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CONTRIBUTION TO THE THEORY OF NONLINEAR POLARIZABILITY OF CRYSTALS

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A new method is proposed for calculating the nonlinear polarizability tensor of crystals $\epsilon_{ijl}(\mathbf{k}\omega', \mathbf{k}'\omega', \mathbf{k}''\omega'')$ which defines third-order nonlinear optical processes in the exciton spectral range. The main difference between the present method and the previous methods is that in the determination of the tensor ϵ_{ijl} real electromagnetic waves are employed in the medium for the zero approximation states. As is well known, the properties of such waves (the dispersion law, polarization) differ significantly from those of the approximate models, such as Coulomb excitons and transverse photons. A relation is established between the tensor ϵ_{ijl} and the cubic anharmonicity coefficients in a normal wave system, and it is demonstrated that the expression for ϵ_{ijl} found in this paper goes over into that found by other authors if the refractive indices of all the normal waves are assumed to be close to unity (or the tensor ϵ_{ij} is assumed to be a unit tensor). The method which is presented can also be used for calculating the nonlinear polarizability tensor ϵ_{ijlm}, etc.

1. INTRODUCTION

IN investigating the propagation of long-wave electromagnetic radiation in material media it is usual to use the phenomenological Maxwell equations augmented by the so-called material equations. The latter are (for instance, in the case of nonmagnetic media) understood to be relations which connect the electrical induction vector **D** and the electric field intensity vector **E**. Generally speaking, these relations are both nonlocal and nonlinear. The presence of nonlocality in these relations leads to the necessity of taking into account the so-called spatial dispersion of the dielectric permittivity tensor (see, for example, [1]). At the same time the presence of a nonlinearity leads to interaction of light waves in the crystal with each other and gives rise to nonlinear optical effects (see, for example, surveys [2-5]).

The relation between the vectors **D** and **E** can up to and including[1] terms quadratic in the field be written in the form:

$$D_i(\mathbf{r},t)=\int\limits_{-\infty}^{t}dt'\int d\mathbf{r}'\,\hat{\varepsilon}_{ij}(\mathbf{r}-\mathbf{r}',\ t-t')E_j(\mathbf{r}',t') + \int\limits_{-\infty}^{t}dt'\int\limits_{-\infty}^{t}dt''\int d\mathbf{r}'\int d\mathbf{r}''\hat{\varepsilon}_{ijl}(\mathbf{r}-\mathbf{r}',\mathbf{r}-\mathbf{r}'',t-t',t-t'') \times E_j(\mathbf{r}',t')E_l(\mathbf{r}'',t''). \tag{1}$$

Here account has been taken of the causality principle by virtue of which the induction at the instant t is determined only by the field $\mathbf{E}(\mathbf{r},t)$ in the past and present, i.e., at the times t', and $t''\le t$. In addition, it has been taken into account in (1) that for spatially homogeneous media, whose properties are in addition unchangeable in time, the kernels of the integral relation (1) depend only on the differences $\mathbf{r}-\mathbf{r}'$, $\mathbf{r}-\mathbf{r}''$, $t-t'$, and $t-t''$.

If one goes over to the Fourier representation

$$\mathbf{D}(\mathbf{r},t)=\int d\mathbf{k}\int d\omega\,\mathbf{D}(\mathbf{k},\omega)e^{i(\mathbf{kr}-\omega t)},$$
$$\mathbf{E}(\mathbf{r},t)=\int d\mathbf{k}\int d\omega\,\mathbf{E}(\mathbf{k},\omega)e^{i(\mathbf{kr}-\omega t)}, \tag{2}$$

relation (1) takes the form:[2]

$$D_i(\mathbf{k},\omega)=\varepsilon_{ij}(\omega,\mathbf{k})E_j(\mathbf{k},\omega)+\int\varepsilon_{ijl}(\mathbf{k}\omega,\mathbf{k}'\omega',\mathbf{k}''\omega'') \times E_j(\mathbf{k}',\omega')E_l(\mathbf{k}'',\omega'')\delta(\omega-\omega'-\omega'') \times\delta(\mathbf{k}-\mathbf{k}'-\mathbf{k}'')d\omega'd\omega''d\mathbf{k}'d\mathbf{k}''. \tag{3}$$

Here $\epsilon_{ij}(\omega,\mathbf{k})$ is the dielectric permittivity tensor with account of spatial dispersion (see [1]), whereas ϵ_{ijl} is the nonlinear polarizability tensor. It is readily seen that this tensor remains unchanged when its arguments $\mathbf{k}'\omega'$ and $\mathbf{k}''\omega''$ are interchanged with simultaneous replacement of the subscripts $j \rightleftarrows l$ (see also [6]).

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[1] Only third-order nonlinear optical processes can be considered in this approximation. We confine ourselves to this case. It is readily seen, however, that higher-order processes can be considered in analogous fashion.

[2] If we retain in (1) along with terms quadratic in the field **E** also cubic terms, then a term with the tensor ϵ_{ijlm} will appear in (3), etc.

The explicit form of the tensors ϵ_{ij} and ϵ_{ijl} can only be obtained in the framework of a microscopic theory. In particular, the tensor $\epsilon_{ij}(\omega, \mathbf{k})$ can, if one is not interested in the long-wavelength absorption edge (see [1, 7]), be found in a calculation of the polarizability of the crystal in the framework of perturbation theory in the approximation linear in the field **E**. If one chooses as the perturbing field the entire macroscopic field $\mathbf{E}(\mathbf{r}, t)$, then one must use as the states of the unperturbed problem states whose wave functions and energies are found without allowance for or in the absence of the field indicated above.[1, 8, 9]

If, on the other hand, only the transverse part of the macroscopic field appears as the perturbing field, then one must use as the states of the zeroth approximation the states of the Coulomb problem, i.e., of the problem corresponding in the case of a crystal to a complete account of the Coulomb interaction of the charges. However, the tensor found in this way $\epsilon^{\perp}_{ij}(\omega, \mathbf{k}) \neq \epsilon_{ij}(\omega, \mathbf{k})$ and is a nonanalytic function of the wave vector $\mathbf{k}$.[10]

In the available literature the nonlinear polarizability tensor ϵ_{ijl} is calculated by means of a procedure which is a direct generalization of the above procedure of calculating the tensors $\epsilon_{ij}(\omega, \mathbf{k})$ (see [11]) and $\epsilon^{\perp}_{ij}(\omega, \mathbf{k})$ (see [12, 13]). The perturbation here is the same macroscopic field, and only the polarizability is calculated more exactly—exact up to and including second-order terms in the perturbation.

Nonlinear optical effects in crystals can also be considered without resorting to the use of the phenomenological Maxwell equations and the tensor ϵ_{ijl}. Such an approach was presented for the exciton region of the spectrum by Ovander.[4] It consists in considering the total Hamiltonian of the crystal (charges + electromagnetic field) exact up to and including third-order terms in the Bose amplitudes of the excitons and photons, and the nonlinear effects appear in this approach because of the presence of anharmonicity in the system under consideration. If, on the other hand, one neglects this anharmonicity, i.e., one omits the cubic terms, then the Hamiltonian of the system (charge + field) can be diagonalized exactly.

As it turns out,[7] the corresponding long-wavelength normal oscillations satisfy the linear phenomenological Maxwell equations, i.e., they represent normal electromagnetic waves in a medium. Their properties (for example, the dispersion law) in the spectral region of interest to us differ essentially both from the properties of the excitons of the Coulomb problem and also from the properties of waves in vacuum. This fact is not strange, because in the approximation quadratic in the Bose operators a considerable part of the interaction between the Coulomb subsystem and the field of the transverse photons has already been taken into account [this part of the interaction leads to the circumstance that the tensor $\epsilon_{ij}(\omega, \mathbf{k}) \neq \delta_{ij}$]. The remaining part of this interaction giving rise to the anharmonicity terms leads to the interaction of normal waves with each other, i.e., to the appearance of nonlinear optical effects. Since the presence in relations (1) and (3) of terms nonlinear in the field **E** leads to the same effects, it is clear that there is a direct connection between the tensors of the nonlinearity polarizability and the anharmonicity coefficients, so that the tensor ϵ_{ijl} is determined by the cubic anharmonicity and the tensor ϵ_{ijlm} by the fourth-order anharmonicity, etc.

In the papers[11-13] mentioned above no account was taken in calculating the nonlinear polarizability tensor of the fact that the tensor $\epsilon_{ij}(\omega, \mathbf{k}) - \delta_{ij}$ leads, generally speaking, to a considerable rearrangement of the spectrum of normal oscillations; this rearrangement is particularly appreciable in the neighborhood of intense exciton absorption lines, and also outside absorption lines in crystals with a large dielectric constant. Therefore the method of calculation used in [11-13], which corresponds to the approximation of weak interaction between radiation and matter, is entirely applicable to gases, but for crystals it is generally speaking very crude.

In connection with this we obtained in this paper the nonlinear polarizability tensor ϵ_{ijl} for the exciton region of the spectrum by another method. In this method the interaction between the charged particles of the crystal and the radiation field existing in the crystal is not assumed to be weak, and the magnitude of the tensor of the nonlinear effects turns out to be proportional to the corresponding anharmonicity coefficients. An analogous method of calculating the linear polarizability tensor has already been employed previously in [14].

2. GENERAL EXPRESSION FOR THE NONLINEAR POLARIZABILITY TENSOR

Dzyaloshinskiĭ and Pitaevskiĭ have shown[15] (see also [16]) that the dielectric permittivity tensor $\epsilon_{ij}(\omega, \mathbf{k})$ can be expressed in terms of the corresponding Fourier component of the retarded Green's function

$$\Pi_{ij}(\mathbf{r}t, \mathbf{r}'t') = -i\langle[\hat{E}_i(\mathbf{r}, t), \hat{E}_j(\mathbf{r}', t')]\rangle\theta(t-t'), \quad (4)$$

where $\hat{\mathbf{E}}(\mathbf{r}, t)$ is the electric field intensity operator at the point $\mathbf{r}$ at the instant t, $\theta(t) = 0$ when $t < 0$ and $\theta(t) = 1$ when $t > 0$. The symbol $\langle [\hat{A}\hat{B}] \rangle$ denotes the statistical average of the commutator of the two operators:

$$[\hat{A}\hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A},$$

$$\langle [\hat{A}\hat{B}] \rangle \equiv \mathrm{Sp}\{\exp[(F-\hat{H})/kT][\hat{A}\hat{B}]\},$$

where $\hat{H}$ is the total Hamiltonian of the system (charges + field), F is the free energy of the system, and T its temperature.

Generalizing, one can also obtain an analogous formula for the nonlinear polarizability tensor ϵ_{ijl}. Indeed, let us substitute relation (3) in Maxwell's equations for the case of a nonmagnetic polarizable medium. We obtain then for plane waves (the density of external charges is assumed to be zero, since the density of external currents $\mathbf{j}^{\mathrm{ext}} \neq 0$):

$$\begin{aligned}&\left[k^2\delta_{ij} - k_ik_j - \frac{\omega^2}{c^2}\varepsilon_{ij}(\omega,\mathbf{k})\right]E_j(\mathbf{k},\omega)\\ &\quad - \frac{\omega^2}{c^2}\int \varepsilon_{ijl}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'')\cdot E_j(\mathbf{k}',\omega')E_l(\mathbf{k}'',\omega'')\\ &\quad \times \delta(\mathbf{k}-\mathbf{k}'-\mathbf{k}'')\delta(\omega-\omega'-\omega'')\,d\omega'd\omega''d\mathbf{k}'d\mathbf{k}''\\ &\quad = i4\pi c^{-2}\omega j_i^{ext}(\mathbf{k},\omega).\end{aligned} \tag{5}$$

If we introduce the notation

$$\Delta_{ij}(\mathbf{k},\omega) = k^2\delta_{ij} - k_ik_j - c^{-2}\omega^2\varepsilon_{ij}(\omega,\mathbf{k}), \tag{6}$$

then the solution of Eq. (5) for $\mathbf{E}(\mathbf{k}, \omega)$ can be set up in the form of an expansion of powers of $\mathbf{j}^{\mathrm{ext}}(\mathbf{k}, \omega)$ as follows:

$$\mathbf{E}(\mathbf{k},\omega) = \mathbf{E}^{(1)}(\mathbf{k},\omega) + \mathbf{E}^{(2)}(\mathbf{k},\omega) + \cdots; \tag{7}$$

$$E_l^{(1)}(\mathbf{k},\omega) = 4\pi i\omega c^{-2}\Delta_{lm}^{-1}(\mathbf{k},\omega)j_m^{ext}(\mathbf{k},\omega), \tag{8}$$

$$\begin{aligned}E_m^{(2)}(\mathbf{k},\omega) &= -\left(\frac{4\pi\omega}{c^3}\right)^2\Delta_{mj}^{-1}(\mathbf{k},\omega)\\ &\times\int \varepsilon_{jj_1j_2}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'')\omega'\omega''\ \Delta_{j_1l_1}^{-1}(\mathbf{k}'\omega')\\ &\times\Delta_{j_2l_2}^{-1}(\mathbf{k}'',\omega'')\mathrm{j}_{l_1}^{ext}(\mathbf{k}',\omega')\mathrm{j}_{l_2}^{ext}(\mathbf{k}'',\omega'')\delta(\mathbf{k}-\mathbf{k}'-\mathbf{k}'')\\ &\times\delta(\omega-\omega'-\omega'')\,d\mathbf{k}'d\mathbf{k}''d\omega'd\omega''.\end{aligned} \tag{9}$$

Let us use a gauge with a zero scalar potential. Then

$$\mathbf{E}(\mathbf{k},\omega) = i\omega c^{-1}\mathbf{A}(\mathbf{k},\omega), \tag{10}$$

and Eqs. (8) and (9) make it possible to find without difficulty the parts of the vector potential linear and quadratic in $\mathbf{j}^{\mathrm{ext}}$. Employing (8), (9), and (10), we find that the Fourier component of the vector potential can, in analogy with (7), be expressed in the form of a sum

$$\mathbf{A}(\mathbf{k},\omega) = \mathbf{A}^{(1)}(\mathbf{k},\omega) + \mathbf{A}^{(2)}(\mathbf{k},\omega) + \cdots. \tag{11}$$

Here $\mathbf{A}^{(1)}$ is proportional to the first power of $\mathbf{j}^{\mathrm{ext}}$. On the other hand, the expression for $\mathbf{A}^{(2)}$ is of the form

$$\begin{aligned}A_l^{(2)}(\mathbf{k},\omega) &= \frac{ic}{4\pi}\int T_{lmn}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'')j_m^{ekt}(\mathbf{k}'\omega')\\ &\times j_n^{ext}(\mathbf{k}''\omega'')\ \delta(\mathbf{k}-\mathbf{k}'-\mathbf{k}'')\delta(\omega-\omega'-\omega'')\\ &\times d\mathbf{k}'d\mathbf{k}''d\omega'd\omega'',\end{aligned} \tag{12}$$

$$\begin{aligned}T_{lmn}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'') &= (4\pi/c^2)^3\omega\omega'\omega''\Delta_{lp}^{-1}(\mathbf{k}\omega)\\ &\times\Delta_{qm}^{-1}(\mathbf{k}'\omega')\Delta_{rn}^{-1}(\mathbf{k}''\omega'')\varepsilon_{pqr}(\mathbf{k}\omega,\ \mathbf{k}'\omega',\ \mathbf{k}''\omega'').\end{aligned} \tag{13}$$

The vector potential of the field in the medium produced by extraneous currents and given by (11) can also be found by another method. Namely, denoting the operators in the presence of external fields with a superscript ext and retaining for the operators in the absence of an external field the notation without the superscript, we have

$$\hat{\mathbf{A}}^{ext}(\mathbf{r},t) = S_{ext}^{-1}(t)\hat{\mathbf{A}}(\mathbf{r},t)S_{ext}(t), \tag{14}$$

$$\hat{\mathbf{A}}(\mathbf{r},t) = e^{i\hat{H}t/\hbar}\hat{\mathbf{A}}(\mathbf{r})e^{-i\hat{H}t/\hbar}. \tag{15}$$

In (15) $\hat{H}$ is the total Hamiltonian of the crystal which includes the radiation field in the absence of external currents and charges, and $\hat{\mathbf{A}}(\mathbf{r})$ is the vector potential operator at the point $\mathbf{r}$ of the medium in the Schrödinger representation.

In relation (14)

$$S_{ext}(t) \equiv S_{ext}(t,-\infty) = T_t\exp\left\{-\frac{i}{\hbar}\int_{-\infty}^{t} H_{ext}(t')dt'\right\}, \tag{16}$$

$$\hat{H}_{ext}(t') = -\frac{1}{c}\int \mathbf{j}^{ext}(\mathbf{r},t')\hat{\mathbf{A}}(\mathbf{r},t')\,d\mathbf{r}, \tag{17}$$

where T_t denotes the sign of the operator ordering in time. It is assumed that $\mathbf{j}^{\mathrm{ext}}(\mathbf{r}, t \to -\infty) \to 0$ (see [16]).

The average value of the operator (14), understood in the sense of statistical averaging, can be expanded in a series of powers of a weak external current $\mathbf{j}^{\mathrm{ext}}$ and one can thus obtain independent expressions for the $\mathbf{A}^{(1)}$, $\mathbf{A}^{(2)}$, etc., which appear in relation (11).

In particular, the term of the expansion of $\langle \hat{\mathbf{A}}^{\mathrm{ext}}(\mathbf{r}, t) \rangle$ quadratic in the external current is of the following form:

$$\begin{aligned}A_l^{(2)}(\mathbf{r},t) &= \frac{1}{\hbar^2c^2}\int d\mathbf{r}'d\mathbf{r}''\left\{\int_{-\infty}^{t}\int_{-\infty}^{t} dt'dt''\langle\hat{A}_m(\mathbf{r}',t')\right.\\ &\times\hat{A}_l(\mathbf{r},t)\hat{A}_n(\mathbf{r}'',t'')\rangle\ j_m^{ext}(\mathbf{r}',t')j_n^{ext}(\mathbf{r}'',t'') - \int_{-\infty}^{t} dt'\\ &\times\int_{-\infty}^{t'} dt''\Big[\langle\hat{A}_e(\mathbf{r},t)\hat{A}_m(\mathbf{r}',t')\hat{A}_n(\mathbf{r}'',t'') + \hat{A}_n(\mathbf{r}'',t'')\\ &\left.\times\hat{A}_m(\mathbf{r}',t')\hat{A}_l(\mathbf{r},t)\rangle j_m^{ext}(\mathbf{r}',t')j_n^{ext}(\mathbf{r}'',t'')\Big]\right\}.\end{aligned} \tag{18}$$

In further calculations of the value of (18) one must take into account that the functions of the form

$$\varphi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') \equiv \langle \hat{A}_l(\mathbf{r}, t)\, \hat{A}_m(\mathbf{r}', t')\, \hat{A}_n(\mathbf{r}'', t'')\rangle \tag{19}$$

in the case of a homogeneous medium under consideration depend under stationary external conditions only on the difference of their arguments, i.e.,

$$\varphi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') \equiv \varphi_{lmn}(\mathbf{r}-\mathbf{r}', \mathbf{r}'-\mathbf{r}'', t-t', t'-t''),$$

so that their expansions in a Fourier series are of the form

$$\varphi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') = \int \varphi_{lmn}(\mathbf{k}_1\nu_1, \mathbf{k}_2\nu_2) \times \exp\{i\mathbf{k}_1(\mathbf{r}-\mathbf{r}') + i\mathbf{k}_2(\mathbf{r}'-\mathbf{r}'') - i\nu_1(t-t') - i\nu_2(t'-t'')\}\, d\mathbf{k}_1 d\mathbf{k}_2 d\nu_1 d\nu_2. \tag{20}$$

The functions φ_{lmn} are not gauge invariant. However, since

$$-\frac{1}{c^3}\frac{\partial^3}{\partial t\partial t'\partial t''}\varphi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') = \psi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t''),$$

where

$$\psi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') = \langle \hat{E}_l(\mathbf{r}, t)\hat{E}_m(\mathbf{r}', t')\hat{E}_n(\mathbf{r}'', t'')\rangle \equiv \psi_{lmn}(\mathbf{r}-\mathbf{r}', \mathbf{r}'-\mathbf{r}'', t-t', t'-t''), \tag{21}$$

it follows that

$$\psi_{lmn}(\mathbf{k}_1\nu_1, \mathbf{k}_2\nu_2) = ic^{-3}\nu_1\nu_2(\nu_2-\nu_1)\varphi_{lmn}(\mathbf{k}_1\nu_1, \mathbf{k}_2\nu_2).$$

Consequently, in accordance with (20),

$$\varphi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') = \frac{i}{c^3}\int \frac{\psi_{lmn}(\mathbf{k}_1\nu_1, \mathbf{k}_2\nu_2)}{\nu_1\nu_2(\nu_1-\nu_2)} \times \exp\{i\mathbf{k}_1(\mathbf{r}-\mathbf{r}') + i\mathbf{k}_2(\mathbf{r}'-\mathbf{r}'') - i\nu_1(t-t') - i\nu_2(t'-t'')\}\, d\mathbf{k}_1 d\mathbf{k}_2 d\nu_1 d\nu_2. \tag{22}$$

Utilizing this relation one can go over in (18) to the Fourier representation and one can determine $\mathbf{A}^{(2)}(\mathbf{k}, \omega)$. At the same time one must take into account that since the external currents $\mathbf{j}^{\text{ext}}(\mathbf{r}, t) \to 0$ for $t \to -\infty$, the integrals over the time

$$\int_{-\infty}^{t} \exp\{i\omega t'\}\, dt'$$

must be understood in the sense

$$\int_{-\infty}^{t} \exp\{i\omega t'\}\, dt' = \int_{-\infty}^{t} \exp\{i\omega t' + \delta t'\}\, dt',$$

where $\delta \to +0$.

Comparison of the expression for $A_l^{(2)}(\mathbf{k}, \omega)$, which can be obtained on the basis of (18), with (12) allows one, in view of the arbitrariness of the function $\mathbf{j}^{\text{ext}}(\mathbf{r}, t)$, to express the tensor $T_{lmn}(\omega\mathbf{k}, \omega'\mathbf{k}', \omega''\mathbf{k}'')$ in terms of the Fourier components of the Green function ψ_{ijl}. This comparison shows that the tensor[3]

$$\begin{aligned} T_{lmn}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'') = {} & [a_{mln}(-\mathbf{k}', \mathbf{k}''; \omega' + i\delta, -\omega'' - i\delta) \\ & + a_{nlm}(-\mathbf{k}'', \mathbf{k}'; \omega'' + i\delta, -\omega' - i\delta)] \\ & + [a_{lmn}(\mathbf{k}, \mathbf{k}''; -\omega - i\delta, -\omega'' - i\delta) + a_{lnm}(\mathbf{k}, \mathbf{k}'; -\omega \\ & - i\delta, -\omega' - i\delta)] + [a_{nml}(-\mathbf{k}'', -\mathbf{k}; \omega'' + i\delta, \omega + i\delta) \\ & + a_{mnl}(-\mathbf{k}', -\mathbf{k}; \omega' + i\delta, \omega + i\delta)], \end{aligned} \tag{23}$$

where, by definition,

$$a_{lmn}(\mathbf{k}, \mathbf{k}'; \omega, \omega') = -\frac{(2\pi)^7}{\hbar^2} \times \int \frac{d\nu_1\, d\nu_2}{\nu_1\nu_2(\nu_2-\nu_1)} \frac{\psi_{lmn}(\mathbf{k}\nu_1, \mathbf{k}'\nu_2)}{(\nu_1+\omega)(\nu_2+\omega')}. \tag{23a}$$

At the same time, in accordance with (13), the tensor

$$\varepsilon_{ijl}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'') = \left(\frac{c^2}{4\pi}\right)^3 \frac{\Delta_{ip}(\mathbf{k}\omega)\Delta_{mj}(\mathbf{k}'\omega')\Delta_{nl}(\mathbf{k}''\omega'')}{\omega\omega'\omega''} \times T_{pmn}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega''). \tag{24}$$

Relation (24) in conjunction with relation (23) makes it possible to express the tensor of nonlinear effects in terms of $\psi_{lmn}(\mathbf{k}\omega, \mathbf{k}'\omega')$ and components of the tensor $\epsilon_{ij}(\omega, \mathbf{k})$ [see (6)]. However, a calculation of all these quantities can be carried out only within the framework of a specific model of a crystal. Below we shall assume the tensor $\epsilon_{ij}(\omega, \mathbf{k})$ to be known and we shall calculate only the quantities $\psi_{lmn}(\mathbf{k}\omega, \mathbf{k}'\omega')$.

3. CALCULATION OF THE TENSOR ϵ_{ijl} FOR THE EXCITON REGION OF THE SPECTRUM

In order to find the Green's function (21), one must know the explicit expression for the total Hamiltonian of the crystal (electromagnetic field + charges), as well as the explicit expression for the electric field intensity operator. The expression for the Hamiltonian of the crystal in the exciton region of the spectrum with account of the radiation field has been obtained in [4, 17]; it can be written in the following form:

$$\hat{H} = \hat{H}_0 + \hat{H}^{\text{III}},$$

where

$$\hat{H}_0 = \sum_{\mathbf{k}, \rho} \mathscr{E}_\rho(\mathbf{k})\xi_\rho^+(\mathbf{k})\xi_\rho(\mathbf{k}),$$

$$\hat{H}^{\text{III}} = \sum_{\mathbf{k}_1, \mathbf{k}_2, \rho_1, \rho_2, \rho_3} \{\alpha(\rho_1\rho_2\rho_3; \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_1+\mathbf{k}_2)\xi_{\rho_1}(\mathbf{k}_1)$$

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[3] Expression (23) satisfies the symmetry requirement $T_{lmn}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'') = T_{lmn}(\mathbf{k}\omega, \mathbf{k}''\omega'', \mathbf{k}'\omega')$.

$$\times \xi_{\rho_2}(\mathbf{k}_2)\xi_{\rho_3}^{+}(\mathbf{k}_1+\mathbf{k}_2) + \beta(\rho_1\rho_2\rho_3; \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_1+\mathbf{k}_2)$$
$$\times \xi_{\rho_1}(\mathbf{k}_1)\xi_{\rho_2}(\mathbf{k}_2)\xi_{\rho_3}(-\mathbf{k}_1-\mathbf{k}_2) + \text{h. c.}\}. \tag{25}$$

Here $\mathbf{k}$ is the wave vector, ρ the number of the branch of the normal electromagnetic wave (the real exciton) in the crystal, $\mathscr{E}_\rho(\mathbf{k})$ is the energy of the corresponding elementary excitation, and α and β are the coefficients of cubic anharmonicity; explicit expressions for these are given in [4].

The electric field intensity operator $\hat{\mathbf{E}}(\mathbf{r}, t)$ in the medium can in the Heisenberg representation be written in the following form:

$$\hat{\mathbf{E}}(\mathbf{r}, t) = \sum_{\mathbf{k}, \rho} \{\mathbf{E}(\mathbf{k}, \rho) e^{i\mathbf{kr}} \xi_\rho(\mathbf{k}, t) + \mathbf{E}^*(\mathbf{k}, \rho) e^{-i\mathbf{kr}} \xi_\rho^{+}(\mathbf{k}, t)\}. \tag{26}$$

A method of calculating the quantities $\mathbf{E}(\mathbf{k}, \rho)$ appearing in (26) is indicated in [14]. Here we shall merely note that each of these quantities, being proportional to the amplitude of the normal electromagnetic wave with a wave vector $\mathbf{k}$ and frequency $\omega = \hbar^{-1}\mathscr{E}_\rho(\mathbf{k})$ [see (25)], satisfies the linear field equations [see also (6)]:

$$\Delta_{ij}(\omega, \mathbf{k}) E_j(\mathbf{k}, \rho)$$
$$\equiv [k^2\delta_{ij} - k_i k_j - \omega^2 c^{-2} \varepsilon_{ij}(\omega, \mathbf{k})] E_j(\mathbf{k}, \rho) = 0. \tag{27}$$

In accordance with the choice of the representation the operator $\xi_\rho(\mathbf{k}, t)$ is determined by the relation

$$\xi_\rho(\mathbf{k}, t) = \exp(i\hat{H}t/\hbar)\xi_\rho(\mathbf{k})\exp(-i\hat{H}t/\hbar).$$

If we take no account of the anharmonicity, i.e., if we assume $\hat{H}^{\mathrm{III}} = 0$, then

$$\xi_\rho(\mathbf{k}, t) = \exp[-i\Omega(\rho, \mathbf{k})t]\xi_\rho(\mathbf{k}), \tag{28}$$
$$\Omega(\rho, \mathbf{k}) = \hbar^{-1}\mathscr{E}_\rho(\mathbf{k}), \tag{29}$$

so that the operator of the electric field intensity

$$\hat{\mathbf{E}}(\mathbf{r}, t) = \hat{\mathbf{E}}^0(\mathbf{r}, t) = \sum_{\mathbf{k}, \rho} \{\mathbf{E}(\mathbf{k}, \rho)\xi_\rho(\mathbf{k})\exp\{i[\mathbf{kr} - \Omega(\rho, \mathbf{k})t]\}$$
$$+ \mathbf{E}^*(\mathbf{k}, \rho)\xi_\rho^{+}(\mathbf{k})\exp\{-i[\mathbf{kr} - \Omega(\rho, \mathbf{k})t]\}. \tag{30}$$

In this approximation the ψ_{lmn} as well as the tensor ϵ_{ijl} vanish. The vanishing of ψ_{lmn} is connected with the fact the averages of products of any three operators belonging to the set $\xi_\rho(\mathbf{k})$, $\xi_\rho^{+}(\mathbf{k}')$, etc. vanish.

With allowance for anharmonicity

$$\psi_{lmn}(\mathbf{r}t, \mathbf{r}'t', \mathbf{r}''t'') = \langle \hat{E}_l(\mathbf{r}, t)\hat{E}_m(\mathbf{r}', t')\hat{E}_n(\mathbf{r}'', t'')\rangle$$
$$= \langle S^{-1}(t, -\infty)\hat{E}_l^0(\mathbf{r}, t)S(t, t')\hat{E}_m^0(\mathbf{r}', t')S(t', t'')$$
$$\times \hat{E}_n^0(\mathbf{r}'', t'')S(t'', -\infty)\rangle, \tag{31}$$

where in the approximation linear with respect to the anharmonicity (namely in the approximation in which we shall carry out the calculations below)

$$S(t, t_0) = T_t \exp\left\{-\frac{i}{\hbar}\int_{t_0}^{t} \hat{H}^{\mathrm{III}}(t')\,dt'\right\} = 1 - \frac{i}{\hbar}\int_{t_0}^{t} \hat{H}^{\mathrm{III}}(t')\,dt'. \tag{32}$$

The operator $\hat{H}^{\mathrm{III}}(t')$ appearing in (32) is determined by the relation

$$\hat{H}^{\mathrm{III}}(t) = \exp(i\hat{H}_0 t/\hbar)\hat{H}^{\mathrm{III}}\exp(-i\hat{H}_0 t/\hbar), \tag{33}$$

Therefore, making use of (25), we find [see also (28)] that

$$\hat{H}^{\mathrm{III}}(t) = \sum_{\substack{\mathbf{k}_1, \mathbf{k}_2, \rho_1 \\ \rho_2, \rho_3}} \{\alpha(\rho_1\rho_2\rho_3; \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_1+\mathbf{k}_2)\xi_{\rho_1}(\mathbf{k}_1, t)\xi_{\rho_2}(\mathbf{k}_2, t)$$
$$\times \xi_{\rho_3}^{+}(\mathbf{k}_1+\mathbf{k}_2, t) + \beta(\rho_1\rho_2\rho_3; \mathbf{k}_1, \mathbf{k}_2, -\mathbf{k}_1-\mathbf{k}_2)\xi_{\rho_1}(\mathbf{k}_1, t)$$
$$\times \xi_{\rho_2}(\mathbf{k}_2, t)\xi_{\rho_3}(-\mathbf{k}_1-\mathbf{k}_2, t) + \text{h. c.}\}. \tag{34}$$

Relations (30), (32), and (34) make it possible to obtain the values of the quantities of interest to us $\psi_{lmn}(\mathbf{k}\omega, \mathbf{k}\omega, \mathbf{k}''\omega'')$ by means of simple calculations. Here one must bear in mind that terms in $\hat{H}^{\mathrm{III}}$ [see (25) and (34)] which contain coefficients of the form $\alpha(\rho_1\rho_2\rho_3; \mathbf{k}_1, 0, \mathbf{k}_1)$ or $\alpha(\rho_1\rho_2\rho_3; 0, \mathbf{k}_2, \mathbf{k}_2)$ need not be taken into account, because they are either proportional to the filling numbers of the normal waves, or are linear in the Bose amplitudes $\xi_\rho(\mathbf{k})$. Terms linear in $\xi_\rho(\mathbf{k})$ can be included in the zeroth-approximation Hamiltonian. On the other hand, as regards terms proportional to the filling numbers, they give in the calculation of the quantities $\langle \xi_{\rho_1}(\mathbf{k}_1, t)\xi_{\rho_2}(\mathbf{k}_2, t')\xi_{\rho_3}(\mathbf{k}_3, t'')\rangle$ a negligible contribution and will not be taken into account below, since in the region of the spectrum of normal oscillations $\hbar\omega \gg kT$. The same circumstance allows one to carry out the subsequent calculations under the assumption that the temperature $T = 0$.[4]

If the values of the quantities ψ_{lmn} obtained by the method indicated above are substituted in the formula (23a), then we obtain

$$a_{lmn}(\mathrm{k}, \mathrm{k}'; \omega, \omega') = \sum_{\gamma=1}^{4} a_{lmn}^{(\gamma)}(\mathrm{k}, \mathrm{k}'; \omega, \omega'), \tag{35}$$

where

$$a_{lmn}^{(1)}(\mathbf{k}, \mathbf{k}'; \omega, \omega')$$
$$= \frac{12\pi}{\hbar}\left(\frac{V}{\hbar}\right)^2 \sum_{\rho_1\rho_2\rho_3} \frac{E_l(\mathbf{k}, \rho_1)E_m(\mathbf{k}'-\mathbf{k}, \rho_2)E_n(-\mathbf{k}', \rho_3)}{\Omega(\rho_1, \mathbf{k}) + \Omega(\rho_2, \mathbf{k}'-\mathbf{k}) + \Omega(\rho_3, -\mathbf{k}')}$$
$$\times \frac{\beta^*(\rho_1\rho_2\rho_3; \mathbf{k}, \mathbf{k}'-\mathbf{k}, -\mathbf{k}')}{\Omega(\rho_1, \mathbf{k}) + \Omega(\rho_2, \mathbf{k}'-\mathbf{k})}$$

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[4] Effects connected with the participation of phonons (for example, Raman scattering) are not considered here for the sake of simplicity.

$$\times \frac{\Omega^{-1}(\rho_1, \mathbf{k})\,\Omega^{-1}(\rho_2, \mathbf{k}'-\mathbf{k})}{[\omega+\Omega(\rho_1,\mathbf{k})][\omega'+\Omega(\rho_1,\mathbf{k})+\Omega(\rho_2,\mathbf{k}'-\mathbf{k})]}, \qquad (36a)$$

$$a_{lmn}^{(2)}(\mathbf{k}, \mathbf{k}'; \omega, \omega') = -\frac{12\pi}{\hbar}\left(\frac{V}{\hbar}\right)^2$$

$$\times \sum_{\rho_1\rho_2\rho_3} \frac{E_l^*(-\mathbf{k},\rho_1)E_m^*(\mathbf{k}-\mathbf{k}',\rho_2)E_n^*(\mathbf{k}',\rho_3)}{\Omega(\rho_1,-\mathbf{k})+\Omega(\rho_2,\mathbf{k}-\mathbf{k}')+\Omega(\rho_3,\mathbf{k}')}$$

$$\times \frac{\beta(\rho_1\rho_2\rho_3; -\mathbf{k}, \mathbf{k}-\mathbf{k}', \mathbf{k}')}{\Omega(\rho_3,\mathbf{k}')+\Omega(\rho_2,\mathbf{k}-\mathbf{k}')}$$

$$\times \frac{\Omega^{-1}(\rho_3,\mathbf{k}')\,\Omega^{-1}(\rho_2,\mathbf{k}-\mathbf{k}')}{[\omega+\Omega(\rho_2,\mathbf{k}-\mathbf{k}')+\Omega(\rho_3,\mathbf{k}')][\omega'+\Omega(\rho_3,\mathbf{k}')]} \qquad (36b)$$

$$a_{lmn}^{(3)}(\mathbf{k}, \mathbf{k}'; \omega, \omega') = -\frac{4\pi}{\hbar}\left(\frac{V}{\hbar}\right)^2$$

$$\times \sum_{\rho_1\rho_2\rho_3} \frac{E_l(\mathbf{k},\rho_1)E_m(\mathbf{k}'-\mathbf{k},\rho_2)E_n^*(\mathbf{k}',\rho_3)}{\Omega(\rho_1,\mathbf{k})+\Omega(\rho_2,\mathbf{k}'-\mathbf{k})-\Omega(\rho_3,\mathbf{k}')}$$

$$\times \frac{\alpha^*(\rho_1\rho_2\rho_3; \mathbf{k}, \mathbf{k}'-\mathbf{k}, \mathbf{k}')}{\Omega(\rho_1,\mathbf{k})[\omega+\Omega(\rho_1,\mathbf{k})]}$$

$$\times \left\{ \frac{\Omega^{-1}(\rho_2,\mathbf{k}'-\mathbf{k})[\Omega(\rho_1,\mathbf{k})+\Omega(\rho_2,\mathbf{k}'-\mathbf{k})]^{-1}}{\omega'+\Omega(\rho_1,\mathbf{k})+\Omega(\rho_2,\mathbf{k}'-\mathbf{k})} \right.$$

$$\left. + \frac{\Omega^{-1}(\rho_3,\mathbf{k}')[\Omega(\rho_1,\mathbf{k})-\Omega(\rho_3,\mathbf{k}')]^{-1}}{\omega'+\Omega(\rho_3,\mathbf{k}')} \right\}, \qquad (36c)$$

$$a_{lmn}^{(4)}(\mathbf{k}, \mathbf{k}'; \omega, \omega') = -\frac{4\pi}{\hbar}\left(\frac{V}{\hbar}\right)^2$$

$$\times \sum_{\rho_1\rho_2\rho_3} \frac{E_l(\mathbf{k},\rho_1)E_m^*(\mathbf{k}-\mathbf{k}',\rho_2)E_n^*(\mathbf{k}',\rho_3)}{\Omega(\rho_1,\mathbf{k})-\Omega(\rho_2,\mathbf{k}-\mathbf{k}')-\Omega(\rho_3,\mathbf{k}')}$$

$$\times \frac{\alpha(\rho_3\rho_2\rho_1; \mathbf{k}', \mathbf{k}-\mathbf{k}', \mathbf{k})}{\Omega(\rho_3,\mathbf{k}')[\omega'+\Omega(\rho_3,\mathbf{k}')]}$$

$$\times \left\{ \frac{\Omega^{-1}(\rho_1,\mathbf{k})[\Omega(\rho_3,\mathbf{k}')-\Omega(\rho_1,\mathbf{k})]^{-1}}{\omega+\Omega(\rho_1,\mathbf{k})} \right.$$

$$\left. + \frac{\Omega^{-1}(\rho_2,\mathbf{k}-\mathbf{k}')[\Omega(\rho_2,\mathbf{k}-\mathbf{k}')+\Omega(\rho_3,\mathbf{k}')]^{-1}}{\omega+\Omega(\rho_3,\mathbf{k}')+\Omega(\rho_2,\mathbf{k}-\mathbf{k}')} \right\}. \qquad (36d)$$

Expressions (23), (24), (35), and (36) complete the solution of the posed problem of the calculation of the tensor of the nonlinear polarizability in the exciton region of the spectrum. However, in order to compare the results of this work with the results of previous papers we shall continue the discussion of the formula for $\epsilon_{ijl}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'')$ obtained above.

4. THE CASE OF LIMITINGLY WEAK EXCITON-PHOTON INTERACTION. DISCUSSION OF RESULTS

We note that the expression obtained above for ϵ_{ijl} is general in the sense that it contains terms responsible not only for the formation of a sum or difference light wave (with $\omega = \omega_1 + \omega_2$ and $\omega = \omega_1 - \omega_2$), but also, for example, the phenomenon of such a two-photon absorption in which the newly produced wave is again longitudinal. Therefore, for comparison with previous calculations of ϵ_{ijl} (see [11-13]), obtained to describe third-order effects in which all participating waves are light waves, we omit below in the expression for a_{lmn} terms in the sum over ρ_1, ρ_2, and ρ_3 in which even a single one of the normal waves is longitudinal.

Furthermore, before we go over in the expression for ϵ_{ijl} to some approximations, we analyze the question of how the interaction between the charges and the radiation field enters into this expression. With this in mind, we introduce a formal small parameter δ, to which the interaction operator of the light radiation with the charges of the crystal is proportional. This parameter enters in the expression for ϵ_{ijl} in two ways. First of all the quantities α and β are proportional to this parameter. In addition, the tensor $\epsilon_{ij}(\omega, \mathbf{k})$ and the energies $\mathscr{E}_\rho(\mathbf{k})$ and the vectors $\mathbf{E}(\mathbf{k}, \rho)$ depend on this parameter. For $\delta \to 0$ the quantities α and β vanish, and $\epsilon_{ij}(\omega, \mathbf{k})$, $\mathscr{E}_\rho(\mathbf{k})$, and $\mathbf{E}(\mathbf{k}, \rho)$ remain finite [$\epsilon_{ij}(\omega, \mathbf{k})$ goes over for $\delta \to 0$ to δ_{ij}, and $\mathscr{E}_\rho(\mathbf{k})$ and $\mathbf{E}(\mathbf{k}, \rho)$ go over into the corresponding quantities for the Coulomb excitons and transverse photons].

We shall show that the expression we obtained for ϵ_{ijl} goes over for $\delta \to 0$ in the lowest approximation in δ into an expression known in the literature (see, for example, [11-13]). Indeed (see [4]), for $\delta \to 0$ the quantities α turn out to be proportional to δ^2, whereas the coefficients β are proportional to δ^4. Therefore for $\delta \to 0$ terms with β can be omitted. Retaining terms with α, we let $\delta \to 0$ in all other quantities. For the transverse waves we then have the following relations:

$$\frac{\Delta_{ij}(\Omega,\mathbf{k})E_j(\mathbf{k},\rho)}{\omega\pm\Omega(\rho,\mathbf{k})} \to \frac{[(\mathbf{k}^2-\omega^2/c^2)\delta_{ij}-k_ik_j]\mathbf{E}_j(\mathbf{k},\rho)}{\omega\pm\Omega(\rho,\mathbf{k})},$$

$$= -c^{-2}[\omega\mp\Omega(\rho,\mathbf{k})]\mathbf{E}_i(\mathbf{k},\rho). \qquad (37)$$

In addition, the transverse-wave spectrum itself also undergoes changes when $\delta \to 0$. When account is taken of the retarded interaction (i.e., for $\delta \neq 0$) a whole set of states ρ, for which the amplitude of the electric field intensity $\mathbf{E}(\mathbf{k}, \rho)$ contains a nonzero transverse component, corresponds to a given arbitrary value of the wave vector $\mathbf{k}$. If, on the other hand, $\delta \to 0$, then the entire aggregate of normal waves splits into the waves of the Coulomb problem [for which the vector $\mathbf{E}(\mathbf{k}, \omega)$ is either longitudinal or vanishes] and transverse photons in vacuum. Since two states of the transverse photons (with energy $\hbar kc$ and different polarization) correspond to a given $\mathbf{k}$, only two terms remain in formulas (36) in each sum over ρ when $\delta \rightarrow 0$.

For the purpose of further simplification we will assume that the frequencies ω, ω', and ω'' appearing in (24) are not arbitrary, but are connected with $\mathbf{k}$, $\mathbf{k}'$, and $\mathbf{k}''$ by the dispersion equation $\omega = ck$ which is valid for transverse photons. In this case the right-hand part of relation (37) vanishes when the upper sign is taken and differs from zero when the lower sign is taken. With these approximations the tensor ϵ_{ijl} takes on the following form:

$$\varepsilon_{ijl}(\mathbf{k}\omega, \mathbf{k}'\omega', \mathbf{k}''\omega'') = \frac{4\pi iN}{V\omega\omega'\omega''}{}_{\mu_1\mu_2} \frac{(0|\hat{p}_i|\mathbf{k}'\mu_1)\,(\mathbf{k}'\mu_1|\hat{p}_j|\mathbf{k}\mu_2)\,(0|\hat{p}_l|\mathbf{k}\mu)^*}{[E^{\text{кул}}_{\mu_1}(\mathbf{k}') - \hbar\omega''][E^{\text{кул}}_{\mu_2}(\mathbf{k}) - \hbar\omega]} + \ldots, \tag{38}$$

where N is the number of unit cells in the crystal, V is its volume, $(0|\hat{p}_j|\mathbf{k}'\mu_1)$ etc., are the matrix elements of the j-th projection of the electron momentum operator of the unit cell, and $C_\mu^{\mathrm{Coul}}(\mathbf{k})$ is the energy of the Coulomb exciton. Comparing this relation with (23) from [13] we see that they coincide fully [for details about Eq. (38), see [13]].

In obtaining Eq. (38) use was made of the expressions for $\alpha(\rho_1\rho_2\rho_3;\, \mathbf{k}_1\mathbf{k}_2\mathbf{k}_3)$ cited by Ovander;[4] we did, however, omit terms corresponding to an account of anharmonicity in the Coulomb subsystem. As has been shown in [4], these terms are particularly important in the vicinity of resonances (frequencies of the Coulomb excitons[11]), where the normal electromagnetic waves are to a large extent mechanical.[5] However, this is not the only circumstance which renders relations (38) unsuitable near resonance. In this region of frequencies the quantities $\alpha(\rho_1\rho_2\rho_3;\, \mathbf{k}_1\mathbf{k}_2\mathbf{k}_3)$ obtained in [4] by a consistent separation of anharmonicity terms have no singularities, since even in finding the spectrum of elementary excitations corresponding to the operator $\hat{H}_0$ [see (25)] there is no intersection of branches of exciton and transverse photon frequencies because of the account of the exciton-photon interaction. However, in order to obtain (38) this effect too has to be ignored.

Thus the expressions for ϵ_{ijl} which were obtained in [11-13] correspond to very crude approximations. They are in particular unsuitable both in the vicinity of exciton absorption bands and also outside the absorption bands if the tensor $\epsilon_{ij}(\omega)$ must not be replaced by δ_{ij}. In this region of the spectrum the normal waves differ appreciably both from states of the Coulomb excitons and from states of transverse photons in vacuum. This difference can be ignored in those cases when the index of refraction of all waves is close to unity ($n - 1 \ll 1$). The expressions for ϵ_{ijl} obtained in [11-13] correspond precisely to this approximation which is only valid in the case of gases.

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[5] As is shown in [8], Sec. 8, for frequencies close to the resonance point the energy of the normal oscillations is a mixture of comparable fractions of mechanical and radiation energy.

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