

were observed conflicted with the criteria in Ref. 6.

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<sup>1)</sup>The  $1/f$  noise includes also the noise obeying the law  $1/f^\alpha$ , where  $\alpha = 1 \pm 0.3$ .

<sup>1)</sup>Sh. M. Kogan, *Usp. Fiz. Nauk* **123**, 131 (1977) [*Sov. Phys. Usp.* **20**, 763 (1977)].

<sup>2)</sup>B. I. Shklovskii, *Solid State Commun.* **33**, 273 (1980).

<sup>3)</sup>A. N. Malakhov and A. V. Yakimov, *Radiotekh. Elektron.* **19**, 2435 (1974).

<sup>4)</sup>Tezisy dokladov II konferentsii po fluktuatsionnym yavleniyam v fizicheskikh sistemakh (Abstracts of Papers presented at Second Conf. on Fluctuation Phenomena in Physical Systems), Vilnius, 1979, pp. 66–89.

<sup>5)</sup>E. C. Jelks, R. M. Walsler, R. W. Bene, and W. H. Neal, *Appl. Phys. Lett.* **26**, 355 (1975).

<sup>6)</sup>A. A. Kokin and G. B. Mikhailov, *Fiz. Tverd. Tela (Leningrad)* **18**, 3384 (1976) [*Sov. Phys. Solid State* **18**, 1970 (1976)].

<sup>7)</sup>A. Alekseyunas, V. Bareikis, V. Bondarenko, and Yu. Lib-

eris, *Fiz. Tverd. Tela (Leningrad)* **20**, 1980 (1978) [*Sov. Phys. Solid State* **20**, 1143 (1978)].

<sup>8)</sup>K. Nagasawa, *Mater. Res. Bull.* **6**, 853 (1971).

<sup>9)</sup>F. N. Hooge, *Phys. Lett. A* **29**, 139 (1969).

<sup>10)</sup>F. N. Hooge, *Physica (Utrecht)* **60**, 130 (1972).

<sup>11)</sup>V. P. Palenskis and Z. L. Shoblitskas, *Tezisy dokladov II konferentsii po fluktuatsionnym yavleniyam v fizicheskikh sistemakh* (Abstracts of Papers presented at Second Conf. on Fluctuation Phenomena in Physical Systems), Vilnius, 1979, p. 70.

<sup>12)</sup>W. H. Rosevear and W. Paul, *Phys. Rev. B* **7**, 2109 (1973).

<sup>13)</sup>A. A. Bugaev, B. P. Zakharchenya, and F. A. Chudnovskii, *Fazovyĭ perekhod metall-poluprovodnik i ego primeneniye* (Metal-Semiconductor Phase Transition and Its Applications), Nauka, M., 1979.

<sup>14)</sup>W. Brückner, U. Gerlach, W. Moldenhauer, H. P. Brückner, B. Thuss, H. Oppermann, E. Wolf, and I. Storbeck, *J. Phys. (Paris)* **37**, Colloq. 4, C4-63 (1976).

<sup>15)</sup>M. Marezio, D. B. McWhan, J. P. Remeika, and P. D. Dernier, *Phys. Rev. B* **5**, 2541 (1972).

<sup>16)</sup>J. M. Reyes, S. L. Segel, and M. Sayer, *Can. J. Phys.* **54**, 1 (1976).

<sup>17)</sup>T. G. Lanskaya, I. A. Merkulov, and F. A. Chudnovskii, *Fiz. Tverd. Tela (Leningrad)* **20**, 336 (1978) [*Sov. Phys. Solid State* **20**, 193 (1978)].

<sup>18)</sup>A. Alekseyunas, V. A. Bareikis, V. M. Bondarenko, Yu. S. Liberis, V. I. Miller, V. A. Perelyaev, and G. P. Shveikin, *Tezisy dokladov II Vsesoyuznogo soveshchaniya po khimii tverdogo tela* (Abstracts of Papers presented at Second All-Union Conf. on Solid-State Chemistry), Sverdlovsk, 1978.

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## Theory of tunnel relaxation in a photon field

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The subbarrier relaxation of a particle located in a nonsymmetric double-well potential is considered. The phonon subsystem plays the role of a thermostat. It is shown that the tunnel kinetics of the particle is characterized by two relaxation times. The relaxation of the occupation numbers is proportional to the square of the overlap integral, whereas the relaxation of the phase correlation is of zero order with respect to the overlap integral and generally proceeds at a faster rate than the relaxation of the occupation numbers. The relative roles of two-phonon and one-phonon relaxation mechanisms are elucidated. Experiments on sound absorption in amorphous media and on tunnel diffusion of mesons are discussed within the framework of the proposed model.

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### 1. INTRODUCTION

In recent years, a whole group of problems has arisen which can in pattern be reduced to sub-barrier tunneling between two potential wells with slightly displaced energy levels, in the presence of an arbitrary interaction with the phonon field. This group includes quantum diffusion of light particles in crystals with defects, the class of low-frequency excitations in amorphous media, orientational transitions, and so on. A characteristic feature of these problems is the presence of a small parameter, such as the overlap integral  $J$  of the wave functions belonging to states in different wells. This

leads to the result that two-well tunnel kinetics differs in principle from the well-known picture of relaxation of two-level systems (spin relaxation). As will be shown below, even in the presence of only one dissipation mechanism—the interaction of the “particle” with the phonon field—a property of two-well kinetics is the simultaneous presence of fast and slow relaxations that differ strongly in magnitude. Using the language of spin kinetics and introducing approximately the concept of longitudinal  $T_1$  and transverse  $T_2$  relaxation times, we can verify that the relaxation of the occupation numbers (longitudinal relaxation) is due to transitions from one well to the other, and the probability of this transition is pro-

portional to  $J^2$ . At the same time, the relaxation of phase correlation of the states (transverse relaxation) is due to phase randomization that takes place in each well separately, and the rate of this process is not connected with tunneling and remains finite even as  $J \rightarrow 0$ .

What has been said above leads to a number of consequences. First, it is clear that the approximation of a single relaxation time, which is usually made in the theory of two-well tunnel kinetics (see, for example, Ref. 1), is not adequate for the problem in the general case. Second, the premise that the presence of widely differing relaxation times  $T_1$  and  $T_2$  ( $T_1 \gg T_2$ ) can be explained only by taking into account the interaction between different pairs of wells (spin-spin interaction) is not true; see, for example, Refs. 2 and 3. It is important that in the case of tunnel relaxation this is already a feature, in principle, of a single isolated pair of wells. Finally, we note that, as will be shown in what follows, the longitudinal relaxation, or the jump of the particle from one well to another, does not reduce in the case of a small distance  $\xi$  between levels simply to a direct inelastic transition but, as also in the case of quantum diffusion,<sup>4,5</sup> is determined by two contributions. Along with transitions that are accompanied directly by excitation of the phonon system (noncoherent diffusion), there exist transitions characterized by purely elastic coherent tunneling into a state that is fluctuating because of interaction with phonons in a single well (coherent diffusion). The second mechanism, which again depends on the damping of the nondiagonal density matrix elements (transverse relaxation) can be predominant at small  $\xi$  over a wide range of the parameters.

The purpose of the present work is the analysis of the picture of two-well tunnel relaxation as a function of the distance between levels and of the temperature  $T$ . We shall consider only one pair of nearby levels in neighboring wells, assuming that the separation between levels in a individual well, which is determined actually by the scale of the zero-point oscillations  $\omega_0$  is large enough so that

$$|\xi| \ll \omega_0; \quad (1.1)$$

here and below,  $\hbar = 1$ .

For the problem under consideration, it is most natural to specify the parameters in the representation of mutually orthogonal functions which correspond to motion of the particle only in one ( $\psi_l$ ) or the other ( $\psi_r$ ) well. In this representation, the Hamiltonian of the particle can be written in the form

$$H = \frac{1}{2}(\xi\sigma_x + \Delta\sigma_z), \quad (1.2)$$

where  $\Delta = 2H_{lr} = \omega_0 J$  (basically, this is the definition of the overlap integral  $J$ ).

The connection between the representation of the eigenfunctions (1.2) of the Hamiltonian (1.2) and the ( $l, r$ ) representation has the well-known form ( $\xi \geq 0$ )

$$\begin{aligned} |1\rangle &= u|l\rangle + v|r\rangle, & E_1 &= \frac{1}{2}\epsilon, \\ |2\rangle &= -v|l\rangle + u|r\rangle, & E_2 &= -\frac{1}{2}\epsilon, \end{aligned} \quad (1.3)$$

$$\begin{aligned} \epsilon &= (\xi^2 + \Delta^2)^{1/2}, & u &= [\frac{1}{2}(1 + \xi/\epsilon)]^{1/2}, & v &= [\frac{1}{2}(1 - \xi/\epsilon)]^{1/2}, \\ C &= u^2 - v^2 = \xi/\epsilon, & S &= 2uv = \Delta/\epsilon. \end{aligned}$$

If we change the level spacing  $\xi$ , for example, by

means of an external field, then the form of the functions  $|l\rangle$  and  $|r\rangle$  is practically unchanged. As a consequence, the nondiagonal matrix elements  $H_{lr}$  and the matrix elements of interaction of the particle with the oscillations of the medium  $V_{lr}, V_{ll}, V_{rr}$  remain unchanged. On the other hand, upon a change in  $\xi$  (if  $\xi$  is not too large in comparison with  $\Delta$ ), the eigenfunctions  $|1\rangle, |2\rangle$  change markedly. This leads to a strong dependence of the matrix elements of the interaction on the level spacing:

$$V_{12} = V_{21} = \frac{1}{2}SV_- + CV_{lr}, \quad (1.4)$$

$$V_{22} - V_{11} = CV_- - 2SV_{lr}, \quad V_- = V_{rr} - V_{ll}$$

(for definiteness, the functions  $|l\rangle$  and  $|r\rangle$  are chosen to be real).

The interaction matrix elements in the ( $l, r$ ) representation have a different order of magnitude relative to the overlap integral:

$$V_{lr} \sim J, \quad V_{ll} \sim J^0, \quad V_{rr} \sim J^0. \quad (1.5)$$

This relation is preserved in the representation (1.2) only if  $\Delta \ll \xi$ . If  $\Delta$  and  $\xi$  are comparable, then the matrix elements  $V_{12}$  and  $V_{11}$  have the same order of magnitude.

This very significant circumstance demonstrates the definite advantage of the ( $l, r$ ) representation, especially in the case of an approximate analysis of the relaxation picture. We shall be able below to demonstrate this lucidly.

## 2. KINETIC EQUATION

For the description of the interwell kinetics under the conditions of interaction with the phonon field, it is necessary to introduce a more general kinetic equation for the density matrix of the tunneling particle. A number of methods can be used to obtain such an equation. We note here, very briefly, one such derivation.

The equation for the density matrix  $\rho$  of the entire system as a whole has the standard form

$$\partial\rho / \partial t + i[H + H_{ph} + V, \rho] = 0. \quad (2.1)$$

Here  $H$  has the value (1.2),  $H_{ph}$  and  $V$  are respectively the Hamiltonians of the phonon subsystem and of the interaction of the particle with the oscillations of the medium. Using the formal integration of this equation with respect to time for the equation of evolution of the density matrix of the particle

$$f = \text{Sp}_{ph}\rho,$$

we find directly

$$\partial f / \partial t + i[H, f] + J = 0, \quad (2.2)$$

where the collision integral  $J$  has the following form:

$$J = \text{Sp}_{ph} \left\{ \left[ V, \int_0^t d\tau e^{-iH_0\tau} [V, \rho(t-\tau)] e^{iH_0\tau} \right] \right\} \quad (2.3)$$

$$H_0 = H + H_{ph}.$$

In these expressions,  $\text{Sp}_{ph}$  represents summation over the states of the phonon subsystem.

To obtain the entire picture of the kinetics, it suffices to take into account the interaction of the particle with the medium in the Born approximation. We can then

neglect [under the integral sign in (2.3)] the correlations between the states of the particle and the phonon subsystem, which is equivalent to the approximation

$$\rho(t) \approx f(t) \rho_{ph}, \quad (2.4)$$

where  $\rho_{ph}$  is the density matrix of the phonons at thermodynamic equilibrium. As a result, Eq. (2.2) becomes closed relative to the density matrix  $f$ .

We now make some remarks. The approximation leading to (2.4) does not impose any actual restrictions on the relation between the magnitude of the interaction of the particle with the phonons and the level space  $\epsilon$ . If the interaction of the particle in an individual well with the medium is sufficiently strong, it is necessary to take the polaron effect into account from the very beginning, and to carry out renormalization of the levels and of the matrix element  $H_{lr}$  of coherent coupling between the wells.

Thus, in the general case the values of the parameters  $\xi$  and  $\Delta$  in (1.2) must imply an explicit account of the polaron renormalization. After this, the interaction with the phonons in the individual well at the new equilibrium position of the particle ( $V_{ll}$ ,  $V_{rr}$ ) can be regarded as weak, while the "jolting" matrix element  $V_{lr}$  is already small because of the fact that it is proportional to the overlap integral [see (1.5)].

It is convenient to solve the general problem of relaxation by the Laplace method. We insert (2.4) in (2.3). Then, Eq. (2.2) gives

$$p f(p) + i[H, f(p)] + J(p) = f(0), \quad (2.2')$$

for the Laplace transform of the density matrix of the particle  $f(p)$ : Here  $f(0)$  is the initial distribution. The Laplace transform of the collision integral operator has in this case the form

$$J(p) = S p_{ph} \left\{ \left[ V, \frac{1}{p+iL} [V, f(p) \rho_{ph}] \right] \right\}. \quad (2.5)$$

For the sake of brevity, a notation is introduced, the sense of which is clear from the definition of the matrix element in the representation of the eigenfunctions of the Hamiltonian  $H_0$ :

$$\left( \frac{1}{p+iL} A \right)_{ik} = \frac{1}{p+i(E_i-E_k)} A_{ik}, \quad (2.6)$$

where  $E_i = E_a + E_\alpha$  are the values of the Hamiltonian  $H_0$  ( $A=1, 2$ ;  $E_\alpha$  is the energy of the phonon system).

The matrix elements of the collision integral can be written in the form

$$J_{cd} = \sum_a \Omega_{ab}^{cd} f_{ab}. \quad (2.7)$$

The direct transformation (2.5) leads in the representation (1.2) to the following expression for the matrix elements of the superoperator  $\Omega$ :

$$\Omega_{ab}^{cd} = S p_{ph} \left\{ \rho_{ph} \langle b | \left[ \frac{1}{p-iL} [ |d\rangle \langle c |, V ], V \right] | a \rangle \right\}. \quad (2.8)$$

The relaxation of the particle is determined by the roots of the homogeneous part of Eq. (2.2') (a system of three independent algebraic equations, defined for the matrix elements  $f_{ab}$ ); In the zeroth approximation, neglecting the interaction with the medium, these roots are equal to

$$p^{(0)} = -i\omega, \quad \omega_{2,3} = \pm \epsilon, \quad \omega_1 = 0. \quad (2.9)$$

The exact values of the roots are shifted from these values in the complex region by a distance of the order of  $V^2$ . Therefore, in the calculation of the matrix elements (2.8), we set

$$[p - i(E_l - E_k)]^{-1} = \pi \delta(\omega + E_l - E_k), \quad (2.10)$$

neglecting by the same token a certain finite width in the conservation law in the integration over the phonon variable in (2.8). The imaginary part is omitted in the right hand side of (2.10); as usual, this part leads to renormalization of the energy levels.

Making use of Eq. (2.10), we can write down in explicit form the expression for the coefficients (2.8):

$$\Omega_{ab}^{cd} = \delta_{bd} \pi \sum_{\alpha\beta\gamma} \rho_\alpha V_{c\gamma}^{\alpha\beta} V_{\alpha\beta}^{\gamma a} \delta(\omega + E_{b\gamma}^{\alpha\beta}) + \delta_{ca} \pi \sum_{\alpha\beta\gamma} \rho_\alpha V_{b\gamma}^{\alpha\beta} V_{\alpha\beta}^{\gamma d} \delta(\omega + E_{a\gamma}^{\beta\alpha}) - \pi \sum_{\alpha\beta} \rho_\alpha V_{bd}^{\alpha\beta} V_{ca}^{\beta\alpha} \{ \delta(\omega + E_{bc}^{\alpha\beta}) + \delta(\omega + E_{da}^{\beta\alpha}) \}; \quad (2.11)$$

here

$$\rho_\alpha = (\rho_{ph})_{\alpha\alpha}, \quad E_{ab}^{\alpha\beta} = (E_a + E_\alpha) - (E_b + E_\beta).$$

In the following, we shall be interested in two-well kinetics under the condition

$$\epsilon \ll \omega_D, \quad (2.12)$$

where  $\omega_D$  is the characteristic frequency of the phonon spectrum.

In this case, the two-phonon (in the general case—multiphonon) processes begin to predominate over the one-phonon ones already at sufficiently low temperatures:

$$T^* < T \ll \omega_D, \quad (2.13)$$

this being due to the low density of the phonon states at the low frequencies. (For the definition of  $T^*$  see Sec. 4.) If we assume simultaneously that

$$\epsilon \ll T, \quad (2.14)$$

then it is easy to understand that we can neglect the quantities  $\omega$  and  $\epsilon$  in the  $\delta$  functions in (2.11) over the entire temperature range (2.13). In this case, the form of the expression (2.11), as also the of initial expression (2.8), becomes invariant relative to the choice of representation for the states of the particles and, in particular, it can be used also for the ( $l$ ,  $r$ ) representation. The fact that in this case expression (2.8) is independent of  $p$  makes the relaxation process purely Markovian.

We note that under the conditions (2.13) and (2.14) we can neglect  $H$  in the Hamiltonian  $H_0$  in the initial collision integral (2.3) and, keeping it in mind that we are interested in those times  $t$  for which  $tT \gg 1$ , we can let the upper limit of integration in time become infinite. Recognizing that in the chosen approximation

$$f(t-\tau) \approx e^{iH\tau} f(t) e^{-iH\tau},$$

we can simultaneously substitute  $f(t-\tau) \rightarrow f(t)$ . As a result, we obtain the kinetic equation which was used earlier in the analysis of the quantum diffusion of particles in a crystal.<sup>4,5</sup>

Keeping it in mind that all the results turn out to be most lucid in the temperature range in which the conditions (2.13) and (2.14) are valid, we begin the analysis with precisely this case.

### 3. TWO-WELL TUNNEL RELAXATION AT $T \gg \varepsilon$ AND $T > T^*$

In correspondence with the arguments advanced in the preceding section, we shall take into account only the two-phonon excitations in the studied temperature range. In the  $(l, r)$  representation, the operator equation (2.2') with the condition integral (2.7), (2.11) can, neglecting quantities of the order of  $\varepsilon$  in the  $\delta$  functions in (2.11), be rewritten in the following explicit form:

$$\begin{aligned} p\chi_l + i/2\Delta(f_{rl} - f_{lr}) + 2W\chi_l + A(f_{rl} + f_{lr}) &= \chi_l(0), \\ pf_{lr} + i\xi f_{lr} - i\Delta\chi_l + \Omega f_{lr} + W(f_{lr} - f_{rl}) + 2A\chi_l &= f_{lr}(0), \\ pf_{rl} - i\xi f_{rl} + i\Delta\chi_l + \Omega f_{rl} - W(f_{lr} - f_{rl}) + 2A\chi_l &= f_{rl}(0). \end{aligned} \quad (3.1)$$

Here  $\chi_l = -\chi_r = f_{ll} - f_{ll}^{(0)}$ ,  $f_{ll}^{(0)}$  is the equilibrium density matrix ( $f_{ll}^{(0)} = f_{rr}^{(0)} = \frac{1}{2}$ );

$$\begin{aligned} \Omega &= \pi \sum_{\alpha, \beta} \rho_{\alpha} |V_{\alpha\beta}|^2 \delta(E_{\alpha} - E_{\beta}), \\ W &= 2\pi \sum_{\alpha, \beta} \rho_{\alpha} |V_{lr\alpha\beta}|^2 \delta(E_{\alpha} - E_{\beta}), \\ A &= \pi \sum_{\alpha, \beta} \rho_{\alpha} V_{lr\alpha\beta} V_{\alpha\beta} \delta(E_{\alpha} - E_{\beta}) \end{aligned} \quad (3.2)$$

[see the notation in (1.4)].

The roots of the homogeneous system in (3.1) are determined by the cubic equation

$$p^3 + 2a_2 p^2 + a_1 p + a_0 = 0, \quad (3.3)$$

where

$$\begin{aligned} a_2 &= \Omega + 2W, & a_1 &= a_2^2 + \varepsilon^2 + 2(W\Omega - 2A^2), \\ a_0 &= \Omega(\Delta^2 - 4A^2) + 4A\xi\Delta + 2W[\xi^2 + \Omega(\Omega + 2W) - 4A^2]. \end{aligned} \quad (3.4)$$

We now make use of the smallness of the overlap integral  $J$ . In correspondence with (1.5), there exists the following relation between the coefficients of (3.2):

$$\Omega \sim J^0, \quad W \approx \Omega J^2, \quad A \approx \Omega J. \quad (3.5)$$

Taking (1.1) into account and the similar inequality  $\Omega \ll \omega_0$ , which is always assumed implicitly to be satisfied, and with account of (3.5), we can materially simplify the expression for the coefficients (3.4); in particular, we can neglect all terms containing  $A$  in the formulas (3.4).

Analyzing Eq. (3.3), we can easily establish the fact that at both  $\Omega \gg \Delta$  and  $\Omega \ll \Delta$ , one of the roots ( $p_1$ ) is always small in comparison with the two others. The value of this purely real root is obtained directly from the relation

$$-p_1 = \gamma_1 \approx a_0/a_1. \quad (3.6)$$

Discarding the small terms in the coefficients of (3.4), we have

$$\gamma_1 \approx \frac{\Delta^2 \Omega}{\varepsilon^2 + \Omega^2} + 2W \left(1 - \frac{\Delta^2}{\varepsilon^2 + \Omega^2}\right). \quad (3.7)$$

The found root describes the slow relaxation of the nonequilibrium distribution to the equilibrium state. In terms of spin kinetics,  $\gamma$ , represents the characteristic time of longitudinal relaxation.

The remaining two complex conjugate roots of Eq.

(3.3) are now found directly:

$$p_{2,3} \approx \pm i\varepsilon - \gamma_2, \quad \gamma_2 \approx \Omega \left(1 - \frac{1}{2} \frac{\Delta^2}{\varepsilon^2 + \Omega^2}\right) + W \left(1 + \frac{\Delta^2}{\varepsilon^2 + \Omega^2}\right). \quad (3.8)$$

The second term in  $\gamma_2$  is small in comparison with the first. However, we shall retain it in this expression for convenience in comparison with the results of the next section. The general form of the dependence of the density matrix on the time can be represented in the following form in this case:

$$f_{\alpha\beta}(t) = C_{\alpha\beta}^{(1)} e^{-\gamma_1 t} + C_{\alpha\beta}^{(2)} e^{+i\omega_0 t - \gamma_2 t} + C_{\alpha\beta}^{(3)} e^{-i\omega_0 t - \gamma_2 t}. \quad (3.9)$$

The oscillating part of the density matrix is damped with frequency  $\gamma_2$ . It is easily understood that the resonance at the frequency  $\omega = \varepsilon$  in an alternating external field will have a width equal to  $\gamma_2$ . Thus  $\gamma_2$  plays the role of the transverse relaxation time.

It is significant that the frequencies of longitudinal and transverse relaxations in the considered temperature range have a different order of magnitude relative to the overlap integral [see (3.7) and (3.8) with account of (3.5)]. The transverse relaxation takes place in the general case more rapidly than the longitudinal relaxation. This occurs over a wide range of parameters, so long as one of the inequalities below is valid:

$$\Delta \ll \varepsilon, \quad \Delta \ll \Omega. \quad (3.10)$$

The second inequality should be specially noted, in spite of the independence of  $\Omega$  from  $J$ , because of the strong falloff of  $\Omega$  with decrease in the temperature. In the case of two-phonon processes,

$$\Omega = \zeta \omega_0 (T/\omega_0)^n, \quad (3.11)$$

where  $n = 7$ , if both wells are different, and  $n = 9$  if the wells are practically identical (see, for example, Refs. 4 and 5).

Thus, two-well tunnel kinetics in the considered region is characterized by the presence of two widely differing longitudinal and transverse relaxation times. It is important that this takes place for an isolated pair of wells.

The slow-relaxation decrement (3.7), which actually determines the frequency of migration of the particle from one well to the other, consists of two terms. In complete analogy with quantum diffusion in a crystal (Refs. 4 and 5), the first term represents coherent migration, and the second the contribution from purely noncoherent transition between the wells. It is of interest that, taking (3.5) into account, we obtain for the ratio of the noncoherent and coherent contributions in (3.7)

$$(\Omega^2 + \varepsilon^2)/\omega_0^2 \ll 1. \quad (3.12)$$

It then follows that the jump from well to well in two-well tunnel kinetics is determined primarily by the coherent process.

It can be concluded from the form of (3.7) that the coherent contribution to the longitudinal relaxation at  $\gg \Delta$  can have a peculiar form. At low temperatures, the transition probability increases strongly with  $T$ , goes through a maximum, and then begins to fall off

sharply. Both slopes have the same temperature dependence (3.11). It is of interest that exactly the same behavior was actually observed in experiments on the diffusion of  $\mu^+$  mesons in crystals (see the corresponding analysis in Ref. 6).

We note that in the case of a significant increase in the temperature, the inequality (3.12) does not hold. This is connected with the fact that  $\omega_0$  is really multiplied by the polaron pre-exponential factor, which falls off with increase in  $T$ . Moreover, the sub-barrier classical jumping sets in. As a result, noncoherent contribution becomes predominant and  $\gamma_1$  again increases with increase in  $T$ . The temperature picture of the behavior of the longitudinal relaxation can be greatly transformed as a function of the relation between the parameters, especially of the value of  $\varepsilon$ .

As is seen from the expressions (3.7) and (3.8), account of the cross terms in the collision integral, which are proportional to  $A$  [see (3.1)], turned out to be unimportant, although it was not possible to exclude them in advance in accord with the degree of the overlap integral. However, these terms can be omitted only in the  $(l, r)$  representation. In the representation (1.2), in the case  $\xi \sim \Delta$ , all the coefficients of (2.11) lead to terms of the same order in the kinetic equation. If we use the well-known approximation of two relaxation times (for the longitudinal and transverse components of the density matrix) in this representation from the very beginning, then we get erroneous results in the general case. In view of the nontriviality of this assertion, we shall consider it in some detail.

In the representation (1.2) in the considered temperature region (2.13), (2.14), the kinetic equation reduces to the system (3.1), (3.2) if we replace the indices  $l$  and  $r$  in the latter by 1 and 2, and set

$$\Delta=0, \quad \xi \rightarrow \varepsilon. \quad (3.13)$$

If we use the transformation (1.4), then it is easy to establish the connection between the kinetic coefficients in the new (superior bar) and old representations:

$$\begin{aligned} \bar{\Omega} &= C^2\Omega + 2S^2W - 4CSA, \\ \bar{W} &= 1/2S^2\Omega + C^2\bar{W} + 2CSA, \\ \bar{A} &= 1/2CS\Omega - CSW + (C^2 - S^2)A. \end{aligned} \quad (3.14)$$

Again finding for the determination of the roots a cubic equation, similar to (3.3), we can easily establish the fact that its coefficients are identical with those of (3.4).

In the two-relaxation-time approximation, which is equivalent to the Bloch equation, we have the following simplified system in place of (3.1) [with account of (3.13)]:

$$\begin{aligned} p\chi_1 + 2\bar{W}\chi_1 &= \chi_1(0), \\ pf_{12} + ie f_{12} + \bar{\Omega} f_{12} &= f_{12}(0), \\ pf_{21} - ie f_{21} + \bar{\Omega} f_{21} &= f_{21}(0). \end{aligned} \quad (3.15)$$

From this system, it is easy to find

$$p_1 = -2\bar{W}, \quad p_{2,3} = \pm ie - \bar{\Omega}. \quad (3.16)$$

Let  $\varepsilon \sim \Delta$ . Then, taking (3.14) into consideration, we have

$$p_1 \approx -\Omega, \quad \text{Re } p_{2,3} \approx -[2W(\Delta/\varepsilon)^2 + (\xi/\varepsilon)^2\Omega],$$

i.e., a result certainly known to be incorrect. Only in the limit  $\varepsilon \gg \Delta, \Omega$  do expressions (3.16) go over to the correct result (3.7), (3.8).

Thus, in the analysis of tunnel relaxation in the ordinary representation (1.2), the two relaxation time approximation in the equation for the density matrix turns out to be inadequate and it is necessary to make use of the exact matrix of the collision integral.

#### 4. TWO-WELL TUNNEL RELAXATION AT $T < T^*$

In the previous section we considered the temperature region  $T > T^*$ , in which the decisive role in kinetics is played by two-phonon processes. With account of (2.14), the scattering of phonons here has a purely elastic character, and the elements of the matrix of kinetic coefficients (2.11) contains only delta functions of the form  $\delta(E_\alpha - E_\beta)$  [see (3.2)]. In the case of preservation of the inequality (2.14), this result is preserved for two-phonon processes and for any arbitrary reduction of temperature. Now, however, inelastic and in particular single-phonon processes can become important. Successive inclusion of such processes in the kinetics requires real values of the arguments of the delta functions in (2.11), which in turn presupposes the use of the representation (1.2).

We estimate the characteristic temperature  $T^*$  at which the single-phonon processes begin to play an important role. For this purpose, we return to the general expression (2.11). It is not difficult using the transformation (1.4) for the matrix elements of the interaction, to ascertain that now the kinetic coefficients, together with terms of the form (3.2), will contain similar terms, but with the substitution  $\delta(E_\alpha - E_\beta) \rightarrow \delta(E_\alpha - E_\beta \pm \varepsilon)$ . For example, in the case of the single-phonon transition, calculating an expression of the form

$$\Omega^{(1)} = \pi \sum_{\alpha\beta} \rho_\alpha |V_{-\alpha\beta}|^2 \delta(E_\alpha - E_\beta \pm \varepsilon)$$

under the assumption that  $T \gg \varepsilon$ , we find

$$\Omega^{(1)} \approx \zeta^{(1)} \omega^{(1)} (\varepsilon/\omega_D)^m T/\omega_D, \quad (4.1)$$

where  $m=2$  if the wells are different, and  $m=4$  in the case of identical wells. The energy parameter  $\omega^{(1)}$  in this expression has the scale of the depth of the well.

We now compare (4.1) with the analogous expression for the two-phonon processes (3.11). Assuming that the wells are not identical we obtain for the temperature  $T^*$  at which case both expressions become equal in value

$$T^* \approx \left( \frac{\zeta^{(1)} \omega^{(1)}}{\zeta} \frac{\omega^{(1)}}{\omega_D} \right)^{1/4} \left( \frac{\varepsilon}{\omega_D} \right)^{1/4} \omega_D. \quad (4.2)$$

The inequality  $T^* \ll \omega_D$  follows from (4.2) by virtue of (2.12) [the first factor in (4.2) is not greatly different from unity; we note that the factors  $\omega^{(1)}/\omega_D \gg 1$  and  $\zeta^{(1)}/\zeta \ll 1$  frequently cancel one another]. At the same time, the weak dependence on the small parameter  $\varepsilon/\omega_D$  (of degree  $1/3$ ) leads to the result that the value of  $T^*$ , which itself depends on  $\varepsilon$ , practically always satisfies the relation

$$T^* \gg \varepsilon, \quad (4.3)$$

therefore, in particular, at  $T \approx T^*$  the assumption used

in the calculation of (4.1) is valid.

An entirely analogous estimate is obtained in the determination of  $T^*$  from a comparison of the single-phonon and two-phonon contributions to other coefficients. It should be noted that when the other matrix elements of the interaction are used [see the expressions for  $W$  and  $A$  in (3.2)] then, in the general case, the temperature dependence of the two-phonon processes is identical with (3.11) with  $n=7$ , and of the single-phonon ones, with (4.1) with  $m=2$ .

At  $T \leq T^*$ , if we assume that

$$(\varepsilon/\omega_D)^{1/2} \ll \omega_D/\omega^{(1)},$$

we find the following directly from (4.1)

$$\Omega^{(1)} \ll \varepsilon. \quad (4.4)$$

It is easy to understand that the inequality actually carries over to all elements of the matrix of kinetic coefficients (2.11), i.e.,

$$|\Omega_{\alpha\beta}^{st}| \ll \varepsilon. \quad (4.5)$$

In the representation (1.2) the damping decrements are determined by the roots of the equation

$$0 = \begin{vmatrix} p - \Omega_{11}^{11} - \Omega_{22}^{11} & \Omega_{12}^{11} & \Omega_{21}^{11} \\ \Omega_{11}^{12} - \Omega_{22}^{12} & p + i\varepsilon + \Omega_{12}^{12} & \Omega_{21}^{12} \\ \Omega_{11}^{21} - \Omega_{22}^{21} & \Omega_{12}^{21} & p - i\varepsilon + \Omega_{21}^{21} \end{vmatrix}. \quad (4.6)$$

With account of the inequality (4.5), the determination of the roots of Eq. (4.6), with accuracy to terms that are linear in  $\Omega$ , becomes trivial:

$$-p_i = \Omega_{i1}^{11}(0) - \Omega_{i2}^{11}(0), \quad -p_{2,3} = \pm i\varepsilon + \Omega_{12}^{12}(\varepsilon). \quad (4.7)$$

Here we have used the general relation

$$\Omega_{21}^{21}(\omega) = \Omega_{12}^{12}(-\omega).$$

We substitute the values of the matrix elements of (2.11) in (4.7). For the damping decrements  $\gamma_1$  and  $\gamma_2$  [see the definitions (3.6) and (3.8)] we find directly

$$\gamma_1 = 2\pi \sum_{\alpha\beta} \rho_{\alpha} |V_{12}^{\alpha\beta}|^2 [\delta(E_{\alpha\beta} + \varepsilon) + \delta(E_{\alpha\beta} - \varepsilon)], \quad (4.8)$$

$$\gamma_2 = \pi \sum_{\alpha\beta} \rho_{\alpha} |V_{11}^{\alpha\beta} - V_{22}^{\alpha\beta}|^2 \delta(E_{\alpha\beta}) + 1/2 \gamma_1.$$

We transform the matrix elements in this representation to the  $(l, r)$  representation, using the relations (1.4). As a result, we get

$$\gamma_1 \approx \frac{\Delta^2}{\varepsilon^2} \Omega_i + 2 \frac{\xi^2}{\varepsilon^2} W_i, \quad (4.9)$$

$$\gamma_2 \approx \frac{\xi^2}{\varepsilon^2} \Omega + 2 \frac{\Delta^2}{\varepsilon^2} W + \frac{1}{2} \gamma_1;$$

here

$$\Omega_i \approx \pi \sum_{\alpha\beta} \rho_{\alpha} |V_{-}^{\alpha\beta}|^2 1/2 [\delta(E_{\alpha\beta} + \varepsilon) + \delta(E_{\alpha\beta} - \varepsilon)],$$

$$W_i \approx 2\pi \sum_{\alpha\beta} \rho_{\alpha} |V_{i+}^{\alpha\beta}|^2 1/2 [\delta(E_{\alpha\beta} + \varepsilon) + \delta(E_{\alpha\beta} - \varepsilon)]. \quad (4.10)$$

The expressions for  $\Omega$  and  $W$  without the index  $i$  have as before the values (3.2), which correspond to the purely elastic case and therefore are determined only by the two-phonon processes. The expressions (4.10) already contain a contribution from both the two-phonon and the single-phonon processes.

It is significant that the temperature ranges where the expressions (4.9) and (3.7), (3.8) are valid, overlap. Actually, by virtue of the inequality (4.3), the expressions (3.7) and (3.8) remain valid right up to  $T \approx T^*$ . By reason of (4.5), the result (4.9), which is obtained only by use of this inequality, is preserved even in a certain region of temperatures above  $T^*$ . In this region the single-phonon processes can be neglected, and  $\Omega_i$  and  $W_i$  coincide with the values  $\Omega$  and  $W$ , respectively. Here, as is easily seen, with account of the condition  $\Omega \ll \varepsilon$ , the expression (3.7) is identical with  $\gamma_1$  in (4.9), and (3.8) with  $\gamma_2$  (4.9).

The existence of a range of temperatures in which both the result (4.9) and the expressions (3.8) and (3.9) are valid simultaneously, permits us to write down single formulas for  $\gamma_1$  and  $\gamma_2$ , which encompass the entire range of temperatures:

$$\gamma_1 \approx \frac{\Delta^2}{\varepsilon^2 + \Omega^2} \Omega_i + 2 \left(1 - \frac{\Delta^2}{\varepsilon^2 + \Omega^2}\right) W_i, \quad (4.11)$$

$$\gamma_2 \approx \left(1 - \frac{\Delta^2}{\varepsilon^2 + \Omega^2}\right) \Omega + 2 \frac{\Delta^2}{\varepsilon^2 + \Omega^2} W + \frac{1}{2} \gamma_1.$$

These expressions completely solve the problem of two-well tunnel kinetics in the case of an arbitrary relation among the parameters  $\Delta$ ,  $\varepsilon$  and  $T$ . The time dependence for any particular problem is determined here by a relation of the form (3.9) with account of the concrete initial conditions.

At  $T < T^*$ , the quantities  $\Omega_i$  and  $W_i$  (4.10) are determined by single-phonon processes and  $\gamma_1$  (4.9) depends on the temperature in linear fashion [see (4.1)]. At  $\varepsilon > \Delta$  the inequality

$$\gamma_2 > \gamma_1, \quad (4.12)$$

which was found in the previous section for  $T > T^*$ , is preserved at temperatures below  $T^*$  right down to the temperature  $T'$  where both roots become equal in value. In the range  $T' < T < T^*$ , the decrement falls off with decrease in temperature more rapidly than  $\gamma_1$  according to the law (3.11), since it is determined as before by the two-phonon processes [the first term in  $\gamma_2$  (4.9)].

Taking into account the analysis given in the previous section in the estimate of  $T'$ , we can keep only the first term in the expression for  $\gamma_1$  (4.9). As a result, we have

$$T' \approx T^*(\varepsilon) (\Delta/\varepsilon)^{1/2} \approx T^*(\Delta). \quad (4.13)$$

We note that  $T'$  does not depend on  $\varepsilon$ .

Thus, the inequality (4.12) at  $\varepsilon \gg \Delta$  is preserved over the entire temperature range  $T > T'$ , while the ratio  $\gamma_2/\gamma_1$  increases with increase in  $T$  in the range  $(T', T^*)$ , reaches a plateau, and then again begins to increase.

At  $T < T'$ , both decrements already depend linearly on the temperature and the ratio  $\gamma_2/\gamma_1$  reaches a constant equal to  $1/2$ .

At  $\varepsilon \sim \Delta$ , the decrements  $\gamma_1$  and  $\gamma_2$  remain of the same order in value at low temperatures, so long as  $T$  does not become greater than some value  $T''$  ( $T'' > T'$ ) determined from the condition  $\Omega(T'') \approx \Delta$ . Taking (3.11) into account, we have

$$T'' \approx \omega_D (\Delta/\xi\omega_D)^{1/2}. \quad (4.14)$$

At  $T > T''$ , the inequality (4.12) again holds, while the ratio  $\gamma_2/\gamma_1$  will increase continuously with increase in  $T$ .

## 5. CONCLUDING REMARKS

1. The results indicate that the tunnel kinetics for a single pair of wells, located in a phonon field has as a characteristic the presence of two completely different relaxation times:  $T_1 = 1/\gamma_1$  and  $T_2 = 1/\gamma_2$  [see (4.11)]. From the physical point of view, these times correspond to the longitudinal and transverse relaxations and in this sense play a role analogous to  $T_1$  and  $T_2$  in the problem of spin relaxation. However, and this is important, the kinetic equation for the density matrix in the considered problem does not reduce to the Bloch equation in the general case, and the times that are found do not correspond to the relaxation times  $T_1$ ,  $T_2$  in the Bloch equation.

2. The nature of the phenomenon of two different relaxation times in the two-well problem is associated with the fact that, in contrast to the spin problem, the initial and final states are localized in different regions of space. As a consequence, the density-matrix elements that are nondiagonal in the index of the wells, are damped because of the difference of the instantaneous interaction with the phonons for both wells; this leads to randomization of the phases. This damping remains finite even upon approach of the overlap integral to zero, since the damping of the diagonal elements is always proportional to  $J^2$ . In the case of an isolated spin the interaction with the phonons actually leads to a single relaxation time in every case.

3. Over a wide range of temperatures  $T > T'$  (4.13), the longitudinal relaxation time is large in comparison with the transverse. At  $T < T'$  both times are comparable in value. In this region of extremely low temperatures,  $\gamma_1$  and  $\gamma_2$  have the same linear dependence on  $T$ . At  $T > T^*$  (4.2)  $\gamma_1$  and  $\gamma_2$  increase sharply with temperature, owing to the two-phonon nature of the relaxation in this region. In the intermediate range of temperatures  $T' < T < T^*$ ,  $\gamma_1$  begins to depend linearly on  $T$ , since the transverse relaxation is as before determined by two-phonon processes.

4. As is well known, the anomalous low-temperature behavior of amorphous media has its explanation in the framework of representations on the existence in such media of two-well configurations with uniform distribution of distances between levels in the wells and tunnel coupling between them (see Refs. 2, 8 and 1).

It is of interest that in experiments on sound absorp-

tion in such media, under conditions of resonance saturation, it has been discovered that the width of the resonance dip [in our problem it corresponds to the quantity  $\gamma_2$ , see the general expression for the density matrix (3.9)] is much greater than the reciprocal time of longitudinal relaxation, i.e.,  $\gamma_1$ , which is measured independently. Since the adopted description of the relaxation picture for an isolated pair of wells contains only a single relaxation time, then the explanation of the emergence of two sharply differing relaxation times requires the introduction of an interaction between the different pairs of wells ("spin-spin" relaxation, see, for example, Ref. 7). In fact, as the results of the present research show, there exists another relaxation mechanism leading to a sharp difference between  $\gamma_1$  and  $\gamma_2$ , and at the same time unconnected with the interaction between different pairs of wells. It would be very interesting to make clear both mechanisms of relaxation experimentally, by investigating the temperature and frequency dependences of the relaxation picture. The very limited experimental results that do exist obviously do not make clear the second relaxation mechanism. Actually, they rather indicate that  $1/T_2$  as well as  $1/T_1$  depend linearly on the temperature, since in the case of an isolated pair of wells, the difference between  $\gamma_1$  and  $\gamma_2$  appears only when two-phonon processes begin to play a role, with their stronger dependence on  $T$ , at least for  $1/T_2$ .

We note that the experiments on the sub-barrier tunnel diffusion of  $\mu^+$  mesons in crystals with defects at low temperatures on the other hand reveal a very strong temperature dependence for the interwell relaxation, which is equivalent precisely to the two-phonon mechanism (see Refs. 9 and 6). To be sure, this takes place at comparatively higher temperatures.

<sup>1</sup>S. Hunklinger and W. Arnold, *Phys. Acoustics*, **12**, 155 (1976).

<sup>2</sup>J. Joffrin and A. Levelut, *J. Physique* **36**, 811 (1965).

<sup>3</sup>J. L. Black and B. I. Halperin, *Phys. Rev.* **B16**, 2879 (1977).

<sup>4</sup>Yu. Kagan and L. A. Maksimov, *Zh. Eksp. Teor. Fiz.* **65**, 622 (1973) [*Sov. Phys. JETP* **38**, 307 (1974)].

<sup>5</sup>Yu. Kagan and M. I. Klinger, *J. Phys.* **C7**, 2791 (1974).

<sup>6</sup>Yu. Kagan, *J. Phys.* **C13**, 1928 (1980).

<sup>7</sup>P. W. Anderson, B. I. Halperin and C. M. Varma, *Phil. Mag.* **25**, 1 (1972).

<sup>8</sup>W. A. Philips, *J. Low Temp. Phys.* **7**, 351 (1972).

<sup>9</sup>V. A. Grebennik, I. I. Gurevich, V. A. Zhukov, A. I. Klimov, V. N. Maĭorov, A. P. Manych, E. V. Mel'nikova, B. A. Nikol'skiĭ, A. V. Pirogov, A. N. Ponomarev, V. I. Selivanov, and V. A. Suetin, *Pis'ma Zh. Eksp. Teor. Fiz.* **25**, 322 (1977) [*JETP Lett.* **25**, 298 (1977)].

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