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Microscopic theory of quantization of radiation in molecular dielectrics. II. Analysis of microscopic field operators

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The microscopic operators have been investigated for radiation and polarization fields within a discrete molecular medium. The medium comprises atoms (or molecules) each containing an arbitrary number of energy levels. Explicit mode expansions have been derived for the quantized microfields in terms of normal Bose operators for polariton creation and annihilation. These microscopic operators have been demonstrated to yield the correct macroscopic and local field operators presented in part I [Phys. Rev. A **53**, 3543 (1996)]. On the other hand, the commutation relations between the expanded microfields differ from the exact commutation relationships. This happens because the microfields have been described in terms of a continuous refractive index. Hence the expanded field operators do not extend to modes with extremely high frequencies ranging over the photon umklapp frequencies cG. In spite of that, the proper commutation relations hold between the resultant macroscopic (averaged) fields. This justifies separate quantization of the slowly modulated (macroscopic) part of the radiation field in linear dielectrics: such an approach is utilized in phenomenological quantization schemes. Finally, although the microscopic operators presented are not complete, the mode expansions adequately represent the quantized microfields associated with the optical modes in condensed molecular media. [S1050-2947(97)02302-0]

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I. INTRODUCTION

The quantum nature of light plays an important role in a number of distinctive phenomena, such as squeezing [1,2] and spontaneous emission [3–7]. In vacuum, the light is quantized by means of a standard procedure as depicted in textbooks on quantum electrodynamics (QED) [8–10] or quantum optics [11]. The quantization of radiation in a condensed medium is a less established issue. Traditionally, the (macroscopic) radiation field is quantized phenomenologically taking into account the influences of the polarizable medium implicitly through the linear [12–17] or nonlinear [18–21] susceptibilities. Although such a procedure is very convenient for utilitarian purposes, the phenomenological approach lacks a rigorous justification. Furthermore, the phenomenological quantization suffers from some difficulties, such as violation of microcausality in a dispersionless medium (as discussed in Refs. [22–26]), or the possible appearance of unphysical photon modes in the case of a nonlinear dispersive medium [20].

In several recent publications [3,22–26], another scheme has been investigated for quantization of the radiation field in linear dielectrics. The approach treats the matter explicitly by invoking the polariton concept [27–33]. Specifically, the electromagnetic field and the matter are considered to constitute a single dynamical system: The quanta of the normal modes for such a system are known as polaritons. The initial theories concentrated on either macroscopic [22–25] or local [3] fields. In the previous paper [26], a combined analysis has been carried out of the macroscopic and local fields in a molecular dielectric. The molecular medium was considered to be discrete and represented by an arbitrary number of energy levels for each constituent molecule (for mode details see part I [26]). Explicit normal-mode expansions have been derived for both local and macroscopic fields. Hence the formalism developed provides a tool for the analysis of propagation of the quantized radiation in molecular dielectrics, as well as consideration of molecule-radiation processes, such as spontaneous emission and linear absorption [34] or bimolecular multiphoton processes [35] in condensed media. Here the previous theory is extended to include the microscopic field operators, in addition to the macroscopic and local ones: This will provide a deeper understanding of the interplay between the microscopic and macroscopic approaches to the quantization of radiation in dielectrics.

The outline of the paper is as follows. In Sec. II the mode expansions are derived for the quantized microfields in terms of normal Bose operators for polariton creation and annihilation. The results are then analyzed in Sec. III. In Sec. III A the microscopic operators are demonstrated to produce the correct macroscopic (averaged) and local field operators. Section III B considers the commutation relations between the expanded microfields, highlighting the deviation from the true commutation relations and giving its explanation . The commutation relations between the averaged (macroscopic) fields are also discussed in Sec. III B. Finally, Sec. IV contains the concluding remarks. Technical details of the calculation of the local polarization field are placed in the Appendix.

II. MICROSCOPIC FIELD OPERATORS

In this section the normal-mode expansions will be obtained for the microscopic operators of radiation and polarization fields in a discrete molecular medium. We shall deal with a nonmagnetic molecular dielectric as described in

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more detail previously [26]. The system is characterized by zero magnetization, i.e.,

$$\mathbf{b}^{\perp}(\mathbf{r})=\mu_0\mathbf{h}^{\perp}(\mathbf{r}), \tag{2.1}$$

$\mathbf{h}^{\perp}(\mathbf{r})$ and $\mathbf{b}^{\perp}(\mathbf{r})$ being the microscopic operators for the magnetic field and induction. On the other hand, the microscopic operator for the transverse displacement field $\mathbf{d}^{\perp}(\mathbf{r})$ is related to the electric and polarization fields in a usual way as [9]

$$\mathbf{d}^{\perp}(\mathbf{r})=\varepsilon_0\mathbf{e}^{\perp}(\mathbf{r})+\mathbf{p}^{\perp}(\mathbf{r}), \tag{2.2}$$

where the full polarization field is, under the electric dipole approximation adopted in part I,

$$\mathbf{p}(\mathbf{r})=\sum_{\zeta}\boldsymbol{\mu}(\zeta)\,\delta(\mathbf{r}-\mathbf{r}_{\zeta}), \tag{2.3}$$

$\mathbf{p}^{\perp}(\mathbf{r})$ being the transverse (divergence-free) component of $\mathbf{p}(\mathbf{r})$. Here $\boldsymbol{\mu}(\zeta)$ is the operator for the electric dipole moment of molecule ζ positioned at $\mathbf{r}_{\zeta}$, and the summation is over the molecules ζ of the system.

The starting point of the present analysis will be the previously derived [26] normal-mode expansion of the averaged (i.e., macroscopic) polarization field:

$$\overline{\mathbf{p}}(\mathbf{r})=i\sum_{\mathbf{k}}\sum_{m}\sum_{\lambda=1}^{2}\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}[(n^{(m)})^2-1]\mathbf{e}^{(\lambda)}(\mathbf{k})\times(e^{i\mathbf{k}\cdot\mathbf{r}}P_{\mathbf{k},m,\lambda}-e^{-i\mathbf{k}\cdot\mathbf{r}}P^{\dagger}_{\mathbf{k},m,\lambda}), \tag{2.4}$$

with $k\ll2\pi/a$ (a being the distance of intermolecular separation). Here V_0 is the quantization volume $P^{\dagger}_{\mathbf{k},m\lambda}(P_{\mathbf{k},m,\lambda})$ is the Bose operator for creation (annihilation) of a polariton characterized by a wave vector $\mathbf{k}$, a polarization λ, and an extra index m, $\mathbf{e}^{(\lambda)}(\mathbf{k})$ being a unit vector for the polariton polarization. The index m labels the branches of polariton dispersion: Altogether there are $M_{\mathrm{mol}}+1$ dispersion branches (depicted in Fig. 2 of part I [26]), where M_{mol} is a number of excitation frequencies contained by each individual molecule of the medium. Here also

$$n^{(m)}\equiv n(\omega_k^{(m)}) \tag{2.5}$$

is the refractive index, presented explicitly in part I, and

$$v_g^{(m)}=c\left\{\frac{d[\omega n(\omega)]}{d\omega}\right\}^{-1}\Bigg|_{\omega=\omega_k^{(m)}}=\frac{d\omega_k^{(m)}}{dk} \tag{2.6}$$

is the group velocity, both quantities being calculated at the polariton frequency $\omega_k^{(m)}$:

$$\omega_k^{(m)}=ck/n(\omega_k^{(m)}). \tag{2.7}$$

At this juncture a couple of remarks need to be made concerning the macroscopic polarization field (2.4). First, the averaged field (2.4) has been determined from first principles on the basis of a microscopic model [26]. Second, the expansion (2.4) accommodates the transverse normal modes $[\mathbf{e}^{(\lambda)}\times(\mathbf{k})\perp\mathbf{k},\lambda=1,2]$ characterized by the first Brillouin zone wave vectors k with $k\ll2\pi/a$ (i.e., large wavelengths $\lambdabar=1/k\gg a$) [36]. The excluded longitudinal excitonlike modes ($\lambda=3$), or the modes with greater k do not play a significant role for most of the optical processes of interest. Furthermore, for modes with $k\cong\pi/a$ neither macroscopic smoothing can be carried out to average the field over the elementary cell of the medium, nor is a description in terms of continuous refractive index relevant. In this way, Eq. (2.4) represents the operator for the transverse $[\overline{\mathbf{p}(\mathbf{r})}\equiv\overline{\mathbf{p}^{\perp}(\mathbf{r})}]$ macroscopic field slowly varying from molecule to molecule.

Due to translational symmetry imposed on the molecular medium [26], the following relationship holds between the microscopic polarization field (2.3) and its macroscopic component $\overline{\mathbf{p}(\mathbf{r})}$:

$$\mathbf{p}(\mathbf{r})=\sum_{\mathbf{G}}\overline{\mathbf{p}(\mathbf{r})}e^{i\mathbf{G}\cdot\mathbf{r}}, \tag{2.8}$$

where the summation extends over the inverse lattice vectors $\mathbf{G}$. Substituting Eq. (2.4) for $\overline{\mathbf{p}(\mathbf{r})}$ into the above equation provides the explicit mode expansion of the microscopic operator:

$$\mathbf{p}(\mathbf{r})=i\sum_{\mathbf{k},\mathbf{G}}\sum_{m}\sum_{\lambda=1}^{2}\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}[(n^{(m)})^2-1]\mathbf{e}^{(\lambda)}(\mathbf{k})\times(e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}P_{\mathbf{k},m,\lambda}-e^{-i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}P^{\dagger}_{\mathbf{k},m,\lambda}) \tag{2.9}$$

for $k\ll2\pi/a$, i.e., we are again concentrating on the optical modes: In what follows this conditions is mostly kept implicit. It is noteworthy that although the macroscopic polarization field (2.4) is completely transverse, its microscopic counterpart (2.9) acquires a longitudinal contribution due to extra short-wavelength terms with $\mathbf{G}\neq\mathbf{0}$. Extracting the transverse component from Eq. (2.9), one finds

$$\mathbf{p}^{\perp}(\mathbf{r},t)=i\sum_{\mathbf{k},\mathbf{G}}\sum_{m}\sum_{\lambda=1}^{2}\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}[(n^{(m)})^2-1]\times\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})(e^{i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P_{\mathbf{k},m,\lambda}-e^{-i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P^{\dagger}_{\mathbf{k},m,\lambda}), \tag{2.10}$$

$$\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})=\sum_{\lambda_1=1}^{2}\mathbf{e}^{(\lambda_1)}(\mathbf{k}+\mathbf{G})[\mathbf{e}^{(\lambda_1)}(\mathbf{k}+\mathbf{G})\cdot\mathbf{e}^{(\lambda)}(\mathbf{k})], \tag{2.11}$$

with $\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{0})=\mathbf{e}^{(\lambda)}(\mathbf{k})$, the time variable emerging through replacement of the initial Schrödinger representation by the Heisenberg one, as

$$P_{\mathbf{k},m,\lambda}\rightarrow e^{-i\omega_k^{(m)}t}P_{\mathbf{k},m,\lambda}\quad\text{and}\quad P^{\dagger}_{\mathbf{k},m,\lambda}\rightarrow e^{i\omega_k^{(m)}t}P^{\dagger}_{\mathbf{k},m,\lambda}. \tag{2.12}$$

Other field operators can then be determined by exploiting the quantum Maxwell equations for the microscopic field operators in the Heisenberg representation:

$$\boldsymbol{\nabla}\times\mathbf{h}^{\perp}(\mathbf{r},t)=\frac{\partial\mathbf{d}^{\perp}(\mathbf{r},t)}{\partial t}, \tag{2.13}$$

$$\nabla\times\mathbf{e}^{\perp}(\mathbf{r},t)=-\frac{\partial\mathbf{b}^{\perp}(\mathbf{r},t)}{\partial t}. \tag{2.14}$$

Combining Eqs. (2.1), (2.2), (2.13), and (2.14) gives

$$\left(c^2\nabla^2-\frac{\partial^2}{\partial t^2}\right)\mathbf{d}^{\perp}(\mathbf{r},t)=c^2\nabla^2\mathbf{p}^{\perp}(\mathbf{r},t), \tag{2.15}$$

$$\varepsilon_0\left(c^2\nabla^2-\frac{\partial^2}{\partial t^2}\right)\mathbf{e}^{\perp}(\mathbf{r},t)=\frac{\partial^2\mathbf{p}^{\perp}(\mathbf{r},t)}{\partial t^2} \tag{2.16}$$

so that calling on Eq. (2.10) for $\mathbf{p}^{\perp}(\mathbf{r},t)$ one arrives at the mode expansions for the microscopic operators for the displacement and electric fields:

$$\begin{aligned}\mathbf{d}^{\perp}(\mathbf{r},t)=i\sum_{\mathbf{k},\mathbf{G}}\sum_{m}\sum_{\lambda=1}^{2}&\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}(n^{(m)})^2\\&\times\frac{c^2k^2-(\omega_k^{(m)})^2}{c^2k^2}\frac{c^2|\mathbf{k}+\mathbf{G}|^2}{c^2|\mathbf{k}+\mathbf{G}|^2-(\omega_k^{(m)})^2}\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})\\&\times(e^{i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P_{\mathbf{k},m,\lambda}\\&-e^{-i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P^{\dagger}_{\mathbf{k},m,\lambda})\end{aligned} \tag{2.17}$$

and

$$\begin{aligned}\mathbf{e}^{\perp}(\mathbf{r},t)=i\sum_{\mathbf{k},\mathbf{G}}\sum_{m}\sum_{\lambda=1}^{2}&\left(\frac{\hbar\omega_k^{(m)}v_g^{(m)}}{2\varepsilon_0cV_0n^{(m)}}\right)^{1/2}\\&\times\frac{c^2k^2-(\omega_k^{(m)})^2}{c^2|\mathbf{k}+\mathbf{G}|^2-(\omega_k^{(m)})^2}\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})\\&\times(e^{i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P_{\mathbf{k},m,\lambda}\\&-e^{-i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P^{\dagger}_{\mathbf{k},m,\lambda}).\end{aligned} \tag{2.18}$$

Here use has been made of the following:

$$[(n^{(m)})^2-1]=(n^{(m)})^2\frac{c^2k^2-(\omega_k^{(m)})^2}{c^2k^2}=\frac{c^2k^2-(\omega_k^{(m)})^2}{(\omega_k^{(m)})^2}. \tag{2.19}$$

The magnetic field operator may now be found utilizing Eqs. (2.1), (2.14), and (2.18):

$$\begin{aligned}\mathbf{h}^{\perp}(\mathbf{r},t)=i\sum_{\mathbf{k},\mathbf{G}}\sum_{m}\sum_{\lambda=1}^{2}&\left(\frac{\hbar\omega_k^{(m)}v_g^{(m)}n^{(m)}}{2\mu_0cV_0}\right)^{1/2}\frac{c^2k^2-(\omega_k^{(m)})^2}{ck}\\&\times\frac{c(\mathbf{k}+\mathbf{G})\times\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})}{c^2|\mathbf{k}+\mathbf{G}|^2-(\omega_k^{(m)})^2}(e^{i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P_{\mathbf{k},m,\lambda}\\&-e^{-i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P^{\dagger}_{\mathbf{k},m,\lambda}).\end{aligned} \tag{2.20}$$

Finally, since

$$\mathbf{e}^{\perp}(\mathbf{r},t)=-\frac{\partial\mathbf{a}^{\perp}(\mathbf{r},t)}{\partial t} \tag{2.21}$$

the expanded operator for the vector potential takes the form

$$\begin{aligned}\mathbf{a}^{\perp}(\mathbf{r},t)=\sum_{\mathbf{k},\mathbf{G}}\sum_{m}\sum_{\lambda=1}^{2}&\left(\frac{\hbar v_g^{(m)}}{2\varepsilon_0c\omega_k^{(m)}V_0n^{(m)}}\right)^{1/2}\\&\times\frac{c^2k^2-(\omega_k^{(m)})^2}{c^2|\mathbf{k}+\mathbf{G}|^2-(\omega_k^{(m)})^2}\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})\\&\times(e^{i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P_{\mathbf{k},m,\lambda}\\&-e^{-i[(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}-\omega_k^{(m)}t]}P^{\dagger}_{\mathbf{k},m,\lambda}),\end{aligned} \tag{2.22}$$

completing a list of the microscopic operators of interest in the Heisenberg representation. Transformation back to Schrödinger representation can be carried out easily by setting $t=0$, so that $\mathbf{p}^{\perp}(\mathbf{r},0)\equiv\mathbf{p}^{\perp}(\mathbf{r}),\mathbf{d}^{\perp}(\mathbf{r},0)\equiv\mathbf{d}^{\perp}(\mathbf{r})$, etc.

III. ANALYSIS OF THE RESULTS

A. Macroscopic and local fields

One can readily ensure that the microscopic field operators provide the correct macroscopic and local operators. Averaging the fields over the elementary cell, only the $\mathbf{G}=\mathbf{0}$ terms remain in the above mode expansions. The resultant macroscopic operators are obviously equivalent to those presented in part I [26]. Consider now the local fields, i.e., the field operators calculated at the molecular sites $\mathbf{r}_{\zeta}$. The operator for the local polarization field, analyzed in the Appendix, reads

$$\begin{aligned}\mathbf{p}^{\perp}(\mathbf{r}_{\zeta})=\frac{i}{3}\sum_{\mathbf{k}}\sum_{m}\sum_{\lambda=1}^{2}&\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}[(n^{(m)})^2-1]\mathbf{e}^{(\lambda)}\\&\times(\mathbf{k})(e^{i\mathbf{k}\cdot\mathbf{r}_{\zeta}}P_{\mathbf{k},m,\lambda}-e^{-i\mathbf{k}\cdot\mathbf{r}_{\zeta}}P^{\dagger}_{\mathbf{k},m,\lambda}),\end{aligned} \tag{3.1}$$

i.e.,

$$\mathbf{p}^{\perp}(\mathbf{r}_{\zeta})=\tfrac{1}{3}\,\overline{\mathbf{p}^{\perp}}(\mathbf{r}_{\zeta}). \tag{3.2}$$

On a similar basis, the microscopic expansion (2.17) produces the local displacement field

$$\begin{aligned}\mathbf{d}^{\perp}(\mathbf{r}_{\zeta})=i\sum_{\mathbf{k}}\sum_{m}\sum_{\lambda=1}^{2}&\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}\left[\frac{(n^{(m)})^2+2}{3}\right]\\&\times\mathbf{e}^{(\lambda)}(\mathbf{k})(e^{i\mathbf{k}\cdot\mathbf{r}_{\zeta}}P_{\mathbf{k},m,\lambda}-e^{-i\mathbf{k}\cdot\mathbf{r}_{\zeta}}P^{\dagger}_{\mathbf{k},m,\lambda}),\end{aligned} \tag{3.3}$$

which is in agreement with the previous result for $\mathbf{d}^{\perp}(\mathbf{r}_{\zeta})$ [26] obtained through bypassing explicit analysis of the microscopic fields. Finally, the remaining fields are characterized by the same local and macroscopic operators, i.e., $\mathbf{e}^{\perp}(\mathbf{r}_{\zeta})=\overline{\mathbf{e}^{\perp}}(\mathbf{r}_{\zeta}),\mathbf{h}^{\perp}(\mathbf{r}_{\zeta})=\overline{\mathbf{h}^{\perp}}(\mathbf{r}_{\zeta})$, etc. Thus one may write, using Eqs. (2.2) and (3.2),

$$\mathbf{d}^{\perp}(\mathbf{r}_{\zeta})=\varepsilon_0\overline{\mathbf{e}^{\perp}}(\mathbf{r}_{\zeta})+\tfrac{1}{3}\,\overline{\mathbf{p}^{\perp}}(\mathbf{r}_{\zeta}). \tag{3.4}$$

Equations (3.2) and (3.4), connecting the local and macroscopic fields, represent the relationships familiar from classic elctrodynamics [37]. It is noteworthy that the relationships have been established at quantum level in the present formalism.

B. Commutation relations

Consider next the equal-time commutators between the microscopic fields. The polariton operators $P_{\mathbf{k},m,\lambda}$ and $P^\dagger_{\mathbf{k},m,\lambda}$ obey the Bose commutation relations. Hence, exploiting the normal-mode expansions (2.17), (2.18), and (2.22), one finds

$$[a_l^\perp(\mathbf{r},t),d_j^\perp(\mathbf{r}',t)]=-\frac{i\hbar}{cV_0}\sum_{\mathbf{G},\mathbf{G}'}\sum_{\mathbf{k}}\sum_{\lambda=1}^{2}S_{ad}(\mathbf{k},\mathbf{G},\mathbf{G}')\times g_l^{(\lambda)}(\mathbf{k},\mathbf{G})\times g_j^{(\lambda)}(\mathbf{k},\mathbf{G}')e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}e^{-i(\mathbf{k}+\mathbf{G}')\cdot\mathbf{r}'} \tag{3.5}$$

and

$$[a_l^\perp(\mathbf{r},t),\varepsilon_0 e_j^\perp(\mathbf{r}',t)]=-\frac{i\hbar}{cV_0}\sum_{\mathbf{G},\mathbf{G}'}\sum_{\mathbf{k}}\sum_{\lambda=1}^{2}S_{ae}(\mathbf{k},\mathbf{G},\mathbf{G})g_l^{(\lambda)}(\mathbf{k},\mathbf{G})\times g_l^{(\lambda)}(\mathbf{k},\mathbf{G}')e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}e^{-i(\mathbf{k}+\mathbf{G}')\cdot\mathbf{r}'}, \tag{3.6}$$

where

$$S_{ad}(\mathbf{k},\mathbf{G},\mathbf{G}')=\sum_m\frac{v_g^{(m)}n^{(m)}}{c^2k^2}\frac{[c^2k^2-(\omega_k^{(m)})^2]^2}{c^2|\mathbf{k}+\mathbf{G}|^2-(\omega_k^{(m)})^2}\times\frac{c^2|\mathbf{k}+\mathbf{G}'|^2}{c^2|\mathbf{k}+\mathbf{G}'|^2-(\omega_k^{(m)})^2} \tag{3.7}$$

and

$$S_{ae}(\mathbf{k},\mathbf{G},\mathbf{G}')=\sum_m\frac{v_g^{(m)}}{n^{(m)}}\frac{[c^2k^2-(\omega_k^{(m)})^2]^2}{c^2|\mathbf{k}+\mathbf{G}|^2-(\omega_k^{(m)})^2}\times\frac{1}{c^2|\mathbf{k}+\mathbf{G}'|^2-(\omega_k^{(m)})^2}. \tag{3.8}$$

On the other hand, the commutation relationships between the microfields should read, according to the general principles of molecular quantum electrodynamics [9],

$$[a_l^\perp(\mathbf{r},t),d_j^\perp(\mathbf{r}',t)]=[a_l^\perp(\mathbf{r},t),\varepsilon_0 e_j^\perp(\mathbf{r}',t)]=-i\hbar\,\delta_{lj}^\perp(\mathbf{r}-\mathbf{r}'), \tag{3.9}$$

where $\delta_{lj}^\perp(\mathbf{r}-\mathbf{r}')$ is the transverse delta function. [8–10,38] The latter function may be represented as

$$\delta_{lj}^\perp(\mathbf{r}-\mathbf{r}')=\frac{1}{V_0}\sum_{\mathbf{k},\mathbf{G}}\frac{[|\mathbf{k}+\mathbf{G}|^2\delta_{lj}-(\mathbf{k}+\mathbf{G})_l(\mathbf{k}+\mathbf{G})_j]}{|\mathbf{k}+\mathbf{G}|^2}\times e^{i(\mathbf{k}+\mathbf{G})\cdot(\mathbf{r}-\mathbf{r}')}. \tag{3.10}$$

It is apparent that the relationships (3.5) and (3.6) differ from the exact commutation relations (3.9) and (3.10). This happens because our normal-mode expansions, described through the refractive index, are not complete. The theory concentrates on the modes with the frequencies $\omega_k^{(m)}$ much less than the photon umklapp frequencies cG $(G\neq 0)$ [39]. In other words, the modes with extremely high frequencies $(\omega_k^{(m)}\cong cG)$ have not been accommodated in the normal-mode expansions. Consequently the commutators (3.5) and (3.6) contain incomplete short-wavelength parts (the terms with $\mathbf{G},\mathbf{G}'\neq\mathbf{0}$). Nevertheless, since [24,26,40]

$$\sum_m v_g^{(m)}n^{(m)}=\sum_m v_g^{(m)}/n^{(m)}=c \tag{3.11}$$

one has

$$S_{ae}(\mathbf{k},\mathbf{0},\mathbf{0})=S_{ad}(\mathbf{k},\mathbf{0},\mathbf{0})=c \tag{3.12}$$

so that both the transformed (3.5) and (3.6) and the original (3.9) and (3.10) commutators are characterized by the same slowly oscillating (macroscopic) parts, represented by the terms with $\mathbf{G},\mathbf{G}'=\mathbf{0}$ and $k\ll 2\pi/a$. Accordingly, the incomplete microscopic commutators (3.5) and (3.6) give the correct macrocommutators, considered previously [26]:

$$[\overline{a_l^\perp}(\mathbf{r},t),\overline{d_j^\perp}(\mathbf{r}',t)]=[\overline{a_l^\perp}(\mathbf{r},t),\varepsilon_0\overline{e_j^\perp}(\mathbf{r}',t)]=-i\hbar\,\overline{\delta_{lj}^\perp}(\mathbf{r}-\mathbf{r}'), \tag{3.13}$$

where

$$\overline{\delta_{lj}^\perp}(\mathbf{r}-\mathbf{r}')=\frac{1}{V_0}\sum_{\mathbf{k}}(\delta_{lj}-\hat{k}_l\hat{k}_j)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \tag{3.14}$$

is the smoothed (coarse-grained) transverse delta function, the summation being over the first Brillouin zone wave vectors with $k\ll 2\pi/a$. It is noteworthy that both the macroscopic and microscopic transverse δ functions, when acting on a smooth function $\overline{F_l}(\mathbf{r})$, produce the equivalent results:

$$\int\overline{F_l}(\mathbf{r}')\overline{\delta_{lj}^\perp}(\mathbf{r}-\mathbf{r}')d\mathbf{r}'\approx\int\overline{F_l}(\mathbf{r}')\delta_{lj}^\perp(\mathbf{r}-\mathbf{r}')d\mathbf{r}'=\overline{F_j^\perp}(\mathbf{r}). \tag{3.15}$$

The same applies to the smoothened (coarse-grained) delta function $\overline{\delta}(\mathbf{r}-\mathbf{r}')$ featured in Eqs. (3.18) and (3.19) below:

$$\int\overline{\mathbf{F}}(\mathbf{r}')\overline{\delta}(\mathbf{r}-\mathbf{r}')d\mathbf{r}'\approx\int\overline{\mathbf{F}}(\mathbf{r}')\delta(\mathbf{r}-\mathbf{r}')d\mathbf{r}'=\overline{\mathbf{F}}(\mathbf{r}), \tag{3.16}$$

where $\delta(\mathbf{r}-\mathbf{r}')$ is an ordinary microscopic delta function.

The above analysis can be extended to other commutators as well. Since

$$\mathbf{b}^\perp(\mathbf{r},t)\equiv\mu_0\mathbf{h}^\perp(\mathbf{r},t)=\nabla\times\mathbf{a}^\perp(\mathbf{r},t) \tag{3.17}$$

the microcommutators $[d_j^\perp(\mathbf{r},t),b_l^\perp(\mathbf{r}',t)]$ and $[e_j^\perp(\mathbf{r},t),b_l^\perp(\mathbf{r}',t)]$ may be found straightforwardly utilizing Eqs. (3.5) and (3.6). Again, the microcommutators obtained are not complete, yet contain the proper macroscopic parts:

$$[\overline{d_j^\perp}(\mathbf{r},t),\overline{b_l^\perp}(\mathbf{r}',t)]=\varepsilon_0[\overline{e_j^\perp}(\mathbf{r},t),\overline{b_l^\perp}(\mathbf{r}',t)]=i\hbar\varepsilon_{jlp}\frac{\partial}{\partial r'_p}\overline{\delta}(\mathbf{r}-\mathbf{r}'), \tag{3.18}$$

with

$$\overline{\delta}(\mathbf{r}-\mathbf{r}')=\frac{1}{V_0}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{3.19}$$

ε_{jlp} being the Levi-Cività unit tensor of rank 3. Note that the macroscopic commutators (3.13) and (3.18) are of the same form as the corresponding microscopic commutators [41], the microscopic δ functions being now replaced by the coarse-grained ones. Finally, the expanded microfields $\mathbf{d}^\perp$,$\mathbf{e}^\perp$ and $\mathbf{p}^\perp$ (as well as the corresponding macrofields $\overline{\mathbf{d}^\perp}$,$\overline{\mathbf{e}^\perp}$, and $\overline{\mathbf{p}^\perp}$) commute at equal times between each other, as required. We conclude that disregard of the high-frequency normal modes does not affect the commutation relationships between the transverse macroscopic fields.

IV. CONCLUSION

The miscroscopic operators for radiation and polarization fields have been investigated from first principles in a discrete molecular medium. Explicit expansions have been derived for the quantized microfields in terms of normal Bose operators for polariton creation and annihilation. The microscopic operators have been demonstrated to produce the correct macroscopic and local field operators. On the other hand, the commutation relations between the expanded microfields differ from the true commutation relationships. This happens because the normal-mode expansions of the microfields, described through the refractive index, are not complete. The modes with extremely high frequencies (ranging over photon umklapp frequencies cG) have not been included. Furthermore, our mode expansions cover the transverse normal modes characterized by relatively small wave vectors $k \ll 2\pi/a$, a being the distance of intermolecular separation.

In spite of that, the proper commutation relations hold for the resultant macroscopic (the averaged) fields. This makes it possible to quantize separately the slowly modulated (the macroscopic) part of the radiation field in linear dielectrics. In fact, such an approach has been utilized in phenomenological schemes [15–17] of the field quantization. It is noteworthy that the macrocommutators have been preserved due to the relationships (3.11) involving summations over the dispersion branches $m=1,2,M_{\mathrm{mol}}+1$, where M_{mol} is the number of molecular frequencies accommodated. Therefore it is the correspondence of more than one normal frequency $\omega_k^{(m)}$ to each wave vector k which is essential for retaining the correct macrocommutators [24–26]. Finally, although the microfields presented are not complete, the omitted modes do not play a significant role for most optical processes of interest. That is, the mode expanions represent adequately the quantized microfields associated with the optical modes in condensed molecular media.

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APPENDIX A: LOCAL POLARIZATION FIELD

Since $\mathbf{e}^{i\mathbf{G}\cdot\mathbf{r}_\zeta}=1$, the operator of the local polarization field reads in Schrödinger representation, through Eq. (2.10),

$$\mathbf{p}^\perp(\mathbf{r}_\zeta)=i\sum_{\mathbf{k}}\sum_{m}\sum_{\lambda=1}^{2}\left(\frac{\varepsilon_0\hbar\omega_k^{(m)}v_g^{(m)}}{2cV_0n^{(m)}}\right)^{1/2}[(n^{(m)})^2-1]$$
$$\times\mathbf{A}^{(\lambda)}(\mathbf{k})(e^{i\mathbf{k}\cdot\mathbf{r}_\zeta}P_{\mathbf{k},m,\lambda}-e^{-i\mathbf{k}\cdot\mathbf{r}_\zeta}P^\dagger_{\mathbf{k},m,\lambda}), \tag{A1}$$

with

$$\mathbf{A}^{(\lambda)}(\mathbf{k})=\sum_{\mathbf{G}}\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G}). \tag{A2}$$

Performing summation over λ_1 in $\mathbf{g}^{(\lambda)}(\mathbf{k},\mathbf{G})$, given by Eq. (2.11), one has for the Cartesian components of $\mathbf{A}^{(\lambda)}(\mathbf{k})$

$$A_j^{(\lambda)}(\mathbf{k})=\sum_{p=1}^{3}e_p^{(\lambda)}(\mathbf{k})\sum_{\mathbf{G}}f_{jp}(\mathbf{k}+\mathbf{G}), \tag{A3}$$

where

$$f_{jp}(\mathbf{k}')=(\tfrac{1}{3}\delta_{jp}-\hat{k}_j\hat{k}_p)+\tfrac{2}{3}\delta_{jp}, \tag{A4}$$

with $\hat{\mathbf{k}}'\equiv\mathbf{k}'/k'$. The sum over $\mathbf{G}$ can be expressed as

$$\sum_{\mathbf{G}}f_{jp}(\mathbf{k}+\mathbf{G})=\frac{1}{N}\sum_{\mathbf{r}_{\zeta'}\neq 0}\left[\sum_{\mathbf{k}'}f_{jp}(\mathbf{k}')e^{i\mathbf{k}'\cdot\mathbf{r}_\zeta}\right]e^{-i\mathbf{k}\cdot\mathbf{r}_s}, \tag{A5}$$

where $\mathbf{k}'$ is no longer restricted to the first Brillouin zone. The sum over $\mathbf{k}'$ in the square brackets may be identified as the tensor for the dipole-dipole coupling. Accordingly, Eq. (A5) represents the familiar dipole sum [33,37] in which the omitted $\mathbf{r}_{\zeta'}=0$ term is due to an infinite self-field.

For $k\ll 2\pi/a$ the sum over $\mathbf{r}_{\zeta'}$ may be replaced by the integral in Eq. (A5), so that one finds from the resultant double Fourier integral

$$\sum_{\mathbf{G}}f_{jp}(\mathbf{k}+\mathbf{G})=\tfrac{1}{3}\delta_{jp}-\hat{k}_j\hat{k}_p. \tag{A6}$$

Here the contribution from the second term of Eq. (A4) has been excluded, as this term generates a δ fucntion at the origin $\mathbf{r}_{\zeta'}=0$. Since $\mathbf{e}^{(\lambda)}(\mathbf{k})\perp\mathbf{k}$ for $\lambda=1,2$, substituting Eq. (A6) into Eq. (A2) one arrives at

$$\mathbf{A}_j^{(\lambda)}(\mathbf{k})=\tfrac{1}{3}\,\mathbf{e}^{(\lambda)}(\mathbf{k}), \tag{A7}$$

which, together with Eqs. (A1), leads to the required results (3.1) or (3.2).

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[36] Furthermore, the dispersion branches with extremely high frequencies have not been covered by the dispersion relationship (2.7). These branches, ranging over photon umklapp frequencies cG (with $G \neq 0$), are well above the frequencies of infrared, optical, or ultraviolet modes of interest.

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[39] In addition, the normal-mode expansions presented are restricted to the transverse modes with $k \ll 2\pi/a$: for more details see Sec. II.

[40] The relationshilps analogous to Eq. (3.11) have also been considered by Ho and Kumar [25] for polaritons with two dispersion branches: $m=1,2$; $M_{\mathrm{mol}}=1$.

[41] Such as those given by Eq. (3.9); the microcommutators corresponding to the macrocommutator (3.18) can be found in the textbooks [8–11].