A quasiclassical theory of multiphonon nonradiative transitions

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A quasiclassical approach to the description of finite-temperature nonadiabatic and tunnel transitions is proposed, based on analytic continuation of the equilibrium density matrix along the lines of Landau's method of complex classical paths. This approach separates out explicitly the exponentially small factors in the transition probabilities and indicates how the coefficients of the exponential terms can be calculated correctly. An extension of the approach to the case of transitions to the continuous spectrum, using a quasiclassical description of the free-electron motion, makes it possible to analyze both multiphonon trapping on an impurity center in a semiconductor and self-trapping of current carriers and excitons from a single viewpoint, including the energy dependences of the rates of these processes. Specific calculations are carried out for simplified models which yield exponential accuracy.

1. INTRODUCTION

Multiphonon nonradiative transitions provide an important channel for electron relaxation in solids, especially when the electronic transition energy is large and intermediate electronic states are absent. Under these circumstances, it becomes necessary to apportion the excitation energy of the "unrelaxed" electron among a large number of lattice vibration quanta (in this work we will not investigate the radiative and Auger processes which compete with these phonon-mediated transitions). Theoretical treatments of this kind of relaxation are based on an adiabatic description of the system, involving rapidly-moving electrons which "track" the much slower motion of the massive nuclei of the lattice. Multiphonon nonradiative transitions between electronic states appear in this context as deviations from adiabaticity (see reviews 1, 2); this nonadiabatic character is shared by nonradiative electronic transitions in molecules[3] and is also encountered in inelastic atomic collisions.[4]

In the simplest (idealized) case of multiphonon transitions between discrete states of a deep impurity center, the usual method of attack is as follows: in the full Hamiltonian of the electron-lattice system, the kinetic energy of the lattice, which is quadratic in the nuclear momenta, is separated out:

$$\hat{H}=K(-i\hbar\partial/\partial\mathbf{q})+\hat{H}_0(\mathbf{q}). \tag{1}$$

The vector $\mathbf{q}$ denotes the ensemble of lattice coordinates q_i; the operator $\hat{H}_0(\mathbf{q})$ does not include differentiation with respect to them, i.e., it depends on $\mathbf{q}$ only parametrically. For each value of $\mathbf{q}$ there is an eigenvalue $U_\alpha(\mathbf{q})$ and an eigenfunction $|\alpha\mathbf{q}\rangle$ of the electronic Hamiltonian; the index α enumerates the terms of the discrete spectrum. Using the Born-Oppenheimer approximation, we can find the eigenfunctions of the full Hamiltonian (1) in the form

$$\Phi_\alpha(\mathbf{q})\,|\alpha\mathbf{q}\rangle, \tag{2}$$

where $\Phi_\alpha(\mathbf{q})$ is an eigenfunction of the "adiabatic" Hamiltonian:

$$\hat{H}_A\Phi_\alpha(\mathbf{q})=E_\alpha\Phi_\alpha(\mathbf{q}), \quad \hat{H}_A\equiv K(-i\hbar\partial/\partial\mathbf{q})+U_\alpha(\mathbf{q}). \tag{3}$$

The wave function (2) is an eigenfunction of the full Hamiltonian (1) if small terms are neglected which come from differentiating the electronic function $|\alpha\mathbf{q}\rangle$ with respect to $\mathbf{q}$; this differentiation arises from the action of (1) on (2). The presence of these terms is usually traced to the action of a so-called "nonadiabaticity" operator $\hat{L}$ defined implicitly by the expression

$$\hat{H}\Phi_\alpha(\mathbf{q})\,|\alpha\mathbf{q}\rangle-E_\alpha\Phi_\alpha(\mathbf{q})\,|\alpha\mathbf{q}\rangle\equiv\hat{L}\Phi_\alpha(\mathbf{q})\,|\alpha\mathbf{q}\rangle.$$

In reality, $\hat{L}$ is not a true operator, since its action on an arbitrary function of the electronic and lattice coordinates is not defined.

The inexactness of the adiabatic approximation leads to mixing of adiabatic wave functions (2) with different α, or in other words, to transitions between terms. The usual approach to calculating the probabilities for these transitions (see Refs. 1, 2), sometimes called "adiabatic perturbation theory" and first used by Kronig[5] for investigating the predissociation of molecules, is to regard the nonadiabaticity operator as a small perturbation which causes the transitions in the usual way. In practical terms, this approach consists of choosing in place of the matrix elements of the full Hamiltonians (1) the set of orthonormal adiabatic functions (2) with different indices and substituting them into the "golden rule" of quantum mechanics for the transition probability:

$$\tau^{-1}=2\pi\hbar^{-1}\sum_f|\langle f|\hat{H}|i\rangle|^2\delta(E_f-E_i), \tag{4}$$

where the summation over the final states $|f\rangle$ is carried out on the assumption that the spectrum is quasicontinuous. Actually, expression (4) is the first term of an iteration series which arises from solving the time-dependent Schrödinger equation in the basis of adiabatic functions (2). The terms of this series are all of equal (exponential) smallness, but do not decrease in magnitude, so that the golden rule (4) when used for nonadiabatic transitions is only an estimate, and not strictly an expansion in the adiabaticity parameter (see, e.g., Ref. 3, §28).

Treatments analogous to the one described above have also been used to investigate the related problems of transitions induced by slowly varying external perturbations and

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high-energy reflection from a barrier. In due course it was established (see Ref. 6, §52, §53, and Refs. 7, 8) that these problems were in essence quasiclassical and thus required investigation in the spirit of Landau's method of complex classical paths. As we will show here, the quasiclassical method must also be used to investigate multiphonon nonradiative transitions; this is because the condition for quasiclassical motion of the lattice, i.e., slow variation of the adiabatic potential energy $U_\alpha(\mathbf{q})$ in (3) over a wave length characteristic of this motion, is in essence identical to the requirement of adiabatic slowness of lattice vibrations compared to electronic motion. Of course, the situation can be complicated by the presence of other small parameters in addition to the adiabaticity parameter. Disregarding these complications, we can assert that a consistent expansion in the adiabaticity parameter leads not to perturbation theory but rather to a quasiclassical approximation which is based on a classical treatment of the lattice motion in the adiabatic potential $U(\mathbf{q})$ corresponding to purely adiabatic variation of the electron wave functions.

The quasiclassical transition amplitude from one point $\mathbf{q}'$ to another $\mathbf{q}$ for a system with kinetic energy $K(\mathbf{q})$ and potential energy $U(\mathbf{q})$, according to general expressions obtained long ago by Fock,[9] equals

$$G(\mathbf{q},t|\mathbf{q}',t')=A\left|\det\left[\frac{\partial^2 S(\mathbf{q},\mathbf{q}')}{\partial q_i\,\partial q_j'}\right]\right|^{1/2}\exp\left[\frac{i}{\hbar}S(\mathbf{q},\mathbf{q}')\right], \tag{5}$$

where $S(\mathbf{q},\mathbf{q}')$ is the value of the classical action

$$S[\mathbf{q}(t)]=\int_{t'}^{t}[K(\dot{\mathbf{q}})-U(\mathbf{q})]\,dt \tag{6}$$

along an extremal path starting at the point $\mathbf{q}'$ at time t' and ending at the point $\mathbf{q}$ at time t; A is a normalization factor which does not depend on $\mathbf{q}$, $\mathbf{q}'$, t or t'. The adiabatic potential energy, regarded as a function of the complex lattice coordinates $\mathbf{q}$, is in general a single many-valued analytic function whose branches are the electronic terms. Each branch represents a specific sheet of a Riemann surface, so that a transition from one sheet to another is effected by a circuit of the corresponding manifolds which define the branch-type singularities (in the one-dimensional case, branch *points*); these singularities are pairwise complex conjugates in view of the reality of $U(\mathbf{q})$ for real $\mathbf{q}$.[1)] A path which describes a nonadiabatic transition must go from one electronic term to another; because this path must encircle the above-mentioned branch singularities, the action along it is complex, causing the transition amplitude to be exponentially small. For this reason, nonadiabatic transitions between terms are analogous to transmission through a potential barrier by tunneling. The fact that the action is complex for a tunneling transition, however, is not a consequence of circling any potential singularities, but rather comes from traversing the classically forbidden region.

The approach we will describe in this work was used by Miller[10] in the theory of slow inelastic atomic collisions. In this case, both initial and final states belong to the continuous spectrum of the adiabatic Hamiltonian (3), and the scattering amplitude is given directly by the Green's function (5) in the momentum representation. The origin of nonadiabatic transitions in impurity centers and molecules, on the other hand, can be ascribed to the quantum-mechanical decay of states in a quasidiscrete spectrum which is associated with the vicinity of some minimum of the adiabatic potential $U(\mathbf{q})$. The point-to-point transition amplitude (5) by itself can say nothing about the transition probability of these processes.

In one-dimensional models, where the "lattice" is described by a single coordinate $\mathbf{q}$ and the classical path is completely determined by the energy, one avoids certain fundamental difficulties characteristic of the general case. The system motion takes the form of oscillation around a potential energy minimum at one of the electron terms, while the transition probability per cycle of oscillation from this term to other terms is given by a transition "coefficient" completely analogous to the tunneling transmission coefficient for a one-dimensional potential barrier (see Ref. 6, §50)

$$D(E)=2\exp\left[2\hbar^{-1}\,\mathrm{Im}\left\{\oint[2M(E-U(q))]^{1/2}dq\right\}\right], \tag{7}$$

where M is the mass of the lattice. The integration in (7) amounts to calculating a "truncated" action [in contrast to (5)] along a contour with real end points located in the classically allowed regions around the initial and final electronic terms. The coefficient 2 of the exponential, like the factor of two which arises in the theory of Landau and Zener (Ref. 6, §90), is connected with the possibility of making a circuit around either of two complex-conjugate branch points of the potential $U(q)$. The imaginary part of the action on these two paths is the same while the real parts differ; this leads to rapid interference oscillations in $D(E)$ which disappear when we average over a small energy interval.

The present report is dedicated to applying the quasiclassical approach to more complicated situations. In section 2, we present a general method for describing nonadiabatic transitions and tunneling in systems with many degrees of freedom at finite temperature. In section 3 this method is generalized and applied to the problem of capture of current carriers by a deep trap in a crystal, while in section 4 the same method is used to investigate barrier autolocalization (self-trapping). In contrast to simple trapping, the quasiclassical description is customary in the theory of autolocalization,[11-13] although autolocalization at finite temperature is investigated for the first time in Ref. 13. The formalism developed in the present report differs from that presented in Ref. 13; a comparison of results is given at the end of section. 4.

Calculations are carried through to the end with exponential accuracy in simplified analytically soluble models. The quasiclassical approach allows us to highlight the exponential smallness of the transition probabilities and to formulate precisely the problem of calculating the argument of the coefficient of the exponential which will be the fundamental object of investigation in the present article. This coefficient, whose calculation is significantly more difficult, is usually estimated (in order of magnitude) using the characteristic vibration frequencies of the system.

It must be noted that first-order adiabatic perturbation

theory, when used with the quasiclassical wave functions of the Born-Oppenheimer approximation, also should yield exponentially small results with accurate values of the exponential arguments (see Ref. 3, ch. 4). In practice, however, this approach usually is accompanied by a quadratic expansion around minima of the adiabatic potential $U(\mathbf{q})$, and sometimes other rather crude approximations are invoked (see Refs. 1, 2). It is clear that the value of the exponent so derived can only be an estimate, since in reality the total action along the path which describes the transition is made up of contributions which can originate quite far from the potential minimum. Exponential accuracy in the results one usually derives will be attained only in those models where the terms interact weakly, and where these terms are purely oscillatory in character everywhere except over a narrow region where the interaction is significant.

2. QUASICLASSICAL DENSITY MATRIX

As is well-know (see, e.g., Ref. 14), quantization of the energy spectrum of a multidimensional potential well by quasiclassical methods is possible only when separation-of-variables techniques can be applied. The question of separability also causes special difficulties in the general investigation of transitions in dynamic systems with more than one degree of freedom. A quasiclassical calculation of the rate of nonadiabatic or tunneling decay of a particular stationary state in a specific valley of the potential $U(\mathbf{q})$ makes no sense; only a transition probability averaged, let us say, over a small energy range of the spectrum, can be calculated quasiclassically. In what follows, we will investigate this averaging procedure using a Gibbs population distribution over all initial states, in the limit of a single valley within which lies some minimum of the potential $U(\mathbf{q})$.

Formulating the problem in this way presupposes the presence of a heat bath with which the system interacts; the interaction should be strong enough to mediate the establishment of thermodynamic equilibrium in the initial valley, but weak enough to neglect in the system equations of motion. In practice, this means that the time τ_0 it takes to establish thermal equilibrium must be small compared to the time τ it takes for the system to leave the valley, but large compared to one of its typical periods of oscillation. For a discrete spectrum of final states, a lower bound on the strength of the interaction with the heat bath which is more important than the one described above is the condition that the spacing between final state levels be small compared to their thermal broadening. Only in this case is it permissible to use the quasiclassical approximation of treating the final-state spectrum as a quasicontinuum. In the case of an impurity center in a crystal, equilibrium is established in the limit of a single term by one-phonon processes whose intensity is usually sufficient to satisfy the conditions outlined above. In molecules, matters can be more complicated (see Ref. 3).

A general method allowing us to find the total probability per unit time for the escape of a system from a metastable valley at finite temperatures has been presented by Langer[15] and involves calculating the imaginary part of the system free energy. By representing the statistical sum in the form of a Feynman integral,[16] and in the latter choosing the required path of steepest descent, we are led to the following exponential expression for the transition probability:

$$\tau^{-1} \propto \exp[i\hbar^{-1}S(\beta)], \tag{8}$$

where $S(\beta)$ is the action (6) taken over one period of that classical path which has a period of $-i\hbar\beta$ (β is the inverse temperature). The potential $U(\mathbf{q})$ is measured from the bottom of the valley; after examining the various extremal paths, we choose the one which gives the largest τ^{-1}. The coefficient of the exponential can also be calculated, though this is much more difficult. A shortcoming of this approach is that it provides no information at all about the partial probabilities for transitions into various final states, which sometimes (see the next section) are of principal interest.

We choose to take a somewhat different approach here, which preserves Langer's method but is free of these deficiencies. The evolution of the density matrix $\hat{\rho}$ of a system initially located in some valley is described quantum-mechanically by the equation of motion

$$i\hbar\dot{\hat{\rho}} \approx \hat{H}\hat{\rho} - \hat{\rho}\hat{H},$$

perturbed by the weak interaction with the heat bath and by processes which cause the sytem to leave the valley. On the time scale τ_0, a distribution is established with a density matrix which, except for exponentially small damping, is stationary (commutes with the Hamiltonian); in the final-state region (i.e., beyond the barrier for tunneling or at some other electronic term in the case of nonadiabatic transitions) the current associated with this density matrix is purely outgoing, i.e., leaving the valley, while deep in this valley the density matrix has the exponential form $\exp(-\beta H)$. Separating out the time dependence

$$\hat{\rho}(t) = \exp(-t/\tau)\,\tilde{\rho}(\mathbf{q}, \mathbf{q}')$$

for $\tau \gg \tau_0$, we arrive at a function $\tilde{\rho}(\mathbf{q}, \mathbf{q}')$ depending only on the coordinates; we will refer to $\tilde{\rho}$ henceforth as the quasi-equilibrium density matrix, which is conveniently normalized to unity by integrating $\tilde{\rho}(\mathbf{q}, \mathbf{q})$ over the initial valley. All possible information about the transition probabilities into various final states from the thermodynamic equilibrium distribution in the initial valley is contained in $\tilde{\rho}(\mathbf{q}, \mathbf{q}')$ or in the corresponding Wigner distribution (see, e.g., Ref. 17):

$$f(\mathbf{p}, \mathbf{q}) \equiv (2\pi\hbar)^{-n} \int \exp(-i\hbar^{-1}\mathbf{p}\boldsymbol{\xi})\,\tilde{\rho}(\mathbf{q}+{}^1/_2\boldsymbol{\xi}, \mathbf{q}-{}^1/_2\boldsymbol{\xi})\,d^n\boldsymbol{\xi} \tag{9}$$

(n is the number of degrees of freedom), which is more transparent in form and contains all the physical information.

The procedure described above can be compared to the use of quasistationary wave functions in the description of the escape of a particle from a potential well by tunneling (see, e.g., Ref. 18, ch. 7), in which one chooses in a similar fashion an analytic continuation of the wave function of some stationary state. An explicit form for $\tilde{\rho}(\mathbf{q}, \mathbf{q}')$ can be obtained in an analogous fashion. We turn to the well-known formulation of the equilibrium density matrix as a Feynman integral[16] with the action (6), i.e.,

$$\rho(\mathbf{q}, \mathbf{q}') = \int \exp\{i\hbar^{-1}S[\mathbf{q}(t)]\}\,\mathcal{D}[\mathbf{q}(t)], \tag{10}$$

along paths which start at the point $\mathbf{q}'$ at time $i\hbar\beta/2$ and arrive at the point $\mathbf{q}$ at time $-i\hbar\beta/2$. In the quasiclassical regime the integration in (10) is performed by the method of steepest descent, and $\rho(\mathbf{q}, \mathbf{q}')$ takes the form of a sum of Gaussian integrals evaluated over a neighborhood around those paths which make the action extremal, i.e., which satisfy the classical equations of motion. The quasiclassical density matrix is one term of this sum, which is chosen in the following way.

Having placed the points $\mathbf{q}$ and $\mathbf{q}'$ at the bottom of the initial valley, let us take the path $\mathbf{q}(t) = \text{const} = \mathbf{q} = \mathbf{q}'$ with these end points, which clearly satisfies the classical equation of motion. Now we investigate complex-conjugate end points $\mathbf{q}$ and $\mathbf{q}'$ (the saddle point in (9) always corresponds to imaginary ξ); we will study in the integral (10) only that saddle point which lies on the aforementioned path and is the analytic continuation from $\mathbf{q}$ to $\mathbf{q}' = \mathbf{q}^*$. The density matrix so obtained commutes with the Hamiltonian (along with contributions from other saddle points) and coincides with (10) deep in the initial valley where the saddle point we have picked dominates. We can therefore identify it with the quasiclassical density matrix, once we have sorted out any ambiguity in the analytic continuation of the extremal path. For a nonadiabatic transition this ambiguity arises because one has the alternative of circling one of two complex-conjugate branch points of the adiabatic potential $U(\mathbf{q})$; note that both circuits must be taken into account with equal weight in $\tilde{\rho}(\mathbf{q}, \mathbf{q}')$. The ambiguity, which is connected with the possibility of surmounting the barrier by either tunneling through it or being activated over it, is removed by the inclusion of supplementary physical considerations (see section 4). We note that analgous questions must also be resolved in the use of Langer's method.[19]

The expression obtained for $\tilde{\rho}(\mathbf{q}, \mathbf{q}')$ is actually the analytic continuation to imaginary times $t = -i\hbar\beta/2$, $t' = i\hbar\beta/2$ of the quasiclassical Green's function (5):

$$\tilde{\rho}(\mathbf{q}, \mathbf{q}') = G(\mathbf{q}, -i\hbar\beta/2 | \mathbf{q}', i\hbar\beta/2), \tag{11}$$

while the distribution function (9) is its mixed (momentum-coordinate space) representation. As regards $S(\mathbf{q}, \mathbf{q}')$ in (5), we need to substitute the action along the saddle-point path described above, while the constant A is determined by invoking the requirement that the quasiclassical density matrix be normalized.

For calculating the transition probability we need to evaluate the distribution function (9) or the current density

$$\mathbf{J}(\mathbf{q}) = -i\frac{\hbar}{2M}\left(\frac{\partial}{\partial \mathbf{q}} - \frac{\partial}{d\mathbf{q}'}\right)\tilde{\rho}(\mathbf{q}, \mathbf{q}')\,|_{\mathbf{q}=\mathbf{q}'} = \int f(\mathbf{p}, \mathbf{q}) M^{-1}\mathbf{p}\, d^n p \tag{12}$$

in the classically allowed region of final states, i.e., for values of the potential $U(\mathbf{q})$ smaller than the system energy on the saddle-point path. If the distribution over some of the degrees of freedom in the final state regions are of no interest, the coordinates in $\mathbf{q}$ and $\mathbf{q}'$ which correspond to them must be set equal to one another and $\tilde{\rho}(\mathbf{q}, \mathbf{q}')$ must be integrated over these q_j. In $f(\mathbf{p}, \mathbf{q})$, this is accomplished by integrating over q_j and the corresponding momenta p_j. Applying the method of steepest descent to this integration imposes on the extremal path the conditions that the values of the coordinates q_j and velocities $\dot{q}_j$ be the same for $t = i\hbar\beta/2$ and $t = -i\hbar\beta/2$; the resulting density matrix and distribution function contain complete information about the remaining degrees of freedom.

The inverse lifetime of the system in the initial valley equals the integral of the current density (12) in the vicinity of this valley over an $(n-1)$-dimensional surface. Evaluating this integral by saddle-point methods, we obtain a classical path with period $-i\hbar\beta$ similar to the one which appears in Langer's method. A general demonstration of the precise equivalence of the expression for τ^{-1} which we have obtained and expressions obtained by other methods has not yet been found.

In the one-dimensional case, from (12), (11), and (5) we obtain the expression

$$\tau^{-1} = \left(\frac{2}{\pi\hbar}\right)^{1/2} \operatorname{sh}\left(\frac{\hbar\Omega_1\beta}{2}\right)\frac{\partial S(q, q')}{M\partial q} \times\left[\frac{\partial^2 S(q, q')}{\partial q\,\partial q'}\right]^{1/2} \exp[i\hbar S(q, q)], \tag{13}$$

where $\Omega_1 \equiv (M^{-1}U'')^{1/2}$ is the frequency of small oscillations around the bottom of the initial valley, from which the energy is measured. In the classically-allowed region $S(q, q)$ does not depend on q and is equal to the action $S(\beta)$ computed along a periodic path with period $-i\hbar\beta$. The coefficient of the exponential is transformed by using the relation

$$M^{-1}\frac{\partial S}{\partial q'}\frac{\partial^2 S}{\partial q\,\partial q'} = \frac{\partial^2 S}{\partial t\,\partial q} = -\left(M^{-1}\frac{\partial S}{\partial q}\right)^{-1}\frac{\partial^2 S}{\partial t^2},$$

where $S \equiv S(q, q', t)$ is the action (6) along a path which carries the particle from point q' to point q with a time t. The result (13) can be cast in the form

$$\tau^{-1} = \left(\frac{2}{\pi\hbar^3}\right)^{1/2} \operatorname{sh}\left(\frac{\hbar\Omega_1\beta}{2}\right)\left[\frac{\partial^2 S(\beta)}{\partial\beta^2}\right]^{1/2} \exp\left[\frac{i}{\hbar}S(\beta)\right]; \tag{14}$$

for a nonadiabatic transition it has an additional coefficient 2 [compare with (7)]. This expression is obtained in Ref. 19 by Langer's method and coincides with the integral

$$\tau^{-1} = (\pi\hbar)^{-1} \operatorname{sh}\left(\frac{\hbar\Omega_1\beta}{2}\right)\int_0^\infty \exp(-\beta E) D(E)\, dE, \tag{15}$$

where $D(E)$ is the tunneling or nonadiabatic transmission coefficient (7).

In order to apply the approach developed so far to a practical calculation of the probability for a nonadiabatic transition, we must first integrate the classical equations of motion, and for complex initial conditions to boot. For a realistic model of an impurity center this can only be done numerically. A considerably simpler situation obtains in a model with one "lattice" degree of freedom; this will also be the case in models which are reducible to one-dimensional problems by separation of variables, since expressions (14) or (15) can be used.

As an illustration we investigate the one-dimensional "spin-oscillator" model[20] with a two-level electron Hamiltonian (see Ref. 1).

$$\hat{H}_0(q) = {}^1/_2 M\omega^2 q^2 + {}^1/_2 E_0\hat{\sigma}_z + {}^1/_2(B_x\hat{\sigma}_x + B_y\hat{\sigma}_y + B_z\hat{\sigma}_z)\,q,$$

where $\hat{\sigma}_x$, $\hat{\sigma}_y$, $\hat{\sigma}_z$ are Pauli matrices, ω, E_0, B_x, B_y, B_z are

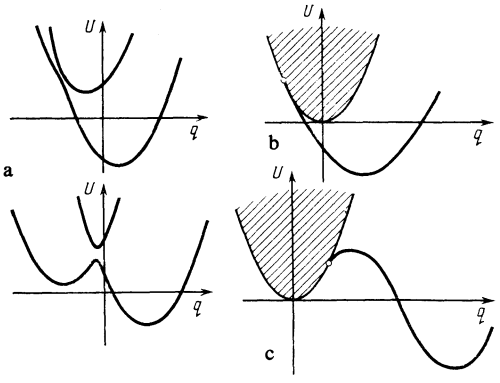

FIG. 1. Energy schemes for various cases of multiphonon nonradiative transitions: a) Two-level model for an impurity center; b) single-level impurity center with a continuous (free-carrier) spectrum (shaded area). Boldface curve: the adiabatic potential of the bound state; c) barrier autolocalization, or an impurity center with a level which appears in the presence of a lattice deformation. At the point where the bound state appears, the slopes of the adiabatic and intrinsic lattice potentials coincide.

parameters. The adiabatic potential energy

$$U(q)={}^1/_2M\omega^2q^2\pm{}^1/_2[(E_0+B_zq)^2+(B_x^2+B_y^2)q^2]^{1/2}$$

for small interaction constant $\alpha\equiv(B_x^2+B_y^2)^{1/2}/B_z\ll 1$ consists of two parabolic terms with equal oscillation frequencies ω, small in keeping with the requirements of the adiabatic approximation. The potential minima are found at the points $\pm B_z/2M\omega^2$; depending on the value of the parameter

$$\gamma\equiv 2M\omega^2E_0/B_z^2,$$

these are located on one (for $\gamma>1$) or on both (for $\gamma<1$) sides of a narrow region around the quasi-intersection $q\approx -E_0/B_z$ (see Fig. 1a). In the case $\gamma>1$, the transmission coefficient (7) from one term to the other for energies lower than the intersection region equals

$$D(E)=2\exp\left\{-\frac{2E_0}{\hbar\omega}\left[\left(\frac{(\gamma+1)^2}{4\gamma}+\frac{E}{E_0}\right)\operatorname{Arth}\left(-\frac{4\gamma E}{(\gamma+1)^2E_0}\right)^{1/2}-\left(\frac{(\gamma-1)^2}{4\gamma}+\frac{E}{E_0}\right)\operatorname{Arth}\left(-\frac{4\gamma E}{(\gamma-1)^2E_0}\right)^{1/2}-\left(-\frac{E}{\gamma E_0}\right)^{1/2}\right]\right\}$$

and does not depend on the interaction constant α. In this expression, the energy is measured from the quasi-intersection point, and before it is substituted into (15) one must first lower the energy origin until it coincides with the bottom of the upper electronic valley, i.e., at the energy $E_0(2\gamma-1)/4\gamma$. Then we obtain for the probability of a nonadiabatic transition from the upper term to the lower

$$\tau^{-1}=\left(\frac{2\omega E_0}{\pi\hbar\gamma}\right)^{1/2}\frac{f}{f+2N+1}\exp\left\{-\frac{2E_0}{\hbar\omega}\left[\operatorname{Arth}\left(\frac{f}{\gamma+1}\right)-\frac{f}{2\gamma}\right]\right\},$$

$$f\equiv[\gamma^2+4N(N+1)]^{1/2}-(2N+1),\quad N\equiv(e^{\hbar\omega\beta}-1)^{-1},\quad (16)$$

where N is the equilibrium occupation number of the lattice oscillator. The inverse transition probability differs from (16) by a factor of $\exp(-\beta E_0)$.

The region in which expression (16) is applicable is determined by two conditions: first, that the separation of the terms in the quasintersection regime, which equals αE_0, must not be so small that the quasiclassical approximation is violated. Second, the energy on the extremal path must be lower than the quasiintersection energy range. These conditions can be formulated quantitatively on the basis of an accurate (nonquasiclassical) investigation (Ref. 21, Ref. 4 ch. 5) of the question of linear quasiintersecting terms; they take the form of inequalities

$$\alpha^2\gamma E_0/\hbar\omega\ll f\ll(\alpha\gamma)^{1/2}(\gamma^2-1)^{1/4}.$$

When the first inequality is violated, the quasiclassical condition is also and we must turn to perturbation theory in the interaction constant α. When the second inequality is violated, we must make use of the exact expression for the transmission coefficient $D(E)$,[21] or for the inverse (strong) inequality use the Landau-Zener formula. In an analogous way, we must investigate the transition between terms for $\gamma<1$. In this case, expression (16) (decreased by half) gives the tunneling transition probability between valleys of the lower term.

Since the terms are strictly parabolic in the model we have just investigated, perturbation theory in the nonadiabaticity operator using an oscillator expansion around any other potential minimum must likewise yield exponential accuracy in a calculation of τ^{-1} (see the introduction). When all the oscillators have equal frequencies, one can (after a detailed investigation) recover the results of Huang and Rhys[22]; however, due to the cumbersomeness of the expressions which result, it is possible to match the exponents in our results with those of Huang and Rhys only asymptotically. Specifically, the high-temperature asymptotic behavior (activated) and zero-temperature limit [see, e.g., formulae (54) and (47) in Ref. 23] of their exponents agree with the corresponding forms derivable from (16). A comparison of the coefficients of the exponentials is meaningless for the reasons discussed in the introduction.

3. CAPTURE

The capture of current carriers on deep traps plays an important role in semiconductor recombination processes (see Ref. 2). A free carrier can fall into a localized trap state whose energy lies in the forbidden gap at a depth which significantly exceeds the limiting phonon energy either by descending a staircase of excited states of the trap ("cascade" capture),[2)] by radiative or Auger processes (which are not discussed here) or by means of multiple phonon emission. In the last case, especially for deep neutral or repulsive traps, the transition is essentially nonadiabatic and can be studied by the approach developed in this paper. The principle difference between these transitions and intracenter transitions lies in the fact that the electronic subsystem, in addition to its discrete spectrum (in the simplest case, a single level), also has a continuous spectrum for which the adiabatic approximation is certainly inapplicable. This same difficulty also appears when one investigates the ejection of a particle from a potential well by a slow external perturbation[24,25]; a well-known method of overcoming it is to describe the free-particle motion quasiclassically, taking into account the influence of the well via boundary conditions on the wave function (see, e.g., Ref. 18, chapter 5, §4).

The approach we will develop below for analyzing cap-

ture on an impurity center makes use of the quasiclassical nature both of the lattice motion, which is based on the adiabatic approximation, and of the free-electron motion. The latter has its origin not in adiabaticity, but simply in the fact that phonons have a rather weak influence on band electron states. The calculations in this and the following sections yield exponential accuracy; this is because they can also be obtained within the framework of adiabatic perturbation theory.

To demonstrate the approach, let us first investigate the capture process for the simplest one-dimensional model of an impurity center, after which it will become clear how to proceed in the general case. The Hamiltonian of a captured particle [see (1)] we take in the form

$$\hat{H}_0(q)=\frac{\hbar^2}{m}\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}-\varkappa(q)\delta(x)\right]+U_0(q), \qquad (17)$$

where m is the particle mass, x is its coordinate. The coefficient $\varkappa(q)$, which has the dimensions of inverse length, determines the dependence of the deep δ-function potential well on the normalized lattice coordinate q, while $U_0(q)$ represents the intrinsic potential energy of the lattice. Corresponding to this latter degree of freedom is a mass which we denote by M. The curve $U_0(q)$ and the adiabatic potential energy

$$U(q)=U_0(q)-\hbar^2\varkappa^2(q)/2m$$

are shown schematically in Fig. 1b.

Because it is easier to calculate, we will first investigate the inverse of the capture process, which is the ionization of a center. This may be regarded as the decay of a bound state due to thermodynamic fluctuations. Once this problem is solved, the capture rate and its dependence on particle energy can be found from the principle of detailed balance. For the reasons described above, we cannot limit ourselves to a simple calculation of the adiabatic binding energy $-\hbar^2\varkappa^2(q)/2m$ of a particle moving in the lattice potential. Therefore the quasi-equilibrium density matrix describing the ionization must be determined from the two-dimensional integral (10) along the path $[x(t), q(t)]$ having the specific values x, q at the time $t=-i\hbar\beta/2$ and x', q' at the time $t'=i\hbar\beta/2$, with the action

$$S[x(t),q(t)]=\int_{i\hbar\beta/2}^{-i\hbar\beta/2}\left[\frac{m\dot{x}^2}{2}-\frac{\hbar^2\varkappa(q)}{m}\delta(x)+\frac{M\dot{q}^2}{2}-U_0(q)\right]dt.$$

In keeping with the assumption of "slowness" of the phonon coordinate, which, let us say, is a consequence of the smallness of the lattice zero-point energy compared to the particle binding energy, it is natural to integrate (10) first over the "rapid" degree of freedom $x(t)$, and treat the "slow" path $q(t)$ as given. Then it is simple to perform the functional integration, yielding a Schrödinger-representation density matrix in the form

$$\rho(x,q|x',q')=\int G\left(x,-\frac{i\hbar\beta}{2}\middle|\,x',\frac{i\hbar\beta}{2}\right)\times\exp\left\{\frac{i}{\hbar}\int_{i\hbar\beta/2}^{-i\hbar\beta/2}\left[\frac{1}{2}M\dot{q}^2(t)-U_0(q(t))\right]dt\right\}\mathcal{D}[q(t)], \qquad (18)$$

where $G(x,t|x',t')$ is the time-dependent Green's function for the Schrödinger equation with the Hamiltonian (17); note that G depends parametrically on the lattice path and the supplementary condition for $t\neq t'$:

$$\frac{\partial G(x,t|x',t')}{\partial t}=-i\frac{\hbar^2}{m}\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}-\varkappa(q)\delta(x)\right]G(x,t|x',t'),$$
$$\frac{\partial G(x,t|x',t')}{\partial t'}=i\frac{\hbar^2}{m}\left[-\frac{1}{2}\frac{\partial^2}{\partial x'^2}-\varkappa(q)\delta(x')\right]G(x,t|x',t'). \qquad (19)$$

Since for all $x\neq 0$ the particle motion is quasiclassical, we can look for a solution to (19) in the form

$$G(x,\,t|x',\,t')\sim\exp[i\hbar^{-1}S(x,\,t|x',\,t')]. \qquad (20)$$

Substituting this into (19) gives an equation of Hamilton-Jacobi type for $S(x,t|x',t')$, analogous to the treatment in Ref. 9, whose solution (see, e.g., Ref. 26) in turn leads to the expression[3)]

$$S(x,t|x',t')=\frac{mx^2}{2(t-t_1)}-\frac{mx'^2}{2(t'-t_1')}+Y(t_1|t_1'), \qquad (21)$$

and where the times t_1 and t_1' are found from the expressions

$$\frac{mx^2}{2(t-t_1)^2}+\frac{\partial Y(t_1|t_1')}{\partial t_1}=0,$$
$$\frac{mx'^2}{2(t'-t_1')^2}-\frac{\partial Y(t_1|t_1')}{\partial t_1'}=0. \qquad (22)$$

The function $Y(t_1|t_1')$, which for a general integral of the Hamilton-Jacobi equation is arbitrary, is here determined by the boundary conditions imposed on the Green's function for the δ-function potential well:

$$\lim_{x\to\pm 0}\frac{\partial G(x,t|x',t')}{\partial x}=\mp\varkappa(q(t))G(0,t|x',t'),$$
$$\lim_{x'\to\pm 0}\frac{\partial G(x,t|x',t')}{\partial x'}=\mp\varkappa(q(t'))G(x,t|0,t'). \qquad (23)$$

Having noted the obvious symmetry of the problem under a sign change of either x or x', we will henceforth assume that x and x' are positive. Substituting (20) into the boundary conditions (23) gives the following conditions for $S(x,t|x',t')$:

$$\left.\frac{\partial S(x,t|x',t')}{\partial x}\right|_{x=0}=i\hbar\varkappa(q(t)),$$
$$\left.\frac{\partial S(x,t|x',t')}{\partial x'}\right|_{x'=0}=i\hbar\varkappa(q(t')).$$

Substituting (16) into these conditions, we obtain

$$\lim_{x\to 0}\frac{mx}{t-t_1}=i\hbar\varkappa(q(t)), \qquad \lim_{x'\to 0}\frac{mx'}{t'-t_1'}=-i\hbar\varkappa(q(t')).$$

By studying (22) in the limit $x\to 0, x'\to 0$ (in this case, obviously, $t\to t_1$, $t'\to t_1'$), we finally obtain an equation for $Y(t_1|t_1')$:

$$\frac{\partial Y(t_1|t_1')}{\partial t_1}=\frac{\hbar^2\varkappa^2(q(t_1))}{2m}, \qquad \frac{\partial Y(t_1|t_1')}{\partial t_1'}=-\frac{\hbar^2\varkappa^2(q(t_1))}{2m},$$

from which

$$Y(t_1|t_1') = \int_{t_1'}^{t_1} \frac{\hbar^2 \varkappa^2(q(t))}{2m} dt.$$

Now, substituting the resulting expression for the Green's function (20) into (18) and ignoring the weak dependence of the coefficient of the exponential in (20) (which in this work is not calculated) on x, t, x', t' and on the path $q(t)$, we are led to the one-dimensional path integral

$$\tilde{\rho}(x, q|x', q') \propto \int \exp\left\{\frac{i}{\hbar} S[q(t)]\right\} \mathcal{D}[q(t)] \qquad (24)$$

with the effective action

$$S[q(t)] = \frac{mx^2}{2(-i\hbar\beta/2 - t_1)} - \frac{mx'^2}{2(i\hbar\beta/2 - t_1')} + \int_{t_1'}^{t_1} \frac{\hbar^2 \varkappa^2(q)}{2m} dt + \int_{i\hbar\beta/2}^{-i\hbar\beta/2} \left[\frac{M\dot{q}^2}{2} - U_0(q)\right] dt \qquad (25)$$

and the following equations for t_1 and t_1':

$$\frac{mx}{-i\hbar\beta/2 - t_1} = i\hbar\varkappa(q(t_1)), \quad \frac{mx'}{i\hbar\beta/2 - t_1'} = -i\hbar\varkappa(q(t_1')). \qquad (26)$$

The physical meaning of this effective action is obvious: the first two terms in (25) are the action accumulated by the particle due to its free motion from x' to 0 and from 0 to x; the third term is due to the bound motion of the particle, and equals the time integral of the binding energy of the particle in the well.

The integration over the paths $q(t)$ can be performed by a simple saddle-point technique, since the lattice motion is quasiclassical for all times. The path through the saddle-point satisfies an equation of motion which corresponds to free motion of the particle on the time intervals $(i\hbar\beta/2, t_1')$ and $(t_1, -i\hbar\beta/2)$, and bound motion on the connecting interval (t_1', t_1). The equations of motion in these regions contain the unperturbed (i.e., no particle present) and adiabatic lattice potential energies, respectively:

$$M\ddot{q} = -\frac{d}{dq} U_0(q), \quad M\ddot{q} = -\frac{d}{dq}\left[U_0(q) - \frac{\hbar^2 \varkappa^2(q)}{2m}\right]. \qquad (27)$$

Variation of the action $S[q(t)]$ at t_1 and t_1' while taking into account (26) yields the condition of conservation of the velocity $\dot{q}$ for a transition from one portion of the path to another, i.e., at the connecting times t_1 and t_1'.

The conditions we have formulated here determine completely the classical path $q(t)$ and the extremal action (25) for given end points x, q, and x', q'. The probability distribution of final values of the lattice coordinate is usually of no interest; integrating (24) over these final values $q = q'$ yields the coincidence conditions on q and $\dot{q}$ at the times $-i\hbar\beta/2$ and $i\hbar\beta/2$. Later on, since we are interested only in the distribution of escaping particles with momentum p, we will transform from the density matrix $\tilde{\rho}(x|x')$ (the arguments q and q' have been integrated out) to the Wigner distribution function in accordance with the recipe (9):

$$f(p, x) \equiv \frac{1}{2\pi\hbar} \int_{-\infty}^{+\infty} \exp\left(-\frac{i}{\hbar} p\xi\right) \tilde{\rho}\left(x + \frac{\xi}{2}, x - \frac{\xi}{2}\right) d\xi, \qquad (28)$$

where once again the integral can be evaluated by the saddle-point method.

Here it must be recalled that the energy origin was chosen to be the energy of the unperturbed lattice plus the energy of a stationary free particle. This implies that in order to obtain the true momentum distribution function for a particle escaping from the center by ionization, expression (28) must still be multiplied by the Gibbs exponent $\exp(\beta U_{\min})$, where $U_{\min} < 0$ is the minimum of the adiabatic potential $U(q)$. However, in the final analysis we are interested in the capture cross-section of a particle with momentum p, which is not strongly dependent on p, and which is determined from the equation of detailed balance; to exponential accuracy this equals

$$\sigma(p) \propto \exp(\beta p^2/2m) f(p),$$

where $f(p)$ is a distribution function (28), independent of x.

Omitting simple operations we write down the final exponential expression for $\sigma(p)$ and the conditions which determine the extremizing trajectory and the argument of the exponential:

$$\sigma(p) \propto \exp[i\hbar^{-1} S(\beta, p)], \qquad (29)$$

$$S(\beta, p) = \int_{-i\theta}^{-i\hbar\beta/2} \left[\frac{1}{2} M\dot{q}^2 - U_0(q)\right] dt + \int_{i\hbar\beta/2}^{i\theta} \left[\frac{1}{2} M\dot{q}^2 - U_0(q)\right] dt + \int_{+i\theta}^{-i\theta} \left[\frac{1}{2} M\dot{q}^2 - U_0(q) + \frac{1}{2m}\hbar^2\varkappa^2(q)\right] dt - \frac{i\theta p^2}{m}, \qquad (30)$$

$$q(-i\hbar\beta/2) = q(i\hbar\beta/2), \quad \dot{q}(-i\hbar\beta/2) = \dot{q}(i\hbar\beta/2), \qquad (31)$$

$$\varkappa(q(-i\theta)) = -i\hbar^{-1}p, \quad \varkappa(q(i\theta)) = i\hbar^{-1}p.$$

The equations of motion (27) together with conditions (31) and the requirement of continuity of $q(t)$ entirely determine the path $q(t)$ and the connecting times $t_1 = -i\theta$ and $t_1' = i\theta$, which can be chosen pure imaginary because of the possibility of arbitrarily shifting the real part without affecting the value of the action.

Let us turn to a specific calculation based on the model described above, considering the lattice as a harmonic oscillator:

$$U_0(q) = M\omega_0^2 q^2/2 \qquad (32)$$

and assume that the dependence $\varkappa(q)$ is linear

$$\varkappa(q) = \varkappa_0 + \varkappa_1 q, \quad \varkappa_1 > 0 \qquad (33)$$

(the interaction constant is chosen positive for definiteness). To complete the final specifications of the system, the bound-state potential is also taken to be an oscillator

$$U(q) = -U^* + {}^1/_2 M\omega^2 (q - Q)^2, \qquad (34)$$

$$U^* \equiv \frac{\hbar^2 \varkappa_0^2 \omega_0^2}{2m\omega^2} > 0, \quad \omega \equiv \left(\omega_0^2 - \frac{\hbar^2 \varkappa_1^2}{mM}\right)^{1/2}, \quad Q \equiv \frac{\hbar^2 \varkappa_0 \varkappa_1}{mM\omega^2},$$

but with a different frequency and a shift in equilibrium posi-

tion. The equation of motion (27) gives a sinusoidal path

$$q(t)=C_0\cos\left[\omega_0(t-\zeta_0\pm i\hbar\beta/2)\right] \tag{35}$$

on the free segment of the motion from $\mp i\theta$ to $\mp i\hbar\beta/2$ and

$$q(t)=Q+C\cos\left[\omega(t-\zeta)\right] \tag{36}$$

on the bound segment. The parameters C_0, ζ_0, C, ζ are all real, due to the symmetry of the path relative to the real axis and the coincidence of q and $\dot q$ for $t=\pm i\hbar\beta/2$.

The final manipulations consist of substituting the path obtained above into the equation

$$\varkappa_0+\varkappa_1 q(-i\theta)=-i\hbar^{-1}p \tag{37}$$

[see (26)] and into the continuity conditions for $\dot q$ at $t=-i\theta$; this leads to a system of equations

$$\begin{aligned}
&C_0\cos(\omega_0\zeta_0)\,\mathrm{ch}\left[\omega_0(^1/_2\hbar\beta-\theta)\right]=-\varkappa_0/\varkappa_1,\\
&C_0\sin(\omega_0\zeta_0)\,\mathrm{sh}\left[\omega_0(^1/_2\hbar\beta-\theta)\right]=-p/\hbar\varkappa_1,\\
&C\cos(\omega\zeta)\,\mathrm{ch}(\omega\theta)=-\varkappa_0\omega_0^2/\varkappa_1\omega^2,\\
&C\sin(\omega\zeta)\,\mathrm{sh}(\omega\theta)=p/\hbar\varkappa_1,\\
&\omega_0C_0\sin(\omega_0\zeta_0)\,\mathrm{ch}\left[\omega_0(^1/_2\hbar\beta-\theta)\right]=\omega C\sin(\omega\zeta)\,\mathrm{ch}(\omega\theta),\\
&\omega_0C_0\cos(\omega_0\zeta_0)\,\mathrm{sh}\left[\omega_0(^1/_2\hbar\beta-\theta)\right]=-\omega C\cos(\omega\zeta)\,\mathrm{sh}(\omega\theta)
\end{aligned} \tag{38}$$

for determining the parameters C_0, ζ_0, C, ζ, and θ (although there are more equations than unknowns, the system still has a solution).

Solving the system of equations (38) and then calculating the action (30) gives the following expression for the capture cross section of a particle:

$$\sigma(p)\propto\exp\left\{\frac{2U^*}{\hbar}\left[\theta-\frac{1}{\omega}\mathrm{th}(\omega\theta)\right]+\frac{p^2}{\hbar m}\theta\right\}, \tag{39}$$

in which out of all the parameters alluded to before only θ remains, determined by the transcendental equation

$$\omega^{-1}\,\mathrm{th}(\omega\theta)=\omega_0^{-1}\,\mathrm{th}\left[\omega_0(\theta-^1/_2\hbar\beta)\right]. \tag{40}$$

We note that θ appears to be negative (for $\omega<\omega_0$), and does not depend on the particle momentum p; this latter property is evidently a characteristic of the specific model under study. The total capture cross section for a thermalized particle, i.e., $\sigma(p)$ integrated over p with a Maxwell distribution, is equal to $\sigma(0)$ to exponential accuracy. Of course, the characteristic momentum of a captured particle is not strictly zero, but is determined by (among other things) how the coefficient of the exponential depends on p.

In Fig. 2 we show schematically a path $q(t)$ for $p=0$ on the imaginary time axis (for $p>0$ the path is complex and therefore difficult to represent graphically). The bound state motion is found by time inversion with respect to the free motion, corresponding to the negative sign of θ.

Expressions (39) and (40) completely describe the capture process for a free particle by a one-dimensional δ-function well in a model which consists of one lattice oscillator and the interaction (28) which is linear in q. Let us proceed to asymptotics, which we will obtain in various limiting cases. In the low-temperature limit $\beta\to\infty$ we have $\theta\to-\omega^{-1}\,\mathrm{Arth}(\omega/\omega_0)$ and the capture cross section approaches

$$\sigma(p)\propto\exp\left\{\frac{2U^*}{\hbar\omega_0}-\frac{2}{\hbar\omega}\left[U^*+\frac{p^2}{2m}\right]\mathrm{Arth}\frac{\omega}{\omega_0}\right\}.$$

In the high-temperature limit $\beta\to0$ (restricted, of course, by the requirement of that the exponential be small, we obtain

$$\sigma(p)\propto\exp\left(-\beta E_A+\theta p^2/m\right),\quad \theta=-(3\beta mM/2\hbar\varkappa_1^2)^{1/3},$$
$$E_A=M\omega_0^2\varkappa_0^2/2\varkappa_1^2.$$

The total capture cross section for thermal particles, given by $\sigma(0)$, depends on temperature in this limit in an activated fashion, where the activation energy E_A is equal to the oscillator energy at the point $q=-\varkappa_0/\varkappa_1$, which is where the binding energy $\hbar^2\varkappa^2(q)/2m$ reduces to zero.

It is also interesting to consider the limit of small $\varkappa_1$ corresponding to weak electron-phonon interaction. In this limit ω_0 and ω approach each other and θ diverges logarithmically:

$$\theta=-(2\omega_0)^{-1}\ln\left[4mM\omega_0^2/\hbar^2\varkappa_1^2(N+1)\right].$$

the expression for the capture cross-section in this case

$$\sigma(p)\propto\left[\frac{\hbar^2\varkappa_1^2(N+1)}{4mM\omega_0^2}\right]^n\exp\left(-\frac{U^*}{\hbar\omega_0}\right),\quad n\equiv\frac{1}{\hbar\omega}\left(U^*+\frac{p^2}{2m}\right)$$

evidently agrees with the results of perturbation theory in the constant $\varkappa_1$. The number n is the number of quanta which a particle with momentum p must emit in order to fall into the bound state. Varying the temperature leads to a change in the oscillator occupation number N.

In spite of the extreme simplicity of the one-dimensional model investigated above, the equations we have obtained also have some applicability in the three-dimensional case for capture of a carrier on an uncharged deep trap from a nondegenerate, not-too-narrow band. Let the carrier (an electron for definiteness) have an isotropic effective mass m; its Hamiltonian clearly depends on the whole ensemble of lattice coordinates q_i. The problem of calculating the quasi-equilibrium density matrix is solved, as before, by first integrating over the electron path, and then over the lattice path. Replacing the first integration by a solution of the Schrödinger equation, we observe that near the connection times the binding energy of an electron on the center is close to zero, and the electron is found with overwhelming probability in the free-particle region (far from the center the lattice is only weakly perturbed). Such weak-coupling states are

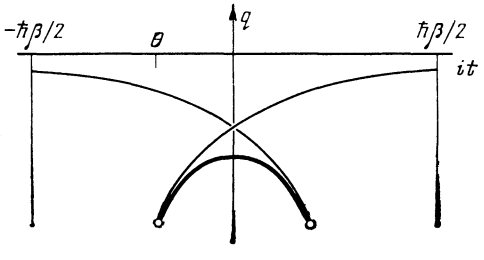

FIG. 2. A characteristic path $q(t)$ which describes the ionization of an impurity center. Boldface curve: the bound-motion segment. The fact that the connection time θ is negative is due to the "overhang" of the barrier as shown in Fig. 1b).

spherically symmetric, and in the spirit of the zero-radius potential method[27] we can in the usual way reduce the three-dimensional Schrödinger equation to a one-dimensional one. The particular characteristics of the center are here contained in the inverse scattering length $\varkappa$, which is defined in terms of a boundary condition for the logarithmic derivative of solutions of the one-dimensional Schrödinger equation in the external region (Ref. 6, §133). This boundary condition (23) has precisely the same form as condition (23), which arose from the δ-function potential in the one-dimensional problem; therefore the integration along the electron path leads once again to expression (24) with the action (25). For q we must substitute the ensemble of all the lattice coordinates, while for $\varkappa(q)$ we need the inverse scattering length for each of these q; for $U_0(q)$ we use the elastic lattice energy.

Here we must recall that, generally speaking, the center can be described by a scattering length only near the capture connecting times, while for other times (i.e., in the bound-state segment of the path) the state must be strongly localized. It is easy to see, however, that in the expression for the action (25), $\varkappa(\mathbf{q})$ enters in only in combinations which involve the well binding energy of the particle. It is therefore clear that in the general case the real binding energy on the center (which depends on $\mathbf{q}$) should appear in (25) in place of $\hbar^2\varkappa^2(\mathbf{q})/2m$. In Eq. (26), which determines the connecting times, the real inverse scattering length $\varkappa(\mathbf{q})$ appears, which can be expressed in terms of the binding energy in view of the one-to-one correspondence between them in this region.

Integrating along the lattice path and then transforming leads to precisely the same result as for the case of a single variable $q(t)$. As a result it is found that the scattering cross section $\sigma(p)$ for a carrier with momentum p on a deep uncharged impurity center is given by expression (29) with the action (30), and by the conditions (31) on the path, where for q we substitute the ensemble of lattice coordinates $\mathbf{q}$; $\varkappa(\mathbf{q})$ is connected with the impurity level energy by the relation

$$\varkappa(\mathbf{q})\equiv\hbar^{-1}[-2mE(\mathbf{q})]^{1/2}, \tag{41}$$

where the branch is chosen to be positive for negative $E(\mathbf{q})$. The lattice path $\mathbf{q}(t)$ consists of two free segments and an intermediate bound segment; the equation of motion on the bound segment has the corresponding form

$$M\ddot{q}_i=-\frac{\partial}{\partial q_i}U_0(\mathbf{q}), \qquad M\ddot{q}_i=-\frac{\partial}{\partial q_i}[U_0(\mathbf{q})+E(\mathbf{q})]. \tag{42}$$

The total path is determined by the continuity condition for $\dot{\mathbf{q}}(t)$ and by Eq. (42), in full analogy to the single-oscillator case. The capture cross section for thermal carriers $\sigma\sim\sigma(0)$, analogous to the one-dimensional model, is determined by the action along the path $\mathbf{q}(t)$, into which the impurity level energy is substituted up to the region of the continuous spectrum at the connecting times $\pm i\theta$.[4)]

The solution to the equations so obtained in the case of more or less realistic models describing capture is accompanied by difficulties in calculation, and it is not possible to describe them here. Further computation using the analytic one-dimensional, single-oscillator model apparently yields a qualitatively accurate picture of capture on a neutral trap with the assistance of phonons of various types; for a local mode the model can approach quantitative accuracy. Evidently one can discern a general regularity in the growth of the scattering cross section with temperature from the result (in agreement with Ref. 28) that the high-temperature law is an activated one. The activation energy for thermal carriers equals the minimum energy of the lattice configuration which ejects the impurity level into the band. Since the adiabatic energy in the bound state $U_0(\mathbf{q})+E(\mathbf{q})$ has no saddle points (see Fig. 1b) which could result in the appearance of an activation path in the quasiequilibrium density matrix, one arrives at an activation law only through a sequence of several steps. The inverse case, which occurs in barrier autolocalization, is investigated in the next section. A less trivial general property of capture processes is the decrease of $\sigma(p)$ with increasing carrier momentum, connected with θ being negative, i.e., with the presence of retrograde motion along the extremal path.

The calculation scheme we propose can be directly generalized to the more complicated cases of anisotropic carrier dispersion laws and multilevel traps. The case of an impurity which has an energy level which is delocalized in the undeformed lattice but becomes localized due to the electron-phonon interaction (see, e.g., Ref. 29) is analogous to autolocalization, which we will investigate in the next section. To investigate capture on a repulsive center we must take into account the Coulomb potential in the external region of quasiclassical particle motion. There is no obstacle in principle to pursuing our approach for the analogous calculation involving two bands; generally speaking, such a calculation is a necessary preliminary to any investigation of carrier recombination in real semiconductors.

4. AUTOLOCALIZATION

Barrier autolocalization of charge carriers and excitons in three-dimensional crystals can also be viewed as a multiphonon-mediated nonradiative capture process (see Refs. 30, 11–13); however, the carrier is not localized on an already present impurity level, but rather in a potential well generated by a concomitant lattice deformation. The existence of self-consistent (autolocalized) states in which the binding energy of a particle in the well exceeds the elastic lattice energy requires a rather strong electron-phonon interaction. An absolute minimum in the adiabatic potential energy $U(\mathbf{q})$ is responsible for this state; a path to it from the undeformed lattice-plus-free-particle state for a short-range electron-lattice interaction goes through energetically unfavorable configurations where a bound state in the well is absent or its binding energy is small (see Fig. 1c). As a result, it is found that the adiabatic lattice energy $U(\mathbf{q})$ has a saddle of height W with a maximum along one principle axis (in $\mathbf{q}$-space) and a minimum along all others except those which are responsible for translation of the well as a whole. The multidimensional potential barrier obtained in this way must be surmounted either by activation or tunneling, which is the topic of this discussion. The question of what degree of influence such barriers have on the rate of autolocalization

in real crystals has not yet been resolved. Experiments, which are described in work which we will cite later on, imply that such barrier heights are completely insignificant.

From the point of view of calculating transition rates, the autolocalization process does not differ in principle from the case of capture on an impurity center which we investigated in the previous section. The lattice equations of motion (42), the connection between the energy of a bound state in the well and the inverse scattering length (41), the formal expression for the action (29) and conditions (30) which determine the connection times, all retain their validity. There are, however, important differences due to the presence of the potential barrier. These differences are expressed in the form of boundary conditions imposed on the lattice path, which lead to the appearance of various types of extrema as mentioned in section 2. The autolocalization rate is determined, depending on the energy band of the captured particles and on temperature, by one or another of these paths.

In view of the impossibility of doing any analytic calculations on realistic models, we turn as in the previous section to the one-dimensionl single-oscillator models (17), (32), and (33), which allow us to demonstrate the qualitative features of more complicated cases. A level in the undeformed lattice will be absent if the condition $\varkappa_0 < 0$ in (33) holds, while an autolocalization barrier is present if

$$\hbar^2\varkappa_1^2/mM > \omega_0^2,$$

so that the system in its bound state is an *inverted* oscillator [see (34)][5]:

$$U(q) = W - {}^1/_2 M\Omega^2 (q-Q)^2, \qquad (43)$$

$$W \equiv \frac{\hbar^2\varkappa_0^2\omega_0^2}{2m\Omega^2} > 0, \quad \Omega \equiv \left(\frac{\hbar^2\varkappa_1^2}{mM} - \omega_0^2\right)^{1/2}, \quad Q \equiv -\frac{\hbar^2\varkappa_0\varkappa_1}{mM\Omega^2} > 0$$

with an imaginary oscillation frequency $\omega = i\Omega$.

As with the case of capture on an impurity center, the "free-particle" motion of the lattice is given by a path of the form (35), while the form of the bound-motion path depends on the relation between the total energy and the barrier heights. The path for the below-barrier case is of the form

$$q(t) = Q + C\,\mathrm{ch}[\Omega(t-\zeta)], \qquad (44)$$

of over-barrier case

$$q(t) = Q - C\,\mathrm{sh}[\Omega(t-\zeta)], \qquad (45)$$

where C and ζ, as before, are the real amplitude and time shift. In the same way as for capture at an impurity center, we will be interested in paths which describe not autolocalization itself but rather the inverse process of fluctuation-induced decay of an autolocalized state. Then we must require that C agree in sign with Q, that is (for $\varkappa_1 > 0$) be positive, so that for negative real times $t < \zeta$ there will be a current which corresponds to escape over the barrier from the region of the autolocalized state. Let us recall that in the previous case we investigated only one type of bound motion, since for a real frequency ω inserted in (35) the cosine is equivalent to a sine with ζ shifted by a quarter period.

Substituting (44) or (45) in place of (36) into equation (37) and into the continuity condition for $\dot{q}(t)$, and then calculating the extremal action, we are led, both for the above-barrier and below-barrier cases, to an exponential expression for the autolocalization probability per unit time $\tau^{-1}(p)$, which coincides with expression (39) for $\sigma(p)$ with $U^* = -W, \omega = i\Omega$. All the subtlety is contained in the fact that Eq. (40) which determines θ now has not one but formally an infinite number of real solutions. The choice of solution is also exactly determined by the character of the motion of the system on the bound segment. The over-the-barrier path (45) carries the particle through the barrier for motion directly along the real time axis. For real time t the below-barrier path describes reflection from the barrier, while the transition through the barrier occurs because of an additional shift along the imaginary axis of $-i\pi/\Omega$. It is therefore clear that the root of Eq. (40) which is smallest in modulus corresponds to the above-barrier case, being negative in sign, while the smallest positive root gives the below-barrier case. The positiveness of the amplitude C in (45) or in (44) yields a condition which defines the regions of applicability of the solution so obtained. As can be shown by investigating systems of equations analogous to (38) on the extremal path, these regions are described by the inequalities

$$p \geqslant -\hbar\varkappa_0\omega_0^2\Omega^{-2}\,\mathrm{tg}(\Omega\theta), \quad p \leqslant -\hbar\varkappa_0\omega_0^2\Omega^{-2}\,\mathrm{tg}(\Omega\theta) \qquad (46)$$

for the above-barrier and below-barrier cases, respectively.

Substituting the values of θ appropriate to each region into (46), it is easy to convince oneself that these regions do not cover all values of the particle momentum p and temperature β^{-1}. In intermediate regions the path which determines the ionization rate does not belong to one of the types (44), (45) which make the action an absolute extremum, but rather can only be a conditionally extremal path. The physical constraint which determines this path is provided by equating the total system energy to the barrier height. On the bound segment, such a path cannot be identified with a unique analytic function, but has the form

$$q(t) = Q + C\exp[\Omega(t \pm i\theta_0)] \qquad (47)$$

(C and θ_0 real) which describes a system motion which crosses the barrier with zero velocity at time $t = -\infty - i\theta_0$. The path parameters are determined as before by matching at the connection times with the free-motion path (35) and using Eq. (31); specifically,

$$\theta_0 = \frac{\hbar}{2}[\beta - \beta_0(p)], \quad \beta_0(p) \equiv \frac{2}{\hbar}\left[\frac{1}{\omega_0}\,\mathrm{Arth}\left(\frac{p\Omega}{\hbar\varkappa_0\omega_0}\right) - \frac{1}{\Omega}\,\mathrm{Arctg}\left(\frac{p\Omega^2}{\hbar\varkappa_0\omega_0^2}\right)\right] \geqslant 0.$$

For a given particle energy $p^2/2m$ (smaller than the barrier height W) the transition from one type of bound-motion path to another is implemented as follows: for high temperatures the extremal path corresponds to motion above the barrier [see (45)]. As the temperature is lowered to a value $\beta_0^{-1}(p)$, the total energy on the extremal path decreases until it reaches the barrier height; from then on it remains constant [the path has the form (47)] right up to

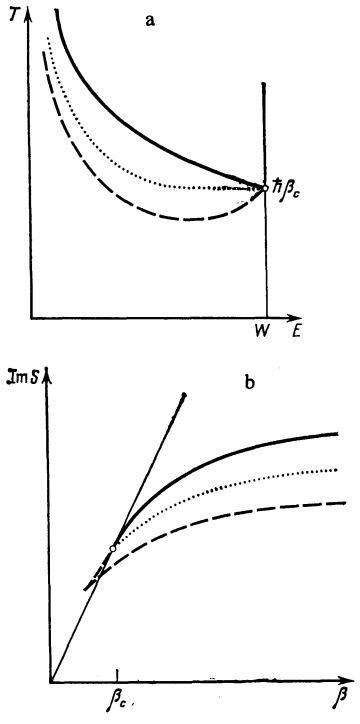

FIG. 3. Various possibilities for a) the energy dependence of the lattice period of the inverted potential and b) the corresponding temperature dependence of the action along a periodic path for the case of tunneling. W is the barrier height, β_c is the critical value of the inverse temperature β. The dotted curve corresponds to the model discussed in the text for autolocalization with no anharmonicity. The straight line on graph b shows the activated dependence corresponding to the path with zero amplitude of oscillation.

the value $\beta=\beta_0(p)+2\pi/\hbar\Omega$, after which point the bound motion corresponds to motion below the barrier.

By following this procedure, we find that the autolocalization probability per unit time of a particle of momentum p in the single-oscillator model under discussion here equals, to exponential accuracy,

$$\tau^{-1}(p)\propto\exp\left\{-\frac{2W}{\hbar}\left[\theta-\frac{1}{\omega_0}\mathrm{th}\left(\omega_0\left(\theta-\frac{\hbar\beta}{2}\right)\right)\right]+\frac{p^2}{\hbar m}\theta\right\}. \tag{48}$$

The value of θ for $\beta\leqslant\beta_0(p)$, and also for $p\geqslant\hbar|\varkappa_0|\omega/\Omega$ (which corresponds to $\beta_0=+\infty$) is given by the equation

$$\Omega^{-1}\,\mathrm{tg}(\Omega\theta)=\omega_0^{-1}\,\mathrm{th}[\omega_0(\theta-\hbar\beta/2)],\quad -\pi/2<\Omega\theta<0, \tag{49a}$$

while for $\beta\geqslant\beta_0(p)+2\pi/\hbar\Omega$, it is given by the other root of the same equation, which lies in the region

$$\pi/2<\Omega\theta\leqslant\pi, \tag{49b}$$

and for $\beta_0(p)\leqslant\beta\leqslant\beta_0(p)+2\pi/\hbar\Omega$, by the expression

$$\theta=\frac{\hbar\beta}{2}-\frac{1}{\omega_0}\mathrm{Arth}\left(\frac{p\Omega}{\hbar|\varkappa_0|\omega_0}\right) \tag{49c}$$

[The argument of the exponential in (48) in this region is linear in β].

Let us now focus our attention on the fact that θ is a continuous function of particle momentum and temperature, while $\tau^{-1}(p)$ is continuous along with its first derivatives with respect to p and β. If we study the dependence of the autolocalization rate on particle momentum for a given temperature, then it is clear that there exists a critical temperature.

$$\beta_c^{-1}=\hbar\Omega/2\pi, \tag{50}$$

below which $\tau^{-1}(p)$ has three analytic segments in p (49a, b, c) while above this temperature it has two such segments (49a, b). On the segment described by expression (49a), $\tau^{-1}(p)$ falls off exponentially with an increase in particle energy $p^2/2m$; on the branch described by (49c) (for $\beta>\beta_c$), it grows exponentially. Hence $\tau^{-1}(p)$ achieves its maximum for

$$p=\hbar|\varkappa_0|\,\omega_0\Omega^{-1}\,\mathrm{th}(\hbar\omega_0\beta/2)$$

and corresponds to the value $\theta=0$ [expression (49b)]. As the temperature falls, the region defined by expression (49b) shrinks while the particle energy which matches the maximum $\tau^{-1}(p)$ tends toward the barrier height W. The average probability per unit time τ^{-1} of autolocalization of thermal carriers, which in the exponential approximation coincides with $\tau^{-1}(0)$, for $\beta\geqslant\beta_c$ is given by expression (48) with $p=0$ and the value of θ from (49c), while for $\beta\leqslant\beta_c$ we are led to the activation expression

$$\tau^{-1}\propto e^{-W\beta}.$$

Having concluded our investigation of a simple model, let us turn to a discussion of the general case of barrier autolocalization. As we noted earlier, there is an analogy between autolocalization and capture on a deep impurity center; this implies that the prescription for finding the extremal path formulated at the end of the previous section is also fully pertinent to autolocalization at finite temperature and given particle energy. However, in addition to solving the classical equation of motion for the lattice, in the autolocalization case it is also necessary to investigate the action for various types of extrema. The physical basis for choosing a path was demonstrated earlier; in the general case we must also arrive at three types of extremal paths. The dependence of the autolocalization rate on particle energy and temperature obtained earlier is apparently general in a qualitative sense. The optimum particle energy for autolocalization at high temperatures is small, while at lower energies it converges towards the height of the saddle point. It is not possible to dwell here on a proof of this assertion.

The case of degeneracy gives rise to certain subtle qualitative regularities in the model under investigation. In order to see this, let us first examine the temperature dependence of the transition rate for a particle to exit a multidimensional potential well of general shape by tunneling. The potential energy $U(\mathbf{q})$ as a function of the particle coordinate $\mathbf{q}$ has a saddle point of height W at some point $\mathbf{Q}$ with a maximum along one direction and a minimum along the others. Calculating the exit probability from this well reduces (see section 2) to finding the classical path for a particle with given imaginary period $-i\hbar\beta$. If we make a variable transformation to imaginary time $\tilde{t}=it$, such a path with energy E will describe the general oscillatory motion in a reversed potential $\tilde{U}(\mathbf{q})=-U(\mathbf{q})$ with energy $-E$. For each value $0<E\leqslant W$ the purely oscillatory solution to the equation of

motion is unique, while for $E > W$ it is absent. The period of these oscillations $T(E)$ for E approaching W is finite and is determined by the frequency Ω of harmonic oscillations of a particle near to the saddle $\tilde{U}(\mathbf{q})$, while as E approaches the bottom of the potential well it diverges logarithmically. The variation of $T(E)$ in the intermediate region is determined by details of the potential $U(\mathbf{q})$, in particular its anharmonicity near the saddle point; however, it clearly is natural to suppose that $T(E)$ grows monotonically as E decreases (see Fig. 3a).

Returning to the usual time domain, we have for the action in one period, which according to (8) determines the lifetime of a particle in the well, the expression

$$-iS(\beta)=\hbar\beta W-\int_{E(\beta)}^{W} T(E)\,dE=\frac{2\pi}{\Omega}W+\int_{2\pi/\Omega}^{\hbar\beta} E(T)\,dT, \qquad (51)$$

where $E(\beta)$ is a root of the equation $T(E)=\hbar\beta$. In addition to the oscillatory path described above, the required condition of periodicity is also satisfied by the trivial solution to the equation of motion $\mathbf{q}(t)=\mathbf{Q}$ with action

$$S(\beta)=i\hbar\beta W.$$

The characteristic dependence of the action on temperature for both paths is displayed in Fig. 3b. For high temperatures the trivial activation path is unique; as the temperature is reduced, there is a concurrent gain in the oscillatory path which describes activated tunneling. For a monotonic dependence of $T(E)$ the transition occurs in a smooth fashion at a critical temperature $\beta_c^{-1}=\hbar\Omega/2\pi$ (see Ref. 19). In the non-monotonic case the dependence (51) of $\mathrm{Im}\,S$ on β has a characteristic "beak," and the transition occurs with a discontinuity in slope at a temperature larger than the critical one. In principle, more elaborate dependence for $T(E)$ and $S(\beta)$ are possible; however, in practice the existence of such forms is doubtful. As the temperature goes to zero, $S(\beta)$ goes to a constant, corresponding to the case of pure tunneling.

With these methods we can study ionization of an autolocalized state, where the role of a potential well is played by the adiabatic potential energy as a function of the lattice coordinates.[6] The character of the temperature dependence of the action described above, which determines the total ionization probability per unit time, and consequently the autolocalization rate for thermal carriers, is also entirely contained in this treatment. There is, to be sure, one subtlety connected with the fact that the adiabatic potential determines only the bound motion of the lattice. The activation and oscillatory paths are confined (with rather small amplitudes) entirely within the bound motion region. However, as the amplitude of oscillatory motion increases, a configuration is reached where the autolocalized particle has zero binding energy, and for larger amplitudes of oscillation the path certainly includes a segment of the free motion (see Fig. 1b). Because there is a specific time at which this free-motion segment first appears, a weak singularity is present in $T(E)$ and in the temperature dependence of the action (51). In a calculation of the autolocalization rate for nonthermal carriers this singularity will be smoothed out due to the complex-valued path.

According to the picture obtained here, as the temperature decreases, the character of the transition undergone by the autolocalization rate (smooth or with a "kink") from activated to activated tunneling (see Fig. 3b) is in fact determined by the anharmonicity of the adiabatic potential energy $U(\mathbf{q})$ near the saddle point. In the single-oscillator model investigated earlier there was no anharmonicity at all [see (43)]; therefore, the dependence of $T(E)$ has a horizontal segment (see Fig. 3a). The temperature dependence of the action is shown in Fig. 3b; the curve which corresponds to tunneling branches off from the activated curve at the point $\beta=\beta_c$, as in the case of monotonic dependence of $T(E)$, only with different slope. The slope discontinuity we obtain is related to the fact that for a transition across the critical temperature (5) ($p=0$), the oscillation amplitude C in (44) grows discontinuously from zero to $|\varkappa_0|\omega_0^2/\varkappa_1\Omega^2$, enclosing the free-motion region $q\leqslant|\varkappa_0|/\varkappa_1$.

Barrier autolocalization for various specific lattice interaction mechanisms was investigated for the case of cold particles at zero temperature in Refs. 11, 12, and for the case of thermal carriers at finite temperatures in the more recent works of Yoselevich and Rashba.[13] The formulation given above, which provides a general prescription for calculating the exponents in expressions for the autolocalization rate τ^{-1} applicable to thermal particles, is equivalent to the one used in Ref. 13; however, the qualitative pictures of the extremal action $S(\beta)$ are somewhat different. According to Ref. 13, both activated and tunneling paths are present; the latter disappear as the temperature increases, "annihilating" with extremals of a third kind—the "long instantons." In such a picture the curve $S(\beta)$ corresponding to a tunneling path intersects the linear activation dependence, so that the change in regimes of temperature dependence of τ^{-1} occurs with a discontinuity. According to the picture set forth in the present work, the tunneling path changes over, generally speaking, in a continuous way to activated at a critical temperature β_c^{-1}, determined by the curvature of the saddle point. At this point the tunneling curve $S(\beta)$ branches off from the activated dependence, in general without a slope discontinuity. The slope discontinuity in the temperature dependence of τ^{-1} due to the intersection at some $\beta<\beta_c$ of the activation and tunneling dependence of $S(\beta)$ therefore does not always occur, but rather appears only in the case of a non-monotonic energy dependence of the oscillation period of the inverted potential $T(E)$, leading to the "beak-shaped" bend in the tunneling curve $S(\beta)$ (see Fig. 3). A precise conclusion about the character of the transition between the activation and tunneling regimes in the temperature dependences of the autolocalization rate for specific models can be obtained on the basis of a rigorous investigation of the anharmonic oscillations near the saddle point.

A quantitative discussion of the true mechanisms of autolocalization falls outside of the framework of this paper, since it requires numerical calculations for all but a few special cases. One such case is the autolocalization of Wannier-Mott excitons in polar crystals when the difference between electron and hole masses is large; in this system (according to Ref. 12) the deformation potential acting on the exciton remains constant in form during the tunneling process,

changing only in strength. Describing the lattice deformation in terms of this strength with a single coordinate, we obtain the single-oscillator model investigated above with parameters

$$m=m_h, \quad -\varkappa_0=\varkappa_1=W_0(2E_0)^{-1/2}a^{-2}, \quad M=2W_0\hbar^4\tilde{\varepsilon}/e^2m_h^2\omega_0^2a^3,$$

where m_h is the hole mass, assumed to be large, a is the exciton Bohr radius determined by the small electron mass, ω_0 is the optical phonon frequency, $\tilde{\varepsilon}$ is the inertial dielectric permittivity of the crystal, e is the electron charge, $W_0\approx 1.07$ and $E_0\approx 3.1$ are constants calculated in Ref. 12. The autolocalization rate for excitons with momentum p (i.e., with energy $p^2/2m_h$) is given to exponential accuracy by expressions (48) and (49). We must keep in mind that certain of the quantitative details predicted here may also appear within the accuracy implied by the approximations used in Ref. 12. This problem is investigated in Ref. 13 for thermalized excitons.

5. CONCLUDING REMARKS

The quasiclassical method we have presented here for calculating the thermodynamic mean probabilities for various processes (within the exponential approximation) reduces to finding the correct method of determining the classical paths. In this work, the coefficient of the exponential is obtained only for the one-dimensional case of a transition between terms [formula (16)]; in the multidimensional case it is necessary to calculate rigorously determinants of the type (5). For transitions involving a portion of the continuous spectrum even this is insufficient; the corresponding development of the method to include this case will be presented in a future publication.

Application of this approach is limited by the usual quasiclassical requirement that large values of the exponential argument appear. As applied to the problems investigated in this paper, i.e., capture on a deep trap and autolocalization, this limitation will in turn give rise to the adiabatic condition $|E(\mathbf{q})| \gg \hbar\,|\theta|^{-1}$ for the characteristic binding energy on the bound segment of the path. Other criteria, which in one way or another reduce to limits on the nonadiabaticity of the system, are pointless even to enumerate in view of their dependence on specific properties of the system under study. An important requirement of a different sort, which essentially bounds from above the admissible values of momentum p for a captured or autolocalized particle, is that the localization of the lattice distortion at the instant of binding be on a scale smaller than $\hbar/p$. In order to remove this restriction, we must give up the use of the zero-radius potential method and attempt to understand in more detail the boundary conditions for integrating along the path of "rapid" motion. We note that large particle energies require an accelerated relaxation rate into the final-state region. When this relaxation rate is too small, trapping or autolocalization states will "decay back" before they can thermalize.

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[1)]It is necessary to emphasize that for such a circuit, the transition between states corresponding to various terms of the electronic subsystem takes place continuously (adiabatically); therefore, it is not necessary to impose any supplementary principles (such as the Franck-Condon principle).

[2)]The staircase spreads out as the energy goes down, so that at the end of the cascade, the transitions can be described as essentially multiphonon transitions.

[3)]The trivial solution $S(x, t; x', t') = m(x-x')^2/2(t-t')$ which corresponds to free particle motion without any interaction with the well, is of no interest to us.

[4)]We point out that for $p>0$ the connection time corresponds formally to a positive energy for the bound state.

[5)]In this model, an energy minimum corresponding to an autolocalized state is absent (it goes to $-\infty$); however, this is irrelevant to an investigation of the autolocalization rate.

[6)]The lack of dependence of $U(\mathbf{q})$ on an arbitrary spatial translation of the lattice configuration as a whole is important only in calculating the coefficient of the exponential. The possibility of secondary extrema of $U(\mathbf{q})$ is likewise unimportant, since the action on trajectories associated with these extrema makes an exponentially small contribution to the whole.

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