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# ON THE THEORY OF ABSORPTION AND DISPERSION OF LIGHT IN CRYSTALS

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The results of the author's earlier papers[1,2] are generalized to the case of finite lifetime for the excited state of the crystal and to several light-absorption bands. Absorption of light is obtained as a result of the finite lifetime for the excited state with respect to thermal transitions. In the limiting case of a very long exciton wave the dependence of its energy on the direction of propagation has been obtained. A general theory of longitudinal polarization waves in crystals has been developed.. By means of a limiting transition to infinite lifetime for the excited states all the principal results of the earlier paper[1] have been obtained (taking into account the additional remarks of reference 4).

## 1. FORMULATION OF THE PROBLEM AND THE GENERAL QUANTUM MECHANICAL THEORY

In our earlier papers[1,2] it was shown that, in the range of exciton absorption of light in crystals, the dispersion has a number of peculiarities: for a given direction of propagation of a plane wave of given frequency and polarization the square of the index of refraction may have not one, but several values (for example, two values in cubic crystals). A proof has been given of the possibility of the existence in the crystal of longitudinal electric waves for which the magnetic field and the electric displacement are both equal to zero. Such waves may be propagated in an unisotropic crystal in certain special directions, and in a cubic crystal in any direction. Essential generalizations were obtained of the fundamental formulas of crystal optics — the Fresnel equations for the passage of light across the boundary between vacuum and the crystal, expressions for the coefficient of reflection of light from the surface of the crystal, formulas expressing the dependence of the indices of refraction on the light frequency, etc.

From the papers referred to above it follows that strong dispersion is possible in the neighborhood of a weak absorption band. In other words, the well-known general relation between the "area under the absorption curve" and the numerator of Sellmeyer's dispersion formula may be violated. This is connected with the fact that, under conditions of steady state illumination of the crystal, the absorption is determined by the finiteness of the lifetime of the exciton with respect to all transitions other than the radiative return of the system to the ground state. For an infinite lifetime of the exciton there will be no absorption at all, no matter how large the value of the oscillator strength of an elementary crystal cell, while the dispersion in this case may be large.[2] Since in the earlier papers the lifetime of the exciton was assumed to be infinite, we were able to calculate in those papers only the dispersion of light, but not its absorption. In the present paper we shall take into account the finite lifetime of the exciton, which will enable us to obtain general formulas for the absorption and corrections to the formulas for the dispersion previously obtained.

In proceeding to set up the Hamiltonian for the system we note that we should not introduce the concept of the dielectric constant of the crystal, since this concept itself is going to be re-examined. Therefore we treat the crystal as a collection of electrons and nuclei interacting with the electromagnetic field in vacuo. The electromagnetic wave perturbing the crystal is assumed to contain many photons and therefore will not be quantized, but will be introduced as a classical perturbing external field defined by a vector potential of the form

$$\mathbf{A} = \mathbf{A}_0 e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + \mathbf{A}_0^* e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)}. \tag{1}$$

The wavelength $2\pi/k$ is assumed to be considerably larger than the lattice constant. $\mathbf{A}$ is assumed to be small and in what follows we take into account effects of order not higher than the first with respect to $\mathbf{A}$.

The operator for the energy of the system may be written in the form $\hat{H} + \hat{W}$; $\hat{H}$ is the usual Schrödinger energy operator for the crystal, consisting of the kinetic energy of the electrons and the nuclei and of the potential energy of their Coulomb (instantaneous) interaction

$$\hat{W} = -\frac{1}{c}\int \hat{\dot{\mathbf{P}}}\cdot\mathbf{A}\,d\tau = \hat{W}'e^{-i\omega t} + \hat{W}''e^{i\omega t},$$

where

$$\hat{W}' = -\frac{1}{c}\int \hat{\dot{\mathbf{P}}}\cdot\mathbf{A}_0 e^{i\mathbf{k}\cdot\mathbf{r}}\,d\tau, \quad \hat{W}'' = -\frac{1}{c}\int \hat{\dot{\mathbf{P}}}\cdot\mathbf{A}_0^* e^{-i\mathbf{k}\cdot\mathbf{r}}\,d\tau,$$

$$d\tau = dx\,dy\,dz; \tag{2}$$

$\hat{\mathbf{P}}$ is the operator for the dipole moment per unit volume of the dielectric polarization of the crystal (cf. below), while $\hat{\dot{\mathbf{P}}}$ is the polarization current density. It should be emphasized that the irrotational part of the electric field of the fictitious charges due to the dielectric polarization, E′, represents the Coulomb interaction between the particles of the crystal and, consequently, is contained in $\hat{H}$. E′ should not appear a second time as a perturbation. Thus, **A** describes the macroscopic electric field with the field E′ subtracted.

Let $\Psi^0$ denote the eigenfunction of $\hat{H}$ which corresponds to the ground state of the crystal, and let $\mathscr{E}^0$ be the energy of the state. This state is not degenerate (we are speaking of a dielectric crystal). Further, let $\Psi_m$ and $\mathscr{E}_m$ be the wave functions and the energies of the excited states of the crystal in the absence of the external perturbation $\hat{W}$. The actually known functions $\Psi_m$ are not exact, but approximate eigenfunctions of $\hat{H}$, since "thermal" transitions between them occur without any external perturbation of the system. Thus, the exciton state, i.e., the excited state which has a single continuous quantum number (the quasi momentum[1]), may as a result of a thermal transition be converted into a "phonon wind" which is a state with many continuous quantum numbers — the momenta of the individual phonons. Thus, the exciton state is only approximately stationary. The same can also be said with respect to the other states appearing in the theory. However, we shall assume that these approximate eigenfunctions of $\hat{H}$ form a sufficiently complete system of orthogonal functions, and this enables us to represent the state of the crystal perturbed by light in the form

$$\Psi = \Psi^0 + \sum_m C_m\Psi_m, \quad |C_m| \ll 1.$$

We assume that the complete dependence on the time is contained in the coefficients $C_m$. We choose the zero for the energy of the system in such a way that $\mathscr{E}^0 = 0$. Thus, $\Psi_0^0$ and $\Psi_m$ do not depend on the time. The time-dependent Schrödinger equation leads to the following system of equations for $C_m$:

$$i\hbar\dot{C}_m = \sum_{m'} H_{mm'}C_{m'} + W_{m0}, \tag{3}$$

where

$$H_{mm'} = \int \Psi_m^* \hat{H}\Psi_{m'}\,d\Omega, \quad W_{m0} = \int \Psi_m^* \hat{W}\Psi^0\,d\Omega,$$

$$d\Omega = \prod_i dx_i\,dy_i\,dz_i. \tag{4}$$

The macroscopic average specific dipole moment due to the dielectric polarization of the crystal can be calculated by means of the formula

$$\mathbf{P}(\mathbf{r}) = \int \Psi^*\Psi\hat{\mathbf{P}}(\mathbf{r})\,d\Omega, \tag{5}$$

where

$$\hat{\mathbf{P}}(\mathbf{r}) = \sum_i e_i(\mathbf{r}_i - \mathbf{r})\,D(\mathbf{r}_i - \mathbf{r}),$$

$$D(\mathbf{r}) = D(xyz) = \begin{cases} 1 & \text{in the range } -1/2 \leqslant x, y, z \leqslant 1/2. \\ 0 & \text{elsewhere} \end{cases} \tag{6}$$

The summation is taken over all the particles of which the crystal is composed; i denotes the particle, $e_i$ denotes its charge, $r_i$ denotes its coordinate r is the coordinate of the macroscopic point in the crystal at which the value of the specific dipole moment is evaluated.

The substitution into (5) of $\Psi$ gives up to terms linear in **A**

$$\mathbf{P}(\mathbf{r}) = \sum_m [C_m\mathbf{P}_m(\mathbf{r}) + C_m^*\mathbf{P}_m^*(\mathbf{r})],$$

$$\mathbf{P}_m(\mathbf{r}) = \int \Psi^{0*}\Psi_m\hat{\mathbf{P}}(\mathbf{r})\,d\Omega. \tag{7}$$

It can be shown in a manner completely analogous to that employed in reference 1 for the transition from the ground to the exciton state, that also in the case of a transition to an arbitrary state with quasi-momentum **f**, i.e., to a state which possesses the translation property $T_n\Psi_m = e^{i\mathbf{f}\cdot\mathbf{n}}\Psi_m$ (**n** is the integral lattice vector), one obtains

$$\mathbf{P}_m(\mathbf{r}) = e^{i\mathbf{f}\cdot\mathbf{r}}\mathbf{P}_m(0), \quad \text{if } |\mathbf{f}a_j| \ll \pi; \tag{8}$$

$a_j$ are the lattice constants, j = 1, 2, 3. The vector **f** can be used as one of the quantum numbers of the state m.

On substituting (2) and (8) into (4) we obtain

$$W_{m0} = W'_{m0}e^{-i\omega t} + W''_{m0}e^{i\omega t}, \quad W'_{m0} = -\frac{i\omega_m V}{c}\mathbf{P}_m^*(0)\,\mathbf{A}_0\delta_{\mathbf{k},\mathbf{f}},$$

$$W''_{m0} = -\frac{i\omega_m V}{c}\mathbf{P}_m^*(0)\,\mathbf{A}_0^*\delta_{-\mathbf{k},\mathbf{f}}. \tag{9}$$

Here we have introduced the notation $\omega_m = \mathscr{E}_m/h$, $V = L^3$, L is the size of the cyclic region in which the wave functions $\Psi^0$ and $\Psi_m$ have been orthonormalized. Each of the components of the vectors **f** and **k** is an integral multiple of $2\pi/L$.

In solving the system of equations (3) we first introduce into the right hand side of (3), for the sake of simplicity, only the first of the two terms appearing in expression (9) — the term proportional to $e^{-i\omega t}$. The result due to the second term proportional to $e^{i\omega t}$ we shall add at the very end, obtaining it by analogy through the substitutions $\mathbf{A}_0 \to \mathbf{A}_0^*$, $\omega \to -\omega$, $\mathbf{k} \to -\mathbf{k}$.

The main task of this section is the determination of the specific polarization of the crystal by means of formulas (7) and (3). In carrying this out it is important to decide what initial conditions are to be adopted for $C_m$ in solving the system of inhomogeneous equations (3). It might appear to be natural to set $C_m = 0$ at time $t = 0$ at which illumination begins, since the system is still in its ground state at that instant. Then, under the perturbing action of the light, $C_m$ varying in accordance with (3) would increase in absolute value with time which, as usual, means that light is absorbed. The process actually occurs in this way, but only during a very short time. This time is considerably shorter than the duration of experiments on the measurement of light absorption in a crystal under conditions of steady-state illumination. Indeed, the increase in the absolute value of $C_m$ with time in accordance with (7) means that the amplitudes of oscillation of the specific dipole moment $\mathbf{P}(\mathbf{r})$ increase with time. In other words, the polarizability of the crystal varies with time. Evidently such a process does not correspond to the experimental conditions of measurement of the steady-state values of the complex dielectric constant of the crystal.

In order that the polarizability of the crystal not depend on the time, the coefficients $C_m$ in formula (7) must vary in time strictly sinusoidally with a frequency $\omega$. The system of equations (7) and (9) does in fact have such a particular solution. However, in this case the absolute values of all the $C_m$ are constant and, consequently, there is no absorption of light at all. This steady-state solution corresponds to such a state of the system when the crystal as a result of absorbing the incident light is heated to such a high temperature that the rate of radiation of light energy equals the absorption rate; the radiation, of course, may have frequencies different from $\omega$. In such a case the energy and the temperature of the crystal will not vary in time. Such a steady state of the system is characterized by optical crystal constants that remain unchanged with time. But evidently this state also does not correspond to the usual conditions of experimental determination of the complex dielectric constant.

The correct initial conditions for $C_m$ must reflect the fact that the usual experimental measurement of the absorption of light in a crystal is necessarily accompanied by the heating of the crystal by the absorbed light (there is no provision for heat removal). Therefore, there appear in the crystal polarization waves of thermal origin with amplitudes that are not proportional to the field of the perturbing light, but depend on the duration of the preceding illumination. The frequencies of these thermal polarization waves have nothing in common with the frequency of the perturbing light. This thermal part of the polarization is also contained in the right-hand side of formula (7), together with the usual polarization which is synchronized with the oscillations of the perturbing light. For the sake of brevity, we shall call the first and second parts of the polarization "thermal" and "synchronous," respectively. It is possible to describe only the synchronous part of the dielectric polarization by means of a complex dielectric constant.

In proceeding to the formulation of the initial conditions for $C_m$, we shall divide all the excited states of the crystal into two groups. In the first group we shall include a comparatively small number of states into which the system may make transitions directly from the ground state under the action of light; these are such states m for which $W_{m0}$ is appreciable. The second group will include those states for which $W_{m0}$ is negligibly small, and into which phototransitions from the ground state are forbidden; the system may go over into these states by means of thermal transitions from states of the first group. The majority of the states of the crystal belong to the second group. We shall continue to denote the states of the first group by the subscript m, while we shall introduce the subscript q to denote the states of the second group. Further, we assume that for the states of the first group the coefficients $C_m$ attain a steady state, i.e., begin to vary sinusoidally with time in accordance with

$$C_m = A_m e^{-i\omega t}. \tag{10}$$

after the lapse of a short time since the beginning of illumination.

The terms corresponding to these states in the right-hand side of (7) give rise to the synchronous part of the polarization. With respect to the states of the second group we assume that $C_q = 0$ at $t = 0$. This means that at the initial instant of time the temperature of the crystal was equal to zero. As t increases $C_q$ become different from zero as a result of thermal transitions of the system from states of the first group into states of the second group. This corresponds to the heating of the crystal by the absorbed light. The terms in formula (7) containing the coefficients $C_q$ comprise the thermal part of the polarization.

We shall now obtain the solution of the system of equations (3) which satisfies the conditions formulated above. We choose tentatively basis functions $\Psi_m$ that make the matrix $H_{mm'}$ diagonal in the subspace of the functions of the first group. This may be easily done, since the number of states of the first group is not large. As a result, the equations in (3) assume the form:

$$i\hbar\dot{C}_m = \mathscr{E}_m C_m + \sum_q H_{mq} C_q + W'_{m0} e^{-i\omega t}; \tag{11}$$

$$i\hbar\dot{C}_q = \mathscr{E}_q C_q + \sum_m H_{qm} C_m. \tag{12}$$

Since $\Psi_m$ and $\Psi_q$ are approximate eigenfunctions of the operator $\hat{H}$, the nondiagonal matrix elements $H_{mq}$ and $H_{qq'}$ are considerably smaller than the diagonal elements $\mathscr{E}_m$ and $\mathscr{E}_q$. We shall regard the diagonal elements as quantities of zero order of smallness, and the nondiagonal elements as quantities of first order. Then it follows from (11), (12) that $C_m$ are quantities of the zero order and $C_q$ are quantities of the first order. Having in mind the determination of $C_m$ from (11) up to terms of the second order inclusive, it is sufficient to retain in (12) terms of only the first order. Therefore, in (12) we have neglected the term $\sum_{q' \neq q} H_{qq'} C_{q'}$.

On substituting (10) into (12) and on taking into account the initial conditions for $C_q$, we obtain

$$C_q = (e^{-i\omega t} - e^{-i\omega_q t}) \sum_m H_{qm} A_m / (\hbar\omega - \mathscr{E}_q),$$
$$\omega_q = \mathscr{E}_q / \hbar. \tag{13}$$

On substituting (10) and (13) into (11) we obtain the system of equations for $A_m$:

$$(\mathscr{E}_m - \hbar\omega) A_m + \sum_{m'} H^a_{mm'} A_{m'} + W'_{m0} = 0, \tag{14}$$

where

$$H^a_{mm'} = \sum_q H_{mq} H_{qm'} \frac{1 - \exp\{-i(\omega_q - \omega)t\}}{\hbar\omega - \mathscr{E}_q}. \tag{15}$$

We transform expression (15) in the following way: we make use of the energy $\mathscr{E}_q$ as one of the quantum numbers of the state q, and we denote the set of the remaining quantum numbers by p. Let $\rho_p(\mathscr{E}_q)\,d\mathscr{E}_q$ be the number of states of the system whose energies are contained within the interval $d\mathscr{E}_q$, while the other quantum numbers are equal to p. Then

$$H^a_{mm'} = \sum_p \int H_{mq} H_{qm'} \times \frac{1 - \exp\{-i(\mathscr{E}_q/\hbar - \omega)t\}}{\hbar\omega - \mathscr{E}_q} \rho_p(\mathscr{E}_q)\, d\mathscr{E}_q. \tag{16}$$

Here the integral has no singularity at $\mathscr{E}_q = \hbar\omega$. If the quantity $(\mathscr{E}_q/\hbar - \omega)t$ is chosen as the new variable of integration then, for not very small values of t, the limits of integration with respect to the new variable can be replaced by $-\infty$ and $+\infty$. By going over from integration along the real axis to integration in the complex plane over the infinite semicircle lying in the lower half plane, it can be easily shown that the integral of the exponential term vanishes. Then, by returning to integration along the real axis, and by deforming the path of integration below the singular point, we obtain

$$H^a_{mm'} = \sum_p \Big\{ \mathrm{P} \int \frac{H_{mq} H_{qm'}}{\hbar\omega - \mathscr{E}_q} \rho_p(\mathscr{E}_q)\, d\mathscr{E}_q - i\pi\rho_p(\hbar\omega)\,[H_{mq} H_{qm'}]_{\mathscr{E}_q = \hbar\omega} \Big\}, \tag{17}$$

where P denotes the principal value of the integral. Thus, $H^a_{mm'}$ turns out not to depend on the time, and consequently $A_m$, defined by (14), also turn out to be constant.

In solving (14) in the zeroth approximation we omit the sums containing nondiagonal elements $H^a_{mm'}$. We then introduce these sums as small perturbations and obtain in the next approximation:

$$A_m = -\frac{1}{\mathscr{E}_m + H^a_{mm} - \hbar\omega} \Big[ W'_{m0} - \sum_{m'}{}' \frac{H^a_{mm'} W'_{m'0}}{\mathscr{E}_{m'} + H^a_{m'm'} - \hbar\omega} \Big] \tag{18}$$

(the prime on the summation sign denotes that the term $m' = m$ should be omitted).

The error in the values of $A_m$ obtained in this manner is of order of magnitude of $(H^a_{mm'})^2$, i.e., $(H_{mq})^4$.

The solutions (10) and (18) have been obtained for the case when in (3) we have taken in place of $W_{m0}$ only the first of the two terms in (9). It is now necessary to take the second term of (9) and to solve the system (3) with this term included. This solution can be immediately written down by analogy with the preceding one: we replace in (10) and (18) $\omega$ by $-\omega$, $\mathbf{k}$ by $-\mathbf{k}$, $A_0$ by $A_0^*$ and $W'_{m0}$ by $W''_{m0}$. The final expression for $C_m$ is given by the sum of the two solutions given above. If the values of $C_m$ obtained in this way are substituted into (7), the latter will give the desired specific polarization of the crystal.

In carrying out these calculations it is necessary to keep in mind the following: (1) $W'_{m0} \neq 0$ only if the quasi-momentum of the state m is $\mathbf{f}_m = \mathbf{k}$; $W''_{n0} \neq 0$, only if $\mathbf{f}_m = -\mathbf{k}$; (2) if $\Psi_m$ is an eigenfunction of $\hat{H}$ which corresponds to the quasi-momentum $\mathbf{f}_m = \mathbf{k}$, then $\Psi_n = \Psi_m^*$ is also an eigenfunction of $\hat{H}$ and corresponds to same energy $\mathscr{E}_n = \mathscr{E}_m$, but does not correspond to the quasi-momentum $\mathbf{f}_n = -\mathbf{k}$; (3) since $\Psi_0$ is assumed to be nondegenerate, it can be considered real. Therefore, in accordance with (7), $\mathbf{P}_n(\mathbf{r}) = \mathbf{P}_m^*(\mathbf{r})$, while in accordance with (9) $W''_{n0} = -W'^{*}_{m0}$; (4) in accordance with the law of conservation of quasi-momentum, $H_{mq} \neq 0$ only if the states m and q have the same quasi-momenta: $\mathbf{f}_m = \mathbf{f}_q$; violations of this rule are possible if the crystal contains defects (impurities); (5) if we introduce the notation

$$H^b_{mm'} = -\sum_q H_{mq} H_{qm'} / (\hbar\omega + \mathscr{E}_q), \tag{19}$$

it can be shown that $H^b_{nn'} = H^{b*}_{mm'}$.

By taking into account these remarks we can express the synchronous part of the specific polarization in the following way:

$$\mathbf{P}(\mathbf{r}) = \mathbf{P}_0 e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + \mathbf{P}_0^* e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)}, \quad \mathbf{P}_0 = \beta \mathbf{E}_0^{\mathrm{ext}}, \tag{20}$$

where $\mathbf{E}_0^{\mathrm{ext}}$ is the amplitude of the intensity of the external part of the electric field:

$$\mathbf{E}^{\mathrm{ext}} = -\frac{1}{c}\dot{\mathbf{A}} = \mathbf{E}_0^{\mathrm{ext}} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + \mathbf{E}_0^{*\mathrm{ext}} e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)},$$
$$\mathbf{E}_0^{\mathrm{ext}} = \frac{i\omega}{c}\mathbf{A}_0. \tag{21}$$

The complex tensor $\beta$ is expressed in the following way:

$$\beta = \sum_m [1/(\mathscr{E}_m + H^a_{mm} - \hbar\omega) - 1/(\mathscr{E}_m + H^b_{mm} + \hbar\omega)]\, a_{mm}/\omega$$
$$- \sum_{m,m'}{}' [H^a_{mm'}/(\mathscr{E}_m + H^a_{mm} - \hbar\omega)(\mathscr{E}_{m'} + H^a_{m'm'} - \hbar\omega)$$
$$- H^b_{mm'}/(\mathscr{E}_m + H^b_{mm} + \hbar\omega)$$
$$\times (\mathscr{E}_{m'} + H^b_{m'm'} + \hbar\omega)]\, a_{mm'}/\omega, \tag{22}$$

where $a_{mm'}$ is a tensor of the second rank in three-dimensional coordinate space. This tensor is determined by the relations

$$\mathbf{P}_m(0)\, W'_{m'0} \equiv -\frac{i}{c} a_{mm'} \mathbf{A}_0$$
$$\text{or} \quad (a_{mm'})_{xy} = \omega_{m'} V (\mathbf{P}_m(0))_x (\mathbf{P}^*_{m'}(0))_y. \tag{23}$$

In formula (22), and also everywhere in the following, the subscripts m and m′ denote only those states of the first group for which $\mathbf{f}_m = \mathbf{f}_{m'} = \mathbf{k}$.

In (22) the second term (containing the double summation) is a small correction to the first term. However, if for any two states m and m′ $\mathscr{E}_m = \mathscr{E}_{m'}$ then for $\hbar\omega \approx \mathscr{E}_m$ the corresponding term in the double sum ceases to be small in comparison with the first term. This difficulty can be avoided if in the choice of the basis states of the first group the functions $\Psi_m$ and $\Psi_{m'}$ are replaced by their linear combinations chosen in such a way that $H^a_{mm'} = 0$. Then the "dangerous" term disappears from the double sum.

It should be emphasized that, generally speaking, it is not possible to identify $\mathbf{E}^{\mathrm{ext}}$ with the total macroscopic field $\mathbf{E}$. As we have already explained earlier, the vector potential $\mathbf{A}$ describes the macroscopic field from which we have subtracted the field $\mathbf{E}'$ produced without retardation by the fictitious charges of the dielectric polarization. The field $\mathbf{E}'$ is the field of the Coulomb interaction of the parts of the system; it has already been included in $\hat{H}$ and should not appear a second time as a perturbation. Thus, the second of equations (20) can be re-written

$$\mathbf{P}_0 = \beta(\mathbf{E}_0 - \mathbf{E}_0'). \tag{24}$$

On the other hand, as is well known, the field of the fictitious charges of the dielectric polarization can be expressed in the following way:

$$\mathbf{E}_0' = -4\pi(\mathbf{P}_0\cdot\mathbf{s})\,\mathbf{s}, \tag{25}$$

where $\mathbf{s} = \mathbf{k}/k$ is the unit vector normal to the wave front. By eliminating $\mathbf{E}_0'$ from (24) and (25) we obtain the usual expression for the specific polarization in terms of the total macroscopic electric field:

$$\mathbf{P}_0 = \varkappa \mathbf{E}_0, \quad \varkappa = (1 - 4\pi\beta\zeta)^{-1}\beta. \tag{26}$$

Here $\zeta = \overline{\mathbf{ss}}$ is a tensor in three-dimensional coordinate space; its components have the form: $\zeta_{xy} = s_x s_y$. Such a tensor is called a dyad; we shall denote the dyadic product by a bar over both letters.

If into the expression (26) for $\kappa$ we substitute $\beta$ from (22), and if we expand the resulting function into partial fractions of the form $C_i/(K_i - \hbar\omega)$, then $\kappa$ assumes its traditional form:

$$\varkappa = \sum_i C_i/(K_i - \hbar\omega). \tag{27}$$

But the parameters $K_i$ occurring in the dispersion formula, generally speaking, do not coincide with the energy levels of the system $\mathscr{E}_m$ even when the small quantities $H^a_{mm'}$ and $H^b_{mm'}$ are neglected. The relation between $K_i$ and $\mathscr{E}_m$ can be obtained if we actually carry out the expansion mentioned above of $(1 - 4\pi\beta\zeta)^{-1}\beta$ into partial fractions, which is possible in practice only in the simplest cases (cf. the next section of this paper). For a special form of excitons — polarization oscillations of the ionic lattice — such a relationship has been established by the author jointly with Krivoglaz.[3]

In concluding this section we generalize the results obtained above to the case of a field which has an arbitrary spatial variation, but which, as before, oscillates in time harmonically at a frequency $\omega$:

$$\mathbf{E} = \mathbf{f}(\mathbf{r})\, e^{-i\omega t} + \mathbf{f}^*(\mathbf{r})\, e^{i\omega t},$$
$$\mathbf{P} = \mathbf{g}(\mathbf{r})\, e^{-i\omega t} + \mathbf{g}^*(\mathbf{r})\, e^{i\omega t}. \tag{28}$$

To do this we expand the field and the polarization into Fourier series of the form

$$\mathbf{f}(\mathbf{r}) = \sum_{\mathbf{k}} \mathbf{E}_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}, \quad \mathbf{g}(\mathbf{r}) = \sum_{\mathbf{k}} \mathbf{P}_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}},$$
$$\text{where } \mathbf{P}_{\mathbf{k}} = \varkappa(\mathbf{k})\,\mathbf{E}_{\mathbf{k}}. \tag{29}$$

Here $\kappa(\mathbf{k})$ is defined by formulas (26) and (22). We assume that $\kappa(\mathbf{k})$ is an analytic function. Then $\kappa(\mathbf{k})\, e^{i\mathbf{k}\cdot\mathbf{r}} = \hat{\kappa}(-i\nabla)\, e^{i\mathbf{k}\cdot\mathbf{r}}$. Consequently, we obtain from (29)

$$\mathbf{g}(\mathbf{r}) = \hat{\varkappa}(-i\nabla)\, \mathbf{f}(\mathbf{r}). \tag{30}$$

It was assumed earlier that the propagation vector of the perturbing light $\mathbf{k}$ in formula (1) is real. In the case when it is complex (damped wave) it may be easily shown by using the general formula (30) that all the results obtained above remain valid, only the complex value of $\mathbf{k}$ should be substituted into them.

Formulas (22), (24), and (26) in principle completely determine the dispersion and the absorption of an electromagnetic wave in a crystal.

## 2. LIMITING TRANSITION TO MACROSCOPIC ELECTRODYNAMICS

In this section we consider the case of an isolated absorption band, i.e., the case when in the single sum of expression (22) one term predominates over all the others. This may occur either if the coefficient $a_{mm}$ in one of the terms is considerably larger than in the others, or if the value of $\omega$ is such that the denominator of one of the terms is close to zero.

It is evident that this is possible only if in (22) $\sum_m$ is a discrete sum and not an integral, i.e., if $\mathbf{f}_m = \mathbf{k}$ is the only continuous quantum number that determines the energy $\mathcal{E}_m$; all the other quantum numbers in the expression for $\mathcal{E}_m$ must be discrete. Such states were denoted in reference 1 as exciton states.

By retaining only the dominant term in (22) we obtain

$$\beta = \frac{Q\overline{\mathbf{x}\mathbf{x}^*}}{\mathcal{E} + H^a - \hbar\omega}, \qquad Q \equiv \frac{\omega_m}{\omega} V\, |\mathbf{P}_m(0)|^2,$$
$$\mathbf{x} \equiv \frac{\mathbf{P}_m(0)}{|\mathbf{P}_m(0)|} \tag{31}$$

(for the sake of brevity we have omitted the subscript m). If we assume the exciton lifetime to be infinite, i.e., if we neglect the small term $H^a$, (31) coincides with the principal result of reference.[1]

The limiting transition to macroscopic electrodynamics reduces to the assumption that the electric field varies very smoothly in space, i.e., $k \to 0$. It is necessary to require that in this case the magnitude of $\kappa$ defined by formula (26) should not depend on the direction of $\mathbf{s}$, since the macroscopic specific polarizability tensor should not depend on the direction of incidence of the wave.

It is well known that an arbitrary function of a product of dyads has the following property:

$$F\,(\overline{\mathbf{x}\mathbf{x}^*}\ \overline{\mathbf{s}\mathbf{s}})\,\overline{\mathbf{x}\mathbf{x}^*} = F\,(\cos^2\alpha)\,\overline{\mathbf{x}\mathbf{x}^*}, \tag{32}$$

where $\alpha$ is the angle between $\mathbf{x}$ and $\mathbf{s}$. Therefore substitution of (31) into the second of formulas (26) leads to the following result:

$$\varkappa = \left(1 - \frac{4\pi Q\cos^2\alpha}{\mathcal{E} + H^a - \hbar\omega}\right)^{-1} \frac{Q\overline{\mathbf{x}\mathbf{x}^*}}{\mathcal{E} + H^a - \hbar\omega}$$
$$= \frac{Q\overline{\mathbf{x}\mathbf{x}^*}}{\mathcal{E} - 4\pi Q\cos^2\alpha + H^a - \hbar\omega}. \tag{33}$$

By identifying this result with expression (27) and by assuming that $\omega_m/\omega \approx 1$, and therefore by neglecting the dependence of Q on $\omega$ we obtain

$$K = \mathcal{E} - 4\pi Q\cos^2\alpha + H^a, \quad C = Q\overline{\mathbf{x}\mathbf{x}^*}. \tag{34}$$

As we have already explained previously, K should not depend on the direction of $\mathbf{s}$, i.e., on $\alpha$. Then it follows from formula (34) that

$$\mathcal{E} = \mathcal{E}_0 + 4\pi Q\cos^2\alpha. \tag{35}$$

Here $\mathcal{E}_0$ does not depend on the direction of $\mathbf{s}$. Strictly epeaking, $\mathcal{E}$ takes on the meaning of exciton energy only if its lifetime is infinite, i.e., when $H^a = 0$. Formula (35) shows that when $\mathbf{f} = \mathbf{k} \to 0$ the energy of the exciton depends on the direction of $\mathbf{f}$, since $\alpha$ is the angle between $\mathbf{f}$ and $\mathbf{x}$. Thus, regarded as a function of the three variables $f_x$, $f_y$, $f_z$, $\beta$ (but not $\kappa$) has a discontinuity at the point $\mathbf{f} = 0$. This, of course, does not exclude the possibility that, for a given direction of $\mathbf{f}$, $\mathcal{E}$ is an analytic function in $|\mathbf{f}|$. The conclusion with respect to the discontinuity in $\mathcal{E}(\mathbf{f})$ follows only if $Q \neq 0$, i.e., $\mathbf{P}_m(0) \neq 0$. Phototransitions are allowed specifically into such states m.

We have already derived Eq. (35) earlier[4] by a completely independent method (by means of a direct calculation of the exciton energy bearing no relation to the theory of dispersion and absorption). In the same paper we have considered cases when in virtue of a definite crystal symmetry the excited state of an elementary crystal cell is degenerate. For such cases we had obtained a generalization of formula (35).

We can now rewrite formula (27) in the form

$$\varkappa = C / (\mathcal{E}_0 + H^a - \hbar\omega), \tag{36}$$

where in $H^a$ we can neglect the first real term of the two terms of expression (17), since it is small in comparison with the other real terms of the denominator. The second term of expression (17) must be retained since it determines the imaginary part of $\kappa$. Formula (36) shows that the region of light absorption, i.e., the region where the real part of $\kappa$ is important is determined by the relation $\hbar\omega \simeq \mathscr{E}_0$, and not by the usual Bohr frequency formula $\hbar\omega \approx \mathscr{E}$. This is not surprising, since in the preceding section of this article in formulating the initial conditions for $C_m$ and $C_q$ it was explained that the exciton state m under discussion, being a state of the first group, is only an intermediate state in the process of photoexcitation of states of the second group q. Formula (13) shows that only those coefficients $C_q$ will increase monotonically with time which correspond to states of energy $\mathscr{E}_q = \hbar\omega$. Thus the Bohr frequency formula is satisfied for phototransitions into states of the second group which give rise to absorption.

From what has been said above it follows that an experimental investigation of the frequency $\omega$ for which the absorption is a maximum allows us to determine, by means of the relation $\hbar\omega = \mathscr{E}_0$, the magnitude of $\mathscr{E}_0$ which, generally speaking, in less than the exciton energy $\mathscr{E}$, since Q is positive in (35). $\mathscr{E}_0$ coincides with the exciton energy only if **s** is normal to **x**.

## 3. PLANE ELECTROMAGNETIC WAVE. DETERMINATION OF THE INDICES OF REFRACTION

Turning from the case of vanishingly small values of **k** to the case of non-zero finite values of **k**, it should be emphasized that the tensor $\kappa$ will now depend both on the absolute value and on the direction of the vector **k**. Whereas in the limiting case $\mathbf{k} \to 0$, the description of the polarization by means of the tensor $\kappa$ was simpler than by means of the tensor $\beta$, since $\kappa$ did not depend on **k** at all, while $\beta$ depended on the direction of **k**, now this technical advantage of $\kappa$ disappears. Now it seems to be more appropriate to describe the polarization by means of the tensor $\beta$, since the latter, in accordance with (31), is expressed in terms of the exciton energy $\mathscr{E}(\mathbf{f})$ — a quantity that determines many other phenomena in the crystal. One could try to determine $\kappa$ without introducing the Hamiltonian for the system; the dependence of $\kappa$ on **k** for the case of small **k** can be phenomenologically given in the form of an expansion (polynomial) in $k_x$, $k_y$, $k_z$, as has been done by Ginzburg.[5] But in this case the expansion coefficients remain as unknown parameters in the theory; it is not clear how they could be calculated or related to other physical phenomena. It is particularly inconvenient that the dependence of $\kappa$ on $\omega$ remains completely undetermined. In the present paper we do therefore not make use of the phenomenological method referred to above, but retain the quantum-mechanical discussion utilized in our original papers.[1,2]

We assume that there are no external charges introduced into the crystal which would give rise to an external irrotational field; the only contribution to the irrotational part of the field is made by the field of the fictitious charges of dielectric polarization **E′**. Then the whole external field $\mathbf{E}^{\mathrm{ext}}$ can be obtained as the rotational part of the total macroscopic field **E**. In the plane-wave case under consideration this corresponds to the following relation between the amplitudes of the fields:

$$\mathbf{E}_0^{\mathrm{ext}} = \eta\mathbf{E}_0, \text{ where } \eta_{xy} = \delta_{xy} - s_x s_y. \tag{37}$$

In the first section of this article $\mathbf{E}^{\mathrm{ext}}$ was introduced as a given external perturbation. The wave vector **k** appeared in it as a given parameter of the perturbing field. It is now necessary to make this perturbing field self-consistent, i.e., to ensure by means of a suitable choice of **k** that this field satisfies the system of Maxwell's equations. If we assume, as usual, that the electric field, the displacement, and the magnetic field are proportional to $e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}$, where $\mathbf{k} = (\omega n/c)\mathbf{s}$, while n is the index of refraction for the wave, then the system of Maxwell's equations reduces to the following relations:

$$\mathbf{D}_0 = n^2[\mathbf{E}_0 - \mathbf{s}(\mathbf{s}\cdot\mathbf{E}_0)] \equiv n^2\eta\mathbf{E}_0,$$
$$\mathbf{D}_0 = \gamma\mathbf{E}_0, \quad \gamma = 1 + 4\pi\beta\eta. \tag{38}$$

One should not identify $\gamma$ with the usual dielectric constant, since it depends not only on $\omega$ but also on the direction of wave propagation **s**. Moreover, $\beta$ depends on $\mathbf{f} = \mathbf{k} = (\omega n/c)\mathbf{s}$. Thus $\gamma$ is a function of the index of refraction n. In this respect Eq. (22) significantly differs from the usual dispersion formula.

If we eliminate $\mathbf{D}_0$ from the system (38) we obtain a system of linear homogeneous equations for the components of $\mathbf{E}_0$. The condition that it have a solution has exactly the same form as in reference 1:

$$(\mathbf{s}, \gamma\mathbf{s})\,n^4 + [(\mathbf{s}, \gamma^2\mathbf{s}) - (\mathbf{s}, \gamma\mathbf{s})\,\mathrm{Sp}\,\gamma]\,n^2 + \begin{vmatrix} \gamma_{xx} & \gamma_{xy} & \gamma_{xz} \\ \gamma_{yx} & \gamma_{yy} & \gamma_{yz} \\ \gamma_{zx} & \gamma_{zy} & \gamma_{zz} \end{vmatrix} = 0. \tag{39}$$

In the usual crystal optics the corresponding equation is a quadratic in $n^2$; it determines two values of $n^2$ (double refraction). But in the theory under discussion $\gamma$ is itself a function of n. Therefore (39) is an equation of higher order with respect to n, it determines not two but more values of $n^2$, and the same number of solutions of Maxwell's equations having the form of a plane wave.

We further consider the case when $\hbar\omega$ approaches one of the exciton energy levels $\mathscr{E}_m = \mathscr{E}_0$. Then we can pick out from (22) the term which "resonates," i.e., which has the small denominator $\mathscr{E}_m + H^a_{mm} - \hbar\omega$. This term varies rapidly with $\omega$. We denote the sum of all the remaining terms by $\beta'$; it is a slowly varying function of $\omega$ and in the region $\hbar\omega \approx \mathscr{E}_m$ can be approximately regarded as a constant. $\beta'$, generally speaking, is complex, and its imaginary part is a quantity of the second order of smallness. Formula (22) can now be simplified:

$$\beta = \beta' + (Q\overline{\mathbf{xx}^*} + \alpha)/(\mathscr{E}(\mathbf{k}) + H^a - \hbar\omega),$$

$$\alpha = \sum_{m'}{}' (H^a_{mm'} a_{mm'} + H^a_{m'm} a_{m'm})/\omega(\mathscr{E}_{m'} - \mathscr{E}_m). \tag{40}$$

Here the summation over $m'$ is taken over all the states except the "resonating" m-th state. Of course, $\beta'$ and $\alpha$ depend on the direction of wave propagation $\mathbf{s}$.

We assume that for a given direction $\mathbf{k}$ the expansion of $\mathscr{E}(\mathrm{k})$ in powers of k is of the following form:

$$\mathscr{E}(k) = \mathscr{E}_s + \hbar^2 k^2/2M_s + \dots \tag{41}$$

Here $\mathscr{E}_{\mathbf{s}}$ and $M_{\mathbf{s}}$ do not depend on k, but depend on the direction of $\mathbf{k}$, i.e., on $\mathbf{s}$. In the case of an isolated exciton absorption band the dependence of $\mathscr{E}_{\mathbf{s}}$ on $\mathbf{s}$ is given by formula (35). We shall call $M_{\mathbf{s}}$ the effective mass of the exciton in the direction $\mathbf{s}$.

If we introduce the notation

$$\gamma' = 1 + 4\pi\beta'\eta, \quad \mu_s = \frac{2M_s c^2}{\hbar\omega}\left(1 - \frac{\mathscr{E}_s + H^a}{\hbar\omega}\right),$$

$$B_s = \frac{8\pi M_s c^2}{\hbar^2\omega^2}[Q\overline{\mathbf{xx}^*} + \alpha], \tag{42}$$

then the result of substituting (31) into the expression (38) for $\gamma$ can be rewritten

$$\gamma = \gamma' + B_s\eta/(n^2 - \mu_s). \tag{43}$$

This formula differs from the corresponding formula of reference 1 only by the fact that the expression for $\mu_{\mathbf{s}}$ has acquired in addition to $\mathscr{E}_{\mathbf{s}}$ the complex term $H^a$, while the expression for $B_{\mathbf{s}}$ has acquired the complex additional term $\alpha$.

If we assume that the exciton lifetime is infinite ($H^a = 0$, $\alpha = 0$), then (42) and (43) reduce exactly to the corresponding equations of reference 1, and yield all the subsequent results of references 1 and 2, subject to the restrictions that follow from reference 4 (cf. also the end of this paper). As has been explained in references 1 and 2, in the case of an infinite exciton lifetime dispersion of light will occur, but there will be no absorption. Thus, absorption appears as a result of taking into account the finite exciton lifetime, which leads to the appearance of additional complex terms $H^a$ and $\alpha$ in the quantities $\mu_{\mathbf{s}}$ and $B_{\mathbf{s}}$. As a result, n also acquires an additional complex term, as can be seen from the expressions for $n^2$, given in references 1 and 2. The formulas derived in references 1 and 2, which express $n^2$ in terms of $\mu_{\mathbf{s}}$ and $B_{\mathbf{s}}$, remain valid, but since $\mu_{\mathbf{s}}$ and $B_{\mathbf{s}}$ become complex these formulas begin to describe the case of an absorbing crystal.

The imaginary part of n determines, in a well known way, the damping of a monochromatic wave in space. But, as has been shown in reference 1, several plane waves arise simultaneously in a crystal, each with its own index of refraction and damping coefficient. These waves are coherent and therefore cannot be investigated independently of each other. The total intensity of light is spatially damped nonexponentially. Therefore, strictly speaking, the phenomenological concept of the coefficient of light absorption (the coefficient in the exponential expression for the spatial damping of light intensity) loses its meaning. But the quantities actually measured by experimenters in investigations of light absorption in crystals completely retain their meaning. These include, for example, the reduction in the intensity of light in passing through a plane parallel plate. This can be easily calculated with the aid of the formulas of Sec. 3 of reference 2, provided we introduce the already-mentioned complex terms added to the indices of refraction in the case of a finite exciton lifetime. This will be done in a later paper. In another paper we shall also obtain expressions for $H^a$ and $\alpha$ for specific exciton models.

## 4. LONGITUDINAL WAVES

Let us investigate the solution of the time-dependent Schrödinger equation for a crystal in the absence of an external perturbing field, i.e., when $\mathbf{A}(\mathbf{r}, t) = 0$. The problem reduces to the solution of the system of equations (11), (12) with $W'_{m0} = 0$. We shall, as before, seek the solutions for $C_m$ in harmonic form (10), but for the coefficients $A_m$ we shall obtain in place of (14) the following system of equations:

$$[\mathscr{E}_m(\mathrm{f}) - \hbar\omega]A_m + \sum_{m'} H^a_{mm'}(\mathrm{f})A_{m'} = 0. \tag{44}$$

By setting the determinant of this system equal to zero we obtain the values of $\omega(\mathbf{f})$, i.e., the frequencies of the characteristic polarization oscillations of the crystal. By finding, for each root $\omega(\mathbf{f})$ in (44), the coefficients $A_m$ and substituting them into (10), we obtain the corresponding $C_m$. If these $C_m$ are substituted into (7), and if (8) is taken into account, the following expression is obtained for the polarization eigenwave in the crystal:

$$\mathbf{P}(\mathbf{r}, t) = \mathbf{P}_0 e^{i(\mathbf{f}\cdot\mathbf{r}-\omega t)} + \mathbf{P}_0^* e^{-i(\mathbf{f}\cdot\mathbf{r}-\omega t)};$$

$$\mathbf{P}_0 = \sum_m A_m \mathbf{P}_m(0), \quad \omega = \omega(\mathbf{f}). \qquad (45)$$

This result is obtained on the basis of Schrödinger's equation, but without taking into account Maxwell's equations. Therefore, it is now necessary to determine the additional limitations imposed by the latter.

It is clear that if the vector $\mathbf{P}_0$ is not parallel to $\mathbf{f}$, the polarization wave (45) will give rise to a magnetic field and, consequently we shall have $\mathbf{A}(\mathbf{r}, t) \neq 0$. This contradicts the initial assumption of this section of the present article, violates the result (45), and brings us back to the case discussed in the preceding sections. But if the vector $\mathbf{P}_0$ is parallel to $\mathbf{f}$, then in this case, according to Maxwell's equations, the polarization wave (45) will give rise only to an irrotational electric field $\mathbf{E}'$ which is produced without retardation by the fictitious charges of the dielectric polarization, and which is equal to

$$\mathbf{E}' = -4\pi\mathbf{P}. \qquad (46)$$

Maxwell's equations will all be satisfied if, in addition to (46), we set

$$\mathbf{D} = \mathbf{H} = \mathbf{E}_\perp = \mathbf{A} = 0, \qquad (47)$$

Thus, the solution (45) — (47) satisfies both the Schrödinger equation and Maxwell's equations. It represents a plane longitudinal wave of the electric field $\mathbf{E}'$ and of the polarization. In this case even though $\mathbf{E}'$ is a macroscopic field, it nevertheless represents the Coulomb interaction of the charges of which the crystal is composed and which, consequently, appears in $\hat{H}$, and not in the perturbing operator $\hat{W}$.

The condition $\mathbf{P}_0 \parallel \mathbf{f}$ determines those directions of $\mathbf{f}$ for which longitudinal waves occur. The number of such direction increases as the symmetry of the crystal becomes higher. It can be shown that, say in a cubic crystal, longitudinal waves can be propagated in an arbitrary direction (it is assumed that the wavelength is considerably greater than the lattice constant).

In the limiting case of long waves it is possible to introduce the concept of the usual macroscopic dielectric constant. In the case of longitudinal waves the latter is equal to zero, since $\mathbf{D} = 0$, while $\mathbf{E} \neq 0$.

It is not difficult to understand why (45) cannot be obtained as one of the solutions of equations (39) and (43). The point is that (39) was obtained as the condition for the existence of such solutions of equations (38) for which $\mathbf{D}$ and $\mathbf{E}_\perp$ differ from zero. But in the case of a longitudinal wave we have just the case $\mathbf{D} = 0$, $\mathbf{E}_\perp = 0$. Therefore the index of refraction of the longitudinal wave does not have to satisfy Eq. (39).

In the case when the system of equations (44) reduces to only one equation (nondiagonal elements of the matrix $H^a_{mm'}$ are neglected and an isolated excited level $\mathscr{E}_m = \mathscr{E}(\mathbf{f})$ is assumed), of the form

$$[\mathscr{E}(\mathbf{f}) + H^a(\mathbf{f}) - \hbar\omega] A_m = 0,$$

we obtain

$$A_m \neq 0, \quad \text{if} \quad \hbar\omega = \mathscr{E}(\mathbf{f}) + H^a(\mathbf{f}). \qquad (48)$$

Since $H^a$ is complex (48) means that, for real $\mathbf{f}$, $\omega$ is complex. This means that the polarization oscillation will be undamped in space but damped in time; its energy will be converted into the energy of excitation of states q of the second group. But it is possible, on assuming a real value of $\omega$, to obtain a complex $\mathbf{f}$ from (48). We then obtain a polarization oscillation which is stationary in time, but damped in space. Such an oscillation may exist, for example, on the surface of a crystal and be damped out with penetration into the crystal; it is maintained in the steady state by an electromagnetic wave that enters the crystal from the vacuum.

In the case of the limiting transition to the infinite lifetime of the excited state, when $H^a \to 0$, (48) reduces to the corresponding result of reference 1.

In conclusion we note a certain omission made in reference 1. In that paper we have uncritically followed other authors by assuming that the exciton energy $\mathscr{E}(\mathbf{k})$ is an analytic function of the components of the quasi-momentum $k_x$, $k_y$, $k_z$, and we expanded $\mathscr{E}(\mathbf{k})$ in powers of these three arguments. But in our subsequent paper[4] it is shown that the interaction of crystal cells situated far from each other, an interaction due to the irrotational part of the electric field accompanying the exciton wave, gives a finite contribution to the exciton energy $\mathscr{E}(\mathbf{k})$. This additional term is a discontinuous function of $k_x$, $k_y$, $k_z$: its value at $\mathbf{k} = 0$ depends on the direction of the vector $\mathbf{k}$, when its modulus tends to zero. Therefore, the expansion used in reference 1 is not justified. In this paper we use, in place of the foregoing expansion, an expansion in terms of $|\mathbf{k}|$ [Eq. (41)] and it is emphasized that $\mathscr{E}_{\mathbf{s}}$ and $M_{\mathbf{s}}$ depend on the direction of propagation of the exciton wave **s** in accordance with reference 4. Moreover, we obtain all the basic results of reference 1. But certain specific assertions of reference 1 must be corrected. Thus, it is shown in reference 4 that in cubic crystals the magnitude of $\mathscr{E}_{\mathbf{s}}$ is different for excitons with longitudinal and transverse polarization. $M_{\mathbf{s}}$ may also differ for excitons of these types. The energy bands of excitons with transverse polarization must overlap in pairs (we are referring to exciton states into which phototransitions from the ground state of the crystal are allowed). For each pair of overlapping bands the magnitudes of $\mathscr{E}_{\mathbf{s}}$ must coincide, and must not depend on the direction of **s**, while the magnitudes of $M_{\mathbf{s}}$ must coincide for two bands only in the directions of the edges and the space diagonals of the basic cube.

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