Role of fluctuational barrier "preparation" in the quantum diffusion of atomic particles in a crystal

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Quantum subbarrier diffusion of atoms in a crystal under conditions when the decisive role is played by the coherent "preparation" of the barriers as a result of the quantum fluctuations of the ambient medium is considered. It is shown that in this case the coherent-tunneling amplitude and the coherent-diffusion coefficient may, in sharp contrast to their usual behavior, increase with increasing temperature. In the general case the actual temperature dependence is the result of the competition between the fluctuational barrier preparation effect and the polaron effect. It is found that the investigated mechanism is also important for incoherent diffusion, which, as is demonstrated, predominates at high temperatures, even in the absence of the polaron effect.

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1. INTRODUCTION

A distinctive feature of the quantum subbarrier diffusion of atomic particles in a crystal is the extreme narrowness of their band, even in the absence of the polaron effect. This is primarily connected with the extreme smallness of their tunneling amplitude because of the large diffusing-particle mass. The vibrations of the lattice lead, at low temperatures (T), to intraband scattering,[1] or, in a wide T range, to the appearance of the phenomenon of "dynamical band destruction,"[2,3] which gives rise to a sharp decrease of coherent diffusion with increasing T in that region of relatively low T where coherent diffusion predominates. The presence of an appreciably strong polaron effect aggravates the situation, leading to a sharp band narrowing that intensifies with increasing T.

In this connection, for the diffusing atomic particles there may become important at low T, at which the over-the-barrier motion is certainly inhibited, a phenomenon that may be called *coherent barrier "preparation."* We are here talking about the quantum fluctuations of the atomic configuration surrounding the particle, fluctuations which may lead to abrupt lowering of the potential barriers for the particle. The probability amplitude for such a fluctuation does not, naturally, depend on the mass of the diffusing particle. Therefore, the heavier the diffusing particle, or, generally, the smaller the tunneling amplitude, is, the more important can be such a fluctuational, coherent, low-barrier preparation. It is important to emphasize that we are here talking about a coherent mechanism, when the state of the lattice (i.e., of the matrix) remains unchanged and the correlation between the tunneling and the fluctuations of the ambient medium is realized on the level of the wave function of the system, with the preservation, naturally, of the coherent (in the limiting case, band) transfer mechanism. The barrier alteration resulting from such fluctuations can significantly affect the magnitude of the incoherent subbarrier diffusion accompanying the change in the vibrational state of the crystal.

If the fluctuational preparation of the barriers is important, then the nature of the T dependence of the band width, i.e., of the coherent-tunneling amplitude Δ, changes (naturally, the quantity $\Delta(T=0)$ also changes in comparison with its value in a rigid lattice). Indeed, the fluctuations grow with increasing T, and this leads, as a result, to the growth of the coherent width Δ, in contrast to the polaron-effect induced decrease of Δ with increasing T. The actual temperature dependence of $\Delta(T)$ will be determined by the relation between the two effects.

An *a priori* judgment about the character of the temperature dependence of Δ can be made in the case of isotopic diffusion. The obvious smallness of the polaron effect in this case may cause the fluctuational barrier preparation effect to play the dominant role and, consequently, make Δ increase with increasing T. Evidently, an example of this kind of system is the isotopic mixture He^3-He^4. The exchange character of the diffusion (the elementary act of interchange of two atoms at neighboring sites), the relatively large mass of the atoms, the low Debye frequency, all this compels us to think that quantum diffusion in this case should be accomplished not through barriers determined by the normal disposition of the surrounding atoms, but through fluctuationally lowered barriers. It should be pointed out here that the growth of $\Delta(T)$ and, consequently, of the diffusion coefficient in the case when the scattering of the diffusing particles by each other predominates[2] can provide another mechanism for the exponentially increasing (with increasing T) diffusion along with the vacancy mechanism usually adopted for the explanation of the exponential growth of the diffusion coefficient with T[4,5] (see also[6]). Notice that, in general, in molecular crystals with a weak van der Waals interaction the fluctuational barrier preparation mechanism will evidently play an important role in the coherent diffusion of the atoms at low T.

The possibility of an effect of the atomic-configuration fluctuations on the potential barrier for a diffusing particle was first pointed out by Flynn and Stoneham.[7] They considered purely incoherent diffusion, adopting the simplest model in which the tunneling amplitude was equal to zero when the displacements of some closest atoms were less than a certain value and equal to a constant finite value otherwise.

In our earlier[2] investigation of coherent transfer, we considered virtually the opposite case when the potential relief (barriers) changes relatively little when the configuration of the surrounding atoms changes, or, which is the same thing, when it is necessary to effect a drastic change in the atomic configuration in order to change the tunneling amplitude appreciably. It is, however, easy to see that all the results (in[2]) pertaining to coherent diffusion are perfectly general in nature, it being only necessary to insert in all the final expressions for the coherent-diffusion coefficient the correctly determined coherent width $\Delta(T)$.

2. COHERENT TUNNELING

Let us consider the role of fluctuational barrier preparation in coherent subbarrier transfer, using as an example the diffusion of atoms in an interstice lattice formed in a regular crystal, i.e., let us consider the same case analyzed in[2]. From all appearances the results obtained below have a general character, and the qualitative picture will be preserved in any conceivable case of coherent diffusion of atoms in a crystal.

Let the temperature be sufficiently low, so that the transfer is accomplished only at the lowest level in the individual potential wells formed by the atoms of the matrix, and let the phonon-induced incoherent diffusion be negligible. Then coherent transfer during scattering of the particles by phonons, by impurities, or by each other can be uniquely determined in terms of the interstitial-transition amplitude which is diagonal with respect to the quantum numbers of the lattice.[2] In the limiting case of weak scattering the corresponding coherent width Δ is determined up to a constant coinciding with the width of the resulting band by the expression

$$\Delta\equiv\Delta(g)=\int(dR)\,\Phi_\nu^{1*}(R)J(R)\Phi_\nu^{1+g}(R), \tag{2.1}$$

$$J(R)=\int d^3r\,\chi^{1*}(\mathbf{r},R)H_1(\mathbf{r},R)\chi^{1+g}(\mathbf{r},R). \tag{2.2}$$

Here χ^1 and χ^{1+g} are the particle wave functions in the two neighboring wells (i.e., at the "sites") 1 and $1+g$ in the absence of tunneling; $H_1(\mathbf{r},R)$ is the difference between the true particle-crystal interaction Hamiltonian and the Hamiltonian describing the locally preferred well (1) for a fixed configuration of the crystal atoms, which is conditionally denoted here by the single letter R (here and below we omit the level index); Φ_ν^1 and Φ_ν^{1+g} are the wave functions of the vibrational motion of the crystal, when the diffusing particle is respectively at the sites 1 and $1+g$, and now, owing to the polaron effect, the atoms of the matrix vibrate near the new equilibrium positions $\tilde{R}^{(1)}$ and $\tilde{R}^{(1+g)}$.

The explicit structure of the expressions (2.2) and (2.1) corresponds to the adiabatic picture, which is appropriate for light-particle diffusion in a "heavy" matrix (for details, see[2]). It should, however, be noted that an expression of the form (2.1) is apparently preserved also in the case when the two masses are of the same order of magnitude, since the act of particle motion under a high barrier is itself a fast process.

Let us use the fact that the determination of the diagonal matrix element with respect to the state of the macroscopic system is equivalent to thermodynamic averaging with the corresponding density matrix. By considering the vibrations of the atoms of the matrix near the new equilibrium positions in the framework of the harmonic problem, and going over to a system of independent real normal coordinates u_β, we can use the well-known Slater formula (see, for example,[8]):

$$\sum_\nu \rho_{ph}^{(0)}(\nu)\Phi_\nu^*(R-\tilde{R}^{(0)})\Phi_\nu(R-\tilde{R}^{(g)})$$

$$=\prod_\beta\left(\frac{\xi_\beta}{2\pi}\right)^{1/2}\exp\Big\{-\frac{1}{2}\sum_\beta\xi_\beta(u_\beta-\eta_\beta{}^g)^2-\Phi^g\Big\}; \tag{2.3}$$

$$\Phi^g\equiv\Phi=\frac{M^2}{2}\sum_\beta(w_\beta{}^g)^2\omega_\beta{}^2\xi_\beta{}^{-1}, \tag{2.4}$$

where

$$\eta_\beta{}^g\equiv{}^1/_2(\tilde{u}_\beta{}^{(0)}+\tilde{u}_\beta{}^{(g)}),\quad w_\beta{}^g\equiv\tilde{u}_\beta{}^{(0)}-\tilde{u}_\beta{}^{(g)},$$
$$\xi_\beta\equiv 2M\omega_\beta\,\mathrm{th}(\omega_\beta/2T),\quad \hbar\equiv 1. \tag{2.5}$$

Here $\tilde{u}_\beta^{(l)}$ is the displacement of the center of the normal oscillator when the diffusing particle is localized in the l-th well (because of the translational symmetry, we take $l=0$), while the factor $\exp(-\Phi^g)$ figuring in the expression (2.3) is the usual polaron exponential function corresponding to the diagonal (with respect to the phonons) transition.

Let us introduce the variables

$$q_\beta=u_\beta-\eta_\beta{}^g. \tag{2.6}$$

Then the expression for the coherent width (2.1) can be rewritten in the form

$$\Delta=e^{-\Phi}\prod_\beta\left(\frac{\xi_\beta}{2\pi}\right)^{1/2}\int(dq)\,\exp\Big\{-\frac{1}{2}\sum_\beta\xi_\beta q_\beta{}^2\Big\}J(q+\eta), \tag{2.7}$$

where q conditionally denotes the set of all the q_β, and, for simplicity of notation, the index g has been dropped.

If the resonance integral J varies slowly during the thermal migration of the surrounding atoms, then we have the case when the fluctuational alteration of the barrier is unimportant for coherent tunneling. The temperature dependence of Δ is then virtually entirely determined by the behavior of the polaron exponential function. This corresponds precisely to the case considered in[2]. In the general case, however, the resonance integral can be very sensitive to the configuration of the surrounding atoms, especially if we bear in mind the scale of the exponential smallness of J for subbarrier penetration by the atomic particles. The q dependence of J then becomes important: The tunneling begins to effectively correlate with the fluctuations of the ambient medium. In other words, coherent barrier preparation becomes important.

Let us write the expression for J in the form

$$J(q+\eta)=\Omega_0\exp[-B(q+\eta)]. \tag{2.8}$$

In the problem under consideration

$$B\gg 1. \tag{2.9}$$

Only the displacement of a definite small group of atoms can naturally lead (at fixed **g**) to a sharp decrease in B. If for this distinct group the variation of B with the displacement is so rapid that the dominant contribution to the integral (2.7) is made by the region in which the displacements significantly exceed the thermal displacements, then we arrive at a typical "steepest-descent" situation with some extremal fluctuation (q^*) in the disposition of the atoms.

Let us restrict ourselves to the consideration of that model for the crystal in the framework of which this extremal configuration is virtually determined by the values of a few normal coordinates q'_λ. (For example, this could be the Einstein model of independent oscillations of the atoms of the matrix.) As to the remaining variables, we shall, in contrast, assume a relatively slow variation of B with the q_β, and expand this quantity in a series near $q_\beta=0$:

$$B=B(\eta,q'+\eta')+\sum_\beta{}' B_\beta q_\beta+\ldots, \tag{2.10}$$

where $B_\beta\equiv\partial B/\partial q_\beta$ (the prime on the summation sign indicates that we do not sum over the "steepest-descent" variables).

We restrict ourselves in (2.9) to the linear terms, assuming that, on account of (2.9),

$$\sum_\beta B_\beta^2\overline{q_\beta^2}\gg\sum_\beta B_{\beta\beta}\overline{q_\beta^2}$$

and that allowance for the higher terms of the expansion will only lead to a correction in the preexponential factor. The extremum with respect to the variables q'_λ is found from the relation

$$\partial B(\eta,\ q'+\eta')/\partial q_\lambda'+\xi_\lambda q_\lambda'=0. \tag{2.11}$$

Determining from this relation the corresponding values of q^*_λ, expanding $B(\eta,\ \eta'+q')$ in a power series in $q'_\lambda-q^*_\lambda$, and retaining in this expansion the terms quadratic in this difference, we find from (2.7) and (2.8) that

$$\Delta=J(\eta)F(T)e^{-\Phi},\quad F(T)\approx A\exp\{\Delta B^*-\varphi(T)+\chi(T)\}. \tag{2.12}$$

Here we have introduced the notation

$$J(\eta)=\Omega_0e^{-B(\eta)},\quad B(\eta)=B(\eta,\ \eta'),\quad \Delta B^*=B(\eta)-B(\eta,\ q^*+\eta'); \tag{2.13}$$

$$\varphi(T)={}^1/_2\sum_\lambda\xi_\lambda q_\lambda^{*2},\quad \chi(T)={}^1/_2\sum_\beta{}' B_\beta^{*2}\xi_\beta^{-1}. \tag{2.14}$$

(All the asterisked quantities are determined at the "steepest-descent" values of q^*_λ.) The form of the preexponential function A, which is not important for the subsequent analysis, can be found directly without difficulty.

Notice that the steepest-descent situation presupposes the validity of the inequalities $\xi_\lambda q_\lambda^{*2}\gg 1$ and, simultaneously,

$$\Delta B^*-\varphi(T)\gg 1. \tag{2.15}$$

The expressions (2.12)–(2.14) allow us to elucidate in the general case the nature of the temperature dependence of the coherent width Δ and, at the same time, that of the coefficient of coherent diffusion for a fixed scattering mechanism.

Let the polaron effect be weak. Then it follows immediately from (2.12)–(2.14) and (2.4) that the *coherent width monotonically increases with increasing temperature*. (Some variation of q^*_λ with temperature, which follows from (2.11), does not change this assertion.)

In those cases when the fluctuational preparation of the barrier plays an important role, the term ΔB^* can partially cancel out the initial barrier exponential function $B(\eta)$. There then arises a strong temperature dependence, determined by the probability for such a fluction, and described by the function $\varphi(T)$. As T increases, this function quite rapidly reaches its classical ($T>\omega_D/2$) limit:

$$\varphi(T)\to\varphi_{cl}(T)=\frac{M}{2T}\sum_\lambda\omega_\lambda^2q_\lambda^{*2}\equiv\frac{Q}{T}, \tag{2.16}$$

giving a pure-activation growth of Δ with temperature that, generally speaking, is not quite characteristic of coherent diffusion.

If the controlling factor turns out to be the alteration of the barrier due to the usual small oscillations of the atoms of the matrix near their equilibrium values, then the temperature dependence of Δ turns out to be different, which is easy to infer from the classical limit for the function $\chi(T)$:

$$\chi(T)\to\chi_{cl}(T)=\frac{T}{2M}\sum_\beta{}' B_\beta^{*2}\omega_\beta^{-2}. \tag{2.17}$$

The polaron effect leads by itself to the decrease of the coherent width with increasing temperature (see (2.4)). In the general case the actual temperature dependence arises as a result of the competition between the two factors, and Δ can increase or decrease with increasing T. Evidently, we should meet with both situations in the case of coherent diffusion of interstitial atoms (for example, of hydrogen isotopes) in a crystal. The heavier the diffusing atoms are, the larger B is and the more serious the role of the fluctuational lowering of the barrier turns out to be, which always leads, in the long run, to the growth of the coherent tunneling with T. If the diffusing atoms are located at the lattice sites and an elementary act of tunneling has an exchange character, then the polaron effect is much weaker, especially in the isotopic case, and the temperature dependence of Δ should virtually always be determined by the fluctuational preparation of the barrier. (The nature of the dependence $\Delta(T)$ in this case will be close to that of (2.12)–(2.13).) Then (see (2.12)–(2.17)) the softer the phonon spectrum of the matrix is, the more vividly expressed the barrier preparation effect will be. Evidently, the diffusion of a helium isotope in the matrix of another isotope pertains to a case of this sort.

Notice that the polaron effect and the fluctuational barrier lowering are, in the general case, dictated by the displacements of different groups of the surrounding atoms. This is connected with the fact that, in accordance with (2.4)–(2.5), the contribution to the polaron effect is made by only those static displacements that turn out to be different when finding the diffusing particle in two neighboring wells.

3. INCOHERENT TUNNELING

Clearly, the fluctuations of the barrier will, in the general case, also play an important role in the incoherent tunneling of particles which is accompanied by an alteration of the phonon system of the crystal. We shall consider this problem, limiting ourselves first to the case when the fluctuations of the matrix atoms within the limits of their thermal displacements constitute the controlling factor. Here we shall assume the validity of the expansion (2.10) not just for the variables indicated there, but for all the variables.

The off-diagonal—with respect to the phonons—matrix element for the transition from one well to a neighboring well is determined by an expression of the type (2.4) if we replace ν by ν' ($\neq \nu$) in one of the wave functions of the crystal. Using the representation defined by (2.8), (2.10), and (2.5), we directly have that

$$\Lambda_{\nu\nu'}=J(\eta)(\hat{\Lambda})_{\nu\nu'},\quad \hat{\Lambda}\equiv\prod_{\beta}\exp\{iw_{\beta}^{\varepsilon}\hat{p}_{\beta}+B_{\beta}q_{\beta}\}. \tag{3.1}$$

where $\hat{p}_\beta$ is the momentum operator for the β-th normal mode. Upon the neglect of the dependence of the barrier on the vibrations of the atoms of the matrix (i.e., for $B_\beta=0$), the generalized displacement operator $\hat{\Lambda}$ assumes the usual meaning corresponding to the pure polaron effect (see[2]).

The incoherent-transition probability

$$W=2\pi J^2(\eta)\sum_{\nu\neq\mu}\rho_{ph}^{(0)}(\nu)\,|\hat{\Lambda}_{\nu\mu}|^2\delta(E_\nu-E_\mu) \tag{3.2}$$

can be found with the aid of a procedure similar to the one used in the theory of polarons of small radius (see, for example, [9,10]). The corresponding computations lead directly to the following result:

$$W=J^2(\eta)\exp[-2\Phi+2\chi]\int_{-\infty}^{\infty}dt\{\exp[\psi_1(t)+i\psi_2(t)]-1\}, \tag{3.3}$$

where

$$\psi_1(t)=\sum_{\beta}\frac{\cos\omega_\beta t}{\operatorname{sh}(\omega_\beta/2T)}\left\{\frac{M\omega_\beta(w_\beta^{\varepsilon})^2}{2}+\frac{B_\beta^2}{2M\omega_\beta}\right\},$$
$$\psi_2(t)=\sum_{\beta}\frac{\sin\omega_\beta t}{\operatorname{sh}(\omega_\beta/2T)}\omega_\beta w_\beta^{\varepsilon}B_\beta. \tag{3.4}$$

As $T\to 0$, the two functions $\psi_{1,2}(t)$ tend to zero, so that, as before, at sufficiently low temperatures the incoherent diffusion is negligible compared to the coherent diffusion (cf.[2]). At temperatures at which the inequality $\psi_1(0)\gg 1$ is valid, only the region of small t is important in the integral (3.3). We can then set $\cos\psi_2(t)\approx 1$ and use the method of steepest descent to evaluate the integral. Then for the classical region $T>\omega_D/2$ we directly have:

$$W\approx J^2(\eta)[\pi/4T(\mathscr{E}+\gamma)]^{1/2}\exp\{-\mathscr{E}/T+\zeta T\}, \tag{3.5}$$

where

$$\mathscr{E}=\frac{M}{8}\sum_{\beta}(w_\beta^{\varepsilon})^2\omega_\beta^2,\quad \zeta=\frac{2}{M}\sum_{\beta}B_\beta^2\frac{1}{\omega_\beta^2},\quad \gamma=\sum_{\beta}\frac{B_\beta^2}{8M}. \tag{3.6}$$

The incoherent-transition probability increases with temperature, owing both to the polaron effect (with an activation energy $\mathscr{E}$) and to the fluctuational alteration of the barrier, although the temperature dependence itself is different in the two cases. (The interference between the two effects is negligibly small in the region of high T, although at low $T\ll\omega_D$ its contribution can be appreciable.) It is interesting to note that the fluctuational preparation of the barrier thus leads to the growth of both the coherent and the incoherent tunneling. It follows from (3.5), (3.6), and (2.17) that the exponent ζT in the incoherent-transition probability is two times greater than the corresponding exponent 2χ for the square of the coherent amplitude, so that at sufficiently high temperatures the incoherent diffusion predominates over the coherent diffusion also in the absence of the polaron effect.

Let us now consider the steepest-descent situation. The off-diagonal (in the phonon numbers) matrix element for the transition from one well into another is then determined by the expression (3.1) if in place of $\hat{\Lambda}$ we introduce the operator

$$\tilde{\Lambda}=\hat{\Lambda}\hat{\Lambda}', \tag{3.7}$$

where

$$\hat{\Lambda}'=\exp\left\{\frac{i}{2}\sum_{\lambda}\hat{p}_\lambda' w_\lambda\right\}\exp\{\Delta B(\eta,q'+\eta')\}\exp\left\{-\frac{i}{2}\sum_{\lambda}\hat{p}_\lambda' w_\lambda\right\} \tag{3.8}$$

is an operator that acts only on the steepest-descent variables. The expression (3.2) for the incoherent-transition probability can then be represented in the form

$$\tilde{W}=J^2(\eta)\int_{-\infty}^{\infty}dt\left\{\mathrm{Sp}(\hat{\rho}_{ph}^{(0)}\tilde{\Lambda}(t)\tilde{\Lambda}^{+}(0))-\sum_{\nu}\rho_{ph}^{(0)}(\nu)\,|\tilde{\Lambda}_{\nu\nu}|^2\right\} \tag{3.9}$$

($\tilde{\Lambda}(t)$ is an operator in the Heisenberg representation).

If the excitation of phonons during the transition is connected primarily with the non-steepest-descent variables, then for $\tilde{W}$ we easily find

$$\tilde{W}\approx WA^2\exp\{2\Delta B^{*}-2\varphi(T)\}, \tag{3.10}$$

where W coincides with (3.3)–(3.6) (the sums over β

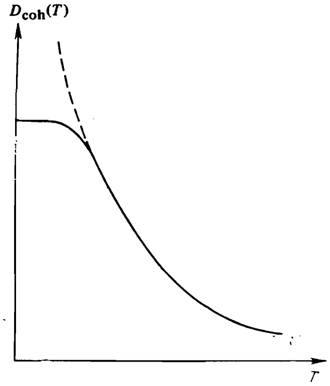

FIG. 1.

pertain in this case only to the non-steepest-descent variables) and the remaining quantities are defined in accordance with (2.12)–(2.14). In particular, in the classical temperature region the probability $\tilde{W}$ will, in comparison with (3.5), contain the additional activation factor $e^{-2Q/T}$, with Q defined in accordance with (2.16). At such T the expression (3.10) virtually generalizes the result obtained in[7] by Flynn and Stoneham, who first considered the question of the influence of the vibrational fluctuation of a lattice on incoherent diffusion for the simple model to which correspond $J=0$ for $q'<q^*$ and a finite $J=J_0$ for $q'>q^*$.

In the general case the off-diagonal (in the phonon numbers) transitions for the normal modes responsible for the steepest-descent situation itself may turn out to be important in (3.9). In this case, however, allowance for the off-diagonal elements of the operator $\hat{\Lambda}$, (3.8), in the expression (3.9) requires knowledge of the explicit form of ΔB. But the unwieldy formulas for this quantity, which are obtainable in their general form, possess little physical transparency, and we shall not give them here.

Thus, the critical dependence of the barrier on the displacement of the surrounding atoms gives rise by itself to a significant growth of the incoherent diffusion with temperature. In the case of a weak polaron effect, it can determine the actual temperature dependence. Allowance for the off-diagonal transitons with respect with to the steepest-descent variables only reinforces the dependence predicted by the expression (3.10), its character remaining virtually the same.

4. DISCUSSION OF THE RESULTS

In accordance with results of our previous paper,[2] the expression for the coherent-diffusion coefficient has the following form (for a matrix of cubic symmetry, z being the number of nearest neighbors):

$$D_{coh}(T)\approx{}^1/_3 za^2\Delta^2(T)/\Omega(T), \quad (4.1)$$

where Ω is the rate of damping of the off-diagonal density matrix elements as a result of the dynamical band destruction[2,3] or the scattering of the diffusing particles by each other or by defects.[2] Similarly, for the incoherent, i.e., phonon-stimulated diffusion we have

$$D_{incoh}(T)\approx{}^1/_3 za^2\tilde{W}(T). \quad (4.2)$$

The general expressions obtained above for Δ, (2.12)–(2.17), and $\tilde{W}(T)$, (3.3)–(3.10), allow us to analyze the general qualitative picture of the behavior of the diffusion coefficient. Let us begin with the region of low T, when the incoherent diffusion is negligible compared to the coherent diffusion. As $T\to 0$, the coherent width Δ, (2.12), tends to a constant limit $\Delta(0)$, which, in the general case, is sensitive to the zero-point vibrations of the matrix. It is worth noting that in many cases the penetration of the barrier between two equivalent wells at $T=0$ will itself also be determined to a decisive degree by the fluctuational preparation of the barrier as a result of the zero-point vibrations of the matrix.

The diffusion coefficient also assumes, as $T\to 0$, a constant value determined by the static scattering of the particles by the defects of the crystal or by each other. At higher T the behavior of the diffusion coefficient will depend to a considerable degree on the relation between the polaron and barrier-preparation effects. If the polaron effect predominates, then $D_{coh}(T)$ monotonically decreases with increasing T, as shown in Fig. 1, the decrease being first according to a power law ($\Omega\propto T^9$) and then according to an exponential law determined by the polaron exponent $2\Phi(T)$ (see (2.12) and (2.4)). If, on the other hand, the decisive rolе is played by the fluctuational alteration of the barrier, then the initial decrease of $D_{coh}(T)$ as a result of the dynamical destruction of the band is replaced by its increase, which rapidly becomes exponential, and the temperature dependence will have the form of the curve 1 in Fig. 2. If the static-scattering rate is sufficiently high, then the minimum may not appear, so that $D_{coh}(T)$ will monotonically increase with T (see the curve 2 in Fig. 2).

For $T\ll\omega_D$ in the case of a weak polaron effect, the increase of $\Delta(T)/\Delta(0)$ is determined by an exponential function with an exponent, $\chi(T)-\varphi(T)$, that, in the case when the acoustic phonons predominate, depends on T as some power of the ratio T/ω_D. (Taking into account the fact that $B_\beta\sim f$, $\beta\equiv \mathbf{f}$, α, where $\mathbf{f}$ is the wave vector and α is the number of the phonon branch, we can easily infer that $\chi(T)-\chi(0)\sim(T/\omega_D)^4$. A similar dependence arises for $\varphi(0)-\varphi(T)$ if we assume that q_λ^* does not depend on f.) At higher T the exponent of the increasing exponential function, $\chi(T)-\varphi(T)$, assumes quite rapidly with increasing T the classical value determined by the relations (2.16) and (2.17), with dif-

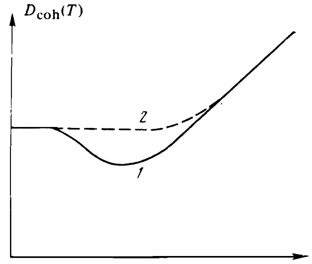

FIG. 2.

ferent temperature dependences for the two terms. (Notice, for comparison, that the polaron parameter $\Phi(T)-\Phi(0)$ in the case when the acoustic phonons predominate depends on T at low T also like $(T/\omega_D)^4$, while at $T>\omega_D$ we have $\Phi(T)-\Phi(0)\propto T$.)

The fluctuational barrier lowering effect should play a very important role in subbarrier diffusion of atomic particles whose mass is large compared to the electron mass. The exponential dependence of the tunneling amplitude on the particle mass m for a fixed barrier and the virtual nondependence on m of the polaron exponent of the exponential function clearly lead to a situation in which the fluctuations of the barrier play a more and more decisive role as the mass of the diffusing particle increases. The effect can become so strong that it virtually amounts to a coherent preparation of a "hole" in the atomic configuration, especially for heavy particles or for "soft" lattices with an appreciable zero-point vibration amplitude. We emphasize the coherent nature of the "hole" (or barrier) preparation, bearing in mind its virtual character and the return of the lattice to the normal (with respect to the phonon number) state. Here we do not, naturally, postulate the actual creation of a vacancy. In this sense, the considered subbarrier-transfer mechanism differs radically from the vacancy mechanism, which has been widely discussed in connection with the problem of diffusion in quantum crystals (see, for example, [6]), although in both cases there arises an exponential growth of the coherent-diffusion coefficient with increasing T (in the case of the vacancy mechanism this growth is connected simply with the growth of the number of real vacancies). In the case of diffusion of He^3 in He^4, such a growth has been experimentally observed[4,5] (see also[6]).

As the temperature increases further, the incoherent or phonon-stimulated diffusion mechanism begins to play a greater and greater role, going over subsequently into the quasiclassical over-the-barrier diffusion. Not dwelling on the picture that arises in the fixed-potential-profile model and, in particular, on the role, discussed in detail in[2], of the many-level nature of the individual potential wells, we only note that, as follows from Sec. 3, the fluctuations of the barrier can sharply enhance the incoherent transfer mechanism. It is significant that such a transfer mechanism becomes predominant at sufficiently high T and in the total absence of the polaron effect, leading to a change in the exponential growth in comparison with $D_{\rm coh}(T)$. The qualitative picture of the dependence $D(T)$ naturally remains the same as shown in Fig. 2.

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Two-dimensional Heisenberg model with weak anisotropy

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A low-temperature phase transition in a two-dimensional Heisenberg model with weak anisotropy is considered. The transition is associated with spontaneous breaking of the symmetry group $O(2)$ of the Hamiltonian. The scaling dimension of the spin operator and the correlation length of the spin fluctuations that take the spin out of the easy-magnetization plane are calculated in the leading logarithmic approximation.

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1. INTRODUCTION

The question of the existence of a phase transition in two-dimensional degenerate systems (the XY-model and Heisenberg model) has been considered in the papers.[1–4] Berezinskiĭ showed that a phase transition exists in the XY-model, even though the spontaneous magnetic moment is equal to zero at all finite temperatures. Arguments in favor of the existence of a phase transition in the Heisenberg model were adduced in[2]. However, it was made clear in[3] that the screening of the interaction which takes place in this case was not taken into account in[2]. The question of the existence of a phase transition in the isotropic Heisenberg model