Electron localization with and without barrier formation

V. V. Kabanov
Laboratory of Neutron Physics, Joint Institute for Nuclear Researches, Head P.O. Box 79, Moscow 101000, Russia

O. Yu. Mashtakov
Institute for Power Engineering, Academy of Sciences of Moldova Republic, Grosul Strasse 5, Kishinev 277028, Moldova
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The possibility of polaron formation within a model of local electron-phonon interactions is considered. It is shown that, unlike the continuous limit, the solution to the Schrödinger equation on a two-dimensional lattice in the adiabatic approximation provides evidence for the presence of an energy barrier that separates a small-size polaron and delocalized electronic state. The polaron stability and metastability regions are determined. The possibility of polaron formation in high-T_c superconductors is discussed.

I. INTRODUCTION

The discovery of high-T_c superconductivity stimulated a large number of theoretical works on this problem. Antiferromagnetic order observed in CuO_2 planes at small carrier concentration indicates that the magnetic subsystem is of much importance to the formation of the superconducting state.[1] Studies of structural phase transitions as well as a number of other experiments[2] provide evidence for the important role of the electron-phonon interaction. The reports concerning the direct observation of polarons in a dielectric phase upon carrier doping[3] and small but nonzero isotope effect[4] lead to the same conclusion. Also it is worth mentioning the recent series of works.[5,6] It has been shown in Ref. 6 that the magnetic polaron in CuO_2 planes has a lower energy of the ground state than any other known magnetic phases. Note that the formation of the magnetic polaron must be accompanied by the usual polaronic effect.[5] One of the indications of the possibility that polarons are formed in high-T_c superconductors at low carrier concentration is the observation of midinfrared peaks in the experiments on the measurement of optical conductivity $\sigma(\omega)$.[7-9] These measurements show two well-pronounced peaks in $\sigma(\omega)$ at $\omega=0.08$ eV and $\omega=0.8$ eV.[7] A qualitative interpretation of these features has been given in Ref. 10 in terms of small-size polaron theory.

The possibility of a small-size polaron (SSP) formation the two-dimensional (2D) case within the model of a local strain interaction between an electron and dispersionless phonons (the Holstein model[11]) is analyzed in this paper. It is shown that, unlike the continuous limit[12-14] taking no account of a finite width of the nonrenormalized electronic band, the formation of a self-trapped state in a 2D lattice is accompanied by the formation of an energy barrier. The effect of the electronic band spectrum on the formation of a self-trapped state is analyzed. Nonadiabatic corrections to the energy of the SSP are calculated in the 2D case.

II. VACUUM EQUATIONS

In order to treat the problem of SSP formation, we consider the following model: An electron interacts locally with a single dispersionless phonon mode Ω_0 (the Holstein model[11]). The Lagrangian of such a system reads:

$$\mathcal{L}=\sum_{\mathbf{i}}\left\{\frac{1}{2\Omega_0}(\partial_\tau\phi_{\mathbf{i}})^2-\frac{\Omega_0}{2}\phi_{\mathbf{i}}^2+i\bar{\psi}_{\mathbf{i}}\partial_\tau\psi_{\mathbf{i}}-\sqrt{2}\Omega_0 g\,\bar{\psi}_{\mathbf{i}}\psi_{\mathbf{i}}\phi_{\mathbf{i}}\right\} -t\sum_{\substack{\langle\mathbf{ij}\rangle\\ \mathbf{i}\neq\mathbf{j}}}\bar{\psi}_{\mathbf{i}}\psi_{\mathbf{j}}\,, \tag{1}$$

where $\bar{\psi}_{\mathbf{i}}(\psi_{\mathbf{i}})$ is the fermionic field describing creation (annihilation) of an electron at a site $\mathbf{i}$, $\phi_{\mathbf{i}}$ is the scalar field corresponding to a local displacement at the site $\mathbf{i}-\phi_{\mathbf{i}}=1/\sqrt{2}(b_{\mathbf{i}}^{\dagger}+b_{\mathbf{i}})$, with $b_{\mathbf{i}}^{\dagger}(b_{\mathbf{i}})$ being local phonon creation (annihilation) operator, t is the amplitude for transferring, the electron from the site $\mathbf{i}$ to the *nearest*-neighbor site $\mathbf{j}$, and g is the dimensionless electron-phonon coupling.

Since the Lagrangian (1) is quadratic in fermionic fields, we can explicitly perform the functional integration over these fields. Formally the problem is reduced to calculation of the fermionic determinant in an arbitrary field ϕ. As a result of this procedure, we obtain effective bosonic action, and its minimum in ϕ is a phonon vacuum. Note that the effective action is not merely a classic quantity, since it involves quantum fluctuations arising from the fermionic determinant. Below we apply the method widely used in the theory of nonzero least action. The main idea of this approximation is to substitute Schrödinger wave functions for the fermionic fields $\psi_{\mathbf{i}}$,[15]

$$\exp(iS_{\text{eff}})\approx\sum_{n_r}\tilde{C}(\{n_r\})\exp\left[iT\left[E_{\text{cl}}(\phi)+\sum_r n_r\varepsilon_r\right]\right]\,, \tag{2}$$

where $\tilde{C}(\{n_r\})$ is a combinatorial factor, $E_{\text{cl}}(\phi)$ is the classical energy of the field $\phi_{\mathbf{i}}$, n_r is the occupation num-

ber, and ε_r is the eigenvalue of the Schrödinger equation in the field ϕ.

Applying variational procedure in ϕ to (2) one can obtain the following set of equations:

$$\phi_i = -\sqrt{2}g \sum_r |\psi_i^{(r)}|^2 n_r \ , \tag{3}$$

$$t \sum_j \psi_{i+j}^{(r)} + \sqrt{2} g \Omega_0 \psi_i^{(r)} \phi_i = \varepsilon_r \psi_i^{(r)} \ . \tag{4}$$

Equations (3) and (4) give the polaron energy and are written within the approximation $\partial\phi/\partial\tau = 0$ (adiabatic approximation[12–14]). Note that the similar equations have been derived in Ref. 6 for a magnetic polaron in an antiferromagnetic background. Equation (4) contains no spatial derivatives, since the "bare" phonon subsystem is supposed to be dispersionless.

Assuming that a single fermionic mode is excited, the solution to Eqs. (3) and (4) can be written as a power series in $t/g^2\Omega_0 (2g^2\Omega_0 \gg t)$:[14]

$$\psi_0(0) \approx 1 - \frac{zt^2}{8(g^2\Omega_0)^2}, \quad \psi_0(1) \approx -\frac{t}{2g^2\Omega_0} \ , \tag{5}$$

$$\varepsilon_0 \approx -2g^2\Omega_0 \ . \tag{6}$$

The total energy of the system is a sum of the electronic energy ε_0 in the strain field ϕ and the energy of the field ϕ itself:

$$E_{\text{tot}} = \varepsilon_0 + \sum_i \frac{\Omega_0}{2} \phi_i^2 = \varepsilon_0 + g^2\Omega_0 \sum_i |\psi_i|^4$$

$$\approx -g^2\Omega_0 - \frac{zt^2}{2g^2\Omega_0} \ . \tag{7}$$

Note that the correction to E_{tot} is of the order of t^2 and consistent with the result obtained in Ref. 16 through the summation of a certain set of diagrams with noncrossing phonon lines. Equations (3) and (4) have the extended solution

$$\psi_l = \frac{1}{\sqrt{N}} \exp[i(l_x + l_y)\pi] \ ,$$

which reaches the minimum at the point $\mathbf{k} = (\pi, \pi)$ of the Brillouin zone and corresponds to the absence of the self-trapped state.

In the 1D case as $g^2\Omega_0 \gg t$, the solution to Eqs. (3) and (4) is given by expressions (5) and (6) with $z = 2$. In the opposite limiting case $g^2\Omega_0 \ll t$, one can obtain the following exact solution:[17]

$$\psi(x) = \left[\frac{|\varepsilon + 2t|}{g^2\Omega_0} \right]^{1/2} ch^{-1} \left[\left[\frac{|\varepsilon + 2t|}{ta^2} \right]^{1/2} x \right] \ ,$$

$$\varepsilon = -2t - \frac{g^4\Omega_0^2}{4ta^2} \ . \tag{8}$$

The results of numerical analysis of Eqs. (3) and (4) in the 1D case are represented in Fig. 1. As can be seen from Fig. 1, the self-trapped state exists in all g region of interest. If $g^2\Omega_0 < t$, the radius of this self-trapped state is restricted to a lattice constant. Further decrease of

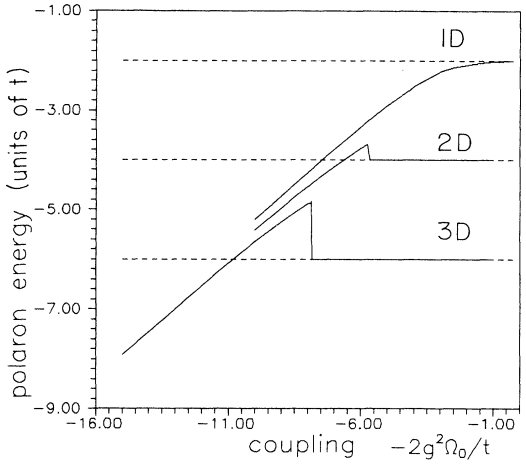

FIG. 1. The total energy of the system as a function of electron-phonon coupling g for different dimensionalities (all quantities are expressed in dimensionless units).

electron-phonon coupling g leads to the increase in the radius. Thus in the 1D case a smooth transition from a SSP with the energy $E_p \sim g^2$ to a large-size polaron with the energy $E_p \sim g^4$ takes place and self-localization occurs without barrier formation. This result is consistent with a scaling approach.[12,13] Since in the 1D case the kinetic energy is proportional to R^{-2} and a gain in the strain energy R^{-1}, the total energy always has a minimum corresponding to the self-trapped state, and the delocalized state is always unstable.

Since one encounters considerable difficulties in solving Eqs. (3) and (4) in 2D and 3D cases analytically, we present the results of numerical analysis of these equations (see Fig. 1). In the 2D case the results differs from the 1D case essentially. There are three critical values of electron-phonon coupling—$g_{c1}^2 = 2.85t/\Omega_0$, $g_{c2}^2/\text{p} = 3.5t/\Omega_0$, and $g_{c3}^2 = 2\pi t/\Omega_0$. The first one sets limits to the region where only the delocalized state exists ($g < g_{c1}$). The SSP is metastable within the range $g_{c1} \le g < g_{c2}$. As $g > g_{c2}$, the SSP becomes the ground state of the system. The total energy of the self-trapped state as a function of its radius at various values of the coupling constant is represented in Fig. 2. This dependence has been calculated within the variational approach by using the simple trial function $\psi(\mathbf{i}) \propto \exp(-\mathbf{i}^2/R^2)$, where $\mathbf{i}^2 = i_x^2 + i_y^2$. It is clear from Fig. 2 that as $g_{c1} < g < g_{c2}$, the SSP is the metastable state separated from the delocalized ground state by an energy barrier. In addition, there exists the third critical value $g_{c3}^2 = 2\pi t/\Omega_0$. When coupling g exceeds this value, the delocalized state becomes absolutely unstable. Within the range $g_{c2} < g < g_{c3}$, the delocalized state is metastable. Thus the formation of the self-trapped state in the 2D case is accompanied by the formation of an energy barrier that separates the self-trapped and delocalized states. Note that the formation of the barrier is connected with a finite width of the nonrenormalized electronic band. Numerical study of Eqs. (3) and (4) in an effective-mass approximation indicates that self-localization occurs

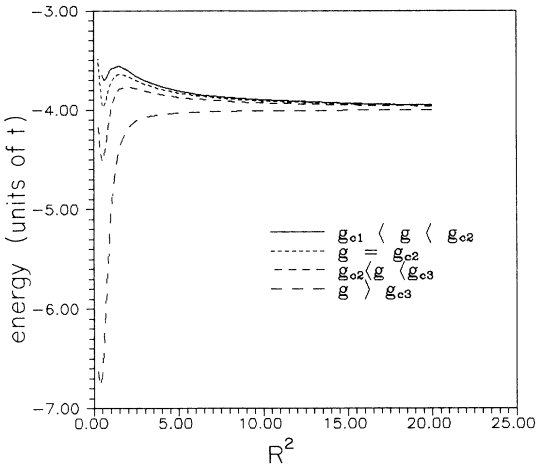

FIG. 2. The total energy of the 2D system as a function of the effective dimensionless radius of the state at various values of coupling g.

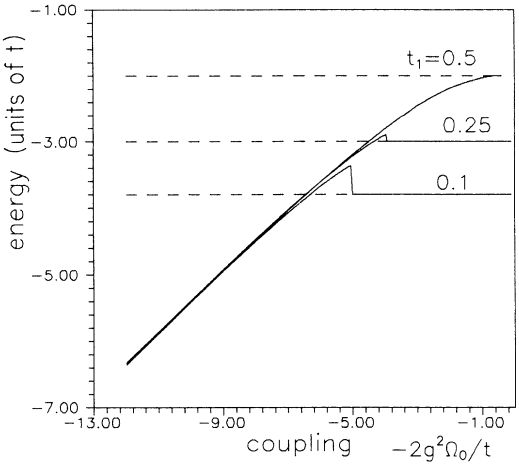

FIG. 3. The total energy of the 2D system as a function of electron-phonon coupling g at various values of the ratio t/t_1, where t and t_1 are the hopping amplitudes to the nearest- and next-nearest lattice sites, respectively.

without the barrier formation. This is closely linked to the fact that on a decrease in a polaron size, the kinetic energy and the strain energy are proportional to $1/R^2$, where R is the radius of the self-trapped state. However, as pointed out in,[14] the criterion of the polaron formation corresponds to existence of the self-trapped state with the radius comparable with interatomic space, and therefore in this case a finite bandwidth (lattice discreteness) plays a major role. This fact means that the kinetic-energy scaling is broken.

In order to clear up the role of the band spectrum in the formation of the self-trapped state, we introduced the additional hopping from a site to the next-nearest neighbor sites with the amplitude t_1. Hence the modified band spectrum has the following form:

$$\varepsilon(\mathbf{k})=2t[\cos(k_x)+\cos(k_y)]+4t_1\cos(k_x)\cos(k_y) .$$

Then the dependence of the strain energy on the radius R remains unchanged. Yet the kinetic energy as a function of the radius R depends on the ratio t_1/t. This dependence results in the obvious fact that the barrier which separates the self-trapped and delocalized states must be a function of t and t_1. As can be seen from Fig. 3, upon increasing the ratio t_1/t from 0 up to 0.5, the energy barrier continuously decreases and finally disappears. This behavior is accounted for by the fact that the presence of the additional term proportional to t_1 leads to a competition between the terms proportional to k^2 and k^4 in the expansion of the kinetic energy near the band bottom. At $t_1=0.5t$, the quadratic terms in the dispersion relation cancel, so that the kinetic energy is proportional to k^4. Therefore, as $R\to\infty$ the behavior of the system is determined by a gain in the strain energy ($\Delta E\propto R^{-2}$). As a consequence, the total energy as a function of R has a minimum corresponding to the formation of the polaronic state. This state is always stable. Numerical study of Eqs. (3) and (4) confirms this result (see Fig. 3).

Note that in the 3D case, lattice discreteness has no qualitative effect on the results obtained in the effective-mass approximation. As $R\to\infty$, the behavior of the total energy $E(R)$ in the 3D case is determined by the kinetic energy proportional to R^{-2}. Thus, the self-trapped state is metastable when $\tilde{g}_{c1}<g<\tilde{g}_{c2}$ and separated by an energy barrier from the delocalized state. As $\tilde{g}_{c2}<g$, the delocalized state becomes metastable. But in contrast to the 2D case the delocalized state is always stable [$\partial E(R)/\partial R<0$ as $R\to\infty$].

Note that the results obtained in an adiabatic approximation and represented in Fig. 1 are in good quantitative agreement with Monte Carlo calculations.[18] The regions of electron-phonon coupling, where the self-trapped state is metastable can be clearly seen on the plots given in that paper.

III. NONADIABATIC CORRECTIONS

As pointed out above, Eqs. (3) and (4) and their solutions (5) and (6) correspond to an adiabatic approximation in polaron theory. The calculation of fluctuation effects in the vicinity of a classic phonon vacuum results in the nonadiabatic corrections to the energy of the ground state of a polaron. As a result, phonon frequencies renormalized due to electron-phonon interaction will be present in the new Lagrangian. In order to calculate the renormalized frequencies, we shall expand the effective action $S_{\text{eff}}(\phi)$ in fluctuations:

$$\delta\phi=\phi(\mathbf{i},\tau)-\phi_0(\mathbf{i}) . \tag{9}$$

Keeping in mind that the minimum of the action $S_{\text{eff}}(\phi)$ is realized on the phonon vacuum ϕ_0 obtained in Sec. II, we calculate the corrections to S_{eff} corresponding to the one-loop approximation:

$$\delta S=ig^2\Omega_0^2\sum_{\substack{\mathbf{ij}\\ \omega\Omega}}\delta\phi_{\mathbf{i}}(\Omega)\delta\phi_{\mathbf{j}}(-\Omega)G_{\mathbf{ij}}(\omega)G_{\mathbf{ji}}(\omega+\Omega) , \tag{10}$$

where

$$G_{ij}(\omega)=\frac{\psi_0(i)\psi_0^*(j)}{\omega-\varepsilon_0+\mu}+\sum_k \frac{\psi_k(i)\psi_k^*(j)}{\omega-\varepsilon_k+\mu}$$

is the Green's function describing the motion of an electron in the vacuum field $\phi_0(i)$, $\psi_0(i)$ is the wave function corresponding to the ground-state energy ε_0, $\varepsilon_k=2t\sum_{i=1}^{d}\cos(k_i a)$, and $\psi_k(i)$ is the wave function of the electron corresponding to the energy ε_k. The direct calculation of (10) gives the expression for δS to lowest order in $t/g^2\Omega_0$:

$$\delta S=\frac{\Omega_0 z t^2}{8(g^2\Omega_0)^2}|\delta\phi_0(\Omega)|^2 , \tag{11}$$

where the subscript 0 refers to the center of the polaron. The additional contribution from (10) gives rise to the occurrence of a renormalized phonon mode (see Refs. 14 and 22):

$$\Omega=\Omega_0\left[1-\frac{zt^2}{2(g^2\Omega_0)^2}\right]^{1/2} . \tag{12}$$

Also note that expression (12) differs from the similar one obtained in Ref. 22, which is likely to be valid as $\Omega_0 \gg t$ (antiadiabatic approximation).

The evaluation of the functional integral over $\delta\phi_i$ yields the nonadiabatic correction to the ground-state energy of the SSP:

$$\Delta E=-\frac{zt^2\Omega_0}{8(8g^2\Omega_0)^2} \quad \text{as } g \gg g_{c2} . \tag{13}$$

In the region of coupling g where the delocalized solution is the most favorable [$\phi_0(i)=0$], the corresponding nonadiabatic correction is well known:[19]

$$\Delta E=-\frac{g^2\Omega_0^2}{\pi^2 t^2} . \tag{14}$$

IV. CONCLUSIONS

It has been shown in this paper that the formation of a polaron in the 2D case within the model of local electron-phonon interaction is accompanied by the formation of an energy barrier attributed to a finite electronic bandwidth (lattice discreteness). Yet lattice discreteness has no influence on a qualitative picture of the self-trapped state formation in 1D and 3D cases.

The criterion of SSP formation has been formulated in the paper. In 2D and 3D cases it has the following form under notations of:[22]

$$\lambda=\frac{g^2\Omega_0}{D}>\lambda_c , \tag{15}$$

where D is the bandwidth ($D=zt$), $\lambda_c\approx 1$.

Note that within the model involved, discontinuous phase transition to the delocalized state occurs at $\lambda=\lambda_c$. The criterion (15) differs from the similar one with $\lambda_c^*\approx 1/\sqrt{2z}$ obtained in Refs. 16 and 19. The present criterion determines a small magnitude of corrections to polaronic shift. The corrections are connected with the hopping matrix element taken into consideration. However, the phase transition to the delocalized state occurs before these corrections become large. The self-trapped state, i.e., the bound state of an electron in a background of static ionic displacements, is formed at $\lambda=\lambda_c^*$, but it is metastable.

The 1D case is essentially different from 2D and 3D ones. The self-trapped state is the ground state of the system at any value of electron-phonon coupling, at $\lambda>\lambda_c^*$, the radius of this state being bounded by the value of a lattice constant (SSP). The radius grows rapidly as $\lambda<\lambda_c^*$ [$r\sim a/\lambda$ as $\lambda\ll 1$, see Eqs. (8)]. Thus, unlike 2D and 3D cases, in the 1D case phase transition from the polaron state to the delocalized electronic state is absent.

It is worthwhile to note that in the 2D case the criterion (15) can be essentially different. If the quadratic terms in the dispersion relation expanded in the vicinity of the band bottom are absent, the self-trapped state is formed at any value of coupling. In this case the radius of the state increases as $\lambda>1$.

Fluctuations around the stationary solutions of Eqs. (3) and (4) give rise to nonadiabatic corrections to the polaronic energy (13). It is worthwhile to point out that a region of strong fluctuations exists near λ_c due to the fact that the effective action has two minima with almost equal energy values. This conclusion puts a limit to the application of the expression (13), i.e., it is valid as $\lambda\gg 1$.

The criterion (15) of SSP formation is rather rigorous. The estimate of g^2 at $D\sim 0.5-1$ eV and the most favorable value of $\Omega_0\sim 0.1$ eV is equal to 5–10. This value results in the picture with the strong renormalization of effective mass of polaron and means that it is practically localized. However, the correlation effects play a major role in high-T_c superconductors and lead to the effective narrowing of electronic bandwidth[16] and even to the formation of magnetic polarons.[5,6,21] These effects may change the criterion (15) essentially. In addition, the formation of the magnetic polarons must result in the enhancement of the usual polaronic effect.[5,23] In this connection it is worth mentioning that an isotope effect has been observed in all high-T_c superconductors.[4] This fact indicates that the phonon subsystems is of great importance to the transition to superconducting state.

It is also interesting to note that medium-size polarons are supposed to be formed in high-T_c superconductors.[24,25] In that case an increase of the radius of such a polaron results in the decrease in its effective mass. Within the Holstein model considered in this paper, the formation of a medium-size polaron is possible if the quadratic terms in the expansion of the dispersion relation near the band bottom are absent. Note that in high-T_c superconductors the band spectrum renormalized by spin fluctuations is highly anisotropic.[20,26,27] Therefore,

the coefficients at the quadratic term are small, so that the criterion (15) is essentially reduced. Upon decreasing the coupling g, this fact results in the decrease in the carrier's effective mass and formation of a medium-size polaron.

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