

Band structure in autolocalization and bipolaron models of high-temperature superconductivity

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(Received 25 May 1994; revised manuscript received 3 March 1995)

We develop the polaron theory taking into account the spatial dispersion of lattice polarizability caused by several phonon branches. This allows us to describe new effects which cannot be obtained in the classical polaron theory because of the limitations of its model medium that has only one dispersionless phonon branch interacting with the carrier. It is demonstrated that if the spatial dispersion is allowed, the carrier spectrum in a medium with many-component polarization proves to have an autolocalization band structure. It occurs due to limitation on the polaron velocity in accordance with Landau's theory of the quantum liquid. This results in several effects. One of them is "Cherenkov" radiation by a sufficiently rapid polaron of the coherent medium vibrations. Owing to this radiation there exists a possibility that two polarons can couple under resonance conditions with the medium vibrations. It leads to the formation of a two-center bipolaron coupled due to the exchange of real phonons. The influence of the autolocalization band structure on the properties of a system with high carrier concentration is also studied. This is of interest as far as the properties of high-temperature superconductors are concerned. Like any complex oxides they are characterized by many-component polarization. The obtained limitation on the velocity of any autolocalized state causes both the existence of maximum polaron concentration (of the order of 10^{20} cm^{-3}) and modification of the Bose-condensation condition for bipolarons.

I. INTRODUCTION

The theory of a large-radius polaron describes the practically important and widespread case of dielectric screening of a charge carrier in a medium with sufficiently strong electron-phonon coupling. Nevertheless, this theory has a surprisingly small number of experimental confirmations and practical applications. One of the most important experimental results indicating the presence of autolocalized carriers of a large-polaron type is the extremely large energy losses of the carrier moving through an Al_2O_3 crystal.¹ Having analyzed all energy-loss mechanisms Tornber and Feynman² came to the conclusion that the only mechanism of such losses can be optical phonon scattering of the carrier which is in a polaronic state. This means that a carrier autolocalized state can be formed even in crystals with intermediate electron-phonon interaction such as the Al_2O_3 crystal which is characterized by a Fröhlich constant of about 3.² Polarons were also observed in several other experiments such as photoinduced infrared absorption,³ normal-state tunneling,⁴ and Hall measurements⁵ on the complex oxides demonstrating high- T_c superconductivity at high carrier concentration. The polaron-based interpretation is also possible for the unusual conductivity behavior in BaTiO_3 .⁶

Why are there so few experiments confirming the polaron theory? It so happened that a factor which has a great influence on the polaron motion was not usually taken into account by the polaron theory. This factor is

spatial dispersion of the lattice polarizability of crystals. Indeed, since the localized carrier can be connected with the coordinate system there exists a critical value for its velocity, as was shown in Landau's theory of the quantum liquid. This critical value is the minimum phase velocity of phonons. When the polaron exceeds this velocity it radiates real phonons so that its state is not stationary in the absence of an electric field. Moreover, it was shown^{7,8} that in principle the polaron with such velocities cannot be formed due to the reconstruction of the phonon vacuum.

Determined by the phonon dispersions the critical velocities are small in most crystals. This causes decomposition of polarons already at a temperature of about 5–15 K (at low carrier concentrations when Boltzmann statistics can be applied to polarons⁹). So the polarons at such concentrations can be observed only at very low temperatures.⁷ This fact was not taken into account by the classic polaron theory. All the basic studies in polaron theory were carried out neglecting the polarizability spatial dispersion, which was often misleading because polaron motion with a velocity higher than the critical one leads to polaron destruction.

Nevertheless, the results obtained in the studies of the polaron effective mass can easily be put on a real base presuming that the phonon dispersion is nonzero and the critical velocity is higher than the velocity of the polaron motion. But there are some works studying polaron braking caused by the polarization field.^{2,10} As will be shown below, for such effects to occur the polaron veloci-

ty must exceed the minimum phase velocity of phonons. If a model medium has one phonon branch^{2,10} we are faced with a contradiction because the carrier localization disappears when the polaron velocity exceeds its critical value (if there is no other factor in the carrier localization).

The simplest way to study such processes correctly is to introduce into the model another branch of optic phonons capable of providing the carrier localization. If the minimum phase velocity of phonons of the latter branch u_2 is higher than that of the first branch u_1 , a carrier moving with the velocity v in the interval $u_1 < v < u_2$ will also be in the polaronic state. Such a model medium allows us to investigate correctly the process of partial breaking of the polarization "cloud" enveloping the auto-localized carrier when its velocity exceeds the minimum phase velocity of phonons of one branch.

II. THE MEDIUM MODEL

In accordance with what was said above we shall consider a model medium with two-component lattice polarization caused by two phonon branches having different dispersion. This model differs from that possessing one branch of optic longitudinal phonons used in the classical polaron theory⁹ constructed for the alkali halides. Our model describes well most ionic crystals, for example, complex oxides (which have many ions in a unit cell and, consequently, many phonon branches).

Let us consider the case when two resonance phonon frequencies Ω_{TO1} and Ω_{TO2} satisfy the inequality $\Omega_{TO1} < \Omega_{TO2}$. The corresponding frequencies of longitudinal vibrations will be denoted as Ω_1 and Ω_2 ($\Omega_1 < \Omega_2$). The dipole moment associated with the vibration of each type will be characterized by inverse effective dielectric constants $c_1 = \epsilon_1^{-1} - \epsilon_0^{-1}$ (for the low-frequency resonance) and $c_2 = \epsilon_\infty^{-1} - \epsilon_1^{-1}$ (for the high-frequency one). Here ϵ_0 and ϵ_∞ are static and high-frequency dielectric constants and ϵ_1 can be determined from the Liddane-Sachs-Teller ratio $\epsilon_1 = \epsilon_\infty \Omega_2^2 \Omega_{TO2}^{-2}$.

Suppose the dispersion of longitudinal phonon branches follows the ordinary law

$$\Omega_i^2(k) = \Omega_i^2(0) + u_i^2 k^2, \quad i = 1, 2. \quad (1)$$

The minimum phase velocities of phonons u_i , $i = 1, 2$, are presumed to satisfy the inequality $u_1 < u_2$. In accordance with this we will denote the two phonon branches as low- and high-velocity (LV and HV) ones.

The Hamiltonian function for such a medium can be written in the form

$$H = \int d^3\mathbf{r} \sum_{i=1,2} \frac{2\pi}{c_i \Omega_i^2} \left[\Omega_i^2 \mathbf{P}_i^2 + \left[\frac{\partial \mathbf{P}_i}{\partial t} \right]^2 - u_i^2 \mathbf{P}_i \nabla_r^2 \mathbf{P}_i \right]. \quad (2)$$

Here \mathbf{P}_1 and \mathbf{P}_2 are the dipole moments associated with the two phonon branches under consideration. The term of the form $\mathbf{P}_i \mathbf{P}_j$ is absent in H because the vibrations belonging to different branches are independent in the har-

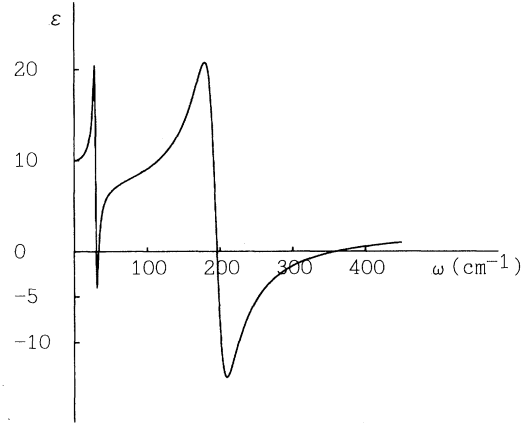


FIG. 1. An example of the dielectric permittivity dispersion in a crystal with two-component lattice polarization.

monic approximation used here.

The motion equation yielded by the Hamiltonian function (2) was usually written in terms of the polarization⁹ or polarization field potential.⁸ But to make the physics of the problem more apparent it will be convenient to write the motion equations for the distributions of the polarization charge density $\rho_i = -\text{div} \mathbf{P}_i$, $i = 1, 2$, associated with each of the two phonon branches:

$$\left[\frac{\partial^2}{\partial t^2} + \Omega_i^2 - u_i^2 \nabla_r^2 \right] \rho_i(\mathbf{r}, t) = 0. \quad (3)$$

Here we assume the medium to be isotropic. The dielectric permittivity of the medium under consideration can be obtained from (2) or (3) in the form

$$\epsilon(\omega, k) = \epsilon_\infty - \frac{c_1 \epsilon_1^2 \Omega_1^2}{\omega^2 - \Omega_{TO1}^2 - k^2 u_1^2 + i\omega\gamma_1} - \frac{c_2 \epsilon_\infty^2 \Omega_2^2}{\omega^2 - \Omega_{TO2}^2 - k^2 u_2^2 + i\omega\gamma_2} \quad (4)$$

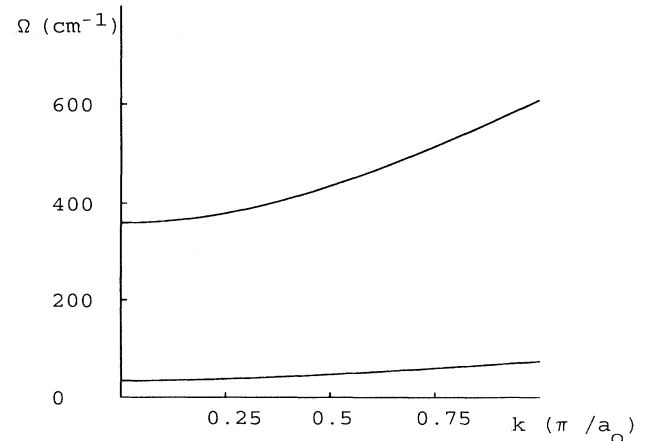


FIG. 2. An example of phonon dispersion satisfying the condition necessary for the autolocalization band structure effects to occur.

(where we make allowance for the decay). The example of the phonon dispersions and dielectric permittivity corresponding to the discussed model medium is demonstrated by Figs. 1 and 2.

III. BAND STRUCTURE OF THE CARRIER SPECTRUM CAUSED BY AUTOLOCALIZATION

The presence of the charge carrier in the system is described by the medium Hamiltonian function with additional terms:

$$H = \int d^3\mathbf{r} \left\{ \Psi^\dagger \left[E_0 - \frac{\hbar^2}{2m^*} \nabla_{\mathbf{r}}^2 \right] \Psi + \sum_{i=1,2} \left[\frac{2\pi}{c_i \Omega_i^2} \left[\Omega_i^2 P_i^2 + \frac{\partial P_i^2}{\partial t} - u_i^2 \mathbf{P}_i \nabla_{\mathbf{r}}^2 \mathbf{P}_i \right] - \mathbf{P}_i \mathbf{D} \right] \right\}, \quad (5)$$

$$\mathbf{D} = -e \nabla_{\mathbf{r}} \int \Psi^\dagger(\mathbf{r}', t) \Psi(\mathbf{r}', t) \frac{d^3\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|},$$

where Ψ is the carrier field operator, m^* is the effective mass of the "free" carrier in the conduction band, and the zero energy level is the bottom of this band. Since the phonon field is supposed to be classical the motion equations will have the form

$$\left\{ i\hbar \frac{\partial}{\partial t} - E_0 + \frac{\hbar^2}{2m^*} \nabla_{\mathbf{r}}^2 - e \int [\rho_1(\mathbf{r}', t) + \rho_2(\mathbf{r}', t)] \frac{d^3\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|} \right\} \psi = 0, \quad (6)$$

$$\left\{ \frac{\partial^2}{\partial t^2} + \Omega_i^2 - u_i^2 \nabla_{\mathbf{r}}^2 \right\} \rho_i(\mathbf{r}, t) = -c_i \Omega_i^2 \psi^2(\mathbf{r}, t) \quad \left. \begin{array}{l} \text{for } i=1 \\ \text{for } i=2, \end{array} \right\} \quad (7a, 7b)$$

where ψ is the real wave function of the autolocalized carrier state.

The system of the motion equations (6) and (7) can be solved in an ordinary manner⁸ using the direct variational method. The functional which must be minimized has the form

$$J = - \int d^3\mathbf{r} \psi(\mathbf{r}) \left\{ \frac{\hbar^2}{2m^*} \nabla_{\mathbf{r}}^2 + \frac{e^2}{2} \int d^3\mathbf{r}_1 \sum_i c_i \int \frac{d^3\mathbf{r}_2}{|\mathbf{r} - \mathbf{r}_1|} \times G_i(\mathbf{r}_1, \mathbf{r}_2) \psi^2(\mathbf{r}_2) \right\} \psi(\mathbf{r}), \quad (8)$$

where the unknown distributions $\rho_1(\mathbf{r}, t)$ and $\rho_2(\mathbf{r}, t)$ are expressed through the carrier charge distribution $\psi^2(\mathbf{r}, t)$ and the corresponding Green functions G_1 and G_2 :

$$\rho_i(\mathbf{r}, t) = -ec_i \Omega_i^2 \int G_i(\mathbf{r} - \mathbf{r}', t) \psi^2(\mathbf{r}', t) d^3\mathbf{r}'. \quad (9)$$

Considering a straight-line translational motion of the carrier with the constant velocity v parallel to the z axis of the cylindrical coordinate system we can find the Green function for Eq. (9) from the equation

$$\left\{ \frac{\partial^2}{\partial t^2} + \Omega_i^2 - u_i^2 \nabla_{\mathbf{r}}^2 \right\} G_i(\mathbf{r}, t) = \delta(\mathbf{r} - \mathbf{v}t). \quad (10)$$

The solution of this equation will give us a Green function which can be called a specialized one since the δ function in the right part of Eq. (10) fixes the form of the trajectory. It can easily be seen that the solution of such an equation is the polarization charge density distribution generated by a moving point-charge particle. It has the form^{7,8}

$$G_i(\mathbf{r}, t) = \begin{cases} \frac{\exp\{-\Omega_i [(z-vt)^2/\beta_{1i}^2 + r^2]^{1/2}/u_i\}}{4\pi u_i^2 \beta_{1i} [(z-vt)^2/\beta_{1i}^2 + r^2]^{1/2}}, & v < u_i, \beta_{1i}^2 = 1 - v^2/u_i^2 \\ \frac{\cos\{\Omega_i [(z-vt)^2/\beta_{2i}^2 - r^2]^{1/2}/u_i\}}{2\pi u_i^2 \beta_{2i} [(z-vt)^2/\beta_{2i}^2 - r^2]^{1/2}}, & v > u_i, z-vt < 0, r < |z-vt|/\beta_{2i} \\ 0, & v > u_i, |z-vt| < 0, r > |z-vt|/\beta_{2i}, |z-vt| > 0, \beta_{2i}^2 = v^2/u_i^2 - 1, \end{cases} \quad (11)$$

where $i=1,2$, $z \equiv z - z'$, and $r \equiv r - r'$. As is clear from (11) the Green function for Eq. (7), $G_i(\mathbf{r}, \mathbf{r}', t)$, experiences sharp reconstruction when the velocity of the carrier changes in the vicinity of the minimum phase velocity u_i : at $v < u_i$ it is localized near the point $\mathbf{r} - \mathbf{r}' - \mathbf{v}t = 0$ and at

$v > u_i$ it is a periodic function oscillating at infinity. As the carrier state is determined by the functional (8) strongly depending on the form of the Green functions there are obviously three different states of the carrier in a medium with two-component polarization and these

states occur in the different velocity intervals $v < u_1$, $u_1 < v < u_2$, and $v > u_2$.

If the translational motion velocity v is within the interval $[0, u_1]$, both polarization charge distributions ρ_1 and ρ_2 are obviously localized in the region of the carrier localization. We shall call this polaron having two polarization "clouds" due to two different phonon branches a double polaron (DP). The participation of both phonon branches in the DP formation can be confirmed by calculation of the total (free and screening) charge of the polaron:

$$q_{\text{tot}} = \frac{e}{\epsilon_\infty} + \int [\rho_1(\mathbf{r}, t) + \rho_2(\mathbf{r}, t)] d^3\mathbf{r}. \quad (12)$$

In the case of $v < u_1$ it turns out to be

$$q_{\text{tot}} = \frac{e}{\epsilon_\infty} - e(c_1 + c_2) = e/\epsilon_0. \quad (13)$$

To make the first approximation to the DP binding energy E_{DP} and radius R_{DP} we may use the formulas of the classical polaron theory with the effective inverse dielectric permittivity $c = c_1 + c_2 = \epsilon_\infty^{-1} - \epsilon_0^{-1}$. For a more precise description it is necessary to solve the system of equations (6) and (7). As for the DP effective mass m_{DP}^* it depends considerably on the frequencies of both phonons forming the polaron. Therefore, without solving the system of motion equations (i.e., using the well-known expressions obtained in the one-branch model medium) it is possible only to point out the interval containing m_{DP}^* .

As soon as the polaron velocity exceeds the u_1 value the term in (10) corresponding to the interaction of LV polarization with the carrier proves to tend to zero. Indeed, at such velocities v it is the result of integration of the rapidly oscillating [with the period $\lambda_z = 2\pi(v^2 - u_1^2)^{1/2}/\Omega_1$ along the z axis] Green function G_1 with the comparatively smooth function ψ^2 . This means that the LV polarization cannot contribute to localization of such a rapid polaron. Nevertheless, due to the interaction with the HV phonons an autolocalized state of the carrier is preserved at such velocities. This carrier state is, of course, different from the DP state because it has no polarization "cloud" of LV phonons. This fact can be demonstrated by the value of the total charge of the polaron moving with the velocity exceeding u_1 : $q_{\text{tot}} = e/\epsilon_1$, i.e., the charge carrier in such a state is screened only by the HV polarization and the carrier localization is maintained only by the HV phonon branch.

It is obvious that such a polaron [we shall call it a single polaron (SP)] has smaller binding energy and effective mass and a larger radius than the DP. To determine SP characteristics classical polaron theory formulas may be used since they are obtained for the case of carrier localization due to the interaction with phonons of one branch. However, it must be pointed out that analytical solution of the system (6) and (7) taking into account the polarizability spatial dispersion was found in the polaron theory only for the slight-dispersion case⁸ while here we shall be interested in the case of strong dispersion of the HV phonon branch.

When the velocity of the carrier translational motion

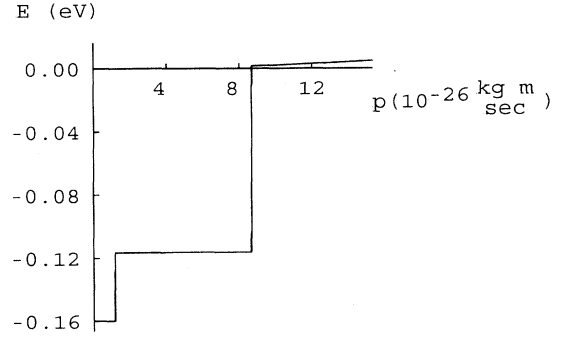


FIG. 3. Autolocalization band structure in the medium with two-component lattice polarization.

exceeds the higher of the two minimum phase velocities of phonons u_2 the Green function G_2 starts oscillating too, so that no factor of the carrier localization at such velocities exists. The higher average velocities are accessible only for delocalized carriers. Thus consideration of the model medium with two-component polarization allows us to describe the partial destruction of the autolocalized state with two polarization "clouds," when its motion is sufficiently rapid. It also enables us to predict the transition from one polaron state to another due to the change of its momentum.

The information obtained can be presented as the carrier energy versus average carrier momentum. Ignoring the carrier motion inside the polaron, the average carrier momentum describes its state with an uncertainty which is greater for the most localized DP, less for the SP, and negligible for the free carrier. An example of such a band structure of the carrier spectrum caused by autolocalization is shown in Fig. 3. There the energies are represented with respect to the bottom of the conduction band and the critical average momenta of the carrier corresponding to transitions between different carrier states are $\bar{p}_1 = m^*u_1$ and $\bar{p}_2 = m^*u_2$. As is clear from Fig. 3 two gaps appear in the carrier spectrum in the medium with two-component lattice polarization.

It is natural that only carrier states with sufficiently large lifetimes are depicted. The DP's, SP's, and free carriers may have momenta from other intervals but for a very short time. If a SP with the velocity $v < u_1$ appears in the medium it will be enveloped by the LV polarization during the time $\tau_{\text{SP}} = 2\pi/\Omega_1$. Similarly, the lifetime of the free carrier with $v < u_2$ is $\tau_e = 2\pi/\Omega_2$. The lifetime of the DP with the velocity $v > u_1$ can be evaluated as $\tau_{\text{DP}} = R_{\text{DP}}/(v - u_1)$.

IV. THE EFFECT OF COHERENT-PHONON RADIATION BY THE MOVING POLARON

The SP is not a stationary state in the whole velocity interval $u_1 < v < u_2$. When on increase of the SP velocity the period of oscillations λ_z of the Green function G_1 exceeds the SP diameter $2R_{\text{SP}}$, the result of integration (9) becomes substantially nonzero. It oscillates in an acoustic cone (named by analogy to the light cone) behind the moving polaron. If the inequality

$$v^2 - u_1^2 \gg [\Omega_1 R_{SP} / (2\pi)]^2 \quad (14)$$

is satisfied these oscillations are quasiharmonic. Such an effect in the motion of the point-charge particle has been described by Myasnikov and Popov.⁷ They pointed out that such a polarization charge distribution appears as a consequence of the coherent radiation of real phonons by a localized charged particle moving with a velocity exceeding the minimum phase velocity of the phonons, which is similar to Cherenkov's effect. In the case of polaron motion the radiation of different parts of the polaron charge density becomes coherent when the wavelength of the radiation becomes much larger than the polaron size.

Thus the single polaron moving with the velocity v satisfying inequality (14) has a tail of radiated LV polarization in the acoustic cone behind the polaron. This resembles a supersonic plane with its boom but here the "boom" can act on the "plane" to a more significant degree. Interaction of the SP with the LV polarization charge radiated by it leads to the slowing of the SP motion. As a consequence the carrier states with corresponding average momenta are not stationary in the absence of an external electric field. Knowing the form of the Green function G_1 (11), it is possible to find the field strength necessary to stabilize SP motion with a certain velocity. Since in the case of $v^2 - u_1^2 \gg 1$ the LV polarization charge concentrates to the highest degree on the SP trajectory, the braking force due to the radiated LV polarization at such velocities can be approximately evaluated from (11) and (9) as $F = c_1 \Omega_1^2 e^2 / v^2$. Equating it to the force caused by the field, one can express the equilibrium SP velocity v as a function of the field strength E : $v(E) = (c_1 \Omega_1^2 e / E)^{1/2}$. Thus in this velocity region the losses decrease with increase of the velocity. It is quite natural since the wavelength of the radiated wave of the LV polarization charge increases with the SP velocity. Bearing in mind that the losses are Ohmic when the polaron velocity is lower than u_1 and continuously increase when the polarization tail is formed, we can assume an approximate dependence $v(E)$.¹¹ Experimental observation of such behavior of the average carrier velocity or corresponding behavior of the carrier mobility versus electric field can be an indication of this effect of the autolocalized band structure.

The exact relationship between the steady-state velocity and the field strength necessary to support it was obtained in Ref. 2 and later in Ref. 10 by directly solving the motion equations written in terms of quantum consideration of the polarization field. However, using the model medium with one phonon branch the authors of Refs. 2 and 10 assumed that the autolocalized state of the carrier still exists when its velocity exceeds the minimum phase velocity of the phonons. Consequently, in their model polarization of one and the same type plays the role of a dissipative subsystem absorbing the energy of the rapid polaron motion and supports the carrier autolocalization. This contradiction can be easily eliminated if another phonon branch with higher dispersion is introduced into the model. In such a case the autolocalized state of the carrier is preserved when its velocity exceeds

the minimum phase velocity of the LV branch, so that both conditions for the coherent radiation turn out to occur: there is a localized particle moving with a velocity exceeding the critical one. Thus, the results of Refs. 2 and 10 are correct for the model medium with two phonon branches of different dispersions.

But the interpretation of these results can be corrected (in the case of Ref. 2) and expanded on the basis of the present study. Tornber and Feynman assumed² that the reason for the thresholdlike increase in energy losses versus average carrier velocity is the one-phonon process of carrier scattering. But numerical calculation² shows that a sharp increase of losses takes place when the average carrier velocity is 10–100 times lower than the velocity corresponding to the threshold of one-phonon scattering. On the basis of the present study that considers the problem dynamically this increase can be attributed to the radiation of real phonons by the moving localized charged particle. This radiation appears when on increase of the particle velocity the inequality

$$2R_{SP} > \lambda_z = \frac{2\pi}{\Omega_1} (v^2 - u_1^2)^{1/2}$$

becomes true, i.e., at the average carrier velocities

$$v^2 > u_1^2 + \left[\frac{2R_{SP}\Omega_1}{2\pi} \right]^2. \quad (15)$$

(In Ref. 2 u_1 is supposed to be zero.) As in most crystals, the threshold for the one-phonon processes in the Al_2O_3 crystals considered in Ref. 2 corresponds to velocities higher than the threshold of "Cherenkov" radiation (15).

In addition the present analysis enables us to clarify the question² of whether the quasiparticle representation is applicable to the system near the threshold of radiation. The authors of Ref. 2 proposed to use the free-electron mass for this velocity region instead of the polaron effective mass, using, however, the polaron wave function, which was contradictory. From the present analysis it can be concluded that coherent radiation (characterized by the decrease of losses with the increase of the carrier average velocity) takes place only if at such velocities there exists a localized state of the carrier (SP). The SP motion with such velocities can be described by its effective mass caused by HV phonons, taking into account the influence of the LV polarization wave separately.

V. RESONANCE EFFECTS IN THE SCREENING OF MOVING POLARONS

The model with two-component polarization will also enable us to consider a different mechanism of dielectric screening of the carrier leading to the formation of different stationary states of the autolocalized carriers. This mechanism takes place in specific types of motion of the autolocalized carriers. These specific types can be found using the results of Sec. III. There it was shown that the divergency of polarization radiated by a sufficiently rapid autolocalized carrier concentrates on its

trajectory. Therefore, when the changes of the SP motion direction are sufficiently large (for instance, due to scattering) the interaction with the LV polarization charge leads to a further change of the polaron trajectory. Thus straight-line motion of the autolocalized carrier with sufficiently high velocity turns out to be unstable. Moreover, it will be demonstrated below that there exist stationary states of the polaron or even two polarons moving in a circular orbit and coupled due to the interaction with the coherent phonon waves radiated by them.

Indeed, we have seen in Sec. III that the state of the carrier moving in a certain trajectory is determined by the form of a specialized Green function for the motion equation. This Green function represents the distribution of the polarization charge radiated by a point-charge particle moving in this trajectory. The authors of Ref. 12 have found an exact solution to the motion equation for a point-charge particle moving in a circular orbit in a polarizing medium. They have shown that there exist stationary solutions for this equation and even for the equation of motion of two charged particles (of the same sign of charge) in a circular orbit. These stationary states occurring due to the interaction with the radiated polarization were found¹² to correspond to a certain set of frequencies of the motion. Using these results let us show the possibility of analogous stationary states where the role of localized charged particles is played by polarons.

To do this let us find the solution for the motion equations (6) and (7) corresponding to the SP motion in a circular orbit of radius $R_0 \gg R_{SP}$ with frequency ω . The velocity of the SP $v_0 = \omega R_0$ is assumed to satisfy the inequality $u_1 \ll v_0 < u_2$. This allows us to make the following simplifying suppositions. Since $v_0 \gg u_1$, the third term in the left-hand side of (7a) is negligible. Equation (7b) may be replaced by the simpler Pecar expression for the polarization charge density in the polaron: $\rho_2(\mathbf{r}, t) = -c_2 \psi^2(\mathbf{r}, t)$. We shall denote the modified equations as (7a') and (7b').

In cylindrical coordinates with the origin at the orbit center and the z axis perpendicular to the orbit plane the specialized Green function for Eq. (7a') corresponding to periodic SP motion on the circular orbit can be obtained as the solution for the equation

$$\left[\frac{\partial^2}{\partial t^2} + \Omega_1^2 \right] G_1(\mathbf{r}, \mathbf{r}', t) = \delta(r - r') \delta(z - z') \delta(\varphi - \varphi' - \omega t) / r'. \quad (16)$$

For the case of the point-charge motion the solution is shown to exist only at frequencies from the set¹²

$$\omega_l = 2\Omega_1 / (2l + 1), \quad l = 0, 1, 2, \dots \quad (17)$$

In our case we shall also confine ourselves to considering such frequencies at which the solution of (16) has the form¹²

$$G_1'(\mathbf{r}, \mathbf{r}', t) = \frac{2l + 1}{4r'\Omega_1^2} \sin \left[\frac{2l + 1}{2} (\varphi - \varphi') - \Omega_1 t \right] \times \delta(r - r') \delta(z - z'). \quad (18)$$

From (17) the conclusion can be made that the stationary state of the charged particle motion appears as a result of resonance with the vibrations of the LV phonon branch. The motion of the localized charged particle plays here the role of a periodic but not harmonic external force. The frequencies (17) are half of the set of the parametric resonance frequencies. The other half corresponds to an infinite increase of the amplitude of the polarization charge wave due to the transmission of the energy of the polaron motion to the lattice.

To solve the system of equations (6), (7a'), and (7b') we can again apply a variational method. The wave function of the ground state must minimize the functional

$$J = - \int d^3\mathbf{r} \psi(\mathbf{r}) \left\{ - \frac{\hbar^2}{2m^*} \nabla_{\mathbf{r}}^2 + \frac{e^2}{2} \int \frac{d^3\mathbf{r}}{|\mathbf{r} - \mathbf{r}_1|} \times \left[c_1 \int d^3\mathbf{r}_2 G(\mathbf{r}_1, \mathbf{r}_2) \psi^2(\mathbf{r}_2) + c_2 \psi^2(\mathbf{r}_1) \right] \right\} \psi(\mathbf{r}). \quad (19)$$

As a test function we used a modified Pecar wave function

$$\psi(\mathbf{r}, t) \sim [\varphi^2 + (1 - R_0/r)^2]^{1/2} (1 + \alpha r') e^{-\alpha r' + \beta \varphi}, \quad (20)$$

$$r' = [(r - R_0)^2 + (r\varphi)^2 + z^2]^{1/2}, \quad \alpha = 0.9794c_2.$$

The expression for parameter α in (20) is the same as in a Pecar wave function.⁹ Besides a centripetal projection, the forces acting on the SP in such a state have projections deforming the polaron. As a result the SP wave function deviates from spherical symmetry. This deviation determines the value of parameter β in (20) which can be obtained by minimization of the functional (19). The third parameter in the wave function (20), the orbit radius R_0 , cannot be varied independently. It is determined by the equilibrium condition for the projections of forces acting on the SP onto the direction normal to the orbit.

The numerical computation performed for the case of $l=0$ in (18) has shown that the functional (19) has a minimum in a wide range of the medium parameters. The quasiparticle corresponding to this minimum is obviously a bound state of the SP and real LV phonons. The binding energy of the quasiparticle with respect to the SP energy can change from zero up to several hundredths of eV for different values of the medium parameters. This quasiparticle may be regarded as an excited state of the DP because it includes real LV phonons whereas the DP contains only virtual LV phonons.

As noted above, the polaron velocity in this quasiparticle must satisfy the inequality $u_1 \ll v < u_2$ and the orbit radius R_0 must exceed the SP radius. But even for the minimum values of R_0 and Ω_1 the polaron velocity in the quasiparticle cannot be lower than approximately 2.6×10^6 cm sec⁻¹ (if Ω_1 is about 35 cm⁻¹). Since there are no crystals with higher minimum phase velocities of phonons we shall not take into account this state in further consideration. This state was analyzed here with the

methodological purpose of simplifying the consideration of a stationary state of two polarons coupled by their radiation.

The Hamiltonian for the system with two carriers is obtained from (5) by adding the term

$$\frac{1}{2} \int \frac{\Psi^+(\mathbf{r})\Psi^+(\mathbf{r}')e^{2\Psi(\mathbf{r}')}\Psi(\mathbf{r})}{\varepsilon_\infty|\mathbf{r}-\mathbf{r}'|} d^3\mathbf{r}'d^3\mathbf{r}$$

corresponding to the interaction of carriers. It is con-

venient to use the self-consistent-field approximation supposing that the states of the carriers in such a system can be described by single-particle wave functions $\psi_1(\mathbf{r}, t)$ and $\psi_2(\mathbf{r}, t)$. The functions ψ_1 and ψ_2 are orthogonal to each other because they do not overlap. Supplementing them to obtain the complete set, substituting the expansion in terms of this set into the motion equation for Ψ , and averaging this equation over the state of the system in which only two carrier states characterized by ψ_1 and ψ_2 are filled, we obtain the following system of equations for ψ_1 and ψ_2 :

$$\left\{ -i\hbar \frac{\partial}{\partial t} + E_0 - \frac{\hbar^2}{2m^*} \nabla_{\mathbf{r}}^2 - e \left[\int [\rho_1(\mathbf{r}', t) + \rho_2(\mathbf{r}', t)] \frac{d^3\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|} + e/\varepsilon_\infty \int \psi_j^2(\mathbf{r}', t) \frac{d^3\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|} \right] \right\} \psi_i(\mathbf{r}, t) = 0, \quad i, j = 1, 2, \quad i \neq j. \quad (21)$$

Owing to the symmetry of the problem, the single-particle wave functions satisfy the condition

$$\psi_i(r, \varphi - \omega t, z) = \psi_j(r, \varphi - \omega t + \pi, z), \quad i, j = 1, 2, \quad i \neq j. \quad (22)$$

After this fact is taken into account the system of equations (21) becomes a pair of identical equations. Two equations for the polarization charge distributions obtained by a procedure similar to that used for Eq. (21) are given by

$$\left[\frac{1}{\Omega^2} \frac{\partial^2}{\partial t^2} + 1 \right] \rho_1(\mathbf{r}, t) = -ec_1 [\psi_1^2(\mathbf{r}, t) + \psi_2^2(\mathbf{r}, t)], \quad (23)$$

$$\rho_2(\mathbf{r}, t) = -ec_2 [\psi_1^2(\mathbf{r}, t) + \psi_2^2(\mathbf{r}, t)]. \quad (24)$$

Equations (21)–(23) form the system of equations which can be solved as in the previous cases if the expression (22) is taken into account.

The specialized Green function for Eq. (23) satisfies the equation

$$J = - \int d^3\mathbf{r} \psi_1(\mathbf{r}) \left\{ \frac{\hbar^2}{2m^*} \nabla_{\mathbf{r}}^2 + \frac{e^2}{2} \int \frac{d^3\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|} \left[c_2 \psi_1^2(\mathbf{r}') - \psi_2^2(\mathbf{r}')/\varepsilon_\infty + c_1 \int G_0(\mathbf{r}', \mathbf{r}_1) \psi_1^2(\mathbf{r}_1) d^3\mathbf{r}_1 \right] \right\} \psi_1(\mathbf{r}). \quad (28)$$

We minimized it numerically, using the test function (20) and equality (22). The parameter β in the wave function was determined by the minimization of functional (28). The orbit radius R_0 is to be obtained from the equilibrium condition for the SP (the sum of normal projections of all forces acting on the SP must be equal to the centripetal force $m_{\text{sp}}^* \Omega_1^2 R_0$). Figure 4 shows this equilibrium condition as the curve in the coordinates $R_0 - c_1$ calculated numerically for a certain set of the medium parameters. As seen from Fig. 4, the equation for the orbit radius has two solutions or it has no solution at all, depending on the medium parameters. The analysis shows

$$\left[\frac{1}{\Omega^2} \frac{\partial^2}{\partial t^2} + 1 \right] G(\mathbf{r}, \mathbf{r}', t) = \frac{1}{r'} \delta(r - r') \delta(z - z') \times [\delta(\varphi - \varphi' - \omega t) + \delta(\varphi + \pi - \varphi' - \omega t)]. \quad (25)$$

Myasnikov and Martynyuk¹² have found that it has stationary solutions of the form

$$G_n(\mathbf{r}, \mathbf{r}', t) = \frac{2n+1}{2r'} \sin[(2n+1)(\varphi - \varphi') - \Omega_1 t] \times \delta(z - z') \delta(r - r') \quad (26)$$

at the frequencies

$$\omega = \omega_n = \Omega_1 / (2n + 1), \quad n = 0, 1, 2, \dots \quad (27)$$

We shall confine ourselves to the consideration of the case of $n=0$. The single-particle wave function corresponding to the bound state is to minimize the functional

that the left branch of the parabola in Fig. 4 corresponds to the solutions stable under the influence of perturbations such as switching on and off the magnetic field, whereas the right one may correspond to unstable solutions. So we shall consider the quasiparticles with radii from the left branch.

The quasiparticle corresponding to the minimum of the functional (28) can be called a Bose quasiparticle (BQ) to emphasize its statistical properties and to distinguish it from the quasiparticle containing one SP. The BQ is a bipolaron but of an entirely different type. Indeed, the BQ contains real phonons (of the LV branch) and is coupled

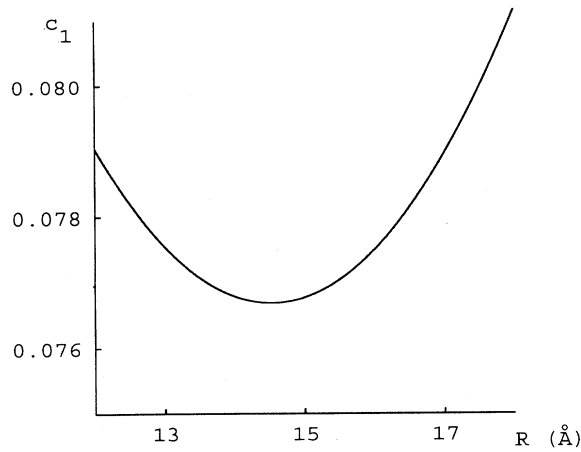


FIG. 4. An example of the calculated curve (in coordinates of BQ radius R_0 and inverse effective dielectric permittivity c_1) on which the sum of forces acting on the SP in the BQ is equal to the centripetal force $m_{sp}^* \Omega_0^2 R_0$.

due to the radiation and absorption of the waves of these phonons by the SP's, which can be regarded as the exchange with real phonons between two SP's. We shall also call the BQ a resonance polaron. As is clear from the comparison of (17) and (27), the velocities of the SP's in the Bose quasiparticle $v_0 = \omega_0 R_0$ are two times lower

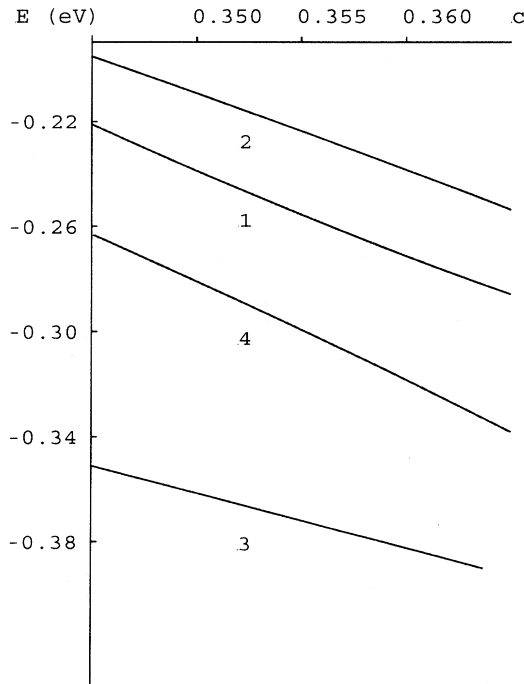


FIG. 5. Energies of the autolocalized carrier states in the medium with two-component polarization (with respect to the conduction-band bottom). Curves 1, the BQ energy; 2, the doubled SP energy; 3, the doubled DP energy; 4, the energy of the conventional bipolaron.

than those in the quasiparticle formed by one polaron so that in crystals with high dispersion of at least one phonon branch the BQ state can occur.

Figure 5 represents the minimum energies of all the autolocalized states occurring in a medium with two-component polarization versus the inverse effective dielectric permittivity $c = \epsilon_\infty^{-1} - \epsilon_0^{-1} = c_1 + c_2$. Zero energy corresponds to the bottom of the free-carrier band. The parameters of the medium in the demonstrated case are the following: $\Omega_1 = 34 \text{ cm}^{-1}$, $\Omega_2 = 360 \text{ cm}^{-1}$, $m^* = m_e$, $\epsilon_\infty = 2.2$, and the ratio $\epsilon_1/\epsilon_0 = 0.557$ is fixed. Curve 1 in Fig. 5 corresponds to the BQ energy calculated numerically; curves 2 and 3 show the doubled minimum energies of the DP and SP, respectively. Curve 4 demonstrates the energy of the most energetically profitable conventional bipolaron. It is a one-center bipolaron considered taking into account a nonzero average distance between the electrons in the potential hole. It was considered in semiclassical approximation in Ref. 13 and in many other works it was analyzed using quantum consideration of the polarization field (for example, Ref. 14). Its energy (calculated from the results of Ref. 13) is shown in order to compare it with the energy of the BQ.

As is seen from Fig. 5, for typical values of the medium parameters the binding energy of the BQ exceeds the doubled binding energy of the SP. Then, although the binding energy of the conventional bipolaron exceeds that of the resonance bipolaron the former states cannot be formed in most media with two (or more) components of polarization characterized by a significantly different retardation. This occurs because the adiabatic condition for conventional bipolaron formation is not satisfied in such media. Indeed, the conventional bipolaron with two polarization "clouds" is formed in such a medium as a result of screening of a bipolaron having one "cloud" (of fast HV polarization) by a comparatively slow LV polarization. Thus, in the discussed medium the adiabatic condition for the conventional bipolaron coincides with the adiabatic condition for the bipolaron formed due to HV polarization only. This condition is much more difficult to satisfy than the adiabatic condition for the SP which, at the same time, is the condition for the BQ formation. Finally, as seen from Fig. 5 the doubled binding energy of the DP exceeds the bipolaron binding energy. However, below we are going to demonstrate that the concentration of polarons is limited due to the limitation on their momenta so that at a carrier concentration of the order of 10^{21} cm^{-3} the band of resonance bipolarons proves to be significantly filled.

VI. THE SYSTEM WITH MANY-COMPONENT POLARIZATION AT HIGH CARRIER CONCENTRATION AND HIGH-TEMPERATURE SUPERCONDUCTIVITY

The above analysis (taking into account the spatial dispersion of lattice polarizability) has significantly changed our knowledge about the carrier spectrum and effects of the carrier motion in ionic crystals with intermediate and strong electron-phonon interaction. It is natural that the demonstrated small width of the bands of

autolocalized states influences the properties of the system with high carrier concentration. In particular, it will be shown below that the limited width of the polaron bands causes a limitation on the polaron concentration in the system.

We should take into account that the carriers must obey Fermi statistics. Since the carriers in the polaron states are localized in coordinate space they can have the same momenta, if their positions are different. In the opposite case the momenta of carriers must be different in accordance with the Pauli principle. Let us consider the process of increasing the carrier concentration when the polaron wave functions become significantly overlapped. Obviously, the new carriers appearing in the system with such a concentration must have average momenta exceeding the uncertainty of the carrier momentum in the polaron. However, this is impossible because the maximum velocity of the polaron is much lower than the uncertainty of the carrier velocity in the polaron states. Thus the maximum concentration of polarons is determined by the degree of degeneracy of the polaron state due to the possibility of the carrier localization in different regions of coordinate space.

This concentration can be obtained as the inverse volume of the polaron. The polaron volume can be estimated using the model of a polaron with constant probability of finding the carrier inside some spheres in coordinate and momentum spaces and zero probability outside them. The radii of these spheres Δr and Δp are determined from the condition of equality of the carrier coordinate and momentum uncertainties in the polaron (we used Pecar's form of the polaron wave function) and in the model. Taking into account that the volume occupied by the carrier in coordinate and momentum spaces is $(2\pi\hbar)^3$ the maximum concentration of polarons can be given by

$$n_{\text{pol}}^0 = 2\frac{4}{3}\pi \frac{(\Delta p_{\text{DP}})^3}{(2\pi\hbar)^3} \quad (29)$$

where 2 is due to spin. For the typical medium parameters the value of n_{pol}^0 is of the order of $5 \times 10^{20} \text{ cm}^{-3}$.

To consider the system at a temperature different from zero it is also necessary to take into account changes in the filling numbers of the polaron states. It will be natural to use for this purpose Fermi's expression in which the filling numbers are determined by the state energy and temperature. But polarons of the same type can have different energy only if their momenta are different. As the momentum of the polaron is proportional to the average momentum of the carrier we must use the latter as characteristic of the state. In addition, we must take into account the possibility of simultaneous existence of some quantity g_{pol} of carriers having the same average momentum but localized in different regions in coordinate space. This statistical weight of the polaron state g_{pol} can be found from the condition of equality of the maximum polaron concentration given by (29) and that obtained by integrating the distribution function over the interval of average carrier momenta corresponding to the polaron states:

$$n_{\text{pol}}^0 = \frac{2g_{\text{pol}}}{(2\pi\hbar)^3} \int_0^{m_e u_2} 4\pi p^2 dp .$$

(The distribution function is supposed to correspond to the complete filling of the polaron band.) Thus the statistical weight of the polaron state can be expressed as

$$g_{\text{pol}} = \frac{(\Delta p_{\text{DP}})^3}{(m_e u_2)^3} . \quad (30)$$

We have shown that the polaron concentration in the system is limited by the value n_{pol}^0 of the order of $5 \times 10^{20} \text{ cm}^{-3}$. As is clear from Fig. 5, when the band of DP's is filled, further increase of the carrier concentration leads to filling the resonance bipolaron band. Thus the extra carriers above the maximum polaron concentration appear in bipolaron states. (It should be noted that at sufficiently high temperature when the filling numbers of the polaron states are different from unity the polaron concentration can even be considerably lower.) The concentration of bipolarons in the system can be calculated from the ordinary Bose distribution function with the only difference that as bipolarons are autolocalized states their impulses are also limited. So the bipolaron concentration can be expressed as

$$n_{\text{bip}} = \frac{g_{\text{bip}}}{2\pi^2\hbar^3} \int_0^{m_{\text{bip}} v_k} \frac{p^2 dp}{\exp[p^2/(2m_{\text{bip}}T) - 2\mu/T] - 1} , \quad (31)$$

where the energy is supposed to be zero at the bottom of the BQ bipolaron band and μ is the chemical potential of the carrier. If we suppose that the bipolarons are resonance bipolarons the statistical weight g_{bip} is equal to 4 (because for the resonance bipolaron the singlet and triplet states have the same energy) and the velocity v_k in the upper limit of the integral is the minimum of two velocities: the minimum phase velocity of phonons of the LV branch u_1 or the difference between the polaron velocity in the BQ v_0 and u_2 : $(u_2 - v_0)$. For the conventional bipolaron v_k is the lower of the two minimum phase velocities of phonons.

Since bipolaron theories of the superconductivity have attracted much attention of late it will be interesting to consider how the spatial dispersion influences the conditions of bipolaron Bose condensation. It is easy to obtain from (31) the bipolaron concentration n_{BC} necessary for their Bose condensation at a certain temperature T_0 presuming in (31) the Bose-condensation condition $\mu=0$. In the ordinary case of $m_{\text{BQ}}v_k^2/T \ll 1$ the exponent is expanded so that the distribution function is easily integrated to give us the resulting expression

$$n_{\text{BC}} = \frac{2}{\pi^2\hbar^3} m_{\text{BQ}}^* v_k T_0 . \quad (32)$$

The contribution of free carriers is negligible because of a large gap. So the carrier concentration necessary for the bipolaron Bose condensation is $n_0 = 2n_{\text{BC}} + n_{\text{pol max}}$.

As seen from (32), n_{BC} depends on the new parameter of the theory v_k which is determined by the phonon dispersion and can be very small. This fact enables us to

predict not high n_0 values (for example, of the order of 10^{21} cm^{-3}) for high condensation temperatures T_0 (100–200 K) even if the bipolaron effective mass is high. In other words, the condensation temperature T_0 found from (32) can be high (for example, 100–200 K) at carrier concentrations typical for the superconducting oxides even at a sufficiently high effective mass of the bipolaron.

Since the question of experimental observation of pairs in high- T_c superconductors at temperatures above the superconducting transition temperature T_0 is still open, it will be useful to study the change in the bipolaron concentration with the temperature at $T > T_0$, taking into account the small width of the bipolaron band. For simplicity let us consider the case when the bottoms of the SP and BQ bands coincide: $2E_{\text{SP}} = E_{\text{BQ}}$. Taking into account the significant width of the SP band in comparison with the BQ one and the large gaps between these two bands and other bands, we can assume that the number of bipolarons will decrease with increase of temperature mainly due to the increase of the SP number. Thus, denoting $\Delta n_{\text{SP}} = n_{\text{SP}}(T) - n_{\text{SP}}(T_0)$ and $\Delta n_{\text{BQ}} = n_{\text{BQ}}(T) - n_{\text{BQ}}(T_0)$ we can write the following approximate equality:

$$\Delta n_{\text{SP}} = -2\Delta n_{\text{BQ}}. \quad (33)$$

The SP concentration in the system has the form

$$n_{\text{SP}}(T) = \frac{2g_{\text{pol}}}{2\pi^2\hbar^3} \int_{m_e u_1}^{m_e u_2} \frac{p^2 dp}{\exp[p^2/(2m_e T) - \mu/T] + 1}. \quad (34)$$

Expressing the $\mu(T)$ function from (33) in which (31) and (34) are taken into account we can substitute it into (31) and easily find the relation

$$\frac{\Delta n_{\text{BQ}}}{n_{\text{BC}}} = \frac{g_{\text{pol}}}{2g_{\text{bip}}} \frac{m_e^4 u_2^5}{80m_{\text{BQ}}^* v_k T_0} \frac{T - T_0}{T}. \quad (35)$$

Depending on many values (g_{pol}, u_2, v_k) which are determined by the medium parameters the value of this relation can vary in a very wide range. In particular, it can be of order of 10. In such a case bipolarons cannot be observed already at the temperatures $T = 1.1T_0$.

VII. CONCLUSION

In conclusion it seems to be useful to discuss in what crystals and experiments the predicted effects can be observed. We pointed out that for the effects of the autolocalization band structure to take place the medium must possess two or more branches of polar phonons of different dispersion. Such a medium turns out to correspond well to complex oxide crystals which are one of the broadest classes of ionic crystals. They have many ions in the unit cell so that there are many branches in their phonon spectrum. To observe the effects of carrier braking by the coherent radiation and polarons pairing under resonance conditions with the medium vibrations, one of these branches must have sufficiently strong dispersion. Although there are very few data of phonon dispersion measurements in complex oxides high dispersion of some branches in complex oxides was nevertheless reported.¹⁵

There are many papers on the computation of phonon dispersion in these crystals. They also report high dispersion for several phonon branches at least in a region of small wave vectors¹⁶ $k < \pi/(2a_0)$ where a_0 is the lattice constant. (This region is essential in our consideration since the polaron radius is 2–3 times larger than a_0 .) The other source of high dispersion in the region of small wave vectors can be the vicinity of a phase transition with softening of one phonon mode, which frequently takes place in complex oxides. Thus such crystals can be expected to exhibit the effects predicted above.

In particular, the effect of ‘‘Cherenkov’’ radiation of a coherent polarization wave by the polarons can reveal itself in measurements of the carrier loss as a function of its velocity and carrier mobility (or equilibrium velocity) versus the applied electric field. In such experiments the carrier concentration is usually low so that polarons obey Boltzmann statistics.⁹ Therefore, to observe these effects we need the temperature $T < m_{\text{SP}} u_2^2/2$, i.e., $T < 10$ K. Moreover, at the temperature $T < m_{\text{DP}} u_1^2/2$ which is less than 1 K the effect of the loss of the LV polarization ‘‘cloud’’ by the DP can be observed in the carrier mobility. The carrier transition from the DP band to the SP one can manifest itself in the temperature dependence of infrared absorption at high carrier concentration. Photoinduced ir absorption measurements enable us not only to demonstrate the presence of autolocalized carriers at low temperature but also to determine their binding energy and effective mass.³ The autolocalization band structure can be observed also in the normal-to-normal-state tunneling spectra of such crystals. But it is difficult to work out predictions for the results of such experiments because a large number of additional circumstances must be taken into account.

As there is the possibility of high- T_c superconductivity by means of resonance bipolarons it is important to compare their properties with experimental data on the pairs in high- T_c superconductors. The size of the resonance bipolaron is determined by the orbit radius in the orbit plane and by the SP radius in the perpendicular direction. These values and anisotropy of the BQ size correspond well to the coherence lengths measured in superconducting complex oxides.¹⁵ The isotopic effect in our model must be, of course, different from the BCS one since here the Bose-condensation condition depends on two phonon frequencies and dispersions.

We have shown that the condition of the bipolaron Bose condensation is strongly determined by phonon dispersion. From this viewpoint it would be useful to check the correlation between the dispersion of relevant phonons, carrier concentration, and the temperature of the superconducting transition in complex oxides and compare it with their relation given by (32). The concentration of carriers necessary for the Bose condensation of the resonance bipolarons coincides in order of magnitude with carrier concentrations in superconducting complex oxides. In addition, we have demonstrated that in the considered model the decrease of the bipolaron concentration above the superconducting transition temperature can be so quick that it may be difficult to observe the pairs as in the superconducting complex oxides.

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