Exponential model in the theory of spatial dispersion. Boundary conditions

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A microscopic model is considered of an excitonic dielectric consisting of oscillators at crystal-lattice sites, the interaction between which decreases exponentially with distance. Account is taken here of the influence of the surface on the constants of the interaction between the oscillators. This model is used to construct a macroscopic phenomenological theory of an excitonic dielectric. An integral equation is obtained, describing the coupled oscillations of a semi-infinite chain of oscillators. A boundary problem equivalent to this integral equation is formulated. In particular, a boundary condition is obtained, which must be superimposed on the polarization. It is inhomogeneous in this model. The solution of the problem of light reflection from the considered semi-infinite dielectric is solved. The coefficients of reflection and transmission of light through the boundary of the medium are obtained. It is shown that the obtained reflection and transmission coefficients satisfy the energy conservation law.

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1. INTRODUCTION

There are a number of papers[1-8] in which an excitonic dielectric is regarded as an aggregate of oscillators situated at the sites of a crystal lattice and interacting with one another. In such models the analysis is usually limited to the interaction of the nearest pairs of neighbors,[2,3] or to N nearest neighbors.[4,5] However, the question of the interaction of light with the oscillator model can be solved exactly if the interaction is not confined to the N neighbors, but decreases exponentially with distance between the oscillators.[5-8] The authors of Refs. 5 and 6 obtained the dispersion relations and light-reflection coefficients in this model. However, no account was taken by them of the influence of the surface on the constants of the interaction between the oscillators, and the boundary condition was obtained for a particular solution of the equations and not in general form. Reference 8 suffers from this same shortcoming.

In the present paper the modeling question is supplemented and takes phenomenologically into account the influence of the surface on the interaction constants. Within the framework of this model, a microscopic analysis is made, on the basis of which a macroscopic theory of light interaction with a semi-infinite crystal is obtained. The boundary-value problem is correctly formulated and, in particular, an additional boundary condition is obtained. For simplicity we consider the one-dimensional problem. As the final result, the coefficients of reflection and transmission are obtained for the case of normal incidence of light on the boundary of the crystal, and it is shown that they satisfy the energy conservation law.

2. FORMULATION OF PROBLEM

We consider a crystal in each site of which are located oscillators that interact with one another, with natural frequency ω_{0n} that depends on n. We confine ourselves here to a one-dimensional lattice. It is known[9] that the problem of the three-dimensional cubic lattice can be reduced to one-dimensional. The equations that describe the oscillations of the polarization in such a lattice are of the form

$$\ddot{p}_n=-\omega_{0n}^2 p_n-\sum_{n'=1}^{\infty} V_{nn'}(p_n-p_{n'})+\frac{e^2}{m}E_n, \tag{1}$$

where p_n is the dipole moment of the oscillator in the site with index n, ω_{0n} is the natural frequency of the oscillator in the site n, E_n is the field acting on the oscillator, and $V_{nn'}$ are the constants of the interaction, which in an infinite lattice would depend on the difference $n-n'$ alone.

When account is taken of the nearest neighbors, it is assumed that all $V_{nn'}=0$, with the exception of those for which $|n-n'|=1$.[2] Allowance for the boundary in this model manifests itself only in a change of V_{12} for one edge cell. However, if all the $V_{nn'}$ differ from zero, then in principle the presence of the boundary should perturb only $V_{nn'}$ in the chain. In the model with exponential interaction, considered in Ref. 5–7 it was assumed that $V_{nn'}=ge^{-\gamma|n-n'|}$, where g and γ are parameters of the model. It is clear, however, that this representation is valid only when the force constants do not change in the medium in which they act. Otherwise, besides the direct interaction of the oscillators, there should appear an additional interaction due to image force and then the interaction constants take the form

$$V_{nn'}=g[e^{-\gamma|n-n'|}+re^{-\gamma(n+n')}], \tag{2}$$

where r is a phenomenological constant characterizing the properties of the boundary that separates the media in our model. In the particular case $r=0$ we obtain the model considered in Refs. 5–8. In addition, we assume that the natural frequencies ω_{0n} are constant in the volume of the medium and are perturbed near the boundary, and that this perturbation is described by the same parameter r:

$$\omega_{0n}^2=\omega_0^2+\frac{1-re^{-\gamma}}{1-e^{-\gamma}}ge^{-\gamma n}. \tag{3}$$

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The last assumption is not obligatory and was chosen here only to simplify the manipulations that follow. In principle, the dependence of ω_{0n} on n can be arbitrary and to be able to write down the form of this dependence we must make the model more specific. Substituting (3) and (2) in (1) and carrying out partial summation, we obtain the equation

$$\ddot{p}_n+\sum_{n'=1}^{\infty} L_{nn'}p_n=\frac{e^2}{m}E_n, \tag{4}$$

in which the off-diagonal elements of the matrix $L_{nn'}$ are equal to $-V_{nn'}$, and the diagonal elements are of the form

$$L_{nn}=\omega_0^2+2ge^{-\gamma}/(1-e^{-\gamma})-gre^{-2\gamma n}.$$

Equation (4) can be solved by the method described in Ref. 3. Then r is connected with the parameter ξ introduced in that paper by the relation $r=(\xi+e^{\gamma})/(\xi+e^{-\gamma})$. We shall not do this, however, and change over in this stage to the continuous-medium approximation. We introduce for this purpose new parameters, the distance a between the oscillators, $\Gamma=\gamma/a, G=g/a, x=an$, and change over from (4) from summation to integration with respect to x. In addition, assuming that the time dependence is determined by the external field and takes the form $\propto e^{-i\omega t}$, we obtain an equation for the macroscopic polarization P:

$$\left(\omega_0^2-\omega^2+\frac{2G}{\Gamma}\right)P(x)-G\int_0^{\infty}K(x,x')P(x')dx'=\frac{\omega_p^2}{4\pi}E(x), \tag{5}$$

where ω_p is the "plasma" frequency and the kernel of the integral transformation takes the form

$$K(x,x')=\exp(-\Gamma|x-x'|)+r\exp(-\Gamma(x+x')).$$

We have obtained an inhomogeneous integral equation of second kind, which describes our system. It must be solved simultaneously with Maxwell's equations, which reduce in the one-dimensional case to a single equation

$$\partial^2E/\partial x^2+\varepsilon_0k_0^2E=4\pi k_0^2P, \tag{6}$$

where $k_0=\omega/c$.

We emphasize that in a system (5), (6) boundary conditions must be imposed only on Eq. (6). Equation (5) contains the necessary boundary conditions. In a number of cases it may turn out that it is more convenient to solve not integral but differential equations with boundary conditions, i.e., a boundary-value problem. To change over from Eq. (5) to the corresponding boundary-value problem, we proceed as follows. We apply to both sides of Eq. (5) the differential operator $\hat{U}_1=(\partial^2/\partial x^2-\Gamma^2)$. Since $\hat{U}_1K(x,x')=-2\Gamma\delta)x-x')$, the integral in (5) degenerates and the following equation holds for the region $x>0$:

$$\left(\omega_0^2-\omega^2+\frac{2G}{\Gamma}\right)\left(\frac{\partial^2}{\partial x^2}-\Gamma^2\right)P+2G\Gamma P=\frac{\omega_p^2}{4\pi}\left(\frac{\partial^2}{\partial x^2}-\Gamma^2\right)E. \tag{7a}$$

In contrast to (5), Eq. (7a) calls for specification of the boundary condition. To obtain it, we proceed as follows. We consider the function

$$f(x,x')=K(x,x')+\frac{1}{k_0T}\frac{\partial}{\partial x}K(x,x')$$
$$=\left[1-\frac{r+1}{r-1}\eta(x-x')\right]\exp(-\Gamma|x-x'|)-\frac{2r}{r-1}\exp(-\Gamma(x+x')),$$

where $T=\Gamma(r-1)/k_0(r+1)$; $\eta(x-x')=1$ at $x-x'>0$ and $\eta(x-x')=-1$ at $x-x'<0$. This function vanishes for all positive x' and at $x=0$, i.e., $f(x=0,x'>0)=0$. Noting this, we apply the operator

$$\hat{U}_2=\left(1+\frac{1}{k_0T}\frac{\partial}{\partial x}\right)$$

to both halves of (5) and put $x=0$ in the obtained equation. Then the integral in this equation vanishes identically for all functions $P(x)$ and $E(x)$, and we obtain the boundary condition

$$\left(\omega_0^2-\omega^2+\frac{2G}{\Gamma}\right)\left(P+\frac{1}{k_0T}\frac{dP}{dx}\right)\bigg|_{x=0}=\frac{\omega_p^2}{4\pi}\left(E+\frac{1}{k_0T}\frac{dE}{dx}\right)\bigg|_{x=0}. \tag{7b}$$

The differential equation (7a), jointly with the boundary condition (7), constitute a correct formulation of the boundary-value problem that is equivalent to the integral equation (5).

The boundary condition obtained by us is inhomogeneous and contains in the right-hand side both the value of the field E and the derivative of the field with respect to the coordinate. This possibility was already discussed in Ref. 1. The considered boundary condition was obtained for a concrete model. We note also that although the inhomogeneity of the boundary condition does reflect in this case the specifics of our model, we did not introduce in the latter any surface charges or currents. Therefore, in contrast to the case considered in Ref. 10, the surface in our model does not disturb Eq. (6), and the boundary conditions imposed on (6) remain the same as before, i.e., they stipulate continuity of the tangential components of the fields on the boundary.

3. THE DIELECTRIC CONSTANT

We rewrite the integral equation (5) in standard form

$$P(x)-\lambda\int_0^{\infty}K(x,x')P(x')dx'=f(x), \tag{8}$$

where

$$\lambda=\frac{G}{\omega_0^2-\omega^2+2G/\Gamma},\quad f(x)=\frac{\omega_p^2\lambda}{4\pi G}E(x).$$

Equation (8) has a continuous eigenvalue spectrum

$$\lambda(k)=(\Gamma^2+k^2)/2\Gamma, \tag{9}$$

where the wave vector k varies in the interval $0<k<\infty$. Corresponding to the eigenvalues (9) is a complete system of orthonormalized eigenfunctions

$$\psi_k(x)=(e^{-ikx}+R(k)e^{ikx})(2\pi R(k))^{-1/2}, \tag{10}$$

where $R(k)=(ik-k_0T)/(ik+k_0T)$. The functions (10) are the solutions of a homogeneous equation, i.e. of Eq. (8) in which $f(x)=0$, and describe "mechanical" excitons.[1] Their dispersion is given by the relation $\lambda(k)=\lambda$, which is shown by the dash-dot line in the figure. It is seen from the figure that in the considered model

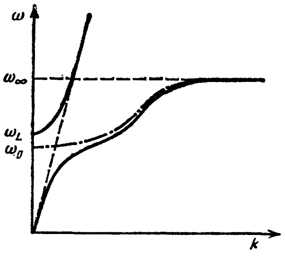

FIG. 1. Excitonic dispersion curves. The dash-dot curve shows the dispersion of the mechanical excitons. The solid curves show the dispersion of the excitonic polaritons.

the exciton band has a finite width even in the limit of a continuous medium, so that t as $k\to\infty$ the frequency tends to a finite limit $\omega^2\to\omega_\infty^2=\omega_0^2+2G/\Gamma_0$ determined by the parameters of the model. The effective mass of the exciton is expressed in terms of the same parameters $m_e^*=\hbar\omega_0\Gamma^3/2G$.

The solution of the inhomogeneous equation (8) can be written in the form

$$P(x)=f(x)+\lambda\int_0^\infty R(x,x',\lambda)f(x')\,dx', \tag{11}$$

where the resolvent $R(x,x',\lambda)$ of Eq. (8) is defined in terms of the eigenfunctions and eigenvalues of the homogeneous equation

$$R(x,x',\lambda)=\int_0^\infty\frac{\psi_k(x)\psi_k^*(x')}{\lambda(k)-\lambda}dk=\frac{\Gamma}{q}\Big[\exp(-q|x-x'|)+\frac{q+k_0T}{q-k_0T}\exp(-q(x+x'))\Big], \tag{12}$$

where $q=(\Gamma^2-2\Gamma\lambda)^{1/2}=\Gamma(\Delta\lambda/G)^{1/2}$ and $\Delta=\omega_0^2-\omega^2$. The integral in (12) can be easily calculated by residue theory. Account must be taken here of the fact that $|r|<1$ and $T<0$. Substituting (12) in (11) we can rewrite the solution of Eq. (8) in the form

$$P(x)=\int_0^\infty\chi(\omega,x,x')E(x')\,dx', \tag{13}$$

where

$$\chi(\omega,x,x')=\frac{\omega_p^2\lambda}{4\pi G}\Big\{\delta(x-x')+\frac{\lambda\Gamma}{q}\Big[\exp(-q|x-x'|)+\frac{q+k_0T}{q-k_0T}\exp(-q(x+x'))\Big]\Big\} \tag{14}$$

has the meaning of the nonlocal susceptibility of the considered medium. The nonlocal dielectric constant is connected with the susceptibility in the usual manner:

$$\varepsilon(\omega,x,x')=\varepsilon_0\delta(x-x')+4\pi\chi(\omega,x,x')=\Big(\varepsilon_0+\frac{\lambda\omega_p^2}{G}\Big)\delta(x-x')+\frac{\lambda^2\Gamma\omega_p^2}{Gq}\Big[\exp(-q|x-x'|)+\frac{q+k_0T}{q-k_0T}\exp(-q(x+x'))\Big], \tag{15}$$

where ε_0 is the "background" dielectric constant. For an infinite medium, the last term in this formula, which contains $x+x'$, vanishes and we can introduce the Fourier transform of the dielectric constant, in the form

$$\varepsilon(\omega,k)=\varepsilon_0+\omega_p^2(k^2+\Gamma^2)/[\Delta(k^2+\Gamma^2)+2Gk^2/\Gamma]. \tag{16}$$

4. REFLECTION AND TRANSMISSION COEFFICIENTS

We shall seek the solution of the system of equations (6) and (7) in the region $x>0$ in the form of a sum of two waves:

$$P(x)=\sum_{j=1}^2 P_je^{ik_jx},\qquad E(x)=\sum_{j=1}^2 E_je^{ik_jx}. \tag{17}$$

Substituting (17) in Eqs. (6) and (7a), we obtain a system of algebraic equations with respect to the amplitudes E_j and P_j:

$$(k_j^2-\varepsilon_0k_0^2)E_j=-4\pi k_0^2P_j, \tag{18a}$$

$$[\Delta+2Gk_j^2/\Gamma(\Gamma^2+k_j^2)]P_j=\omega_p^2E_j/4\pi, \tag{18b}$$

from which follows the dispersion relation for the polaritons

$$k_j^2=\varepsilon_0k_0^2+k_0^2\omega_p^2(k_j^2+\Gamma^2)/[\Delta(k_j^2+\Gamma^2)+2Gk_j^2/\Gamma]. \tag{19a}$$

At each fixed ω it has two solutions for k_j^2:

$$k_{1,2}^2=\frac{u-\Delta\Gamma^2\pm[(u+\Delta\Gamma^2)^2+8k_0^2\omega_p^2G\Gamma]^{1/2}}{2(\Delta+2G/\Gamma)}, \tag{19b}$$

where $u=k_0^2(\Delta\varepsilon_0+2G\varepsilon_0/\Gamma+\omega_p^2)$. In the frequency region $\omega_L^2<\omega^2<\omega_\infty^2$, where $\omega_L^2+\omega_0^2+\omega_p^2/\varepsilon_0$, the two wave vectors k_j are real. Outside this interval one is real and the other imaginary. The dispersion branches of the excitonic polaritons defined in (19b) are shown in the figure by solid lines.

Substituting (17) into the boundary condition (7b) and using (18b), we obtain an equation that connects the amplitudes of the two waves:

$$\sum_{j=1}^2 P_j\Big(\frac{1}{\Gamma+ik_j}-\frac{r}{\Gamma-ik_j}\Big)=0. \tag{20}$$

A relation equivalent to (20) with $r=0$ was obtained in Refs. 5, 6, and 8, and was called by their authors the supplementary boundary conditions. It is seen from the foregoing analysis, however, that relation (20) follows from the boundary condition (7b) only for a particular solution of (17). If, however, the solution differs from (17) in form, for example, at a definitely specified right-hand side of (5), then Eq. (20) is also changed. Equation (20) can therefore not be used in the general case as the boundary condition in the boundary-value problem. In addition (20) is valid in the entire volume of the medium, and not only on the boundary. It is clear therefore that (20) is not a boundary condition and constitutes in fact, jointly with (17) and (19), a solution of a boundary-value problem.

With the aid of (20) and the condition of continuity of the tangential components of the electric and magnetic fields on the boundary, we obtain in the usual manner the amplitude coefficients of reflection R_0 and transmission T_{01} and T_{02} of a light wave through the boundary of the medium:

$$R_0=\frac{1}{Q}(\alpha_1-\alpha_2-\alpha_1n_2+\alpha_2n_1),\qquad T_{01}=2\alpha_2/Q,\quad T_{02}=-2\alpha_1/Q, \tag{21}$$

where

$$Q=\alpha_1-\alpha_2+\alpha_1 n_2-\alpha_2 n_1, \quad \alpha_j=(k_0 T+ik_j)/\Phi_j,$$
$$\Phi_j=\Delta(\Gamma^2+k_j^2)+2Gk_j^2/\Gamma, \quad n_j=k_j/k_0.$$

5. ENERGY CONSERVATION LAW

We shall show that the obtained coefficients (21) satisfy the energy conservation law. To this end we assume that an electromagnetic wave of unit amplitude is incident on the boundary, and consider the energy fluxes propagating away from the boundary. We consider first the case $\omega_L^2<\omega^2<\omega_\infty^2$, when the two wave vectors in the medium are real. In the region $x<0$ the energy is carried away by a wave of amplitude R_0. The energy flux carried away by this wave is given by the Poynting vector $S_R=(c/8\pi)|R_0|^2$. Energy is carried into the interior of the medium by two waves with amplitudes T_{0j}, each of which consists of an electromagnetic-energy flux

$$S_j^{(0)}=\frac{cn_j}{8\pi}|T_{0j}|^2$$

and a mechanical-energy[1] flux

$$S_j^{(1)}=-\frac{\omega}{16\pi}\frac{\partial\varepsilon(\omega,k)}{\partial k}|T_{0j}|^2=\frac{\omega k_j}{8\pi}\frac{2G\Gamma\omega_p^2}{\Phi_j^2}|T_{0j}|^2. \tag{22}$$

Since we neglect the losses in our model, the incident energy flux should equal the sum of the energies of the waves that move away from the boundary. After cancelling the factor $c/8\pi$, this condition takes the form

$$|T_{01}|^2n_1\psi_1+|T_{02}|^2n_2\psi_2+|R_0|^2=1, \tag{23}$$

where $\psi_j=(1+2G\Gamma\omega_p^2k_0^2/\Phi_j^2)$. Substituting in this relation the formulas for the amplitudes and multiplying both sides by the common denominator $|Q|^2$, we get

$$4|\alpha_2|^2\psi_1+4|\alpha_1|^2\psi_2+|\alpha_1-\alpha_2-\alpha_1n_2+\alpha_2n_1|^2=|\alpha_1-\alpha_2+\alpha_1n_2-\alpha_2n_1|^2.$$

Expanding the brackets and dividing both halves of the equation by four:

$$|\alpha_2|^2\psi_1+|\alpha_1|^2\psi_2=(\alpha_1'-\alpha_2')(\alpha_1'n_2-\alpha_2'n_1)+(\alpha_1''-\alpha_2'')(\alpha_1''n_2-\alpha_2''n_1),$$

where $\alpha_j'=T/\Phi_j$, $\alpha_j''=k_j/\Phi_j$. We gather like terms:

$$2G\Gamma k_0^2\omega_p^2(|\alpha_2|^2k_1/\Phi_1^2+|\alpha_1|^2k_2/\Phi_2^2)=-(n_1+n_2)(\alpha_1'\alpha_2'+\alpha_1''\alpha_2'').$$

Substituting here the expressions for the real and imaginary parts of α_j and carrying the necessary cancellations, we obtain the identity

$$-2G\Gamma\omega_p^2k_0^2=\Phi_1\Phi_2, \tag{24}$$

which can be easily proved by substituting in the right-hand side the expressions for Φ_1 and Φ_2 with allowance for the formulas for the wave vectors k_j^2. A proof of an analogous identity for the model with nearest-neighbor interaction is given in Ref. 10. Thus, in the case when two waves propagate in the medium, the energy conservation law is satisfied.

We consider now the case when the frequency ω lies outside the frequency interval $[\omega_L, \omega_\infty]$. In this case one of the wave vectors, say k_2, is imaginary, $k_2=i\varkappa_2$, and the energy flux in the interior of the medium is produced only by one wave. The equality of the energy fluxes is then given by

$$|T_{01}|^2n_1\psi_1+|R_0|^2=1. \tag{25}$$

We substitute here the formulas for the amplitudes of the waves and multiply both halves by $|Q|^2$:

$$4|\alpha_2|^2n_1\psi_1+|\alpha_1-\alpha_2-i\varkappa_2\alpha_1+n_1\alpha_2|^2=|\alpha_1-\alpha_2+i\varkappa_2\alpha_1-n_1\alpha_2|^2.$$

We expand the brackets, divide both sides by four, and recognize that α_2 is real in this case:

$$2G\Gamma\omega_p^2k_0^2\alpha_2n_1=\Phi_1^2(\varkappa_2\alpha_1''-n_1\alpha_1').$$

Substituting here the expressions for α_j and making the necessary cancellations, we obtain again the identity (24), which is valid also for imaginary k_2. We see therefore that the energy conservation law is satisfied at all frequencies ω.

6. CONCLUSION

We see that the model considered above permits formulation of an exactly solvable problem, and from this point of view it is of definite interest and is worthy of further study. The analysis presented here shows that the equations that describe a given medium, and in particular the supplementary boundary conditions, can be obtained only from a microscopic model of the medium and of its boundary layer. At the same time, it is also possible to construct a phenomenological macroscopic theory, which proves to be more convenient for actual calculations. The exponential model, in contrast to the model with the nearest-neighbor interaction, is of the two-parameter type. The spatial dispersion is characterized in it not only by the effective mass $m_e^*=\hbar\omega_0\Gamma^3/2G$ but also by the width of the exciton band $\omega_\infty^2-\omega_0^2=2G/\Gamma$. In this sense, the model describes more accurately the process of light reflection by a resonant medium and it is possible that for crystals in which the width of the band is comparable with the longitudinal-transverse splitting it leads to better agreement between theory and experiment than the effective-mass approximation.

[1]V. M. Agranovich and V. L. Ginzburg, Kristallooptika s uchetom prostranstvennoi dispersii i teoriya éksitonov (Spatial Dispersion in Crystal Optics and the Theory of Excitons), Nauka, 1979.
[2]V. M. Agranovich and V. I. Yudson, Opt. Commun. **7**, 121 (1973).
[3]C. W. Deutsche and C. A. Mead, Phys. Rev. **138**, A63 (1965).
[4]C. D. Mahan and G. Obermair, Phys. Rev. **183**, 834 (1969).
[5]C. A. Mead, Phys. Rev. B **15**, 519 (1977).
[6]J. E. Sipe and J. Van Kranendonk, Can. J. Phys. **53**, 2095 (1975).
[7]O. Litzman and V. Holy, Phys. Status Solidi B **93**, 135 (1979).
[8]S. I. Pekar, Zh. Eksp. Teor. Fiz. **74**, 1458 (1978) [Sov. Phys. JETP **47**, 762 (1978)].
[9]C. A. Mead and M. R. Philpott, Phys. Rev. B **17**, 914 (1978).
[10]N. N. Akhmediev and V. V. Yatsyshen, Solid State Commun. **27**, 357 (1978).

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