Long-wave tail of light absorption by polar crystals in an electric field

A. S. Ioselevich and A. V. Maslov

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted 1 February 1982)
Zh. Eksp. Teor. Fiz. **83**, 368–379 (July 1982)

Light absorption by a polar crystal in an electric field was investigated at a light frequency $\hbar\Omega$ smaller than the band gap E_g. The manner in which the intrinsic absorption (in a zero field), previously investigated and describable in a number of cases by the Urnach rule, goes over in a strong field into the Keldysh-Franz effect is traced. It is shown that not too strong an electric field orients somewhat and deforms ("polarizes") the optimal fluctuations. The field and temperature dependences of the weak-field correction to the absorption coefficient agree with the experimental data. When the field reaches a certain critical value the optimal fluctuation dissociates; the electron is detached from the hole, is delocalized, and goes over into a tunnel state. With further increase of the field the role of the light particle (electron) increases and that of the heavy one (hole) decreases, while the expression for the absorption coefficient comes closer to the relation typical of the Keldysh-Franz effect.

PACS numbers: 78.20.Jq

1. INTRODUCTION

As shown by Keldysh[1] and Franz,[2] in the presence of an external electric field the light absorption by a crystal in the forbidden band acquires and exponential tail due to the tunneling of an electron and a hole (Fig. 1). This phenomenon has been investigated thoroughly both experimentally and theoretically (see, e.g., the review of Aronov and Ioselevich[3] and the references therein). In the case of simple bands, the absorption coefficient K_a is described by the expression

$$K_a\sim\exp\left(-\frac{4\sqrt{2}}{3}\frac{\Delta^{3/2}m^{*1/2}}{\hbar eE}\right). \quad (1)$$

here $\Delta=E_g-\hbar\Omega$ is the phonon energy deficit, E is the electric field, $m^*=(m_e^{-1}+m_h^{-1})^{-1}$ is the reduced effective mass, and m_e and m_h are respectively the electron and hole masses, which we assume in this paper to differ greatly:

$$\gamma=m_e/m_h\ll 1, \quad (2)$$

so that $m^*\approx m_e$. We have not included in (1) the pre-exponential factor and the terms resulting from the Coulomb attraction of the electron and the hole (see Ref. 3).

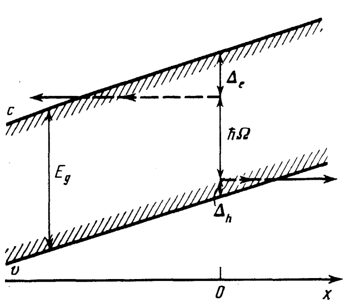

FIG. 1. Picture of the process of electron-hole production in the Keldysh-Franz effect. The electron energy (c) and the hole (v) energy bands are inclined to the electric field. The electron and hole are produced at the point $x=0$, in a classically inaccessible region, and then tunnel along the paths shown by the arrows. The hole energy deficit Δ_h is small compared with the electron Δ_e to the extent that γ is small.

In many substances, however, the exponential absorption tail is observed also in the absence of external fields. Such tails, as a rule, satisfy the well known Urbach rule,[4] which states that the logarithm of the absorption coefficient K_a depends linearly on the light frequency Ω. A detailed review of the available data can be found in Refs. 5 and 6. The Urbach tails are apparently the result of thermal vibrations of the lattice: a certain number of phonons is absorbed simultaneously with the phonon, so that their combined energy is sufficient to produce an electron-hole pair. If the required number of phonons is small (one or two), indirect absorption thresholds are observed. In this region, the dependence of the absorption coefficient on the frequency and on other parameters has been well investigated and is not exponential. An Urbach tail is observed, on the other hand, if many phonons take part in the process. The theory of light absorption in this case (for polar crystals, where the principal role is played by LO phonons) was developed in Refs. 7 and 8. Reference 7 contains an analysis of the earlier theories and the necessary bibliography. Without allowance for the exciton effects and the pre-exponential factor, the following formula holds for K_a:

$$K_a\sim\exp\left\{-\frac{\Delta}{\hbar\omega}f_2(\omega\tau_s)-\left(\frac{\gamma\Delta}{\hbar\omega}\right)^{1/2}f_3(\omega\tau_s)\right\}, \quad (3)$$

$$\tau_s=\omega^{-1}(\Delta/\hbar\omega)^{1/2}/\alpha^*N, \quad (4)$$

where ω is the frequency of the LO phonons, $N=[\exp(\hbar\omega/T)-1]^{-1}$ is the phonon occupation number, $\alpha^*=(\alpha_e^2+\alpha_h^2)^{-1/2}\approx\alpha_h$, α_e and α_h are the polaron-coupling constants for the electron and hole respectively, and τ_s is the characteristic time of LO-phonon absorption by an electron-hole pair with energy on the order of Δ. Plots of the computer-calculated functions f_2 and f_3 are given in Ref. 7. $f_2(x)=1.17x$, $f_3(x)=7x^{1/2}$, at $x\ll 1$ and $f_2(x)=\ln 2.77x$, $f_3(x)=8.55$ at $x\gg 1$. Formula (3) was derived under the assumption that both terms in the argument of the exponential are large (see Ref. 7 for

 0038-5646/82/070201-06$04.00

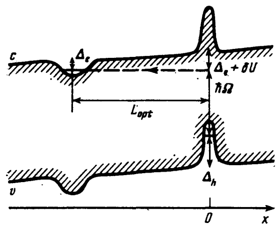

FIG. 2. Structure of optimal fluctuation in the overscreened case (in a weak field $E < E_2$). The optimal distance L_{opt} between wells is much larger than the size of the wells. The pair is produced at the point $x=0$, so that the hole is immediately located in its own well, and the electron must tunnel into its well along the path shown by the arrow. The energy shift in the electric field is $\delta U = -eL_{opt}E$. The hole interacts more strongly with the phonons, therefore $\Delta_e \sim \gamma\Delta_h \ll \Delta_h$.

details). The first term always exceeds the second. To make the second term likewise large compared with unity, it is necessary that γ be not too small:

$$\hbar\omega/\Delta f_s^2(\omega\tau_s) \ll \gamma \ll 1. \qquad (5)$$

The role of the LO phonons in the absorption of light reduces to formation of several (generally speaking nonstationary) fluctuation potential wells separately for the electron and for the hole (Fig. 2). At $\omega\tau_s \ll 1$ the wells are stationary, and at $\omega\tau_s \gg 1$ they vary strongly with time. As $\gamma \to 0$ (the left-hand side of inequality (5) is violated), no electron well is produced at all, and the electronic state remains free and delocalized.

The structure of the optimal fluctuation, shown in Fig. 2, can change if account is taken of the Coulomb attraction of the electron and hole.[8] At a small value of the exciton Rydberg, $R < 3\gamma\Delta$, the state has a spontaneous dipole moment eL_{opt} (the overscreened situation), which decrease with increasing R like

$$eL_{opt}^{R} = eL_{opt}^{R=0}\left(1-(R/3\gamma\Delta)^{1/2}\right)^{1/2} \qquad (6)$$

and vanishes identically at $R > 3\gamma\Delta$ (non-overscreened situation). In this case the fluctuation is spherically symmetric (Fig. 3). The presence or absence of a dipole moment becomes, of course, extremely important when the field is turned on.

It was shown in Ref. 8 that the condition (2) makes it possible to separate the motions of the electron and the hole, so that the problem can be solved with allowance for exciton effects. At the same time, the lighter particle (electron) plays only a secondary role in the Urbach absorption, which is determined mainly by the hole, which interacts more strongly with the LO phonons. In the Keldysh-Franz effect, on the contrary, the decisive role is played by the light electron, whose tunneling probability is much higher than that of the hole. Thus, if the masses differ greatly it is possible to separate to a large degree the field and phonon effects in absorption. Whereas the former affect mainly the electronic component of the electron-hole wave functions, the latter affect the hole component.

It is important to note the substantial difference between the proposed theory and that developed by Dow and Redfield[9] and already discussed by us in Ref. 7. When explaining the Urbach rule, Dow and Redfield attempt to reduce the interaction of the electron-hole pair with the phonon to the Keldysh-Franz effect in a random feld, which calls for assuming that the electric fields corresponding to the absorbed LO phonons are sufficiently static and homogeneous. As shown in Ref. 7, this assumption cannot be satisfied. One of the consequences ofthe Dow-Redfield theory is that the Urbach-absorption coefficient depends only on the reduced mass of the electron and hole, i.e., predominance of the light particle, just as in the Keldysh-Franz effect. In fact, however (see Ref. 7), the Urbach absorption depends mainly on the translational mass $M = m_h + m_e (M \approx m_h$ at $\gamma \ll 1)$, whereas the Keldysh Franz absorption depends on the reduced mass $m^* = (m_e^{-1} + m_h^{-1})^{-1} \approx m_e$. To compare the theory with experiment it is therefore not enough to know the values of m_e, which are known for many polar crystals (see Refs. 6, 10, and 11), and we must know also m_h, yet the data on the effective masses of holes in dielectrics are quite scanty.

When the field is turned on the absorption coefficient, of course, will not be a simple sum of the absorption coefficients described by expressions (1) and (3) and corresponding separately to each of the mechanisms (tunneling and fluctuation). For example, the following situation is possible: a heavy hole that interacts strongly with phonons is localized in a fluctuation well, while the light electron tunnels in the electric field (Fig. 4). It will be shown below that this is precisely the absorption mechanism realized in a sufficiently strong field.

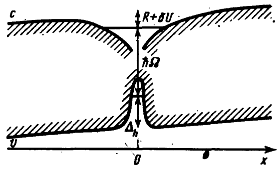

FIG. 3. Structure of optimal fluctuation in the non-overscreened case. The hole is localized in a narrow fluctuation well, and the electron forms near it a broad excitonic state (with size of the order of the exciton Bohr radius and with energy equal to the exciton Rydberg R). The weak electric field changes the exciton energy by δU.

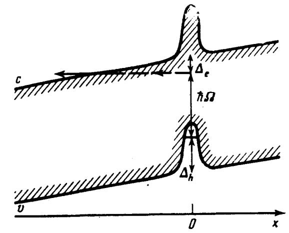

FIG. 4. Tunnel state at $E > E_2$. After production, the hole is in the well and the electron tunnels as in the Keldysh-Franz effect.

In a weak field the situation is different. Indeed, the tunnel state of the electron (Fig. 4) differs qualitatively from the situation in the fluctuation well (Figs. 2 and 3) used to describe the Urbach tail. It is clear at the same time that a weak enough electric field cannot alter qualitatively the structure of the state, and should simply distort somewhat the form of the optimal fluctuation, lowering the energy of the electron-hole pair. It turns out the tunnel state becomes advantageous only fields stronger than a certain critical field E_2, the hole remains localized, and only the light electron tunnels (Fig. 4). The hole delocalization takes place in a still stronger field, when the phonons are no longer effective in practice, and is therefore not considered here. Usually the critical field is apparently quite strong ($E_2 \sim 10^6$ V/cm), we therefore analyze in greatest detail (in Sec. 2) the case of weak fields $E < E_2$ and discuss the available experimental data.[11] The mechanisms for perturbing the optimal fluctuation with a weak field are quite diverse and differ greatly, depending on the structure of the initial unperturbed state. All these mechanism are analyzed and compared with experiment in Sec. 2. In Sec. 3 we describe the tunnel state. The main deductions are summarized in the Conclusion.

2. LIGHT ABSORPTION COEFFICIENT IN A RELATIVELY WEAK ELECTRIC FIELD

In this section we describe in detail the fluctuation distortion of an electric field in the simplest quasistatic case ($\omega\tau_s \ll 1$), when the fluctuation can be regarded as static. All the described effects occur also in the dynamic case ($\omega\tau_s \gg 1$), so that in the description of the latter we present only the final result and dwell only on specific dynamic effects.

Quasistatic case

We begin with the overscreened situation, when the fluctuation has a dipole moment. Obviously, in this case the energy of the state changes in the field **E** by an amount

$$\delta U = -e\mathbf{L}_{opt}\mathbf{E}. \tag{7}$$

In a weak field $E \ll E_2^{(0)}$ it can be assume that L_{opt} is not altered by the field and that the polarizing action of the field reduces to orienting a dipole moment of fixed magnitude. The absorption coefficient $K_a(E)$ is described at $E \ll E_2^{(0)}$ by the expression

$$K_a(E) = K_a(0)\frac{E_1^{(0)}}{E}\,\mathrm{sh}\,\frac{E}{E_1^{(0)}}, \tag{8}$$

$$E_1^{(0)} = 0.29\left[\frac{\gamma}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{3/2}(1-(R/3\gamma\Delta)^{1/2})\right]^{-1/2} E_2^{(0)}, \ll E_2^{(0)}, \tag{9}$$

$$E_2^{(0)} = 1.6(\hbar\omega^3\gamma m_e)^{1/2}\alpha_h N/e = 1.1(\varepsilon_\infty^{-1}-\varepsilon_0^{-1})\frac{em_e}{\hbar}\omega N. \tag{10}$$

In Eq. (10) we have expressed α_h in terms of the static and high-frequency dielectric constants ε_0 and ε_∞, respectively (see Ref. 7).

When the field E becomes comparable with $E_2^{(0)}$, the value of L_{opt} begins to increase, and the field stretches the dipole:

$$L_{opt}(E) = L_{opt}(0)(1-E/E_2^{(0)})^{-1/2} \tag{11}$$

Since $E_2^{(0)} \gg E_1^{(0)}$, the possibilities of orientational polarization have already been exhausted at $E \lesssim E_2^{(0)}$, and the dipole is oriented exactly along the field. For $K_a(E)$ at $E \lesssim E_2$ we have

$$K_a(E) = K_a(0)\frac{E_1^{(0)}}{2E}(1-E/E_2^{(0)})^{1/2} \times\exp\left\{7\left[\frac{\gamma}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{3/2}\left(1-\left(\frac{R}{3\gamma\Delta}\right)^{1/2}\right)\right]^{1/2}[1-(1-E/E_2^{(0)})^{1/2}]\right\}. \tag{12}$$

It follows from (11) that at $E = E_2^{(0)}$ the value of L_{opt} becomes infinite and the dipole is broken. As will be shown below, a tunnel state is produced in this case. At $E_1^{(0)} \ll E \ll E_2^{(0)}$ Eqs. (8) and (12) are joined together.

The derivation of Eqs. (7)–(11) is based on the following physical premises. It was shown in Refs. 7 and 8 that the electron-hole interaction energy takes, when the fluctuation overscreening is taken into account, the form

$$U = \frac{\hbar}{L}\left\{\left(\frac{6\Delta}{m_h}\right)^{1/2} - \left(\frac{2R}{m_e}\right)^{1/2}\right\}.$$

The probability of the onset of a fluctuation with the combined energy of the electron and hole, equal to $-\varepsilon$, is described by the expression

$$W \sim \exp\left\{-\frac{1.17}{\alpha^* N}\left(\frac{\varepsilon}{\hbar\omega}\right)^{3/2}\right\}. \tag{13}$$

The absorption coefficient is proportional to the product of W and the square $|J|^2$ of the overlap integral of the electron and hole wave functions:

$$|J|^2 \sim \exp\left\{-\frac{2Lm_e}{\hbar}\left(\frac{2\Delta}{m_h}\right)^{1/2}\right\}.$$

Substituting $\Delta + U$ in (13) in place of ε, expanding in powers of $U/\Delta \ll 1$, and the finding L_{opt} from the condition that the product $W|J|^2$ be a maximum, we obtain expression (3) at $\omega\tau_s \ll 1$. In the presence of an electric field, U must be replaced by $U + \delta U$, where δU is described by expression (7). We then obtain Eq. (11) for L_{opt} and, after integrating over all the possible directions of L, we obtain Eqs. (8) and (12) for $K_a(E)$. We do not present here the detailed derivation, since it is perfectly analogous to the calculations of Ref. 7.

In the overscreened situation (when $R > 3\gamma\Delta$) the dipole moment of the state is zero. At $R \gg 3\gamma\Delta$ the absorption coefficient K_a is described by formula (13), where ε must be replaced by $\bar{\Delta} = \Delta - R$ and α^* by α_h. It makes sense to take the excitonic effects into account if $R\partial \ln K_a/\partial\Delta \gg 1$, i.e., the Coulomb attraction leads to an exponential change of the absorption. The shift of the exciton ground-state energy is quadratic in the field[12]:

$$\delta U = -{}^9/_{16}\hbar^2 e^2 E^2/m_e R^2. \tag{14}$$

Replacing ε in (13) by $\bar{\Delta} + \delta U$ we get

$$K_a(E) = K_a(0)\exp\{-E^2/6\bar{E}_1^{(0)2}\}, \tag{15}$$

$$\bar{E}_1^{(0)} = 0.41\frac{R}{e}\left(\frac{m_e^2\omega^3}{\hbar(\Delta-R)}\right)^{1/4}(\alpha_h N)^{1/2}. \tag{16}$$

The field at which the tunnel state becomes more expedient is obtained in Sec. 3:

$$\tilde{E}_2^{(0)}=2.8\frac{(m_e\omega^3\hbar)^{1/2}}{e}\left(\frac{R}{\Delta-R}\right)^{1/2}\alpha_h N. \quad (17)$$

Comparing (16) with (17) we see that in the overscreened case $\tilde{E}_2^{(0)} \ll \tilde{E}_1^{(0)}$, therefore Eq. (15) must be expanded. It is also easy to verify that $\tilde{E}_2^{(0)} \ll R/ea_B$, i.e., $E_2^{(0)}$ is much weaker than the field at which the exciton is ionized ($\delta U \sim R$), thereby justifying the use of Eq. (14).

We emphasize that the influence of the electric field on the absorption reduces to a shift of the energy of the exciton ground state only in the overscreened case, when one can speak of the creation of an exciton that exists as a relatively stable and unchangeable formation during the entire absorption process.

The dynamic case ($\omega\tau_s \gg 1$)

In the overscreened case at $\omega\tau_s \ll 1$, just as $\omega\tau_s \gg 1$, the electric field orients and stretches the fluctuation. The quantity $L_{\text{opt}}(E)$ is determined as before by an expression such as (11), but the corresponding characteristic fields are then

$$E_1^{(\infty)}=0.65(m_e\hbar\omega^3)^{1/2}/\ln\left(\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\right), \quad (18)$$

$$E_2^{(\infty)}=2.8(m_e\gamma\Delta\omega^2)^{1/2}/\ln\left(\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\right) \gg E_1^{(\infty)}. \quad (19)$$

The difference between the expressions for K_a from Eqs. (8) and (12), however, does not reduce generally speaking to a simple replacement of $E_1^{(0)}$ and $E_2^{(0)}$ by $E_1^{(\infty)}$ and $E_2^{(\infty)}$. The point is that besides the effects considered in the preceding section the dynamic effect is characterized by one other mechanism whereby the electric field acts on the fluctuation.

As shown in Ref. 7, at $\omega\tau_s \gg 1$ the depth of the electron potential well decreases exponentially with an imaginary time. Therefore in the presence of the field it will become convenient for electron, at a certain instant of time, to tunnel from its "shallowed" well rather than continue to stay there. This phenomenon is similar to the tunneling, discussed in Ref. 8, of an electron from its well and formation of an exciton in the partially overscreened case. In other words, this combined process proceeds as follows: the electron first absorbs a certain optimal number of phonons, and then tunnels with a decreasing energy deficit. The ratio of this residual deficit to the initial one (Δ) turns out to be small if $E \ll E_2^{(\infty)}$.

We confine ourselves to the case of a weak field ($E \ll E_2^{(\infty)}$). An expression for the field correction to the below-barrier action is given in the Appendix. The final result is

$$K_a(E)=K_a(0)\frac{E_1^{(\infty)}}{E}\operatorname{sh}\frac{E}{E_1^{(\infty)}}\exp\left\{0.018\left(\frac{E}{E_1^{(\infty)}}\right)^2\right.$$
$$\left.\times\ln^3\left[\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\left(\frac{E}{E_2^{(\infty)}}\right)^2\right]\Big/\ln^2\left[\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\right]\right\}. \quad (20)$$

In the derivation of the last factor we have assumed that the field is still not too small ($E \gg E_1^{(\infty)}$), so that this factor is the principal one and determines the field dependence of K_a. In the opposite case (at $E \lesssim E_1$) it becomes numerically small and therefore unimportant.

In the overscreened situation, just as in the preceding section, the influence of the field reduces to replacement of R by $R+\delta U$, where δU is determined by Eq. (14). In analogy with (15) we obtain

$$K_a(E)=K_a(0)\exp\{-E^2/6\tilde{E}_1^{(\infty)2}\}, \quad (21)$$

$$\tilde{E}_1^{(\infty)}=0.54\frac{R}{e}\left(\frac{m_e\omega}{\hbar}\right)^{1/2}\Big/\ln^{1/2}\left[\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\right]. \quad (22)$$

The field at which the transition into the tunnel state takes place is

$$\tilde{E}_2^{(\infty)}=4.9(m_e\omega^2 R)^{1/2}/e\ln\left[\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\right]. \quad (23)$$

Comparison with the experimental data

Typical ionic crystals such as CuCl and TlCl are characterized by the large value $E_2^{(0)} \sim 10^6$ V/cm (according to the data of Ref. 11). Therefore from the experimental point of view greatest interest attaches to the case of "superweak" fields $E \ll E_1$. In this case Eqs. (8), (15), (20), and (21) can be expanded in powers of E/E_1, and for the relative absorption coefficient we have

$$\delta K_a=\frac{K_a(E)-K_a(0)}{K_a(0)}=\frac{E^2}{6E_1^2}, \quad (24)$$

where E_1 must be replaced by $E_1^{(0)}$, $\tilde{E}_1^{(0)}$, $E_1^{(\infty)}$ or $\tilde{E}_1^{(\infty)}$, depending on the relation between the parameters.

Mohler and Thomas[11] investigated experimentally the dependence of δK_a on the electric field and on the temperature. With changing temperature, the light frequency changed in such a way that $K_a(0)$ remained constant. On the basis of their measurements, Mohler and Thomas arrived at the relation

$$\delta K_a|_{K_a(0)=\text{const}} \sim E^2T^{-2}. \quad (25)$$

They used this result in Ref. 11 to compare the theoretical Urbach-absorption models proposed on the one hand by Dow and Redfield,[9] and on the other by Sumi and Toyozawa.[13] Mohler and Thomas reach the conclusion that the first model leads in accord with (25) to the relation $\delta K_a \sim E^2T^{-2}$, and the second to $\delta K_a \sim E^2T^{-1}$, causing them to prefer the first model. Their reasoning, however, is based on the incorrect premises of Ref. 9 and are therefore not satisfactory.

To compare Eq. (24) with the empirical (25) we consider separately the case of high and low temperatures.

At $T \gg \hbar\omega$ we have $N=T/\hbar\omega$ and $\omega\tau_s \ll 1$. From (3) and from the condition $K_a(0)=\text{const}$ we obtain $\Delta^{3/2}T^{-1}=\text{const}$, while from (9) we get $E_{(1)}^0 \sim (\Delta^{3/2}T^{-1})^{-1/2}T \sim T$. We arrive thus at the conclusion that (24) agrees with the rule (25) in the overscreened case. An analogous analysis for the non-overscreened case yields

$$\tilde{E}_1^{(0)} \sim (\Delta-R)^{-1/4}T^{1/2} \sim T^{-1/4}T^{1/2}=T^{1/4},$$

which contradicts (25).

At $T \ll \hbar\omega$ we have $N=\exp(-\hbar\omega/T)$, $\omega\tau_s \ll 1$. From (3) and the condition $K_a(0)=\text{const}$ we get $\Delta T^{-1}=\text{const}$, and from (28) we get $E_1^{(\infty)} \sim T$, which again agrees with (25). In the non-overscreened case $\tilde{E}_1^{(\infty)} \sim T^{1/2}$ in contradiction with (25).

The character of the field and temperature dependences of δK_a in the overscreened case thus coincides with that observed experimentally by Mohler and Thomas at both low and high temperatures.

A more accurate comparison of (24) with the results of Mohler and Thomas is unfortunately difficult, since the frequencies used in the experiment are not cited in Ref. 11.

3. ABSORPTION IN A STRONG FIELD

As can be seen from (6), at $E \geqslant E_2$ the electron is detached from the hole and the fluctuation "dissociates," The tunnel state already mentioned in the introduction sets in (Fig. 4). The structure of this state is quite simple, so that the derivation can be carried out for an arbitrary value of $\omega\tau_s$.

We divide the energy deficit Δ in two parts: let the hole be localized in a state with energy $-\Delta_h$, and let the electron tunnel with a deficit Δ_e; thus $\Delta_e + \Delta_h = \Delta$. The absorption coefficient is then proportional (with exponential accuracy) to the product of the electron tunneling probability (Eq. (1), where the substitution $\Delta \to \Delta_e$, $m^* \to m_e$ must be made) by the hole-localization probability (Eqs. (3) and (4), where $\Delta \to \Delta_h$, $\alpha^* \to \alpha_h$, and the second term in the exponential is discarded). As a result we have[1)]

$$K_a \sim \exp\left(-\frac{4\sqrt{2}}{3}\frac{\Delta_e^{3/2} m_e^{1/2}}{eE\hbar}\right)\exp\left(-\frac{\Delta_h}{\hbar\omega} f_2(\omega\tau_s^h)\right). \tag{26}$$

The values of Δ_e and Δ_h should be obtained from the condition of the maximum of (26). We begin with the case of "not too strong" a field $E_2 < E \ll E_3$, when it turns out that $\Delta_e \ll \Delta_h$ and the relative role of the field in the absorption is still small. Here

$$E_3 = \frac{2.8\omega(m_e\Delta)^{1/2}}{e\tau(\omega\tau_s)} = \gamma^{-1/2}E_2 \gg E_2; \tag{27}$$

$$\tau(x) = f_2(x) + \frac{x}{2} f_2'(x), \quad E_2(\omega\tau_s \ll 1) = E_2^{(0)}, \quad E_2(\omega\tau_s \gg 1) = E_2^{(\infty)}.$$

If $\Delta_e \ll \Delta_h$ it is easy to find the maximum of (26) by expanding $(\Delta/\hbar\omega) f_2(\omega\tau_s^h)$ in powers of Δ_e/Δ. We ultimately get

$$K_a(E) \sim K_a(0)\exp\left(\frac{\Delta}{3\hbar\omega}\frac{\tau(\omega\tau_s)E^2}{E_3^2}\right). \tag{28}$$

Comparing (28) with (3) at $\omega\tau_s \ll 1$ we easily verify that the tunnel state of the electron is indeed favored at $E > E_2^{(0)}$, and the localized state at $E < E_2^{(0)}$. The transition from the localized state to the tunnel state at $E = E_2^{(0)}$ is practically jumplike, with a very narrow transition region. The same holds also for the nonoverscreened case, where we obtain for the limiting field $E = \tilde{E}_2^{(0)}$ at $\omega\tau_s \ll 1$ and $E = \tilde{E}_2^{(\infty)}$ at $\omega\tau_s \gg 1$.

The situation is different in the dynamic ($\omega\tau_s \gg 1$) overscreened case. Here the transition is gradual, for as the field approaches the critical $E_2^{(\infty)}$ from below an ever decreasing fraction of the electron energy deficit $\Delta_e = \gamma\Delta$ is replenished by phonon absorption, and an ever larger one by tunneling.

We obtain now the maximum of expression (26) at an arbitrary ratio of E and E_3. Introducing the dimensionless quantities $\omega\tau_s = x$ and $\omega\tau_s^h = x_h$, we obtain

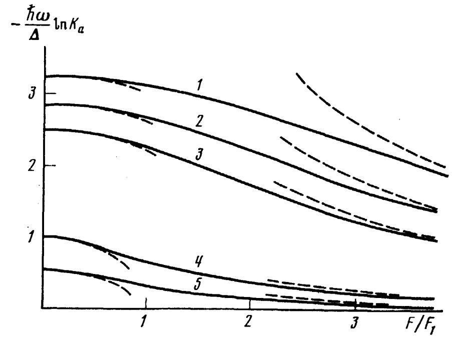

FIG. 5. Dependence of the logarithm of the absorption coefficient on the dimensionless field $E/E_3^{(0)}$ (for the case of strong fields $E > E_2$) at various of the parameter $\omega\tau_s$: 1 – $\omega\tau_s$ = 10, 2—7, 3—5, 4—1, 5—0.5. The dashed lines show the asymptotic values of the weak field ($E_2 < E \ll E_3$) and very strong field ($E \gg E_3$—the Keldysh-Franz effect).

$$K_a \sim \exp\left\{-\frac{\Delta}{\hbar\omega}\tilde{f}_2\left(x, \frac{E}{E_3^{(0)}}\right)\right\}, \tag{29}$$

$$\tilde{f}_2(x, y) = x^{-2}\min_{x_h}\{x_h^2 f_2(x_h) + 1{,}17y^{-1}(x^2 - x_h^2)^{3/2}\} \quad (0 \leqslant x_h \leqslant x).$$

The function $\tilde{f}_2$ was calculated with a computer and is shown in Fig. 5. It takes the simplest form in the quasistatic case, when $x_h = x(1 + y^2)^{-1/2} \ll 1$. Then

$$K_a \sim \exp\left(-\frac{4\sqrt{2}}{3}\frac{\Delta^{3/2} m_e^{1/2}}{e\hbar(E^2 + E_3^{(0)2})^{1/2}}\right). \tag{30}$$

At first glance Eq. (30) can be interpreted (in the sense of Dow and Redfield[9]) as the result of electron tunneling in a homogeneous field that has besides a dc component also a random component with a characteristic value $E_3^{(0)}$ determined by the temperature. We note, however, that with this interpretation it would be utterly impossible to explain the dependence of the field $E_3^{(0)}$ on the hole mass which, as mentioned in the Introduction, should drop out of the answer.

In fact, the physical picture of absorption in the case described (Fig. 4) differs from the Keldysh-Franz "pure tunnel" effect (Fig. 1), and the quantity $E_3^{(0)}$ is due entirely to interaction of the hole with the phonons.

The dynamic case ($x_h \gg 1$) is realized in practice only at $E < E_3$, when Eq. (28) is valid.

4. CONCLUSION

We formulate now our main results.

As was shown earlier,[7,8] in the absence of a field the light is absorbed with formation of an optimal self-consistent fluctuation of the displacements of the crystal lattice. The fluctuation can be regarded as stationary or nonstationary, depending on the ratio of the parameters. The character of the influence of the electric field on the optimal fluctuation and on the absorption depends on the relation between the field E and the characteristic fields $E_1, E_2, E_3 (E_1 \ll E_2 \ll E_3)$ so that:

1. In the region $E \ll E_2$ the influence of the field reduces to orienting the dipole moment of the fluctuation by the electric field.

2. At $E \ll E_1 \ll E_2$ this orientation leads to a small absorption increment quadratic in the field and described by Eq. (24). The temperature and field dependences of the correction agree qualitatively with the experimental data.[11]

3. At $E \lesssim E_2$ the fluctuation is stretched out by the electric field (its dipole moment is increased), and at $E = E_2$ it is completely broken (it "dissociates"). In addition, in the dynamic case ($\omega\tau_s \gg 1$) the region $E \lesssim E_2$ is characterized by the combined character of the electron absorption: only part of the electron energy deficit is covered by phonon absorption, and the electron tunnels with the remaining deficit.

4. At $E > E_2$ a tunnel state sets in: the hole is localized in the fluctuation well, and the light electron tunnels. If at the same time $E < E_3$, the principal role in the absorption is played by the hole, while at $E > E_3$ the electronic process predominates. In the region $E \gg E_3$ the absorption is described by the Keldysh-Franz formula.

The authors thank A. G. Aronov for a discussion of the work and D. E. Khmel'nitskiĭ for a valuable remark.

APPENDIX

As shown in Ref. 7, the absorption coefficient is expressed in terms of the below-barrier action of the electron-hole pair:

$$K_a \sim \exp(-\Delta\tau - \tilde{S}(\tau)), \tag{A1}$$

where the below-barrier time τ is determined from the condition

$$\Delta = -d\tilde{S}/d\tau.$$

The action $\tilde{S}$ is resolved into electron and hole components: $\tilde{S} = \tilde{S}_e + \tilde{S}_h$, where at $\gamma \ll 1$ it turns out that $S_e \ll S_h$ and $\tilde{S}_h$ is hardly changed by the field.

To calculate the below-barrier action of the electron in the "combined" absorption mechanism, we break up the total time of the absorption process τ into two parts: the time τ^* during which the electron is in the well, and the time $t = \tau - \tau^*$ during which the electron tunnels. The action $\tilde{S}_e$ is accordingly also divided into two terms:

$$\tilde{S}_e = \tilde{S}_{e1} + \tilde{S}_{e2}. \tag{A2}$$

The value of $\tilde{S}_{e1}$ at $\omega\tau^* \gg 1$ was calculated in Ref. 8:

$$\tilde{S}_{e1} = -{}^1/_2 \alpha_e^2 N e^{2\omega\tau} (0.048 - {}^1/_2 e^{-\omega\tau^*}). \tag{A3}$$

The below-barrier action for tunneling in an electric field is well known (see Ref. 14):

$$\tilde{S}_{e2} = -e^2 E^2 t^3 / 24 m_e \hbar. \tag{A4}$$

We obtain τ^* from the condition that $\tilde{S}_e$ be a minimum:

$$\partial\tilde{S}_{e1}/\partial\tau^* = \partial\tilde{S}_{e2}/\partial t,$$

whence, using the definition of the field $E_2^{(\infty)}$ (19) and the fact that

$$\omega\tau = \ln\left[\frac{4.56}{\alpha_h N}\left(\frac{\Delta}{\hbar\omega}\right)^{1/2}\right],$$

we obtain

$$\omega\tau^* = \ln\left[5.3\left(\frac{\tau E_2^{(\infty)}}{tE}\right)^2\right], \quad t = \tau - \tau^*. \tag{A5}$$

It is seen that the condition $\omega\tau^* \gg 1$ is satisfied at $E \ll E_2^{(\infty)}$. Substituting (A5) in (A3) and (A4) in (A2) we have at $\omega t \gg 1$

$$\tilde{S}_e \approx \tilde{S}_e^{(0)} - \frac{e^2 E^2 t^3}{24 m_e \hbar} = \tilde{S}_e^{(0)} - 0.018\left(\frac{E}{E_1^{(\infty)}}\right)^2 \frac{(\omega t)^3}{(\omega\tau)^2},$$

which leads to the final expression (20).

[1)]We have disregarded here (just as in Eq. (1)) the Coulomb attraction between the electron and the hole, which is known (see Ref. 3) to change the electron-tunneling probability. It can be taken into account easily when necessary merely by allowing for the fact that the interaction of the electron with the fluctuation well of the hole is screened by the attraction to the hole itself. Accordingly, the resultant correcting Coulomb logarithm in exponential can become negative.[8]

[1]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **34**, 1138 (1958) [Sov. Phys. JETP **7**, 788 (1958)].

[2]W. Franz, Z. Naturforsch. **13a**, 1138 (1958).

[3]A. G. Aronov and A. G. Ioselevich, Exciton Electro-optics, in: Excitons, E. I. Rashba and M. D. Sturge, Ed., North-Holland, 1982, Chap. 7, p. 267.

[4]F. Urbach, Phys. Rev. **92**, 1324 (1953).

[5]R. S. Knox, Theory of Excitons, Academic, 1963.

[6]T. Tomiki, T. Myjata, and H. Tsukamoto, Z. Naturforsch. **29a**, 145 (1974).

[7]A. S. Ioselevich, Zh. Eksp. Teor. Fiz. **81**, 1508 (1981) Sov. Phys. JETP **54**, 800 (1981) .

[8]A. S. Ioselevich, *ibid.* **82**, 601 (1982) [**55**, 358 (1982)].

[9]J. D. Dow and D. Redfield, Phys. Rev. **B5**, 594 (1972).

[10]D. Appel, transl. in: Polyarony (Polarons), Yu. A. Firsov, ed., Nauka, 1975, p. 23.

[11]E. Mohler and B. Thomas, Phys. Rev. Lett. **44**, 543 (1980).

[12]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics, Nonrelativistic Part), Fizmatgiz, 1963, p. 324 [Pergamon, 1968].

[13]A. Sumi and Y. Toyozawa, J. Phys. Soc. Jpn. **31**, 342 (1971).

[14]A. I. Baz' Ya. B. Zel'dovich, and A. M. Perelomov, Scattering Reactions, and Decay in Nonrelativistic Quantum Mechanics, Nauka, 1971, p. 231 [Wiley, 1969].

Translated by J. G. Adashko