Optical-absorption tail in polar crystals and the Urbach rule

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The light absorption tail in a polar crystal is considered for the case when the photon energy deficit $\Delta = E_g - \hbar\Omega$ (E_g is the width of the forbidden band and Ω is the light frequency) is large compared with the phonon energy $\hbar\omega_0$. It is shown that two different mechanisms of multiphonon absorption may occur, quasistatic and dynamic. In the first case the absorption occurs in a fluctuation state that does not manage to change during the absorption time (equal to the exciton lifetime τ_s in the fluctuation state). The test of the quasistatic absorption is $\omega_0\tau_s \ll 1$. In the opposite (dynamic) limit simultaneous absorption of n phonons takes place, where $n = [(E_g - \hbar\Omega - |E_0|)]/\hbar\omega_0$ (E_0 is the exciton binding energy including polaron effect). Self-consistent field equations are derived which are valid for all cases in which a large number of phonons participate, and describe both limiting cases in a unified manner. It is shown that fluctuation "overscreening" of the Coulomb attraction between the electron and hole should be possible. As a result the electron and hole repel each other and exciton effects turn out to be insignificant. The dependence of the light absorption coefficient on frequency Ω turns out to be somewhat stronger than that predicted by the well-known Urbach law and is consistent with the latter only at low temperatures.

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I. INTRODUCTION

An exponential light-absorption tail is observed in the forbidden band of many substances. This phenomenon was first observed by Urbach[1] in AgBr. Urbach has noted that the dependence of the absorption coefficient $K_a(\Omega)$ on the photon energy $\hbar\omega$ and on the temperature is well described by the empirical formula (the Urbach rule)

$$K_a(\Omega) = K_0 \exp\left[\sigma(\hbar\Omega - E_0)/T\right], \tag{1}$$

where E_0 is a certain energy of the order of the band gap E_g and σ is a constant. In alkali-halide crystals $\sigma = 0.7$–1.5, and in semiconductors $\sigma = 3$–5.

A similar behavior was subsequently observed in many alkali-halide crystals (reviews can be found in Refs. 2 and 3), semiconductors,[4,5] and molecular crystals (see Ref. 6). It turned out that σ is frequently a function of the temperature, and it appears that the ratio $\sigma(T)/T$ tends sometimes as $T \to 0$ to a finite limit; this corresponds to finite absorption at $T = 0$.

In some cases the absorption tail may be due to impurities, but in pure crystals the relation (1) is attributed as a rule to the thermal vibrations of the lattice. Favoring this assumption is the temperature dependence (1). We shall consider hereafter only pure crystals and disregard the possible influence of impurities.

The theoretical justification of the rule (1) is the subject of a large number papers,[7–15] which can be arbitrarily divided into two groups. In the first group[7–10] the interaction with the phonon is reduced to the interaction of an exciton with homogeneous and constant electric fields (or with strains, depending on the type of considered phonons). The final expression is then averaged with the distribution function of these fields (or strains). Dexter[7] considered the homogeneous variation of the band gap under the influence of the strain. The expression he obtained for $\ln K_a(\Omega)$ is quadratic in $\Delta = E_g - \hbar\Omega$ and does not coincide with (1). To satisfy the rule (1), Toyozawa[8] investigated a variation of E_g quadratic in the strain. This assumption, however, seems to deviate from the known data.[16]

In another paper[9] Dexter assumed the cause of the rule (1) to be the quadratic Stark effect on excitons. A quadratic dependence of the level shift on the electric field, however, calls for the assumption that the field is homogeneous. It would be obviously more convenient to produce an inhomogeneous field fluctuation, thus ensuring a linear level shift.

Dow and Redfeld[10] interpreted the Urbach rule (1) as a Kelydsh-Franz effect averaged over random fields. They, however, started out from a linear dependence of $K_a(\Omega)$ on Δ in a constant homogeneous electric field, which is not the case in the Kelydsh-Franz effect. On the basis of numerical calculations,[17] Dow and Redfeld have proposed that the exciton effects alter radically the Keldysh-Franz effect and lead to such a linear dependence. The latest investigations[18] do not confirm this assumption. We note that even if we start out from the premises of Dow and Redfeld, the final expression for $K_a(\Omega)$ has neither the same frequency nor the same temperature dependence as (1).

It will be shown in the present paper that the assumption that the fields (and strains) are static, which is made in the first group of papers, can be satisfied under certain (rather stringent) conditions on the temperature T and on the energy Δ. On the other hand, the assumption that the fields are homogeneous is always incorrect. The characteristic wavelength of the phonons responsible for the absorption process (i.e., the size of the field fluctuations) coincides with the wavelength of the electron and hole, so that their interaction with the field has essentially a quantum nature in both the static and dynamic case. Therefore the results of Refs. 7–10 and the very concept of homogeneous fields or strains are not correct.

In the second group of papers the interaction of the

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electron-hole pair with phonons is considered directly. Some papers[11-13] use the approach adopted in the theory of light absorption by an infinitely heavy impurity center. The need for satisfying the momentum-conservation law when a phonon is absorbed is practically ignored. As a result, contributions to the absorption are made by phonons having momenta that differ substantially from the electron momenta. A detailed critical analysis of Refs. 11–13 was carried out by Kagan,[14] who obtained an estimate of $K_a(\alpha)$ within the framework of an approximation that takes into account interaction between the phonons and only one of the particles. The lowest order of perturbation theory in the phonons, ensuring the possibility of absorption of a photon with energy $\hbar\Omega$ was considered.

We shall show in the present paper that the possibility of considering only the lowest order of perturbation theory arises in a case that is the alternative of the quasistatic approximation.

Kagan[14] has noted that at $T=0$ the absorption in the region of frequencies lying below the ground bound state should vanish exactly. Therefore at sufficiently low temperature the statistical approximation cannot be valid in this frequency region, since it leads to finite absorption. Nonetheless, in a number of experiments the absorption coefficient $K_a(\Omega)$ remains finite as $T\to 0$. If we exclude from consideration the influence of the impurities, then it remains to assume that the frequency interval corresponding to the rule (1) lies higher than the ground state in these experiments. The latter is possible for a strong exciton-phonon coupling, when the energy of the localized exciton lies much lower than the energy of the free exciton.

Such a situation was considered by Sumi and Toyozawa[15], where the behavior of $K_a(\Omega)$ was investigated, using the Frenkel-exciton model, in a frequency region much lower than the free-exciton energy and much higher than the energy of the localized exciton.

II. QUALITATIVE ANALYSIS AND PRINCIPAL RESULTS

We assume in this paper that all the characteristic energies of the problem are much lower than the band width, and the characteristic dimensions are much larger than the lattice constant. This allows us to use the effective-mass method for the electron and for the hole, and to neglect the dispersion of the optical phonons. At the same time we shall assume that $\Delta\gg\hbar\omega_0$, and regard the electron and hole as being in a state that varies adiabatically with the motion of the lattice. We consider interaction with longitudinal optical (LO) phonons.

1. Quasistatic fluctuations

We study first the conditions of applicability of the quasistatic approximation, where the premise is that during the entire absorption act the fluction of the electric field remains unchanged. According to the Franck-Condon principle, the state of the lattice does not manage to change during the time $\sim\Omega^{-1}$ of the optical transition (since $\Omega\gg\omega_0$). Therefore, in order for an exciton to be produced in a certain polaron well with an energy level $-\Delta$, the polaron jacket corresponding to this state should be prepared beforehand by a fluctuation method. At sufficiently high jacket energy, the probability of such a fluctuation is exponentially small, and this provides in principle a possibility of explaining the rule (1).

The probability of the onset of any fluctuation corresponding to any value of Δ is finite also at $T=0$ (on account of the zero-point oscillations of the vacuum). This, however, still does not mean that the absorption is finite. Indeed, the duration of the absorption act is in fact not the time Ω^{-1} of the optical transition, but the lifetime τ_s of the final state, after which the coherence of the exciton state with the photon is violated as a result of some scattering act. However, it is meaningful to speak not simply of an optical transition but of absorption of a photon and creation of an exciton in a given fluctuation state only if the lifetime of the exciton in this state is much shorter than the time during which the fluctuation itself changes its form, i.e., if $\omega_0\tau_s\ll 1$.

To explain this, we consider in greater detail the case $T=0$. If the energy $-\Delta$ lies below the ground state of the system, then a departure from the fluctuation state into a stationary state is possible only with increase of the exciton energy. The system contains no phonons capable of being absorbed; on the other hand, it is impossible to draw energy from the zero-point oscillations. Therefore $\tau_s\to\infty$ and the static approximation is certainly not applicable. If an optical transition to such a fluctuation state does take place, the system must of necessity again emit a photon (without loss of coherence), and such a transition will contribute only to the real part of the dielectric constant, but not to the absorption.

If $T\neq 0$, then $\tau_s\sim N^{-1}$, where $N=[\exp(\hbar\omega_0/T)-1]^{-1}$ is the phonon occupation number, and at a certain temperature the condition $\omega_0\tau_s\ll 1$ will be satisfied.

When the energy $-\Delta$ lies above the ground state of the system, the static approximation may turn out to be valid even at $T=0$. Indeed, in this case transitions with emission of phonons are possible and can take place also at $T=0$.

Thus, the Franck-Condon principle (which is valid at $\Omega^{-1}\omega_0\ll 1$) does not necessarily lead generally speaking to applicability of the quasistatic approximation, the condition for which is $\omega_0\tau_s\ll 1$, and the Franck-Condon principle by itself, of course, does not contradict the requirement that an exact absorption edge exist.

We assume initially for simplicity that the electron is much lighter than the hole. In this case, owing to the larger polaron coupling constant of the hole,

$$\alpha_i=\frac{1}{2}(\varepsilon_\infty^{-1}-\varepsilon_0^{-1})\frac{e^2}{\hbar\omega_0}\left(\frac{2m_i\omega_0}{\hbar}\right)^{1/2}$$

it turns out to be convenient to ensure the greater part of the decrease of the exciton energy on account of the hole and not on account of the electron. The fluctuation of the electric field should have a hole level with energy

$E_h = -\Delta$, and the electron energy is close to zero. The energy of the hole interacting with the polarization is described by a functional of the energy[19]

$$E_h = \min_{\psi}\left\{\frac{1}{2m_h}\int |\nabla\psi|^2 d^3\mathbf{r} - \int \mathbf{P}(\mathbf{r})\mathbf{D}(\mathbf{r})d^3\mathbf{r}\right\}, \quad (2)$$

$$\mathbf{D}(\mathbf{r}) = -e\int \nabla_r(|\mathbf{r}-\mathbf{r}'|^{-1})|\psi(\mathbf{r}')|^2 d^3\mathbf{r}', \quad (3)$$

$\mathbf{D}(\mathbf{r})$ is the electric induction produced by the hole in a state with wave function ψ, and $\mathbf{P}(\mathbf{r})$ is the polarization.

The condition for the applicability of expressions (2) and (3) is a large value of the energy $|E_h|$ compared with $\hbar\omega_0$, i.e., $\Delta \gg \hbar\omega_0$. The energy of the polarization is of form[19]

$$E_{pol} = \frac{2\pi}{\varepsilon_\infty^{-1} - \varepsilon_0^{-1}}\int \mathbf{P}^2(\mathbf{r})d^3\mathbf{r}. \quad (4)$$

Since the probability of the fluctuation increases with decrease of E_{pol}, the optimal fluctuation should ensure a minimum of E_{pol} upon satisfaction of the condition $E_h = -\Delta$. It is easy to estimate $\min E_{pol}$. Let the hole potential well have a depth U_h and a width a_h (Fig. 1). The characteristic polarization energy density in the region of the well is then $w \sim (\varepsilon_\infty^{-1} - \varepsilon_0^{-1})^{-1}(U_h/ea_h)^2$, and the volume of the region in which the energy is concentrated is $\sim a_h^3$. Therefore

$$E_{pol}^h(\Delta) \sim wa_h^3 \sim (\varepsilon_\infty^{-1} - \varepsilon_0^{-1})^{-1}(U_h/e)^2 a_h. \quad (5)$$

It is clear that $U_h \sim |E_h| = \Delta$. From (5) it is seen that E_{pol} decreases with decreasing dimension a_h of the fluctuation. However, when a_h becomes comparable with the hole wavelength $\hbar(2m_h\Delta)^{-1/2}$, the level E_h begins to be pushed out of the well. Therefore the optimal size of the fluctuation is determined by the wavelength of the hole with energy Δ. Consequently

$$E_{pol}^h(\Delta) = \text{const}\cdot(\varepsilon_\infty^{-1} - \varepsilon_0^{-1})^{-1}\hbar\Delta^{3/2}/e^2 m_h^{1/2} = \text{const}\cdot\Delta^{3/2}/\alpha_h(\hbar\omega_0)^{1/2}. \quad (6)$$

A variational calculation (see Sec. IV) yields a value const = 1.167.

The light absorption coefficient is proportional to the probability $W(E_{pol}^h(\Delta))$ for the onset of the optimal fluctuation:

$$K_a(\Omega) \sim W \sim \exp\left[-\frac{E_{pol}^h(\Delta)}{\hbar\omega_0(N+1/2)}\right] = \exp\left[-\frac{1.167}{\alpha_h(N+1/2)}\left(\frac{\Delta}{\hbar\omega_0}\right)^{3/2}\right]. \quad (7)$$

At high temperature ($T \gg \hbar\omega_0$ we have $N + 1/2 \approx T/\hbar\omega_0$ and expression (7) coincides with the usual Boltzmann formula $\exp(-E/T)$. At low temperature the fluctuation is due to the zero-point oscillations of the phonon vacuum. As shown above, this is possible without violation of the condition $\omega_0\tau_s \ll 1$ only if $\Delta < |E_0|$, where E_0 is the energy of the ground state of the system.

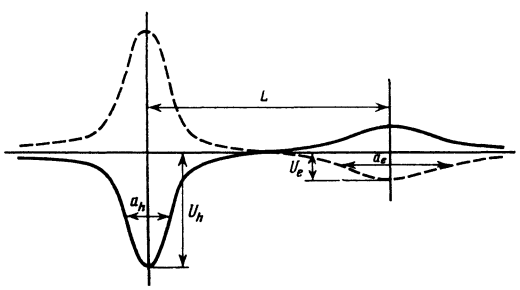

FIG. 1. Form of the optimal fluctuation. $U_{e,h}$ and $a_{e,h}$ are the depth and width, respectively, of the electron hole wells. The solid and dashed lines show the potential energies of the hole and of the electron, respectively.

We consider now the case when the masses of the electron and of the hole are comparable. Two fluctuation wells should be produced then, electron and hole (Fig. 1). Since the charges of the electron and of the hole are opposite, the hole well is a potential "hump" for the electron, and vice versa. It is clear therefore that it is impossible to place both an electron and a hole in a single well (provided that the exciton interaction between them is not too large, see below).

Let the wells be separated by a large distance L, so that the interaction can be neglected in the zeroth approximation. It is then easy to determine the energy of the optimal fluctuation:

$$E_{pol}^*(\Delta, L=\infty) = \min_{E_e+E_h=-\Delta}\{E_{pol}^e(-E_e) + E_{pol}^h(-E_h)\}, \quad (8)$$

where $E_{pol}^{e,h}$ are determined by expression (6). As a result $E_{pol}^*(\Delta, L=\infty)$ differs from (6) and that α_h is replaced by $\alpha^* = (\alpha_h^2 + \frac{2}{e})^{1/2}$. For the energies we obtain

$$E_e = -(m_e/M)\Delta, \quad E_h = -(m_h/M)\Delta, \quad M = m_e + m_h.$$

To take into account the interaction it is necessary to find the effective charges q_e and q_h of the polarization wells, which screen the "bare" charges of the electron and the hole. Obviously, $q_e > 0$, $q_h < 0$, and their values can be estimated from the relation $|E_i| \sim e|q_i|/a_i$ or $|q_i| \sim (h/e)(|E_i|/m_i)^{1/2}$. Using the obtained expressions for E_1 (in the zeroth approximation in the interaction) we have

$$q_e = -q_h = q \sim (\hbar/e)(\Delta/M)^{1/2}. \quad (9)$$

At $q = e/\varepsilon_\infty$ the screening charge coincides with the "bare" charge and the effective dielectric constant becomes infinite (complete screening), while at $q > e/\varepsilon_\infty$ it becomes negative, and overscreening sets in. We emphasize that we are dealing here not with the equilibrium static dielectric constant, but with the fluctuating one, which has a finite lifetime. With the aid of (9) we can write the over screening condition in the form

$$\Delta > \Delta_0 = \text{const}\cdot M^2 R/4m_e m_h, \quad (10)$$

where R is the exciton Ryberg. Calculation yields const = 1.3. At $m_e \ll m_h$ it turns out that $\Delta_0 \gg R$, but at $m_e \ll m_h$ the behavior of the electron is of no importance at all (its energy is low). At $m_e \sim m_h$ we have $\Delta_0 \sim R$ and overscreening sets in at all energies, in the exciton region. At $\Delta \gg \Delta_0$ (strong overscreening), the bare charge e/ε_∞ can be neglected compared with q.

In the present paper we consider for simplicity just the case of strong overscreening, neglecting the exciton effects. We note, however, that in the case of weak overscreening ($\Delta \gtrsim \Delta_0$) the exciton effects, while substantially altering the value of $K_a(\Omega)$, do not change the character of the dependence of $K_a(\Omega)$ on Δ and on N. The situation here is the same as in the Keldysh-Franz

effect, namely, the exciton effects lead to an additional term in the argument of the exponential. This term is large compared with unity but small compared with the fundamental non-Coulomb term and therefore does not change strongly the frequency and field dependences of the effect.[18] A strong restructuring of $K_a(\Omega)$ as a result of exciton effects can be expected only at $\Delta < \Delta_0$.

Allowance for a strongly overscreened interaction makes it necessary to add the interaction energy $-\Delta = E_e + E_h + qe/L$ to the condition that the total energy of the pair be equal to $-\Delta$ ($E_{e,h}$ are the single-particle energies). As a result we have

$$E^*_{pol}(\Delta, L) = E^*_{pol}(\Delta + qe/L, \infty).$$

Thus, the fluctuation energy decreases with increasing distance between the wells (in the case of overscreening). It must be remembered, however, that $K_a(\Omega)$ is proportional not only to the probability $W(E^*_{pol})$ of the onset of the fluctuation (density of states), but also to the square of the overlap integral $|J(L)|^2$ of the electron and hole wave functions, which decreases exponentially with increasing distance between wells. Obviously, $J(L)$ is determined by the tails of the wave function of the lighter electron in the region of the polarization of the heavier hole, therefore

$$J(L) \sim \exp\left[-L(2m_e|E_e|)^{1/2}/\hbar\right].$$

Consequently, the optimal fluctuations will be the one determined by those $L = L_{opt}$ which ensures a maximum of the product $|J(L)|^2 W(E^*_{pol})$:

$$K_a(\Omega) \sim \max_L |J(L)|^2 W(E^*_{pol})$$

$$\sim \exp\left\{-\frac{1{,}17}{\alpha^*(N+1/2)}\left(\frac{\Delta}{\hbar\omega_0}\right)^{1/2} - 7.0\left[\frac{m_e}{M}\left(\frac{\Delta}{\hbar\omega_0}\right)^{1/2}\frac{1}{\alpha^*(N+1/2)}\right]^{1/2}\right\}. \quad (11)$$

The second term in the exponential of (11) need be taken into account only when it is large compared with unity (this means $L_{opt} \gg a_e$); as $m_e/m_h \to 0$ it becomes small and (11) coincides with (7).

We now write out explicitly the expression for τ_s. Calculation of the free-path time of a particle i with energy E_i, interacting with phonons, yields (see, e.g., Ref. 20)

$$\frac{1}{\tau_{si}} \sim \frac{\alpha_i(\hbar\omega_0)^{1/2}}{\hbar|E_i|^{1/2}} N$$

at $|E_i| > |E_0|$ (transitions with phonon absorption) and

$$\frac{1}{\tau_{si}} \sim \frac{\alpha_i(\hbar\omega_0)^{1/2}}{\hbar|E_i|^{1/2}}\left(N + \frac{1}{2}\right)$$

at $|E_i| < |E_0|$ (transitions with both absorption and emission of phonons). We have

$$\frac{1}{\tau_s} = \frac{1}{\tau_{se}} + \frac{1}{\tau_{sh}} \sim \frac{\alpha^*(\hbar\omega_0)^{1/2}}{\hbar\Delta^{1/2}} \begin{cases} N & \text{if } \Delta > |E_0| \\ N+1/2 & \text{if } \Delta < |E_0| \end{cases}. \quad (12b)$$

The optimal-fluctuation method used above is valid if $E^*_{pol} \gg \hbar\omega_0(N + 1/2)$; in the opposite case the contribution to the absorption is made by non-optimal fluctuations and $K_a(\Omega)$ ceases to be an exponential function. With the aid of (6) and (12) it is easy to verify that the last requirement coincides with the condition that the quantum state of the electron-hole pair is "well-defined," $\tau_s\Delta \gg \hbar$. We note also that by virtue of the very same condition the second term in the argument of the exponential in (11) is always smaller than the first, so that we can regard the interaction as a small increment to the single-particle energies.

Thus, the condition for the applicability of the quasistatic approximation is of the form

$$\Delta \gg \hbar\tau_s^{-1} \gg \hbar\omega_0. \quad (13)$$

2. Dynamic fluctuations

We consider now another limiting situation:

$$\Delta \gg \hbar\omega_0 \gg \hbar\tau_s^{-1}.$$

In this case the fluctuation can certainly not be regarded as static. However, the smallness of the parameter $1/\omega_0\tau_s$ means effective weakness of the interaction with the phonons, and by the same token the possibility of using perturbation theory. At $\Delta \gg \hbar\omega_0$ only high order (n-th and higher, where $n = [\Delta/\hbar\omega_0] + 1 \gg 1$, and $[\ldots]$ is the integer part of the number) of perturbation theory contribute. The contributions of lower order are equal to zero because of the impossibility of satisfying the energy conservation law. At $\omega_0\tau_s \gg 1$ it is necessary to take into account only the n-th lowest of the nonvanishing orders of perturbation theory. The absorption coefficient is thus proportional to the probability of simultaneous absorption of n phonone[14]:

$$K_a(\Omega) \sim \left(\frac{\text{const}}{\omega_0\tau_s}\right)^n \sim \exp\left\{-\frac{\Delta}{\hbar\omega_0}\ln\left[\frac{\text{const}}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega_0}\right)^{1/2}\right]\right\}, \quad (14)$$

and the optimal dynamic fluctuation constitutes simply n phonons.

To illustrate the estimate (14), we put again $m_h > m_e$ and calculate the imaginary part of some perturbation theory diagram of p-th ($p \geq n$) order for the hole Green's function, for example G_p shown in Fig. 2. This diagram contains p phonon lines, $2p+1$ hole lines, $2p$ vertices, and p integrations over the phonon momenta. It is clear that all the momenta of both the phonons (Q^{ph}) and the holes (Q^h) in the G_p diagram are of the same order. Also of the same order are all the hole energies ε_h:

$$\varepsilon_h \sim \Delta, \quad Q^{ph} \sim Q^h \sim Q = (m_h\Delta)^{1/2}. \quad (15)$$

Allowance for the different momenta and energies leads to a change in the constant in (14); we disregard the constant in the present estimate. Substituting (15) in G_p, we obtain

$$\operatorname{Im} G_p \sim [N\alpha_h(\hbar\omega_0/\Delta)^{1/2}]^p.$$

Since all the phonon momenta are of the same order,

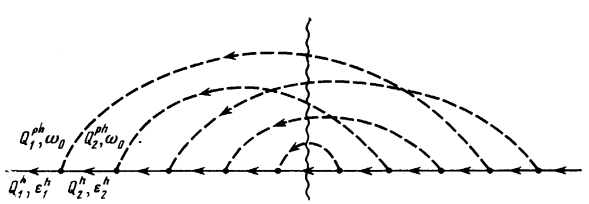

FIG. 2. Typical Feynman diagram of G_p, corresponding to the absorption of $p = 5$ phonons. Dashed lines—phonon Green's functions (at finite temperature), solid line—Green's function of free hole.

permutations of the ends of the phonon lines should not lead to a strong change of G_p. Therefore approximately equal contributions are made to $K(\Omega)$ by all $p!$ diagrams of p-th order in which the sequence of phonon absorption is arbitrary. The difference between these contributions will influence only the constant in (14), and we shall therefore assume them to be the same. As a result, the contribution of all the diagrams of p-th order is $\mathrm{Im}\tilde{G}_p \approx p!\,\mathrm{Im}G_p$. Putting $p=n+n_1$, where $n_1 \ll n$, we obtain

$$\mathrm{Im}\,G_p \sim [N\alpha_h(\hbar\omega_0/\Delta)^{1/2}]^{n+n_1},$$

or, comparing with (12)

$$G_p \sim (1/\omega_0\tau_s)^{n+n_1}.$$

Thus, the parameter of the perturbation theory in this situation is indeed $1/\omega_0\tau_s$; at $1/\omega_0\tau_s \ll 1$ we can neglect all the $\tilde{G}_p$ except $\tilde{G}_n$, and the estimate (14) is valid.

The calculation of the constant in (14) as well as of all the two particle effects by the diagram technique is difficult. They will be calculated in Sec. IV by the self-consistent-field method. We note here only that at $E_0 \gg \hbar\omega_0$ is it necessary to take into account the renormalization of the bare Green's functions. As a result, in the calculation of τ_s it is necessary to replace Δ in (12a) by $\tilde{\Delta}=\Delta-|E_0|$. This corresponds to using as the zeroth approximation not free holes but polarons (or excitons–depending on the type of bond that predominates in E_0).

It must be emphasized that the possibility of using perturbation theory with respect to phonons in the calculation of $K_a(\Omega)$ is not connected with such a possibility in the calculation of the energy of the ground state E_0. In the former case the parameter is $1/\omega_0\tau_s$, and in the latter it is α_h, and smallness of the first does not mean smallness of the second and vice versa. The point is that the absorption is due only to real phonons with momenta $Q \sim (m_h\Delta)^{1/2}$, and the self-energy is due to virtual phonons with momenta that do not depend on Δ and are determined only by E_0 itself.

3. Self-consistent-field approximation, principal results

The two limiting cases considered above, which describe different physical situations, have one common feature; a large number of phonons participate in the process. This feature is connected with the condition $\Delta \gg \hbar\omega_0$ and does not depend on the value of $\omega_0\tau_s$. In Sec. III of the present paper we obtain a method that makes it possible to calculate $K_a(\Omega)$ using only the condition that the number of phonons be large. A nonlinear Schrödinger equation is derived, which describes the motion of an electron-hole pair in a self-consistent field that is not regarded as static but varies adiabatically slowly in comparison with the pair-motion frequency. The adiabaticity is ensured by the condition $\Delta \gg \hbar\omega_0$. The interaction of the pair with the field of the optimal fluctuations is reduced to the effective self-action. At $t=0$, the obtained equation is similar in structure to the equation introduced by Iordanskiĭ and Rashba[21] for the description of tunnel autolocalization.

The developed method makes it possible to take into account the interaction of an electron with a hole, to determine all the numbers in the exponential, and to calculate the pre-exponential factor. The final expression for $K_a(\Omega)$ at $\Delta > |E_0|$ is of the form

$$K_a(\Omega) = \frac{4\pi e^2 s^2 (\hbar\omega_0)^{1/2}}{\Omega c n_0 \hbar^3} C_1 m_e^{3/2} \left(\frac{m_e}{M}\right)^{1/2} \times f_1(\omega_0\tau_s)\exp\left\{-\frac{\Delta}{\hbar\omega_0} f_2(\omega_0\tau_s) - \left(\frac{m_e}{M}\frac{\Delta}{\hbar\omega_0}\right)^{1/2} f_3(\omega_0\tau_s)\right\}. \quad (16)$$

Here C_1 is a certain number of the order of unity, τ_s is defined by (12a), s is the interband matrix element of the velocity operator, and n_0 is the refractive index. The functions f_1, f_2, and f_3 are shown in Fig. 3. Their quasistatic ($x \ll 1$) and dynamic ($x \gg 1$) asymptotics are

$$f_1(x)\approx 2.45x^{-1/2}, \quad f_2(x)\approx 1.167x, \quad f_3(x)\approx 7.00x^{1/2}, \quad x \ll 1, \quad (17)$$

$$f_1(x)\approx 1.24, \quad f_2(x)\approx \ln 2.77x, \quad f_3(x)\approx 8.55, \quad x \gg 1. \quad (18)$$

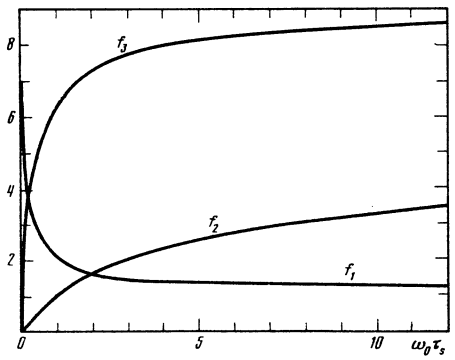

FIG. 3. Plots of f_1, f_2, and f_3 [see expressions (16) and (50)].

Figure 4a shows in the $T\Delta$ plane the regions of applicability of the expressions obtained for the case of weak and intermediate coupling ($\alpha^* \lesssim 6, R \lesssim \hbar\omega_0, E_0 \lesssim \hbar\omega_0$).

Equation (16) is valid everywhere except in the region $\Delta \lesssim \hbar\omega_0$, in which the absorption is determined by a small number of phonons, and in the region $(\Delta/h_0)^{3/2} \times \hbar\omega_0/\alpha^* T \lesssim 1$ ($\tau_s\Delta \lesssim h$), where the very high temperature causes the absorption to be large and to depend nonexponentially on Δ.

The region of applicability of the quasiclassical

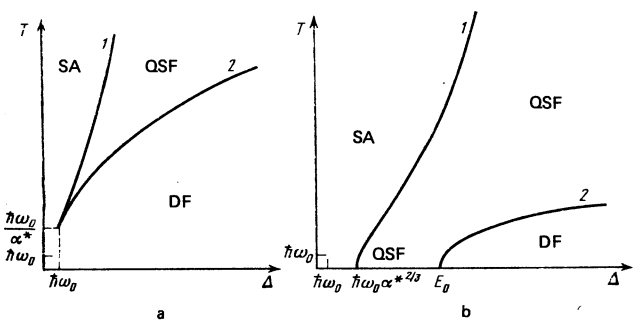

FIG. 4. Region of applicability of the quasistatic and dynamic approximations in the $T\Delta$ plane. a) Weak and intermediate coupling; b) strong coupling. 1) $\tau_s\Delta \sim \hbar$, 2) $\tau_s\omega_0 \sim 1$. E_0 is the energy of the free state. QSF and DF are the regions of quasistatic and dynamic fluctuations, and SA is the region of strong (non-exponential) absorption.

asymptote ($\omega_0\tau_s \ll 1$) (16) is quite narrow in the case of weak coupling and lies entirely in the region of high temperatures ($T \gg \hbar\omega_0$) and very large deficits Δ. At ordinary temperatures the quasistatic situation cannot be realized in the case of weak and intermediate coupling. The entire region of low and medium temperatures is described by the dynamic asymptotic relation (16).

Figure 4b deals with the case of strong coupling. In this case Eq. (16) describes only a frequency lying below E_0 ($\Delta > |E_0|$). In expressions (12a) and (16) it is necessary here to replace Δ by $\Delta = \Delta - |E_0|$. Let us analyze the region $\Delta < |E_0|$. At high temperature ($T \gg \hbar\omega_0$) this entire region (until the condition $\tau_s \Delta \gg \hbar$ is violated) is described by the quasistatic equation (11). At low temperature, the quasistatic approximation is valid only in a narrow region $[|E_0|(\hbar\omega_0)^2]^{1/3} \ll \Delta \ll |E_0|$, and at $\Delta \lesssim |E_0|$ there is realized an intermediate case which, however, is not described by expression (16), since the principal role is played in this case by virtual phonons. This situation will not be studied in detail in the present paper. We note, however, that the self-consistent-field approximation (expressions (32)–(34) is valid here, too.

III. DERIVATION OF THE SELF-CONSISTENT-FIELD EQUATIONS

To calculate the absorption coefficient we use the known connection between the complex dielectric constant and the exact Green's function (GF) of an electron pair:

$$K_\omega(\Omega) = \frac{\Omega}{cn_0} \operatorname{Im} \varepsilon(\Omega) = \frac{4\pi e^2 s^2}{\Omega c n_0} g(\Delta), \tag{19}$$

$$g(\Delta) = \int d^3\mathbf{r} \operatorname{Im} G_{-\Delta}(\mathbf{r}, \mathbf{r}; 0, 0). \tag{20}$$

Here $\varepsilon(\Omega)$ is the complex dielectric constant, $G_E(\mathbf{R}_e, \mathbf{R}_h; \mathbf{R}_e', \mathbf{R}_h')$ is the exact GF of the electron-hole pair with energy E and takes into account both the direct Coulomb interaction of the electron and hole as well as their interaction with the phonon at the temperature T. Expression (20) contains the GF for identical coordinates of the electron and hole, since the electron and hole are created at the same point.

To obtain the self-consistent-field equations we represent the time-dependent GF in terms of a functional integral over the electron-hole-pair wave function $\psi(t, \mathbf{R}_e, \mathbf{R}_h)$:

$$G(\mathbf{R}_e, \mathbf{R}_h; \mathbf{R}_e', \mathbf{R}_h'; t) = -i \frac{\int D\psi\psi(t, \mathbf{R}_e, \mathbf{R}_h)\psi^*(0, \mathbf{R}_e', \mathbf{R}_h') \exp(iS\{\psi\})}{\int D\psi \exp(iS_0\{\psi\})}, \tag{21}$$

$$S_0\{\psi\} = \int_0^t dt_1 \langle \psi_1 | i\frac{\partial}{\partial t_1} - \hat{H}_0 | \psi_1 \rangle, \quad \hat{H}_0 = -\frac{\nabla_e^2}{2m_e} - \frac{\nabla_h^2}{2m_h} - \varkappa |\mathbf{R}_e - \mathbf{R}_h|^{-1}, \tag{22}$$

$$S = S_0 + S_{int}, \quad S_{int} = \frac{1}{2}\int_0^t\int_0^t dt_1\, dt_2\, K(t_1 - t_2) \times \langle\langle \psi_1^* \psi_2^* | U(\mathbf{R}_{e1}, \mathbf{R}_{h1}; \mathbf{R}_{e2}, \mathbf{R}_{h2}) | \psi_1 \psi_2 \rangle\rangle, \tag{23}$$

$$K(t_1 - t_2) = i\{(N+1)\exp(-i|t_1 - t_2|) + N\exp(i|t_1 - t_2|)\}, \tag{24}$$

$$U(\mathbf{R}_{e1}, \mathbf{R}_{eh}; \mathbf{R}_{e2}, \mathbf{R}_{h2}) = 2^{-1/2}\alpha_0\{-|\mathbf{R}_{e1} - \mathbf{R}_{e2}|^{-1} - |\mathbf{R}_{h1} - \mathbf{R}_{h2}|^{-1} + |\mathbf{R}_{e1} - \mathbf{R}_{h2}|^{-1} + |\mathbf{R}_{h1} - \mathbf{R}_{e2}|^{-1}\}. \tag{25}$$

Here and below we use units in which $\hbar = \omega_0 = m_0 = 1$ (m_0 is the mass of the free electron), α_0 is the polaron coupling constant divided by the mass of the free electron, and $\varkappa = e^2 m_0^{1/2}/\hbar^{3/2}\omega_0^{1/2}\varepsilon_\infty$ is the Coulomb coupling constant with account taken of the high-frequency part of the dielectric constant.

In (22) and (23) we put

$$\psi_1 \equiv \psi(t_1, \mathbf{R}_{e1}, \mathbf{R}_{h1}), \quad \psi_2 \equiv \psi(t_2, \mathbf{R}_{e2}, \mathbf{R}_{h2}), \quad D\psi \equiv D \operatorname{Re}\psi D \operatorname{Im}\psi,$$

the angle brackets denote integration with respect to $\mathbf{R}_e$ and $\mathbf{R}_h$; $S_0\{\psi\}$ is the action for the free exciton, and $S_{int}\{\psi\}$ describes the exciton effective retarded self-action with the aid of exchange of polarization phonons. The electron located at the instant t_1 at the point R_{e1} produces a lattice polarization that acts in succeeding instants of time both on the electron itself [the first term of (25)] and on the hole (third term). The factor $K(t_1 - t_2)$ describes the retardation effect. Similarly, the second and fourth terms of (25) describe respectively the self-action of the hole and its action (via the phonon) on the electron. Obviously, since the electron and hole have opposite signs, they are oppositely acted upon by one and the same polarization, therefore the signs of the corresponding terms in (25) are opposite.

Thus, S_{int} contains both the self-action of the electron and hole (the polaron effect) and their effective interaction. If the electron and hole are at rest (or move slowly enough), and the lattice manages to reach equilibrium, this interaction screens partially the "right-hand" Coulomb attraction, as a result of which ε_0 appears in the expression for $\varkappa$ in place of ε_∞.

The possibility of excluding the phonon fields and reducing the interaction of the exciton with the phonons to its effective self-action is the consequence of only the quadratic and linear dependences of the Lagrangian of the electron-phonon system on the phonon coordinates.[22,23] Consequently, it is possible to carry out integration over the phonon field in the initial functional integral. The result is Eq. (21).

We note also that the action used in the normalization factor [the denominator in (21)] was S_0 rather than the total action S, in contrast to standard field theory (see, e.g., Ref. 24). The reason is that in our problem only one electron and one hole take part—no pairs are produced, so that all the vacuum loops are identically equal to zero and all the diagrams in the expansion of (21) are connected. This is ensured by the use of retarded GF.[24]

When the energy deficit $\Delta \gg 1$, only diagrams with a large number of phonons (n and more) contribute to the absorption coefficient. The large number of phonons participating in the process make it possible to apply the saddle-point method in the calculation of the functional integral (21); this method is equivalent in the present situation to the self-consistent-field approxi-

mation.

By varying the argument of the exponential in (21), we obtain the following nonlinear Schrödinger equation for the saddle-point wave function:

$$\left\{ i\frac{\partial}{\partial t} - \hat{H}_0 - V\{\psi_0\} \right\} \psi_0 = 0, \tag{26}$$

$$V\{\psi_0\} = \int_0^\infty K(t_1-t_2)\, dt_2 \langle \psi_{02}^* | U(\mathbf{R}_{e1}, \mathbf{R}_{h1}; \mathbf{R}_{e2}, \mathbf{R}_{h2}) | \psi_{02} \rangle. \tag{27}$$

We note that when the action is independently varied with respect to ψ and ψ^*, an additional condition that the potential $V\{\psi_0\}$ be real is imposed. We, however, do not need this condition, for ultimately imaginary times will turn out to be essential and the potential is real automatically.

Since the pair energy is of the order of $\Delta \gg 1$, and the potential V changes within a time of the order of unity (or ω_0^{-1} in ordinary unit), the solution of (26) will be sought in the adiabatic approximation

$$\psi_0(t_1) = \exp\left(-i\int^{t_1} E(t')\,dt'\right) \nu^{1/2} \varphi(t_1). \tag{28}$$

Here $\varphi(t_1)$ is the wave function normalized to unity and slowly varying in time, and $E(t)$ is the energy, which also varies slowly in time. The factor $\nu^{1/2}$ was introduced because integration is carried out in (21) over all the wave functions with arbitrary normalization, and it is not obvious beforehand that the saddlepoint function is normalized to unity. From the Schrödinger equation (26) follows only the independence of the normalization of the time.

Carrying out in the integral (21) a shift by ψ_0, we obtain

$$G(\mathbf{R}_e', \mathbf{R}_h'; \mathbf{R}_e, \mathbf{R}_h, t) = -iF\{\varphi, \nu\} \nu \varphi(\mathbf{R}_e', \mathbf{R}_h', t) \varphi^*(\mathbf{R}_e, \mathbf{R}_h, 0) \exp(i\mathcal{S}\{\varphi, \nu\}), \tag{29}$$

$$\mathcal{S}\{\varphi, \nu\} = \int_0^t \left\{ (\nu-1)E(t_1) - \langle \varphi_1^* | \nu \hat{H}_0 + \frac{\nu^2}{2} V\{\varphi\} | \varphi_1 \rangle \right\} dt_1, \tag{30}$$

$$F\{\varphi, \nu\} = \frac{\int D\bar{\psi} \exp\left\{ i\int_0^t dt_1 \langle \bar{\psi}_1^* | i\frac{\partial}{\partial t_1} - \hat{H}_0 - \nu V\{\varphi\} | \bar{\psi}_1 \rangle \right\}}{\int D\psi \exp\{iS_0\{\psi\}\}}. \tag{31}$$

In (31), $\bar{\psi} = \psi - \psi_0$. When calculating the action we have neglected the time derivatives of the slowly varying quantities $\varphi(t_1)$ and $E(t_1)$.

The absence of vacuum loops from the theory makes the factor F that describes the vacuum polarization equal to unity. Indeed, all the diagrams in the expansion of (31) in V are identically equal to zero (since they do not contain free ends). From this we get $F = 1$.

The normalization constant ν is determined from the condition for the extremum of expression (30). It is easily seen that

$$\frac{\partial \mathcal{S}}{\partial \nu}\Big|_{\nu=1} = \int_0^t dt_1 \{ E(t_1) - \langle \varphi_1^* | \hat{H}_0 + V\{\varphi\} | \varphi_1 \rangle \}.$$

The right-hand side of the equation vanishes by virtue of Eqs. (26) and expression (28) in which we must put $\nu = 1$. Consequently, the stationary point is $\nu = 1$.

As will be shown later on, the essential role in the Fourier transformation of the GF is played by imaginary times ($t = -i\tau$). Making the substitution $t = -i\tau$, we obtain

$$G(\mathbf{R}_e', \mathbf{R}_h'; \mathbf{R}_e, \mathbf{R}_h, \tau) = -i\varphi(\mathbf{R}_e', \mathbf{R}_h', \tau)\varphi^*(\mathbf{R}_e, \mathbf{R}_h, 0)\exp(-\mathcal{S}\{\varphi\}), \tag{32}$$

where

$$\mathcal{S}\{\varphi\} = \int_0^\tau \langle \varphi_1^* | H_0 | \varphi_1 \rangle d\tau_1 + \frac{1}{2}\int_0^\tau \int_0^\tau d\tau_1 d\tau_2 K_1(\tau_1-\tau_2) \times \langle\langle \varphi_1^* \varphi_2^* | U(\mathbf{R}_{e1}, \mathbf{R}_{h1}; \mathbf{R}_{e2}, \mathbf{R}_{h2}) | \varphi_1 \varphi_2 \rangle\rangle, \tag{33}$$

and φ is determined from the condition that S be a minimum under the conditions $\langle \varphi^* | \varphi \rangle = 1$;

$$\left\{ E(\tau_1) - \hat{H}_0 - \int_0^\tau K_1(\tau_1-\tau_2) d\tau_2 \langle \varphi_2^* | U(\mathbf{R}_{e1}, \mathbf{R}_{h1}; \mathbf{R}_{e2}, \mathbf{R}_{h2}) | \varphi_2 \rangle \right\} \varphi_1 = 0, \tag{34}$$

$$K_1(\tau_2-\tau_1) = (N+1)\exp(-|\tau_1-\tau_2|) + N\exp|\tau_1-\tau_2|. \tag{35}$$

We emphasize once more than the condition for the applicability of self-consistent-field approximation is a large number of phonons (real or virtual) participating simultaneously in the process. It does not matter whether the cause of the presence of the large number of phonons is the impossibility of realizing the process (in this case–absorption of a photon with an energy deficit $\Delta \gg 1$) by a small number of phonons, the high temperature, or the large value of the coupling constant α.

IV. SOLUTION OF THE SELF-CONSISTENT-FIELD EQUATIONS

For the reason discussed in Sec. II, we shall disregard the Coulomb interaction in the Hamiltonian $\hat{H}_0$. In addition, we neglect initially the contribution of the virtual phonons [unity in the first term of (35)].

Obviously, this can be done at high temperature, when $N \gg 1$, but it is valid also at $N \lesssim 1$, inasmuch as the important role in the integral (33) is played by $|\tau_1 - \tau_2|$ that are so large that the entire first term with the damped exponential in (35) can be neglected compared with the second (with the rising exponential). The possibility of neglecting unity in (35) is a reflection of the already noted need for participation of a large (compared with unity) number of phonons in the process.

Let $L \gg a_e, a_h$ be the distance between the electron and hole wells. Then the interaction can be regarded as weak and $\varphi(\mathbf{R}_e, \mathbf{R}_h, \tau) = \varphi_e(\mathbf{R}_e, \tau)\varphi_h(\mathbf{R}_h, \tau)$, while expression (33) takes the form

$$\mathcal{S} = \mathcal{S}_e + \mathcal{S}_h + \frac{\alpha_0}{\sqrt{2}L}\int_0^\tau \int_0^\tau d\tau_1 d\tau_2 K_1(\tau_1-\tau_2), \tag{36}$$

$$\mathcal{S}_i = \min\left\{ \int_0^\tau d\tau_1 \int \frac{1}{2m_i} |\nabla\varphi_i(\tau_1, \mathbf{R}_1)|^2 d^3\mathbf{R}_1 - \frac{\alpha_0}{2\sqrt{2}}\int_0^\tau \int_0^\tau K_1(\tau_1-\tau_2) d\tau_1 d\tau_2 \cdot \int\int |\mathbf{R}_1-\mathbf{R}_2|^{-1} |\varphi_i(\tau_1, \mathbf{R}_1)\varphi_i(\tau_2, \mathbf{R}_2)|^2 d^3\mathbf{R}_1 d^3\mathbf{R}_2 \right\}. \tag{37}$$

Making in (37) the substitutions $\varphi_i = (m_i\alpha_0 N)^{3/2}\varphi(\mathbf{r})$ and $\mathbf{R} = (m_i\alpha_0 N)^{-1}\mathbf{r}$, which preserves the normalization, and neglecting the virtual phonons, we obtain

$$\tilde{S}_i = -m_i\alpha_0^2 N^2 I(\tau), \tag{38}$$

$$I(\tau) = \max\left\{-\int_0^\tau d\tau_1 \int \frac{1}{2}|\nabla\varphi(\mathbf{r}_1,\tau_1)|^2 d^3\mathbf{r}_1 + \frac{1}{\sqrt{2}}\int_0^\tau\int_0^\tau \mathrm{ch}(\tau_1-\tau_2)\,d\tau_1 d\tau_2 \int\int |\mathbf{r}_1-\mathbf{r}_2|^{-1}|\varphi(\mathbf{r}_1,\tau_1)\varphi(\mathbf{r}_2,\tau_2)|^2 d^3\mathbf{r}_1 d^3\mathbf{r}_2\right\}. \tag{39}$$

The maximum should be obtained under the condition $\int d^3\mathbf{r}_1 |\varphi(\mathbf{r}_1,\tau_1)|^2 = 1$.

The Schrödinger equation in the region of the well is of the form

$$\left\{E(\tau_1) + \frac{\nabla_{\mathbf{r}_1}^2}{2} + \sqrt{2}\int \mathrm{ch}(\tau_1-\tau_2)\,d\tau_2 \int d^3\mathbf{r}_2 |\mathbf{r}_1-\mathbf{r}_2|^{-1} \times |\varphi(\mathbf{r}_2,\tau_2)|^2\right\}\varphi(\mathbf{r}_1,\tau_1) = 0. \tag{40}$$

Substituting (38) in (36), we obtain

$$\tilde{S} = -\alpha^{*2}N^2 I(\tau) + 2\sqrt{2}\alpha_0 N(\mathrm{ch}\,\tau - 1)/L. \tag{41}$$

The first term is due to the self-action, and the second is due to the interaction of the electron and the hole and is smaller than the first in all cases (see Sec. II).

Let for the sake of argument $m_h > m_e$. It is then obvious that the probability of finding the electron and hole at the same point is ensured by the tunneling of the electron from its localization region into the hole localization region. We write down for the electron a Schrödinger equation that is valid far from its localization region,

$$\left\{E_e(\tau_1) + \frac{\nabla^2}{2m_e} + \alpha_0 N\sqrt{2}\int_0^\tau d\tau_2\,\mathrm{ch}(\tau_2-\tau_1) \times [|\mathbf{R}_e|^{-1} - |\mathbf{L}-\mathbf{R}_e|^{-1}]\right\}\varphi_e(\tau_1,\mathbf{R}_e) = 0. \tag{42}$$

The first term in the potential energy describes the interaction with "its own" polarization well, and the second with that of the hole. Both interactions have a Coulomb form at large distances, with the time-dependent interaction constants of equal absolute value and of opposite sign. $\mathbf{R}_e$ is reckoned from the center of the electron jacket.

The wave function of the electron in the localization region of the hole can be obtained from (42):

$$(L) = \frac{\mathrm{const}}{L a_e^{1/2}}\exp(-L/a_e), \qquad a_e(\tau_1) = [2m_e|E_e(\tau_1)|]^{-1/2}. \tag{43}$$

Here $a_0(\tau_1)$ is the radius of the electron well. Expression (43) is valid at $L \gg a_e$. The constant in (43) depend on the type of wave function at short distances. Although (42) contains a Coulomb interaction with polarization wells, there is no Coulomb logarithm in (43), since the Coulomb logarithms corresponding to attraction and repulsion in (42) cancel each other.

It is seen from (23) that $\varphi(\tau_1) = \varphi(\tau - \tau_1)$, and in particular $\varphi(0) = \varphi(\tau)$. Substututing (43) and (41) in (32) and integrating, we obtain

$$g(\tau) = -i\int \varphi_h(\mathbf{R},0)\varphi_h^*(\mathbf{R}+\mathbf{r},0)\varphi_e(\mathbf{L}+\mathbf{R},0)\varphi_e^*(\mathbf{L}+\mathbf{R}+\mathbf{r},0)\,e^{-\tilde{S}}\,d^3\mathbf{r}.$$

Here $\mathbf{R}$ is the coordinate of the center of the hole well.

It is clear that the integral with respect to $\mathbf{r}$ converges in the region $\mathbf{r} \sim a_h(0)$. In this region φ_e does not have time to change greatly (at comparable masses m_e and m_h, the value of φ_e changes by an amount equal to itself, and if the masses differ greatly it does not change at all). Therefore, accurate to a number of the order of unity in the pre-exponential factor, we can write

$$g(\tau) = -i\cdot\mathrm{const}\cdot|\varphi_e(L)|^2 e^{-\tilde{S}} = -i\frac{\mathrm{const}}{L^3 a_e(0)}\exp\left\{\alpha^{*2}N^2 I(\tau) - \frac{2\sqrt{2}\alpha^* N(\mathrm{ch}\,\tau - 1)}{LM^{1/2}} - \frac{2L}{a_e(0)}\right\}. \tag{44}$$

We have chosen $\mathbf{R} = 0$, since this ensures a maximum of $g(\tau)$. The value of L must also be chosen from the condition that the argument of the exponential in (44) be a maximum. As a result we have

$$L_{opt} = [\sqrt{2}\alpha^* N(\mathrm{ch}\,\tau - 1)a_e(0)/M^{1/2}]^{1/3},$$

$$g(\tau) = -i\cdot\mathrm{const}\frac{M^{1/2}}{\alpha^* N a_e^2(0)(\mathrm{ch}\,\tau - 1)} \times \exp\left\{\alpha^{*2}N^2 I(\tau) - 4\left[\frac{\sqrt{2}\alpha^* N(\mathrm{ch}\,\tau-1)}{M^{1/2}a_e(0)}\right]^{1/3}\right\}. \tag{45}$$

To determine $E_e(0)$ and $a_e(0)$ we shall use the following relation:

$$E(0) = E(\tau) = -dI(\tau)/d\tau. \tag{46}$$

Indeed, differentiating (39) we obtain

$$\frac{dI(\tau)}{d\tau} = \frac{\partial I(\tau,\varphi=\varphi_0)}{\partial\tau} + \int d\tau_1 \left\langle \frac{\partial\varphi_0}{\partial t}\middle| \frac{\delta I(\tau,\varphi=\varphi_0)}{\delta\varphi}\right\rangle. \tag{47}$$

Here $I(\tau,\varphi)$ is the varied functional that stands in the right-hand side of (39), and φ_0 is the extremal value. The second term in (47) is zero because $I(\tau,\varphi_0)$ is extremal, and the first coincides with $-E(\tau)$, as can be easily verified by direct differentiation and comparison with (40).

From (46) we obtain

$$a_e(0) = (2m_e|E_e(0)|)^{-1/2} = [(m_e/M^{1/2})\alpha^* N]^{-1}(2I')^{-1/2}. \tag{48}$$

Substituting (48) in (45), we obtain the final expression for $g(\tau)$:

$$g(\tau) = -i\cdot\mathrm{const}\cdot m_e^{3/2}(m_e/M)^{1/6}\alpha^* N I'(\tau)(\mathrm{ch}\,\tau-1)^{-1} \times \exp\{\alpha^{*2}N^2 I(\tau) - 4\alpha^* N(m_e/M)^{1/3}[2(\mathrm{ch}\,\tau-1)I''^{1/2}(\tau)]^{1/3}\}.$$

To calculate the absorption coefficient, we must take the Fourier transform $g(\tau)$ of the retarded:

$$g(\Delta) = -\int_0^\infty g(it)e^{-i\Delta t}dt.$$

The saddle point $t = -i\tau_0$ lies on the imaginary axis, and the integration contour should have the form shown in Fig. 5. Only the horizontal section of the contour contributes to the imaginary part of $g(\Delta)$. The saddle point τ_0 is determined from the equation

$$\frac{dI(\tau)}{d\tau}\Big|_{\tau_0} = x^2, \quad x = \omega_0\tau_e = \Delta^{1/2}/\alpha^* N. \tag{49}$$

[To find the saddle point it is necessary to take into account only the principal (first) term in the exponential.] Carrying out the integration by the saddle-point method, we obtain

$$\mathrm{Im}\,g(\Delta) = -\mathrm{const}\cdot\mathrm{Im}\,g(\tau_0)(\alpha^* N)^{-1}(I''(\tau_0))^{-1/3}e^{-\Delta\tau_0}.$$

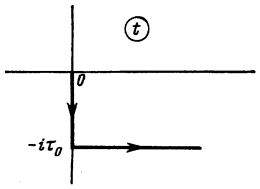

FIG. 5. Integration contour for the Fourier transform of $g(t)$.

We arrive ultimately at expression (16), where

$$f_1(x)=\frac{4x^2}{[I''(\tau_0)]^{1/3}(\operatorname{ch}\tau_0-1)},\quad f_2(x)=\tau_0-x^{-2}I(\tau_0),$$

$$f_3(x)=4\left[\frac{2(\operatorname{ch}\tau_0-1)}{x}\right]^{1/3}. \tag{50}$$

The functions I, f_1, f_2, and f_3 were calculated by a direct variational method with a computer. When the functional (39) was maximized, the trial functions were chosen in the form

$$\varphi_\tau(\tau_1,\mathbf{r})=A(\tau,\tau_1)\left(1+\frac{r}{2a(\tau_1,\tau)}\right)\exp\left(-\frac{r}{2a(\tau,\tau_1)}\right),$$

where

$$a(\tau_1,\tau)=\frac{a(\tau)}{\operatorname{ch}[\gamma(\tau)(\tau_1-\tau/2)]},$$

$A(\tau,\tau_1)$ was determined from the conditions that φ be normalized.

In the quasistatic case ($x\ll 1$), τ_0 is also small ($\tau_0\sim x$). Therefore $\cosh(\tau_1-\tau_2)\approx 1$ and φ can be regarded as independent of time. Then

$$I(\tau)=\max\left\{-\tau\int\frac{1}{2}|\nabla\varphi|^2d^3\mathbf{r}_1+\frac{\tau^2}{\sqrt{2}}\int\int|\mathbf{r}_1-\mathbf{r}_2|^{-1}\times|\varphi(\mathbf{r}_1)\varphi(\mathbf{r}_2)|^2d^3\mathbf{r}_1d^3\mathbf{r}_2\right\}.$$

Making the substitutions $\varphi=\tau^{3/2}\psi$ and $\mathbf{r}=\tau^{-1}\rho$, we obtain $I(\tau)=\tau^3I_1$. From (49) we get

$$\tau_0=x(3I_1)^{-1/3}, \tag{51}$$

$$I_1=\max\left\{-\int\frac{|\nabla\psi|^2}{2}d^3\rho_1+\frac{1}{\sqrt{2}}\int\int|\rho_1-\rho_2|^{-1}\times|\psi(\rho_1)\psi(\rho_2)|^2d^3\rho_1d^3\rho_2\right\}=0.109$$

is the energy functional investigated by Pekar[19] in the theory of a strong-coupling polaron. Using (50) and (51), it is easy to obtain the quasistatic asymptotics (17) for f_1, f_2, and f_3.

In the dynamic case ($\varphi x\gg 1$) we have $\tau_0\sim\ln x>1$. The integrals with respect to time in (39) converge near the end points of the interval $(0,\tau)$. We can then write

$$I(\tau)=2\max\left\{-\int_0^\infty d\tau_1\int\frac{1}{2}|\nabla\varphi(\mathbf{r}_1,\tau_1)|^2d^3\mathbf{r}_1+\frac{1}{\sqrt{2}}\int\int\frac{1}{2}e^{\tau-\tau_1-\tau_2}d\tau_1d\tau_2\iint|\mathbf{r}_1-\mathbf{r}_2|^{-1}|\varphi(\mathbf{r}_1,\tau_1)\varphi(\mathbf{r}_2,\tau_2)|^2d^3\mathbf{r}_1d^3\mathbf{r}_2\right\}.$$

We have subdivided the interval $(0,\tau)$ in to two: $(0,\tau/2)$ and $(\tau/2,\tau)$, and in the second we made the substitution $\tau_1\to\tau-\tau_1$ and extended the integration to infinity by virtue of the rapid convergence of the integral.

Making now the substututions $\varphi=((1/2)e^\tau)^{3/2}\psi$ and $\mathbf{r}=((1/2)e^\tau)^{-1}\rho$, we obtain $I(\tau)=(1/2)e^{2\tau}I_2$. From (49) we get

$$\tau_0=\ln(x/I_2^{1/3}),$$

$$I_2=\max\left\{-\int_0^\infty d\tau_1\int\frac{1}{2}|\nabla\psi(\rho_1,\tau_1)|^2d^3\rho_1+\frac{1}{\sqrt{2}}\int_0^\infty\int_0^\infty e^{-\tau_1-\tau_2}d\tau_1d\tau_2\iint|\rho_1-\rho_2|^{-1}|\psi(\rho_1,\tau_1)\times\psi(\rho_2,\tau_2)|^2d^3\rho_1d^3\rho_2\right\}=0.048. \tag{52}$$

From (52) and (50) we get the dynamic asymptotics (18) for f_1, f_2, and f_3.

Let us see now what results from allowance for the virtual phonons. As already noted, below the renormalized absorption edge the quasistatic case can be realized only if $N\gg 1$. Consequently, in this situation the virtual phonons need not be considered. In the dynamic case $\tau\gg 1$, therefore the first term in (35) can be substantial only if $N\ll 1$. We shall show that in this case allowance for the virtual phonons [unity in (35)] leads to a renormalization of the absorption edge, as a result of which it is necessary to make the substitution $\Delta\to\tilde{\Delta}=\Delta-|E_0|$ in all the expressions. Indeed, the second term in (35) attenuates exponentially with increasing distance between τ_1 or τ_2 and the end points of the interval $(0,\tau)$ in its inner region, but in the inner region the principal role is played just by the term with unity in (35), which does not contain the small factor N. Thus, the action correction due to the virtual phonons is determined by the inner region of the interval $(0,\tau)$ and is given by

$$\delta S_i=\min\left\{\int_0^\tau d\tau_1\int\frac{|\nabla\varphi_i|^2}{2m_i}d^3r_1-\frac{\alpha_0}{2\sqrt{2}}\int_0^\tau\int_0^\tau e^{-|\tau_1-\tau_2|}d\tau_1d\tau_2\int\int|\mathbf{r}_1-\mathbf{r}_2|^{-1}|\varphi_{i1}\varphi_{i2}|^2d^3\mathbf{r}_1d^3\mathbf{r}_2\right\},$$

φ_i does not depend on the time, since $\tau\gg 1$. As a result, integrating with respect to time, we obtain again the Pekar energy functional[19] and ultimately

$$\delta S_i=-m_i\alpha_0^2I_1\tau,\ \delta S=-\alpha^{*2}I_1\tau=-|E_0|\tau. \tag{53}$$

Allowance for δS leads thus to replacement of Δ by $\tilde{\Delta}=\Delta-|E_0|$, i.e., to a renormalization of the absorption energy. In addition, it must be noted here that far from the end points of the interval $(0,\tau)$ the Coulomb interaction is no longer overscreened, so that E_0 should be taken to be not $-\alpha^{*2}I_1$, but the binding energy that takes into account the Coulomb interaction of the electron and the hole. The renormalization is important only in the case of strong coupling ($E_0\gg 1$).

V. CONCLUSIONS

The main results of the present paper are reduced to the following.

1. We have demonstrated the existence of two limiting cases of interband absorption of light with participation

of a large number of LO phonons: quasistatic and dynamic. In the first case, the lattice does not manage to change its configuration during the time of the absorption, while in the second, on the contrary, it executes many oscillations. The criterion that separates these two situations is the quantity $\omega_0\tau_s$, where ω_0 is the phonon frequency and τ_s is the lifetime of the state.

In the quasistatic case the absorption probability is determined mainly by the probability of the onset of a fluctuation well having a level with an energy equal to the photon energy deficit. In the dynamic case, the absorption coefficient is proportional to the probability of simultaneous absorption of n phonons, where $n=[\Delta/\hbar\omega_0]+1$ is the smallest number needed to make up for the energy deficit.

The characteristic size of the fluctuation well (or the phonon wavelength) coincides with the wavelength of the particle (electron or hole), therefore the fluctuations cannot be regarded as homogeneous, and the particles have essentially a quantum motion.

2. We have demonstrated the possibility of fluctuation overscreening of the Coulomb attraction, as a result of which large distances between the electron and hole are energywise more profitable. This makes it possible to simplify the problem greatly, by regarding the interaction between them as weak. The possibility of overscreening is due to the specifics of the long-range interaction with LO phonons.

3. We derived self-consistent-field equations that make it possible to solve in a unified manner the problem in both limiting cases, as well as in the intermediate region. The condition for the applicability of the method is a large number of phonons participating in the process. The equations are valid also in the absence of overscreening. The method makes possible a calculation of not only the exponential factor of the absorption coefficient, but also the pre-exponential one. Similar equations can be easily derived also for processes in which phonons of any other type participate.

4. In the present paper, the self-consistent-field equations were solved in the strong-overscreening approximation (when the Coulomb attraction can be entirely neglected). A single answer was obtained, capable of describing the entire range of temperatures and light frequencies lying below the absorption edge renormalized on account of the polaron effect at an arbitrary binding force. In the case of strong coupling the self-consistent-field method is applicable also for absorption above the renormalized edge. This situation is considered in the paper only under the condition that the quasistatic approximation is valid.

It is of interest to investigate exitonic effects that are essential in the non-overscreened situation, and also to consider in greater detail the absorption above the renormalized edge of the band. We emphasize that all this can be done within the framework of the self-consistent-field approximation proposed in the present paper.

5. The expression obtained for the light absorption coefficient below the renormalized edge corresponds to the Urbach rule (1) only at low temperatures, in which case $\sigma \to 1$ and does not depend on T. With increasing temperatures, the frequency dependence of $K(\Omega)$ becomes stronger than in Eq. (1): The linear dependence on Δ in the exponential gives way first to $\Delta\ln(\Delta^{1/2})$ and next to $\Delta^{3/2}$.

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[1]F. Urbach, Phys. Rev. **92**, 1324 (1953).
[2]R. S. Knox, Theory of Excitons, Academic, 1963.
[3]T. Tomiki, T. Miyata, and H. Tsukamoto, Z. Naturforsch. **29a**, 145 (1974).
[4]V. V. Galavanov, N. I. Novak and O. S. Shevchuk, Fiz. Tekh. Poluprov. **5**, 2365 (1971) [Sov. Phys. Semicond. **5**, 2075 (1971)].
[5]J. R. Dixon and J. M. Ellis, Phys. Rev. **123**, 1560 (1961).
[6]A. S. Davydov, Theory of Molecular Excitons, Plenum, 1971.
[7]D. L. Dexter, Nuovo Cim. Suppl. **7**, 245 (1958).
[8]Y. Toyozawa, Prog. Theor. Phys. **22**, 445 (1959); **27**, 89 (1962).
[9]D. L. Dexter, Phys. Rev. Lett. **19**, 1383 (1967).
[10]J. D. Dow and D. Redfield, Phys. Rev. **B5**, 594 (1972).
[11]G. O. Mahan, Phys. Rev. **145**, 602 (1966).
[12]E. P. Sinyavakiĭ, Fiz. Tverd. Tela (Leningrad) **13**, 2089 (1971) [Sov. Phys. Sol. State **13**, 1750 (1972)].
[13]A. S. Davydov and A. F. Lubenko, Doklady Akad. Nauk SSSR **179**, 1301 (1968) [Sov. Phys. Dokl. **13**, 325 (1968)].
[14]V. D. Kagan, Fiz. Tverd. Tela (Leningrad) **17**, 2578 (1975) [Sov. Phys. Sol. State **25**, 1717 (1975)].
[15]A. Sumi and Y. Toyozawa, J. Phys. Soc. Jpn. **35**, 137 (1973).
[16]V. M. Agranovich, Teoriya eksitonov (Exciton Theory), Nauka, 1968.
[17]J. D. Dow and D. Redfield, Phys. Rev. **B1**, 3351 (1970).
[18]I. A. Merkulov and V. I. Perel, Phys. Lett. **45a**, 83 (1973).
[19]S. I. Pekar, Issledovaniya po elektronnoi teorii kristallov (Research into the Crystal Theory of Crystals), Gostekhizdat, 1951 [US AEC-tr-5575, 1963].
[20]A. I. Ansel'm, Vvedenie v teoriyu poluprovodnikov (Introduction to the Theory of Semiconductors), Nauka, 1978.
[21]S. V. Iordanskiĭ and É. I. Rashba, Zh. Eksp. Teor. Fiz. **74**, 1872 (1978) [Sov. Phys. JETP **47**, 975 (1978)].
[22]R. P. Feynman, Phys. Rev. **97**, 660 (1955).
[23]D. Appel, transl. in: Polyarony (Polarons), Yu. A. Firsov, ed., Nauka, 1975.
[24]A. N. Vasil'ev, Funktsional'nye metody v kvantovoĭ teorii polya i statistike (Functional Methods in Quantum Field Theory and Statistics), Leningrad Univ. Press, 1976.

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