[3] M. Blume and Y. Y. Hsieh, J. Appl. Phys. 40, 1249 (1969).
[4] M. Nauciel-Bloch, G. Sarma, and A. Castets, Phys. Rev. B5, 4603 (1972).
[5] H. H. Chen and P. M. Levy, Phys. Rev. Lett. 27, 1383 (1971).
[6] S. V. Tyablikov, Metody kvantovoĭ teorii magnetizma (Methods in the Quantum Theory of Magnetism), Nauka, M., 1965 (English translation published by Plenum, N. Y., 1967).
[7] S. B. Haley and P. Erdös, Phys. Rev. B5, 1106 (1972).
[8] M. E. Lines and E. D. Jones, Phys. Rev. 141, 525 (1966).
[9] J. Sivardiere and M. Blume, Phys. Rev. B5, 1126 (1972).
[10] N. P. Grazhdankina, Usp. Fiz. Nauk 96, 291 (1968) [Sov. Phys.-Uspekhi 11, 727 (1969)].
[11] P. M. Levy and H. H. Chen, Phys. Rev. Lett. 27, 1385 (1971).
[12] G. Will and W. Schäfer, J. Phys. C4, 811 (1971).

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169

Polarons in the strong coupling limit

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The problem of the continuum model of a polaron is considered in the strong-coupling limit. The polaron-phonon interaction Hamiltonian is derived from the electron-phonon interaction Hamiltonian. A formula is derived for the zero-order (with respect to the coupling constant) correction to the polaron ground-state energy. It is shown that the polaron mobility is determined by two-phonon processes, viz., the scattering of phonons by the polaron. An upper limit is obtained for the mobility.

A polaron is customarily defined as an electron interacting with the non-dispersive lattice vibrations in an ionic crystal. The formulation of this problem is due to Pekar.[1] The parameters of the problem are the electron mass m and charge e, the frequency ω_0 of the optical phonons, and the values of the crystal dielectric constant in the low- and high-frequency limits, ϵ_0 and ϵ_∞. The dimensionless combination of these quantities

$$\alpha=\frac{e^2}{\hbar\omega_0}\left(\frac{m\omega_0}{2\hbar}\right)^{1/2}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right) \tag{1}$$

determines the strength of the coupling between the electron and the lattice.

We shall be solely interested in the case of strong coupling, $\alpha \to \infty$. In this limit Pekar[1] found that the polaron's ground state energy was given by $E_0 = -0.1094\alpha^2\hbar\omega_0$. The polaron mass, $M = 0.023\alpha^4 m$, was calculated by Landau and Pekar.[2] The numerical coefficients in the formulas for E_0 and M are determined from the solution of the dimensionless Schrödinger equation (see Eq. (4) below). It is necessary to assume that the lowering of the ground state energy and the increase of the mass at the expense of the interaction with phonons takes the major part of the electron-phonon interaction into account. The remaining part of the interaction must determine the corrections to E_0 and M, and it must also cause the scattering of the polaron by real phonons. It is necessary, in particular, to take this scattering into account in order to calculate the polaron mobility. A systematic strong-coupling theory of the polaron should give a method for calculating physical quantities in the form of a power series in the reciprocal coupling constant. Such an approach has been developed by Bogolyubov[3] and Tyablikov.[4] They reproduced the results of Pekar and Landau for the polaron's energy and mass as the first-approximation in their approach. The major difficulty in this approach[3,4] has turned out to be the introduction of polaron coordinates together with the electron coordinates. The condition that the total number of independent dynamical variables must be conserved upon introducing the polaron coordinates required the determination of three subsidiary conditions on the phonon coordinates. This fact led to very cumbersome calculations, and the investigation of the polaron's kinetics has not been completed. The articles by Pekar,[1] Feynman, Hellwarth, Iddings, and Platzman,[5] Kadanoff,[6] and Feynman and Thornber,[7] are devoted to the investigation of the polaron's kinetics. Without going into the details of these articles, we note the striking disagreement between the results obtained in these articles for the polaron mobility in the limit $\alpha \to \infty$. The indicated circumstances justify an attempt to construct a theory of the polaron by means of a systematic expansion in powers of the inverse coupling constant. The ultimate goal is to investigate the kinetics of the polaron.

In the present work a polaron-phonon Hamiltonian will be derived which is suitable for the description of weakly excited states of the crystal near the energy E_0 in the limit $\alpha \to \infty$, and the corrections, some of which do not have the form of a Hamiltonian, are estimated. The calculation of the correction to the ground state energy E_0 will be reduced to a variational principle. We shall show that the polaron mobility is determined by two-phonon processes, namely, the scattering of a phonon by the polaron. It will be shown that the momentum transfer associated with a collision is small, so that the polaron mobility is related to the amplitude for the scattering of a phonon by a polaron by a simple relationship. Analysis of the equation for the scattering amplitude enables us to obtain an upper bound for the polaron mobility.

DERIVATION OF THE POLARON-PHONON HAMILTONIAN

The Lagrangian of an electron interacting with the phonons in an ionic crystal has the form[8]

$$\mathscr{L}=\frac{1}{2}\dot{\mathbf{r}}^2+\frac{1}{2}\sum_{\mathbf{k}}(\dot{q}_{\mathbf{k}}\dot{q}_{-\mathbf{k}}-q_{\mathbf{k}}q_{-\mathbf{k}})+(2\sqrt{2}\pi\alpha)^{1/2}\sum_{\mathbf{k}}\frac{q_{\mathbf{k}}}{k}e^{i\mathbf{k}\mathbf{r}}. \tag{2}$$

Here $\mathbf{r}$ and $\dot{\mathbf{r}}$ denote the position and velocity of the electron, $q_{\mathbf{k}}$ and $\dot{q}_{\mathbf{k}}$ represent the coordinate and velocity of a phonon with quasi-momentum $\mathbf{k}$, and α is the electron-phonon constant. For simplicity we set $\hbar = m = \omega_0 = 1$. The factor $V^{-1/2}$ in front of the last term in Eq. (2) has also been replaced by unity, so that

$$\sum_{\mathbf{k}} \to \int\frac{d\mathbf{k}}{(2\pi)^3}.$$

The quantum formalism in the Lagrangian formulation is described in[8] and requires an averaging of $\{\mathbf{r}(t), q_{\mathbf{k}}(t)\}$ over the classical trajectories, where the relative weight of each trajectory is determined by the classical action, calculated for our problem with the aid of the Lagrangian (2).

Now let us describe a method of overcoming the principal difficulty in the polaron problem, which is the transition from electron coordinates to polaron coordinates. In addition to an integration over the trajectories of the electron and of the phonons, we introduce one more integration—over the trajectories $\mathbf{R}(t)$. Since $\mathbf{R}(t)$ is not contained in expression (2), the integration will give a numerical factor, which cancels out upon evaluating the thermodynamic averages. One further transformation, which does not change the value of the path-integrals, consists in replacing $q_{\mathbf{k}}(t)$ by $q_{\mathbf{k}}(t) + q_{\mathbf{k}}^0(t)$, where $q_{\mathbf{k}}^0(t) = q_{\mathbf{k}}^0 \exp[-i\mathbf{k}\cdot\mathbf{R}(t)]$ and $q_{\mathbf{k}}^0$ does not depend on the time. Now the Lagrangian explicitly depends on $\mathbf{R}(t)$ and $\dot{\mathbf{R}}(t)$, but this dependence is fictitious—it corresponds to the transformation to a moving reference frame and can be eliminated by the inverse shift of $q_{\mathbf{k}}(t)$. Let us choose

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$q_{\mathbf{k}}^0$ in such a way that the individual terms in the Lagrangian (or in the corresponding Hamiltonian) can be classified according to the powers of the coupling constant α to which they are proportional. Having separated out the major part of the Hamiltonian and the perturbation, and having calculated the corrections to the ground level of the Hamiltonian according to perturbation theory, we arrive at an approximate description of the weakly excited states in the crystal; the electron coordinates vanish and the Hamiltonian of a particle with coordinate $\mathbf{R}$, interacting with phonons, is obtained.

In the intermediate calculations it is convenient to utilize the Routhian, written down in terms of quantum operators for certain coordinates and in terms of classical variables for the other coordinates. The interpretation of the corresponding expressions is obvious: it is necessary to calculate the trace with regard to the quantum variables for a fixed classical trajectory and then average over the trajectories.

Let us make the replacement $q \to q + q^0$ in Eq. (2) and go over to the operator expression for the electron. We obtain the following expression for the Routhian:

$$\mathscr{R}=\frac{p^2}{2}-(2\sqrt{2}\pi\alpha)^{1/2}\sum_{\mathbf{k}}\frac{q_{\mathbf{k}}^0}{k}e^{i\mathbf{kr}}+\frac{1}{2}\sum_{\mathbf{k}}q_{\mathbf{k}}^0q_{-\mathbf{k}}^0-\frac{1}{2}\sum_{\mathbf{k}}(\dot{q}_{\mathbf{k}}\dot{q}_{-\mathbf{k}}+2\dot{q}_{\mathbf{k}}^0\dot{q}_{-\mathbf{k}}-q_{\mathbf{k}}q_{-\mathbf{k}})-\frac{1}{2}\sum_{\mathbf{k}}\dot{q}_{\mathbf{k}}^0\dot{q}_{-\mathbf{k}}^0-\sum_{\mathbf{k}}q_{\mathbf{k}}\left[(2\sqrt{2}\pi\alpha)^{1/2}\frac{e^{i\mathbf{kr}}}{k}-q_{-\mathbf{k}}^0\right]. \quad (3)$$

If $\mathbf{R}(t)$ is assumed to be time independent, the sum of the first three terms determines the energy of the crystal when the electron interacts with the polarization well produced by the displacements $q_{\mathbf{k}}^0$, and the coordinate $\mathbf{R}$ determines the position of the well in space. The problem of determining the minimum value of this energy has been treated in detail by Pekar.[1] We shall assume that the wave functions $\psi_n(\mathbf{r})$ and the energies E_n of the electron in the polarization well are known, these values corresponding to the ground state of the system and the well being located at the origin of coordinates The Schrödinger equation for these quantities is obtained by minimizing with respect to $q_{\mathbf{k}}^0$ the sum of the first three terms in (3) under the condition that the electron exists in the state $\psi_0(\mathbf{r})$:

$$\left(-\frac{\Delta}{2}-\frac{\alpha}{\sqrt{2}}\int\frac{d\mathbf{r}'\psi_0^2(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}+\frac{\alpha}{2\sqrt{2}}\iint\frac{d\mathbf{r}'d\mathbf{r}''\psi_0^2(\mathbf{r}')\psi_0^2(\mathbf{r}'')}{|\mathbf{r}'-\mathbf{r}''|}\right)\psi_n(\mathbf{r})=E_n\psi_n(\mathbf{r}). \quad (4)$$

Only the nonlinear equation for ψ_0 is self-consistent. The functions ψ_n with $n > 0$ are the solutions of Eq. (4) which minimize the electron energy for a fixed shape of the well. The function $\psi_0(\mathbf{r})$ doesn't have any singularities on the real axis—it can be expanded in a series in powers of r^2. It is clear from Eq. (4) that the characteristic values of $r \sim \alpha^{-1}$; therefore it is natural to assume that in the complex r plane the singularity of the function $\psi_0(\mathbf{r})$ which is closest to the real axis is located at a distance $\sim\alpha^{-1}$ from the axis.

For the equilibrium values of the phonon coordinates we have $q_{\mathbf{k}}^0 = v_{00}(\mathbf{k})$, where we have introduced the notation

$$v_{nm}(\mathbf{k})=(2\sqrt{2}\pi\alpha)^{1/2}\frac{1}{k}\int d\mathbf{r}\psi_n(\mathbf{r})e^{i\mathbf{kr}}\psi_m(\mathbf{r}). \quad (5)$$

The state ψ_0 with energy E_0 is the lowest; the following states are separated from it in energy by an amount $\sim\alpha^2$, and we construct a perturbation theory based on the functions ψ_n in order to finally derive a new Hamiltonian which describes the crystal at energies close to E_0, and the new Hamiltonian doesn't contain any terms which are large according to the coupling constant α.

The immediate goal is to write down expression (3) in terms of the electron functions ψ_n for the case when $q_{\mathbf{k}_0}^0$ is time-dependent, namely, the case when $q_{\mathbf{k}}^0(t) = q_{\mathbf{k}}^0\exp[-i\mathbf{k}\cdot\mathbf{R}(t)]$. The second term in (3) will then correspond to the polarization well being located at the point $\mathbf{R}(t)$, and we can rewrite (3) in terms of the functions $\psi_n(\mathbf{r}-\mathbf{R}(t))$. The motion of the well causes the appearance of an additional term in the Hamiltonian; we can understand the form of this term in the following manner. Let the Hamiltonian $\mathscr{H} = \mathscr{H}(\mathbf{r}-\mathbf{R}(t))$. After making the substitution $\psi = \sum_n c_n(t)\psi_n(\mathbf{r}-\mathbf{R}(t))$ the Schrödinger equation,

$$i\frac{\partial\psi}{\partial t}=\mathscr{H}(\mathbf{r}-\mathbf{R}(t))\psi \quad (6)$$

takes the following form in the representation of eigenfunctions of the Hamiltonian:

$$\left(i\frac{\partial c_n}{\partial t}=E_nc_n-\dot{\mathbf{R}}\sum_m\mathbf{p}_{nm}c_m, \quad \mathbf{p}_{nm}=\int\psi_n\mathbf{p}\psi_m\,d\mathbf{r}.\right) \quad (7)$$

It is clear that the motion of the well is taken into consideration by the term $-\dot{\mathbf{R}}\cdot\mathbf{p}$ in the Hamiltonian, so that in the representation of the operators a_n^+ and a_n which create and annihilate an electron in the state ψ_n and after changing from the velocities $\dot{q}_{\mathbf{k}}$ to the momenta $p_{\mathbf{k}} = (\dot{q}_{-\mathbf{k}} + \dot{q}_{-\mathbf{k}}^0)$ we obtain

$$\mathscr{R}=\sum_n E_na_n^+a_n-\dot{\mathbf{R}}\sum_{nm}\mathbf{p}_{nm}a_n^+a_m-\sum_{\mathbf{k}nm}q_{\mathbf{k}}e^{i\mathbf{kR}}[v_{nm}(\mathbf{k})-v_{00}(\mathbf{k})\delta_{nm}]a_n^+a_m+\frac{1}{2}\sum_{\mathbf{k}}[(p_{\mathbf{k}}-\dot{q}_{-\mathbf{k}}^0)(p_{-\mathbf{k}}-\dot{q}_{\mathbf{k}}^0)+q_{\mathbf{k}}q_{-\mathbf{k}}]-\frac{1}{2}M\dot{R}^2. \quad (8)$$

The quantity M, defined by the formula

$$M=\frac{1}{3}\int\frac{d\mathbf{k}}{(2\pi)^3}k^2q_{\mathbf{k}}^0q_{-\mathbf{k}}^0=\frac{8\sqrt{2}\pi^2}{3}\alpha\int_0^\infty\psi_0^4r^2\,dr\approx 0{,}023\alpha^4, \quad (9)$$

is the effective mass of the polaron, calculated by Landau and Pekar.[2]

The Routhian (8) was obtained as the result of exact transformations. Now let us consider the case when the energy of the crystal differs very little from E_0. Namely, we shall assume that $M\dot{R}^2 \lesssim 1$, we shall consider the electron to be in its ground state, and we shall calculate the corrections to its energy E_0 according to perturbation theory. Considering that the motion of the phonons is α^2 times slower than the electron's motion, we shall neglect the time dependence of $q_{\mathbf{k}}$ and R in calculating the corrections to E_0. This corresponds to the adiabatic approximation and should lead to relative errors of the order of α^{-2}.

Now let us prove that the second term in (8) can be neglected for large values of α. In fact, in second-order perturbation theory the correction to the energy E_0 due to this term is given by

$$\dot{R}^2\frac{1}{3}\sum_n{}'\frac{|\mathbf{p}_{0n}|^2}{E_0-E}\sim\dot{R}^2, \quad (10)$$

so that $p_{n0} \sim \alpha$ and $E_n - E_0 \sim \alpha^2$. By comparing $\dot{R}^2$ with $M\dot{R}^2 \sim \alpha^4\dot{R}^2$, we see that the relative correction to the mass turns out to be $\sim\alpha^{-4}$. Considering that the second term in (8) by itself doesn't cause any polaron scattering, it will give corrections to the scattering process of the same order of magnitude as the correction to the mass which has just now been estimated. Being interested in the case $\alpha \to \infty$, we neglect these corrections.

Let us return the phonon operator in (8) to its standard form by a canonical transformation with the aid of the operator $\exp[i\sum_{\mathbf{k}} q_{\mathbf{k}}\dot{q}_{-\mathbf{k}}^0(t)]$. This transformation depends on the time; therefore the following term is added to the Hamiltonian:

$$\sum_{\mathbf{k}}q_{\mathbf{k}}\ddot{q}_{-\mathbf{k}}^0(t)=\sum_{\mathbf{k}}q_{\mathbf{k}}q_{-\mathbf{k}}^0e^{i\mathbf{kR}}[-(\mathbf{k}\dot{\mathbf{R}})^2+i\mathbf{k}\ddot{\mathbf{R}}]. \quad (11)$$

First let us consider the term involving $(\mathbf{k}\cdot\dot{\mathbf{R}})^2$. It gives corrections of two types—it changes the kinetic energy of the polaron and leads to the scattering of the polaron by phonons. In second-order perturbation theory the correction to the kinetic energy of the polaron coming from the term involving $(\mathbf{k}\cdot\dot{\mathbf{R}})^2$ in (11) is negative, and its order of magnitude is given by

$$\dot{R}^4\sum_{\mathbf{k}}q_{\mathbf{k}}^0q_{-\mathbf{k}}^0k^4\sim M\dot{R}^2\cdot\alpha^2\dot{R}^2. \quad (12)$$

The negativeness of this correction indicates a reduction in the effective mass of the polaron with increasing values of its velocity, which corresponds to a decrease in the number of virtual phonons forming the polaron. One can say that the coupling constant decreases with increasing velocity of the polaron. The correction (12) to the kinetic energy becomes important when $\dot{R} \sim \alpha^{-1}$. In this connection $M\dot{R}^2 \sim \alpha^2$, which proves the statement made in[2] that a parabolic dispersion law is valid for the polaron as long as the kinetic energy is smaller than the ground state energy. In what follows we shall confine our attention to the case $M\dot{R}^2 \lesssim 1$, and therefore we neglect corrections of the type (12). The contribution of the interaction (11) to the scattering of the polaron will be estimated later, when the principal term in the polaron-phonon interaction will be written down. Having in mind the transition to the Hamiltonian form of notation with respect to the coordinate $\mathbf{R}$, the difficulties of realizing this transition for expressions of the type (11), and also the fact that this expression will be discarded in the final analysis, we shall not write down the corresponding term in the Hamiltonian. With what has been said taken into account, it is now necessary to neglect the second term in the Routhian (8), and the fourth term is replaced by the free-phonon Hamiltonian:

$$\mathscr{R}=-\frac{1}{2}M\dot{R}^2+\frac{1}{2}\sum_{\mathbf{k}}(p_{\mathbf{k}}p_{-\mathbf{k}}+q_{\mathbf{k}}q_{-\mathbf{k}})+\sum_n E_na_n^+a_n-\sum_{\mathbf{k}}q_{\mathbf{k}}e^{i\mathbf{kR}}[v_{nm}(\mathbf{k})-v_{00}(\mathbf{k})\delta_{nm}]a_n^+a_m. \quad (13)$$

The last term in Eq. (13) should be treated as a perturbation. Evaluating the correction to the energy due to this term in second-order perturbation theory, and also changing from the velocity $\mathbf{R}$ to the momentum $\mathbf{P}$ of the polaron, we obtain the final expression for the polaron-phonon Hamiltonian:

$$\mathscr{H}=E_0+\frac{P^2}{2M}+\frac{1}{2}\sum_{\mathbf{k}}(p_{\mathbf{k}}p_{-\mathbf{k}}+q_{\mathbf{k}}q_{-\mathbf{k}})+\sum_{\mathbf{kk}'}q_{\mathbf{k}}q_{-\mathbf{k}'}V(\mathbf{k},\mathbf{k}')e^{i(\mathbf{k}-\mathbf{k}')\mathbf{R}}, \quad (14)$$

where the following notation has been introduced:

$$V(\mathbf{k},\mathbf{k}')=\sum_{m>0}\frac{v_{0m}(\mathbf{k})v_{m0}(-\mathbf{k}')}{E_0-E_m}. \quad (15)$$

We write down the following estimate for the correction to the Hamiltonian in third-order perturbation theory:

$$-\sum_{\mathbf{kk}'\mathbf{k}''}q_{\mathbf{k}}q_{\mathbf{k}'}q_{\mathbf{k}''}e^{i(\mathbf{k}+\mathbf{k}'+\mathbf{k}'')\mathbf{R}}\sum_{l,m>0}\frac{v_{0l}(\mathbf{k})v_{lm}(\mathbf{k}')v_{m0}(\mathbf{k}'')}{(E_0-E_l)(E_0-E_m)}. \quad (16)$$

Later on the following property of the function $V(\mathbf{k},\mathbf{k}')$ will be important for us:

$$\int\frac{d\mathbf{k}''}{(2\pi)^3}V(\mathbf{k},\mathbf{k}'')V(\mathbf{k}'',\mathbf{k}')\sim V(\mathbf{k},\mathbf{k}'), \qquad k,k'\sim\alpha. \quad (17)$$

We shall talk about such functions since they are of the order of unity.

Now let us sum up the situation. Having started from expression (2), describing an electron interacting with phonons, we changed over to the new coordinate $\mathbf{R}$, determining the position of the polaron, and we derived the Hamiltonian (14) for the polaron + phonons system. The electron coordinates were eliminated in connection with the transition from (12) to (14) at the expense of the fact that we can only apply the derived Hamiltonian for states of the crystal which are close in energy to E_0. One should consider expressions (11) and (16) as corrections to this Hamiltonian. We note that the principal term of the polaron-phonon interaction is quadratic in the phonons, the correction (11) is linear, and the correction (16) is cubic.

Tyablikov[4] obtained a term in the Hamiltonian which differs from the last term in (14) by the replacement of $\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}]$ by unity. This led to the erroneous conclusion that the phonons are not coupled to the polaron in the present approximation. The presence of the indicated factor is obvious from considerations of translational invariance.

The derivations which we have carried out make sense only in that case when the resulting polaron-phonon interaction which we have obtained is weak in comparison with the original electron-phonon interaction. Namely, the relative corrections to the energy and mass of the polaron due to the polaron-phonon interaction must be small, and the polaron damping due to the interaction with phonons—emission, absorption, and scattering of phonons—must be small in comparison with the polaron's energy. In the meanwhile we have proved that the correction from expression (11) changes the kinetic energy of the polaron for $M\dot{R}^2 \sim 1$ by an amount $\sim\alpha^{-2} \ll 1$. Now let us consider the corrections to the energy coming from the third term in expression (14). Having averaged it over the ground state, we obtain a quantity of the order of

$$\int V(\mathbf{k},\mathbf{k})\frac{d\mathbf{k}}{(2\pi)^3}=\sum_{m>0}\frac{1}{E_0-E_m}\int\frac{d\mathbf{k}}{(2\pi)^3}|v_{0m}(\mathbf{k})|^2\sim 1, \quad (18)$$

because $v_{nm}(\mathbf{k}) \sim \alpha^{-1/2}$, and momenta $k \sim \alpha$ give the essential contribution to the integral. This estimate shows that the interaction of the polaron with the phonons is of the order of unity as $\alpha \to \infty$. We recall that the correction to the energy, evaluated for the electron-phonon Hamiltonian, was of order α, so that we have obtained an effective weakening of the interaction. All of the terms of the perturbation theory series with regard to the last term in expression (14) are of the order of unity.

Now let us go on to a more convenient notation, in order to distinguish the small parameter existing in the Hamiltonian (14). This parameter corresponds to the smallness of the recoil energy of the polaron associated with the emission of a phonon. In fact, in view of the fact that $M \sim \alpha^4$ and $k \sim \alpha$, the recoil energy $k^2/2M \sim \alpha^{-2}$. Let us change in expression (14) to second-quantized operators for the phonons and let us carry out the transformation of Lee, Low, and Pines,[9] thus eliminating the polaron coordinate from the Hamiltonian:

$$\mathscr{H}=E_0+\frac{1}{2M}\Big(\mathbf{P}-\sum_{\mathbf{k}}\mathbf{k}b_{\mathbf{k}}{}^{+}b_{\mathbf{k}}\Big)^2+\frac{1}{2}\sum_{\mathbf{k}}(b_{\mathbf{k}}{}^{+}b_{\mathbf{k}}+b_{\mathbf{k}}b_{\mathbf{k}}{}^{+})$$
$$+\frac{1}{2}\sum_{\mathbf{k}\mathbf{k}'}V(\mathbf{k},\mathbf{k}')\,(b_{\mathbf{k}}{}^{+}+b_{-\mathbf{k}})\,(b^{+}_{-\mathbf{k}'}+b_{\mathbf{k}'}). \qquad (19)$$

The Hamiltonian now depends parametrically on the total momentum P of the system, which is just a number. As long as the energy of the crystal differs from the ground state energy by an amount which is smaller than the energy of a phonon, the momentum of the crystal coincides with the polaron momentum. In the transformed Hamiltonian the last two terms represent a quadratic form which cannot be diagonalized in general. At the same time, calculation of the coefficients $V(\mathbf{k}, \mathbf{k}')$ requires knowledge of the wave functions of the excited states of the electron in the polarization well. The problem of the diagonalization of literally this same quadratic form was treated in a different notation in work by the author and Rashba.[10] It was shown that an infinite number of local phonon levels exists near the polaron, and a variational principle was formulated to calculate the wave functions and frequencies of these levels. In this connection it was possible to transform the variational functional in such a way that it was expressed only in terms of the polaron's ground state wave function $\psi_0(\mathbf{r})$:

$$\frac{1-\omega^2}{16\pi}=\max_{f}\Big\{\int d\mathbf{r}\,\psi_0{}^2(\nabla f)^2\Big/\int d\mathbf{r}[\nabla(\Delta f+2\nabla f\nabla\ln\psi_0)]^2\Big\}. \qquad (20)$$

Here f is the variational function, and ω is the frequency of the local vibration. The variational functional does not contain any parameters; therefore ω differs from unity numerically. The ground state energy of the polaron now takes the form

$$E=E_0+\frac{1}{2}\sum(\omega_\alpha-1). \qquad (21)$$

Formulas (20) and (21) enable us to calculate the correction to E_0, which is of the order of unity, and moreover the problem of calculating the frequencies ω_α reduces to a convenient form.

Now let us prove that the corrections to Eq. (21) are small. The corrections arising from the term containing $(\mathbf{k}\cdot\dot{\mathbf{R}})^2$ in Eq. (11) is of the order of

$$\sum_{\mathbf{k}} q_{\mathbf{k}}{}^0 q_{-\mathbf{k}}{}^0\Big(\frac{k^2}{M}\Big)^2\sim\alpha^{-6}, \qquad (22)$$

if it is assumed that the mean square fluctuation of the velocity $\overline{\dot{\mathrm{R}}^2}\sim(\mathrm{k}/\mathrm{M})^2$, where k is the characteristic phonon momentum $\mathrm{k}\sim\alpha$. Taking into consideration that the characteristic frequencies are equal to unity, we have $\ddot{\mathrm{R}}^2\sim(\mathrm{k}/\mathrm{M})^2\sim\alpha^{-6}$ for the mean square acceleration, so that the correction to the energy coming from the term $\mathbf{k}\cdot\ddot{\mathbf{R}}$ is of the order of α^{-2}. The term $(\sum_{\mathbf{k}}\mathbf{k}b^{+}_{\mathbf{k}}b_{\mathbf{k}})^2/2\mathrm{M}$ in Eq. (19) serves (at P = 0) as one more source of a correction to the energy; it is necessary to combine this term with the last term. The first nonvanishing correction is of order α^{-4}. Finally, the cubic correction (in the phonon variables) to the Hamiltonian (16) gives a contribution $\sim\alpha^{-2}$ in second-order perturbation theory.

The correction to the polaron mass is obtained, for example, if the term $-\mathbf{P}\cdot(\sum_{\mathbf{k}}\mathbf{k}b^{+}_{\mathbf{k}}b_{\mathbf{k}})/\mathrm{M}$ in Eq. (19) is taken into account in perturbation theory. It has a relative order α^{-2}.

The estimates presented here prove that one can regard the polaron-phonon interaction as weak in the sense that the correction to the ground state energy, having the order of unity, can be isolated and a method for its calculation is indicated, the remaining corrections are at least as small as α^{-2}, and the corrections to the mass are also of relative order α^{-2}.

POLARON MOBILITY

The kinetics of the polaron is determined by its interaction with real phonons. According to the Hamiltonian (14), processes are possible involving the simultaneous emission or absorption of a pair of phonons and the scattering of a phonon by the polaron with a change of the momentum. The correction (11) leads to the absorption or emission of a single phonon, and the correction (16) to the Hamiltonian gives rise to three-phonon processes. The intensities of the enumerated types of scattering are quite diverse. In connection with the emission or absorption of a single phonon, a polaron with energy of the order of unity must give up or gain an amount of momentum $\mathrm{P}\sim\alpha^2$. According to Eq. (11), the matrix element for the absorption or emission contains the quantity $q^0_{\mathbf{k}}$ which for $\mathrm{k}\sim\mathrm{P}\sim\alpha^2$ must be at least exponentially small in the coupling constant:

$$q_{\mathbf{k}}{}^0=\int\psi_0{}^2(\mathbf{r})\,e^{i\mathbf{k}\mathbf{r}}\,d\mathbf{r}\sim e^{-ck/\alpha},\qquad k\gg\alpha, \qquad (23)$$

where c is the number determining the distance from the real axis to the nearest singularity of the function $\psi_0(\mathbf{r})$ in units of α^{-1}. (We indicated earlier that $\psi_0(\mathbf{r})$ does not have any singularities on the real axis, and it was assumed that it has singularities in the complex r plane. If all of the singularities of $\psi_0(\mathbf{r})$ are located at infinity, then $q^0_{\mathbf{k}}\sim\exp(-c_1\mathrm{k}^2/\alpha^2)$ which makes $q^0_{\mathbf{k}}$ even smaller for $\mathrm{k}\sim\alpha^2$.) In similar fashion, all real processes involving a change in the number of phonons will be suppressed by a factor that is exponential in the coupling constant. In contrast to emission and absorption, the scattering of phonons does not involve any large momentum transfers, and its amplitude decreases only in power-law fashion. To estimate the mobility it is therefore necessary to take only the processes of phonon scattering by the polaron into account.

Let us consider the polaron mobility in the temperature interval $1>\mathrm{T}>\alpha^{-2}$. The condition $\mathrm{T}<1$ guarantees that the number of real phonons will be small, so that their collisions with the polaron can be regarded as independent. Under the conditions $\mathrm{T}>\alpha^{-2}$ the polaron momentum $\mathrm{P}\sim\sqrt{\mathrm{MT}}\sim\alpha^2\mathrm{T}^{1/2}$ exceeds the momentum transfer during the collision $|\mathbf{P}-\mathbf{P}'|\sim\alpha$. One can therefore assume that the polaron's velocity $\mathbf{v}=\mathbf{P}/\mathrm{M}$ does not change during the phonon scattering process, and determine the amplitude $\mathrm{F}(\mathbf{v}, \mathbf{k}, \mathbf{k}')$ for the scattering of the phonon from the state $\mathbf{k}$ into the state $\mathbf{k}'$ by a polaron moving with constant velocity $\mathbf{v}$. The mobility of the polaron is expressed in terms of $\mathrm{F}(\mathbf{v}, \mathbf{k}, \mathbf{k}')$ by the formulas

$$\mu=\frac{1}{3T}\int v^2\tau_{tr}(v)\,e^{-Mv^2/2T}\,d\mathbf{v}\Big(\int e^{-Mv^2/2T}\,d\mathbf{v}\Big)^{-1}, \qquad (24)$$

$$\frac{1}{\tau_{tr}(v)}=2\pi e^{-1/T}\int\frac{d\mathbf{k}\,d\mathbf{k}'}{(2\pi)^6}|F(\mathbf{v},\mathbf{k},\mathbf{k}')|^2\delta[\mathbf{v}(\mathbf{k}-\mathbf{k}')]\frac{(\mathbf{k}-\mathbf{k}')^2}{2M^2v^2}. \qquad (25)$$

The transition to a polaron moving with constant velocity is accomplished by neglecting the terms in the Hamiltonian (19) which are quadratic in the phonon momenta. The resulting Hamiltonian

$$H=E_0+\frac{P^2}{2M}+\sum_{\mathbf{k}}(1-\mathbf{k}\mathbf{v})\Big(b_{\mathbf{k}}{}^{+}b_{\mathbf{k}}+\frac{1}{2}\Big)$$

$$+\frac{1}{2}\sum_{\mathbf{k}\mathbf{k}'}V(\mathbf{k},\mathbf{k}')\,(b_{\mathbf{k}}{}^{+}+b_{-\mathbf{k}})\,(b^{+}_{-\mathbf{k}'}+b_{\mathbf{k}'}) \qquad (26)$$

describes the scattering of phonons with the spectrum $1-\mathbf{k}\cdot\mathbf{v}$ by a fixed center, where the Born scattering amplitude is given by the quantity $\mathrm{V}(\mathbf{k}, \mathbf{k}')$. The energy denominators in the perturbation theory expressions for $\mathrm{F}(\mathbf{v}, \mathbf{k}, \mathbf{k}')$ are small, since $\mathrm{v}|\mathbf{k}-\mathbf{k}'|\sim\mathrm{v}\alpha\sim\mathrm{T}^{1/2}\alpha^{-1}\ll1$; therefore, the order of magnitude of F can be estimated from the equation

$$F(\mathbf{v},\mathbf{k},\mathbf{k}')=V(\mathbf{k},\mathbf{k}')-\frac{1}{v}\int\frac{V(\mathbf{k},\mathbf{k}'')F(\mathbf{v},\mathbf{k}'',\mathbf{k}')}{\mathbf{n}(\mathbf{k}''-\mathbf{k}')-i\delta}\frac{d\mathbf{k}''}{(2\pi)^3}, \qquad (27)$$

where $\mathbf{n}=\mathbf{v}/\mathrm{v}$.

Let us begin the investigation of this equation with an estimate of $\mathrm{V}(\mathbf{k}, \mathbf{k}')$. As was indicated earlier, for k, k' $\sim\alpha$, V is of the order of unity in the sense of Eq. (17), which gives the following estimate for its absolute magnitude:

$$V(\mathrm{k},\mathrm{k}')\sim\alpha^{-3};\quad k,k'\sim\alpha. \qquad (28)$$

For k, k' $\gg\alpha$ and $|\mathbf{k}-\mathbf{k}'|\sim\alpha$, one can approximately calculate the vertex $\mathrm{V}(\mathbf{k}, \mathbf{k}')$. In order to do this, in expression (15) for V it is sufficient to use plane waves with momenta ~k, k' as the intermediate states of the continuum. As a result we obtain

$$V(\mathbf{k},\mathbf{k}')\approx-\frac{4\sqrt{2}\pi\alpha}{k^4}\psi_0{}^2[\mathbf{k}-\mathbf{k}'],\qquad k,k'\gg\alpha,\qquad|\mathbf{k}-\mathbf{k}'|\sim\alpha, \qquad (29)$$

where

$$\psi_0{}^2[\mathbf{k}]\equiv\int\psi_0{}^2(\mathbf{r})\,e^{i\mathbf{k}\mathbf{r}}\,d\mathbf{r}. \qquad (30)$$

From Eq. (29) it is clear that $\mathrm{V}(\mathbf{k}, \mathbf{k}')$ decreases with increasing values of k and k'; therefore, at large momenta the integral term in Eq. (27) becomes inessential and F approximately coincides with V. Equation (27) can be solved by iterations in this momentum range. An estimate based on the first iteration, with Eq. (29) and the fact that the momentum transfer $|\mathbf{k}''-\mathbf{k}'|\sim\alpha$ taken into consideration, leads to our conclusion about the validity of the iterative solution in the region $\mathrm{k}^4>\alpha^3\mathrm{v}^{-1}$, where

$$F(\mathbf{v},\mathbf{k},\mathbf{k}')\approx-\frac{4\sqrt{2}\pi\alpha}{k^4}\psi_0{}^2[\mathbf{k}-\mathbf{k}'],\qquad k^4>\alpha^3v^{-1}. \qquad (31)$$

The integrand in (25) is positive; therefore, one can obtain an upper bound on the mobility from Eqs. (24) and (25). It is convenient to change to the variables $\mathbf{q}=\mathbf{k}-\mathbf{k}'$ and $\mathbf{k}$ in the integral (25). We shall assume that the integral over $\mathbf{q}$ converges for $\mathrm{q}\sim\alpha$; this assumption can only lead to an underestimate of the value of the integral. Let us carry out the integration with respect to $\mathbf{k}$ over the region $\mathrm{k}^4>\alpha^3\mathrm{v}^{-1}$, where formula (31) is applicable. Finally we obtain the estimate $\mu\lesssim\mathrm{e}^{1/\mathrm{T}}\mathrm{T}^{7/8}\alpha^{-7/4}$, which can be improved immediately. The point is that when the asymptotic expression (29) for V is substituted in (27), the latter turns out to be an equation of the convolution type and can be solved without any difficulty, so that the series in powers of the parameter $\alpha^3/\mathrm{k}^4\mathrm{v}$ is completely gathered together. Substitution of the solution into formula (25) shows, however, that all of the terms of the series in powers of the parameter $\alpha^3/\mathrm{k}^4\mathrm{v}$ cancel each other in the integrand. The reason for this lies in the fact that the series in $\alpha^3/\mathrm{k}^4\mathrm{v}$ adds up to form the purely imaginary phase of the function $\mathrm{F}(\mathbf{v}, \mathbf{k}, \mathbf{k}')$, which drops out from the absolute value. Thus, the integrand in Eq. (25) must be expanded at least in powers of the parameter $\alpha^4/\mathrm{k}^5\mathrm{v}$, which permits us to use expression (31) for F in the region $\mathrm{k}^5\gtrsim\alpha^4/\mathrm{v}$. Integration over this region leads to the estimate

$$\mu\lesssim e^{\omega_0/T}T/m\omega_0{}^2\alpha^2, \qquad (32)$$

where we have returned to dimensional units (also see[11]).

The upper bound on the temperature is due to the requirement that the number of real phonons should be small. The lower bound on T is written down in order to guarantee a small momentum transfer in the collision in comparison with the total momentum of the polaron; this enables us, in particular, to write down the simple expression (24) for the mobility. In fact the criterion $\mathrm{T}>\alpha^{-2}$ also has a deeper meaning—it is only under this condition that a kinetic equation exists for the polaron. In fact, it was indicated earlier that the uncertainty in the polaron's momentum due to the interaction with the zero-point vibrations of the phonons is of the order of α. The concept of the polaron as a particle having a definite momentum, which changes only as a result of collisions with real phonons, and which is distributed on the average according to Maxwell's formula, is valid as long as the thermal momentum $\mathrm{P}\sim\alpha^2\mathrm{T}^{1/2}$ is larger than the momentum fluctuations α. Thus, we have shown that the extension of this theory to temperatures on either side of the region $1>\mathrm{T}>\alpha^{-2}$ is not a trivial problem.

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[1] S. I. Pekar, Issledovaniya po élektronnoĭ teorii kristallov (Investigations in the Electronic Theory of Crystals), Gostekhizdat, 1951.
[2] L. D. Landau and S. I. Pekar, Zh. Eksp. Teor. Fiz. **18**, 419 (1948).
[3] N. N. Bogolyubov, Ukr. mat. zhurn. **2**, 3 (1950).
[4] S. V. Tyablikov, Zh. Eksp. Teor. Fiz. **21**, 377 (1951).
[5] R. P. Feynman, R. W. Hellwarth, C. K. Iddings, and P. M. Platzman, Phys. Rev. **127**, 1004 (1962).
[6] L. P. Kadanoff, Phys. Rev. **130**, 1364 (1963).
[7] K. K. Thornber and R. P. Feynman, Phys. Rev. **B1**, 4099 (1970).
[8] R. P. Feynman and A. R. Hibbs, Quantum Mechanics and Path Integrals, McGraw-Hill, 1965 (Russ. Transl., Mir, 1968).
[9] T. D. Lee, F. E. Low, and D. Pines, Phys. Rev. **90**, 297 (1953).
[10] V. I. Mel'nikov and É. I. Rashba, ZhETF Pis. Red. **10**, 95 (1969) [JETP Lett. **10**, 60 (1969)]; Erratum in **10**, 359 (1969) [JETP Lett. **10**, 228 (1969)].
[11] G. E. Volovik, V. I. Mel'nikov, and V. M. Édel'shteĭn, ZhETF Pis. Red. **18**, 138 (1973) [JETP Lett. **18**, 81 (1973)].

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170