, with T given by

$$\mathsf{T}(\mathbf{r}, \mathbf{r}', \omega) = -(2\hbar\omega)^{-1/2} \int \mathrm{d}\mathbf{r}'' \mathsf{U}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathsf{T}_0(\mathbf{r}'', \mathbf{r}', \omega). \tag{A.11}$$

Since T_0 is real, one finds that introducing T in this way implies that it fulfils the relation

$$\int \mathrm{d}\mathbf{r}'' \tilde{\mathsf{T}}(\mathbf{r}'', \mathbf{r}, \omega) \cdot \mathsf{T}^*(\mathbf{r}'', \mathbf{r}', \omega) - \mathrm{c.c.} = 0 \tag{A.12}$$

which is consistent with (but somewhat stronger than) conditions (8) and (22) imposed on T in the main text. Adopting the above stronger relation implies that the susceptibility (36) acquires an additional symmetry property on a par with (37) and (38), namely $\chi(\mathbf{r}, \mathbf{r}', z) = \chi(\mathbf{r}, \mathbf{r}', -z)$. As the validity of (A.12) is not essential in setting up our Hamilton formalism, we have refrained from using it in the main text.

Appendix B. Evaluation of the tensorial coefficients $\mathbf{f}_i$

In this appendix, we will show how equations (27)–(30) can be solved. We start by using (27) to eliminate f_1 from (28). As a result we obtain the differential equation:

$$\Delta'\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega)+\frac{\omega^2}{c^2}\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega)-\mu_0\hbar\int \mathrm{d}\mathbf{r}''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot[\mathsf{F}(\mathbf{r}'',\mathbf{r}')]_{\mathrm{T}'}$$
$$-\,\mathrm{i}\mu_0\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'')]_{\mathrm{T}'}$$
$$-\,\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega'')]_{\mathrm{T}'}\}=0. \qquad \text{(B.1)}$$

To get an expression for the last two terms we use (29) and (30). First, we multiply (29) by $\mathsf{T}^*(\mathbf{r}',\mathbf{r}'''',\omega')$ and integrate over $\mathbf{r}'$. Relabelling the dummy variables we get

$$-\mathrm{i}\hbar\omega'\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\tilde{\mathsf{T}}(\mathbf{r}''',\mathbf{r}'',\omega')\cdot\mathsf{T}^*(\mathbf{r}''',\mathbf{r}',\omega')$$
$$-\,(\omega-\omega')\int \mathrm{d}\mathbf{r}''\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega')\cdot\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega')$$
$$+\,\frac{\hbar}{\varepsilon_0}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\int \mathrm{d}\mathbf{r}''''\int_0^\infty \mathrm{d}\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}$$
$$+\,\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}\}\cdot\tilde{\mathsf{T}}(\mathbf{r}'''',\mathbf{r}''',\omega')\cdot\mathsf{T}^*(\mathbf{r}'''',\mathbf{r}',\omega')=0. \qquad \text{(B.2)}$$

A similar relation is obtained by multiplying (30) by $\mathsf{T}(\mathbf{r}',\mathbf{r}'''',\omega')$ and integrating over $\mathbf{r}'$:

$$-\mathrm{i}\hbar\omega'\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\tilde{\mathsf{T}}^*(\mathbf{r}''',\mathbf{r},'',\omega')\cdot\mathsf{T}(\mathbf{r}''',\mathbf{r}',\omega')$$
$$-\,(\omega+\omega')\int \mathrm{d}\mathbf{r}''\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega')\cdot\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega')$$
$$-\,\frac{\hbar}{\varepsilon_0}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\int \mathrm{d}\mathbf{r}''''\int_0^\infty \mathrm{d}\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}$$
$$+\,\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}\}\cdot\tilde{\mathsf{T}}^*(\mathbf{r}'''',\mathbf{r}''',\omega')\cdot\mathsf{T}(\mathbf{r}'''',\mathbf{r}',\omega')=0. \qquad \text{(B.3)}$$

We add (B.2) and (B.3). Upon integrating over ω' and using (8) and (23) we get

$$-\mathrm{i}\hbar\int \mathrm{d}\mathbf{r}''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathsf{F}(\mathbf{r}'',\mathbf{r}')-\omega\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''[\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'')$$
$$+\,\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega'')]+\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''\omega''[\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')$$
$$\cdot\,\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'')-\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega'')]=0. \qquad \text{(B.4)}$$

By taking the transverse part of this relation with respect to $\mathbf{r}'$ we get an identity that can be used to rewrite (B.1) in the form

$$-[\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\nabla}']\times\overleftarrow{\nabla}'+\frac{\omega^2}{c^2}\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega)-\mathrm{i}\mu_0\omega\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')$$
$$\cdot\,[\mathsf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'')]_{\mathrm{T}'}+\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathsf{T}(\mathbf{r}'',\mathbf{r}',\omega'')]_{\mathrm{T}'}\}=0. \qquad \text{(B.5)}$$

Here we used the transversality of $\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega)$ in its second argument to write the first term as a repeated vector product, with the spatial derivative operator ∇' acting to the left on the argument $\mathbf{r}'$ of the function $\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega)$.

The integral in (B.5) contains the transverse parts of T and T^* only. A more natural form of the differential equation, with the full tensors T and T^*, is obtained by introducing instead of $\mathbf{f}_2$ a new tensor $\mathbf{g}$ defined as

$$\mathbf{g}(\mathbf{r},\mathbf{r}',\omega) \equiv \mathrm{i}\omega\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega) - \frac{1}{\varepsilon_0}\int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega'' \{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'')]_{\mathrm{L}'} + \mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}(\mathbf{r}'',\mathbf{r}',\omega'')]_{\mathrm{L}'}\}. \tag{B.6}$$

It satisfies a differential equation, which follows from (B.5) as:

$$-[\mathbf{g}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\nabla}']\times\overleftarrow{\nabla}' + \frac{\omega^2}{c^2}\mathbf{g}(\mathbf{r},\mathbf{r}',\omega) + \mu_0\omega^2\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''[\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'') \cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'') + \mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathbf{T}(\mathbf{r}'',\mathbf{r}',\omega'')] = 0. \tag{B.7}$$

The integral contribution still depends on $\mathbf{f}_3$ and $\mathbf{f}_4$, so that the differential equation is not yet in closed form. However, we may rewrite the integral in such a way that its relation to $\mathbf{g}$ becomes obvious. This can be achieved with the help of the identity:

$$\int \mathrm{d}\mathbf{r}''\int_0^\infty \mathrm{d}\omega''[\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'') + \mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathbf{T}(\mathbf{r}'',\mathbf{r}',\omega'')] = \varepsilon_0\int \mathrm{d}\mathbf{r}''\mathbf{g}(\mathbf{r},\mathbf{r}'',\omega)\cdot\chi(\mathbf{r}'',\mathbf{r}',\omega-\mathrm{i}0) + \mathbf{s}(\mathbf{r},\mathbf{r}',\omega), \tag{B.8}$$

which contains a tensor $\mathbf{s}(\mathbf{r},\mathbf{r}',\omega)$ that arises while avoiding a pole in the complex frequency plane, as we shall see below. Furthermore the right-hand side contains the susceptibility tensor χ that has been defined in (36). In (B.8) the frequency is chosen to be in the lower half of the complex plane just below the real axis. Correspondingly, the term $-\mathrm{i}0$ is an infinitesimally small number on the negative imaginary axis.

To prove (B.8) we divide (B.2) by $\omega'-\omega+\mathrm{i}0$, with $\mathrm{i}0$ an infinitesimally small imaginary number. The result is

$$-\mathrm{i}\hbar\frac{\omega'}{\omega'-\omega+\mathrm{i}0}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\tilde{\mathbf{T}}(\mathbf{r}''',\mathbf{r}'',\omega')\cdot\mathbf{T}^*(\mathbf{r}''',\mathbf{r}',\omega') + \int \mathrm{d}\mathbf{r}''\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega')\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega') + \frac{\hbar}{\varepsilon_0}\frac{1}{\omega'-\omega+\mathrm{i}0} \times\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\int \mathrm{d}\mathbf{r}''''\int_0^\infty \mathrm{d}\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''} + \mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}\}\cdot\tilde{\mathbf{T}}(\mathbf{r}'''',\mathbf{r}''',\omega')\cdot\mathbf{T}^*(\mathbf{r}'''',\mathbf{r}',\omega') = \delta(\omega-\omega')\mathbf{s}(\mathbf{r},\mathbf{r}',\omega). \tag{B.9}$$

On the right-hand side, we have introduced a term proportional to the delta function $\delta(\omega-\omega')$ to account for the fact that the division by $\omega'-\omega+\mathrm{i}0$ yields a singular result for $\omega=\omega'$, as discussed in [28]. The coefficient $\mathbf{s}$ is as yet unknown. Likewise, upon dividing (B.3) by $\omega+\omega'$ we obtain

$$-\mathrm{i}\hbar\frac{\omega'}{\omega+\omega'}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\tilde{\mathbf{T}}^*(\mathbf{r}''',\mathbf{r}'',\omega')\cdot\mathbf{T}(\mathbf{r}''',\mathbf{r}',\omega') - \int \mathrm{d}\mathbf{r}''\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega')\cdot\mathbf{T}(\mathbf{r}'',\mathbf{r}',\omega') - \frac{\hbar}{\varepsilon_0}\frac{1}{\omega+\omega'}\int \mathrm{d}\mathbf{r}''\int \mathrm{d}\mathbf{r}'''\int \mathrm{d}\mathbf{r}'''' \times\int_0^\infty \mathrm{d}\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''} + \mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'') \cdot[\mathbf{T}(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}\}\cdot\tilde{\mathbf{T}}^*(\mathbf{r}'''',\mathbf{r}''',\omega')\cdot\mathbf{T}(\mathbf{r}'''',\mathbf{r}',\omega') = 0. \tag{B.10}$$

We subtract (B.10) from (B.9) and integrate over ω'. Inspecting the contributions from the last terms on the left-hand sides we find it useful to introduce the susceptibility tensor (36).

Upon employing moreover (8), we get as a result of combining (B.9) and (B.10):

$$
\begin{aligned}
-\mathrm{i}\varepsilon_0\omega \int d\mathbf{r}''\mathbf{f}_2(\mathbf{r},\mathbf{r}'',\omega)\cdot\chi(\mathbf{r}'',\mathbf{r}',\omega-\mathrm{i}0) + \int d\mathbf{r}''\int_0^\infty d\omega''[\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega'')\\
+\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot\mathbf{T}(\mathbf{r}'',\mathbf{r}',\omega'')]\\
+\int d\mathbf{r}''\int d\mathbf{r}'''\int_0^\infty d\omega''\{\mathbf{f}_3(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}\\
+\mathbf{f}_4(\mathbf{r},\mathbf{r}'',\omega,\omega'')\cdot[\mathbf{T}(\mathbf{r}'',\mathbf{r}''',\omega'')]_{\mathrm{L}'''}\}\cdot\chi(\mathbf{r}''',\mathbf{r}',\omega-\mathrm{i}0) = \mathbf{s}(\mathbf{r},\mathbf{r}',\omega). \quad \text{(B.11)}
\end{aligned}
$$

When (B.6) is invoked to eliminate $\mathbf{f}_2$ in favour of $\mathbf{g}$, we recover (B.8).

Having established (B.8) we insert it in (B.7) so as to arrive at an inhomogeneous wave equation for $\mathbf{g}$:

$$
\begin{aligned}
-[\mathbf{g}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\nabla}']\times\overleftarrow{\nabla}' + \frac{\omega^2}{c^2}\mathbf{g}(\mathbf{r},\mathbf{r}',\omega) + \frac{\omega^2}{c^2}\int d\mathbf{r}''\mathbf{g}(\mathbf{r},\mathbf{r}'',\omega)\cdot\chi(\mathbf{r}'',\mathbf{r}',\omega-\mathrm{i}0)\\
= -\mu_0\omega^2\mathbf{s}(\mathbf{r},\mathbf{r}',\omega). \quad \text{(B.12)}
\end{aligned}
$$

To solve $\mathbf{g}$ from this differential equation we employ the Green function $\mathbf{G}(\mathbf{r},\mathbf{r}',z)$ associated with the operator on the left-hand side. It has been defined in (35). In terms of this Green function the solution of (B.12) reads

$$
\mathbf{g}(\mathbf{r},\mathbf{r}',\omega) = -\mu_0\omega^2\int d\mathbf{r}''\mathbf{s}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{G}(\mathbf{r}'',\mathbf{r}',\omega-\mathrm{i}0). \quad \text{(B.13)}
$$

With the use of the above expression for $\mathbf{g}$ in terms of $\mathbf{s}$, we can evaluate the coefficients $\mathbf{f}_i$ successively. Let us start with $\mathbf{f}_2$. From (B.7) it follows that the integral contribution in (B.6) is proportional to the longitudinal part $[\mathbf{g}(\mathbf{r},\mathbf{r}',\omega)]_{\mathrm{L}'}$ of $\mathbf{g}$. As a consequence, (B.6) implies that $\mathbf{f}_2$ equals $-(\mathrm{i}/\omega)[\mathbf{g}(\mathbf{r},\mathbf{r}',\omega)]_{\mathrm{T}'}$. Hence, upon using (B.13) and introducing the tensor $\mathbf{u}$ by writing $\mathbf{s}$ as

$$
\mathbf{s}(\mathbf{r},\mathbf{r}',\omega) = \int d\mathbf{r}''\mathbf{u}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}',\omega) \quad \text{(B.14)}
$$

we obtain

$$
\mathbf{f}_2(\mathbf{r},\mathbf{r}',\omega) = \mathrm{i}\mu_0\omega\int d\mathbf{r}''\int d\mathbf{r}'''\mathbf{u}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega)\cdot[\mathbf{G}(\mathbf{r}''',\mathbf{r}',\omega-\mathrm{i}0)]_{\mathrm{T}'}. \quad \text{(B.15)}
$$

On a par with this expression we get from (27)

$$
\mathbf{f}_1(\mathbf{r},\mathbf{r}',\omega) = \frac{\omega^2}{c^2}\int d\mathbf{r}''\int d\mathbf{r}'''\mathbf{u}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega)\cdot[\mathbf{G}(\mathbf{r}''',\mathbf{r}',\omega-\mathrm{i}0)]_{\mathrm{T}'}. \quad \text{(B.16)}
$$

Expressions for $\mathbf{f}_3$ and $\mathbf{f}_4$ follow from (B.9) and (B.10) upon using (B.13)–(B.15) and the longitudinal part of (B.7). We get

$$
\begin{aligned}
\mathbf{f}_3(\mathbf{r},\mathbf{r}',\omega,\omega') = \delta(\omega-\omega')\mathbf{u}(\mathbf{r},\mathbf{r}',\omega)\\
-\mu_0\hbar\omega\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\mathbf{u}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega)\\
\cdot[\mathbf{G}(\mathbf{r}''',\mathbf{r}'''',\omega-\mathrm{i}0)]_{\mathrm{T}''''}\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}'''',\omega')\\
+\mu_0\hbar\frac{\omega^2}{\omega-\omega'-\mathrm{i}0}\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\mathbf{u}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{T}^*(\mathbf{r}'',\mathbf{r}''',\omega)\\
\cdot\mathbf{G}(\mathbf{r}''',\mathbf{r}'''',\omega-\mathrm{i}0)\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}'''',\omega') \quad \text{(B.17)}
\end{aligned}
$$

$$\mathbf{f}_4(\mathbf{r}, \mathbf{r}', \omega, \omega') = \mu_0\hbar\omega \int d\mathbf{r}'' \int d\mathbf{r}''' \int d\mathbf{r}''''\mathbf{u}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathbf{T}^*(\mathbf{r}'', \mathbf{r}''', \omega) \cdot [\mathbf{G}(\mathbf{r}''', \mathbf{r}'''', \omega - i0)]_{T''''} \cdot \tilde{\mathbf{T}}^*(\mathbf{r}', \mathbf{r}'''', \omega') - \mu_0\hbar\frac{\omega^2}{\omega + \omega'} \int d\mathbf{r}'' \int d\mathbf{r}''' \int d\mathbf{r}''''\mathbf{u}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathbf{T}^*(\mathbf{r}'', \mathbf{r}''', \omega) \cdot \mathbf{G}(\mathbf{r}''', \mathbf{r}'''', \omega - i0) \cdot \tilde{\mathbf{T}}^*(\mathbf{r}', \mathbf{r}'''', \omega'). \quad \text{(B.18)}$$

The introduction of $\mathbf{u}$ instead of $\mathbf{s}$ has led to the simple form of the first term on the right-hand side of (B.17).

Appendix C. The tensor u

In appendix B, the coefficients $\mathbf{f}_i$ have been obtained. They are all proportional to the tensor $\mathbf{u}(\mathbf{r}, \mathbf{r}', \omega)$. In this section, we shall show how this tensor can be determined.

After the insertion of the coefficients in expression (26) it follows that the diagonalizing operator $\mathbf{C}(\mathbf{r}, \omega)$ itself is proportional to $\mathbf{u}$ as well. Since $\mathbf{C}(\mathbf{r}, \omega)$ and its Hermitian conjugate must satisfy canonical commutation relations of the general form (4), $\mathbf{u}$ has to fulfil a constraint that is obtained by evaluating the commutator $[\mathbf{C}(\mathbf{r}, \omega), \mathbf{C}^\dagger(\mathbf{r}', \omega')]$. In fact, substituting expression (26) and its Hermitian conjugate in the commutator and employing (2) and (4) we arrive at the following condition:

$$i\hbar \int d\mathbf{r}''[\mathbf{f}_1(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathbf{f}}_2^*(\mathbf{r}', \mathbf{r}'', \omega') - \mathbf{f}_2(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathbf{f}}_1^*(\mathbf{r}', \mathbf{r}'', \omega')] + \int d\mathbf{r}'' \int_0^\infty d\omega''[\mathbf{f}_3(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot \tilde{\mathbf{f}}_3^*(\mathbf{r}', \mathbf{r}'', \omega', \omega'') - \mathbf{f}_4(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot \tilde{\mathbf{f}}_4^*(\mathbf{r}', \mathbf{r}'', \omega', \omega'')] = \mathbf{I}\,\delta(\mathbf{r} - \mathbf{r}')\delta(\omega - \omega'). \quad \text{(C.1)}$$

After the insertion of the formulae for $\mathbf{f}_i$ we arrive at a bilinear condition for $\mathbf{u}$ of the form

$$\int d\mathbf{r}'' \int d\mathbf{r}'''\mathbf{u}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathbf{M}(\mathbf{r}'', \mathbf{r}''', \omega, \omega') \cdot \tilde{\mathbf{u}}^*(\mathbf{r}', \mathbf{r}''', \omega') = \mathbf{I}\,\delta(\mathbf{r} - \mathbf{r}')\delta(\omega - \omega'). \quad \text{(C.2)}$$

The explicit form of the tensorial integral kernel $\mathbf{M}$ follows by evaluating (C.1) with (B.15)–(B.18). It contains contributions with a variable number of Green functions $\mathbf{G}$. The simplest contribution $\mathbf{M}_0$ is that without a Green function, which is found to be

$$\mathbf{M}_0(\mathbf{r}, \mathbf{r}', \omega, \omega') = \mathbf{I}\,\delta(\mathbf{r} - \mathbf{r}')\delta(\omega - \omega'). \quad \text{(C.3)}$$

The next contribution $\mathbf{M}_1$ consists of all terms containing a single Green function:

$$\mathbf{M}_1(\mathbf{r}, \mathbf{r}', \omega, \omega') = -\mu_0\hbar \int d\mathbf{r}'' \int d\mathbf{r}''' \,\mathbf{T}^*(\mathbf{r}, \mathbf{r}'', \omega) \cdot \Big\{\omega[\mathbf{G}(\mathbf{r}'', \mathbf{r}''', \omega - i0)]_{T'''} + \omega'[\tilde{\mathbf{G}}^*(\mathbf{r}''', \mathbf{r}'', \omega' - i0)]_{T''} - \frac{\omega^2}{\omega - \omega' - i0}\mathbf{G}(\mathbf{r}'', \mathbf{r}''', \omega - i0) + \frac{\omega'^2}{\omega - \omega' - i0}\tilde{\mathbf{G}}^*(\mathbf{r}''', \mathbf{r}'', \omega' - i0)\Big\} \cdot \tilde{\mathbf{T}}(\mathbf{r}', \mathbf{r}''', \omega'). \quad \text{(C.4)}$$

The first two terms contain the transverse part of the Green function, whereas the last two depend on the full Green function.

Finally, we have got the contributions with two Green functions. Since again both the full Green function and its transverse part show up, we can distinguish various types of terms.

The terms with two transverse Green functions become upon invoking (8)

$$\mathbf{M}_{2TT}(\mathbf{r},\mathbf{r}',\omega,\omega') = \frac{\mu_0\hbar}{c^2}\omega\omega'(\omega+\omega')\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\,\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot[\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)]_{\mathrm{T}'''}\cdot[\tilde{\mathbf{G}}^*(\mathbf{r}'''',\mathbf{r}''',\omega'-\mathrm{i}0)]_{\mathrm{T}'''}\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}'''',\omega'). \tag{C.5}$$

The terms with one transverse and one full Green function are

$$\begin{aligned}\mathbf{M}_{2T}(\mathbf{r},\mathbf{r}',\omega,\omega') = \frac{\mu_0\hbar}{c^2}\omega\omega'\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\int d\mathbf{r}^{v}\,\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\\ \cdot\{\omega\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\cdot\chi(\mathbf{r}''',\mathbf{r}'''',\omega-\mathrm{i}0)\cdot[\tilde{\mathbf{G}}^*(\mathbf{r}^{v},\mathbf{r}'''',\omega'-\mathrm{i}0)]_{\mathrm{T}''''}\\ +\omega'[\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)]_{\mathrm{T}'''}\cdot\tilde{\chi}^*(\mathbf{r}'''',\mathbf{r}''',\omega'-\mathrm{i}0)\cdot\tilde{\mathbf{G}}^*(\mathbf{r}^{v},\mathbf{r}'''',\omega'-\mathrm{i}0)\}\\ \cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}^{v},\omega')\end{aligned} \tag{C.6}$$

where we introduced the susceptibility tensor (36). The last set of terms we have to consider is that with two full Green functions. Again using (36) we get

$$\begin{aligned}\mathbf{M}_{2}(\mathbf{r},\mathbf{r}',\omega,\omega') = \frac{\mu_0\hbar}{c^2}\frac{\omega^2\omega'^2}{\omega-\omega'-\mathrm{i}0}\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\int d\mathbf{r}^{v}\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\\ \cdot\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\cdot[\chi(\mathbf{r}''',\mathbf{r}'''',\omega-\mathrm{i}0)-\tilde{\chi}^*(\mathbf{r}'''',\mathbf{r}''',\omega'-\mathrm{i}0)]\\ \cdot\tilde{\mathbf{G}}^*(\mathbf{r}^{v},\mathbf{r}'''',\omega'-\mathrm{i}0)\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}^{v},\omega').\end{aligned} \tag{C.7}$$

Having obtained all contributions to the integral kernel $\mathbf{M}$ we are now in a position to evaluate their sum. We start by investigating the terms with transverse Green functions, as given by (C.5), (C.6) and part of (C.4). Taking all terms together we may write them as

$$\begin{aligned}\mu_0\hbar\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot\Big\{-\omega\left[\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\right]_{\mathrm{T}'''}\\ \cdot\Big[\mathbf{I}\,\delta(\mathbf{r}'''-\mathbf{r}'''')-\frac{\omega'^2}{c^2}\tilde{\mathbf{G}}^*(\mathbf{r}'''',\mathbf{r}''',\omega'-\mathrm{i}0)\\ -\frac{\omega'^2}{c^2}\int d\mathbf{r}^{v}\tilde{\chi}^*(\mathbf{r}^{v},\mathbf{r}''',\omega'-\mathrm{i}0)\cdot\tilde{\mathbf{G}}^*(\mathbf{r}'''',\mathbf{r}^{v},\omega'-\mathrm{i}0)\Big]\\ -\omega'\Big[\mathbf{I}\,\delta(\mathbf{r}''-\mathbf{r}''')-\frac{\omega^2}{c^2}\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\\ -\frac{\omega^2}{c^2}\int d\mathbf{r}^{v}\mathbf{G}(\mathbf{r}'',\mathbf{r}^{v},\omega-\mathrm{i}0)\cdot\chi(\mathbf{r}^{v},\mathbf{r}''',\omega-\mathrm{i}0)\Big]\\ \cdot[\tilde{\mathbf{G}}^*(\mathbf{r}'''',\mathbf{r}''',\omega'-\mathrm{i}0)]_{\mathrm{T}'''}\Big\}\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}'''',\omega').\end{aligned} \tag{C.8}$$

The sets of terms that multiply the transverse Green functions are transverse themselves, as follows from (35). Since the integral of the scalar product of a longitudinal and a transverse function vanishes, we can replace each of these transverse Green functions by their full Green function counterparts. Subsequently, upon using (35) and performing partial integrations we may rewrite (C.8) in the form

$$\mu_0\hbar(\omega+\omega')\int d\mathbf{r}''\int d\mathbf{r}'''\int d\mathbf{r}''''\mathbf{T}^*(\mathbf{r},\mathbf{r}'',\omega)\cdot\{[\mathbf{G}(\mathbf{r}'',\mathbf{r}''',\omega-\mathrm{i}0)\times\overleftarrow{\nabla}''']\times\overleftarrow{\nabla}''''\}\cdot\tilde{\mathbf{G}}^*(\mathbf{r}'''',\mathbf{r}''',\omega'-\mathrm{i}0)\cdot\tilde{\mathbf{T}}(\mathbf{r}',\mathbf{r}'''',\omega'). \tag{C.9}$$

An alternative form for this expression is found upon splitting it in two terms by writing the factor $(\omega+\omega')$ as the difference of $\omega^2/(\omega-\omega'-\mathrm{i}0)$ and $\omega'^2/(\omega-\omega'-\mathrm{i}0)$. Subsequently, we carry out partial integrations in the first term, while we leave the second as it stands. Finally, we use (35) to eliminate the double spatial derivatives. We end up with a set of terms that precisely cancel (C.7) and the remainder of (C.4) (i.e., the terms without the transverse Green functions).

Collecting the results we find that we are left with (C.3). The result for M is thus quite simple:

$$\mathsf{M}(\mathbf{r}, \mathbf{r}', \omega, \omega') = \mathsf{I}\,\delta(\mathbf{r} - \mathbf{r}')\delta(\omega - \omega'). \tag{C.10}$$

As a consequence, condition (C.2) becomes

$$\int \mathrm{d}\mathbf{r}''\mathsf{u}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathsf{u}}^*(\mathbf{r}', \mathbf{r}'', \omega) = \mathsf{I}\,\delta(\mathbf{r} - \mathbf{r}'). \tag{C.11}$$

In other words, the tensorial integral kernel $\mathsf{u}(\mathbf{r}, \mathbf{r}', \omega)$ must be unitary. For convenience we choose from now on

$$\mathsf{u}(\mathbf{r}, \mathbf{r}', \omega) = \mathsf{I}\,\delta(\mathbf{r} - \mathbf{r}') \tag{C.12}$$

so that u is independent of the frequency and diagonal in the spatial variables. A different choice for u leads to a unitarily equivalent form of the diagonalizing operator $\mathbf{C}(\mathbf{r}, \omega)$, as follows from (26) with (B.15)–(B.18). Upon inserting (C.12) in (B.15)–(B.18) we finally arrive at expressions (31)–(34) of the main text.

As a final check of the expressions for $\mathbf{f}_i$ we may verify that the commutator $[\mathbf{C}(\mathbf{r}, \omega), \mathbf{C}(\mathbf{r}', \omega')]$ vanishes for all position and frequency arguments. To that end we have to check whether the condition

$$\begin{aligned} i\hbar \int \mathrm{d}\mathbf{r}''[\mathbf{f}_1(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathbf{f}}_2(\mathbf{r}', \mathbf{r}'', \omega') - \mathbf{f}_2(\mathbf{r}, \mathbf{r}'', \omega) \cdot \tilde{\mathbf{f}}_1(\mathbf{r}', \mathbf{r}'', \omega')] \\ + \int \mathrm{d}\mathbf{r}'' \int_0^\infty \mathrm{d}\omega''[\mathbf{f}_3(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot \tilde{\mathbf{f}}_4(\mathbf{r}', \mathbf{r}'', \omega', \omega'') \\ - \mathbf{f}_4(\mathbf{r}, \mathbf{r}'', \omega, \omega'') \cdot \tilde{\mathbf{f}}_3(\mathbf{r}', \mathbf{r}'', \omega', \omega'')] = 0 \end{aligned} \tag{C.13}$$

is satisfied. Along similar lines as above one verifies that this is indeed true.

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