Theory of rate of nonradiative trapping

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A theory of the rate w of nonradiative trapping of carriers and excitons in dielectrics and semiconductors is developed. The theory covers the cases of intrinsic self-trapping and of multiphonon trapping by various defects (or extrinsic self-trapping and trapping by "normal" defects). Capture of both thermalized and hot particles is considered. The self-trapping barrier is assumed high enough for the trapping probability to be exponentially small. An exponential rate for thermalized particles was investigated by the authors earlier [JETP Lett. **40**, 1151 (1984); Sov. Phys. JETP **61**, 1110 (1985)]. The coefficient of the exponential is a product of several factors. The first, due to averaging over the Maxwellian distribution of the trapped particles, determines the dependence of w on T at low temperatures. The second is determined by the effective volumes in which the particles are trapped from the band state to a discrete level. The third is due to the presence of a zero temporal mode; in the continuum approximation it is due also to the existence of spatial zero modes. The last two factors cause the coefficient multiplying the exponential to substantially exceed the chracteristic lattice frequency. In the low-temperature region, the coefficient decreases with increasing T. The initial energy E of the hot particles can affect the probability of surmounting the barrier (this probability increases in the case of self-trapping), but only if this energy is coherently transferred to the lattice. The probability of this transfer at high E is exponentially small, so that particles with $E \lesssim W$ are predominantly trapped. The optimal self-trapping energy decreases with increasing T; the optimum for trapping by normal defects is always reached at $E = 0$. Specific differences of the temperature dependence of w for self-trapping (intrinsic and extrinsic) and for trapping by normal defects are also discussed.

1. INTRODUCTION

This article is closely related to our preceding papers[1,2] on the theory of electron and hole self-trapping rate in homopolar crystals, as well as self-trapping of excitons in crystals of all types. The formalism developed in Refs. 1 and 2 pertained to a very general model of electron-lattice interaction. The only serious constraint was in fact the assumption that the electron-phonon coupling is linear in the phonon amplitudes. The process that determines the rate of self-trapping was taken to be the surmounting of the self-trapping barrier beween the free and self-trapped states. Such a barrier always exists in three-dimensional systems if the particle (electron, exciton) is coupled to the phonon field by a short-range interaction.[3] The penetration of the barrier (by tunneling or activation) was considered in Ref. 2 in an exponential approximation. A common formalism was used for three specific models corresponding to three types of electron-phonon coupling.

In the present paper we generalize the model further, waiving the requirement that the electron-phonon interaction be linear, and analyze the coefficient of the exponential: we estimate its value and find its temperature dependence.

Our aim is best illustrated by using the simplest diagram that describes self-trapping. It is shown in Fig. 1 and corresponds to the presence of one lattice degree of freedom (one configuration coordinate Q). Generalization to the case of many coordinates is trivial in the case of an isolated trapping center: the point $\mathscr{W}$ is changed from a maximum into a saddle, and br is changed from a point into a surface. In the presence of translational symmetry there are many equivalent points—as many as there are unit cells in the crystal.

Linear electron-phonon coupling is a widely accepted approximation. It is not valid, however, at the core of a self-trapped state, where the displacements have the dimension of the lattice constant a_0.[3] The extent to which it is valid in the region of the barrier (Fig. 1), where the principal events connected with electron capture into a self-trapped state, is not clear at present. From general considerations one might assume that W is of the order of atomic, ~ 1 eV ($W \sim E_{\text{at}} \sim E_b$), and the spatial scale of the barrier is $r_b \sim a_0$. Many experimental results seem to indicate, however, that W for excitons is noticeably smaller in a number of crystals. Typical values are $W \lesssim 100$ meV (in alkali-halide crystals,[4,5] solidified noble gases,[6] and others). At $W \sim 1$ eV the exciton lifetime would simply be too short for self-trapping to occur. The only presently known argument[3] capable of explaining the low value of W is based on the assumption that the coupling constant in the investigated crystals is $\Lambda \simeq E_{\text{FC}}/E_B \gg 1$ (see Fig. 1 for the notation). Under these conditions $W \sim E_B/\Lambda^2 \ll E_B$ is indeed small, and the characteristic dimension of the barrier is large, $r_b \sim \Lambda a_0 \gg a_0$. At $r_b \gg a_0$ the displacements are small and the linear-coupling approximation suffices. This is why a continuum approximation and a linear coupling were used in the preceding papers.[1,2,7] There

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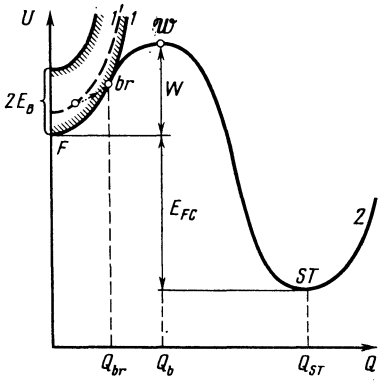

FIG. 1. Schematic dependence of the adiabatic potential U on the configuration coordinate Q for a crystal with an electron. 1—potential energy of crystal with free electron at the bottom of the electron band; 1'—the same curve, but shifted by an amount equal to the itinerant-electron energy E; 2—adiabatic potential for a crystal with an electron on a local level; W—height of self-trapping barrier; E_{FC}—Franck-Condon lattice-deformation energy; $2E_B$—width of electron band. The labeled values of the coordinate Q have the following meaning: Q_{ST}—in the self-trapping state, Q_b—in the barrier state, Q_{br}—at the branch point corresponding to formation of a local level for an exciton. The free state F corresponds to $Q=0$.

is, however, no direct experimental evidence that $r_b \gg a_0$, and a typical value is $\Lambda \sim 3$ (Ref. 6), i.e., not very large. One cannot exclude the possibility that $r_b \sim a_0$ actually happens by chance. It must also be taken into account that the minimum dimension on the tunnel trajectory can be less than r_b. Indeed, it was shown in Ref. 7 that such a small quantity does exist and can be either numerical or parametric. It is therefore likely that W can be small owing to the relatively large values of Λ, but tunnel trajectories can nonetheless pass through configurations of size $\sim a_0$, for which the nonlinearity is appreciable. A generalization of the model is therefore urgently needed. Of course, the results become consequently less specific. On the other hand, it will be possible to make certain assertions of general (model-independent) character. Some equations for specific models can be borrowed from Refs. 1 and 2.

The determination of the coefficients of the exponentials in the equations for the self-trapping rate is important for comparison of theory with experiment, especially for interpretation of the temperature dependences. The point is that for $T \ll \omega$, where ω is the characteristic frequency of the phonons, the temperature dependence of the argument of the exponential is very weak,[1,2] and the T dependence of the coefficient is therefore decisive. At high temperatures, $T \gtrsim \omega$, the exponential itself is equal (or close, see Sec. 5) to $\exp(-W/T)$, and if W is small ($W \sim 3\omega$), the contribution of the coefficient to the temperature dependence can be quite appreciable. It must be taken into account if W is to be correctly determined from the experimental data.

We shall not determine the numerical coefficients in the coefficients, and we confine ourselves to finding how they depend on the principal parameters, especially on temperature. It is practically impossible to determine the numerical coefficients in self-trapping problems. In fact, the self-energy part Σ for an itinerant electron is of the order of the Debye frequency ω_D (if all the estimates are made from first principles), and the main contribution is made by large momenta $q \sim a_0^{-1}$, when calculations are impractical. An uncertainty $\sim \omega_D$ in the value of Σ leads to a comparable uncertainty in W, and hence to an uncertainty in the numerical coefficient, since the tunneling probability at $T=0$ is proportional to $\exp(-bW/\omega)$, where ω is the characteristic value of the vibrational frequency and $b \sim 1$ (Ref. 7). An uncertainty of the same order is introduced also by other factors. A possible exception is the case of models of trapping by impurity centers, in which it is assumed that there is no interaction with an ideal lattice, and coupling exclusively to a local mode is considered.[8-10]

We return now to Fig. 1, which is an adiabatic diagram whose individual segments can be related to definite stages of the self-trapping process. This will allow us to trace the reasoning in the article and to discern the similarities and differences in related studies.

Since $W \gg \omega$, the lattice motion is quasiclassical and the adiabatic approximation is valid over the greater part of its path. In contrast, however, to the theory of multiphonon nonradiative transitions in impurity centers, which dates back to the paper of Huang and Rhys[11] (see the review by Kovarskiĭ *et al.*[12]), the dependence of the electron wave function on Q is significant in the entire region of the barrier and must be consistently taken into account. The configuration-curve scheme in Fig. 1 does not take account of translational invariance, and hence of the fact that intrinsic self-trapping can take place in any cell of the crystal, so that an infinite number of degrees of freedom is involved in the process. A consistent description must therefore be based on field-theoretical methods.[13,14] The adiabatic approximation is not valid at the point *br*, where the bound-state energy vanishes and near which electrons are trapped from the continuum to an adiabatic level. This stage of the process has an analog in the theory of atomic collisions,[15] where it is described by the zero-radius-potential method.[16] It determines the dependence of the self-trapping rate on the electron energy E.

A related group of problems deals with tunneling under conditions of interaction with a heat bath (e.g., phonons); work along these lines was initiated in Ref. 17. Our problem is different because the barrier is formed by the very same phonon interaction that determines the scattering. Under these conditions, the time of tunneling with a trapped electron is not a free parameter, but is always of the order of ω^{-1}. This time is too short for dissipative forces to manifest themselves, since interaction of a "dressed" electron with phonons is strongly suppressed.

Our problem being quasiclassical, the self-trapping rate is determined mainly by an "imaginary" action $S(\beta)$, where $\beta = T^{-1}$. This quantity is real and is equal to that imaginary part of the total classical action which is due both to below-barrier motion and to Gibbs averaging. The variety of situations notwithstanding, the possible $S(\beta)$ dependences are apparently of two principal types, illustrated in Figs. 2a and 2b. Each includes a curve A corresponding to the classical activational (Arrhenius) surmounting of the barrier, and a

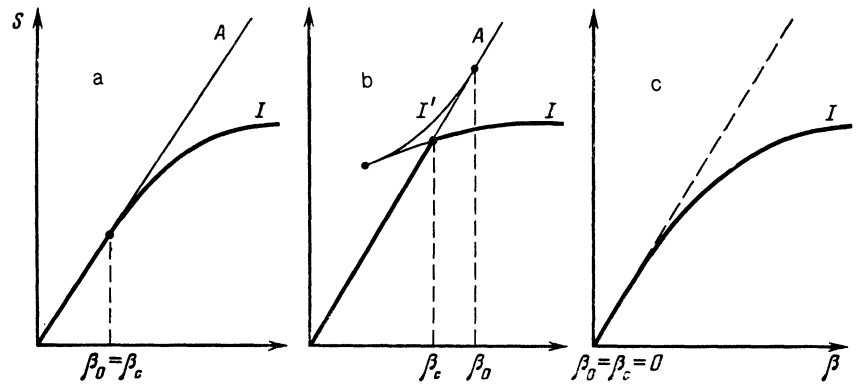

FIG. 2. Main types of $S(\beta)$ dependences: A—activation (Arrhenius) regime; I, I'—instanton; $\beta_c = T_c^{-1}$—regime switch over point. Cases a and b pertain to self-trapping (both intrinsic and extrinsic), case c—to trapping by a "normal" recombination center.

curve I that describes thermally activated tunneling (instanton). These curves are tangent at the point $\beta_0 = 2\pi/\omega_{W_0}$. The frequency ω_{W_0} has a simple meaning. In the multidimensional case the point $\mathscr{W}$ is the lowest saddle point of the adiabatic-potential surface. This surface is represented in the vicinity of $\mathscr{W}$ by a quadratic form[1)] that has a single negative eigenvalue corresponding in imaginary time to the frequency ω_{W_0}. The tangency of the curves at the point β_0 was analyzed in detail by Affleck,[19] while the scheme in Figs. 2a and 2b was discussed by Meshkov.[10] For the one-dimensional model it is simply interpreted in terms of the energy dependence of the period of the quasiclassical oscillations. A similar picture appears in the theory of quantum nucleation.[20] We found the intersection of curves A and I (Fig. 2b) in Refs. 1 and 2 for specific models. The form of segment I' of the curve, however, interpreted there in terms of the "long instanton," was incorrectly obtained (this was noted by Meshkov[10]). Incidentally, the form of segment I' is unimportant, since the action on I' is larger than on I and A. It appears that cases 2a and 2b are experimentally distinguishable: the switch from the Arrhenius to the instanton temperature dependence is more abrupt in the second.

In the next two sections we obtain coefficients of the exponentials in the low- and high-temperature regions. In Sec. 4 we consider self-trapping of hot electrons, while in Sec. 5 the theory is transferrred to multiphonon trapping of carriers by impurity centers (recombination and extrinsic self-trapping; the latter term was introduced in Ref. 21). Some of the results that follow were published, in part without proof, in Refs. 22 and 23.

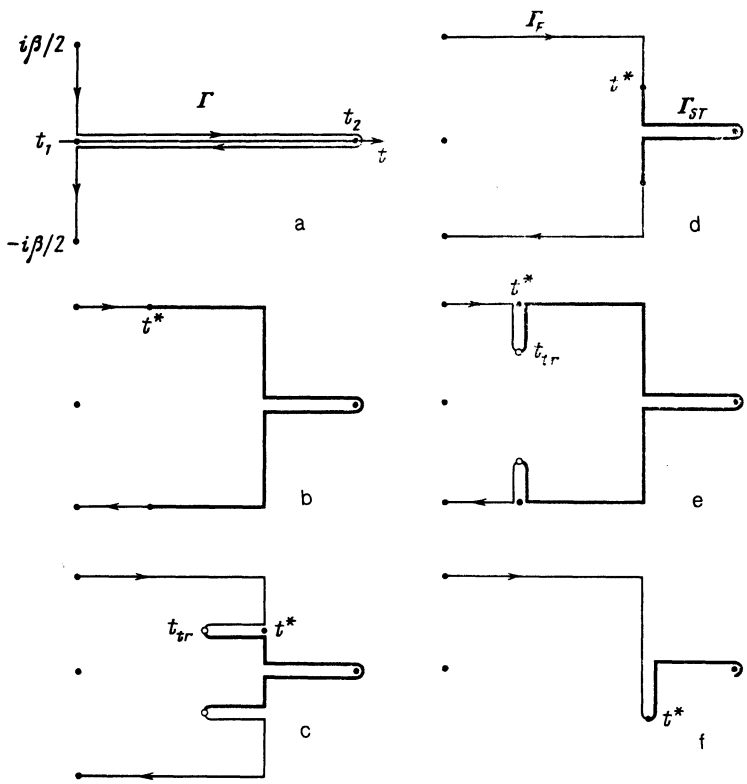

FIG. 3. Integration contours Γ: a) initial contour; b) contour for low-temperature region. Γ_F—free motion of lattice (thin line), t^*—instant when a discrete level is produced. c) Contour for high-temperature region. The motion becomes loaded in the classically accessible region long before the barrier is surmounted. d) Contour for self-trapping of hot electrons, high-temperature region. 3) The same for the low-temperature region. f) Contour for tunneling in a recombination center (the upper half of the contour is shown).

2. LOW-TEMPERATURE REGION

The low-temperature region is bounded by the inequality $T<T_c=\beta_c^{-1}$. The self-trapping in this region is by the instanton mechanism.

Since we seek below, in contrast to Refs. 1 and 2, instanton solutions in explicit form, whereas the coefficients are most naturally obtained by integrating with respect to the phonon coordinates rather than the electron wave functions, we modify somewhat the formalism used in Refs. 1 and 2. We do not exclude the phonon coordinates $Q(t)$ from the path integrals (I.9) and I.10)[2)] but, on the contrary, exclude formally the electron functions Ψ for thermalized elecrons, averaging, with allowance for (I.8)–(I.11), Eq. (I.6) over the Boltzmann distribution of the electrons, we obtain

$$(t_2-t_1)\,w_T=Z^{-1}\int\frac{d\mathbf{k}}{(2\pi)^3}\int d\mathbf{r}_1\,d\mathbf{r}_2\,d\mathbf{r}_1'\,d\mathbf{r}_2'\,\psi_{ST}^*(\mathbf{r}_2)\psi_{ST}(\mathbf{r}_2')$$
$$\times\int\mathcal{D}Q\,G^R_{\{Q\}}(\mathbf{r}_2t_2,\mathbf{r}_1t_1)\,G^A_{\{Q\}}(\mathbf{r}_1't_1,\mathbf{r}_2't_2)\,\psi_{\mathbf{k}}(\mathbf{r}_1)\psi_{\mathbf{k}}^*(\mathbf{r}_1')$$
$$\times\exp\left\{i\int_\Gamma L_{lat}(Q)\,dt-\beta E(\mathbf{k})\right\},\quad(1)$$

where $Z=Z_{\text{lat}}Z_e$ is the normalization factor:

$$Z_{lat}=\int\mathcal{D}Q\exp\left\{i\int_\Gamma L_{lat}(Q)\,dt\right\},$$
$$Z_e=\int\frac{d\mathbf{k}}{(2\pi)^3}\exp\{-\beta E(\mathbf{k})\}.\quad(2)$$

If the measure is properly chosen, Z_{lat} is equal to the lattice partition function. Here $G^R_{\{Q\}}$ and $G^A_{\{Q\}}$ are the retarded and advanced Green's functions of the electron and depend functionally on the lattice trajectory $Q(t)$, L_{lat} is the free-lattice Lagangian, and the contour Γ is shown in Fig. 3a. The usual periodic conditions $Q(t_1+i\beta/2)=Q(t_1-i\beta/2)$ are imposed on Q and will hereafter be called the conditions of periodicity in imaginary lines, $\mathbf{k}$ is the momentum of the free electron, $E(\mathbf{k})$ is its energy, and ψ_{ST} and $\psi_{\mathbf{k}}$ are the Schrödinger wave function of the self-trapped and itinerant electrons, respectively. Electron Green's functions that depend on the lattice trajectory were previously used in other problems.[24]

Self-trapping sets in at some instant of time between t_1 and t_2. It is convenient therefore to express $G^A_{\{Q\}}$ and $G^A_{\{Q\}}$ in a mixed representation:

$$G^R_{\{Q\}}(\mathbf{r}_2t_2,\mathbf{r}_1t_1)$$
$$=V^{-1/2}\sum_m\int\frac{d\mathbf{k}}{(2\pi)^3}\,\psi_{m,Q(t_2)}(\mathbf{r}_2)\,u_{m,\mathbf{k}}(\{Q\})\,\psi_{\mathbf{k}}^*(\mathbf{r}_1)$$
$$\times\exp\left\{-i\int_{t_1}^{t_2}E_m(Q(t))\,dt\right\},\quad(3)$$

where V is the volume of the crystal. The functions $\psi_{m,Q(t)}(\mathbf{r})$ and $E_m(Q(t))$ comprise the complete set of Schrödinger wave functions and the energies corresponding to them in the time domain t corresponding to self-trapping (loaded motion of the lattice); ψ_{ST} corresponds to the lowest $E_m(Q)$. In the region of free lattice motion, we must substitute $E_m(Q)\Rightarrow E(\mathbf{k})$. The symbol $\{Q\}$ in the arguments of the various quantities means that they depend on the coordinates Q nonlocally with respect to time, or in other words, that they depend on the entire trajectory of the lattice. The factor $u_{m,\mathbf{k}}$ is the amplitude of the transition from the state $\mathbf{k}$ to the state m. Substituting (3) in (1) we obtain

$$(t_2-t_1)\,w_T=(VZ)^{-1}\int\frac{d\mathbf{k}}{(2\pi)^3}\int\mathcal{D}Q\,|u_{ST,\mathbf{k}}(\{Q\})|^2$$
$$\times\exp\left\{i\int_\Gamma L_{\mathbf{k}}(Q)\,dt\right\},\quad(4)$$

where

$$L_{\mathbf{k}}(Q)=\begin{cases}L_{lat}(Q)-E_{ST}(Q) & \text{on }\Gamma_{ST}\\ L_{lat}(Q)-E(\mathbf{k}) & \text{on }\Gamma_F\end{cases}.\quad(5)$$

The correspondingly deformed contour is shown in Fig. 3b (cf. Ref. 2). The electron is trapped at the point t^* located on the vertical section of the contour Γ. The entire vertical section corresponds to lattice tunneling within a time $\beta/2$. The duration $\text{Im}\{t^*\}=\tau_0\sim\omega^{-1}$ of the loaded motion for $T\ll\omega$ is considerably shorter than $\beta/2$ and is independent of T in this region.

We calculate the path integral by the saddle-point method, using the presence of the large paraemeter $W/\omega\gg1$. We can then take $u_{\text{ST},\mathbf{k}}$ of the coefficient outside the integral on the extremal trajectory; we denote this quantity by $u(\mathbf{k})$. For slow electrons with $E(\mathbf{k})\sim T$ we can neglect the dependence of $u(\mathbf{k})$ on $\mathbf{k}$ and write simply $u(0)$ (see the Appendix). The dependence on $E(\mathbf{k})$ is completely separated in the form of a factor $\exp\{-(\beta-2\tau_0)E(\mathbf{k})\}$. Thermal electrons obey the quadratic dispersion law $E(\mathbf{k})=k^2/2m$. The integral with respect to $\mathbf{k}$ can therefore be calculated, and its ratio to Z_e is equal to

$$p_e(T)=\langle\exp(2\tau_0E(\mathbf{k}))\rangle_T=(1-2\tau_0T)^{-3/2}.\quad(6)$$

We express the Lagrangian $L(Q)$, with $E(\mathbf{k})$ left out, in the form

$$L(Q)=\frac{1}{2}\sum_j(\partial_tQ_j)^2-U(Q),\quad(7)$$

where the adiabatic potential $U(Q)$ is equal to the lattice potential energy $U_m(Q)$ of the lattice on the free-motion segment (curve 1, Fig. 1), while on the loaded-motion segment we have $U(Q)=U_0(Q)+E_{\text{ST}}(Q)$ (curve 2, Fig. 1). The subscript j numbers the degrees of freedom of the lattice. The potential $U(Q)$ cn be expanded near the extremal trajectory $Q_I(T)$ in powers of $q=Q-Q_I$. As a result, the contribution w_{cell} of this extremal to w_T is equal to

$$(t_2-t_1)\,w_{cell}$$
$$=p_e|u(0)|^2e^{-S_I}(VZ_{lat})^{-1}\int\mathcal{D}q\exp\left\{i\int_\Gamma l(q,t)\,dt\right\},\quad(8)$$

where

$$l(q,t)=\frac{1}{2}\sum_j(\partial_tq_j)^2-\frac{1}{2}\sum_{jj'}q_jq_{j'}\delta^2U(Q_I(t))/\delta Q_j\,\delta Q_{j'}.\quad(9)$$

Here $S_I\sim W/\omega$ is the instanton "imaginary" action at

$E(\mathbf{k})=0$. Its temperature dependence was considered in Refs. 1 and 2. The eigenvectors of form $l(q,t)$, which are periodic in imaginary time, will be designated by $q^{(s)}(t)$, and the corresponding eigenvalues by $(-\lambda_s)$. The λ_s depend on T via the periodicity conditions. The eigenvalues of the operator l for the free lattice [i.e., with $Q_I(t)\equiv 0$] will be designated $(-\lambda_s^{(0)})$. The meaning of the index s for the free lattice is easy to interpret. It numbers both the normal coordinates of the lattice and all the natural oscillations (periodic in imaginary time) corresponding to each of these coordinates. There is no such simple classification for loaded motion, but the total number of eigenvalues remains unchanged. It can be shown in the usual fashion that one of the λ_s is negative ($\lambda_0<0$) and one is zero ($\lambda_1=0$). the corresponding zero mode $q^{(1)}\propto \partial_t Q_I(t)$. This zero mode is time dependent. It corresponds to translation in time and its contribution to the integral in (8) is $(t_2-t_1)S_I^{1/2}$. Gaussian integration in (8) and cancellation of (t_2-t_1) yields

$$w_{cell}\sim p_e V^{-1}|u(0)|^2 S_I^{1/2} e^{-S_I}\omega_{eff},$$

$$\omega_{eff}=\left|\prod_s{}'\lambda_s^{(0)}\Big/\prod_s{}'\lambda_s\right|^{1/2}. \tag{10}$$

A prime on the product symbol means that the zero mode is left out. If no soft modes other than long-wave acoustic phonons are included among the λ_s then, since the characteristic values satisfy $\lambda_s\sim\lambda_s^{(0)}\sim\omega^2$, the estimate $\omega_{\text{eff}}\sim\omega$ is valid, with ω_{eff} independent of T as $T\to 0$. Since all the lattice cells are equivalent, the number of extremals with identical action is V/v, where v is the unit-cell volume. Therefore $w_T=(V/v)w_{\text{cell}}$. Using Eqs. (A1) and (A2) for $|u(0)|^2$, we obtain ultimately

$$w_T\sim p_e v^{-1}\omega(m\omega)^{-1/2}\exp(-S_I)\sim p_e\omega(E_B/\omega)^{3/2}\exp(-S_I), \tag{11}$$

where we used the estimate $E_B\sim m^{-1}v^{-2/3}$.

Equation (11) is invalid when the instanton radius $r_I\sim a_0$. With increasing r_I there appear among the λ_s three anomalously small eigenvalues corresponding to three translational soft modes (with frequencies ω_{soft}). Then $\omega_{\text{eff}}\sim\omega(\omega/\omega_{\text{soft}})^3$ and w_T increases substantially. As $\omega_{\text{soft}}\to 0$, the expansion (9) in terms of the corresponding variables is no longer sufficient, and ω_{eff} no longer increases. In the continuum limit ($r_b\gg a_0$) the soft modes are transformed into three spatial zero modes. A standard procedure[14] leads to the equation $\omega_{\text{eff}}\sim\omega(v/r_I^3)S_I^{3/2}$, since the intergration over the instanton coordinate is limited to the unit cell. For interaction with nonpolar optical phonons we have $r_I\sim r_b$ (Ref. 7), and then

$$w_T\sim p_e(T)\omega S_I^3\exp(-S_I). \tag{12}$$

For acoustic phonons, the instanton has two scales in the continuum approximation.[2] The internal scale satisfies $r_I<r_b$, with $r_I\neq 0$ only because the lattice is discrete. In the strict continuum limit, when $r_I\to 0$ and the electron interacts exclusively with acoustic phonons, the system acquires as $T\to 0$ an additional symmetry with respect to the four-dimensional scale transformation of the instanton.[7] This symmetry should correspond to an additional zero mode. All this can change, and more readily increase, the coefficient in Eq. (12).

Large factors, $(E_B/\omega)^{3/2}\gg 1$ and $S_I^3\gg 1$, multiply ω in both equations (11) and (12). This increases w_T considerably with the usual elementary estimate. It is convenient to interpret first the physical meaning of these factors as applied to Eq. (11), which we rewrite in the form

$$w_T\sim p_e\omega S_I^{1/2}(r_{tr}^3/v)e^{-S_I},\quad r_{tr}^3=|u(0)|^2. \tag{13}$$

The factor $S^{1/2}\gg 1$ always appears in problems involving tunneling from an oscillator-type potential well. It stems from the fact that the tunneling is not from the bottom of the well but from a lower level with energy $\omega/2$. The factor $r_{\text{tr}}^3/v\gg 1$ contains the effective radius r_{tr} of the trapping region (see the Appendix). This factor reflects the possibility of the onset of instanton fluctuation in any of the cells within the trapping region. The additional factor $S_I^{1/2}\gg 1$ in Eq. (12) is due to the fact that the translation group becomes continuous. Equations (11) and (12) can be rewritten in the form

$$w_T=p_e r_{tr}^3 d_I,\quad d_I\sim\begin{cases}(\omega/v)S_I^{1/2}\exp(-S_I)\\(\omega/r_b^3)S_I^2\exp(-S_I)\end{cases}. \tag{14}$$

The quantity d_I is called the instanton density[25] and has the meaning of the probability of the onset of an instanton fluctuation per unit volume and per unit time. The upper and lower equation for d_I pertain to the discrete and continuous limits, respectively.

At $T\ll 1$ the temperature dependence of S_I is weak: the temperature-dependent contributions to S_I are proportional to T^4 and T^2 respectively for deformation and piezolectric interactions with acoustic phonons, and to $\exp(-\omega/T)$ for nonpolar interaction with optical phonons.[1,2] The temperature dependence of $p_e(T)$ [Eq. (6)] should therefore be dominant here. It appears that it is just the factor $p_e(T)$ which describes the temperature dependence of the rate of exciton self-trapping in Xe at $T\lesssim 30$ K (Ref. 26), as follows from the analysis of Ref. 23.

3. HIGH-TEMPERATURE REGION

At high temperatures $T>T_c$ the self-trapping is by the activation mechanism. In the region $T\gtrsim T_c$, nevertheless, an appreciable fraction of the flux passes by tunneling near the top of the barrier.

It is convenient to rewrite Eq. (4) in terms of the lattice Green's functions $D^{R(A)}$. They should in principle be labeled by two indices corresponding to the numbers of the sheets of the adiabatic-potential surface in the initial and final states. Since self-trapping corresponds only to off-diagonal elements (such as $D_{21}^{R(A)}$), we shall omit the subscripts. Passage through the point br introduces into D the amplitude of the passage between the sheets, which coincides with the electron-trapping amplitude $V^{-1/2}u(\mathbf{k})$. In this notation, (4) takes the form

$$(t_2-t_1)w_T=Z^{-1}\int\frac{d\mathbf{k}}{(2\pi)^3}\int dQ_1\,dQ_2\,D^A(Q_1,t_1-i\beta/2|Q_2,t_2)\times D^R(Q_2,t_2|Q_1,t_1+i\beta/2). \tag{15}$$

The integration over $\bar{d}Q_1$ extends here to the region from which flow behind the barrier is possible, while the integration of $\bar{d}Q_2$ extends only to a region behind the barrier from which return to the free state is already practically impossible. The integrands take in the energy representation the form

$$\int \frac{dE\,d\varepsilon}{(2\pi)^2} D^{A}_{E+\varepsilon/2}(Q_1,Q_2)\,D^{R}_{E-\varepsilon/2}(Q_2,Q_1)\exp\{-\beta E+i(t_2-t_1)\varepsilon\}. \tag{16}$$

Since the coefficient of the exponential in (16) oscillates rapidly on the scale $\varepsilon\sim(t_2-t_1)^{-1}$, the function $D^{R(A)}_{E\pm\varepsilon/2}$ can be expanded in powers of ε. It is necessary here to make use of the quasiclassical character of the motion (with the possible exception of the immediate vicinity of the point $\mathscr{W}$), as a result of which the fastest factor in D^R is $\exp\{iS_0(E\,|Q_2,Q_1)\}$, where S_0 is the reduced action. Since Q_1 and Q_2 are on opposite sides of the barrier, the reflected wave need not be taken into account. Expanding the action in powers of ε, recognizing that $\partial S_0(E\,|Q_2,Q_1)/\partial E$ is equal to the time $T_E(Q_2,Q_1)$ of the motion between Q_1 and Q_2, and then integrating over ε, we transform (16) into

$$\int \frac{dE}{2\pi}\,|D_E^R(Q_2,Q_1)|^2\exp(-\beta E)\,\delta(t_2-t_1-T_E(Q_2,Q_1)). \tag{17}$$

Our problem is to separate in the right-hand side of (15) the time dependence in the form of a factor t_2-t_1. To this end it is convenient to partition the entire multidimensional space by means of a hypersurface $\varepsilon_{\mathscr{W}}$ that passes through the point $\mathscr{W}$ near which the main flux is concentrated, and is normal to the coordinate q_0 corresponding to a negative eigenvalue ($-\omega w_0^2$) of the adiabatic potential in the vicinity of the point $\mathscr{W}$. The main flux of the particles is oriented along q_0. We denote by $Q_{\mathscr{W}}(Q_1,Q_2)$ the point where a classical trajectory drawn from Q_1 to Q_2 intersects the surface $\Sigma_{\mathscr{W}}$. We can then write $T_E(Q_2,Q_1)=T_E(Q_2,Q_{\mathscr{W}})+T_E(Q_{\mathscr{W}},Q_1)$ and respresent (15) with the aid of an elementary transformation in the form

$$(t_2-t_1)\,w_T=\frac{1}{Z}\int_{t_1}^{t_2}dt\int\frac{d\mathbf{k}}{(2\pi)^3}\frac{dE}{2\pi}\int\bar{d}Q_1\,\bar{d}Q_2\,|D_E^R(Q_2,Q_1)|^2$$
$$\times\exp(-\beta E)\,\delta(t_2-t-T_E(Q_2,Q_{\mathscr{W}}))\,\delta(t-t_1-T_E(Q_{\mathscr{W}},Q_1)). \tag{18}$$

To interpret the inner integral, it is convenient to consider the auxiliary expression

$$Z_{lat}^{-1}\frac{e^{-\beta E}}{2\pi}\bar{d}Q_1\,\delta(f_1(Q_1))\,(\mathbf{p}_1\nabla f_1)\,|D_E^R(Q_2,Q_1)|^2$$
$$\times(\mathbf{p}_2\nabla f_2)\,\delta(f_2(Q_2))\,\bar{d}Q_2, \tag{19}$$

where $f_1(Q_1)=0$ and $f_2(Q_2)=0$ are the equations of the two hypersurfaces Σ_1 and Σ_2, while $\mathbf{p}_1$ is the multidimensional momentum of the lattice at the point Q_1 for the trajectory from Q_1 to Q_2. If Σ_1 intersects the volume element dQ_1, then

$$d\Pi_1=Z_{lat}^{-1}\exp(-\beta E)\,\bar{d}Q_1\nu(E,Q_1)\,\delta(f_1(Q_1))\,(\mathbf{p}_1\nabla f_1)$$

is equal to the spectral density of the flux, near the energy E, which passes through an elementary surface area Σ_1. Here $\nu(E,Q_1)=\pi^{-1}\operatorname{Im}D_E^R(Q_1,Q_2)$ is the density of states. From the equation $(\hat{H}-E+i0)D_E^R(Q,Q_1)=-\delta(Q-Q_1)$, which defines the function D_E^R, it follows that the stationary density produced at the point Q by a δ-function source located in Q_1 is equal to $|D_E^R(Q,Q_1)|^2$. It follows at the same time from a time-dependent Schrödinger equation having the same right-hand side that the source produces a total flux $2\operatorname{Im}D_E^R(Q_1,Q_1)$.[27] In the quasiclassical situation the flux is concentrated near a classical trajectory, and the density dN_2 at the point Q_2, produced by a flux $d\Pi_2$ at the point Q_2, is equal to

$$dN_2=Z_{lat}^{-1}\exp(-\beta E)\,\pi^{-1}\operatorname{Im}D_E^R(Q_1,Q_1)\,\delta(f_1(Q_1))\,(\mathbf{p}_1\nabla f_1)$$
$$\times\frac{|D_E^R(Q_2,Q_1)|^2}{2\operatorname{Im}D_E^R(Q_1,Q_1)}\bar{d}Q_1$$
$$=Z_{lat}^{-1}\exp(-\beta E)\,(2\pi)^{-1}|D_E^R(Q_2,Q_1)|^2$$
$$\times\delta(f_1(Q_1))\,(\mathbf{p}_1\nabla f_1)\bar{d}Q_1. \tag{20}$$

It is now clear that (19) is the elementary flux from surface Σ_1 to surface Σ_2. It can be verified that for surfaces Σ_1 and Σ_2 defined by the δ functions in (18) we have $|(\mathbf{p}_i\nabla f_i)|=1$, $i=1,2$. Indeed, separating from the set Q_i the coordinate q_{0i} corresponding to motion along a classical trajectory tangent to $\mathbf{p}$, we get $p_i\partial f_i/\partial q_{0i}=(\mathbf{p}_i\nabla f_i)=\pm 1$. To obtain the last equation we used the well-known classical formula

$$\partial T_E(Q_i,Q_{\mathscr{W}})/\partial q_{0i}=\pm p_i^{-1}.$$

Thus, the integral over Q_1 and Q_2 in (18) is the spectral density of the flux between the surfaces Σ_1 and Σ_2.

By virtue of flux conservation, this integral remains unchanged in the quasiclassical region if Σ_1 and Σ_1 are displaced along the flux, and is therefore independent of t. Consequently, the integral with respect to t in (18) reduces to multiplication by t_2-t_1, and the surfaces Σ_1 and Σ_2 can be brought close to $\Sigma_{\mathscr{W}}$ to permit only quadratic expansion of $U(Q)$ near $\mathscr{W}$. Next it is convenient to transform with respect to all coordinates except q_0 to the quantum-number representation $Q\to q_0,\{n_i\},i\geqslant 1$. We can similarly write for the energy

$$E=W+E_0+\sum_{i\geqslant 1}E_i,$$

where E_0 is the energy corresponding to the degree of freedom q_0 and measured from the top of the barrier. The Green's function for the degree of freedom q_0 is of the form

$$D_{E_0}^R(q_{02},q_{01})=(p(q_{01},E_0)\,p(q_{02},E_0))^{-1/2}$$
$$\times\exp\{iS_0(E_0|q_{02},q_{01})\}V^{-1/2}u(\mathbf{k})\,d(E_0). \tag{21}$$

The first two factors are the standard form for the quasiclassical Green's function, the factor $V^{-1/2}u(\mathbf{k})$ is the contribution from the trapping stage, and $|d(E_0)|^2$ is the transparency of the barrier. This last factor takes into account the possible deviation from quasiclassical behavior near $\mathscr{W}$. Taking (21) and (A2) into account, we can integrate in (18) with respect to $\mathbf{k}$. It is unusual that the integral with

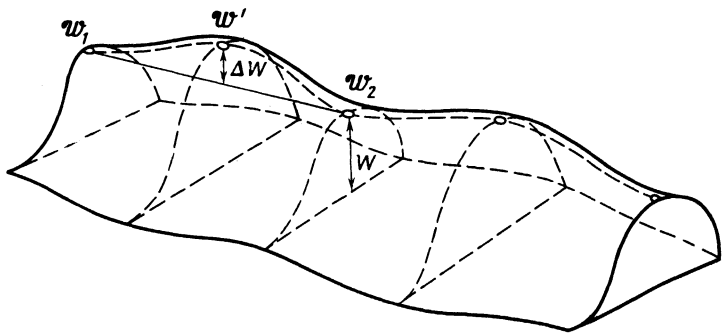

FIG. 4. Schematic form of adiabatic-potential surface: $\mathscr{W}_1$ and $\mathscr{W}_2$—lowest saddles corresponding to a self-trapping barrier for two neighboring unit cells; $\mathscr{W}'$—taller saddle that acts as a col between $\mathscr{W}_1$ and $\mathscr{W}_2$.

respect to $\mathbf{k}$ converges not to $E(\mathbf{k})\sim T$, but to $E(\mathbf{k})\sim\Omega\sim(\omega^2 W)^{1/3}$, which is the characteristic capture energy. As a result, the dimensionless phase volume of self-trapping particles is of order $(\Omega/T)^{3/2}$. In the integration with respect to $\mathbf{k}$ the factor $\exp(-E(k)/T)$ is not manifested in the numerator of (18), since $E(\mathbf{k})$ is a fraction of the total energy E contained in the Gibbs factor in (18); the integration with respect to E is carried out below. The integration with respect to Q_1 and Q_2 in (18) reduces to calculation of the partition function with respect to n_i and integration with respect to q_{0i} with $i=1$ and 2. The last two integrations eliminate the δ functions contained in the arguments of $T_{E_0}(Q_i, Q_{\mathscr{W}})$, and cancel out simultaneously the momenta that enter in (21). As a result, we have for the contribution ω_{cell} in w_T from the self-trapping of an electron in a defnite unit cell:

$$w_{cell}=\frac{2}{V}\left(\frac{2\pi}{mT}\right)^{3/2}e^{-W/T}\frac{\prod_i \operatorname{sh}(\omega_i/2T)}{\prod_{i\neq 0}\operatorname{sh}(\omega_{Wi}/2T)}\times\int\frac{dE_0}{2\pi}|d(E_0)|^2 e^{-\beta E_0}. \tag{22}$$

Here ω_i are the free-lattice frequencies, and ω_{W_i} are the vibrational frequencies on the barrier. Using for $d(E_0)$ the familiar expression $|d(E_0)|^2=\{1+\exp(-2\pi E_0/\omega_{W0})\}^{-1}$, corresponding to a parabolic barrier, and multiplying by the degeneracy factor V/v (see Sec. 2), we get

$$w_T=\left[v^{-1}\left(\frac{2\pi}{mT}\right)^{3/2}\right]\frac{\omega_{eff}}{2\pi}\exp\left(-\frac{W}{T}\right),$$
$$\omega_{eff}=\{\omega_{W0}/\sin(\omega_{W0}/2T)\}\left\{\prod_i \operatorname{sh}(\omega_i/2T)\Big/\prod_{i\neq 0}\operatorname{sh}(\omega_{Wi}/2T)\right\}. \tag{23}$$

This expression for w_T differs from its analog for activation surmounting of a barrier by nonlinear multimode system[19] in that the latter contains in square brackets a factor responsible for trapping under activation conditions. The meaning of this factor becomes clear if it is rewritten in the form $(r_{\text{tr}}^3/v)(\Omega/T)^{3/2}$. Its order of magnitude is $(E_B/T)^{3/2}$. If $T\gtrsim T_c$, we have $\omega_{\text{eff}}\sim\omega$ and

$$w_T\sim v^{-1}\omega(mT)^{-3/2}\exp(-W/T)\sim(E_B/T)^{3/2}\omega\exp(-W/T). \tag{24}$$

The coefficient of the exponential is thus large and depends on temperature.

Although (23) is written as an exact equality, in view of the renormalization of the free-electron spectrum it is in fact accurate only to within a factor of order unity (as explained in Sec. 1).

The estimate (24) is valid for a small radius of the barrier state, when the individual saddles are well separated. As r_b increases the potential relief (Fig. 4) is smoothed out and the height ΔW of the barriers between neighboring saddles decreases rapidly at an exponential rate. Three anomalously low frequencies Δ_{soft} appear then among the frequencies $\omega_{W_i}(i\neq 0)$. The oscillations corresponding to them are generators of zero-point modes (cf. Sec. 2). If $\Delta W>T$, then $\omega_{\text{eff}}\sim\omega(\omega/\omega_{\text{soft}})^3$, and we get in place of (24)

$$w_T\sim(E_B/T)^{3/2}(\omega/\omega_{soft})^3\omega\exp(-W/T), \tag{25}$$

i.e., w_T increases appreciably. With further decrease of ΔW, when $\Delta W<T$, the situation approaches that of a continuum. ω_{eff} is then obtained from the corresponding equation of Sec. 2 by making the substitutions $r_I\to r_b$ and $S_I\to S_A\equiv W/T$. The result is

$$\omega_{eff}\sim\omega(v/r_b^3)S_A^{3/2},\quad w_T\sim(W/T)^3\omega\exp(-W/T). \tag{26}$$

The temperature dependence of the coefficient of the exponential becomes stronger than in (24).

The meaning of the derivation of Eqs. (25) and (26) requires the coefficient in (26) to be much larger than in (24). This is indeed so if the low values of W for excitons ($W\ll E_B$), reported for a number of crystals, are due only to the large $\Lambda\gg 1$. In the transition region $\Lambda\sim 1$ the barrier height is then $W\sim E_B$, and the inequality of the coefficients is well satisfied. If, however, for reasons unknown at present, the numerical values are such that $W\ll E_B$ also in the region $\Lambda\sim 1$, the use of W as the energy scale in the coefficient of the exponential in (26) is not justified at $\Lambda\sim 1$. It is then apparently more correct to use in this region the estimate $(E_B/T)^3$. This remark holds also for Eq. (12).

4. HOT ELECTRONS

The carriers and excitons are produced in most experiments with an initial energy significantly higher than thermal. The question is: how are they thermalized and how does the branching of the process take place, viz., what fraction of

the particles is thermalized into a free state, and what fraction is self-trapped prior to thermalization? It is also very important to identify the relaxation stage at which the main outflow of particles to the self-trapping state takes place. To answer these questions, we must calculate the self-trapping rate $w(E,T)$ as a function of the particle energy E.

We shall carry out all the calculations in the exponential approximation. Calculation of the coefficients is not urgent at present, since the methods of exciting fast carriers and excitons are difficult to control and the nonequilibrium distribution function of the particles is unknown.

The self-trapping barrier blocks not the electron but the lattice degrees of freedom. Nonetheless, the initial electron energy can in principle be used to permit the lattice to surmount the barrier. If, however, the fast electron loses energy gradually, emitting a cascade of incoherent phonons, this will in no way facilitate its self-trapping, which requires that the energy be transferred from the electron to the lattice coherently. Since, however, the lattice frequencies are low, the probability of such a transfer is exponentially small. We consider here, for the technique used above, a modification that is needed to be able to calculate this probability.

If any of the contours of Figs. 3a to 3c are used, the coordinates remain real at all times, and the transition from surface 1′ to the point *br* (Fig. 1) entails a jump of the electron energy. It is just this jump which determines the amount of energy that must be coherently transferred to the lattice, and the problem consists of calculating the probability of this transfer. In the spirit of the adiabatic theory of quantum transitions,[28] the contour must be deformed so as to prevent this jump. It follows from (5) that it is necessary that $E_{ST}(Q)$ which is negative for real Q, satisfy at a certain instant of time the condition $E_{ST}(Q)=E$, where $E>0$ is the free electron energy. This is possible only if Q is complex, i.e., the contour must be deformed.

We make one general remark. In the present section, in contrast to Secs. 2 and 3, no Gibbs averaging over the electron energy is carried out. In this calculation method, the results of Eqs. (4) and (5) must be multipled by the factor $\exp(\beta E)$.

Consider first the activation regime. A discrete level is produced at the instant t^* as the lattice moves in the direction of real time. At times t close to t^* the electron energy varies as

$$E_{ST}(t)=-\Omega^3(t-t^*)^2 \tag{27}$$

(see the Appendix). The equation $E_{ST}(t)=E$ is satisfied at

$$t_{tr}=t^*-i\Omega^{-3/2}E^{1/2}. \tag{28}$$

The contour must therefore be provided with an appendage that passed through the point t_{tr}. The increment to the imaginary action due to circuiting by this appendage, is equal to

$$\Delta S_A=2\,\mathrm{Im}\int_{t_{tr}}^{t^*}(E-E_{ST}(t))\,dt=\frac{4}{3}\left(\frac{E}{\Omega}\right)^{3/2}. \tag{29}$$

The integration in (29) was carried out with (27) and (28) taken into account. The factor 2 is due to the presence of two appendages, upper and lower (Fig. 3d). It can be seen from (29) that the probability of coherent energy transfer from the electron to the lattice is indeed exponentially small at $E>\Omega\sim(\omega^2W)^{1/3}$.

For imaginary $t-t^*$ the energy $E_{ST}(t)$ can be interpreted as the location of a quasilocal level above the continuous spectrum of the electron. This level is formed in a complex potential produced by complex displacements Q of the lattice. At the instant t_{tr} the level energy coincides with the electron energy and the latter is resonantly captured on the level. Equation (29) is outwardly similar to the usual equation of adiabatic perturbation theory,[28,24] where adiabatic energies which are two branches of one analytic function are located on the two edges of the cut, and the contour can be deformed and still bypass the branch point. There is nevertheless a substantial difference. The energies E_{ST} and E in (29) are actually two independent functions, so that the contour cannot be shifted away from the point t_{tr}.

As a result, the imaginary action for hot particles can be written in the form

$$S_A(E,T)=(W-E)/T+{}^4/_3(E/\Omega)^{3/2}. \tag{30}$$

The first term is a modification of the action $S_A=W/T$ that enters in the arguments of the exponentials in Eqs. (23)–(25). This modification results from the already mentioned omission of Gibbs averaging over the electron energy. The physical meaning of the first term is easily understood. The electron contributes an energy E to the total energy W needed to surmount the barrier. The heat bath need therefore supply only the energy deficit $W-E$. The second term determines the probability of transferring an energy E from the electron to the lattice, and agrees with Demkov's exact solution[29] (cf. the Appendix). S_A has a nonmonotonic dependence on E. At small E it always decreases linearly. The minimum is reached at $E_{\min}\sim(\omega/T)^2W$, with

$$S_A(0,T)-S_A(E_{min},T)\sim(\omega/T)^2(W/T).$$

The equations obtained are valid if (27) can be used, meaning that $\omega|t^*-t_{tr}|\ll 1$ or $E\ll W$. Only under these conditions can recoil be neglected and electron capture considered for a given motion of the lattice.

We change now to the instanton regime. Since the point t^* is now on a vertical segment, the potential well becomes deeper as we move downward from the point t^*, i.e., in the direction of imaginary time. We have therefore in place of (27) and (28)

$$E_{ST}(t)=\Omega^3(t-t^*)^2,\quad t_{tr}=t^*-\Omega^{-3/2}E^{1/2}, \tag{31}$$

and the contour takes the form shown in Fig. 3e. The same integral as in (29) determines ΔS_I, but its imaginary part is zero in this case, so that $\Delta S_I=0$. As a result, the action for hot carriers is equal to

$$S_I(E,T)=S_I(T)-2\tau_0E,\quad \tau_0=\mathrm{Im}\{t^*\}. \tag{32}$$

This equation, just as (30), is valid if $E\ll W$.

For $E\sim W$ it is impossible to obtain the dependence of S on E in general form. If $E\gg W$, however, estimates can be

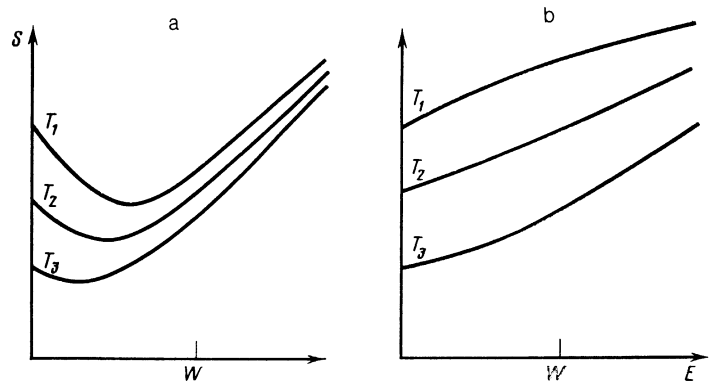

FIG. 5. Schematic dependence of S on the electron energy E at various lattice temperatures ($T_1 < T_2 < T_3$): a) intrinsic and extrinsic self-trapping; b) recombination centers.

made from scaling considerations. The results are found to depend on the model. For two models (deformation interaction with acoustic and optical phonons) it was shown in Ref. 22 that S_A and S_I increase as powers of E in this region. It follows from all the foregoing that $E_{\min} \sim W$ in the low-temperature region and $E_{\min} \ll W$ at high temperatures. Approximate plots of $S(E,T)$ are shown in Fig. 5a.

One general relation valid for all E and T can be established. The shape of the contour Γ and in particular the values of t_{tr} are determined by E and T. Formally, however, it is convenient to regard the imaginary action as a function, $S = S(E,T,t_{tr})$, with t_{tr} chosen to meet the condition $\partial S / \partial t_{tr} = 0$ [it can be shown that Eqs. (28) and (31) satisfy this condition]. The total derivative is therefore $dS/dE = \partial S / \partial E$. The explicit dependence of S on T is obtained by integrating $(-E)$ with respect to time along Γ_E. The resulting contribution is equal to $(-2E\,\mathrm{Im}\{t_{tr}\})$. As a result,

$$dS/dE = -2\,\mathrm{Im}\{t_{tr}\}. \tag{33}$$

It follows hence, in particular, that t_{tr} is real at the point $E = E_{\min}$. In the region where S increases with E we have $\mathrm{Im}\{t_{tr}\} < 0$. This shows that at $E \gtrsim W$ the contours become considerably more complicated than those shown in Figs. 3d and 3e (see Ref. 22).

Averaging the self-trapping probabilities given by (30) and (32) over an equilibrium Gibbs distribution leads to the equations of Secs. 2 and 3.

Returning to the questions raised at the beginning of the present section, we can assert that the self-trapping rate increases in a definite range of electron energies E. It remains, nevertheless, exponentially small. The argument of the exponential, however, decreases noticeably and this can lead to a considerable diversion of electrons to the self-trapping state prior to their thermalization.

5. TRAPPING BY IMPURITY CENTERS

For electrons interacting with impurity centers there exist adiabatic-potential surfaces of two basic types.[8,30,31] They are shown in Figs. 6a and 6b for the one-dimensional model. Those of the first type are called intrinsic self-trapping centers,[21] and of the second "normal" defects or recombination centers.

The adiabatic-potential surfaces for the centers of the first type differ from the analogous surface for intrinsic self-trapping in the bulk (Fig. 1) in having no translational symmetry. Specific centers in $Al_x Ga_{1-x} As$ were ascribed in Ref. 30 to both types. The qualitative difference between them is that the points *br* and $\mathscr{W}$ are separated in case a but are superposed in case b ($\mathscr{W}$ lies on the *br* surface in the multidimensional case.)

Interest in multiphonon trapping was stimulated recently by Henry and Lang's analysis[8] of long-time photoconductivity in terms of the theory of multiphonon transitions. Capture by centers of type b was analyzed in detail by Abakumov *et al.*[9] In a model where an electron interacts with one local mode. A comparative analysis of the picture of

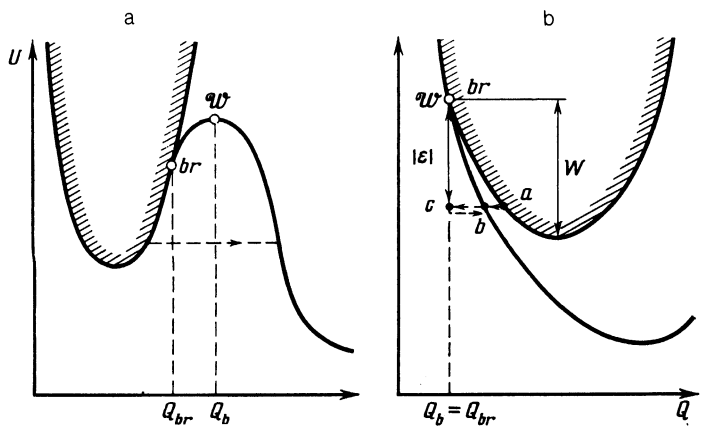

FIG. 6. Adiabatic-potential curves for impurity centers: a) extrinsic self-trapping, b) recombination center. The shaded lines show the tunneling trajectories.

capture by centers of both types was made by Meshkov[10] on the basis of a similar model. The important step made in Refs. 9 and 10 is the consistent description of the system motion in the vicinity of the point *br*. The methods used in Refs. 9 and 10 for impurity centers and in Ref. 22 for intrinsic self trapping are close to one another.

The task of the present section is to examine the types of behavior that are typical of multimode systems and therefore are lost sight of in the one-mode description. Another task is to determine the features that are common and are not connected with particular models.

The rate of nonradiative capture of electrons by impurity centers, averaged over the thermal distribution, is equal to

$$w_T = N_i \langle v_e \sigma(v_e) \rangle, \tag{34}$$

where N_i is the density of the centers, v_e the electron velocity, and σ the capture cross section. The general formalism of Refs. 2 and 23 and of Secs. 2 and 3 of the present paper can be directly applied to impurity centers.

We consider first the temperature dependence of w_T in the exponential approximation $w_T \propto \exp(-S)$, where $S(T)$ is the imaginary action. The value of $S(0)$ is determined by the detailed structure of the centers, and local modes can make an appreciable, possibly even decisive, contribution. The low-temperature dependence of $S(T)$ is, however, always determined by interaction with acoustic phonons, which are the only low-frequency modes of the system. The low-temperature correction $\Delta S(T)$ to the action can be satisfied by making the motions in the system (made up of long-wave acoustic modes and the instanton formed by the short-wave and local modes) self-consistent so as to satisfy the least-action condition.[3)] Naturally, this correction depends on the space-time form of the instanton, so that the numerical coefficient cannot be found in general form. The equations for deformation and piezoacoustic electron-phonon interactions differ greatly. Estimating ΔS in analogy with Refs. 2 and 23 and assuming $\tau_0 \sim \omega^{-1}$, we get

$$\Delta S_{def}(T) \sim \frac{C^2}{\rho s^5 \omega^2} T^4, \quad \Delta S_{p\text{-}ac} \sim \frac{e^2 \beta_p^2}{\rho s^3 \omega^2} T^2. \tag{35}$$

Here C is the deformation potential, β_p the piezomodulus, ρ the density, and s the speed of sound.

The analysis of case a is quite similar to that of intrinsic self-trapping. The only difference is that the number of extremals is not V/v but VN_i. As a result we obtain in lieu of (11) and (24), respectively,

$$w_T \sim p_e(T) N_i \omega (m\omega)^{-1/2} \exp(-S_I(T)),$$
$$w_T \sim N_i \omega (mT)^{-1/2} \exp(-W/T), \tag{36}$$

$p_e(T)$ is determined as before by Eq. (6). Just as for intrinsic self-trapping, the temperature dependence is monotonic, and at the very lowest temperature it is determined by the electronic factor $p_e(T)$.

Case b differs in one important respect from case a: here $\tau_0 < 0$ (see Fig. 3f). The reason is the following. It can be seen from Fig. 6b that on the tunneling trajectory the point *br* is a turning point.[33,9] At the same time, the "imaginary" velocity $\dot{Q}(\tau)$ (τ is the imaginary time) is not zero at this point. To ensure continuity of $\dot{Q}(\tau)$ it is therefore necessary to invert the contour at the point t^*. Complex trajectories with backward motion in time were considered in detail for this problem in Ref. 10. Since $\tau_0 < 0$, $p_e(T)$ becomes a decreasing function of the temperature [see Eq. (6)]. This makes the temperature dependence of w_T [Eq. (36)] nonmonotonic with a minimum in the low-temperature region ($T_{\min} \ll \omega$).

The specifics of case b are most clearly manifested at high temperatures ($\omega\beta \gg 1$). In contrast to the case a, the process retains the properties of thermally activated tunneling right up to the highest temperatures, and there is no transition to the activation regime. The optimum tunneling energy increases with T and approaches W asymptotically. As a result, the tunneling correction always causes the imaginary action S_I to be somewhat smaller than $S_A = W/T$. (In case a at $T \gtrsim T_c$ there is likewise a tunneling contribution to the current, but this contribution alters only the coefficient of the exponential and does not affect its argument.)

The activation regime is absent because the point $\mathscr{W}$ has moved to the *br* surface. The lowest saddle is therefore reached not at the point where the function $U(Q)$ is analytic, but on a multidimensional sharply peaked crest. Since the trajectories of importance at $\beta\omega \ll 1$ are those passing in a narrow region near $\mathscr{W}$, the variables can be separated (see Sec. 3). Introducing a longitudinal coordinate q_0 measured from $\mathscr{W}$, and transverse coordinates Q_i, we obtain for the two branches of the adiabatic potential in the vicinity of $\mathscr{W}$:

$$U_1 \approx A_1 q_0^2 - B_0 q_0 + \frac{1}{2} \sum_{i \neq 0} \omega_{Wi}^2 Q_i^2,$$
$$U_2 \approx A_2 q_0^2 - B_0 q_0 + \frac{1}{2} \sum_{i \neq 0} \omega_{Wi}^2 Q_i^2,$$
$$A_1 \sim A_1 - A_2 \sim \omega^2, \quad B_0 \sim \omega W^{1/2}. \tag{37}$$

A_1, A_2, and B_0 were estimated under the assumption that the dependence of U_1 on q_0 does not deviate greatly from parabolic, and that U_1 and U_2 have the same scale. For the reduced action corresponding to a transition between different points (see Fig. 6b), the following estimates are valid:

$$S_0(ac|\varepsilon) \approx S_0(bc|\varepsilon) \sim |\varepsilon|^{3/2}/\omega W^{1/2},$$
$$S_0(\varepsilon) \equiv S_0(ab|\varepsilon) = S_0(ac|\varepsilon) - S_0(bc|\varepsilon) \sim |\varepsilon|^{5/2}/\omega W^{3/2}. \tag{38}$$

Here the energy ε is measured from W. The minimum of the imaginary action $S_I(\varepsilon) = 2S_0(\varepsilon) + \beta(W + \varepsilon)$ is reached at

$$\varepsilon_{min} \sim -(\beta\omega)^{2/3} W \tag{39}$$

and is equal to

$$S_I(T) \approx (W/T)(1 - c(\beta\omega)^{2/3}), \quad c \sim 1. \tag{40}$$

Equations (38)–(40) are valid when the tunneling correction to the action is large: $(W/T)(\beta\omega)^{2/3} \gg 1$. This quasiclacissism condition is met up to $T \sim \omega (W/T)^{3/2}$. The region of higher temperature is of no practical importance. Thus, an instanton solution exists at practically all T, but there are no activation solutions at all (i.e., $T_c = \infty$). As $\varepsilon \to 0$, however, the difference between the behaviors typical of the instanton

and activation solutions vanishes for the most part (see Fig. 2c).

We show now that the recoil of thermalized electrons captured by a discrete level can be neglected (in analogy with Secs. 2 and 3). To this end we put again $E_{ST}(\tau) = -\Omega_m^2(\tau-\tau_0)^2$ and determine Ω_m from the following estimates: according to (37), $E_{ST} = (A_1 - A_2)q_0^2$. On the other hand,

$$\tau_0(\varepsilon_{min}) = dS_0(bc|\varepsilon)/d\varepsilon|_{\varepsilon=\varepsilon_{min}} \sim -\omega^{-1}(\beta\omega)^{1/3}. \quad (41)$$

Calculating $E_{ST}(\tau)$ for $|\tau-\tau_0| \sim |\tau_0|$, we obtain the estimate

$$\Omega_m \sim (\beta\omega)^{2/3}(\omega^2 W)^{1/3}. \quad (42)$$

It can be seen that Ω_m, in contast to Ω [Eq. (A1)], depends on T. It is easy to verify that the adibaticity condition for an electron level at times $\sim|\tau_0|$, which takes the form $\Omega_m|\tau_0| \gg 1$, coincides with the quasiclassical condition obtained above.

For thermalized particles, p_e is calculated in analogy with Eq. (6), but in this case $\tau_0 < 0$ and $\exp\{(-2|\tau_0|E(\mathbf{k})\}$ is averaged out. It follows from (41) that $|\tau_0| \gg \beta$. As a result, the integral over the Gibbs distribution converges to energies $\sim|\tau_0| \ll T$. The particles predominantly captured have therefore energy $\sim|\tau_0|^{-1}$, which is much lower than thermal. From the inequality

$$E \lesssim |\tau_0|^{-1} \sim \omega(\beta\omega)^{-1/3} \ll |\varepsilon_{min}|$$

it follows that recoil can be neglected.

The capture probability can be written in the form (cf. Sec. 3)

$$w_T \approx (N_i V) p_e \frac{|u(0)|^2}{V} \frac{\prod_i \operatorname{sh}(\beta\omega_i/2)}{\prod_{i\neq 0} \operatorname{sh}(\beta\omega_{Wi}/2)} \int \frac{d\varepsilon}{2\pi} e^{-\beta(W+\varepsilon)-2S_0(\varepsilon)}. \quad (43)$$

Calculating the integral by the saddle-point method and recognizing that $p_e \sim |\beta/2\tau_0|^{3/2} \sim \beta\omega$, $|u(0)|^2 \sim (m\Omega_m)^{-3/2}$ and $d^2S_0/d\varepsilon^2 \sim (\beta\omega)^{-1/3}W\omega$, we obtain ultimately

$$w_T \sim N_i\omega(mT)^{-1/3}\exp\{-S_I(T)\}. \quad (44)$$

It is of interest to note that, to within the difference between $S_I(T)$ given by (4) and S_A, Eq. (44) agrees with the second equation of (36), obtained for case a by considering the activation mechanism. The coefficients agree in both limiting cases with those obtained by Abakumov *et al.*[9] for a special model of the recombination center.

From the difference in the signs of τ_0 for extrinsic self-trapping ($\tau_0 > 0$) and for recombination centers ($\tau_0 < 0$) it follows that the trapping rates of the hot particles depend differently on their energies E. It follows from (32) that when E is low, $S_I(E,T)$ increases with E for recombination centers. Since at high E the action also increases with E, as shown in Ref. 22, $S_I(E,T)$ can be regarded as a monotonically increasing function of E (Fig. 5b) and $w(E,T)$ as a monotonically decreasing one. Therefore slow carriers recombine most effectively via "normal" centers. A similar result was obtained in Ref. 10 within the framework of a single-mode model.

6. CONCLUSION

The probability of multiphonon capture of an electron in a crystal (self-trapping, recombination) can be written in the form

$$w = \omega B \nu \exp(-S). \quad (45)$$

Here ω is the characteristic frequency of the phonons, and $S \gg 1$ is the classical action; the vector ν is the dimensionless density of the capture centers. For intrinsic self-trapping we have $\nu = 1$, and for capture by various impurity centers $\nu = N_i v$ (as before, N_i is the density of the centers and v is the cell volume). This notation is valid both for the activation regime (high T) and for thermoactivated tunneling (low T). The question is what are the values of S and B and how do they depend on the lattice temperature T and on the electron energy E? It turns out that $B \gg 1$ in all cases.

Three nontrivial factors must be taken into account: 1) the multiplicity of the lattice modes; 2) the presence of a temporal zero mode, and 3) the nonadiabaticity of the process during the electron-capture stage.

The lattice mode multiplicity determines primarily the low-temperature behavior of S. It is formed by low-frequency acoustic phonons and is different for the deformation and piezoelectric interactions [Eq. (35)]. If surmounting of the barrier can be described by the continuum model, three spatial zero modes appear for the intrinsic self-trapping and as a result the mode multiplicity increases B by a factor $S^{3/2} \gg 1$. The temporal zero mode always introduces in B a factor $S^{1/2}$ at low temperatures. The non-adiabaticity during the trapping stage causes B to contain in all cases the large factor $r_{tr}^3/v \gg 1$. Here r_{tr} is the radius of the electron ψ function at the boundary of the adiabaticity region.

Allowance for the initial energy E of the thermalized carriers in the low-temperature region introduces in B the factor $p_e(T)$ [Eq. (6)]. This factor determines the temperature dependence of w_T as $T \to 0$, which has opposite signs for self-trapping (intrinsic or extrinsic) and "normal" recombination centers. Systematic experiments in this temperature range are highly desirable.

In the high-temperature region, Gibbs averaging over E leads to the relation $B \propto T^{-3/2}$. If spatial zero modes exist, we have $B \propto T^{-3}$ (the action at high T is $S = W/T$, where W is the height of the barrier). Since semilog plots of many experimental data have shown the surprisingly low values $W \sim (2-3)\omega$, correct allowance for $B(T)$ is very urgent; it will lead to a noticeable increase of W.

The estimates of B can be generalized in the following manner. At low temperatures we have

$$B(T) \propto p_e(T) \begin{cases} (W/\omega)^3 \text{—in the continuum limit for intrinsic self-trapping,} \\ (E_B/\omega)^{3/2} \text{—in all other cases,} \end{cases} \quad (46)$$

where E_B is the halfwidth of the band. At high temperatures, we replace ω by T and put $p_e = 1$.

If the electrons are not thermal, allowance for their initial energy leads to the even more important consequence that S is changed. This is due to simultaneous action of two mechanisms: suppression of the fast-electron trapping probability and an influence of the electron's initial energy of the lattice motion. These mechanisms compete in the self-trapping process; as $T \to 0$, therefore,the largest w is obtained for electrons with $E \sim W$. For recombination centers these mechanisms are additive, and w is a maximum at $E = 0$.

The most difficult problem is to determine the absolute values of B and S. The numerical coefficients depend strongly on the model. Omitting initially the calculation, we tried to express all the quantities in terms of the three parameters W, E_B, and ω. Naturally, estimates of all the quantities involved in the theory in terms of these three are bound to be crude. It is impossible, however, to improve on them without resorting to specific models.

As for nonradiative capture phenomena, the comparison of the theory with experiment is at present in its initial stage. It appears that the experimental data are so far insufficient even to determine the position of the point T_c at which the regimes change over. Light may be cast on the situation by measurement of the capture rate in a wide range of temperatures.

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APPENDIX

Consider an electron in a potential well that becomes monotonically deeper with time and is produced by a moving lattice. Assume that the well parameters vary smoothly over a time $\sim\omega^{-1}$ without change of scale. Let a local level be produced at the instant $t = 0$ and let it deepen with increase of t such that after a time $\sim\omega^{-1}$ it reaches a depth $\sim W \gg \omega$. At times $t \ll \omega^{-1}$ the level energy depends quadratically on t and can be expressed as $E = -\Omega^3 t^2$, where

$$\Omega \sim (\omega^2 W)^{1/3}. \tag{A1}$$

The level is adiabatic at $T > \Omega^{-1}$, so that the particle is trapped within a time $t \lesssim \Omega^{-1} \ll \omega^{-1}$. Consequently, during the entire capture stage the level remains shallow ($|E| \ll W$) and can be described by the zero-radius-potential method.[16] The problem was solved by Demkov[29] and i follows from his result that the capture probability is equal to $|u(\mathbf{k})|^2/V$, where

$$|u(\mathbf{k})|^2 = \frac{8\pi^2}{(2m\Omega)^{3/2}} \exp\left\{-\frac{4}{3}\left(\frac{k^2}{2m\Omega}\right)^{3/2}\right\},$$

$$\int |u(\mathbf{k})|^2 \frac{d\mathbf{k}}{(2\pi)^3} = 1. \tag{A2}$$

The quantity $|u(\mathbf{k})|^2$ has the dimension of the volume from which the particle is effectively trapped on a level. For slow particles the trapping radius is $r_{tr} = |u(0)|^{2/3} \sim (m\Omega)^{-1/2}$ and has the meaning of the radius of the wave function at the limit of the adiabaticity region, i.e., at $t \sim \Omega^{-1}$.

[1)]The situation for "normal" recombination centers is special; it is considered in Sec. 5. In this case $\beta_0 = 0$ and there is no activation segment (Fig. 2c).

[2)]Numbered equations of Ref. 2 are preceded here and below by the Roman numeral I.

[3)]Increasing the tunneling probability by optimal displacement of neighboring atoms was named "preparation of the barrier" in Ref. 32.

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