INTERBAND OPTICAL TRANSITIONS IN LOW MOBILITY SEMICONDUCTORS

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Optical phenomena connected with the transition of electrons from inner shells to a low-mobility band are investigated. A graph technique for the calculation of the complex conductivity, which is applicable to strong electron-phonon interaction, is developed. Just as in the F-center problem, the main absorption curve peak has a maximum at a distance on the order of $\omega_p = \Delta E_p/\hbar$ (ΔE_p is the polaron shift) from the absorption band edge. The peak width $\delta\omega$ is determined exclusively by the nature of the transition ($\delta\omega \sim (kT\Delta E_p/\hbar^2)^{1/2}$ for $T > T_0$ and $\delta\omega \sim (\omega_0 \Delta E_p/\hbar)^{1/2}$ for $T < T_0$) and is in no way related to the width of the polaron band or of the initial electron band (for which the polaron effect is not taken into account). When $T < T_0$ two additional peaks are observed at the edge of the absorption band, the distance between them being ω_0. The low frequency peak has a Lorentz shape and its width is determined by the relaxation time in the transport equation for small-radius polarons. The shape of the second peak is not a Lorentz one. At low temperatures the final height of the peak is determined by the possibility of decay of an optical phonon into two acoustic phonons, and hence is not related to the characteristics of the polaron or initial electron bands. The refractive index possesses singularities (of the discontinuity type) in the same frequency ranges.

INTRODUCTION

THE static electric conductivity of semiconductors with low mobility was recently considered in several theoretical papers[1-3], where it was established that the observed activation growth of mobility with increasing temperature can be the consequence of the strong interaction between the carriers and polarization vibrations (in semiconductors of this type, for example in NiO and in Fe_2O_3, the bond has a pronounced ionic character). In the present paper we consider optical phenomena in semiconductors of this type, connected with transitions from a deep atomic level into a band in which the carriers have low mobility.

We shall calculate here the contribution $\Delta\epsilon(\omega)$ made to the complex dielectric constant of the medium by the processes indicated above. The total dielectric constant $\epsilon(\omega)$ can be represented in the form $\epsilon(\omega) = \bar{\epsilon}(\omega) + \Delta\epsilon(\omega)$, $\epsilon = \epsilon' + i\epsilon''$ where $\bar{\epsilon}$ includes all the other mechanisms. The absorption and reflection coefficients $K(\omega)$ and $n(\omega)$ are expressed in terms of the real and imaginary parts ϵ' and ϵ'' of ϵ:

$$K(\omega) = \frac{\omega}{c}\varkappa, \quad \varkappa(\omega) = \left\{\frac{1}{2}[-\varepsilon' + (\varepsilon'^2 + \varepsilon''^2)^{1/2}]\right\}^{1/2},$$

$$n(\omega) = \left\{\frac{1}{2}[\varepsilon' + (\varepsilon'^2 + \varepsilon''^2)^{1/2}]\right\}^{1/2}.$$

In the case of weak absorption (when $\epsilon''/\epsilon' \ll 1$) we have $\kappa \approx \epsilon''/2\sqrt{\epsilon'}$ and $n = \sqrt{\epsilon'}$. In the frequency region where absorption due to the mechanism in question is significant, $\Delta\epsilon$ experiences abrupt changes. If the remaining mechanisms do not produce noticeable absorption in this frequency region, then $\bar{\epsilon}$ varies smoothly, and since $\bar{\epsilon}'' \approx 0$ the value of κ is determined by $\Delta\epsilon''$, viz., $\kappa = \Delta\epsilon''/2\sqrt{\epsilon'}$. If furthermore $\Delta\epsilon'/\bar{\epsilon}' \ll 1$, then the refractive index will be of the form $n = \bar{n} + \Delta n$, where $\bar{n} = (\bar{\epsilon}')^{1/2}$ and $\Delta n = \Delta\epsilon'/2\bar{n}$, with $\bar{n}$ varying weakly in the region under consideration.

Thus, calculation of $\Delta\epsilon$ enables us to obtain the values of K and Δn, which can be measured experimentally. It is more convenient to calculate the corresponding addition $\Delta\sigma$ to the complex conductivity of the medium. $\Delta\epsilon$ and $\Delta\sigma$ are connected by the simple relation $\Delta\epsilon = 4\pi i\Delta\sigma/\omega$.

1. FORMULATION OF THE PROBLEM

Let us consider the optical transitions from a deep electron level, the band width of which can be neglected, into a narrow band (we shall henceforth refer simply to the level and to the band). It is assumed that the level and the band have been determined in the single-electron approximation. If the semiconductor contains polarization phonons that interact strongly with the electrons, the single-electron states do not constitute a good

initial system of functions, for the polarization of the lattice by the electron becomes appreciable by virtue of the strong electron-phonon interaction (polaron effect).

For simplicity we neglect the interaction between the phonons and the electrons at the level. The band is assumed to be narrow, so that we deal with a polaron of small radius, i.e., $J/\Delta E_p < 1$, where J—exchange integral between the nearest neighbors (which determines the width of the single-electron band), and ΔE_p—polaron level shift[1]. The distance between the bottom of the band and the level is denoted $\hbar\Omega_0$; we shall henceforth confine ourselves to the case $\hbar\Omega_0/kT \gg 1$.

The Hamiltonian of the electron-phonon system under consideration is of the form

$$\mathcal{H} = \sum_{\alpha m} \mathcal{E}_\alpha a_{\alpha m}^+ a_{\alpha m} + \sum_{mg} J(g)\, a_{1m}^+ a_{1m+g} + \sum_q \hbar\omega_q b_q^+ b_q + N^{-1/2} \sum_{qm} \hbar\omega_q (\gamma_q e^{iqR_m} b_q^+ + \gamma_q^* e^{-iqR_m} b_q)\, a_{1m}^+ a_{1m};$$

$$|\gamma_q|^2 = \frac{2^{3/2}\pi e^2}{q^2 d^3 \hbar\omega_0} (\varepsilon_\infty^{-1} - \varepsilon_0^{-1}) \quad \text{for } qd \ll 1; \tag{1}$$

only the interaction with the polarization phonons is taken into account, $\mathcal{E}_0 = 0$, $\mathcal{E}_1 = \hbar\Omega_0$, ϵ_∞ and ϵ_0 are respectively the electronic and static dielectric constants, the summation over m is summation over all the lattice sites, R_m—corresponding lattice vector, summation over g denotes summation over the nearest neighbors of the given site, d^3—volume of unit cell, ω_0—limiting frequency of the polarization phonon, q—phonon wave vector, α takes on values 0 and 1 for the level and for the band respectively, and J(g)—exchange integral between the neighboring lattice sites. Only terms diagonal in the site number are retained in the Hamiltonian of the interaction between the electrons and the phonons[2]. The interaction of electrons of species 1 ($\alpha = 1$) is assumed strong:

$$\bar{\gamma} = N^{-1} \sum_q |\gamma_q|^2 \gg 1.$$

Let us calculate the complex conductivity $\sigma(\omega)$, using the Kubo formula

$$\sigma(\omega) = V^{-1} \int_0^\infty e^{-i\omega t}\, dt \int_0^\beta \langle j(-i\hbar\lambda)\, j(t)\rangle\, d\lambda \tag{2}$$

($V = Nd^3$—volume of crystal, j—current operator, $\beta = 1/kT$). Formula (2) is best rewritten in a form that does not contain the double integral:

$$\sigma = -(V\hbar\omega)^{-1} \int_0^\infty e^{-i\omega t} \langle j(t)\, j(0) - j(0)\, j(t)\rangle\, dt + i(V\omega)^{-1} \int_0^\beta \langle j(-i\hbar\lambda)\, j(0)\rangle\, d\lambda, \tag{3}$$

which can be readily verified by writing the correlators in terms of the matrix elements of the operators in the total-Hamiltonian eigenfunction representation.

Inasmuch as we are considering transitions from the level to the band, we retain in the current operator only the terms responsible for these transitions. In addition, we assume the dipole transition $0 \to 1$ to be allowed. The corresponding contribution to the current operator is

$$j' = \sum_m (j_{0m;\,1m}\, a_{0m}^+ a_{1m} + j_{1m;\,0m}\, a_{1m}^+ a_{0m}). \tag{4}$$

Then the correlator $\langle j'(t)\, j'(0)\rangle$, which is contained in (3), is of the form

$$\langle j'(t)\, j'(0)\rangle = |j_{0m;\,1m}|^2 \sum_m [\langle a_{1m}(t)\, a_{1m}^+\rangle \langle a_{0m}^+(t)\, a_{0m}\rangle + \langle a_{0m}(t)\, a_{0m}^+\rangle \langle a_{1m}^+(t)\, a_{1m}\rangle]. \tag{5}$$

In (5) we take account of the fact that

$$\langle a_{0m}(t)\, a_{0m}\rangle = \langle a_{0m}^+(t)\, a_{0m}^+\rangle = 0.$$

Noting that

$$\langle a_{0m}^+(t)\, a_{0m}\rangle = \langle \hat{n}_0\rangle, \quad \langle a_{0m}(t)\, a_{0m}^+\rangle = \langle 1 - \hat{n}_0\rangle$$

(inasmuch as we are referring everything to the lower level, $a_{0m}(t) = a_{0m}$, since $\hat{\mathcal{E}}_0 = 0$), and recognizing that practically all the lower levels are filled, we can assume that $\langle \hat{n}_0\rangle = 1$ and $\langle 1 - \hat{n}_0\rangle = 0$. Then only the first term remains in (5), and we obtain

$$V^{-1} \langle j'(t)\, j'(0)\rangle = V^{-1} |j_{0m;\,1m}|^2 \sum_m \langle a_{1m}(t)\, a_{1m}^+\rangle = n_0 |j_{0m;\,1m}|^2 \langle a_{10}(t)\, a_{10}^+\rangle, \quad n_0 = NV^{-1}. \tag{6}$$

Account is taken in (6) of the fact that $\langle a_{1m}(t)\, a_{1m}^+\rangle$ does not depend on m, by virtue of the translation symmetry of the lattice[2].

In order to include the principal part of the interaction between the electrons and the phonons in the zero-order Hamiltonian, we make use of the canonical transformation[2]

$$\tilde{\mathcal{H}} = e^S \mathcal{H} e^{-S}, \quad S = N^{-1/2} \sum_{qm} (b_q^+ u_{qm} - b_q u_{qm}^*)\, a_{1m}^+ a_{1m}; \quad u_{qm} = \gamma_q \exp(iqR_m). \tag{7}$$

[1] We assume ΔE_p to be small compared with the width of the forbidden band; in this case the polaron effect does not lead to an entanglement of the initial single-electron states.

[2] If the dipole transition is forbidden, then $\langle j(t)j\rangle$ will have also the form (6), but in place of $|j_{0m;\,1m}|^2$ we shall have $|j_{0m;\,1m+g}|^2$. In this case indirect transitions with participation of an intermediate electron level, the transitions to which are allowed[4], may become important.

The electron operators are transformed in accordance with the law

$$\tilde{a}_{1\mathbf{m}} = a_{1\mathbf{m}} \exp\left[-N^{-1/2}\sum_{\mathbf{q}}(b_{\mathbf{q}}^{+}u_{\mathbf{qm}} - b_{\mathbf{q}}u_{\mathbf{qm}}^{*})\right],$$

$$\tilde{a}_{1\mathbf{m}}^{+} = a_{1\mathbf{m}}^{+} \exp\left[N^{-1/2}\sum_{\mathbf{q}}(b_{\mathbf{q}}^{+}u_{\mathbf{qm}} - b_{\mathbf{q}}u_{\mathbf{qm}}^{*})\right]. \qquad (8)$$

In the transformed Hamiltonian we go over to the k-representation

$$a_{1\mathbf{m}} = N^{-1/2}\sum_{\mathbf{k}} a_{1\mathbf{k}} \exp(i\mathbf{k}\mathbf{R}_{\mathbf{m}}),$$

after which we obtain

$$\tilde{\mathcal{H}} = \sum_{\mathbf{k}}[\hbar\Omega + \mathcal{E}(\mathbf{k})]\, a_{\mathbf{k}}^{+}a_{\mathbf{k}} + \sum_{\mathbf{q}}\hbar\omega_{\mathbf{q}}b_{\mathbf{q}}^{+}b_{\mathbf{q}} + \sum_{\mathbf{k}_1\mathbf{k}_2} a_{\mathbf{k}_1}^{+}a_{\mathbf{k}_2}$$
$$\times\left\{-\mathcal{E}(\mathbf{k}_1)\,\delta_{\mathbf{k}_1\mathbf{k}_2} + \frac{1}{N}\sum_{\mathbf{mq}} J(\mathbf{g})\exp[i\mathbf{k}_2\mathbf{R}_{\mathbf{m}} - i\mathbf{k}_1(\mathbf{R}_{\mathbf{m}}+\mathbf{g})\right.$$
$$\left. + N^{-1/2}\sum_{\mathbf{q}}(b_{\mathbf{q}}^{+}\Delta u_{\mathbf{qm};\,m+g} - b_{\mathbf{q}}\Delta u_{\mathbf{qm};\,m+g}^{*})]\right\}$$
$$\equiv \tilde{\mathcal{H}}_{0e} + \tilde{\mathcal{H}}_{0ph} + \tilde{\mathcal{H}}_{int}, \quad \hbar\Omega = \hbar\Omega_0 - \Delta E_p, \qquad (9)$$

where

$$\Delta E_p = \frac{1}{N}\sum_{\mathbf{q}}|\gamma_{\mathbf{q}}|^2\hbar\omega_{\mathbf{q}}$$

is the polaron shift and $\Delta u_{\mathbf{qm};\,m+g} = u_{\mathbf{qm}+\mathbf{g}} - u_{\mathbf{qm}}$; we shall henceforth leave out everywhere the index $\alpha = 1$ in the operators $a_{1\mathbf{m}}$, $a_{1\mathbf{k}}$. The electronic part of the Hamiltonian $\mathcal{H}_{0e}$ contains an additional quantity (which is simultaneously subtracted from the electron-formula interaction)[2]

$$\mathcal{E}(k) = \sum_{g} e^{-ikg} J(\mathbf{g})\, e^{-S_T},$$

$$e^{-S_T} = \langle\exp\left[N^{-1/2}\sum_{\mathbf{q}}(b_{\mathbf{q}}^{+}\Delta u_{\mathbf{qm};\,m+g} - b_{\mathbf{q}}\Delta u_{\mathbf{qm};\,m+g})\right]\rangle.$$

After the transformation (7), $\langle a_0(t)\, a_0^{+}(0)\rangle$ assumes the form

$$\langle a_0(t)\exp[-N^{-1/2}\sum_{\mathbf{q}}(b_{\mathbf{q}}^{+}(t)\,u_{\mathbf{q}0} - b_{\mathbf{q}}(t)\,u_{\mathbf{q}0}^{*})]$$
$$\times a_0\exp[N^{-1/2}\sum_{\mathbf{q}}(b_{\mathbf{q}}^{+}u_{\mathbf{q}0} - b_{\mathbf{q}}u_{\mathbf{q}0}^{*})]\rangle, \qquad (10)$$

where the averaging $\langle\ldots\rangle$ is over a Gibbs distribution with transformed Hamiltonian $\tilde{\mathcal{H}}$.

Following the canonical transformation (7), the "residual" electron-phonon interaction $\tilde{\mathcal{H}}_{int}$ becomes proportional to a small exchange integral J. When J = 0, the present problem is completely analogous to the problem of the optical properties of F-centers[5,6]. Thus the formula for $\Delta\sigma$ is of the form

$$\Delta\sigma(\omega) = \Lambda\frac{\Omega\omega_0}{\omega}\left[\int_0^{\infty} e^{-i\omega t}\langle\tilde{a}_0(-t)\,\tilde{a}_0^{+}\rangle\, dt\right.$$
$$\left. - \int_0^{\infty} e^{-i\omega t}\langle\tilde{a}_0(t)\,\tilde{a}_0^{+}\rangle\, dt + i\hbar\int_0^{\beta}\langle\tilde{a}_0(-i\hbar\lambda)\,\tilde{a}_0^{+}\rangle\, d\lambda\right],$$

$$\Lambda = \frac{e^2 n_0 f_{01}}{2m\omega_0} \qquad (11)$$

(we have introduced here the dimensionless oscillator strength $f_{01} = 2m\,|j_{01}|^2/e^2\hbar\Omega$).

2. EXPANSION OF THE CORRELATOR IN POWERS OF $\tilde{\mathcal{H}}_{int}$

We confine ourselves to zeroth order terms in the electron density in the band, i.e., we put $n_1 = \langle a_{1\mathbf{m}}^{+}a_{1\mathbf{m}}\rangle \ll 1$. We can then leave out the term with $\tilde{\mathcal{H}}_{int}$ in the Gibbs density matrix and in the left bracketing exponential of the Heisenberg operator in (11)[3]. It is sufficient to calculate one of the correlators (11), for example $\langle\tilde{a}_0(t)\,\tilde{a}_0^{+}\rangle$. Expanding the right bracketing exponential in a series in $\tilde{\mathcal{H}}_{int}$ and replacing everywheve $\exp(i\,\mathcal{E}i\tau/\hbar)$ by unity[2], we obtain

$$\langle\tilde{a}_0(t)\,a_0^{+}\rangle = e^{-i\Omega t}\sum_{n=0}^{\infty}\left(-\frac{iJ}{\hbar}\right)^n\frac{1}{N}\sum_{\mathbf{k};\mathbf{g}_1\ldots\mathbf{g}_n} e^{i\mathbf{k}(\mathbf{g}_1+\ldots+\mathbf{g}_n)}\int_0^{t} d\tau_n$$
$$\ldots\int_0^{\tau_2} d\tau_1\,\langle P_0(t)\,[P_{\mathbf{g}_1+\ldots+\mathbf{g}_n}^{+}(\tau_n)\,P_{\mathbf{g}_2+\ldots+\mathbf{g}_n}(\tau_n) - e^{-S_T}]$$
$$\times[P_{\mathbf{g}_2+\ldots+\mathbf{g}_n}(\tau_{n-1})\,P_{\mathbf{g}_3+\ldots+\mathbf{g}_n}(\tau_{n-1}) - e^{-S_T}]$$
$$\ldots[P_{\mathbf{g}_n}^{+}(\tau_1)P_0(\tau_1) - e^{-S_T}]\,P_0^{+}(0)\rangle. \qquad (12)$$

Here*

$$P_{\mathbf{R}}^{+} = \exp[N^{-1/2}\sum(b_{\mathbf{q}}^{+}u_{\mathbf{qR}}e^{i\omega_{\mathbf{q}}\tau} - b_{\mathbf{q}}u_{\mathbf{qR}}^{*}e^{-i\omega_{\mathbf{q}}\tau})],$$
$$P_{\mathbf{R}}(\tau) = \exp[-N^{-1/2}\sum(b_{\mathbf{q}}^{+}u_{\mathbf{qR}}e^{i\omega_{\mathbf{q}}\tau} - b_{\mathbf{q}}u_{\mathbf{qR}}^{*}e^{-i\omega_{\mathbf{q}}\tau})],$$
$$S_T = N^{-1}\sum|\gamma_{\mathbf{q}}|^2\,\mathrm{cth}\,\frac{\beta\hbar\omega_{\mathbf{q}}}{2}(1-\cos\mathbf{q}\mathbf{g}).$$

In the course of the derivation of (12) it is necessary to go over to the k-representation for the operators $a_{\mathbf{m}}$ and $a_{\mathbf{m}}^{+}$, and then sum over k with allowance for

$$\sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}} = N\delta_{\mathbf{k};0}.$$

The averaging of the phonon operators in (12) is carried out in the same manner as before[2]. The rules which enable us to write down in explicit form the n-th term of the expansion (12) are best formulated in graphic form: the n points corresponding to the intermediate times τ_i are arranged

3)Inasmuch as $\tilde{\mathcal{H}}_{int}$ contains Fermi operators in the order $a^{+}a$, while (11) contains them in the reverse order (aa^{+}), the account of $\tilde{\mathcal{H}}_{int}$ in these expressions would yield corrections $\sim n_1, n_1^2$, etc.

*cth = coth.

in accordance with their order in (12) on a horizontal straight line, the left end of which corresponds to the instant t and the end of which corresponds to zero (we call the left and the right points the terminals). Each internal point τ_i is set in correspondence with the vector g_i. We draw lines [4] interconnecting pairwise all the points of the diagram, including the terminals, with the exception of the lines joining a bracket with a bracket. Each line is assumed to be directed from the earlier instant of time to the later one. Each internal point corresponds to a factor $(-iJ/\hbar)\exp(ik\cdot g_i - S_T)$. Each line going from τ_s to τ_r ($s < r$) is set in correspondence with a factor $T_{rs}(\tau_r - \tau_s)$:

$$T_{rs}(\tau_r - \tau_s; g_r \dots g_s) = \exp\left[N^{-1}\sum_q \Gamma_q \cos\left[\omega_q(\tau_r - \tau_s + i\hbar\beta/2)\right] f(g_r \dots g_s; q)\right], \quad (13)^*$$

$$\Gamma_q = |\gamma_q|^2 \operatorname{csch}(\beta\hbar\omega_q/2);$$

$$f(g_r \cdots g_s; q) = \cos[q(g_r + g_{r-1} + \cdots + g_{s+1} + g_s)] - \cos[q(g_{r-1} + \dots + g_{s+1} + g_s)] - \cos[q(g_r + g_{r-1} + \dots + g_{s+1})] + \cos[q(g_{r-1} + \dots + g_{s+1})]. \quad (14)$$

Each line going from the internal point τ_s to the right terminal t is set in correspondence with a factor $F(t - \tau_s; g_s \dots g_n)$:

$$F(t - \tau_s; g_s \dots g_n) = \exp\left[\frac{1}{N}\sum_q \Gamma_q \cos\left[\omega_q(t - \tau_s + i\hbar\beta/2)\right] \chi(g_s \dots g_n; q)\right], \quad (15)$$

$$\chi(g_s \dots g_n; q) = \cos[q(g_{s+1} + \dots + g_n)] - \cos[q(g_{s+1} + \dots + g_n)]. \quad (16)$$

Each line going from the terminal 0 to the internal point τ_r is assigned a factor $F(\tau_r; g_1 \dots g_r)$.

If the term with $\mathcal{E}(k)$ in the zeroth Hamiltonian were not separated in (9), then the construction of the term of the n-th order would be determined by the rules indicated above (we call this the initial diagram). The separation of this term leads to the need for adding to the initial diagram $2^n - 1$ more diagrams of the same order, obtained by modification from the initial one [5]. The modified diagrams are formed from the initial diagram in the following fashion. We first form the n diagrams which are obtained from the initial diagram by removing all the phonon lines from each internal point τ_i (we call these diagrams with one empty point). The resultant n expressions are added and the sum multiplied by (-1). We then form the sum of all the diagrams which are obtained from the initial one by removing all the lines from any pair of internal points (diagrams with two empty points), and multiply it by $(-1)^2$. Continuing further, we form the sums of all the diagrams with 3, 4, ..., l, ..., n empty points, and multiply each sum respectively by $(-1)^3$, $(-1)^4$, ..., $(-1)^l$, ..., $(-1)^n$. All the obtained sums are added to the expression corresponding to the initial diagram. We integrate over the internal times in the sequence indicated in (12), sum over all the g_i, sum over k, and divide by N. We thus obtain the n-th term of the series, which is of the form

$$M^{(n)} = \frac{1}{N}\sum_k M_k^{(n)}(t).$$

Finally, in order to obtain (12) we must multiply

$$M(t) = \sum_{n=0}^{\infty} M^{(n)}(t)$$

by the expression Q(t), where

$$Q(t) = \exp[-S_T^{(0)} - i\Omega t + F_0(t)],$$

$$S_T^{(0)} = \frac{1}{N}\sum_q |\gamma_q|^2 \operatorname{cth}\frac{\beta\hbar\omega_q}{2};$$

$$F_0(t) = \frac{1}{N}\sum_q \Gamma_q \cos\left[\omega_q\left(t + \frac{i\hbar\beta}{2}\right)\right]. \quad (17)$$

By way of an example we shall write out the sum of the diagrams for $M^{(2)}$ (see Fig. 1). We note that in the zeroth approximation in $\tilde{\mathcal{H}}_{int}$ the correlator is given by the expression (17). Inasmuch as $F_0(t) \to 0$ as $t \to \infty$, the quantity (17) oscillates as $t \to \infty$ and its Fourier component will in any case have a δ-like singularity at $\omega = \Omega$ [and in the case of polarization phonons also a singularity of the type $(\omega - \Omega \pm \omega_0)^{-1/2}$ [6]]. Allowance for $\tilde{\mathcal{H}}_{int}$ leads to a decrease in the correlator as $t \to \infty$, and this leads to a "smearing out" of the singularities.

3. PARTIAL SUMMATION OF THE SERIES (12)

In view of the fact that T_{rs} and F tend to unity as $t \to \infty$ [7], the contribution of the initial diagram

4) These lines correspond to phonon convolutions for the case of multiphonon interaction.

5) The n-th order term in (12) is the result of averaging of n binomials; the first term of each binomial is the operator P+P, while the second is the c-number $\exp(-S_T)$. The initial diagram will correspond to a term with a product of all the first terms, while the modified diagrams are obtained when a certain number of second terms is encountered in the product; the total number of different terms will be 2^n.

*csch = cosech.

6) See Sec. 4 of the present paper.

7) More accurately, for $t \gg (\Delta\omega)^{-1}$, where $\Delta\omega$—phonon dispersion.

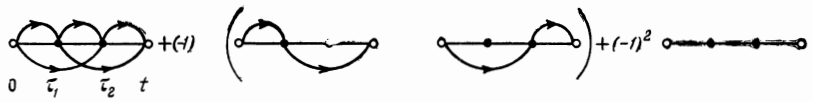

FIG. 1

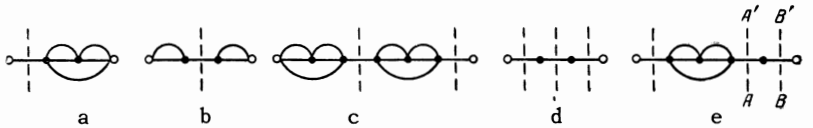

FIG. 2

to $M^{(n)}$ behaves as $t \to \infty$ like a polynomial of degree n in t, and accordingly its Fourier transform behaves like a polynomial of degree $n+1$ in ω^{-1}. (The complete expression for $M^{(n)}$, including all the modified diagrams, will be a polynomial of order lower than n, since the added modified diagrams cancel some of the coefficients of some lower powers of t [8], particularly t^n). To determine the behavior of N(t) as $t \to \infty$ it is necessary to sum an infinite number of terms of the series. In view of the complexity of $M^{(n)}$, such a summation cannot be made directly. However, if we put

$$\sum_{n=0}^{\infty} M^{(n)} = \sum (M^{(n)} - \mu^{(n)}) + \sum \mu^{(n)} = M_R + \mu,$$

where $\mu^{(n)}$ has a simpler structure than $M^{(n)}$ and is chosen such as to make $M^{(n)} - \mu^{(n)}$ decrease as $t \to \infty$, then we can confine ourselves in M_R to the lower terms of the expansions, and the summation of $\mu^{(n)}$ may be realizable. We present below a method of constructing $\mu^{(n)}$ for arbitrary n.

We shall assume that the diagram contains a free section (s; s + 1) if a vertical line drawn between the points τ_s and τ_{s+1} does not cross any phonon line (Fig. 2).

Such diagrams break up into several phonon blocks joined by free sections [9]. By a block is means a part of the diagram which does not contain free sections; blocks containing one of the terminals will be called vertices; blocks not containing any terminals will be called compact parts. Analysis shows that in order to obtain $\mu^{(n)}$ it is necessary to make up from the initial diagram all possible $M^{(n)}$ diagrams $\mu_i^{(n)}$ with free sections, with the exception of those where two free sections are separated by an empty point (Fig. 2e, sections AA′ and BB′). In each block of such a diagram, modification is carried out independently by forming empty points, the sum of all the diagrams with free sections, obtained from the given diagram, is then subtracted (within the block), each of the blocks of these diagrams is again subjected to a similar procedure, etc. Each diagram constructed in this fashion consists of blocks, the expressions for which tend to zero whenever any τ in the integrand of the given block tends to infinity. We can show that [10]

$$M^{(n)}(t) - \sum_i \mu_i^{(n)}(t) \to 0 \quad \text{as} \quad t \to \infty.$$

The series $\Sigma_n \Sigma_i \mu_i^{(n)}$ can be readily summed over n (in Fig. 3a the series is written in graphic form [11]), and its convolution into an integral equation is

$$\mu_k(t) = \int_0^t d\tau' \Phi_k(t-\tau')\,\pi_k(\tau'), \tag{18}$$

$$\pi_k(\tau) = \int_0^\tau d\tau' \Phi_k(\tau') + \int_0^\tau d\tau_2 \int_0^{\tau_2} d\tau_1 L_k(\tau_2 - \tau_1)\,\pi_k(\tau_1). \tag{19}$$

Here $\Phi_k(\tau)$—total vertex part, $L_k(\tau)$—total compact part (see Figs. 3b, c). Performing the Fourier transformation

$$\psi(\omega) = \int_0^\infty e^{i\omega t}\psi(t)\,dt,$$

we obtain

$$\pi_k(\omega) = i\Phi_k(\omega)[\omega - iL_k(\omega)]^{-1},$$

$$\mu_k(\omega) = \Phi_k(\omega)\pi_k(\omega). \tag{20}$$

Thus, the expression for the Fourier transform of the correlator is represented in the form of a sum of two expressions: $M_R(\omega)$ (which we call the Holstein term) and $\mu_k(\omega)$, which is expressed in

[8] For example, for $M^{(2)}(t)$ we have $M^{(2)}(t) \sim t$ as $t \to \infty$.

[9] Diagrams with free sections have the property that each free section in their Fourier transforms corresponds to a factor ω^{-1}, i.e., the singularities are present in simple form, making it possible to sum the series.

[10] To this end it is convenient to go over to the Fourier representation for $M^{(n)}(t)$ and make the substitution $z_1 = t + \tau_1 + \cdots + \tau_n$, $z_2 = \tau_1 + \cdots + \tau_n$, $\cdots$, $z_{n+1} = \tau_1$; integration over each z_i is between the limits $(0; \infty)$. Then the integrand tends to zero for any $z_i \to \infty$.

[11] We present for illustration the explicit form of the second diagram of Fig. 3a:

$$\int_0^t d\tau' \int_0^{\tau'} d\tau_2 \int_0^{\tau_2} d\tau_1 \int_{\tau'}^{\tau_1} \Phi(\tau) L(\tau_2 - \tau_1) \Phi(t-\tau')\,d\tau.$$

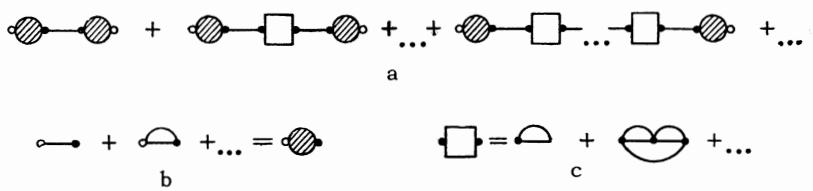

FIG. 3

terms of the values of Φ_k and L_k. The expressions for M_R, Φ_k, and L_k are power series in some parameters containing J [12]. We shall confine ourselves henceforth to the lowest term for the vertex $\Phi_k = 1$, and choose for L_k the largest term of the series [13]. Then

$$\mu_k(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{-i\omega t}\,[\omega - iL_k^{(4)}]^{-1}\,d\omega.$$

It is difficult to determine the dependence of $L^{(4)}$ on ω, but we can state that no abrupt changes occur in $L_k^{(4)}(\omega)$ in the region of frequencies that are not too large compared with ω_0.[15] We then get

$$\mu_k(t) \approx \exp(-w_0 t), \quad t > 0, \quad w_0 = L_k^{(4)}(0). \tag{21}$$

The quantity w_0 was calculated previously [2,3], namely:*

$$w_0 = (\eta_1^4\omega_0^2/\Delta\omega)\,\mathrm{sh}^{-2}(\beta\hbar\omega_0/2), \qquad \eta_1 = J/\Delta E_p.$$

We ultimately obtain for the correlator

$$\langle \tilde{a}_0(t)\,\tilde{a}_0^{+}\rangle = \begin{cases} \exp\left\{-S_T^{(0)} - i\Omega t - w_0 t + N^{-1}\sum_q \Gamma_q \cos\left[\omega_q(t+i\hbar\beta/2)\right]\right\}, & t>0 \\ 0, & t<0 \end{cases}. \tag{22}$$

The Holstein terms have been left out, since they are proportional to J^2.

4. CALCULATION OF THE COMPLEX CONDUCTIVITY

It is convenient to calculate $\Delta\sigma$ for $\omega > 0$. The values of $\Delta\sigma$ for $\omega < 0$ are obtained from the symmetry conditions

$$\Delta\sigma'(-\omega) = \Delta\sigma'(\omega), \quad \Delta\sigma''(-\omega) = -\Delta\sigma''(\omega).$$

When $\omega > 0$, the second and third terms of (11) do not contain singularities in the vicinity of $\omega \sim \Omega$, and the corresponding correlators can be taken in the zeroth approximation (17). The calculations lead to the following contribution to $\Delta\sigma$ (under the condition $\Delta E_p/\hbar\Omega \ll 1$):

$$i\Lambda\omega_0(2\Omega + \omega)/\Omega(\Omega + \omega). \tag{23}$$

Let us consider the first term in (11). When $w_0 = 0$ it becomes infinite at $\omega = \Omega$ and $\omega = \Omega \pm \omega_0$.[14] Therefore the heights and the widths of the peaks are determined by the value of w_0. Noting that the singularities are connected with the first two terms of the expansion of the exponential [16]

$$\exp\left\{\frac{1}{N}\sum_q \Gamma_q \cos\left[\omega_q\left(t - \frac{i\hbar\beta}{2}\right)\right]\right\},$$

we can separate the diverging terms and represent the integral in the form of a sum $J_1 + J_2$, where

$$J_1 = e^{-S_T^{(0)}}\int_0^\infty e^{i(\Omega-\omega)t - w_0 t} \times\left\{1 + \frac{1}{N}\sum_q \Gamma_q \cos\left[\omega_q\left(t - \frac{i\hbar\beta}{2}\right)\right]\right\} dt,$$

$$J_2 = e^{-S_T^{(0)}}\int_0^\infty e^{i(\Omega-\omega)t - w_0 t} \times\left\{\exp\left[\frac{1}{N}\sum_q \Gamma_q \cos\left(\omega_q\left(t - \frac{i\hbar\beta}{2}\right)\right)\right] - 1 - \frac{1}{N}\sum_q \Gamma_q \cos\left(\omega_q\left(t - \frac{i\hbar\beta}{2}\right)\right)\right\}. \tag{24}$$

J_2 contains no singularities and has a smooth variation in the vicinity of $|\omega - \Omega| \approx \omega_0$. For estimating purposes, we calculate J_2 for $\omega = \Omega$. Accurate to $\exp(-S_T^{(0)})$ we get

12) M_R, Φ_k, and L_k are power series in η_1, η_2, and $\exp(-S_T)$ [2,3].

13) The largest term in the series for L_k is the one for $n = 4$ [2].

14) These singularities are connected with the singularities $|\gamma_q|^2 \sim q^{-2}$ and $|\partial q/\partial\omega| \sim q^{-1/2}$ as $q \to 0$ in the case of polarization phonons. When the phonon dispersion law is linear in the essential region, as in the case of degeneracy, there should be no such peaks at $\omega = \Omega \pm \omega_0$.

15) An account of the dependence of L on ω may influence the form of the absorption curve in a frequency region of width w_0 at the maximum.

16) In the remaining terms of the expansion, the singularities are contained in the derivatives, in view of the presence of at least double integration with respect to q.

*sh = sinh.

$$J_2 = \left(\frac{2}{\pi}\frac{1}{N}\sum_{\mathbf{q}}\Gamma_{\mathbf{q}}\omega_{\mathbf{q}}^2\right)^{-1/2}\exp\left(-\frac{E_a^{(0)}}{kT}\right) + i\left(\frac{1}{N}\sum_{\mathbf{q}}|\gamma_{\mathbf{q}}|^2\omega_{\mathbf{q}}\right)^{-1} \quad \text{for } T > T_0,$$

$$J_2 = i\left(\frac{1}{N}\sum_{\mathbf{q}}|\gamma_{\mathbf{q}}|^2\omega_{\mathbf{q}}\right)^{-1} \quad \text{for } T < T_0;$$

$$\frac{E_a^{(0)}}{kT} = \frac{1}{N}\sum_{\mathbf{q}}|\gamma_{\mathbf{q}}|^2 \operatorname{th}\frac{\beta\hbar\omega_{\mathbf{q}}}{4}, \qquad (24')^*$$

T_0 is determined from the equality $N^{-1}\Sigma\Gamma_{\mathbf{q}} = 1$. For J_1 we have

$$J_1 = e^{-S_T^{(0)}}\left\{\frac{1}{w_0 + i(\omega - \Omega)} + \frac{1}{2N}\sum_{\mathbf{q}}\Gamma_{\mathbf{q}}\exp\left(\frac{\beta\hbar\omega_{\mathbf{q}}}{2}\right)\frac{1}{w_0 + i(\omega - \Omega - \omega_{\mathbf{q}})} + \frac{1}{2N}\sum_{\mathbf{q}}\Gamma_{\mathbf{q}}\exp\left(-\frac{\beta\hbar\omega_{\mathbf{q}}}{2}\right)\frac{1}{w_0 + i(\omega - \Omega + \omega_{\mathbf{q}})}\right. \qquad (25)$$

The first term of (25) has a sharp peak at $\omega = \Omega$, and its real part is described by a Lorentz curve; it does not depend on the phonon dispersion. The frequency dependence of the second and third terms (peaks at $\omega = \Omega + \omega_0$ and $\omega = \Omega - \omega_0$) is determined by the phonon dispersion. By way of illustration we have calculated (25) for the simplest model of phonon spectrum $\omega = \omega_0 - \Delta\omega(qd)^2$. We consider the second term in (25). Bearing in mind that

$$(2\pi)^3\sum_{\mathbf{q}} = Nd^3\int d^3q,$$

and assuming the dispersion to be small, $\Delta\omega/\omega_0 \ll 1$, we take out the quantity $\exp(\beta\hbar\omega_{\mathbf{q}}/2)\operatorname{cosech}(\beta\hbar\omega_{\mathbf{q}}/2)$, which varies little with q, outside the integral sign, replacing it by $\exp(\beta\hbar\tilde{\omega}/2)\operatorname{cosech}(\beta\hbar\omega/2)$, where $\tilde{\omega} \sim \omega_0$. We also put $|\gamma_{\mathbf{q}}|^2 = \bar{\gamma}(dq)^{-2}$. Going over to integration with respect to ω, we find that the contribution of this term to the admittance is

$$\delta\sigma(\omega) = \Psi(\omega)\,\delta\sigma_0,$$

$$\delta\sigma_0 = \frac{\sqrt{2}\Lambda\bar{\gamma}\omega_0}{8\pi(\omega_0\Delta\omega)^{1/2}}\frac{\exp(\beta\hbar\tilde{\omega}/2 - S_T^{(0)})}{\operatorname{sh}(\beta\hbar\tilde{\omega}/2)}, \qquad (26)$$

$$\Psi(\omega) = (2\pi)^{-2}\left(\frac{\Omega}{\omega}\right)\int_0^{x_0}\frac{x^{-1/2}\,dx}{1 + ix + i(\omega - \Omega - \omega_0)/w_0},$$

$$x_0 = \frac{(q_0 d)^2\,\Delta\omega}{w_0} \qquad (27)$$

(q_0—limiting value of q). For $|(\omega - \Omega - \omega_0)/\Delta\omega| \ll 1$ we have

$$\Psi(\omega) = \frac{\Omega}{\omega}\left[1 + \left(\frac{\omega - \Omega - \omega_0}{w_0}\right)^2\right]^{-1/4} \times \begin{cases}\sin(\varphi/2) - i\cos(\varphi/2), & \omega - \Omega - \omega_0 > 0,\\ \cos(\varphi/2) - i\sin(\varphi/2), & \omega - \Omega - \omega_0 < 0\end{cases}$$

$$\varphi = \operatorname{arc tg}\left|\frac{w_0}{\omega - \Omega - \omega_0}\right|. \qquad (28)^*$$

(The expression for (27) is quite cumbersome in the general case.)

The third term in J_1 shows an analogous dependence in the region $\omega = \Omega - \omega_0$, but contains a small factor $\exp(-\beta\hbar\tilde{\omega})$. The approximate variations of Re $\delta\sigma$ and Im $\delta\sigma$ near $\omega \sim \Omega + \omega_0$ is shown in Figs. 4a and b. Re $\delta\sigma$ increases on the low-frequency side and $|\text{Im}\,\delta\sigma|$ decreases on the high-frequency side, both like $|\omega - \Omega - \omega_0|^{-1/2}$, and the decrease of Re $\delta\sigma$ towards the higher frequencies and the increase of $|\text{Im}\,\delta\sigma|$ on the side of the lower frequencies are like $|\omega - \Omega - \omega_0|^{-3/2}$. The maxima are attained at $\omega - \Omega = \omega_0$, and have absolute values that are equal to $\delta\sigma_0\Omega/(\Omega + \omega_0)$ for both Re $\delta\sigma$ and Im $\delta\sigma$. The width of the peaks is $\sim w_0$. When $|\omega - \Omega - \omega_0| \gg \Delta\Omega$, the value of Re $\delta\sigma$ is small, like w_0/Ω, and Im $\delta\sigma$ behaves like $(\omega - \Omega - \omega_0)^{-1}$.

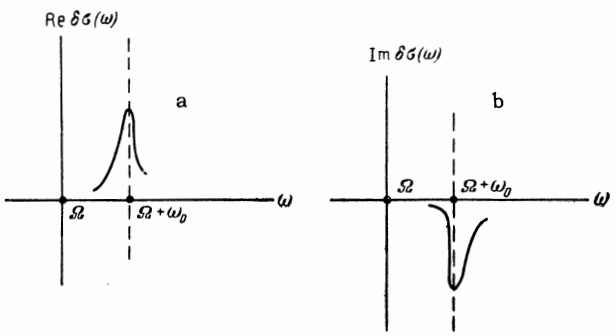

FIG. 4

If the mechanisms that ensure a finite height of the peaks at $\omega = \Omega$ and $\omega = \Omega + \omega_0$ are the same, then the ratio of the heights of the first and second peaks will be $\gamma^{-1}(w_0/\Delta\omega)^{1/2}$ (see Sec. 5). These peaks are superimposed on the dependence $\Delta\sigma(\omega)$, which is smooth in the remaining frequency region. Re $\Delta\sigma$ is described by a bell-shaped curve of width $(kT\Delta E_p/\hbar^2)^{1/2}$ at $T > T_0$ and $(\omega_0\Delta E_p/\hbar)^{1/2}$ for $T < T_0$[5,6]. Its values at the maximum points are

$$\Lambda(\hbar^2\omega_0^2/4kT\Delta E_p)^{1/2} \text{ for } T > T_0,$$
$$\Lambda(\hbar\omega_0/4\pi\Delta E_p)^{1/2} \text{ for } T < T_0;$$

Im $\Delta\sigma$ has a smooth variation and vanishes at $\omega \sim \Omega + \Delta E_p/\hbar$.

5. DISCUSSION OF RESULTS

We have calculated in this paper the contribution made by $\Delta\sigma$ to the complex conductivity and due to

*th = tanh.

*arc tg = $\tan^{-1}$.

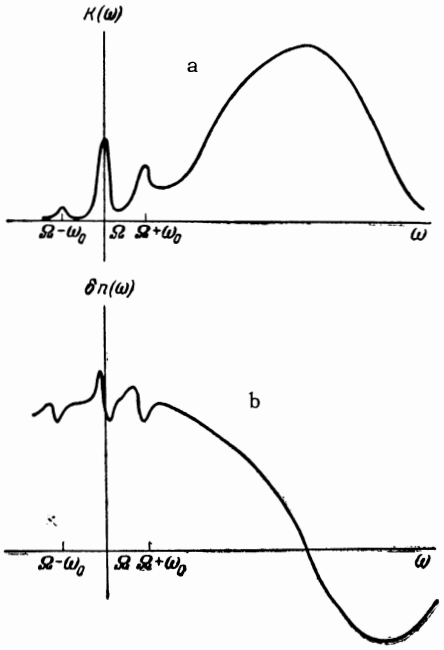

FIG. 5. Absorption curves (not to scale). It must be borne in mind that in the region $(\Omega - \omega_0, \Omega + \omega_0)$ the structure part should be exponentially small (by a factor $\exp[S_T]$) compared with the region of the principal maximum.

transitions from a discrete level to a band with low mobility; this, as pointed out in the introduction, makes it possible to obtain $K(\omega)$ and $\Delta n(\omega)$. The character of $K(\omega)$ is shown in Fig. 5. The principal maximum of the curve is located at $\omega \sim \Omega + \Delta E_p/\hbar$; its height is

$$\frac{4\pi\Lambda}{c\bar{n}}\left(\frac{\hbar^2\omega_0^2}{4kT\Delta E_p}\right)^{1/2}, \quad T > T_0; \quad \frac{4\pi\Lambda}{c\bar{n}}\left(\frac{\hbar\omega_0}{4\pi\Delta E_p}\right)^{1/2}, \quad T < T_0;$$

The width of the curve is of the order of

$$(kT\Delta E_p/\hbar^2)^{1/2}, \quad T > T_0; \qquad (\Delta E_p\omega_0/\hbar)^{1/2}, \quad T < T_0$$

($\bar{n}$ was defined in the introduction, Λ and ΔE_p in Sec. 1, and E_a and T_0 in Sec. 4). Additional peaks occur at $\omega = \Omega$, $\omega = \Omega + \omega_0$, and $\omega = \Omega - \omega_0$. Their heights are respectively of the order of

$$\frac{4\pi\Lambda\omega_0}{c\bar{n}w_0}e^{-S_T^{(0)}}, \quad \frac{4\pi\delta\sigma_0}{c\bar{n}}\left(\frac{w_0}{\tilde{w}_0}\right)^{1/2},$$

$$\frac{4\pi\delta\sigma_0}{c\bar{n}}\left(\frac{w_0}{\tilde{w}_0}\right)^{1/2}\exp\left(-\frac{\hbar\omega_0}{kT}\right);$$

w_0—escape probability, which enters in the transport equation; $w_0 = w_0 + w_{anh}$, where w_{anh}—of the order of the polarization phonon decay probability connected with the anharmonicity (see below); at low temperatures $w_{anh} \gg w_0$. The width of the first peak is $\sim w_0$, and that of the second and third $\sim \tilde{w}_0$; the first peak has a Lorentz shape, while the second and third have essentially different shapes (see Sec. 4). When $T < T_0$ the smoothly varying background in the vicinity of these peaks is small, while for $T > T_0$ it is of the order of

$$[4\pi\Lambda\hbar / c\bar{n}(kT\Delta E_p)^{1/2}]\exp(-E_a^0/kT),$$

and the peaks themselves broaden considerably in view of the increase in w_0. The easiest to observe are the first and second peaks.

We note that in the case of a strong electron-phonon interaction the width of the absorption band is not determined at all by the widths of the initial electron or polaron bands. The addition to the refractive index is determined by the quantity $\mathrm{Im}\,\Delta\sigma$, which displays a nonmonotonic dependence in the vicinity of the frequencies Ω and $\Omega + \Delta E_p/\hbar$ and, in particular, reverses sign at $\omega \sim \Omega + \Delta E_p/\hbar$ (where $\mathrm{Re}\,\Delta\sigma$ has a maximum). Near $\omega = \Omega$ and $\omega = \Omega \pm \omega_0$ there are superimposed on the smooth variation of the curve some sharp sign-reversing peaks (of the discontinuity type). The amplitudes and the widths of these peaks are of the same order as of the corresponding peaks in $\mathrm{Re}\,\delta\sigma$. Compared with the background, the peaks are small like $\exp(-S_T^{(0)})$ (see Fig. 5b).

We note that we have assumed in this paper that the smearing of the singularities of $\Delta\sigma$ is due only to the "residual" electron-phonon interaction. Another cause of the smearing may be the anharmonic interaction between the optical phonons and the acoustic phonons, which leads to a finite lifetime of the optical phonon. Such a mechanism can lead to an additional smearing of the peaks at $\omega = \Omega \pm \omega_0$. Indeed, in this case one phonon participates in the transition, and it can decay into two acoustic phonons, giving a finite lifetime even when $T = 0$, whereas the mechanism considered in the present work gives a sharp decrease of w_0 at low temperatures, $w_0 \sim \exp(-\hbar\omega_0/kT)$. Therefore the anharmonicity predominates, at least at low temperatures.

As to the peak at $\omega = \Omega$, the presence of anharmonicity does not affect its width, because the peak is due to transitions between states that do not contain real phonons (phononless transition); the smearing appears only as a result of residual interaction, and in the low-temperature limit also as a result of the tunnel effect, when the condition $J\exp(-S_T) > \hbar\omega_0$ begins to be satisfied. The width of the peak is then $\delta E \sim J\exp(-S_T)$, and the height is $\sim (\delta E)^{-1}$. The shape of the peak is not Lorentzian.

Inclusion of the nonadiabatic terms in the Hamiltonian can also lead to a broadening of the phononless peak resulting, for example, from nonradiative transitions (which are due to terms

$b^+a_0^+a_1$ etc.), but their contribution will be proportional to $(\gamma\omega_0/\Omega)^{\Omega/\omega_0}$, which is small in the case under consideration, when $AE_p/\hbar\Omega \ll 1$.

Let us point out the analogy between the phononless peak and the Mössbauer line in nuclear transitions; the quantity $\exp(-S_T^{(0)})$ is analogous to the Debye-Waller factor and determines the intensity of the peak.

The present results can be generalized to include the case when both initial electron states (0 and 1) strongly interact with the phonons (neglecting the terms $b^+a_0^+a_1$, see above) under the condition that the band width of one of the levels is small compared with the width of the other. In this case it is sufficient to replace $|\gamma_{\mathbf{q}}|^2$ by $|\gamma_{\mathbf{q}}^{(0)} - \gamma^{(1)}|^2$ in $Q(t)$, and substitute in w_0 the value of η_1 pertaining to the broader band [17].

Comparison of the present results with experiments on the absorption of light in NiO[7] shows qualitative agreement with the theoretically deduced shape of the absorption curve. At 300°K (see Fig. 2 in the paper of Newman and Chrenko[7]) the quantity $K(\omega)$ shows traces of a peak at $\hbar\omega \approx 0.75$ eV, which agrees in order of magnitude with the possible position of the peak at $\omega = \Omega$. For a sharp resolution of such a peak it would be necessary to carry out measurements at lower temperatures, but this frequency interval has been left out from the 77°K curve of the cited paper[7].

In conclusion the authors thank A. I. Ansel'm, V. L. Gurevich, L. I. Korovin, and G. E. Pikus for useful discussions.

17) Eagles [4] considered an analogus problem of absorption of light by a crystal containing polarons of small radius. The coefficient of absorption was calculated using the nonstationary perturbation theory for multiphonon transitions, developed in the F-center theory[5, 6]. The phonon dispersion was not taken into account, and the singularities of the refractive index were not investigated. The authors confined themselves to the general form of the absorption line and the additional peaks were not investigated, but it was pointed out that a peak may exist at $\omega = \Omega$.

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