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Nonlinear polarizability tensor of an anisotropic medium

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Inversion of the retarded Green's function of the electromagnetic field of an anisotropic medium yielded a correct solution of the problem of the limiting transition in the nonlinear polarizability tensor (NPT) from the independent variables $\{\mathbf{k}, \omega\}$ to variables $\{\mathbf{k}, \omega_\rho(\mathbf{k})\}$ that satisfy the dispersion law of normal electromagnetic waves; this corresponds to the conditions of resonance between the extraneous sources and the natural oscillations of this system. An exact limiting expression having no poles over the entire interval of variation of its frequency arguments is obtained for the total NPT of a medium of arbitrary symmetry. The intensity of generation of the summary harmonic is calculated in the given-field approximation.

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I. INTRODUCTION

A microscopic calculation of the total nonlinear polarizability tensor (NPT) for concrete models of a crystal medium remains one of the vital problems of nonlinear optics. This is due to the need for knowing the NPT to solve Maxwell's equations for a nonlinear medium, as well as to obtain the probabilities (intensities) of various nonlinear processes.

It is known that normal electromagnetic waves in an anisotropic medium are in the general case neither longitudinal nor transverse,[1] and that this division is quite arbitrary. Therefore the choice of such models as optically isotropic cubic crystals[2] or a weakly anisotropic medium,[3] in which account is taken of the contribution of only transverse waves, is not always satisfactory for the calculation of the limiting value of the total NPT. First, the transverse-wave approximation leads not to a total but to a transverse NPT $\varepsilon^{\perp}_{ijl}$, having poles at the frequencies of the Coulomb exciton, i.e., in those frequency regions where the transverse-wave approximation itself cannot be used because the amplitudes of the electric fields of the corresponding transverse waves vanish. Second, in addition to the contribution of the transverse waves it is necessary to take into account also the contribution of the longitudinal waves, which turns to exert a substantial influence on the dispersion properties of the NPT ε_{ijl} even in the case of low anisotropy.

II. CONNECTION OF NPT WITH THE ANHARMONICITY COEFFICIENTS

With the aid of a Fourier transformation, Maxwell's equations for the electric field in a nonlinear homogeneous medium, with both the quadratic and linear polarizabilities taken into account in the material relations, can be represented in the form

$$\Delta_{ij}(\mathbf{k},\omega)E_j(\mathbf{k},\omega)=\frac{\omega^2}{c^2}\int\varepsilon_{ijl}(\mathbf{k},\omega;\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)E_j(\mathbf{k}_1,\omega_1)\times E_l(\mathbf{k}_2,\omega_2)\,\delta(\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2)\,\delta(\omega-\omega_1-\omega_2)\,d\mathbf{k}_1\,d\mathbf{k}_2\,d\omega_1\,d\omega_2, \tag{1}$$

where $\Delta_{ij}(\mathbf{k},\omega)=k^2\delta_{ij}-k_ik_j-\omega^2\varepsilon_{ij}(\mathbf{k},\omega)/c^2$, while $\varepsilon_{ij}(\mathbf{k},\omega)$ and $\varepsilon_{ijl}(\mathbf{k},\omega;\mathbf{k}_1\omega_1;\mathbf{k}_2,\omega_2)$ are the permittivity and nonlinear polarizability tensors.

The method of extraneous currents[4] was used in Refs. 3, 5, and 6 to establish the connection between the NPT and the anharmonicity coefficients in a polariton system:

$$\varepsilon_{ijl}(\mathbf{k},\omega;\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=\frac{V^2c^6\Delta_{im}(\mathbf{k},\omega)\,\Delta_{nj}(\mathbf{k}_1,\omega_1)\,\Delta_{pl}(\mathbf{k}_2,\omega_2)}{(4\pi)^2\hbar^3\omega_1\omega_2\omega}$$
$$\times\sum_{\rho_1\rho_2\rho_3}\left\{\frac{S^m_{\rho_3}(\mathbf{k})\,S^{*n}_{\rho_1}(\mathbf{k}_1)\,S^{*p}_{\rho_2}(\mathbf{k}_2)[W(\mathbf{k}_1\rho_1;\mathbf{k}_2\rho_2;\mathbf{k}\rho_3)+W(\mathbf{k}_2\rho_2;\mathbf{k}_1\rho_1;\mathbf{k}\rho_3)]}{\omega_{\rho_3}(\mathbf{k})\,\omega_{\rho_1}(\mathbf{k}_1)\,\omega_{\rho_2}(\mathbf{k}_2)[\omega-\omega_{\rho_1}(\mathbf{k}_1)-\omega_{\rho_2}(\mathbf{k}_2)][\omega_2-\omega_{\rho_2}(\mathbf{k}_2)]}\times\frac{1}{[\omega-\omega_{\rho_3}(\mathbf{k})]}+\ldots\right\}, \tag{2}$$

where $\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2$, $\omega=\omega_1+\omega_2$, the three dots denote five additional terms of similar structure, and $S\rho^i(\mathbf{k})$ is the amplitude of the electric field intensity of the polariton mode:

$$S_\rho{}^i(\mathbf{k})=S_\rho(\mathbf{k})\,l_i{}^{\mathbf{k}\rho}=-\left(\frac{2\pi\hbar\omega_\rho{}^2(\mathbf{k})}{Vkc^2}\right)^{1/2}\left(\frac{\mathbf{v}_{\rho\,\mathrm{gr}}(\mathbf{k})\mathbf{s}}{c}\right)^{1/2}g_{\mathbf{k}\rho}l_i{}^{\mathbf{k}\rho}. \tag{3}$$

Here V is the cyclicity volume, $\mathbf{s}=\mathbf{k}/|\mathbf{k}|$; $g_{\mathbf{k}\rho}$, $l_i^{\mathbf{k}\rho}$ and $\mathbf{v}_{\rho\mathrm{gr}}(\mathbf{k})=\partial\omega_\rho(\mathbf{k})/\partial\mathbf{k}$ are respectively the normalization factor, the unit polarization vector, and the group velocity of the ρ-the normal mode, the explicit forms of which are given in the Appendix.

The relation (2) is not accidental, since the use, on the one hand, of phenomenological equations (1) and, on the other, the Hamiltonian of a crystal accurate to three-particle interactions

$$H=\sum_{k\rho}\hbar\omega_\rho(\mathbf{k})\,\xi_\rho{}^+(\mathbf{k})\,\xi_\rho(\mathbf{k})+\sum_{\substack{\mathbf{k}_1\mathbf{k}_2\\ \rho_1\rho_2\rho_3}}\{W(\mathbf{k}_1\rho_1;\mathbf{k}_2\rho_2;\mathbf{k}_1+\mathbf{k}_2\rho_3)\times\xi_{\rho_3}{}^+(\mathbf{k}_1+\mathbf{k}_2)\,\xi_{\rho_2}(\mathbf{k}_2)\,\xi_{\rho_1}(\mathbf{k}_1)+\mathrm{H.c.}\} \tag{4}$$

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corresponds to one and the same approximation. In the last Hamiltonian, $\xi_\rho^+(\mathbf{k})$, $\xi_\rho(\mathbf{k})$ and $\hbar\omega_\rho(\mathbf{k})$ are respectively the creation, annihilation, and energy operators of the polariton of branch ρ with wave vector $\mathbf{k}$; W stands for the cubic anharmonicity coefficients, explicit expressions for which and whose connection with the nonlinear-effect tensor are given by Ovander.[7]

In expression (2), the arguments of $\mathbf{k}$ and ω are independent of each other. However, physical interest attaches to the value of the NPT at those argument values which satisfy the dispersion law of the normal electromagnetic field, and when the synchronism conditions are satisfied in the unbounded crystal.[7] For these arguments, the factors $\Delta_{ij}[\mathbf{k}, \omega_\rho(\mathbf{k})]S_\rho^j(\mathbf{k})$ vanish in accordance with the homogeneous Maxwell's equations for normal waves. Furthermore, owing to satisfaction of the energy and momentum conservation laws, the corresponding energy denominators also vanish. It is precisely these denominators which contain the term of (2) which is written up in explicit form and which gives a nonzero contribution to the limiting value of the NPT. This raises the question of finding the value of the limit

$$L_{\rho_0}{}^i(\mathbf{k})=\lim\frac{\Delta_{ij}(\mathbf{k},\omega)S_{\rho_0}{}^j(\mathbf{k})}{\omega-\omega_{\rho_0}(\mathbf{k})},\quad \omega\to\omega_{\rho_0}(\mathbf{k}) \tag{5}$$

for an anisotropic crystal.

III. LIMITING VALUE OF THE TOTAL NPT

Attempts to obtain the limiting value of the NPT were made numerous times. Thus, Obukhovskii and Strizhevskii[3] determined the limiting value for a crystal of cubic symmetry using the explicit expression for the phenomenological permittivity. In Agranovich's monograph[2] this limit was calculated for transverse waves in the approximation $\varepsilon_{ij}(\mathbf{k},\omega)=\delta_{ij}$ and a normal-wave dispersion law $\omega=ck$ that is valid only for transverse photons in vacuum. The limiting value of the NPT obtained in this manner is not the complete but the transverse NPT and agrees with the formula that can be obtained for the tensor $\varepsilon_{ijl}^{\perp}$ by calculating the polarizability induced in the crystal by transverse electromagnetic fields, using as the zeroth-approximation states not polaritons but Coulomb excitons. The question of the possibility of taking the limit for less rough approximations of the normal-wave spectrum also remained unclear. On the other hand, the value of the limit (5) has not yet been found for an anisotropic crystal.

The problem of the limiting transition can be solved correctly by taking into account the connection between the linear Maxwell operator $\Delta_{ij}(\mathbf{k},\omega)$ and the Fourier transform $D_{ij}(\mathbf{k},\omega)$ of an electromagnetic field in a medium

$$\Delta_{ij}(\mathbf{k},\omega)=-4\pi\omega^2D_{ij}^{-1}(\mathbf{k},\omega)/c^2. \tag{6}$$

In the exciton region of the spectrum

$$D_{ij}(\mathbf{k},\omega)=\frac{V}{\hbar}\sum_\rho\left\{\frac{S_\rho{}^i(\mathbf{k})S_\rho{}^j(\mathbf{k})}{\omega-\omega_\rho(\mathbf{k})}-\frac{S_\rho{}^{*i}(-\mathbf{k})S_\rho{}^j(-\mathbf{k})}{\omega+\omega_\rho(\mathbf{k})}\right\}+4\pi\delta_{ij}. \tag{7}$$

It is known[8] that the inversion of $D_{ij}(\mathbf{k},\omega)$ in the case of an anisotropic medium is a complicated problem. To solve it, we separate from the sum over the polariton states in (7) the resonant term with $\rho=\rho_0$, and include all the remaining terms of this sum, which have no singularities when the limit is taken, in the tensor $\Pi_{ij}(\mathbf{k},\omega)$. Then

$$D_{ij}(\mathbf{k},\omega)=\frac{V}{\hbar}\frac{S_0{}^iS_0{}^{*j}}{\omega-\omega_0}+\Pi_{ij}(\mathbf{k},\omega)+4\pi\delta_{ij}. \tag{8}$$

Here and below $\mathbf{S}_0=\mathbf{S}_{\rho0}(\mathbf{k})$ and $\omega_0=\omega_{\rho0}(\mathbf{k})$. For the inverse tensor $D_{ij}^{-1}(\mathbf{k},\omega)$ the following expression is valid:

$$D_{ij}^{-1}(\mathbf{k},\omega)=\left[\frac{C_{ij}(\mathbf{k},\omega)}{\omega-\omega_0}+F_{ij}(\mathbf{k},\omega)\right]\left[\frac{A(\mathbf{k},\omega)}{\omega-\omega_0}+B(\mathbf{k},\omega)\right]^{-1}, \tag{9}$$

where the quantities $C_{ij}(\mathbf{k},\omega)$, $F_{ij}(\mathbf{k},\omega)$, $A(\mathbf{k},\omega)$, and $B(\mathbf{k},\omega)$ contain no singularities when the limit is taken, and are equal to

$$C_{ij}(\mathbf{k},\omega)=(V/\hbar)e_{ikl}e_{jmn}S_0{}^nS_0{}^{*l}[4\pi\delta_{mk}+\Pi_{mk}(\mathbf{k},\omega)],$$
$$F_{ij}(\mathbf{k},\omega)={}^1/{}_2e_{ikl}e_{jmn}[4\pi\delta_{nl}+\Pi_{nl}(\mathbf{k},\omega)][4\pi\delta_{mk}+\Pi_{mk}(\mathbf{k},\omega)],$$
$$A(\mathbf{k},\omega)=(V/\hbar)F_{ij}(\mathbf{k},\omega)S_0{}^{*i}S_0{}^j,$$
$$B(\mathbf{k},\omega)={}^1/{}_3\,\mathrm{Sp}\,[\hat{F}(\mathbf{k},\omega)(4\pi\hat{I}+\hat{\Pi}(\mathbf{k},\omega))],$$

e_{ikl} is the fully antisymmetrical Levi-Civita tensor.

Using the property of the matrix $C_{ij}(\mathbf{k},\omega)$

$$C_{ij}(\mathbf{k},\omega_0)S_0{}^j=0, \tag{10}$$

which is the consequence of the homogeneous Maxwell's equations, and substituting relation (9) in (5) with allowance for the conduction (6), we obtain an expression for the limit

$$L_{\rho_0}{}^i(\mathbf{k})=-\frac{4\pi\omega_{\rho_0}{}^2(\mathbf{k})\hbar}{Vc^2}\left[\frac{S_{\rho_0}{}^i(\mathbf{k})}{|S_{\rho_0}(\mathbf{k})|^2}+Q_{\rho_0}{}^i(\mathbf{k})\right]. \tag{11}$$

The vector $\mathbf{Q}_{\rho0}(\mathbf{k})$ is orthogonal to the vector $\mathbf{S}^*_{\rho0}(\mathbf{k})$

$$Q_{\rho_0}{}^i(\mathbf{k})S_{\rho_0}{}^{*i}(\mathbf{k})=0$$

and is equal to

$$Q_{\rho_0}{}^i(\mathbf{k})=[(4\pi)^{-2}F_{jq}(\mathbf{k},\omega_0)S_0{}^q-S_0{}^j]e_{jkn}e_{kli}S_0{}^nS_0{}^{*l}.$$

Changing over in (2) to the corresponding limits and knowing their explicit form (11), we obtain a simple connection between the limiting value of the NPT of an anisotropic medium with the anharmonicity coefficients

$$\varepsilon_{ijl}(\omega_{\rho_3}(\mathbf{k}),\mathbf{k};\omega_{\rho_1}(\mathbf{k}_1),\mathbf{k}_1;\omega_{\rho_2}(\mathbf{k}_2),\mathbf{k}_2)=-\frac{4\pi}{V}[W(\mathbf{k}_1\rho_1;\mathbf{k}_2\rho_2;\mathbf{k}\rho_3)$$
$$+W(\mathbf{k}_2\rho_2;\mathbf{k}_1\rho_1;\mathbf{k}\rho_3)]\left[\frac{S_{\rho_3}{}^i(\mathbf{k})}{|\mathbf{S}_{\rho_3}(\mathbf{k})|^2}+Q_{\rho_3}{}^i(\mathbf{k})\right]\left[\frac{S_{\rho_1}{}^{*j}(\mathbf{k}_1)}{|\mathbf{S}_{\rho_1}(\mathbf{k}_1)|^2}+Q_{\rho_1}{}^{*j}(\mathbf{k}_1)\right]$$
$$\times\left[\frac{S_{\rho_2}{}^{*l}(\mathbf{k}_2)}{|\mathbf{S}_{\rho_2}(\mathbf{k}_2)|^2}+Q_{\rho_2}{}^{*l}(\mathbf{k}_2)\right], \tag{12}$$

where $\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2$. In the approximation of optically isotropic crystals, with account taken of the contribution of the transverse waves, expression (12) goes over into the well known formula for the transverse NPT,[2,3] which for convenience we write in the form

$$\varepsilon_{ijl}^{\perp}(\mathbf{k},\omega_{\rho_3}(\mathbf{k});\mathbf{k}_1,\omega_{\rho_1}(\mathbf{k}_1);\mathbf{k}_2,\omega_{\rho_2}(\mathbf{k}_2))$$
$$=-(4\pi/V)[W(\mathbf{k}_1\rho_1;\mathbf{k}_2\rho_2;\mathbf{k}\rho_3)+W(\mathbf{k}_2\rho_2;\mathbf{k}_1\rho_1;\mathbf{k}\rho_3)]$$
$$\times[S_{\rho_3}{}^{\perp i}(\mathbf{k})/|\mathbf{S}_{\rho_3}{}^{\perp}(\mathbf{k})|^2][\mathbf{S}_{\rho_1}^{*\perp j}(\mathbf{k}_1)/|\mathbf{S}_{\rho_1}{}^{\perp}(\mathbf{k}_1)|^2][S_{\rho_2}^{*\perp l}(\mathbf{k}_2)/|\mathbf{S}_{\rho_2}{}^{\perp}(\mathbf{k}_2)|^2]. \tag{13}$$

The quantities $\mathbf{Q}_\rho(\mathbf{k})$ do not enter in (13), since they vanish identically in the class of transverse waves $S_\rho^\perp(\mathbf{k})$.

IV. INTENSITY OF GENERATION OF SUMMARY HARMONIC

To solve the phenomenological Maxwell's equations for the field $\mathbf{E}^s(\mathbf{k},\omega)$ of the summary harmonic, we use the given-field approximation,[9] in which the amplitudes of the primary high-power waves $\mathbf{E}^0$ are regarded as specified classical functions. In this approximation, Eq. (1) takes the form

$$\Delta_{ij}(\mathbf{k},\omega)E_j^s(\mathbf{k},\omega)=\frac{\omega^2}{c^2}\int \varepsilon_{ijl}(\mathbf{k},\omega;\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)E_j^0(\mathbf{k}_1,\omega_1)E_l^0(\mathbf{k}_2,\omega_2)$$
$$\times\delta(\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2)\delta(\omega-\omega_1-\omega_2)d\mathbf{k}_1\,d\mathbf{k}_2\,d\omega_1\,d\omega_2. \tag{14}$$

The complete solution of (14), satisfying the zero initial conditions

$$\mathbf{E}^s(\mathbf{r},0)=\dot{\mathbf{E}}^s(\mathbf{r},0)=0$$

takes the form

$$E_i^s(\mathbf{r},t)=\sum_{\mathbf{k}\rho}\int_{-\infty}^{\infty}d\omega\, e^{i\mathbf{kr}}\,l_i^{\mathbf{k}\rho}E_\rho^s(\mathbf{k},\omega)\left\{e^{-i\omega t}-\left[\frac{\omega_\rho(-\mathbf{k})+\omega}{\omega_\rho(-\mathbf{k})+\omega_\rho(\mathbf{k})}\right]e^{-i\omega_\rho(\mathbf{k})t}\right.$$
$$\left.-\left[\frac{\omega_\rho(\mathbf{k})-\omega}{\omega_\rho(-\mathbf{k})+\omega_\rho(\mathbf{k})}\right]e^{i\omega_\rho(-\mathbf{k})t}\right\}, \tag{15}$$

here

$$E_\rho^s(\mathbf{k},\omega)=\varepsilon_{ij}(\mathbf{k},\omega_\rho(\mathbf{k}))E_i^s(\mathbf{k},\omega)\,l_j^{\mathbf{k}\rho}/\varepsilon_{ij}(\mathbf{k},\omega_\rho(\mathbf{k}))\,l_i^{\mathbf{k}\rho}l_j^{\mathbf{k}\rho}$$

is the ρ-th component of the Fourier transform of the particular solution of the inhomogeneous equation (14).

The energy transferred to the summary harmonic per unit time is equal to the work performed by the primary light waves and determines the integrated intensity of the process

$$I=-\int d\mathbf{r}\,E_i^s(\mathbf{r},t)j_i(\mathbf{r},t). \tag{16}$$

The current induced by the nonlinear polarizability of a unit volume of the medium is

$$j_i(\mathbf{r},t)=\partial P_i(\mathbf{r},t)/\partial t=-(i/4\pi)\int\varepsilon_{ijl}(\mathbf{k},\omega;\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$$
$$\times E_j^0(\mathbf{k}_1,\omega_1)E_l^0(\mathbf{k}_2,\omega_2)e^{i(\mathbf{kr}-\omega t)}\,\omega\,d\mathbf{k}_1\,d\mathbf{k}_2\,d\omega_1\,d\omega_2, \tag{17}$$

where $\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2$ and $\omega=\omega_1+\omega_2$.

To simplify the calculations, the primary fields $\mathbf{E}^0$ will be assumed to be monochromatic

$$E_i^0(\mathbf{k},\omega)=N^{1/2}S_\rho(\mathbf{k})\,l_i^{\mathbf{k}\rho}\delta(\mathbf{k}-\mathbf{k}_0)\delta(\omega-\omega_0)$$

and we change from summation over $\mathbf{k}$ to integration introducing the density of the final states of the field of the summary harmonic. In the final stage of the calculations it is necessary to go, for all the frequencies, to the limit as $\omega_0\to\omega_{\rho0}(\mathbf{k}_0)$, inasmuch as in the case of a weak nonlinearity the fields that take part in the process are the normal waves in the medium.

Substituting (15) and (17) in (16) and taking (2) and (12) into account, we obtain for the intensity of the summary harmonic

$$I=\frac{\bar{V}I_1I_2}{(2\pi)^2\hbar^3\omega_{\rho_1}(\mathbf{k}_1)\omega_{\rho_2}(\mathbf{k}_2)v_{\rho_1\,\mathrm{rp}}(\mathbf{k}_1)v_{\rho_2\,\mathrm{rp}}(\mathbf{k}_2)}$$
$$\times\int d\Omega'\,\frac{\omega^3_{\rho_3}(\mathbf{k}_1+\mathbf{k}_2)\,|\bar{W}(\mathbf{k}_1\rho_1;\mathbf{k}_2\rho_2;\mathbf{k}_1+\mathbf{k}_2\rho_3)+W(\mathbf{k}_2\rho_2;\mathbf{k}_1\rho_1;\mathbf{k}_1+\mathbf{k}_2\rho_3)|^2}{v_{\rho_3\,\mathrm{rp}}(\mathbf{k}_1+\mathbf{k}_2)\,v^2_{\Phi\,\rho_3}(\mathbf{k}_1+\mathbf{k}_2)}. \tag{18}$$

Here $I_{1,2}=\hbar\omega_{\rho1,2}(\mathbf{k}_{1,2})v_{\rho1,2\mathrm{gr}}(\mathbf{k}_{1,2})N_{1,2}/V$ are the intensities of the primary waves, $d\Omega'$ is the element of the solid angle in the direction of the group velocity vector of the summary harmonic $\mathbf{v}_{\rho3\mathrm{gr}}(\mathbf{k}_1+\mathbf{k}_2)$; $\mathbf{v}_{\rho\mathrm{gr}}(\mathbf{k})$ and $\mathbf{v}_{\mathrm{ph}\rho}(\mathbf{k})$ are the group and phase velocities of the normal waves. Formula (18) coincides with the expression for the intensity of the generation of the summary harmonic, which can be easily obtained by following the quantum-mechanical "golden rule" for the scattering.

It should be noted that both the limiting value of the NPT (12) and the intensity (18) remain finite over the entire interval of variation of their frequency arguments. The reason is that the spectrum of the normal waves in the medium differs greatly both from the spectrum of the Coulomb excitons and from the spectrum of the transverse photons in vacuum.

V. APPENDIX

We obtain now some useful formulas and relations by comparing the phenomenological and microscopic aspects of the solution of the problem of normal electromagnetic waves in an anisotropic dispersive medium.

In the absence of extraneous charges and currents we determine unambiguously from Maxwell's equations

$$\Delta_{ij}(\mathbf{k},\omega_\rho(\mathbf{k}))\,l_j^{\mathbf{k}\rho}=0 \tag{19}$$

the polarization vectors $l_i^{\mathbf{k}\rho}$ and the dispersion law $\omega=\omega_\rho(\mathbf{k})$ of the normal waves, where $\rho=1,2,\ldots$ numbers the roots of the equation

$$\det|\Delta_{ij}(\mathbf{k},\omega)|=0 \tag{20}$$

at a given wave vector $\mathbf{k}$.

The structure of the unit polarization vectors $l_i^{\mathbf{k}\rho}$ is[10]

$$l_i^{k\rho}=g_{k\rho}^{-1}\left[\tau_i^{\mathbf{k}\rho}-k_i\frac{k_j\tau_s^{\mathbf{k}\rho}\varepsilon_{js}(\mathbf{k},\omega_\rho(\mathbf{k}))}{k_jk_s\varepsilon_{js}(\mathbf{k},\omega_\rho(\mathbf{k}))}\right], \tag{21}$$

where $\tau_i^{\mathbf{k}\rho}$ is a unit transverse polarization vector, and satisfies the orthogonality condition

$$\varepsilon_{ij}(\mathbf{k},\omega_\rho(\mathbf{k}))\,l_i^{\mathbf{k}\rho}l_j^{\mathbf{k}\rho'}=\delta_{\rho\rho'}n_\rho^2(\mathbf{k})/g_{\mathbf{k}\rho}^2, \tag{22}$$

from which in fact we determine $g_{\mathbf{k}\rho}$.

In the general case the dispersion equation (20) breaks up into a product of two factors, the roots of which determine two sets of normal waves $\{\rho_1\}$ and $\{\rho_2\}$. In the particular case of an optically uniaxial crystal, these are the ordinary and extraordinary waves.

In the microscopic approach, the same spectrum of the normal electromagnetic waves (nonlongitudinal polaritons) is determined from a system of homogeneous equations for the Bogolyubov-Tyablikov[2] canonical-transformation coefficients

$$\sum_{j'=1}^{2}[N_{jj'}(\mathbf{k},\omega_\rho(\mathbf{k}))-\Delta^2(\mathbf{k},\omega_\rho(\mathbf{k}))\delta_{jj'}]U_{\mathbf{k}j'}(\rho)=0 \tag{23}$$

by equating to zero the determinant

$$\det|N_{jj'}(\mathbf{k},\omega)-\Delta^2(\mathbf{k},\omega)\delta_{jj'}|=0. \tag{24}$$

The coefficients $U_{\mathbf{k}j}(\rho)$, which determine the amplitude of the electric field of the normal wave, the anharmonicity coefficients, etc., are simply connected with the phenomenological characteristics of the anisotropic medium,[11] such as the refractive index $n_\rho(\mathbf{k})=kc/\omega_\rho(\mathbf{k})$ and the group velocity

$$U_{\mathbf{k}j}(\rho)=i\frac{n_\rho(\mathbf{k})+1}{2n_\rho(\mathbf{k})}\left(\frac{\mathbf{v}_{\rho\,\mathrm{gr}}(\mathbf{k})\mathbf{s}}{c}\right)^{1/2}\eta_j(\mathbf{k}\rho), \tag{25}$$

where we have used the relation $\partial\omega_\rho(\mathbf{k})/\partial k=v_{\rho\,\mathrm{gr}}(\mathbf{k})\mathbf{s}$. The difference between formula (25) and the corresponding formula in Ref. 11 lies in the absence of the factors $\eta_j(\mathbf{k}\rho)$, which have the property

$$\sum_{j=1}^{2}\eta_j^2(\mathbf{k}\rho)=1$$

and which are equal to

$$\eta_1(\mathbf{k}\rho)=\left[1+\frac{\Delta^2(\mathbf{k},\omega_\rho(\mathbf{k}))-N_{11}(\mathbf{k},\omega_\rho(\mathbf{k}))}{\Delta^2(\mathbf{k},\omega_\rho(\mathbf{k}))-N_{22}(\mathbf{k},\omega_\rho(\mathbf{k}))}\right]^{-1/2},$$
$$\eta_2(\mathbf{k}\rho)=\left[1+\frac{\Delta^2(\mathbf{k},\omega_\rho(\mathbf{k}))-N_{22}(\mathbf{k},\omega_\rho(\mathbf{k}))}{\Delta^2(\mathbf{k},\omega_\rho(\mathbf{k}))-N_{11}(\mathbf{k},\omega_\rho(\mathbf{k}))}\right]^{-1/2}. \tag{26}$$

Even in an anisotropic medium, the matrix $N_{jj}(\mathbf{k},\omega)$ can be reformulated in terms of the principal axis. Equation (24) is then represented as a product of two factors

$$[\Delta^2(\mathbf{k},\omega)-N_{11}(\mathbf{k},\omega)][\Delta^2(\mathbf{k},\omega)-N_{22}(\mathbf{k},\omega)]=0, \tag{27}$$

which define two families of normal waves $\{\rho_1\}$ and $\{\rho_2\}$. The unit transverse polarization vector of the polariton mode

$$\tau_i^{\mathbf{k}\rho}=\sum_{j=1}^{2}l_{\mathbf{k}j}{}^i\eta_j(\mathbf{k}\rho) \tag{28}$$

is then directed along one of the two principal axes of the tensor N_{jj}, since the quantities $\eta_j(\mathbf{k},\rho)$, according to (26), at the roots of the dispersion equation (27), are equal to

$$\eta_j(\mathbf{k},\{\rho_{j'}\})=\delta_{jj'}.$$

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