These estimates are made under the assumption that $\mathbf{k}_1$, $\mathbf{k}_2$, $\mathbf{k}_3$, $\mathbf{k}_4$ are of the same order of magnitude. We note that in the expression (5.4), the chief contribution is made by the region of integration $k' \sim L^{-1}$, and in (5.5), by the region $k' \sim k_1$.

The turbulence spectrum is determined from Eq. (2.14) for $l_{\mathbf{k},\omega}$. The increment to the vertex $\delta T^{(1)}_{12|34} \approx M^2 T_{12|34}$ has the same index of homogeneity as the vertex $T_{12|34}$ for the incompressible liquid. Therefore, it does not lead to a change in the Kolmogorov exponent. The increment

$$\delta T^{(2)}_{12|34} \approx T_{12|34} M^2 (kL)^{-4/3}$$

leads to the appearance in the equation for $l_{\mathbf{k}\omega}$ of additional terms with the parameter of smallness $M^2(kL)^{2/3}$. They should be cancelled by the terms which arise because of the correction (5.1) to the spectrum J_k. Thus $f(\mathbf{k}) \approx (kL)^{-2/3}$,

$$J_k = J_k^0 + \delta J_k, \quad \delta J_k \approx J_k^0 M^2/(kL)^{1/3}. \tag{5.6}$$

This result has a simple meaning; in correspondence with the Kolmogorov hypothesis that the spectrum interaction is local, the value of J_k in the inertial interval of the scales ($kL > 1$) cannot depend on the velocity of motion v_T in the scale L, i.e., on $M^2 = v_T^2/c_s^2$. Only the fluctuations of density due to motions of the liquid $v(k)$ of the same scale $1/k$ are important. Taking it into account that in the inertial interval $v(k) \approx v_T (kL)^{-1/2}$, we obtain

$$\frac{\delta\rho_T(k)}{\rho_0} \approx \frac{v^2(k)}{c_s^2} \approx M^2 (kL)^{-2/3}.$$

It can be shown that the relative correction to the spectrum ($\delta J_k / J_k$) is of the order of the square of the Mach number calculated from the circular velocity of motion of vortices of scale $1/k$.

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Auxiliary boundary conditions in the theory of additional light waves and excitons in bounded crystals

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Frenkel excitons [Phys. Rev. **37**, 17, 1276 (1931)] and corresponding optical excitons (optical waves, polaritons) are considered for a semiinfinite crystal of any symmetry but possessing an inversion center. It is shown that the solutions for a semiinfinite crystal can be constructed as linear combinations of solutions for an infinite crystal only in the case of normal incidence or for selected wave polarizations, and if the interactions of close (A) or distant (B) cells in a crystal predominate. The coefficients of such linear combinations are found from auxiliary boundary conditions whose justification is the main task of the present paper. It is shown that these auxiliary boundary conditions, proved by the present author for the case of degenerate excitons and restricted crystal symmetries [S. I. Pekar, Sov. Phys. JETP **6**, 785, (1958)], are always valid in case A, but in case B they apply only for normal incidence and certain polarizations of light. Auxiliary boundary conditions of a new type are obtained for all other cases and for dipole-forbidden radiative transitions. The results obtained agree, in the appropriate special cases, with microcalculations based on the simplest model of a crystal.

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1. INTRODUCTION

According to the conventional theory of birefringence, two orthogonal waves of the same frequency and with the same direction of propagation may travel in a crystal. Their amplitudes can be described uniquely in terms of the amplitude of a wave incident on a crystal from vacuum, using the well-known conditions of continuity of the tangential projections of an electron and magnetic fields. However, in the vicinity of the ex-

citon absorption band, there may exist additional optical waves in a crystal, i.e., there may be more than two linearly independent solutions of the Maxwell equations. In this case, the above continuity conditions are insufficient and one has to impose auxiliary boundary conditions. Clearly, this does not imply any new assumptions because the reaction of a crystal to an optical perturbation is described completely by the Schrödinger equation.[1)] All that is necessary is to use the Schrödinger wave function for a bounded crystal. However, such a crystal is a spatially inhomogeneous system: it does not have the translational and point symmetries of an infinite crystal, and its wave functions are not characterized by three-dimensional quasimomentum, so that we are not allowed to introduce a coordinate-independent permittivity, assume that the exciton quasimomentum is equal to the wave vector of light, etc. Therefore, it is technically simpler to use the solutions for an infinite crystal as a basis and then, at the end of the calculations, obtain with the aid of auxiliary boundary conditions those linear combinations of solutions which correspond to a bounded crystal.

It has been possible to develop a theory of additional light waves in an infinite crystal without specifying excitons but defining them as "excitations in an ideal insulator characterized by single continuous quantum number, which is the quasimomentum $\mathbf{k}$ (all the other quantum numbers are discrete)."[1] However, as pointed out in that theory,[1] the auxiliary boundary conditions may vary with the exciton model. The conditions obtained in the author's theory[1] for Frenkel excitons were subject to the following restrictions: a) the excitation in a unit cell of a crystal was assumed to be nondegenerate; b) an exciton radiative transition was assumed to be allowed in the dopole approximation; c) the surface of a crystal coincided with one of its mirror symmetry planes; d) the exciton theory allowed only for the interaction between neighboring unit cells in a crystal because the importance of the interaction between the more distant cells became apparent later.[2]

In this theory, the author also calculated the reflection coefficient of light of the surface of a crystal and the transparency coefficient of a crystal boundary.

In the present paper, we shall impose auxiliary boundary conditions also in the case of Frenkel excitons but without all the above restrictions. We shall assume that the crystal has an inversion center. It must be stressed that the auxiliary boundary conditions follow uniquely from the type of Schrödinger wave function of an exciton in a bounded crystal (see below). Like this function, the conditions are approximate. The following reasoning shows that is it impossible to bypass this purely mechanical problem: away from an exciton resonance in the direction of red or violet frequencies or in the limit $c \to \infty$ (Ref. 3), some of the optical waves (optical excitons) are known to transform into Schrödinger excitons and the theory of their reflection from the surface of a crystal simply reduces to the theory of exciton reflection. Thus, the purely mechanical problem mentioned above has to be considered even in asymptotically limiting cases, and this applies even more to the general case.

2. PRINCIPAL DEFINITIONS

Let $\mathbf{a}_1$, $\mathbf{a}_2$, and $\mathbf{a}_3$ denote the lattice periods, where integral vectors $\mathbf{n}=n_1\mathbf{a}_1+n_2\mathbf{a}_2+n_3\mathbf{a}_3$ give the positions of the centers of unit cells; $\psi_{\mathbf{n}}$ is the ground state of a cell and $\varphi_{\mathbf{n}\nu}$ represents excited states of the cell; ν labels the degenerate states ($\nu=1,2,\ldots,s$);

$$\Phi_{\mathbf{m}\mu}=S\varphi_{\mathbf{m}\mu}\prod_{\substack{\mathbf{n}\\ \mathbf{n}\neq\mathbf{m}}}'\psi_{\mathbf{n}}$$

is the state of the main periodic part of a crystal which contains an excited cell $\mathbf{m}$; S is the operation of antisymmetrization of a function with respect to the electron coordinate.

The wave function of an exciton is

$$\Psi=\sum_{\mathbf{m}\mu}a_{\mathbf{m}\mu}\Phi_{\mathbf{m}\mu}. \tag{1}$$

The coefficients $a_{\mathbf{m}\mu}$ are found from the system of equations

$$\sum_{\mathbf{n}\nu}H_{\mathbf{m}\mu,\mathbf{n}\nu}a_{\mathbf{n}\nu}=\mathscr{E}a_{\mathbf{m}\mu}, \tag{2}$$

where $H_{\mathbf{m}\mu,\mathbf{n}\nu}\equiv H_{\mu\nu,\mathbf{n}-\mathbf{m}}$ represents the familiar matrix elements;[4] $\mathscr{E}$ is the exciton energy. For an unbounded crystal, the quantities m_1, m_2, m_3, n_1, n_2, and n_3 can have any integral values from $-\infty$ to ∞.

The translational symmetry of the system (1) suggests the type of solution:

$$a_{\mathbf{n}\nu}=a_{0\nu}e^{i\mathbf{k}\mathbf{n}}, \tag{3}$$

where $\mathbf{k}$ is the exciton quasimomentum. Substituting Eq. (3) in Eq. (2), we obtain

$$\sum_{\nu=1}^{s}H_{\mu\nu}(\mathbf{k})a_{0\nu}=\mathscr{E}a_{0\mu};\quad H_{\mu\nu}(\mathbf{k})=\sum_{\mathbf{n}'}H_{\mu\nu\mathbf{n}'}e^{i\mathbf{k}\mathbf{n}'},\quad \mathbf{n}'=\mathbf{n}-\mathbf{m}. \tag{4}$$

From this system of equations and the normalization condition of the function (1), we find the quantities $\mathscr{E}_l(\mathbf{k})$ and $a_{0\nu}^{(l)}(\mathbf{k})$ (l is the number of the solution, $l=1,2,\ldots,s$). We also find that

$$\sum_{\nu=1}^{s}a_{0\nu}^{(l)*}(\mathbf{k})a_{0\nu}^{(l')}(\mathbf{k})=\frac{1}{G^3}\delta_{ll'}, \tag{5}$$

where G^3 is the number of cells in the main part of the crystal considered.

Allowing for the overlap of the wave functions and the exchange interaction between the nearest neighbors alone, i.e., assuming $\mathbf{n}-\mathbf{m}=\pm\mathbf{a}_i$ ($i=1,2,3$), we can regard the potential energy of the interaction between the more distant cells as of the dipole–dipole type, given by

$$F(\mathbf{p}_{\mathbf{m}}',\mathbf{p}_{\mathbf{n}}',\mathbf{n}-\mathbf{m})=\frac{(\mathbf{p}_{\mathbf{m}}',\mathbf{p}_{\mathbf{n}}')}{|\mathbf{n}-\mathbf{m}|^3}-3\frac{(\mathbf{p}_{\mathbf{m}}',\mathbf{n}-\mathbf{m})(\mathbf{p}_{\mathbf{n}}',\mathbf{n}-\mathbf{m})}{|\mathbf{n}-\mathbf{m}|^5}, \tag{6}$$

where $\mathbf{p}_{\mathbf{n}}'$ is the dipole moment of a cell $\mathbf{n}$, expressed in

terms of the coordinates of the particles in the cell. Introducing $\mathbf{p}_\nu = \langle \psi_{\mathbf{n}} | \mathbf{p}'_{\mathbf{n}} | \varphi_{\mathbf{n}\nu} \rangle$, we can show[2] that

$$H_{\mu\nu\mathbf{n}'} = F(\mathbf{p}_\mu^*, \mathbf{p}_\nu, \mathbf{n}') + \mathcal{H}_{\mu\nu\mathbf{n}'}, \quad \mathbf{n}' \neq 0, \tag{7}$$

where $\mathcal{H}_{\mu\nu\mathbf{n}'} \neq 0$ only for the neighboring cells (for $\mathbf{n}' = \pm\mathbf{a}_i$) and, in these cases, this last quantity supplements the right-hand side of Eq. (7) so that the correct value is obtained.[2)] More rigorously, we have to ensure that the terms (48) supplement the expression in the braces in Eq. (47) until the correct value is obtained.

The substitution of Eq. (7) into Eq. (4) makes it possible to interpret formally the sum

$$\sum_{\mathbf{n}'} F(\mathbf{p}_\mu^*, \mathbf{p}_\nu, \mathbf{n}') e^{i\mathbf{k}\mathbf{n}'} \quad (\mathbf{n}' \neq 0)$$

as the potential energy of the interaction of a dipole $\mathbf{p}_\mu^*$, located at the origin of the coordinate system with the dipoles $\mathbf{p}_\nu e^{i\mathbf{k}\mathbf{n}'}$ located at all other lattice sites $\mathbf{n}'$. We are interested in small values of $\mathbf{k}$ for which $(\mathbf{k}, \mathbf{a}_i) \sim 10^{-2} \ll 1$ and we can then replace the sum over $\mathbf{n}'$ by an integral and regard the system of dipoles as continuous.[2] In this way, we obtain

$$\sum_{\mathbf{n}'(\neq 0)} F(\mathbf{p}_\mu^*, \mathbf{p}_\nu, \mathbf{n}') e^{i\mathbf{k}\mathbf{n}'} = \frac{4\pi}{v}(\mathbf{s}, \mathbf{p}_\mu^*)(\mathbf{s}, \mathbf{p}_\nu) + O[(\mathbf{k}, \mathbf{a}_i)^2], \quad \mathbf{s} \equiv \frac{\mathbf{k}}{|\mathbf{k}|}, \tag{8}$$

where v is the volume of a unit cell, and

$$H_{\mu\nu}(\mathbf{k}) = H_{\mu\nu 0} + \frac{4\pi}{v}(\mathbf{s}, \mathbf{p}_\mu^*)(\mathbf{s}, \mathbf{p}_\nu) + \sum_{i=1}^{3} (\mathcal{H}_{\mu\nu\mathbf{a}_i} e^{i\mathbf{k}\mathbf{a}_i} + \mathcal{H}_{\mu\nu,-\mathbf{a}_i} e^{-i\mathbf{k}\mathbf{a}_i}) + O[(\mathbf{k}\mathbf{a}_i)^2]. \tag{9}$$

For example, in the absence of degeneracy, introducing for a fixed direction $\mathbf{s}$ the effective exciton mass $M(\mathbf{s})$ and, assuming that $s = 1$ and $\mu = \nu = 1$ in Eqs. (4) and (9), we obtain

$$\mathcal{E} = H_{11}(\mathbf{k}) = H_{110} + \frac{4\pi}{v}|(\mathbf{s}, \mathbf{p}_1)|^2 + \sum_{i=1}^{3} (\mathcal{H}_{11\mathbf{a}_i} + \mathcal{H}_{11,-\mathbf{a}_i}) + \frac{\hbar^2 \mathbf{k}^2}{2M(\mathbf{s})}. \tag{10}$$

In Eqs. (9) and (10), an estimate of the ratio of the third to the second term (for $\mathbf{s}$ not perpendicular to $\mathbf{p}_1$) gives

$$\mathrm{III/II} \propto v \sum_{i=1}^{3} (\mathcal{H}_{\mu\nu\mathbf{a}_i} + \mathcal{H}_{\mu\nu,-\mathbf{a}_i}) / 4\pi |\mathbf{p}_1|^2. \tag{11}$$

This ratio plays an important role in the subsequent treatment.

3. FRENKEL EXCITONS IN A SEMIINFINITE CRYSTAL AND RELEVANT BOUNDARY CONDITIONS

Let us assume that the surface of a crystal is the plane xy parallel to the vectors $\mathbf{a}_1$ and $\mathbf{a}_2$. The crystal is in the half-space $z > 0$ and the half space $z < 0$ is a vacuum.

There are now no molecules at the lattice sites n characterized by $n_3 \leq 0$. Consequently, in the system of equations (2), the quantities n_2 and m_3 assume only the positive values $1, 2, 3, \ldots$. We shall call these the truncated equations. We shall seek their solution in the form of a linear combination of solutions of the problem of an infinite crystal, all corresponding to the same energy $\mathcal{E}$. Since, along the x and y directions, there is still translational symmetry, we shall include in our linear combination only those solutions which have the same given tangential projection of the quasimomentum equal to $\mathbf{k}_\tau$.

If the constant-energy surface of an exciton l is ellipsoidal in the $\mathbf{k}$ space and if it is also true of other cases (see below), then, for given values of $\mathcal{E}$ and $\mathbf{k}_\tau$, the quasimomentum may assume two values which we shall denote by $\mathbf{k}^{(l)}$ and $\tilde{\mathbf{k}}^{(l)}$. In the first of these states, the z projection of the group velocity of the excitons is directed toward the surface of the crystal and we shall call it the incident wave. In the second state, the z projection is directed into the crystal and we shall call it the reflected wave.

This linear combination is of the form

$$\begin{aligned} &a_{n\nu} = \sum_{l=1}^{s} [C_l a_{0\nu}^{(l)} e^{i\mathbf{k}^{(l)}\mathbf{n}} + \tilde{C}_l \tilde{a}_{0\nu}^{(l)} e^{i\tilde{\mathbf{k}}^{(l)}\mathbf{n}}], \\ &\mathcal{E}_l(\mathbf{k}^{(l)}) = \mathcal{E}_{l_1}(\tilde{\mathbf{k}}^{(l_1)}) = \mathcal{E}, \quad \mathbf{k}_\tau^{(l)} = \tilde{\mathbf{k}}_\tau^{(l)} = \mathbf{k}_\tau, \\ &a_{0\nu}^{(l)} = a_{n\nu}^{(l)}(\mathbf{k}^{(l)}), \quad \tilde{a}_{0\nu}^{(l)} = a_{0\nu}^{(l)}(\tilde{\mathbf{k}}^{(l)}). \end{aligned} \tag{12}$$

This combination is definitely a solution of the complete system of equations (2). Therefore, after substituting it in the truncated equation, the remaining residue is

$$R_{\mathbf{m}\mu} = -\sum_{\substack{\nu \\ n_3 \leq 0}} H_{\mathbf{m}\mu,\mathbf{n}\nu} a_{n\nu}, \quad m_3 = 1, 2, 3, \ldots. \tag{13}$$

It is necessary to select the coefficients C_l and $\tilde{C}_l$ in such a way that this residue vanishes for all values of μ and $\mathbf{m}$ with $m_3 \geq 1$.

A. Case of large III/II ratios

We shall consider the case when the ratio (11) is much greater than unity, i.e., when the small number of matrix elements of the interaction of close (adjacent) cells makes a much greater contribution to the sum (2) than the infinitely large number of all the other terms representing the interaction of the more distant cells. We can then confine the sum (13) to just a few initial values of the index n_3 and replace $\exp\{in_3(\mathbf{k}^{(l)}\mathbf{a}_3)\}$ in Eq. (12) with unity. The substitution of Eq. (12) into Eq. (13) and the requirement $R_{\mathbf{m}\mu} = 0$ gives the following system of equations:

$$\sum_{\substack{\nu \\ n_3 \leq 0}} H_{\mathbf{m}\mu,\mathbf{n}\nu} \exp(i\mathbf{k}_\tau \mathbf{n}) \sum_{l=1}^{s} [C_l a_{0\nu}^{(l)} + \tilde{C}_l \tilde{a}_{0\nu}^{(l)}] = 0, \quad m_3 = 1, 2, 3, \ldots, \tag{14}$$

and hence we obtain the required relationship between the exciton amplitudes C_l and $\tilde{C}_l$:

$$\sum_{l=1}^{s} [C_l a_{0\nu}^{(l)} + \tilde{C}_l \tilde{a}_{0\nu}^{(l)}] = 0, \quad \nu = 1, 2, 3, \ldots, s. \tag{15}$$

Thus, on condition that Eq. (15) is satisfied, the linear combination (12) is the solution of the problem of a semiinfinite crystal. The corresponding wave function (1) becomes

$$\Psi_{\mathcal{E}_{\mathbf{k}_\tau}} = \sum_{l=1}^{s} [C_l \Psi_{l\mathbf{k}(l)} + \bar{C}_l \Psi_{l\bar{\mathbf{k}}(l)}], \tag{16}$$

where $\Psi_{l\mathbf{k}}$ are the solutions for an infinite crystal taken in the main periodic part of the crystal, one of whose faces is the plane xy. In Eqs. (12) and (16), the amplitudes of all the incident waves C_l can be specified in an arbitrary manner but the amplitudes of all the reflected waves $\bar{C}_l$ are expressed uniquely in terms of C_l on the basis of the conditions in Eq. (15).

The perturbation of a semiinfinite crystal by any time-dependent infinitesimally weak electromagnetic field, deformation, etc. with smooth variation in space creates a state

$$\Psi = \Psi^0 + \sum_{l\mathbf{k}} C_{l\mathbf{k}}(t)\Psi_{l\mathbf{k}} + \Psi', \quad |C_{l\mathbf{k}}| \ll 1, \tag{17}$$

where the coefficients $C_{l\mathbf{k}}$ of the reflected waves are expressed in terms of the coefficients of the incident waves,

$$\Psi^0 = S\prod_{\mathbf{n}} \psi_{\mathbf{n}}$$

is the ground state of the crystal, and Ψ' is a small sum of all the other states orthogonal to Ψ^0 and to all $\Psi_{\mathcal{E}_{\mathbf{k}_\tau}}$. In the state (17), a specific smooth macroscopic electric polarization appears in the crystal:

$$\mathbf{P}(\mathbf{r}, t) = \langle \Psi | \hat{\mathbf{P}}(\mathbf{r}) | \Psi \rangle,$$

where $\mathbf{r}$ is the coordinate of a macroscopic point in the crystal. The exciton contribution to the polarization is

$$\mathbf{P}_{ex}(\mathbf{r}, t) = \sum_{l\mathbf{k}} C_{l\mathbf{k}}(t) \langle \Psi^0 | \hat{\mathbf{P}}(\mathbf{r}) | \Psi_{l\mathbf{k}} \rangle + \text{c.c.}, \quad \hat{\mathbf{P}}(\mathbf{r}) = \frac{1}{v}\mathbf{p}_{\mathbf{r}}'. \tag{18}$$

We can easily show[1,5] that

$$\mathbf{P}_{ex}(\mathbf{r}, t) = \sum_{\nu l\mathbf{k}} C_{l\mathbf{k}}(t) a_{0\nu}^{(l)}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}} \{\mathbf{P}_\nu + i(\mathbf{k}, A_\nu)\} + \text{c.c.}, \tag{19}$$

where the braces contain the first and second terms of the expansion in $(\mathbf{k}, \mathbf{a}_i)$, A_ν is an asymmetric second-rank tensor, and $\mathbf{P}_\nu \equiv \mathbf{p}_\nu / v$. If $\mathbf{P}_\nu \neq 0$, i.e., if the exciton transition is allowed in the dipole approximation, we can simplify the expression in braces by dropping the term linear in $\mathbf{k}$. We then find from Eqs. (19) and (15) that

$$[\mathbf{P}_{ex}(\mathbf{r}, t)]_{z=0} = 0. \tag{20}$$

This is the auxiliary boundary condition obtained by the present author earlier[1] for the more restricted case. If only some of the projections of $\mathbf{P}_\nu$ differ from zero, the auxiliary boundary condition (20) only applies to these $\mathbf{P}_{ex}(\mathbf{r}, t)$ projections.

For the projections such that $\mathbf{P}_\nu = 0$, the polarization of Eq. (19) is governed—for all values of ν—by the term in braces which is linear in $\mathbf{k}$. This "quadrupole" term of the polarization will be denoted by $\mathbf{P}'_{ex}(\mathbf{r}, t)$. This term does not obey the boundary condition (20) but can be represented as the divergence of a second-rank tensor $Q(\mathbf{r}, t)$:

$$Q(\mathbf{r}, t) \equiv \sum_{\nu l\mathbf{k}} C_{l\mathbf{k}}(t) a_{0\nu}^{(l)} e^{i\mathbf{k}\mathbf{r}} A_\nu + \text{c.c.}, \quad \mathbf{P}_{ex}'(\mathbf{r}, t) = (\nabla, Q). \tag{21}$$

Since, in this case, the coefficients $C_{l\mathbf{k}}$ are related by the conditions (15), it follows that

$$[Q(\mathbf{r}, t)]_{z=0} = 0. \tag{22}$$

This is the auxiliary boundary condition for dipole-forbidden excitons (or for those polarizations of light for which an exciton transition is forbidden in the dipole approximation).

There is an infinite set of tensors Q which have the same divergence and which give the same term $\mathbf{P}'_{ex}(\mathbf{r}, t)$. However, the condition (22) should only be applied to Q given by Eq. (21). This requires knowledge of the characteristic tensor of a crystal

$$A^{(l)} \equiv \sum_\nu a_{0\nu}^{(l)} A_\nu.$$

The same tensor is required in the determination of the refractive indices of additional light waves of polarization in which a dipole exciton transition is forbidden. In this sense, the formulation of the auxiliary boundary condition given by Eq. (22) does not require determination of any new parameters of a crystal.

The number of conditions given by Eq. (15) is s. The number of auxiliary boundary conditions of Eqs. (20) and (22) can be much greater than s, but not all are independent. We can easily show that they are all equivalent to Eq. (15).

B. Case of small III/II ratios

We shall now discuss the case when the ratio (11) is much smaller than unity, i.e., when a small number of matrix elements of the interaction between close cells makes a much smaller contribution to the sum (2) than the infinitely large number of other terms representing the interaction of the more distant cells. In this case, we can simplify the right-hand side of Eq. (7) by dropping the second term and then substituting Eqs. (7) and (12) into Eq. (13); in this way, we obtain the residue of the truncated equation

$$R_{\mathbf{m}\mu} = \sum_{\nu, l} [C_l a_{0\nu}^{(l)} R_{\mathbf{m}\mu\nu}(\mathbf{k}^{(l)}) + \bar{C}_l \bar{a}_{0\nu}^{(l)} R_{\mathbf{m}\mu\nu}(\bar{\mathbf{k}}^{(l)})], \tag{23}$$

$$R_{\mathbf{m}\mu\nu}(\mathbf{k}) \equiv -\sum_{\substack{\mathbf{n} \\ n_3 \leq 0}} F(\mathbf{p}_\mu^*, \mathbf{p}_\nu, \mathbf{n}-\mathbf{m}) e^{i\mathbf{k}\mathbf{n}}. \tag{24}$$

Here, the sum over $\mathbf{n}$ is again interpreted as the energy of the electrostatic interaction of a dipole of moment $\mathbf{p}_\mu^*$, located at a point $\mathbf{m}$, with a system of dipoles of moments $\mathbf{p}_\nu e^{i\mathbf{k}\mathbf{n}}$, located at lattice sites $\mathbf{n}$ in the half-space $n_3 \leq 0$. If we regard this system of dipoles as a continuous medium with a specific polarization

$$\mathbf{P}(\mathbf{r}) = \Theta(-z)\mathbf{P}_\nu e^{i\mathbf{k}\mathbf{r}}, \quad \Theta(\xi) = \begin{cases} 1 & \text{for } \xi > 0 \\ 0 & \text{for } \xi < 0 \end{cases}, \tag{25}$$

we can find its potential $\varphi(\mathbf{r})$ from the Poisson equation, in which the space charge density is $\rho = -\nabla \mathbf{P}(\mathbf{r})$. Consequently, the potential in the region $z > 0$ is

$$\varphi = -\frac{2\pi}{|\mathbf{k}_\tau|} \frac{L(\mathbf{P}_\nu)}{|\mathbf{k}_\tau| + ik_z} e^{L(\mathbf{r})}, \quad L(\mathbf{r}) = i\mathbf{k}_\tau \mathbf{r} - |\mathbf{k}_\tau| z. \tag{26}$$

Next, we find that

$$R_{\mathbf{m}\mu\nu}(\mathbf{k}) = -v(\mathbf{P}_\mu^*, \nabla\varphi)_{\mathbf{r}=\mathbf{m}} = \frac{2\pi v}{|\mathbf{k}_\tau|} \frac{L(\mathbf{P}_\mu^*) L(\mathbf{P}_\nu)}{|\mathbf{k}_\tau| + ik_z} e^{L(\mathbf{m})}. \tag{27}$$

Since the linear combination (12) only includes waves with a given value of $\mathbf{k}_\tau$, the substitution of Eq. (27) into Eq. (23) gives

$$R_{\mathbf{m}\mu} = \frac{2\pi v}{|\mathbf{k}_\tau|} L(\mathbf{P}_\mu^*) e^{L(\mathbf{m})} \sum_l \left[\frac{C_l L(\mathbf{P}_l)}{|\mathbf{k}_\tau| + ik_z^{(l)}} + \frac{\tilde{C}_l L(\tilde{\mathbf{P}}_l)}{|\mathbf{k}_\tau| + i\tilde{k}_z^{(l)}} \right], \tag{28}$$

$$\mathbf{P}_l = \sum_{\nu=1}^{s} a_{0\nu}^{(l)} \mathbf{P}_\nu, \quad \tilde{\mathbf{P}}_l = \sum_{\nu=1}^{s} \tilde{a}_{0\nu}^{(l)} \mathbf{P}_\nu. \tag{29}$$

Now, in contrast to case A considered in the preceding subsection, the condition $R_{\mathbf{m}\mu} = 0$ reduces to just one equation

$$\sum_l \left[\frac{C_l L(\mathbf{P}_l)}{|\mathbf{k}_\tau| + ik_z^{(l)}} + \frac{\tilde{C}_l L(\tilde{\mathbf{P}}_l)}{|\mathbf{k}_\tau| + i\tilde{k}_z^{(l)}} \right] = 0, \tag{30}$$

which is insufficient in the $s > 1$ case to express the amplitudes of all the reflected waves $\tilde{C}_l$ in terms of the amplitudes of the incident waves C_l. A way out of this difficulty can be found in many special cases, as shown below.

1. Normal incidence ($|\mathbf{k}_\tau| \to 0$). In this case, $L(\mathbf{P}_\mu^*)$ and $L(\mathbf{P}_l)$ are infinitely small quantities of the order of $|\mathbf{k}_\tau|$. Therefore, the residue of the truncated equations (28) vanishes for any value of C_l and $\tilde{C}_l$. Consequently, the distant dipole–dipole interaction gives accidentally zero contribution to the residue $R_{\mathbf{m}\mu}$ (although it makes a large contribution to the exciton energy). Then, $R_{\mathbf{m}\mu}$ is governed by the contribution of the close interactions, considered in case A above, and C_l and $\tilde{C}_l$ are related by Eq. (15), i.e., by Eq. (20).

2. Exciton polarization $\mathbf{P}_{l'} = \tilde{\mathbf{P}}_{l'}$ perpendicular to the plane of incidence. This may occur for symmetry reasons (for example, if the plane of incidence coincides with the plane of the reflection symmetry of a crystal) or in the case when $s = 1$, etc. We then have again $L(\mathbf{P}_{l'}) = L(\tilde{\mathbf{P}}_{l'}) = 0$, i.e., the exciton amplitudes l' ($C_{l'}$ and $\tilde{C}_{l'}$) do not occur in Eqs. (28) and (30). If we assume the amplitudes of all the other excitons to vanish, we can see that the contribution (28) of the distant interactions to $R_{\mathbf{m}\mu}$ again vanishes and $R_{\mathbf{m}\mu}$ is governed by the contribution of the close interactions, which gives the conditions of Eq. (20) or the relationships of Eq. (15). It follows from $\mathbf{P}_{l'} = \tilde{\mathbf{P}}_{l'}$ that $a_{0\nu}^{(l')}(\mathbf{k}) = \tilde{a}_{0\nu}^{(l')}(\tilde{\mathbf{k}})$. Therefore, the relationships (15) become

$$C_{l'} + \tilde{C}_{l'} = 0. \tag{30a}$$

Hence, $\tilde{C}_{l'}$ can be expressed uniquely in terms of $C_{l'}$. An l' exciton reflects without transformation into other types of exciton.

In cases 1 and 2 discussed above, the ratio III/II can have any value. However, we shall now consider only case B and oblique incidence.

3. $s = 3$. As is known, in this case, $\varphi_{\mathbf{m}\nu}$ can be selected in such a way that the basis vectors $\mathbf{P}_\nu$ ($\nu = 1, 2, 3$) are mutually orthogonal and have the same magnitude. In the case of an infinite crystal, an analysis of the functions (1) and (3) as an approximation for the wave function with the parameters $a_{0\nu}$ in the direct variational method readily yields the energy functional in the form

$$\mathcal{E}(\mathbf{k}, \ldots a_{0\nu} \ldots) = \mathcal{E}_\perp + 4\pi v |(\mathbf{s}, \mathbf{P}_l)|^2, \quad \mathcal{E}_\perp \equiv H_{110} + \sum_{i=1}^{3} (\mathcal{H}_{11a_i} + \mathcal{H}_{11,-a_i}),$$

where the parameters $a_{0\nu}$ occur only in $\mathbf{P}_l$, in accordance with Eq. (29). We can determine them by extremizing the energy $\mathcal{E}(\mathbf{k}, \ldots a_{0\nu} \ldots)$ subject to an additional orthonormalization condition (5), i.e., the condition $|\mathbf{P}_l|^2 = \text{const}$. Thus, it remains to select the extremal directions of the vector $\mathbf{P}_l$. This procedure is much simpler and clearer than the solution of the system of equations (4) and (9). Extremization yields a nondegenerate longitudinal exciton ($\mathbf{P}_l \| \mathbf{s}$), whose energy is $\mathcal{E}_l = \mathcal{E}_\parallel \equiv \mathcal{E}_\perp + 4\pi v |\mathbf{P}_l|^2$ and two transverse excitons ($\mathbf{P}_{l'}, \mathbf{P}_{l''} \perp \mathbf{s}; \mathbf{P}_{l'} \perp \mathbf{P}_{l''}$), whose energy is $\mathcal{E}_{l'} = \mathcal{E}_{l''} = \mathcal{E}_\perp$. The nature of the ellipsoidal constant-energy surfaces of the excitons can be determined by calculating their energy more accurately, including terms of the order of $|\mathbf{k}|^2$. The longitudinal-transverse splitting $4\pi v |\mathbf{P}_l|^2$ in case B is large and, therefore, a longitudinal exciton reflected from the surface of a crystal cannot transform into transverse excitons or vice versa. Consequently, linear combinations of Eq. (12) should include only terms representing longitudinal excitons or transverse excitons.

In the case of a longitudinal exciton, Eq. (30) does not include a sum over l and this boundary condition then becomes

$$\frac{C_l L(\mathbf{P}_l)}{|\mathbf{k}_\tau| + ik_z^{(l)}} + \frac{\tilde{C}_l L(\mathbf{P}_l)}{|\mathbf{k}_\tau| + i\tilde{k}_z^{(l)}} = 0. \tag{30b}$$

Hence, $\tilde{C}_l$ can be expressed uniquely in terms of C_l.

In the case of transverse excitons, the sum (30) has two terms: l' and l''. We shall only consider the case when one of these excitons, for example, l', fits the above case 2, $L(\mathbf{P}_{l'}) = L(\tilde{\mathbf{P}}_{l'}) = 0$, so that the term l' in Eq. (30) disappears and, therefore, in the case of the exciton l'', we obtain a condition of the type (30b), which defines uniquely $\tilde{C}_{l''}$ in terms of $C_{l''}$.

4. $s = 2$. In this case, there are two mutually orthogonal equal basis vectors $\mathbf{P}_\nu$ ($\nu = 1, 2$), which define a plane in which the variation of the parameters $a_{0\nu}$ causes the vector $\mathbf{P}_l$ to rotate without change in its magnitude. The energy functional $\mathcal{E}(k, \ldots a_{0\nu} \ldots)$ is of the same form as in case 3. However, its extremum corresponds to a transverse exciton ($\mathbf{P}_{l_1} \perp \mathbf{s}$), whose energy is

$\mathcal{E}_{l'} = \mathcal{E}_\perp$. A second extremum occurs when $\mathbf{P}_l$ is rotated in the same plane and it forms the smallest (or largest) angle with $\mathbf{s}$. It corresponds to an inclined exciton ($\angle \mathbf{P}_{l_2}, \mathbf{s} = a$), whose energy is

$$\mathcal{E}_{l_2} = \mathcal{E}_\perp + 4\pi v |\mathbf{P}_l|^2 \cos^2 \alpha.$$

If α differs greatly from $\pi/2$, then $\mathcal{E}_{l_2}$ differs considerably from $\mathcal{E}_{l_1}$. Therefore, on the right-hand side of Eq. (12) and in Eq. (30), there is no sum but only one of these excitons occurs there and Eq. (30) reduces to Eq. (30b). The amplitude of a reflected wave is then expressed uniquely in terms of the amplitude of the incident wave.

For $\alpha \approx \pi/2$, we have $\mathcal{E}_{l_1} \approx \mathcal{E}_{l_2}$. We shall consider only the case when one of these excitons fits case 2, discussed above, and its reflection law has the form (30a). In Eq. (30), there is then only the second exciton and it reduces to Eq. (30b).

5. $s = 1$. In this case, Eq. (30) changes directly into Eq. (30b) and there is no ambiguity in the reflection law.

For the types of point symmetry encountered in crystals, the degree of degeneracy does not usually exceed 3 (accidential degeneracy will be ignored). In all the special cases discussed above, the amplitude of reflected excitons is expressed uniquely in terms of the amplitude of the incident wave. The sum (30) then retains no more than one term, i.e., the condition (30) reduces to Eq. (30b) or it is replaced by the condition (30a).

In general, the solution of the problem of a semiinfinite crystal cannot be represented as a linear combination of solutions for an infinite crystal. This is demonstrated in the next section for optical excitons but it can be shown in exactly the same way for excitons.

4. OPTICAL WAVES (OPTICAL EXCITONS, POLARITONS) IN A SEMIINFINITE CRYSTAL AND AUXILIARY BOUNDARY CONDITIONS

The formulas (17) and (19), subject to the additional conditions (15), and the expressions (20) and (22) deduced from them, apply to a semiinfinite crystal perturbed by any weak slowly varying in space electromagnetic, acoustic, or other field. It is clear from Eqs. (19) and (15) that, for a semiinfinite crystal, we cannot expect at all a polarization proportional to $\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$, even if the perturbation is an electric field proportional to $\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$. Hence, it follows that a linear polarization response of a semiinfinite crystal cannot be described by the permittivity $\epsilon(\omega, \mathbf{k})$ and the Maxwell equations do not have particular solution proportional to $\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$ even before the application of the Maxwell boundary conditions.

An analysis shows that, in the presence of spatial dispersion and external field sources, the solution of the Maxwell equations for a semiinfinite crystal becomes much more complex. However, in the absence of external sources, this solution can frequently be obtained as a linear combination of solutions for an infinite crystal, in the same way as has been done for excitons in the preceding section. We shall justify this method of solution and fine the conditions similar to Eqs. (15) and (30), which apply in the case of optical waves (optical excitons).

We shall consider that an exciton radiative transition is allowed in the dipole approximation. The energy of the interaction of a crystal with an external electromagnetic field is

$$W = -\frac{1}{c}\sum_{\mathbf{n}} \dot{\mathbf{p}}_{\mathbf{n}} \mathbf{A}(\mathbf{n}, t),$$

where $\mathbf{A}(\mathbf{r}, t)$ is the vector potential describing only the rotational part of the field (div $\mathbf{A} = 0$) and the sum over n is taken over the main part of the crystal. The state of a field-perturbed crystal (17) can be described in the form

$$\Psi = \Psi^0 + \sum_{m\mu} b_{m\mu}(t)\,\Phi_{m\mu} + \Psi'. \tag{31}$$

It is assumed that the incident light is of frequency $\omega \approx \mathcal{E}_l(\mathbf{k})/\hbar$ and that the states included in Ψ' have energies sufficiently far from $\mathcal{E}_l(k)$ so that Ψ' can be regarded as small compared with the second term on the right-hand side of Eq. (31) and the contribution of the states Ψ' to the polarization of the crystal can be calculated ignoring spatial dispersion but introducing some "background" polarizability $\beta_0(\omega)$, which varies slowly with ω (Ref. 5).

If only the terms of the first order of smallness in respect of the perturbation W are retained in the secular Schrödinger equation, this equation becomes

$$i\hbar \dot{b}_{m\mu} - \sum_{n,\nu=1}^{s} H_{m\mu,n\nu} b_{n\nu} = -\frac{v}{c}\dot{\mathbf{P}}_\mu \mathbf{A}(\mathbf{m}, t). \tag{32}$$

The Maxwell equations can be written in the form

$$\Delta \mathbf{A} - \frac{1}{c^2}\ddot{\mathbf{A}} = -\frac{4\pi}{c}\dot{\mathbf{P}}_\perp, \quad \mathbf{E}_\perp = -\frac{1}{c}\dot{\mathbf{A}}, \quad \mathbf{E}_\parallel = -4\pi \mathbf{P}_\parallel, \tag{33}$$

where $\mathbf{P}(r, t)$ is the specific polarization of a crystal given by

$$\mathbf{P} = \beta_0\left(i\frac{\partial}{\partial t}\right)\mathbf{E}_\perp + \sum_{\mu=1}^{s} \mathbf{P}_\mu (b_{m\mu})_{m\to r}. \tag{34}$$

Here, the term with the tensor $\beta_0(i\partial/\partial t)$ represents the contribution to the polarization which does not require allowance for the spatial dispersion and the subscripts "$\perp$" and "$\parallel$" represent rotational and irrotational parts of a vector.

The system (32)–(34) describes a light wave in the exciton resonance region, i.e., an optical exciton (polariton). From this equation, we can determine $b_{m\mu}(t)$, $\mathbf{A}(\mathbf{r}, t)$ and $\mathbf{P}(\mathbf{r}, t)$.

In the case of an infinite crystal, we seek the solution of Eqs. (32)–(34) in the form

$$\mathbf{A}=\mathbf{A}_0 e^{i(\mathbf{kr}-\omega t)}, \quad \mathbf{A}_0 \perp \mathbf{k}, \quad b_{\mathbf{m}\mu}=b_{0\mu}e^{i(\mathbf{km}-\omega t)}, \tag{35}$$

and expand an s-dimensional vector $b_{0\mu}$ ($\mu = 1, 2, \ldots, s$) in terms of a complete system of orthogonal s-dimensional vectors $a_{0\mu}^{(l)}$ [see Eqs. (4) and (5)]:

$$b_{0\mu}=\sum_{l=1}^{s} B_l a_{0\mu}^{(l)}(\mathbf{k}). \tag{36}$$

Substituting Eqs. (35) and (36) in Eq. (32), we find that allowance for Eq. (4) gives

$$B_l=\frac{i\omega_l}{c}\frac{V(\mathbf{P}_l^*, \mathbf{A}_0)}{\mathscr{E}_l(\mathbf{k})-\hbar\omega}, \tag{37}$$

where $\omega_l = \mathscr{E}_l(\mathbf{k})/\hbar$ and V is the volume of the main periodic part of the crystal. The substitution of Eqs. (36) and (37) into Eq. (34) gives

$$\mathbf{P}=\beta\mathbf{E}_\perp, \quad \beta(\omega, \mathbf{k})=\beta_0(\omega)+\sum_{l=1}^{s}\frac{\omega_l V T^{(l)}}{\omega[\mathscr{E}_l(\mathbf{k})-\hbar\omega]}. \tag{38}$$

Here, $T^{(l)}$ is the dyad tensor: $T_{xy}^{(l)} \equiv (P_l)_x (P_l^*)_y$. Equation (38) is identical with that obtained in the earlier papers[1,5] for the general case without specifying the exciton model.

Finally, the substitution of Eq. (38) into the first expression in Eq. (33) gives the following equation for $\mathbf{A}_0$:

$$(n^2-1)\mathbf{A}_0=4\pi\eta\beta\mathbf{A}_0; \quad \eta_{xy}=\delta_{xy}-s_x s_y, \quad \mathbf{s}=\mathbf{k}/|\mathbf{k}|, \quad n=|\mathbf{k}|c/\omega. \tag{39}$$

Here, n is the refractive index of the light wave and η is a tensor projecting a vector on a plane perpendicular to $\mathbf{s}$. The condition of solubility of the homogeneous equation system (39) is the dispersion equation. Its various forms and solution have already been considered by the present author[1,5-7] and by others. Each root of the dispersion equation $n_j(\omega)$ [or $k_j(\omega)$], where j is the root number, and the corresponding solution of the system (39) given by $\mathbf{A}_{0j}$ represent a light wave. The number of such roots N exceeds two and it implies the existence of additional light waves. For example, $N = 5$ in one of the earlier treatments by the present author.[7]

In the case of a semiinfinite crystal, Eqs. (33) and (34) retain their form and only Eq. (32) changes: this equation becomes truncated, i.e., in the sum over $\mathbf{n}$, ν, there are no terms with $n_3 \leq 0$, and m_3 assumes only the positive values $1, 2, 3, \ldots$. Therefore, any solution of the problem of an infinite crystal $b_{\mathbf{m}\mu}$, $\mathbf{A}(\mathbf{r}, t)$, substituted into the equations for a semiinfinite crystal, satisfies Eqs. (33) and (34) and Eq. (32) then has a residue

$$R_{\mathbf{m}\mu}' = -\sum_{\substack{\mathbf{n}\nu \\ n_3\leq 0}} H_{\mathbf{m}\mu,\mathbf{n}\nu} b_{\mathbf{n}\nu}, \quad m_3=1, 2, 3, \ldots \tag{40}$$

This is analogous to the residue (13) in the exciton problem.

In view of the linearity and homogeneity of Eqs. (32)–(34), their solutions are any linear combinations of particular solutions. We shall seek a solution of the problem for a semiinfinite crystal in the form of a linear combination of the solutions of Eqs. (35)–(39) for an infinite crystal. The coefficients of the linear combination have to be selected so that the residue (40) disappears for all values of μ and $\mathbf{m}$ with $m_3 \geq 1$. This method of solution of the truncated equations has been used already by the present author,[1] by Mahan and Obermair,[8] and by others.

Since the coefficients in the equations (32)–(34) are independent of t and invariant in the case of translation along $\mathbf{a}_1$ and $\mathbf{a}_2$, we can seek the solution of the problem for a semiinfinite crystal in the form $A \propto \exp\{i(\mathbf{k}_\tau \mathbf{r} - \omega t)\}$, $b_{\mathbf{m}\mu} \propto \exp\{i(\mathbf{k}_\tau \mathbf{m} - \omega t)\}$, i.e., a linear combination can only include solutions with identical values of ω and $\mathbf{k}_\tau$. Therefore, in a dispersion equation, it is necessary to fix ω and $\mathbf{k}_\tau$ and then find the roots k_{zj} ($j = 1, 2, \ldots, N$) and the corresponding values of $\mathbf{A}_{0j}$, B_{ij}, and $b_{\mathbf{m}\mu j}$. An arbitrary coefficient in this solution is the absolute value of the vector $\mathbf{A}_{0j}$ (amplitude of a light wave).

A. Case of large III/II ratios

In exactly the same way as in the derivation of Eqs. (14) and (15), we can show that, in the case of the j-th solution, the residue (40) is

$$R_{\mathbf{m}\mu}' = -\sum_{\substack{\mathbf{n}\nu \\ n_3\leq 0}} H_{\mathbf{m}\mu,\mathbf{n}\nu} b_{0\nu j} \exp\{i(\mathbf{k}_\tau \mathbf{n}_\tau - \omega t)\}. \tag{41}$$

Hence, it follows that the required superposition of solutions corresponding to $R_{\mathbf{m}\mu}' = 0$ is given by the conditions

$$b_{0\nu}=\sum_{j=1}^{N} b_{0\nu j}=\sum_{j=1}^{N}\sum_{l=1}^{s} B_{lj} a_{0\nu}^{(l)}(\mathbf{k}_j)=0, \quad \nu=1, 2, \ldots, s. \tag{42}$$

The above equation represents the auxiliary boundary conditions for light waves. They are completely analogous to the conditions of Eq. (15) because the latter can also be written in the form $a_{0\nu}=0$ using Eq. (12). The number of these conditions in Eq. (42) is s, i.e., it is identical with the number of additional light waves in case A. Therefore, these conditions together with the Maxwell boundary conditions are sufficient for the amplitudes of all the waves appearing in a semiinfinite crystal and for the amplitude of wave reflected from this crystal to be expressed uniquely in terms of the amplitude of a wave incident on the crystal.

Substituting Eq. (37) into Eq. (42), we can rewrite the auxiliary boundary conditions in the form of equations for the wave amplitudes A_{0j}:

$$\sum_{j=1, l=1}^{N, s}\left[\frac{\omega_l(\mathbf{k})\, a_{0\nu}^{(l)}(\mathbf{k})\,(\mathbf{P}_l^*, \mathbf{A}_{0j})}{\mathscr{E}_l(\mathbf{k})-\hbar\omega}\right]_{\mathbf{k}=\mathbf{k}_j}=0, \quad \omega_l=\frac{1}{\hbar}\mathscr{E}_l(\mathbf{k}), \quad \mathbf{k}_j=\mathbf{k}_\tau+\mathbf{k}_{z_j}, \quad \nu=1, 2, \ldots, s. \tag{43}$$

The contribution of the excitons in question to the specific polarization of a crystal induced optically, $\mathbf{P}_{ex}(\mathbf{r}, t)$, is equal to the second term on the right-hand side of Eq. (34). Hence, it is clear that Eq. (20) always follows from the auxiliary boundary conditions of Eq. (42) and

that Eq. (42) follows from Eq. (20).

We shall now consider the case of dipole-forbidden excitons. In this case, we have $\mathbf{P}_\mu = 0$ and, therefore, the matrix elements of the dipole moment have to be considered more rigorously, allowing for the overlap of the wave functions of neighboring cells in a crystal. Consequently, Eqs. (32)–(34) are modified as follows: instead of $\mathbf{P}_\mu$, we have now to substitute an operator (∇, A_μ), where A_μ is the same second-rank tensor which occurs in Eq. (19) and ∇ represents differentiation with respect to r or m if the function of r or m is found on the right-hand side. The residue of Eq. (32) still has the form given by Eq. (40). Therefore, the auxiliary boundary conditions retain the form of Eq. (42) or (43). In the formulas (29), (37), (38), and (43), we have to replace $\mathbf{P}_\nu$ with $i(\mathbf{k}, A_\nu)$.

The exciton contribution to the polarization $\mathbf{P}'_{ex}(\mathbf{r}, t)$ is equal to the second term on the right-hand side of Eq. (34), which now becomes

$$\mathbf{P}_{ex}'(\mathbf{r}, t) = \nabla Q(\mathbf{r}, t), \quad Q(\mathbf{r}, t) = \sum_{\mu=1}^{s} A_\mu (b_{\mathbf{m}\mu})_{\mathbf{m}\to\mathbf{r}}. \tag{44}$$

Hence, we can see that the auxiliary boundary conditions of Eq. (42) follow from the corresponding conditions of Eq. (22).

B. Case of small III/II ratios

If Eq. (40) is modified by substituting

$$b_{\mathbf{n}\nu} = \sum_j b_{\mathbf{n}\nu j},$$

where $b_{\mathbf{n}\nu j}$ are given by Eqs. (35), (36), and (39), we find—in full analogy with the derivation of Eq. (28)—that

$$R_{\mathbf{m}\mu}' = \frac{2\pi\nu}{|\mathbf{k}_\tau|} L(\mathbf{P}_\mu^*) e^{L(\mathbf{m})-i\omega t} \sum_{j,l=1}^{N_0} \left[\frac{B_{lj} L(\mathbf{P}_l)}{|\mathbf{k}_\tau| + ik_z}\right]_{\mathbf{k}=\mathbf{k}_j}. \tag{45}$$

Hence, we obtain the auxiliary boundary conditions for light waves in the form of a single equation

$$\sum_{j,l} \left[\frac{B_{lj} L(\mathbf{P}_l)}{|\mathbf{k}_\tau| + ik_z}\right]_{\mathbf{k}=\mathbf{k}_j} = 0. \tag{46}$$

Substituting here B_{lj} in the form of Eq. (37), we find the required relationship between the values of A_{0j}.

The number of additional waves may be greater than one. In this case, a single auxiliary boundary condition (46) is insufficient. The way out of this difficulty will be demonstrated below for specific cases, exactly as at the end of Sec. 3 after the derivation of Eq. (30). It involves supplementing the auxiliary boundary conditions of Eq. (46) with some conditions from Eq. (43). However, it is convenient to consider first the case of arbitrary values of the ratio III/II so as to prove this rigorously.

C. Case of arbitrary III/II ratios

In this case, it is necessary to retain both terms on the right-hand side of Eq. (7). Consequently, the auxiliary boundary conditions for light waves become

$$R_{\mathbf{m}\mu}' \exp\{-i(\mathbf{k}_\tau \mathbf{m} - \omega t)\} = \sum_{j=1,l=1}^{N_0} B_{lj} \Big\{ \sum_{\nu=1}^{s} h_{\mu\nu m_3} a_{0\nu}^{(l)}(\mathbf{k}) + \frac{2\pi\nu L(\mathbf{P}_\mu^*) L(\mathbf{P}_l)}{|\mathbf{k}_\tau|(|\mathbf{k}_\tau| + ik_z)} \exp(-|\mathbf{k}_\tau| a_{33} m_3) \Big\}_{\mathbf{k}\to\mathbf{k}_j} = 0, \tag{47}$$

where $m_3 = 1, 2, 3, \ldots$,

$$h_{\mu\nu m_3}(\mathbf{k}) = -\sum_{n_1=-\infty}^{\infty} \sum_{n_2=-\infty}^{\infty} \sum_{n_3=-\infty}^{0} \mathcal{H}_{\mu\nu, n_1, n_2, n_3 - m_3} e^{i\mathbf{k}\mathbf{n}}. \tag{48}$$

In Eq. (47), the first and second terms in braces on the right represent the contributions of the terms $\mathcal{H}_{\mu\nu\mathbf{n}'}$ and $F(\mathbf{p}_\mu^*, \mathbf{p}_\nu, \mathbf{n}')$ on the right-hand side of Eq. (7). Since $h_{\mu\nu m_3}$ decreases rapidly on increase in m_3, we find that, in the limit $m_3 \to \infty$, we have $R'_{\mathbf{m}\mu} \to 0$ identically, i.e., for any value of A_{0j}. This means that, inside a crystal at a distance of many wavelengths from the surface (!), the solution of the truncated system (32)–(34) is an arbitrary linear combination of the solutions for an infinite crystal and, in particular, it can be a simple exponential wave (35). This means that inside the crystal we can use the permittivity $\epsilon(\omega, \mathbf{k})$. Moreover, it justifies the formulation of the problem of reflection from the surface and partial escape into vacuum of waves which inside the crystal tend asymptotically to the exponential form given by Eq. (35).

In determining the reflection coefficient of such waves, it is necessary to know the solution of the truncated equations (32)–(34) and in the surface layer of a crystal. There is no doubt about the existence of this solution in the general case C discussed above although it has not as yet been found. Therefore, it is important to consider at least those cases when this solution, valid right up to the surface, can be represented as some specific linear combination of solutions for an infinite crystal. This can be done if we can ensure that the residue (47) vanishes.

If in Eq. (47) it is necessary to retain both terms in braces, then—because m_3 assumes all positive integral values—the number of such equations is infinite and they cannot be satisfied by a finite number N of the amplitudes A_{0j}. In those cases, the solution of the problem for a semiinfinite crystal cannot be represented as a linear combination of solutions for an infinite crystal.

The only way of satisfying Eq. (47) is to neglect in some of the equations the second term in braces so that these equations reduce to the auxiliary boundary conditions (42) for case A and, in the remaining equations, to drop the first term from the braces, which reduces them to the auxiliary boundary conditions (46) for case B. In both ways, the main aim is achieved: the equations cease to depend on m_3. The total number of auxiliary boundary conditions then becomes equal to s, i.e., to the number of additional light waves. These limiting forms of the auxiliary boundary conditions will be obtained below for certain specific cases. Case A has already been discussed above.

1. Normal incidence of light ($|\mathbf{k}_\tau| \to 0$). In this case, as shown in Sec. 3, the second term in the braces in

Eq. (47) is of the order of $|\mathbf{k}_\tau|$ i.e., it vanishes. These equations reduce to Eq. (42), i.e., to Eq. (43) and, therefore, to Eq. (20).

2. The direction of one of the principal axes $\mathbf{e}$ *of the tensors* $\beta_0(\omega)$ *and* $T^{(l)}$ *[see Eq. (38)] is governed by the crystal symmetry (is independent of* ω*) and is perpendicular to the plane of incidence, and* $\mathbf{P}_{l'} \| \mathbf{e}$. The other vectors are $\mathbf{P}_l \perp \mathbf{e}$ for $l \neq l'$. It then follows from Eqs. (38) and (39) that, after quadratic expansion of $\mathcal{E}_{l'}(\mathbf{k})$ in respect of $|\mathbf{k}|$, there are, for given values of $\mathbf{k}_\tau$ and ω, two incident and two reflected waves ($j = 1, 2, 3, 4$) with the same polarization $\mathbf{A}_{0j} \| \mathbf{e}$. For these waves, it follows from Eq. (37) that $B_{l'j} \neq 0$ but, for the rest, we have $B_{lj} = 0$ when $l \neq l'$. Therefore, only the terms with $l = l'$ remain in Eq. (47). However, there is now no second term in the braces because $L(\mathbf{P}_{l'}) = 0$ ($\mathbf{P}_{l'} \perp \mathbf{k}_\tau, 0z$). Thus, the contribution of the dipole–dipole interactions to $R_{m\mu'}$ for the four ways in question is accidentally zero; $R_{m\mu'}$ is governed by the short-range interactions. In this case, retaining only the first terms in braces in Eq. (47), we obtain the auxiliary boundary conditions (42) for case *A*, i.e., Eq. (20). For the four waves in question, we have $\mathbf{P}_{l'}$ and, consequently, $a_{0\nu}^{(l)}(\mathbf{k}_j)$ are independent of j. Therefore, the conditions (42) become

$$\sum_{j=1}^{4} B_{l'j} = 0. \tag{49}$$

The above expression is analogous to Eq. (30a). However, the auxiliary boundary condition of Eq. (49) is sufficient for the determination of the reflection laws of all the four waves mentioned above. These waves may transform into one another as a result of reflection but not into any other waves.

In cases 1 and 2, the ratio III/II can have any value. However, we shall now consider only case *B* and oblique incidence.

3. $s = 3$. Bearing in mind the points made about excitons in the corresponding subsection (B3) above and retaining the same notation, we shall denote a longitudinal exciton by l and transverse excitons by l' and l''. In the case of an infinite crystal, the system (32)–(34) has a solution in the form of a longitudinal light wave:

$$\mathbf{A} = 0, \quad \mathbf{P} = \mathbf{P}_\parallel = -\frac{1}{4\pi}\mathbf{E}_\parallel = \mathbf{P}_l e^{i(\mathbf{k}\mathbf{r} - \omega t)} \tag{50}$$

The system (32) for $b_{m\nu}$ is then identical with the system (2) for $a_{m\nu}$, i.e.,

$$b_{m\nu} = a_{m\nu}^{(l)}(\mathbf{k}), \quad \omega = \mathcal{E}_l(\mathbf{k})/\hbar. \tag{51}$$

Hence, it is clear that such a light wave is identical with a longitudinal Schrödinger exciton (see also one of the author's papers[7]). According to Eq. (37), we have $B_l = 0/0$, i.e., this quantity can be an arbitrary constant which we can identify with C_l.

For a semiinfinite crystal in case B, the reflection law for a longitudinal exciton has the form (30b):

$$\sum_{j=1}^{2} [B_{lj} L(\mathbf{P}_l)/(|\mathbf{k}_\tau| + ik_z)]_{\mathbf{k}=\mathbf{k}_j} = 0, \text{ i.e., } [(\mathbf{P}_{ex})_\tau]_{z=0} = 0. \tag{52}$$

Here, $j = 1$ and $j = 2$ are, respectively, the indices of the incident and reflected waves. The auxiliary boundary conditions of Eq. (52) allow us to express uniquely the amplitude of an incident wave, and there is no transformation of longitudinal into transverse waves.

In the case of an infinite crystal, for a given direction $\mathbf{s}$, Eqs. (32)–(34) or (38) and (39) also have solutions in the form of transverse waves. For these waves, the spatial dispersion is considerable in the spectral range $\omega \approx \mathcal{E}_\perp/\hbar$. We shall only consider case 2, when four transverse waves have the polarization $\mathbf{A}_{0j} \| \mathbf{P}_{l'} \| \mathbf{e}$ and, therefore, they make no contribution to the residue (45). The auxiliary boundary conditions for these waves have the form (49). The other four transverse waves ($j = 5, 6, 7, 8$) are polarized in the plane of incidence: $\mathbf{A}_{0j} \| \mathbf{P}_{l''} \perp \mathbf{P}_{l'}, \mathbf{s}$. According to Eq. (37), the following relationships apply to these waves: $B_{l''j} \neq 0$, $B_{lj} = B_{l'j} = 0$. Therefore, the auxiliary boundary conditions (46) for such waves become

$$\sum_{j=5}^{8} [B_{l''} L(\mathbf{P}_{l''})/(|\mathbf{k}_\tau| + ik_z)]_{\mathbf{k}=\mathbf{k}_j} = 0, \text{ i.e., } [(\mathbf{P}_{ex})_\tau]_{z=0} = 0. \tag{53}$$

These, together with the Maxwell boundary conditions, are sufficient to obtain a single-valued solution of the problem of wave reflection.

4. $s = 2$. Bearing in mind the conclusions reached about excitons in the corresponding subsection (B4) above and retaining the same notation as before, we shall denote a transverse exciton by l_1 and an inclined exciton by l_2. If $\mathcal{E}_{l_2}$ differs considerably from $\mathcal{E}_{l_1}$, then, in the spectral range when one of these excitons makes a considerable contribution to the spatial dispersion [i.e., when the denominator of the relevant term in Eq. (38) is close to zero], the second exciton makes practically no contribution to the spatial dispersion and the corresponding term in Eq. (38) can be removed from the sum and included in $\beta_0(\omega)$. Consequently, the sum in Eq. (38) consists of just one term and, for a given $\mathbf{s}$, there is only one additional wave and the auxiliary boundary conditions (46) assume the form given by Eq. (52).

If $\mathcal{E}_{l_1} \approx \mathcal{E}_{l_2}$, we need only consider case 2, when some of the waves are reflected in accordance with the auxiliary boundary conditions (49) and other waves in accordance with the conditions of type (53).

5. $s = 1$. In this case, Eq. (46) transforms directly to the auxiliary boundary conditions (53) and there is no ambiguity in the reflection law.

The above analysis can be summarized as follows. The solution of Eqs. (32)–(34) for optical excitons in a semiinfinite crystal can be obtained as a linear combination of solutions of these equations for any infinite crystal only in those special cases when the dependence on m_3 disappears from Eq. (47). Consequently, some of the equations of this type transform to the auxiliary boundary conditions (42), i.e., to Eq. (20) or (22),

whereas others transform to the auxiliary boundary conditions (46). These conditions determine the relationship between the coefficients of linear combinations. The solution of the Maxwell equations in vacuum should be related to the solution for a semiinfinite crystal by the Maxwell boundary conditions.

The solution of equations for an infinite crystal mentioned above can be obtained, in particular, using the theoretically calculated or experimentally determined permittivity $\epsilon(\omega,\mathbf{k})$. Only in this sense can we use $\epsilon(\omega,\mathbf{k})$ in solving the optical exciton problem for a semiinfinite crystal.

5. COMPARISON OF RESULTS. DISCUSSION

In the last 20 years, very many contradictory papers have been published on the question of the auxiliary boundary conditions. In particular, there have been statements to the effect that these conditions are quite unnecessary. In answer to this objection, we point out that, if optical exciton solutions are deduced as linear combinations of solutions for an infinite crystal, then the auxiliary boundary conditions are necessary. If the polarization response of a semiinfinite crystal, for example,

$$\mathbf{P}_{sc}(\mathbf{r},t)=\int_{-\infty}^{t}dt'\int_{z'>0}d^2r'\chi(t-t',x-x',y-y',z,z')\mathbf{E}(\mathbf{r}',t'), \tag{54}$$

is known, then no auxiliary boundary conditions are necessary in the subsequent solution of Eqs. (33) and (54); in the above expression, χ is found from the equations of mechanics for a semiinfinite crystal, for example, using the functions (16), where C_l now satisfies the auxiliary boundary conditions for a Schrödinger exciton. In the case of Frenkel excitons, the results are identical with those obtained in Sec. 4.

It is interesting to compare the results of Sec. 4 with those obtained by other authors. However, we must make a preliminary selection among the numerous contradictory results.

In a large group of papers,[9-13] use is made of Eq. (54), but it is assumed arbitrarily that z and z' occur in χ in the form of the difference $z-z'$, in spite of the fact that there is no translational symmetry along the z axis in a semiinfinite crystal. This assumption is in conflict with the dependence of χ on z and z', obtained from the equations of mechanics for specific exciton models. Justified criticisms of this group of papers can also be found elsewhere.[14-17] We thus see that the results reported in this group of papers can hardly be correct.

In a second group of papers,[8,15-19] there are no arbitrary assumptions and the polarization response of a crystal is found from the equations of mechanics. Only the exciton model, simplifying the solution of these equations, is specified. The model seriously limits the range of validity of the quantitative results but the results are still rigorous. This group of papers includes the author's own work.[1] Preference should clearly be given to a comparison with the results reported in this group of papers.

We shall first compare the above results with the exact microcalculations carried out for a model consisting of point dipoles (one-dimensional harmonic oscillators) located at the sites of a cubic lattice in the half-space $z>0$. The crystal planes are defined by $z=am_3$, where $m_3=1,2,3,\ldots$. In each plane, the dipoles oscillate in synchronism and with the same amplitudes. A light wave is incident normally. The potential energy of the interaction between the layers is proportional to the product of the dipole moments of the oscillators in these layers and its dependence on the distance between the layers is in the form $\exp(-\gamma|m_3-m_3'|)$. This one-dimensional problem is solved by Sipe and Van Kranenkonk[17] and Mead[15] without any approximations. It is shown that, in an appropriate infinite medium, there are two identical polarized waves with the wave vectors $\mathbf{k}_j$, where $j=1,2$. In the case of a semiinfinite medium, the solution is a linear combination of these waves but their amplitudes B_j are related by auxiliary boundary conditions of the type

$$\sum_{j=1}^{2}\frac{B_j}{\exp(-\gamma-ik_{z_j}a)-1}=0. \tag{55}$$

It is shown by Philpott[16,18] that, for these models and in the case of oblique incidence of light, the separation of the dependences of the field and polarization on x and y in the form $\exp(i\mathbf{k}_\tau\cdot\mathbf{r})$ gives the one-dimensional problem discussed above.

In comparison with the auxiliary boundary conditions (55), our formulas have to be simplified by assuming that $s=1$. We then find from Eqs. (4) and (5) that $a_{0\nu}^{(l)}=a_{01}^{(l)}=G^{-3/2}$, and from Eq. (29) we find that $\mathbf{P}_l=\mathbf{P}_1$ is independent of $\mathbf{k}$. In case A, we find that $e^{-\gamma}\ll 1$ and Eq. (55) becomes

$$\sum_{j=1}^{2}B_j=0, \tag{56}$$

which is identical with Eq. (42), i.e., it gives Eq. (20). In case B, we have $\gamma\ll 1$ and Eq. (55) becomes

$$\sum_{j=1}\frac{B_j}{\gamma+ik_{z_j}a}=0\,. \tag{57}$$

This is identical with Eq. (46) if we bear in mind that, in our case, the interaction between the layers decreases proportionally to $\exp\{-|\mathbf{k}_\tau||m_3-m_3'|a\}$,[3)] i.e., $\gamma=|\mathbf{k}_\tau|a$. The quantity γ in the papers of Mead[15] and Sipe and Van Kranendonk[17] is an arbitrary specified parameter of the model, whereas in the present paper the value of γ is found by considering the Coulomb interaction of the particles and depends on the angle of incidence of light. However, this difference does not minimize the importance of the agreement of the results, which shows that, in case B, the "exponential model" is close to reality.

In the case of other models postulating point dipoles[8,16,18] it is usual to introduce a real dipole–dipole interaction between the lattice sites but the problem

is solved approximately: the interaction between monomolecular layers separated from one another by distances exceeding $\mathfrak{N}a$ ($\mathfrak{N}$ approximation) is ignored. All these treatments give an unacceptable result: even when an infinite crystal is considered, it is found that the number of waves is $N=\mathfrak{N}+1$, i.e., the number of roots of the dispersion equation rises without limit on increase in the precision of the calculation when allowance is made for the interactions of more and more distant layers. This result is in conflict with the exact "exponential model" calculations and with the results of the present and other authors on the theory of additional light waves in an infinite crystal, according to which we have $N=2$ for nondegenerate excitons.

The paradoxical result just reported arises because, when the dispersion (or other equation) contains a function of an unknown, expansion of this function as a series and retention of a finite number of terms give rise to real as well as fictitious roots which have nothing in common with the solutions of the original equation. These roots are fictitious because, for the appropriate values of the unknown, the rejected residue of the series is large. For example, for the equation $e^x=1.1$, the exact root is $\bar{x}=0.0953\ldots$. If the exponential is expanded as a linear series in x, the root of this equation is $x_1=0.1$, which is an approximation to $\bar{x}$. If the expression is quadratic, the roots are $x_1'=0.0954\ldots$ and $x_2'=2.0954\ldots$. The root x_1' is an even better approximation to $\bar{x}$ but x_2' is fictitious. In a cubic expansion of the exponential function, we obtain three roots, two of which are fictitious, and so on. The greater the number of terms retained in the expansion, the greater is the number of the fictitious roots.

We can show that the above $\mathfrak{N}$ approximation is equivalent to the expansion of the dispersion equation as a series and that only two roots among $\mathfrak{N}+1$ have physical meaning and the rest are fictitious.

However, if $\mathfrak{N}=1$, there are no fictitious roots in the dispersion equation and the results reported in some of these papers[8,16,18] are valid. These results apply to the limiting case A. All these papers give one auxiliary boundary condition which is identical with Eq. (20).

We shall conclude by noting that our treatment ignores distortions of the crystal lattice in the surface layer. In some cases, these distortions may give rise to surface excitons[11] and may greatly alter the auxiliary boundary conditions.[20,21]

[1)] All the optical properties of a gas molecule are known to be described completely by specifying the system of its stationary wave functions. A bounded crystal can be regarded as a large molecule.

[2)] Allowance for the terms $\mathcal{K}_{\mu\nu\mathbf{n}'}$ in the nearest coordination spheres presents no difficulty but does not alter the results basically.

[3)] This is proved in the same way as Eq. (26).

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