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DOI
10.1209/epl/i2004-10131-8

Publication date
2004

Document Version
Final published version

Published in
Europhysics Letters

EUROPHYSICS LETTERS 1 September 2004

Europhys. Lett., **67** (5), pp. 766–772 (2004)

DOI: 10.1209/epl/i2004-10131-8

Fano diagonalization of a polariton model for an inhomogeneous absorptive dielectric

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(received 2 June 2004; accepted in final form 30 June 2004)

PACS. 42.50.Nn – Quantum optical phenomena in absorbing, dispersive and conducting media.
PACS. 71.36.+c – Polaritons (including photon-phonon and photon-magnon interactions).
PACS. 03.70.+k – Theory of quantized fields.

Abstract. – The Hamiltonian of a polariton model for an inhomogeneous linear absorptive dielectric is diagonalized by means of Fano's diagonalization method. The creation and annihilation operators for the independent normal modes are explicitly found as linear combinations of the canonical operators. The coefficients in these combinations depend on the tensorial Green function that governs the propagation of electromagnetic waves through the dielectric. The time-dependent electromagnetic fields in the Heisenberg picture are given in terms of the diagonalizing operators. These results justify the phenomenological quantization of the electromagnetic field in an absorptive dielectric.

Introduction. – To describe quantum optical phenomena in dielectrics it is essential to have available a quantization procedure for the electromagnetic field in ponderable matter. Preferably, such a quantization should be based on the standard canonical quantization method of quantum field theory. To apply that method to the fields in linear dielectrics, one should start from a Hamiltonian description, in which both the field and the dielectric are given in terms of canonical variables that are coupled bilinearly. The normal modes in such a system are the well-known polaritons [1]. If the dielectric is absorptive and dispersive, damping of the polariton modes may be taken into account by coupling the dielectric degrees of freedom to a suitable bath of harmonic oscillators with a continuous range of frequencies. Working along these lines, Huttner and Barnett [2] were the first to formulate a damped-polariton model for an absorptive dielectric in an electromagnetic field and to study its properties. By using a diagonalization method due to Fano [3], they were able to find the full time dependence of the electromagnetic field operators for their model.

The method of diagonalization employed in [2] is based on a separation of longitudinal and transverse degrees of freedom, and on a Fourier decomposition of the canonical variables. Both of these means are only expedient for homogeneous systems with translation invariance. In fact, when the damped-polariton model is taken to be inhomogeneous, with an arbitrary spatial dependence of the material properties, the longitudinal and transverse degrees of freedom get coupled, while the Fourier components of the canonical variables for different wave vectors start interacting as well. Hence, the diagonalization procedure in [2] runs into difficulties for the inhomogeneous case. Since inhomogeneities are unavoidable in any quantum optical experiment involving dielectrics, this is a serious drawback. It is the purpose of this letter to show that diagonalization of the inhomogeneous version of the damped-polariton model is possible, and that explicit expressions for the diagonalizing operators in terms of canonical variables can be found.

Model. – In the damped-polariton model the polarization density is coupled to a bath of harmonic oscillators with a continuous range of eigenfrequencies. The electromagnetic field interacts with the dielectric according to the standard minimal-coupling scheme. The Hamiltonian of the model is [2]

$$\begin{aligned} H \;=\; & \int \mathrm{d}\boldsymbol{r} \left[\frac{1}{2\epsilon_0}\Pi^2 + \frac{1}{2\mu_0}(\boldsymbol{\nabla}\times\boldsymbol{A})^2 + \frac{1}{2\rho}P^2 + \frac{1}{2}\rho\tilde{\omega}_0^2 X^2 + \frac{1}{2\rho}\int_0^\infty \mathrm{d}\omega\, Q_\omega^2 + \right. \\ & \left. + \frac{1}{2}\rho\int_0^\infty \mathrm{d}\omega\,\omega^2 Y_\omega^2 + \frac{\alpha}{\rho}\boldsymbol{A}\cdot\boldsymbol{P} + \frac{\alpha^2}{2\rho}A^2 + \frac{1}{\rho}\int_0^\infty \mathrm{d}\omega\, v_\omega \boldsymbol{X}\cdot\boldsymbol{Q}_\omega \right] + \\ & + \int \mathrm{d}\boldsymbol{r}\,\mathrm{d}\boldsymbol{r}'\,\frac{\boldsymbol{\nabla}\cdot(\alpha\boldsymbol{X})\,\boldsymbol{\nabla}'\cdot(\alpha'\boldsymbol{X}')}{8\pi\epsilon_0|\boldsymbol{r}-\boldsymbol{r}'|}\,. \end{aligned} \tag{1}$$

The transverse part of the electromagnetic field is determined by the vector potential $\boldsymbol{A}(\boldsymbol{r})$, for which the Coulomb gauge is adopted. Its conjugate canonical momentum is $\boldsymbol{\Pi}(\boldsymbol{r})$. The linear dielectric, with a space-dependent density $\rho(\boldsymbol{r})$, is described by the harmonic displacement variable $\boldsymbol{X}(\boldsymbol{r})$ and its canonical momentum $\boldsymbol{P}(\boldsymbol{r})$. The associated (renormalized) eigenfrequency $\tilde{\omega}_0(\boldsymbol{r})$ is generally space dependent as well. The electromagnetic field is coupled to the dielectric variable $\boldsymbol{X}$ in the usual way. In terms of the polarization density $-\alpha\boldsymbol{X}$, with a space-dependent coupling parameter $\alpha(\boldsymbol{r}) > 0$, the minimal coupling scheme leads to an electrostatic contribution and to a bilinear interaction term with $\boldsymbol{A}\cdot\boldsymbol{P}$. Finally, damping is introduced in the system by a continuum bath of harmonic oscillators with canonical variables $\boldsymbol{Y}_\omega(\boldsymbol{r})$, $\boldsymbol{Q}_\omega(\boldsymbol{r})$ and with eigenfrequencies ω. These bath oscillators are coupled to $\boldsymbol{X}(\boldsymbol{r})$ with a strength $v_\omega(\boldsymbol{r}) > 0$. We used the notation $\boldsymbol{X}' = \boldsymbol{X}(\boldsymbol{r}')$, and likewise α' and $\boldsymbol{\nabla}'$.

The canonical variables obey the standard commutation relations:

$$\begin{aligned} [\boldsymbol{\Pi}(\boldsymbol{r}), \boldsymbol{A}(\boldsymbol{r}')] \;&=\; -i\hbar\,\boldsymbol{\delta}_{\mathrm{T}}(\boldsymbol{r}-\boldsymbol{r}'), \qquad [\boldsymbol{P}(\boldsymbol{r}), \boldsymbol{X}(\boldsymbol{r}')] = -i\hbar\,\mathsf{I}\,\delta(\boldsymbol{r}-\boldsymbol{r}'), \\ [\boldsymbol{Q}_\omega(\boldsymbol{r}), \boldsymbol{Y}_{\omega'}(\boldsymbol{r}')] \;&=\; -i\hbar\,\delta(\omega-\omega')\,\mathsf{I}\,\delta(\boldsymbol{r}-\boldsymbol{r}'), \end{aligned} \tag{2}$$

while all other commutators of the canonical variables vanish. Here I is the three-dimensional unit tensor, while $\boldsymbol{\delta}_{\mathrm{T}}(\boldsymbol{r}) = \mathsf{I}\delta(\boldsymbol{r}) + \boldsymbol{\nabla}\boldsymbol{\nabla}(4\pi r)^{-1}$ is the transverse delta-function.

The electric field operator $\boldsymbol{E}$ is the sum of a transverse part depending on $\boldsymbol{\Pi}$ and a longitudinal part that is proportional to the polarization density:

$$\boldsymbol{E}(\boldsymbol{r}) = -\frac{1}{\varepsilon_0}\,\boldsymbol{\Pi}(\boldsymbol{r}) + \frac{1}{\varepsilon_0}\,[\alpha\boldsymbol{X}(\boldsymbol{r})]_{\mathrm{L}}\,. \tag{3}$$

The longitudinal part of a vector (or a tensor) is obtained by a convolution with the longitudinal delta-function $\boldsymbol{\delta}_{\mathrm{L}}(\boldsymbol{r}) = -\boldsymbol{\nabla}\boldsymbol{\nabla}(4\pi r)^{-1}$. The displacement field

$$\boldsymbol{D}(\boldsymbol{r}) = -\boldsymbol{\Pi}(\boldsymbol{r}) - [\alpha\boldsymbol{X}(\boldsymbol{r})]_{\mathrm{T}} \tag{4}$$

is purely transverse.

In the following we will show how the Hamiltonian (1), with canonical operators satisfying the commutation relations (2), can be brought in diagonal form.

Fano diagonalization. – The Hamiltonian is quadratic in the canonical variables. Hence, it should be possible to find a diagonal representation of the form

$$H = \int \mathrm{d}\boldsymbol{r} \int_0^\infty \mathrm{d}\omega \hbar\omega\, \boldsymbol{C}^\dagger(\boldsymbol{r},\omega) \cdot \boldsymbol{C}(\boldsymbol{r},\omega), \tag{5}$$

where we omit a zero-point-energy term. The operators $\boldsymbol{C}(\boldsymbol{r},\omega)$ are annihilation operators, which (together with the associated creation operators) satisfy the commutation relations

$$\left[\boldsymbol{C}(\boldsymbol{r},\omega), \boldsymbol{C}^\dagger(\boldsymbol{r}',\omega')\right] = \delta(\omega-\omega')\,\mathsf{I}\,\delta(\boldsymbol{r}-\boldsymbol{r}'), \qquad \left[\boldsymbol{C}(\boldsymbol{r},\omega), \boldsymbol{C}(\boldsymbol{r}',\omega')\right] = 0. \tag{6}$$

Each canonical operator can be written as a linear combination of the annihilation and creation operators. For instance, one has

$$\begin{aligned} \boldsymbol{A}(\boldsymbol{r}) &= \int \mathrm{d}\boldsymbol{r}' \int_0^\infty \mathrm{d}\omega\, \mathsf{f}_A(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot \boldsymbol{C}(\boldsymbol{r}',\omega) + \text{h.c.},\\ \boldsymbol{Q}_\omega(\boldsymbol{r}) &= \int \mathrm{d}\boldsymbol{r}' \int_0^\infty \mathrm{d}\omega'\, \mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega,\omega')\cdot \boldsymbol{C}(\boldsymbol{r}',\omega') + \text{h.c.}, \end{aligned} \tag{7}$$

with tensorial coefficients f_A and f_Q. The coefficients f_Π, f_X, f_P and f_Y are defined analogously. Both f_A and f_Π are transverse in $\boldsymbol{r}$. The electric field follows from (3) as

$$\boldsymbol{E}(\boldsymbol{r}) = \int \mathrm{d}\boldsymbol{r}' \int_0^\infty \mathrm{d}\omega\, \mathsf{f}_E(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot \boldsymbol{C}(\boldsymbol{r}',\omega) + \text{h.c.}, \tag{8}$$

with the coefficient

$$\mathsf{f}_E(\boldsymbol{r},\boldsymbol{r}',\omega) = -\frac{1}{\varepsilon_0}\,\mathsf{f}_\Pi(\boldsymbol{r},\boldsymbol{r}',\omega) + \frac{1}{\varepsilon_0}\,\left[\alpha\, \mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega)\right]_{\mathrm{L}}. \tag{9}$$

From eq. (6) it follows that the coefficients are equal to commutators. For instance, one has

$$\mathsf{f}_A(\boldsymbol{r},\boldsymbol{r}',\omega) = \left[\boldsymbol{A}(\boldsymbol{r}), \boldsymbol{C}^\dagger(\boldsymbol{r}',\omega)\right], \qquad \mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega,\omega') = \left[\boldsymbol{Q}_\omega(\boldsymbol{r}), \boldsymbol{C}^\dagger(\boldsymbol{r}',\omega')\right]. \tag{10}$$

Inversely, each $\boldsymbol{C}(\boldsymbol{r},\omega)$ is a linear combination of the canonical operators

$$\begin{aligned} \boldsymbol{C}(\boldsymbol{r},\omega) = -\frac{i}{\hbar}\int \mathrm{d}\boldsymbol{r}' \Big\{ & \boldsymbol{A}(\boldsymbol{r}')\cdot \mathsf{f}^*_\Pi(\boldsymbol{r}',\boldsymbol{r},\omega) - \boldsymbol{\Pi}(\boldsymbol{r}')\cdot \mathsf{f}^*_A(\boldsymbol{r}',\boldsymbol{r},\omega) + \\ & + \boldsymbol{X}(\boldsymbol{r}')\cdot \mathsf{f}^*_P(\boldsymbol{r}',\boldsymbol{r},\omega) - \boldsymbol{P}(\boldsymbol{r}')\cdot \mathsf{f}^*_X(\boldsymbol{r}',\boldsymbol{r},\omega) + \\ & + \int_0^\infty \mathrm{d}\omega' \left[\boldsymbol{Y}_{\omega'}(\boldsymbol{r}')\cdot \mathsf{f}^*_Q(\boldsymbol{r}',\boldsymbol{r},\omega',\omega) - \boldsymbol{Q}_{\omega'}(\boldsymbol{r}')\cdot \mathsf{f}^*_Y(\boldsymbol{r}',\boldsymbol{r},\omega',\omega)\right]\Big\}. \end{aligned} \tag{11}$$

Fano's method to diagonalize the Hamiltonian amounts to finding the tensorial coefficients in these expressions by solving a set of equations that follows from the commutator of $\boldsymbol{C}$ with the Hamiltonian. In fact, eqs. (5) and (6) imply

$$[\boldsymbol{C}(\boldsymbol{r},\omega), H] = \hbar\omega\, \boldsymbol{C}(\boldsymbol{r},\omega). \tag{12}$$

Upon inserting eqs. (1) and (11), employing (2) and comparing the coefficients of the canonical operators, we arrive at the following set of linear relations:

$$i\omega\,\mathsf{f}_A(\boldsymbol{r},\boldsymbol{r}',\omega) = -\frac{1}{\varepsilon_0}\,\mathsf{f}_\Pi(\boldsymbol{r},\boldsymbol{r}',\omega), \tag{13}$$

$$i\omega\,\mathsf{f}_\Pi(\boldsymbol{r},\boldsymbol{r}',\omega) = -\frac{1}{\mu_0}\,\Delta\,\mathsf{f}_A(\boldsymbol{r},\boldsymbol{r}',\omega) + \left[\frac{\alpha^2}{\rho}\,\mathsf{f}_A(\boldsymbol{r},\boldsymbol{r}',\omega)\right]_{\mathrm{T}} + \left[\frac{\alpha}{\rho}\,\mathsf{f}_P(\boldsymbol{r},\boldsymbol{r}',\omega)\right]_{\mathrm{T}}, \tag{14}$$

$$i\omega\,\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega) = -\frac{\alpha}{\rho}\,\mathsf{f}_A(\boldsymbol{r},\boldsymbol{r}',\omega) - \frac{1}{\rho}\,\mathsf{f}_P(\boldsymbol{r},\boldsymbol{r}',\omega), \tag{15}$$

$$i\omega\,\mathsf{f}_P(\boldsymbol{r},\boldsymbol{r}',\omega) = \rho\tilde{\omega}_0^2\,\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega) + \frac{\alpha}{\varepsilon_0}\,\left[\alpha\,\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega)\right]_{\mathrm{L}} + \frac{1}{\rho}\int_0^\infty \mathrm{d}\omega'\,v_{\omega'}\,\mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega',\omega), \tag{16}$$

$$i\omega\,\mathsf{f}_Y(\boldsymbol{r},\boldsymbol{r}',\omega',\omega) = -\frac{1}{\rho}\,v_{\omega'}\,\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega) - \frac{1}{\rho}\,\mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega',\omega), \tag{17}$$

$$i\omega\,\mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega',\omega) = \rho\omega'^2\,\mathsf{f}_Y(\boldsymbol{r},\boldsymbol{r}',\omega',\omega), \tag{18}$$

with α, ρ, $\tilde{\omega}_0$ and v_ω depending on $\boldsymbol{r}$. To solve these equations, we shall first consider the purely algebraic equations (13), (15)-(18). This will lead to the introduction of the susceptibility.

Introduction of the susceptibility. – By elimination of f_Y from eqs. (17) and (18) we get

$$\left(\omega^2 - \omega'^2\right)\,\mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega',\omega) = \omega'^2 v_{\omega'}\,\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega). \tag{19}$$

To obtain f_Q, we have to impose a prescription for the pole at $\omega = \omega'$:

$$\mathsf{f}_Q(\boldsymbol{r},\boldsymbol{r}',\omega',\omega) = \frac{\omega'^2\,v_{\omega'}}{(\omega + i0)^2 - \omega'^2}\,\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega) + \mathsf{s}(\boldsymbol{r},\boldsymbol{r}',\omega)\,\delta(\omega - \omega'), \tag{20}$$

with $\omega + i0$ a complex frequency in the upper half-plane and infinitesimally close to the real axis. The last term contains an as yet unknown tensor $\mathsf{s}(\boldsymbol{r},\boldsymbol{r}',\omega)$. Substituting eq. (20) in (16), and eliminating f_P and f_A with the help of eqs. (13) and (15), we get an equality which expresses f_Π in terms of f_X and s. If we use (9) to eliminate f_Π in favor of f_E, this equality may be rewritten as a linear relation between the coefficients of the polarization density $-\alpha\boldsymbol{X}$ and the electric field $\boldsymbol{E}$:

$$-\alpha\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega) = \varepsilon_0\,\chi(\boldsymbol{r},\omega)\,\mathsf{f}_E(\boldsymbol{r},\boldsymbol{r}',\omega) + \frac{\varepsilon_0}{\rho\alpha}\,v_\omega\,\chi(\boldsymbol{r},\omega)\,\mathsf{s}(\boldsymbol{r},\boldsymbol{r}',\omega). \tag{21}$$

The proportionality constant is the position- and frequency-dependent susceptibility:

$$\chi(\boldsymbol{r},\omega) = -\frac{\alpha^2}{\varepsilon_0\rho}\left[\omega^2 - \tilde{\omega}_0^2 - \frac{1}{\rho^2}\int_0^\infty \mathrm{d}\omega'\,\frac{\omega'^2\,v_{\omega'}^2}{(\omega + i0)^2 - \omega'^2}\right]^{-1}. \tag{22}$$

Having solved the algebraic equations of the set (13)-(18), we now turn to the differential equation (14).

General form of the tensorial coefficients. – After elimination of f_A and f_P with the help of (13) and (15) we get from (14)

$$\Delta\,\mathsf{f}_\Pi(\boldsymbol{r},\boldsymbol{r}',\omega) + \frac{\omega^2}{c^2}\mathsf{f}_\Pi(\boldsymbol{r},\boldsymbol{r}',\omega) = -\frac{\omega^2}{c^2}\,\left[\alpha\mathsf{f}_X(\boldsymbol{r},\boldsymbol{r}',\omega)\right]_{\mathrm{T}}. \tag{23}$$

Since $\mathsf{f}_\Pi(\boldsymbol{r},\boldsymbol{r}',\omega)$ is purely transverse in $\boldsymbol{r}$, we may write the first term on the left-hand side as $-\boldsymbol{\nabla}\times[\boldsymbol{\nabla}\times\mathsf{f}_\Pi]$. Subsequently, we introduce f_E by means of (9), and eliminate f_X with the help of (21). The result is an inhomogeneous wave equation for f_E:

$$-\boldsymbol{\nabla}\times\big[\boldsymbol{\nabla}\times\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)\big]+\frac{\omega^2}{c^2}\,[1+\chi(\boldsymbol{r},\omega)]\;\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)=-\frac{\omega^2}{\rho\alpha c^2}\,v_\omega\,\chi(\boldsymbol{r},\omega)\,\mathsf{s}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big). \tag{24}$$

To solve it, we introduce the tensorial Green function G of this wave equation, which is defined as

$$-\boldsymbol{\nabla}\times\big[\boldsymbol{\nabla}\times\mathsf{G}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)\big]+\frac{\omega^2}{c^2}\,[1+\chi(\boldsymbol{r},\omega)]\;\mathsf{G}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)=\mathsf{I}\,\delta\big(\boldsymbol{r}-\boldsymbol{r}'\big). \tag{25}$$

In terms of G the solution of (24) reads

$$\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)=-\frac{\omega^2}{c^2}\int\mathrm{d}\boldsymbol{r}''\,\frac{1}{\rho''\alpha''}\,v''_\omega\,\chi\big(\boldsymbol{r}'',\omega\big)\,\mathsf{G}\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{s}\big(\boldsymbol{r}'',\boldsymbol{r}',\omega\big). \tag{26}$$

Now that we have found f_E in terms of s, it is straightforward to express all other tensorial coefficients in s. We first list the results for the coefficients of the field and polarization variables:

$$\begin{aligned}
\mathsf{f}_A\big(\boldsymbol{r},\boldsymbol{r}',\omega\big) &= -\frac{i}{\omega}\,\big[\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)\big]_{\mathrm{T}}\,, &(27)\\
\mathsf{f}_\Pi\big(\boldsymbol{r},\boldsymbol{r}',\omega\big) &= -\varepsilon_0\,\big[\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)\big]_{\mathrm{T}}\,, &(28)\\
\mathsf{f}_X\big(\boldsymbol{r},\boldsymbol{r}',\omega\big) &= -\frac{\varepsilon_0}{\rho\alpha^2}\,v_\omega\chi(\boldsymbol{r},\omega)\,\mathsf{s}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)-\frac{\varepsilon_0}{\alpha}\,\chi(\boldsymbol{r},\omega)\,\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big), &(29)\\
\mathsf{f}_P\big(\boldsymbol{r},\boldsymbol{r}',\omega\big) &= \frac{i\varepsilon_0}{\alpha^2}\,\omega\,v_\omega\chi(\boldsymbol{r},\omega)\,\mathsf{s}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)+i\alpha\,\frac{1}{\omega}\,\big[\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)\big]_{\mathrm{T}}+\\
&\quad+\frac{i\varepsilon_0\rho}{\alpha}\,\omega\,\chi(\boldsymbol{r},\omega)\,\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big), &(30)
\end{aligned}$$

where (26) should be inserted. The coefficients for the bath variables are

$$\begin{aligned}
\mathsf{f}_Y\big(\boldsymbol{r},\boldsymbol{r}',\omega',\omega\big) &= \frac{i}{\rho\omega}\,\mathsf{s}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)\,\delta\big(\omega-\omega'\big)-\frac{i\varepsilon_0}{\rho^2\alpha^2}\,\frac{\omega v_\omega v_{\omega'}}{(\omega+i0)^2-\omega'^2}\,\chi(\boldsymbol{r},\omega)\,\mathsf{s}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)-\\
&\quad-\frac{i\varepsilon_0}{\rho\alpha}\,\frac{\omega v_{\omega'}}{(\omega+i0)^2-\omega'^2}\,\chi(\boldsymbol{r},\omega)\,\mathsf{f}_E\big(\boldsymbol{r},\boldsymbol{r}',\omega\big), &(31)\\
\mathsf{f}_Q\big(\boldsymbol{r},\boldsymbol{r}',\omega',\omega\big) &= -i\rho\frac{\omega'^2}{\omega}\,\mathsf{f}_Y\big(\boldsymbol{r},\boldsymbol{r}',\omega',\omega\big). &(32)
\end{aligned}$$

The expressions listed here still depend on the tensor s, which is not yet known. It can be determined by employing the commutation relations (6).

Determination of the tensor $\mathsf{s}(\boldsymbol{r},\boldsymbol{r}',\omega)$. – The general form (11) should satisfy the commutation relations (6). The first of these leads upon using eqs. (2) to the following constraint:

$$\begin{aligned}
&\int\mathrm{d}\boldsymbol{r}''\Big\{\mathsf{f}_A^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{f}_\Pi\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)-\mathsf{f}_\Pi^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{f}_A\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)+\\
&\quad+\mathsf{f}_X^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{f}_P\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)-\mathsf{f}_P^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{f}_X\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)+\\
&\quad+\int_0^\infty\mathrm{d}\omega''\big[\mathsf{f}_Y^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega'',\omega\big)\cdot\mathsf{f}_Q\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'',\omega'\big)-\mathsf{f}_Q^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega'',\omega\big)\cdot\mathsf{f}_Y\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'',\omega'\big)\big]\Big\}=\\
&\quad=-i\hbar\,\delta\big(\omega-\omega'\big)\,\mathsf{I}\,\delta\big(\boldsymbol{r}-\boldsymbol{r}'\big), \qquad (33)
\end{aligned}$$

where $\mathsf{f}^\dagger(\boldsymbol{r},\boldsymbol{r}')$ equals $\mathsf{f}^*(\boldsymbol{r}',\boldsymbol{r})$ with interchanged tensor indices. If the expressions (27)-(32) are substituted on the left-hand side, we find contributions with either no factor s, or one or two such factors. We first consider the contribution that depends quadratically on s. In the $\boldsymbol{r}''$-integrand in (33) one encounters a frequency integral, which can be rewritten in terms of $\chi(\boldsymbol{r}'',\omega)$ and $\chi(\boldsymbol{r}'',\omega')$. After doing so, one finds that most terms depending quadratically on s drop out. A single term of this type survives. It yields the following contribution to the left-hand side of (33):

$$-\frac{2i}{\omega}\,\delta\big(\omega-\omega'\big)\int \mathrm{d}\boldsymbol{r}''\,\frac{1}{\rho''}\,\mathsf{s}^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{s}\big(\boldsymbol{r}'',\boldsymbol{r}',\omega\big). \tag{34}$$

The remaining contributions on the left-hand side of (33) can likewise be evaluated by first rewriting the frequency integrals in terms of the susceptibility. Subsequently, one finds that the integrands contain terms with the transverse part $[\mathsf{f}_E^\dagger(\boldsymbol{r},\boldsymbol{r}'',\omega)]_{\mathrm{T}''}$ of $\mathsf{f}_E^\dagger(\boldsymbol{r},\boldsymbol{r}'',\omega)$. Upon adding all such terms, one establishes that this transverse part is multiplied by the combination

$$\big[1+\chi\big(\boldsymbol{r}'',\omega'\big)\big]\,\mathsf{f}_E\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)+\frac{1}{\rho''\alpha''}\,v''_{\omega'}\,\chi\big(\boldsymbol{r}'',\omega'\big)\,\mathsf{s}\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big). \tag{35}$$

This combination is itself purely transverse in $\boldsymbol{r}''$, as one proves directly from (24). Hence, in the integral over $\boldsymbol{r}''$ one may replace the transverse part $[\mathsf{f}_E^\dagger(\boldsymbol{r},\boldsymbol{r}'',\omega)]_{\mathrm{T}''}$ by the full coefficient $\mathsf{f}_E^\dagger(\boldsymbol{r},\boldsymbol{r}'',\omega)$. A similar remark applies to the terms with the transverse part of $\mathsf{f}_E(\boldsymbol{r}'',\boldsymbol{r}',\omega')$. After these replacements, one arrives at an integral expression, which, after use of (24), becomes proportional to

$$\int\mathrm{d}\boldsymbol{r}''\Big[\big\{\boldsymbol{\nabla}''\times\big[\boldsymbol{\nabla}''\times\mathsf{f}_E^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\big]\big\}\cdot\mathsf{f}_E\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)-\mathsf{f}_E^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\big\{\boldsymbol{\nabla}''\times\big[\boldsymbol{\nabla}''\times\mathsf{f}_E\big(\boldsymbol{r}'',\boldsymbol{r}',\omega'\big)\big]\big\}\Big]. \tag{36}$$

A partial integration shows that this integral vanishes. Hence, the only terms that contribute to the left-hand side of (33) are those depending quadratically on s, as given in (34). As a consequence, the tensor s has to fulfill the condition

$$\int \mathrm{d}\boldsymbol{r}''\,\frac{1}{\rho''}\,\mathsf{s}^\dagger\big(\boldsymbol{r},\boldsymbol{r}'',\omega\big)\cdot\mathsf{s}\big(\boldsymbol{r}'',\boldsymbol{r}',\omega\big)=\frac{\hbar\omega}{2}\,\mathsf{I}\,\delta\big(\boldsymbol{r}-\boldsymbol{r}'\big). \tag{37}$$

Hence, the general form for s is

$$\mathsf{s}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big)=\sqrt{\frac{\hbar\omega\rho}{2}}\,\mathsf{U}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big). \tag{38}$$

The frequency-dependent tensor $\mathsf{U}(\boldsymbol{r},\boldsymbol{r}',\omega)$ must be unitary, so that it has the property $\int \mathrm{d}\boldsymbol{r}''\,\mathsf{U}^\dagger(\boldsymbol{r},\boldsymbol{r}'',\omega)\cdot\mathsf{U}(\boldsymbol{r}'',\boldsymbol{r}',\omega)=\mathsf{I}\,\delta(\boldsymbol{r}-\boldsymbol{r}')$. One may check that the second commutation relation in (6) does not lead to new constraints on s.

Upon inserting (38) and (26) in (27)-(32), and substituting the latter in (11), we obtain the diagonalizing operators $\boldsymbol{C}(\boldsymbol{r},\omega)$ and $\boldsymbol{C}^\dagger(\boldsymbol{r},\omega)$. They are determined up to a unitary transformation, which leaves both (5) and (6) invariant. The simplest choice for U is a local diagonal tensor $\exp[i\psi(\boldsymbol{r},\omega)]\,\mathsf{I}\,\delta(\boldsymbol{r}-\boldsymbol{r}')$, with an arbitrary phase factor $\exp[i\psi(\boldsymbol{r},\omega)]$. A convenient form for this phase factor is $\exp[i\psi(\boldsymbol{r},\omega)]=i\,\chi^*(\boldsymbol{r},\omega)/|\chi(\boldsymbol{r},\omega)|$, as it enables us to eliminate v_ω from the tensorial coefficient (26). In fact, from (22) one can prove the identity $v_\omega=\alpha\,[2\rho\,\mathrm{Im}\,\chi(\boldsymbol{r},\omega)/(\pi\varepsilon_0\omega)]^{1/2}/|\chi(\boldsymbol{r},\omega)|$.

Now that the diagonalizing operators of the model have been found, we can switch to the Heisenberg picture and derive explicit expressions for the time-dependent operators representing the electric field and the displacement field. With the above choice for the unitary transformation and the associated phase factor we obtain

$$\boldsymbol{E}(\boldsymbol{r},t) = -i\mu_0 \int \mathrm{d}\boldsymbol{r}' \int_0^\infty \mathrm{d}\omega\, \omega\, e^{-i\omega t}\, \mathsf{G}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big) \cdot \boldsymbol{J}\big(\boldsymbol{r}',\omega\big) + \text{h.c.}, \tag{39}$$

$$\begin{aligned}\boldsymbol{D}(\boldsymbol{r},t) = & -\frac{i}{c^2} \int \mathrm{d}\boldsymbol{r}' \int_0^\infty \mathrm{d}\omega\, e^{-i\omega t}\, \omega\, [1+\chi(\boldsymbol{r},\omega)]\, \mathsf{G}\big(\boldsymbol{r},\boldsymbol{r}',\omega\big) \cdot \boldsymbol{J}\big(\boldsymbol{r}',\omega\big) + \\ & + i \int_0^\infty \mathrm{d}\omega\, e^{-i\omega t}\, \frac{1}{\omega}\, \boldsymbol{J}(\boldsymbol{r},\omega) + \text{h.c.},\end{aligned} \tag{40}$$

with a current density $\boldsymbol{J}$ that is proportional to the diagonalizing operator:

$$\boldsymbol{J}(\boldsymbol{r},\omega) = \sqrt{\frac{\hbar\varepsilon_0\, \mathrm{Im}\,\chi(\boldsymbol{r},\omega)}{\pi}}\, \omega\, \boldsymbol{C}(\boldsymbol{r},\omega). \tag{41}$$

Similar expressions can be found for the Heisenberg operators that represent other canonical variables.

Expressions (39) and (40) agree with those found before by means of a Laplace-transform technique [4]. The electric field $\boldsymbol{E}(\boldsymbol{r},t)$ at the position $\boldsymbol{r}$ and the time t is given by an integral transform, which involves the current density $\boldsymbol{J}(\boldsymbol{r}',\omega)$ as a source and the Green function $\mathsf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)$ propagating a disturbance from $\boldsymbol{r}'$ to $\boldsymbol{r}$. The displacement field $\boldsymbol{D}(\boldsymbol{r},t)$ is the sum of two terms. First, it contains a contribution that is closely analogous to the expression for the electric field $\boldsymbol{E}(\boldsymbol{r},t)$, with the local permeability $1+\chi(\boldsymbol{r},\omega)$ as an extra factor in the integral transform. The second term in the polarization density depends on the local current density $\boldsymbol{J}(\boldsymbol{r},\omega)$ only, without an intervening permeability, and without propagation effects. For the homogeneous case expressions (39) and (40) reduce to those found in [2]. In a phenomenological quantization scheme [5], forms like (39) and (40) are postulated without proof. In that context $\boldsymbol{J}(\boldsymbol{r},\omega)$ is called a noise-current density, which is introduced without giving its precise connection to the fundamental dynamical variables of the system. In contrast, the present model leads to the specific expression (41) with (11) for $\boldsymbol{J}(\boldsymbol{r},\omega)$.

Conclusion. – By using Fano's diagonalization procedure we have succeeded in finding the ladder operators $\boldsymbol{C}$ and $\boldsymbol{C}^\dagger$, which annihilate and create the normal-mode excitations of the inhomogeneous damped-polariton model. Knowledge of these fundamental operators suffices to determine the full time dependence of the dynamical operators that describe the field and the dielectric. The general structure of these time-dependent operators agrees with that postulated in a phenomenological quantization scheme. Hence, the present diagonalization of the inhomogeneous damped-polariton model gives a justification of the phenomenological quantization method for inhomogeneous absorptive dielectrics.

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