Electronic polaron effect and the quantum diffusion of heavy particle in a metal

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A study is made of the polaron effect that arises during tunneling of a heavy particle in the electron fluid of a metal. The problem of adiabaticity in a metal is analyzed with a consistent allowance for the intra-well motion of the particle. That part of the perturbed electronic wave function which is associated with virtual excitations having an energy δE greater than the level separation ω in the well, is found to readjust itself adiabatically to the particle, whereas the other part, with $\delta E < \omega$, does not follow the particle but is directed toward the center of the well. The anti-adiabatic character of the second part of the intra-well wave function gives rise to a polaron effect which, however, is governed by the ratio T/ω rather than T/ε_F. In the presence of a relative shift ξ of the levels in adjacent wells the polaron effect decreases and is governed by the parameter ξ/ω. The problem of the tunneling jump of a heavy particle with shake-up is solved for an arbitrary relationship between ξ and T. The $\xi = 0$ and T less than the effective width Δ_* of the band, a diffusion under conditions of band-type motion arises and is characterized by a constant value of Δ_* and by an intraband scattering by electrons which are not involved in the polaron effect. This effect is manifested only in the size of Δ_*.

1. INTRODUCTION

A careful analysis of the motion of a heavy particle in a metal meets with substantial difficulties. On the one hand, the characteristic velocity of the electrons is large compared to that of the heavy particle, and this makes it natural to use the approximation of an adiabatic readjustment of the electrons to the instantaneous position of the particle. It might seem that there would be here an insignificant renormalization, of the order of m/M, to the mass M of the particle (m is the electron mass). On the other hand, even the very slow motion of a heavy particle in a degenerate Fermi fluid can give rise to electron-hole pairs of low energy, and this, in and of itself, contradicts the assumption of a complete adiabatic readjustment. Meanwhile, there is a class of phenomena for which the question of whether or not there is an adiabatic readjustment of the electrons to the moving heavy particle is of absolutely fundamental significance.

In connection with the quantum diffusion of a heavy particle in a metal, this problem was first pointed out by Kondo[1,2] (Ref. 2 contains a review of Kondo's more recent papers). Kondo considered the motion of a heavy particle in a narrow band arising in the periodic potential of a crystal in the tight-binding approximation with allowance for the interaction with the electron fluid. This interaction causes a renormalization of the tunneling matrix element of the transition from cell $\mathbf{l}$ to $\mathbf{l} + \mathbf{g}$; this renormalized matrix element, which determines the actual width of the band, is then obtained as

$$\tilde{\Delta}_0 = \Delta_0 \Lambda_{\mathbf{l},\, \mathbf{l}+\mathbf{g}}, \tag{1.1}$$

where

$$\Lambda_{\mathbf{l},\, \mathbf{l}+\mathbf{g}} = \langle \psi_{\mathbf{l}}(\mathbf{r}) | \psi_{\mathbf{l}+\mathbf{g}}(\mathbf{r}) \rangle \tag{1.2}$$

is the overlap integral of the electronic states corresponding to a fixed position of the particle in sites $\mathbf{l}$ and $\mathbf{l} + \mathbf{g}$, and Δ_0 is the tunneling matrix element corresponding to the original potential relief. The structure of expressions (1.1) and (1.2) is exactly the same as the familiar result for the ordinary small-radius polaron if $\Psi_{\mathbf{l}}$ is understood to be the phonon wave function.

Overlap integral (1.2), at least for $T \to 0$, goes to zero, in a formal sense, to macroscopic accuracy. This result is a consequence of the so-called "orthogonality catastrophe" of Anderson, or, in other words, of the presence of a large number of electron-hole pairs of low excitation energy in the structure of the many-particle wave function $\Psi_{\mathbf{l}}$ considered in the basis of unperturbed electronic states.[3–7] It is the admixture of these low-energy states that leads to the infrared divergence that makes (1.2) go to zero. As a result, the width of the coherent band narrows to zero or, in any case (when all the physical factors that smear out the divergence are taken into account), to a very small value compared to Δ_0. This same infrared divergence causes the difficulties noted by Nozieres and Iche[8] in the description of the Brownian motion of a heavy particle in a Fermi fluid.

Actually, results (1.) and (1.2) imply that during the time of the under-barrier tunneling of the heavy particle there is no readjustment of the electron cloud of the heavy particle in the intial unit cell. *This corresponds to a completely anti-adiabatic picture.* In the case of a small-radius polaron, the light electron passes under the barrier in a time during which the polarized cloud formed by the displacement of the atoms in the crystal is unable to readjust. In the case under consideration, which is just the opposite, this is obviously not so. It is easily inferred that, on the contrary, in the case of a total adiabatic readjustment of the electrons the overlap integral in (1.1) in general goes to unity.

The recent papers of Kondo[9,10,2] have cast doubt upon the validity of the adiabatic approximation in metals. This doubt was based on the fact that if one takes as the variational wave function the purely adiabatic wave function

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$$\Phi(\mathbf{r}, \mathbf{R}) = \varphi(\mathbf{R}) \psi(\mathbf{r}, \mathbf{R}),$$

where $\varphi(\mathbf{R})$ is the wave function of the particle in an isolated potential well and $\psi(\mathbf{r},\mathbf{R})$ is the wave function of the electrons at a fixed value of $\mathbf{R}$, then the energy diverges. This divergence is a result of the action of the kinetic-energy operator of the particle on the function $\psi(\mathbf{r},\mathbf{R})$. In fact,

$$\begin{aligned} &\langle \psi(\mathbf{r}, \mathbf{R}) | -\partial_{\mathbf{R}}{}^2/2M | \psi(\mathbf{r}, \mathbf{R}) \rangle \\ &= -(\partial_{\mathbf{R}'}{}^2/2M) \langle \psi(\mathbf{r}, \mathbf{R}) | \psi(\mathbf{r}, \mathbf{R}') \rangle |_{\mathbf{R}'=\mathbf{R}}, \end{aligned} \tag{1.3}$$

and, using for the overlap integral the explicit form obtained in Ref. 7 for $T = 0$, we immediately find that this part of the total energy diverges macroscopically. (Here and below the symbol $\partial_{\mathbf{R}}$ means a differentiation with respect to $\mathbf{R}$ that operates on the electron wave function only). This result also led to the conclusion that the interaction of the heavy particle with the electrons is of a nonadiabatic character. In fact, this is actually the case for the model Hamiltonian used by Kondo in analysis of the problem,

$$\hat{H} = \hat{H}^{el} + \Delta_0 \sum_{\mathbf{l},\mathbf{g}} \hat{c}_{\mathbf{l}}^{+} \hat{c}_{\mathbf{l}+\mathbf{g}} + \sum_{\mathbf{l}} \hat{V}_{\mathbf{l}} \hat{c}_{\mathbf{l}}^{+} \hat{c}_{\mathbf{l}}, \tag{1.4}$$

where $\hat{c}_{\mathrm{l}}^{+}$ and $\hat{c}_{\mathrm{l}}$ are the creation and absorption operators for a particle at site l and $\hat{V}_{\mathrm{l}}$ is the operator, fixed at site l, for the interaction of the electrons with the particle. In this Hamiltonian the particle has no intra-well degrees of freedom. Thus it was implicitly assumed that the excited levels of the particle in the well are separated from the ground state by an amount ω that is greater than the Fermi energy ε_F of the electrons. But this is just the obvious anti-adiabaticity condition. It is not accidental that in considering a more general variational problem with allowance for the intra-well dynamics of the particle Kondo[9] obtained a criterion of nonadiabaticity in the form

$$\omega \gg \varepsilon_F / \ln(M/m). \tag{1.5}$$

It was concluded that under the opposite inequality there is no polaron effect.

Actually, the adiabatic approximation is valid in metals and does not lead to a divergence of the energy.[11] It was shown in Ref. 11 that when the nonadiabatic operator $\hat{B} = -\partial_R^2/2M$ is taken into account it is necessary to simultaneously take into account another nonadiabatic operator $\hat{A} = -\partial_R \nabla_R / M$ as well. Then the divergence in the energy is eliminated and instead the corrections to the energy of the adiabatic state turn out to be small (see the following section for details). We emphasize that the operators $\hat{A}$ and $\hat{B}$, which differ in their structure and the effect of their operation on the adiabatic wave function, derive from the same origin and should always be taken into account simultaneously. Because the matrix elements of the operator $\hat{A}$ are nonzero only for the transitions that are non-diagonal in the state of the particle in the well, allowance for this operator necessarily requires consideration of a system of levels in the potential well, i.e., allowance for the intra-well motion of the particle. This circumstance is of a fundamental nature.

Thus, the divergence of the energy of the adiabatic state due to (1.3) is spurious, and Hamiltonian (1.4), which does not take the intra-well transitions into account, does not give a realistic description of the problem in general.

In this analysis of the problem, Kondo actually proceeded from the assumed validity of (1.4) and (1.5) and thus considered the completely anti-adiabatic case. In ordinary metals the inequality opposite to (1.5) always holds, in accordance with the standard small parameter

$$\varkappa^2 = (m/M)^{1/2} \sim \omega/\varepsilon_F \ll 1 \tag{1.6}$$

[as the ratio M/m in (1.5) is formally increased, it should be remembered that, in a fixed potential relief, $\omega \sim M^{-1/2}$]. Inequality (1.5) actually fails also for the lighter kinds of heavy particles, including the μ^+ meson (the lightest of all), whose quantum diffusion in metals has been studied intensively in recent years.

Although in the energy sense the validity of the adiabatic approximation in metals is not in question, the actual picture, as elucidated in the present study, turns out to be substantially more complicated. In the many-particle wave function arising upon the introduction of a heavy particle, the admixture of virtual excitations having frequencies $\delta E > \omega$ adjusts to the particle and follows it adiabatically. At the same time, excitations with $\delta E < \omega$ remain, and as the particle moves in the potential well the corresponding part of the wave function adjusts to the center of the well. By virtue of (1.6), the overwhelming majority of the virtual transitions are adiabatic, and this fraction of the transitions determines the state energy, the screening, the effective adiabatic potential, etc. The low-energy excitations, which because of their small phase volume do not substantially influence these characteristics, nevertheless predetermine the occurrence of an appreciable polaron effect owing precisely to the nonadiabatic part of the wave function that is directed toward the center of the potential well. This is because the infrared catastrophe due to the virtual creation of low-energy electron-hole pairs remains, though the sharp decrease in the nonadiabaticity interval from $(0,\varepsilon_F)$ to $(0,\omega)$ causes the large characteristic parameter $\ln(\varepsilon_F/T)$ to be replaced by $\ln(\omega/T)$. Because this factor is in the exponent, the polaronic narrowing turns out to be several orders of magnitude smaller.

In this paper we develop a technique of constructing the many-particle wave function in the presence of intra-well motion of the heavy particle through the use of the adiabatic basis and a perturbation theory in the nonadiabaticity parameter $\varkappa^2$ (1.6). We systematically trace the separation of the electronic wave function into two parts, one of which actually does not follow the particle but turns out to be directed toward the center of the potential well. The results enable us to determine the polaron effect due to the electrons and to solve the problem of the quantum diffusion of a heavy particle in a metal both in the ideal case and in the presence of a static shift between the levels of adjacent wells.

2. ADIABATIC APPROXIMATION IN A METAL

Suppose that a heavy particle in a metal is moving in a potential relief $U(\mathbf{R})$. For simplicity we assume that the electron fluid is uniform. The initial Hamiltonian is then written in the form

$$\hat{H}=\hat{H}_M+\hat{H}^{el}+\hat{V}(\mathbf{r},\ \mathbf{R}),\quad \hat{H}_M=-\nabla_\mathbf{R}^2/2M+U(\mathbf{R}) \tag{2.1}$$

(where $\hat{V}(\mathbf{r},\mathbf{R})$ is the interaction of the electrons with the particle). We use the scheme developed in Ref. 11 for the adiabatic theory of the metal. In this theory the adiabatic wave function (zeroth approximation) is of the form

$$\Phi_{\alpha n}^{(0)}=\varphi_\alpha(\mathbf{R})\psi_n(\mathbf{r},\mathbf{R}), \tag{2.2}$$

where ψ_n is the solution of the Schrödinger equation for the electrons at a fixed $\mathbf{R}$:

$$[\hat{H}^{el}+\hat{V}(\mathbf{r},\ \mathbf{R})]\psi_n(\mathbf{r},\ \mathbf{R})=E_n\psi_n(\mathbf{r},\ \mathbf{R}), \tag{2.3}$$

and $\varphi_\alpha(\mathbf{R})$ is the eigenfunction of the Hamiltonian $\hat{H}_M$. At first we shall neglect the weak tunneling between the potential wells forming the periodic potential relief $U(\mathbf{R})$. Then the wave functions φ_α will refer to an individual well, for which we shall assume, without loss of generality, that the harmonic approximation holds. In the case of a uniform electron fluid, E_n is independent of $\mathbf{R}$ and the function φ_α is independent of the index n.

The system of functions $\Phi_{\alpha n}^{(0)}$ is complete in $(\mathbf{r},\mathbf{R})$ space. Therefore, an arbitrary solution can be represented as

$$\Phi(\mathbf{r},\mathbf{R})=\sum_{\alpha,n}\xi_{\alpha n}\varphi_\alpha(\mathbf{R})\psi_n(\mathbf{r},\mathbf{R}). \tag{2.4}$$

By operating on $\Phi(\mathbf{r},\mathbf{R})$ with the total Hamiltonian $\hat{H}$ (2.1), we arrive at a system of perturbation-theory equations which determine the corrections to the adiabatic solution:

$$(E-E_n-E_\alpha)\xi_{\alpha n}=\sum_{\beta,m}C_{\alpha n,\beta m}\xi_{\beta m}, \tag{2.5}$$

where for the nonadiabaticity operator $\hat{C}=\hat{A}+\hat{B}$ we have

$$\begin{aligned}\hat{A}_{\alpha n,\beta m}&=\langle\alpha|-\langle n|\partial_\mathbf{R}|m\rangle\nabla_\mathbf{R}/M|\beta\rangle,\\ \hat{B}_{\alpha n,\beta m}&=\langle\alpha|-\langle n|\partial_\mathbf{R}^2|m\rangle/2M|\beta\rangle.\end{aligned} \tag{2.6}$$

Using Eq. (2.3), we can convert the matrix element $\langle n|\partial_\mathbf{R}|m\rangle$ to a form which is convenient for further analysis. To do this, we operate with $\partial_\mathbf{R}$ on both sides of Eq. (2.3) and then, after multiplying by ψ_n^*, integrate over the coordinates of the electrons. As a result we find

$$\langle n|\partial_\mathbf{R}|m\rangle=-\frac{\langle n|\partial V/\partial\mathbf{R}|m\rangle}{E_n-E_m},\quad \langle n|\partial_\mathbf{R}|n\rangle=0. \tag{2.7}$$

The solution of system of equations (2.5) permits us to find the nonadiabatic readjustment of the wave function and the corresponding corrections to the energy. Let us begin with the second.

The operator for the interaction of the electrons with the heavy particle in (2.1) and (2.7) can be written in the second-quantization representation as

$$\hat{V}=\sum_{\mathbf{k},\mathbf{k}',\sigma}V_{\mathbf{k},\mathbf{k}'}(\mathbf{R})\hat{a}^{+}_{\mathbf{k},\sigma}\hat{a}_{\mathbf{k}',\sigma},\quad V_{\mathbf{k},\mathbf{k}'}(\mathbf{R})=V_{\mathbf{k},\mathbf{k}'}(0)e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{R}}. \tag{2.8}$$

In calculating matrix element (2.7) we keep only the first term of the expansion in the small deviation of the particle from the equilibrium position in the potential well, setting $\mathbf{R}=\mathbf{R}_0=0$. Then the operator $\hat{B}$ turns out to be diagonal with respect to the states of the particle in the well, while the matrix elements of the operator $\hat{A}$ decompose into products of independent matrix elements:

$$\hat{A}_{\alpha n,\beta m}=-\langle n|\partial_\mathbf{R}|m\rangle\langle\alpha|\nabla_\mathbf{R}/M|\beta\rangle. \tag{2.9}$$

It follows from the form of (2.8) that matrix elements (2.7) are nonzero only for transitions involving the creation of a single electron-hole pair:

$$\begin{aligned}&\langle n|\partial_\mathbf{R}|m\rangle\to-d_{\mathbf{k},\mathbf{k}'}/(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'}),\\ &d_{\mathbf{k},\mathbf{k}'}=\langle\mathbf{k}|\nabla_\mathbf{R}\hat{V}(\mathbf{r}-\mathbf{R})|\mathbf{k}'\rangle=-i(\mathbf{k}-\mathbf{k}')V_{\mathbf{k},\mathbf{k}'}.\end{aligned} \tag{2.10}$$

Let us calculate the correction to the energy of the adiabatic state. The first-order correction $E^{(1)}$ is determined by the diagonal matrix element of the operator $\hat{B}$. This matrix element can be found immediately by using the completeness of the electron wave functions and the vanishing of the diagonal matrix element of $\partial_\mathbf{R}$:

$$\langle n|\partial_\mathbf{R}^2|n\rangle=\sum_s\langle n|\partial_\mathbf{R}|s\rangle\langle s|\partial_\mathbf{R}|n\rangle.$$

After using (2.10), we have

$$E^{(1)}=\frac{1}{2M}\sum_{\mathbf{k},\mathbf{k}',\sigma}\frac{|d_{\mathbf{k},\mathbf{k}'}|^2}{(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})^2}n_\mathbf{k}(1-n_{\mathbf{k}'}). \tag{2.11}$$

This correction to the energy agrees exactly with the result obtained from (1.3). It contains a pole of order two, which gives rise to a logarithmic divergence at $T=0$:

$$E^{(1)}\sim\ln N,$$

where N is the number of electrons in the system.

A contribution from the operator $\hat{A}$ arises only in the second order of perturbation theory. If we take it into account that ∇_R has nonzero matrix elements only for transitions between adjacent levels

$$\langle N_\lambda\pm1|\nabla_\mathbf{R}/M|N_\lambda\rangle=\mp e_\lambda[\omega(N_\lambda+{}^1/_2\pm{}^1/_2)/2M]^{1/2} \tag{2.12}$$

(e_λ, ω and N_λ are the polarization vector, the frequency, and number of the level of a degenerate three-dimensional oscillator; $\lambda=1, 2, 3$), then we easily find that

$$\begin{aligned}E_A^{(2)}=\frac{1}{2M}\sum_{\substack{\mathbf{k},\mathbf{k}'\\ \sigma,\lambda}}\frac{|e_\lambda d_{\mathbf{k},\mathbf{k}'}|^2}{(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})^2}n_\mathbf{k}(1-n_{\mathbf{k}'})\left\{\frac{\omega^2}{(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})^2-\omega^2}\right.\\ \left.+(2N_\lambda+1)\frac{\omega(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})}{(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})^2-\omega^2}\right\}.\end{aligned} \tag{2.13}$$

We can easily verify that for $|\varepsilon_k-\varepsilon_k'|\ll\omega$ the first term in this expression cancels exactly the first-order correction (2.11) $\Sigma_\lambda\ e_\lambda^i e_\lambda^j=\delta_{i,j}$). The divergence is thereby removed. This cancellation, which occurs in all orders of perturbation theory, is by no means accidental. It reflects the fact that both nonadiabatic operators $\hat{A}$ and $\hat{B}$ derive from the same origin and should always be taken into account together. A model which formally introduces only one level in the well eliminates the contribution of the operator $\hat{A}$ because of (2.12) and thereby causes the nonadiabatic correction to the energy to become infinite (see the Introduction).

Combining (2.11) and (2.13), we get for the leading correction, quadratic in the parameter $\varkappa^2$ (1.6), to the adiabatic energy

$$\Delta E=\frac{1}{2M}\sum_{\substack{\mathbf{k},\mathbf{k}'\\ \sigma,\lambda}}\frac{|e_\lambda d_{\mathbf{k},\mathbf{k}'}|^2}{(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})^2-\omega^2}n_\mathbf{k}(1-n_{\mathbf{k}'})\times\left\{1+(2N_\lambda+1)\frac{\omega}{\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'}}\right\}. \tag{2.14}$$

A direct calculation of (2.14) gives the estimate

$$\Delta E\sim\varepsilon_F(m/M)\ln(\varepsilon_F/\omega)\sim\omega(\omega/\varepsilon_F)\ln(\varepsilon_F/\omega).$$

Thus the nonadiabatic corrections to the energy not only turn out to be finite but they are small, in accordance with the adiabaticity parameter $\varkappa^2$ (1.6), as well.

3. NONADIABATIC READJUSTMENT OF THE ELECTRONIC WAVE FUNCTION

Let us now consider the nature of the readjustment of the adiabatic wave function under the influence of a nonadiabatic perturbation. For this we return to the representation of the wave function in the form an expansion in the adiabatic basis (2.4) and seek the coefficients $\xi_{\alpha n}$ from the solution of equation (2.5). Let the original unperturbed state have indices $\alpha=0$ and m_0. Then, in the first order of a perturbation theory in $\hat{A}$ we have

$$\xi_{1m_1}=\left(\frac{\omega}{2M}\right)^{1/2}\frac{(\mathbf{e}_\lambda d_{\mathbf{k},\mathbf{k}'})n_{\mathbf{k}'}(1-n_\mathbf{k})}{(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'}+\omega)(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{k}'})}. \tag{3.1}$$

Here the state m_1 differs from m_0 by one electron-hole pair $(\mathbf{k},\mathbf{k}')$.

In this approximation the admixture of states having an excitation of the first level in the well is given by [see Eq. (2.10)]

$$\gamma=\sum_{m_1}|\xi_{1m_1}|^2$$

$$\approx\varkappa^2[V\rho(\varepsilon_F)]^2\int\int\frac{d\varepsilon\, d\varepsilon' n_{\varepsilon'}(1-n_\varepsilon)\omega^2}{(\varepsilon-\varepsilon')^2(\varepsilon-\varepsilon'+\omega)^2}. \tag{3.2}$$

Here V is the characteristic scale of the interaction of the particle with the electrons, and $\rho(\varepsilon_F)$ is the density of states at the Fermi surface. The main contribution to this integral is from electron-hole pairs with energy $|\varepsilon-\varepsilon'|<\omega$. The contribution of the virtual excitations from all the rest of the phase volume is small, amounting in order of magnitude to $\varkappa^2$. Therefore,

$$\gamma\approx\varkappa^2[V\rho(\varepsilon_F)]^2J_\omega,\quad J_\omega=\int_{-\omega}^{\omega}\int\frac{d\varepsilon d\varepsilon' n_{\varepsilon'}(1-n_\varepsilon)}{(\varepsilon-\varepsilon')^2}. \tag{3.3}$$

We note that to go over from the sum to an integral in (3.2) it is necessary to formally supplement the definition of the integration procedure because of the presence of a pole of order two; in particular, we must introduce a cutoff of the familiar logarithmic divergence at $T=0$. We shall return to this later. For now it is important only that $J_\omega\gg1$.

To clarify the structure of the resulting wave function, let us first assume that the parameter $\varkappa^2$ is so small that $\gamma\ll1$. We find the coefficients $\xi_{\alpha n}$ to lowest order in this parameter. Purely for simplicity of exposition, we shall consider a particle in a one-dimensional potential relief (the generalization to the three-dimensional case is immediate). Then α coincides with the number of the level, and, to lowest order in γ, $\xi_{\alpha n}$ arises on the α-fold application of the operator $\hat{A}$ (the operator $\hat{B}$ is diagonal in the vibrational states):

$$\xi_{\alpha m_\alpha}=\sum_{m_1\ldots m_{\alpha-1}}(-1)^\alpha\frac{\langle\alpha m_\alpha|\hat{A}|\alpha-1m_{\alpha-1}\rangle}{\alpha\omega+E_{m_\alpha}-E_{m_0}}\cdots\frac{\langle1m_1|\hat{A}|0m_0\rangle}{\omega+E_{m_1}-E_{m_0}} \tag{3.4}$$

In accordance with (2.9) and (2.10), operating with $\hat{A}$ changes the number of electron-hole pairs by one (the state $|m_0\rangle$ is taken to be the vacuum state) or leads to the rescattering of an electron or hole. If α pairs form, then the intermediate summation in (3.4) drops out, and

$$\sum_{m_\alpha}|\xi_{\alpha m_\alpha}|^2\approx\gamma^\alpha. \tag{3.5}$$

Here, as in going from (3.2) to (3.3), in each integral over $d\varepsilon_s d\varepsilon_s'$ for an individual pair, the main contribution is from the region of excitation energies $|\varepsilon_s-\varepsilon_s'|$ that are small compared to ω. Excitations with $|\varepsilon_s-\varepsilon_s'|>\omega$ give a negligible contribution.

This same result (3.5) is obtained in the case when the absorption of pairs occurs in the intermediate transitions and the state $|m_\alpha\rangle$ corresponds to fewer than α pairs. In fact, in this case the intermediate summation over the momenta of the created and then absorbed pair $\mathbf{k}_s$, $\mathbf{k}_s'$ remains in (3.4). Then the integrand again has a pole of order two, $(\varepsilon_s-\varepsilon_s')^{-2}$, which makes it possible to neglect the energies of the electronic excitations in comparison with ω, and the integral gives (3.3).

As a result, every such "annihilation" leads to a factor of γ_2 in (3.5) which compensates for the lack of summation over the momenta of the two pairs in Σ_{m_α}.

Let us now show that the rescattering processes can be neglected in comparison with the creation or absorption of electron-hole pairs. The virtual rescattering of an electron in (3.4) corresponds to the creation of a pair $\hat{a}_s+\hat{a}_{k'}$ in one step and a pair $\hat{a}_k+\hat{a}_s$ in a later step; the opposite order corresponds to the rescattering of a hole. If a pair $(\mathbf{k},\mathbf{k}')$ is contained in state $|m_\alpha\rangle$, then in amplitude (3.4) there is one intermediate integration over $d\varepsilon_s$. Again it is easily inferred that the energy of the electron-hole pairs can be neglected in comparison with ω, and the sum of the two rescattering channels leads to an integral of the form

$$\int d\varepsilon_s\left[\frac{1-n_s}{(\varepsilon_s-\varepsilon_{k'})(\varepsilon_k-\varepsilon_s)}-\frac{n_s}{(\varepsilon_k-\varepsilon_s)(\varepsilon_s-\varepsilon_{k'})}\right]$$

$$=\frac{2}{\varepsilon_{k'}-\varepsilon_k}\ln\frac{\varepsilon_{k'}}{\varepsilon_k}. \tag{3.6}$$

This expression is nonsingular at $\varepsilon_k=\varepsilon_{k'}$. If we now evaluate the sum in (3.5), we are easily satisfied that each intermediate rescattering removes a factor $J_\omega \gg 1$ and so, instead of two powers of γ, the substantially smaller factor $\gamma \varkappa^2$ appears. This result allows us to neglect rescattering processes in finding the amplitude (3.4).

We note that by increasing the order of perturbation theory when finding $\xi_{\alpha m_\alpha}$, we could also have put in the operator $\hat{B}$ in the intermediate stage; since it is diagonal in α, the exact same transition occurs as when two operators $\hat{A}$ are used. The sum of two such channels, by analogy with the correction to the energy (see the previous section), largely cancels, and an extra small factor that is clearly smaller than γ^2 arises in sum (3.5).

From these results we can conclude that substantial readjustment of the adiabatic wave function involves the creation of electron-hole pairs with an energy that is small compared to ω. Therefore, in finding the amplitude $\xi_{\alpha m_\alpha}$ in (3.4) we can neglect the energy of the electronic excitations in the denominators whenever it occurs together with ω. We substitute (2.9) into (3.4) and use the well-known relation for the matrix elements

$$\langle\alpha|\nabla_x/M\omega|\beta\rangle=-\langle\alpha|(x-x_0)|\beta\rangle.$$

Then, remembering that ∂_x operates only on the electronic states, we find

$$\xi_{\alpha m_\alpha}=(-1)^\alpha \sum_{m_1\ldots m_{\alpha-1}}{}' \frac{\langle\alpha m_\alpha|(x-x_0)\partial_x|\alpha-1 m_{\alpha-1}\rangle}{\alpha}$$

$$\ldots\frac{\langle 1m_1|(x-x_0)\partial_x|0m_0\rangle}{1}$$

$$=\frac{(-1)^\alpha}{\alpha!}\langle\alpha m_\alpha|\{(x-x_0)\tilde{\partial}_x\}^\alpha|0m_0\rangle; \tag{3.7}$$

here

$$\tilde{\partial}_x=P\partial_x P, \tag{3.8}$$

where P is the projection operator that selects from the total set of electronic states $\{m_\alpha\}$ (at fixed $\mathbf{R}$) only those states which differ from the initial state m_0 by the presence of electron-hole pairs with energies smaller than ω [the primed sum in (3.7) corresponds to this same selection of intermediate states]. The cutoff introduced on the frequency ω has logarithmic accuracy, as follows directly form (3.3) [see also Eqs. (5.5) and (5.7) below].

To this same order in the parameter γ, the expression for $\xi_{\alpha m_\alpha}$ can be written in the form

$$\xi_{\alpha m_\alpha}=\langle\alpha m_\alpha|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}|0m_0\rangle. \tag{3.9}$$

Here we have given directly the expression for the three-dimensional oscillator—the corresponding generalization of (3.7) presents no difficulty (α is now a set of three quantum numbers).

Actually, result (3.9) is more general and does not rely on the parameter γ being small. In fact, if we recognize at the very beginning that it is electronic excitations with energies small compared to ω that are important in the structure of the perturbation theory, we can rewrite the initial equation (2.5) in the form

$$(E-E_\alpha)\xi_{\alpha n}=\sum_{\beta m}(\hat{A}+\hat{B})_{\alpha n,\beta m}\xi_{\beta m}. \tag{3.10}$$

Let us show that (3.9) is a solution of equation (3.10) with $E=E_{\alpha=0}=E_0$. For this we substitute (3.9) into (3.10) and transform the left- and right-hand sides separately [see Eq. (2.1)]:

$$(E_0-E_\alpha)\langle\alpha n|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}|0m_0\rangle$$

$$=\langle\alpha n|[\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\},H_M]|0m_0\rangle$$

$$=\langle\alpha n|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}\frac{\tilde{\partial}_\mathbf{R}{}^2}{2M}$$

$$-\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}\frac{\tilde{\partial}_\mathbf{R}}{M}\nabla_\mathbf{R}|0m_0\rangle. \tag{3.11}$$

In transforming the right-hand side we use form (2.6) for the nonadiabaticity operators:

$$\sum_{\beta m}\left\langle\alpha n\left|-\frac{\partial_\mathbf{R}{}^2}{2M}-\frac{\partial_\mathbf{R}\nabla_\mathbf{R}}{M}\right|\beta m\right\rangle\langle\beta m|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}|0m_0\rangle$$

$$=\left\langle\alpha n\left|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}\left(-\frac{\partial_\mathbf{R}{}^2}{2M}+\frac{\partial_\mathbf{R}\tilde{\partial}_\mathbf{R}}{M}\right)\right.\right.$$

$$\left.\left.-\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}\frac{\tilde{\partial}_\mathbf{R}}{M}\nabla_\mathbf{R}\right|0m_0\right\rangle. \tag{3.12}$$

Since the state $|n\rangle$ in (3.10) belongs to the subspace projected by the operator P, the product $\partial_\mathbf{R}\tilde{\partial}_\mathbf{R}$ in the curly brackets is automatically the same as $\tilde{\partial}^2_\mathbf{R}$, in operating on a state from this space, leads either directly to the creation of two electron-hole pairs belonging to this same subspace or to the creation of one such pair and a rescattering in the intermediate stage. In the latter case, however, it can be assumed by virtue of result (3.6) that no departure from the subspace under consideration will occur. Therefore, the operator $\partial^2_\mathbf{R}$ in the curly brackets can be replaced by $\tilde{\partial}^2_\mathbf{R}$. Now by comparing (3.11) and (3.12), we see that the two results agree. Thus Eq. (3.9) is actually a solution of equation (3.10).

Importantly, this demonstration implies that a solution of the form (3.9) is valid not only for arbitrary γ but also for an arbitrary form of the potential well, and also that it does not rely on the approximate representation (2.9) of the matrix elements of the operator $\hat{A}$.

Knowing solution (3.9), we can now find the perturbed wave function (2.4):

$$\mathbf{\Phi}(\mathbf{r},\mathbf{R})$$

$$=\sum_{\alpha n}\langle\alpha n|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}|0m_0\rangle\varphi_\alpha(\mathbf{R}-\mathbf{R}_0)\psi_n(\mathbf{r},\mathbf{R}). \tag{3.13}$$

We note here that

$$\sum_{\alpha n}|\langle\alpha n|\{\exp-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_\mathbf{R}\}|0m_0\rangle|^2=1$$

and, consequently, function (3.13) is automatically normalized.

Had $\partial_\mathbf{R}$ operated throughout the whole space of wave functions, i.e., if we had $P = 1$ in (3.8), then the operator $\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_{\mathbf{R}}\}$ would be the usual displacement operator. Then

$$\langle \alpha n|\exp\{-(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_{\mathbf{R}}\}|0m_0\rangle = \langle \Phi^{(0)}_{\alpha n}(\mathbf{r},\mathbf{R})|\Phi^{(0)}_{0m_0}(\mathbf{r},\mathbf{R}_0)\rangle$$

and function (3.13) would turn out to be

$$\Phi(\mathbf{r},\mathbf{R}) = \Phi^{(0)}_{0m_0}(\mathbf{r},\mathbf{R}_0),$$

i.e., the initial adiabatic function would become a typical anti-adiabatic function—the electrons would not follow the position of the particle but would be directed toward the center of the well.

In the situation under consideration, only the part of the wave function associated with low-energy virtual excitations turns out to be directed toward the center of the well. This can be seen most clearly in the case when interaction (2.8) is weak. Then, taking (2.10) into account, we can expand the exponential in (3.9) and (3.13) in a series, keeping only two terms. Then in the same approximation we have

$$\begin{gathered}\langle \alpha n|-P(\mathbf{R}-\mathbf{R}_0)\tilde{\partial}_{\mathbf{R}}P|0m_0\rangle \\ = \langle \Phi^{(0)}_{\alpha n}(\mathbf{r},\mathbf{R})|P\Phi^{(0)}_{0m_0}(\mathbf{r},\mathbf{R}_0)\rangle, \quad \alpha n\neq 0m_0, \\ \Phi(\mathbf{r},\mathbf{R}) = \Phi^{(0)}_{0m_0}(\mathbf{r},\mathbf{R})\{1-\langle \Phi^{(0)}_{0m_0}(\mathbf{r},\mathbf{R})|\Phi^{(0)}_{0m_0}(\mathbf{r},\mathbf{R}_0)\rangle\} \\ +\varphi_0(\mathbf{R}-\mathbf{R}_0)P\psi_{m_0}(\mathbf{r},\mathbf{R}_0).\end{gathered} \tag{3.14}$$

Operating with P on the function $\psi_{m_0}(\mathbf{r},\mathbf{R}_0)$ reduces to selecting from the expansion of this function in the adiabatic functions (2.2) only those states with $\delta E < \omega$.

Thus, in this case wave function (3.13) decomposes into the sum of two functions, one of which is purely adiabatic and the other anti-adiabatic.

4. TRANSITION MATRIX ELEMENT

Let us now find the matrix element for the transition of a particle from unit cell $\mathbf{l}$ to $\mathbf{l}+\mathbf{g}$. Assuming that the amplitude of the tunneling transition of the particle is extremely small, we can say that within each cell there is sufficient time for the formation of an electronic wave function of the type found in the previous section (this statement is refined somewhat in the next section). Here the nonadiabatic part of the electronic wave function gives rise to a polaron effect which reflects the fact that a part of the perturbed electron cloud does not follow the particle when it tunnels. We note that the inverse time (imaginary) for the passage of the particle under the barrier is of the same order of magnitude as ω. For simplicity we shall not differentiate between these two characteristics.

Let $\hat{H}'(\mathbf{R})$ be the Hamiltonian responsible for the tunneling transition between adjacent cells. Then, assuming that the motion is in the lowest vibrational level ($\alpha = 0$) and taking (3.13) into account, we have for a transition involving excitation of the electronic subsystem

$$\begin{gathered}M_{nm}(\mathbf{g}) = (\Phi^{(\mathbf{l}+\mathbf{g})}_{0n}|\hat{H}_1(\mathbf{R})|\Phi^{(\mathbf{l})}_{0m}) \\ = \sum_{\alpha,\beta,s}\langle 0_{\mathbf{l}+\mathbf{g}}n|\exp\{(\mathbf{R}-\mathbf{R}_{\mathbf{l}+\mathbf{g}})\tilde{\partial}_{\mathbf{R}}\}|\alpha_{\mathbf{l}+\mathbf{g}}s\rangle\langle\alpha_{\mathbf{l}+\mathbf{g}}|\hat{H}_1|\beta_{\mathbf{l}}\rangle \\ \times\langle\beta_{\mathbf{l}}s|\exp\{-(\mathbf{R}-\mathbf{R}_{\mathbf{l}})\tilde{\partial}_{\mathbf{R}}\}|0_{\mathbf{l}}m\rangle \\ = \langle 0_{\mathbf{l}+\mathbf{g}}|\langle n|\exp\{-\mathbf{g}\tilde{\partial}_{\mathbf{R}}\}|m\rangle|\hat{H}_1|0_{\mathbf{l}}\rangle.\end{gathered}$$

Here the states in the angle brackets correspond to the purely adiabatic wave function (2.2); for the true wave functions (in parentheses) we retain the notation of the initial adiabatic states.

Evaluating the electronic matrix element at the external point $\mathbf{R}_*$ between the two cells, we find

$$M_{nm}(\mathbf{g}) \approx \Delta_0\langle n|\exp\{-\mathbf{g}\tilde{\partial}_{\mathbf{R}}\}|m\rangle_{\mathbf{R}_*}, \quad \Delta_0 = \langle 0_{\mathbf{l}+\mathbf{g}}|\hat{H}_1|0_{\mathbf{l}}\rangle. \tag{4.1}$$

We note that for purely adiabatic states only the diagonal matrix element, which is simply equal to Δ_0, is nonzero.

The wave functions $\psi_m(\mathbf{r},\mathbf{R})$ is a solution of equation (2.3). We operate from the left on both sides of this equation with the operator $\exp\{-\mathbf{g}\tilde{\partial}_R\}$. Introducing the function

$$\Psi_m(\mathbf{r},\mathbf{R}) = \exp\{-\mathbf{g}\tilde{\partial}_{\mathbf{R}}\}\psi_m(\mathbf{r},\mathbf{R}), \tag{4.2}$$

we find

$$\begin{gathered}[\hat{H}^{el}+\hat{V}(\mathbf{r},\mathbf{R})]\Psi_m(\mathbf{r},\mathbf{R})+\Delta\hat{V}(\mathbf{r},\mathbf{R})\Psi_m(\mathbf{r},\mathbf{R}) = E_m\Psi_m(\mathbf{r},\mathbf{R}), \\ \Delta\hat{V}(\mathbf{r},\mathbf{R}) = \exp\{-\mathbf{g}\tilde{\partial}_{\mathbf{R}}\}\hat{V}(\mathbf{r},\mathbf{R})\exp\{g\tilde{\partial}_{\mathbf{R}}\}-\hat{V}(\mathbf{r},\mathbf{R}).\end{gathered} \tag{4.3}$$

Because of the projection operators in definition (3.8), the operator $\Delta\hat{V}$ has nonzero matrix elements only between states having an energy of the electron-hole pairs that is less than ω. Using the explicit form of the interaction from (2.8), we have

$$\begin{aligned}\Delta\hat{V} = \sum_{\mathbf{k},\mathbf{k}',\sigma} V_{\mathbf{k},\mathbf{k}'}\{&\exp[-i(\mathbf{k}-\mathbf{k}')(\mathbf{R}-\mathbf{g})] \\ &-\exp[-i(\mathbf{k}-\mathbf{k}')\mathbf{R}]\}P\hat{a}_{\mathbf{k}\sigma}{}^{+}\hat{a}_{\mathbf{k}'\sigma}P.\end{aligned} \tag{4.4}$$

The solution of equation (4.3) can be written

$$\Psi_m = \hat{\Lambda}\psi_m, \quad \hat{\Lambda} = T\exp\left\{-i\int_{-\infty}^{0}\Delta\hat{V}(t)\,dt\right\}. \tag{4.5}$$

The operator $\Delta\hat{V}(t)$ is given in the interaction representation. Here the Hamiltonian in the absence of the interaction is $\hat{H}^{el}+\hat{V}(\mathbf{r},\mathbf{R})$, i.e., the electronic Hamiltonian for a fixed position of the particle at point $\mathbf{R}$. The eigenfunctions of this Hamiltonian are precisely the functions ψ_m, and the structure of expression (4.5) corresponds to the assumption that the interaction is turned on adiabatically.

In considering an electron gas without the interaction, we take the vertex in (2.8) and (4.4) to mean the screened potential or pseudopotential (which takes into account the readjustment of the one-electron wave function around the particle).

Thus, matrix element (4.1) with allowance for (4.2) has actually reduced to the matrix element

$$M_{nm}(\mathbf{g}) = \Delta_0\langle n|\hat{\Lambda}|m\rangle_{\mathbf{R}_*}. \tag{4.6}$$

We now advance an important argument. As was shown in Sec. 3, in the amplitude for the creation of an arbitrary number of electron-hole pairs, the governing channels are those in which only new pairs are created or absorbed, while the intermediate rescattering of excited pairs entails a sharp decrease of this amplitude. If we neglect the rescattering and rewrite $\Delta\hat{V}$ from (4.4) in the form

$$\Delta\hat{V}=\sum_{s>s'}{}'\Delta V_{ss'},\quad \Delta V_{ss'}=C_{ss'}\hat{a}_s^+\hat{a}_{s'}+C_{s's}\hat{a}_{s'}^+\hat{a}_s,\qquad(4.7)$$

then it is easy to understand that each term in the series of the ordinary *S*-matrix expansion will contain factors of $\Delta V_{ss'}(t_i)$, which commute with each other. This allows us to remove the chronological operator T in (4.5), whereupon the integration over time can be done directly in the argument of the exponential function. [Actually, the operators $\Delta V_{ss'}(t)$ for the same modes (s,s') at different times do not commute with each other. However, allowance for this noncommutativity will lead to the unimportant phase factor

$$-i\Delta E\int_{-\infty}^{0}dt,$$

which actually cancels out the analogous phase factor that was dropped in going from (4.2) to (4.5)]:

$$\hat{\Lambda}=\exp\left\{-\sum_{s>s'}{}'\left[\frac{C_{ss'}}{\varepsilon_s-\varepsilon_{s'}-i\delta}\hat{a}_s^+\hat{a}_{s'}+\frac{C_{s's}}{\varepsilon_{s'}-\varepsilon_s-i\delta}\hat{a}_{s'}^+\hat{a}_s\right]\right\}.\qquad(4.8)$$

By the same considerations, this expression can be rewritten in the form of a product:

$$\hat{\Lambda}=\prod_{s>s'}{}'\hat{\Lambda}_{ss'},$$

$$\hat{\Lambda}_{ss'}=\exp\left\{-\left[\frac{C_{ss'}}{\varepsilon_s-\varepsilon_{s'}-i\delta}\hat{a}_s^+\hat{a}_{s'}-\frac{C_{ss'}^*}{\varepsilon_s-\varepsilon_{s'}+i\delta}\hat{a}_{s'}^+\hat{a}_s\right]\right\}\qquad(4.9)$$

Here we have made use of the fact that according to (4.4) and (4.7), $C_{s's}=C_{ss'}^*$.

The evaluation of matrix element (4.6) can now be done without difficulty if it is taken into account that to macroscopic accuracy only the first three items must be kept in the series expansion of $\hat{\Lambda}_{ss'}$ in powers of the argument of the exponential function.

5. QUANTUM DIFFUSION IN A METAL

Let us begin our discussion of the diffusion problem with the case of a nonideal metal in which the static scatter $\xi_{1,1+g}$ of the energy levels in the neighboring wells, though small compared to ω, is larger than the tunneling energy width Δ_0 (more precisely, the effective width $\tilde{\Delta}_0$, see below). The probability of a transition from cell 1 to cell 1 + g with allowance for the results of the previous section is given by the expression

$$W_{1,1+g}=2\pi\Delta_0^2\sum_{n,m}\rho_m^{el}|\langle n|\hat{\Lambda}|m\rangle|^2\delta(E_m-E_n+\xi_{1,1+g})$$
$$=\Delta_0^2\int_{-\infty}^{+\infty}dt\,\mathrm{Sp}\{\hat{\rho}^{el}\hat{\Lambda}^+(t)\hat{\Lambda}(0)\}\exp(i\xi_{1,1+g}t).\qquad(5.1)$$

Taking (4.9) into account, we have for the diagonal matrix element of the product $\tilde{\Lambda}_{ss'}^+(t)\tilde{\Lambda}_{s's}(0)$, averaged over the electron distribution (see the remark at the end of Sec. 4),

$$1-\frac{|C_{ss'}|^2}{|\varepsilon_s-\varepsilon_{s'}-i\delta|^2}\{\bar{n}_s(1-\bar{n}_{s'})[1-\exp(i(\varepsilon_s-\varepsilon_{s'})t)]$$
$$+\bar{n}_{s'}(1-\bar{n}_s)[1-\exp(-i(\varepsilon_s-\varepsilon_{s'})t)]\},$$

where $\bar{n}_s$ is the Fermi distribution function. Using this result, we immediately find

$$W_{1,1+g}=\Delta_0^2\int_{-\infty}^{+\infty}dt\exp(-\chi(t))\exp\{i\xi_{1,1+g}t\},$$

$$\chi(t)=\sum_{ss'}\frac{|C_{ss'}|^2}{(\varepsilon_s-\varepsilon_{s'})^2}\bar{n}_s(1-\bar{n}_{s'})[1-\exp\{i(\varepsilon_s-\varepsilon_{s'})t\}]\qquad(5.2)$$

(here we have the total sum over s and s', so there is no factor of 2).

We note that the procedure used to obtain expression (5.2) is analogous to the familiar procedure in the theory of the phononic small-radius polaron (see, e.g., Ref. 12) and is a direct consequence of the possibility of representing the operator $\hat{\Lambda}$ in the form of a product as in (4.9).

Substituting the explicit form of the expression for $C_{ss'}$ from (4.4) and (4.7) and averaging over the directions of the vectors **k** and **k**′, we obtain

$$\chi(t)\approx 2b\int\int_{-\omega}^{\omega}\frac{d\varepsilon\,d\varepsilon'}{(\varepsilon-\varepsilon')^2}n_\varepsilon(1-n_{\varepsilon'})(1-e^{i(\varepsilon-\varepsilon')t}),\qquad(5.3)$$

$$b=2\rho^2(\varepsilon_F)\overline{|V(\mathbf{k}-\mathbf{k}')|^2[1-\cos(\mathbf{k}-\mathbf{k}')\mathbf{g})}.\qquad(5.4)$$

The limits of integration in (5.3) reflect the fact that only states with excitation energies smaller than ω contribute to expressions (4.4)–(4.9). In integral (5.3) there is no pole of order two, and all the calculations can be done immediately. Suppose $T\ll\omega$. Then, making the change of variables $\varepsilon-\varepsilon'=y$ and recognize that the integration with respect to ε' is an interval that is small compared to ω, we have

$$\chi(t)\approx 2b\int_0^{\omega}\frac{dy}{y}\left[(1-\cos yt)\,\mathrm{cth}\frac{y}{2T}+i\sin yt\right].$$

The expression in square brackets can be rewritten in the form

$$\left[\mathrm{ch}\frac{y}{2T}-\mathrm{ch}\,iy\left(t+\frac{i}{2T}\right)\right]\Big/\,\mathrm{sh}\frac{y}{2T}.$$

In the initial integral (5.2) we make the change of variables $t\rightarrow t-i/2T$ and then shift the integration contour onto the real axis. As a result we find

$$W_{1,1+g}=\Delta_0^2\exp\left(\frac{\xi_{1,1+g}}{2T}\right)\int_{-\infty}^{+\infty}dt\exp\left\{-2b\int_0^{\omega}\frac{dy}{y}\left[\frac{\mathrm{ch}(y/2T)-1}{\mathrm{sh}(y/2T)}\right.\right.$$
$$\left.\left.+\frac{1-\cos yt}{\mathrm{sh}(y/2T)}\right]\right\}\exp(i\xi_{1,1+g}t).\qquad(5.5)$$

The integral of the first term in square brackets is independent of t and equal to $\ln(\omega/\eta T)$, $\eta\approx 2$. Taking it into account that the integral of the second term converges rapidly, we replace the upper limit of the integration over y by ∞. Taking then the integral, we get

$$W_{l,l+g}=2\tilde{\Delta}_0^2(T)\exp\left(\frac{\xi_{l,l+g}}{2T}\right)\int_0^\infty dt\cos(\xi_{l,l+g}t)\frac{1}{(\operatorname{ch}\pi Tt)^{2b}},\tag{5.6}$$

$$\tilde{\Delta}_0(T)=\Delta_0(\eta T/\omega)^b.\tag{5.7}$$

Evaluating the resulting integral and making some straightforward manipulations, we finally obtain

$$W_{l,l+g}=\frac{2\tilde{\Delta}_0^2(T)\Omega_T}{\xi^2_{l,l+g}+\Omega_T^2}\pi^{1/2}\frac{|\Gamma(1+b+i\xi_{l,l+g}/2\pi T)|^2}{\Gamma(b+1)\Gamma(b+1/2)}\times\exp\left(\frac{\xi_{l,l+g}}{2T}\right),\quad\Omega_T=2\pi bT.\tag{5.8}$$

If we set $\xi=0$ and $\omega=\varepsilon_F$ in this expression, the result goes over to that found in Refs. 10 and 13.

Before embarking on a general analysis of expression (5.8), let us first consider the case of a weak interaction of the particle with the electrons of the metal, i.e., $b\ll1$. In this limit

$$W_{l,l+g}=\frac{2\Delta_0^2\Omega_{l,l+g}}{\xi^2_{l,l+g}+\Omega_T^2},\quad\Omega_{l,l+g}=\Omega_T\frac{\xi_{l,l+g}/T}{1-\exp(-\xi_{l,l+g}/T)}.\tag{5.9}$$

This result has the same structure as the general expression for the transition probability in the problem of quantum diffusion in an irregular medium in the presence of an interaction of the particle with phonons.[14,15] Expression (5.9) reflects the competition between the relative shifts of static ($\xi_{l,l+g}$) and dynamic (Ω_T) origin between the levels in adjacent wells. The dynamic shift is due to the difference in the energy of the levels in adjacent wells on account of the interaction of the particle with the same fluctuation in the electronic system ($T\ll\omega$).

For $\Omega_T\gg\xi_{l,l+g}$ we have $\Omega_{l,l+g}\approx\Omega_T$ and

$$W_{l,l+g}\approx2\Delta_0^2/\Omega_T.\tag{5.10}$$

We thereby arrive at a diffusion that is uniform over the crystal and is characterized by $D\sim1/T$ if the above inequality holds everywhere. In the opposite limiting case we have

$$W_{l,l+g}\approx\frac{2\Delta_0^2}{\xi_{l,l+g}}\frac{2\pi b}{1-\exp(-\xi_{l,l+g}/T)}.\tag{5.11}$$

Let us now return to the general case of arbitrary b. In the limit $\Omega_T\gg|\xi_{l,l+g}|$ expression (5.8), up to a numerical factor of the order of 1, becomes

$$W_{l,l+g}\approx2\tilde{\Delta}_0^2(T)/\Omega_T.$$

Hence, on the assumption of cubic symmetry, we have for the coefficient of quantum diffusion

$$D=\frac{za^2}{3}\frac{\tilde{\Delta}_0^2(T)}{\Omega_T},\tag{5.12}$$

where z is the number of equivalent sites in the immediate environment.

Comparison with (5.10) with allowance for (5.7) implies that *there is actually a polaronic decrease in the amplitude of the tunneling transition, but on a scale determined by the ratio T/ω rather than T/ε_F* (cf. Refs. 2, 10, 13, and 16). This result was actually predetermined by the limits of integration in (5.3), which reflect the actual narrow energy interval of nonadiabaticity $(0,\omega)$, contrary to the assumption that this interval extends throughout an entire energy band of the order of ε_F or of the width of the electronic conduction band. Importantly, even by $T\sim\omega$ the polaronic narrowing has vanished altogether.

We note that $\omega\sim M^{-1/2}$ in a fixed potential relief, and the polaron effect diminishes with increasing particle mass. In particular, the existing paradox concerning the nonphysical behavior of the polaron effect in the formal limit $M\to\infty$ is eliminated. For $T\ll\omega$

$$D\sim T^{-(1-2b)}\tag{5.13}$$

and for $b<\frac{1}{2}$, as is assumed in the absence of resonance scattering of the electrons on the particle, the diffusion coefficient increases with decreasing T, albeit slowly, especially in comparison with the phonon case (see, e.g., Refs. 14, 15, and 17). The dependence in (5.13) agrees with that found in Refs. 10 and 13.

In the case $|\xi_{l,l+g}|\gg\Omega_T$, again to within a factor of the order of 1,

$$W_{l,l+g}\approx\frac{2\Delta_0^2}{\xi_{l,l+g}}\left(\frac{|\xi_{l,l+g}|}{\omega}\right)^{2b}\frac{2\pi b}{1-\exp(-\xi_{l,l+g}/T)}.\tag{5.14}$$

By comparing this expression with (5.10) we see that the polaron effect has now ceased to depend on T altogether:

$$\tilde{\Delta}_0\approx\Delta_0(|\xi_{l,l+g}|/\omega)^b.\tag{5.15}$$

Here, as follows from comparison with (5.7), the size of the polaron narrowing has also decreased. This is because a real transition with a change in the particle energy includes the time scale $\sim1/\xi$, rendering ineffective the contribution to the polaron effect from excitations with energies smaller than $\xi_{l,l+g}$. On the other hand, since the phase volume created in the inelastic interaction of electron-hole pairs is now determined by the quantity $|\xi_{l,l+g}|$, rather than T, the frequency $\Omega_{l,l+g}$ (5.9) for $\xi_{l,l+g}>0$ ceases to depend on temperature (for $\xi_{l,l+g}<0$ a trivial Gibbs factor arises). As a result, in this case the transition probability (5.14) does not depend on T at all.

In the general case the macroscopic diffusion coefficient should be found from the solution of the equation for the distribution function f_l of the particles in the site representation (see Ref. 15 for details):

$$\frac{\partial f_l}{\partial t}+\sum_g[W_{l,l+g}f_l-W_{l+g,l}f_{l+g}]=0.$$

The problem actually reduces to that of finding the conductance of a three-dimensional network with a random distribution of the resistances of the individual links. The answer depends on the nature of the statistical distribution of the relative level shifts $\xi_{l,l+g}$.

In obtaining the result of this and the previous sections, we proceeded from the implicit assumption that the state of the particle in each well is described by a stationary function and thus that the formation of the polaron cloud of the particle involves the participation of excitations of arbitrarily low energies. In fact, in a self-consistent approach it is necessary to take it into account that the particle spends a finite time τ in an individual cell and so components with $\delta E < 1/\tau$ do not have time to form in the anti-adiabatic part of the wave function (3.13). Therefore, the nonadiabatic energy interval is bounded not only from above but also from below. However, for $|\xi_{\mathbf{l},\mathbf{l}+\mathbf{g}}| \gg \tilde{\Delta}_0$

$$1/\tau \ll |\xi_{\mathbf{l},\mathbf{l}+\mathbf{g}}|$$

and the real low-end cutoff occurs at the much higher frequencies $|\xi_{\mathbf{l},\mathbf{l}+\mathbf{g}}|$ or Ω_T. Moreover, for $\Omega_T \gg z^{1/2}\tilde{\Delta}_0$ the results are valid even for $\xi_{\mathbf{l},\mathbf{l}+\mathbf{g}} \to 0$. In fact, as follows from (5.6), the correlator appearing in the initial expression (5.1) falls off at long times as

$$\exp(-\Omega_T t). \tag{5.16}$$

This means that the integral builds up within a time $\sim 1/\Omega_T$. From the result obtained above, however, it follows that

$$1/\tau \approx 2z(\tilde{\Delta}_0/\Omega_T)^2\Omega_T \ll \Omega_T$$

and the cutoff again occurs at frequencies much larger than $1/\tau$. Thus all the results are valid not only for a static disruption of the band ($\xi \gg \tilde{\Delta}_0$) but also for a dynamic one ($\Omega_T\tilde{\Delta}_0$). The quantity Ω_T is just the characteristic frequency of the relative jitter of the levels in adjacent wells [see Eq. (5.20) below]. We note that the asymptotic form (5.16) is a consequence of the anomalous behavior of the integrand in the argument of the exponential function in (5.5) as $y \to 0$ on account of the finite density of states for excitations near the Fermi surface.

The situation is fundamentally different when

$$\Omega_T < \tilde{\Delta}(T) = 2z\tilde{\Delta}_0(T) \tag{5.17}$$

and at the same time there is no static shift of the levels. In this case there is a itinerant motion and the particle leaves the unit cell after a time $\tau \sim 1/\tilde{\Delta}(T)$. Now components with $\delta E < \tilde{\Delta}(T)$ do not have time to form in the site wave function of the particle.

To describe the itinerant motion it is necessary to determine the band width Δ_* corresponding to coherent motion of the particle (tunneling without excitation of the electronic system), which is governed by the diagonal matrix elements of the operator in (4.8). In evaluating the latter we must take into account the low-end cutoff $\delta E \sim \Delta_*$ on the energies of the electron-hole pairs. Then to logarithmic accuracy we have

$$\Delta_* = 2z\Delta_0^*, \quad \Delta_0^* = \Delta_0 \exp\left[-b\ln\frac{\omega}{(\Delta_*, T)_{max}}\right]. \tag{5.18}$$

To describe the itinerant motion in the general case it is necessary to use the Bloch-function representation. However, as was found for $T > \Delta_*$ [which is compatible with (5.17) for $b \ll 1$], the kinetic equation for the density matrix $\hat{\rho}$ of the particle admits a unitary transformation to the site representation, in which the kinetic equation is of the form (see Refs. 14, 17, and 18)

$$\frac{\partial\rho_{\mathbf{n},\mathbf{n}+\mathbf{l}}}{\partial t} + i\Delta_0^* \sum_{\mathbf{g}} (\rho_{\mathbf{n}+\mathbf{g},\mathbf{n}+\mathbf{l}} - \rho_{\mathbf{n},\mathbf{n}+\mathbf{l}+\mathbf{g}}) = -I_{\mathbf{n},\mathbf{n}+\mathbf{l}}. \tag{5.19}$$

For $\mathbf{l} \neq 0$ the collision integral, with the scattering taken into account by perturbation theory ($b \ll 1$), is given by the expression

$$I_{\mathbf{n},\mathbf{n}+\mathbf{l}} = \Omega_{\mathbf{l}}\rho_{\mathbf{n},\mathbf{n}+\mathbf{l}},$$

where for the case of an interaction with electrons [see Eq. (2.8)] we have

$$\Omega_{\mathbf{l}} = \pi \sum_{\mathbf{k},\mathbf{k}',\sigma} |V_{\mathbf{k},\mathbf{k}'}(0) - V_{\mathbf{k},\mathbf{k}'}(\mathbf{l})|^2 \bar{n}_{\mathbf{k}}(1-\bar{n}_{\mathbf{k}'})\delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}). \tag{5.20}$$

In the mixed representation that arises upon the Fourier transformation of (5.19) with respect to $\mathbf{n}$, we have

$$\frac{\partial\rho_{\mathbf{l}}(\mathbf{q})}{\partial t} + i\Delta_0^* \sum_{\mathbf{g}} \rho_{\mathbf{l}+\mathbf{g}}(\mathbf{q})(e^{-i\mathbf{q}\mathbf{g}} - 1) = -\Omega_{\mathbf{l}}\rho_{\mathbf{l}}(\mathbf{q}). \tag{5.21}$$

In treating macroscopic diffusion, when the concentration changes over distances $L \gg a$ and when $qa \ll 1$, we can take

$$qa\Delta_0^*/\Omega_{\mathbf{g}} \ll 1. \tag{5.22}$$

In this case the density matrix $\rho_{\mathbf{l}}(q)$ falls off rapidly with increasing $\mathbf{l}$, and system of equations (5.21) can be reduced to the equations for $\mathbf{l} = \mathbf{g}$ and $\mathbf{l} = 0$, in which for $\mathbf{l} \neq 0$ one can drop the time derivative (see Refs. 14, 17, and 18 for details). The equation for $\mathbf{l} = 0$ reduces to the ordinary diffusion equation

$$\partial\rho_0(\mathbf{q})/\partial t = -q^2 D(q)\rho_0(\mathbf{q})$$

with a diffusion coefficient

$$D = \frac{za^2}{3}\frac{(\Delta_0^*)^2}{\Omega_{\mathbf{g}}}. \tag{5.23}$$

Evaluating $\Omega_{\mathbf{g}}$ from (5.20), we immediately find $\Omega_{\mathbf{g}} = 2\pi bT$. This agrees with expression (5.8) for Ω_T. Interestingly, the coefficient of hopping diffusion (5.12) in the case under discussion goes over directly to the coefficient of band diffusion (5.23). This result is due to the specific character of the electronic polaron effect and does not obtain in the case of the ordinary phononic polaron effect (cf. Ref. 17). Here two circumstances are important. On the one hand, under condition (5.22) only the nondiagonal density-matrix elements of the form $\rho_{\mathbf{n},\mathbf{n}+\mathbf{g}}$, i.e., only the correlations for adjacent sites, are important in the itinerant motion. On the other hand, in both the coherent and hopping diffusion the governing role is played by relative fluctuations of the levels in adjacent wells (see Refs. 17 and 18); this circumstance is reflected in the fact that $\Omega_{\mathbf{g}}$ and Ω_T are the same.

For $T < T' = \Delta_*(T')$ the width of the band goes to a constant value which is determined self-consistently from relation (5.18):

$$\Delta_* = \Delta(\Delta/\omega)^{b/(1-b)}, \tag{5.24}$$

where $\Delta = 2z\Delta_0$ is the width of the band in the absence of the polaron effect.

For $T, \Omega_T \ll \Delta_*$ the particle moves in itinerant manner and is scattered by electronic excitations of energy T which do not take part in the formation of the polaron effect, as is clear from the preceding discussion. In this case one can find the diffusion coefficient by using the ordinary quasiclassical kinetic equation and treating the interaction with the thermal electronic excitations by perturbation theory ($b < \frac{1}{2}$). Let $T \ll \Delta_*$. Then it can be assumed that the particle has a quadratic dispersion relation $\varepsilon(p) = p^2/2M_*$, where $M_* = 3/z\Delta_0^* a^2$, after which the solution is found by the standard method. The momentum transferred by the particle to the electrons is limited to the scale of the thermal momentum $p_T \approx (2M_* T)^{1/2}$. Therefore, for $p_T \ll p_F$ the inverse mean free time is $\sim \Omega_T (p_T/p_F)^2$, and the diffusion coefficient, with allowance for the mean square velocity $\langle v^2 \rangle \sim T/M_*$ of the particle, turns out to be

$$D = \beta \frac{a^2 (\Delta_0^*)^2}{\Omega_T} (p_F a)^2 \sim \frac{1}{T}. \qquad (5.25)$$

An analogous temperature dependence was found in Ref. 19.

In the case of semimetals and degenerate semiconductors, the opposite limiting case $p_F \ll p_T$ or $p_F a \ll (T/\Delta_0^*)^{1/2} < 1$ can become important in a certain temperature interval. Here the particle in colliding with the electrons is scattered at a small angle $\theta \sim p_F/p_T$, but there is no restriction on the scattering angles of the electrons. In this case, the inverse transport mean free time is $\sim \Omega_T (p_F/p_T)^2$, and instead of (5.25) we have a dependence

$$D(T) \approx \beta' \frac{a^2}{(p_F a)^2} \frac{T^2}{\Omega_T} \sim T. \qquad (5.26)$$

There are two points of interest here: the vanishing of the dependence on the width of the band and the decrease of the diffusion coefficient with decreasing T. In the limit of extremely low T, dependence (5.25) is restored.

6. CONCLUDING REMARKS

The results obtained in the previous section solve the problem of the quantum diffusion of a heavy particle in the electron fluid of a metal at all temperatures. What is remarkable in these results is that although the adiabatic approximation is energywise valid for a metal to an accuracy of the order of $\omega/\varepsilon_F \ll 1$, there is an electronic polaron effect in the tunneling transition of the particle to an adjacent cell. Granted, the scale of the polaron effect turns out to be substantially diminished and is determined by the ratio of the inverse time $\sim\omega$ for passage of the heavy particle under the barrier to the largest of the following three parameters: the relative shift ξ of the levels, the temperature T, and the renormalized band width Δ_*. The electronic polaron effect vanishes when T is comparable to ω. In obtaining these results it was of fundamental importance to allow for the nature of the intra-well and under-barrier motion of the particle, with the possibility of virtual transitions between energy levels of the particle in an individual well. In this connection it should be noted that the usual allowance for only one level in each well, as is typically made, for example, when the spin Hamiltonian model is used for the two-well problem (see, e.g., Ref. 20), implicitly corresponds to the assumption of nonadiabaticity and therefore leads to an incorrect result.

The treatment given in the present study actually corresponds to allowance for the interaction with the electrons in the Born approximation. In principle, to find the polaron effect the problem can be generalized to take into account the screening of the potential of the particle[7] or to make a consistent allowance for scattering of arbitrary strength.[5,6,7,21]

In the actual case the particle interacts with phonons as well as with electrons. This leads to another overlap integral, now over the wave functions of the atoms forming the lattice in which the particle is moving. The two overlap integrals are independent, and the true value of $\tilde{\Delta}_0(T)$ is obtained by simply multiplying the quantities found in the previous section by the ordinary phononic polaron exponential J_{ph}. In the three-dimensional case the argument of this exponential does not have any singularities as $T \to 0$. Therefore, the overlap integral that is diagonal in the phonon occupation numbers is finite at any T.

A characteristic feature of the interaction with phonons in the case of narrow bands is that the polaron effect is due to single-phonon interaction, whereas the relative fluctuations of the levels in adjacent wells, at least for small ξ, is due to two-phonon interaction.[14,15,17] (For $\xi = 0$, single-phonon scattering processes are forbidden by the conservation laws.) It was found in the papers cited that the transition probability in the case of a purely phononic mechanism of interaction is described for $T > \xi$ by an expression of form (5.9) with

$$\begin{aligned} \Omega_{ph}^{(2)} &\approx 10^6 \Theta_D (T/\Theta_D)^9, \quad T \ll \Theta_D, \\ \Omega_{ph}^{(2)} &\approx B_2 \Theta_D (T/\Theta_D)^2, \quad T \gg \Theta_D, \end{aligned} \qquad (6.1)$$

where Θ_D is the Debye temperature, and $B_2 \sim 1$.

When ξ is appreciable, single-phonon processes also become possible. Here (see, e.g., Ref. 15)

$$(\Omega_{ph}^{(1)})_{1,1+g} \approx B_1 \left(\frac{\xi_{1,1+g}}{\Theta_D} \right)^4 \frac{\xi_{1,1+g}}{1 - \exp(-\xi_{1,1+g}/T)}. \qquad (6.2)$$

If the particle diffuses via interstitial sites which are energetically equivalent but are inequivalent from the standpoint of the principal axes of the strain tensor, then the transport effect goes away, and to a first approximation the exponent 9 in (6.1) is replaced by 7 (simultaneously, the numerical factor changes from 10^6 to 10^4), and the exponent 4 in (6.2) is replaced by 2 (see Refs. 22 and 23).

From a comparison of (6.1) and (6.2) with $\Omega_{1,1+g}$ (5.9) and Ω_T (5.8), we easily conclude that in ordinary metals with $(p_F a) \sim 1$ at low temperatures (actually up to $T \sim \Theta_D$) the governing role is played by the interaction with the electrons. This means that quantum diffusion in ordinary metals will be described by the expressions found in the preceding section if only the following replacement is made:

$$\tilde{\Delta}_0(T), \ \Delta_0^* \to \tilde{\Delta}_0(T) J_{ph}(T), \quad \Delta^* (J_{ph}(0))^{1/(1-b)}. \qquad (6.3)$$

We note that for $\xi = 0$ the electronic polaron effect becomes weaker with increasing temperature (5.7), whereas the phonon effect becomes stronger. Granted, at appreciable values of T, comparable to Θ_D, it becomes necessary to take into account the tunneling transitions that are nondiagonal in the phonons ("phonon shake-up"), and this alters the picture.

In semimetals and degenerate semiconductors the density of electrons and holes is small, and therefore $(p_Fa) \ll 1$. Then it follows immediately from (5.4) that

$$b \sim (p_F a)^2 \ll 1. \tag{6.4}$$

This result has two consequences. First, the electronic polaron effect is largely eliminated. Second, the phononic scattering mechanism can become predominant. By comparing (5.8) in this case with (6.1), we can determine the temperature T_2 above which the two-phonon scattering predominates:

$$T_2 = \Theta_D (10^6 n_e^{-2/3})^{-1/8} \quad \text{or} \quad T_2 = \Theta_D (10^4 n_e^{-2/3})^{-1/6} \tag{6.5}$$

(the second expression holds in the absence of the transport effect).

The relationship between $\Omega_{1,1+g}$ (5.9) and $\Omega^{(1)}_{ph\,1,1+g}$ (6.2) depends on the size of the shift of the levels in adjacent wells. While in typical metals the electron scattering is practically always dominant, in semimetals, on the contrary, the large-shift region in which $\Omega^{(1)}_{ph} \gg \Omega_{1,1+g}$ always obtains.

We note that in an ideal semimetal or degenerate semiconductor the character of the temperature dependence of the diffusion coefficient is quite nontrivial. In fact, for $T > T_2$ the diffusion coefficient D goes as $\sim 1/T^9$. On decreasing temperature, when T becomes smaller than T_2, this dependence is replaced by $D \sim T^{2b-1}$ [see Eq. (5.13)]. For $T \sim \Delta_*(T)$ it goes over, according to (5.26), to a dependence $D \sim T$ and, finally, for $T < \Delta_*(p_Fa)^2$, the dependence $D \sim 1/T$ (5.25) is established.

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