Pairing theory of polarons in real and momentum space

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A consistent pairing theory of carriers in real (r) and k space is developed. The pairing of different free (F), delocalized (D), and localized (L) carriers in r space leading to the formation of various bipolaronic states is considered within the continuum model and adiabatic approximation, taking into account the combined effect of the short- and long-range components of the electron-lattice interaction with and without electron correlation. We calculate the formation possibility of D and L bipolarons and determine their concrete formation criteria as a function of $\varepsilon_\infty/\varepsilon_0$. The pairing scenarios of carriers in k space leading to the formation of different bipolarons (including also Cooper pairs or dynamic bipolarons) are considered within a generalized BCS-like model, taking into account the combined phonon- and polaron-bag-mediated processes. It is shown that the pure BCS pairing picture is the particular case of a more general BCS-like one, and Bose-Einstein condensation of r-space or ideal-Bose-gas bipolarons is irrelevant to the superconductivity phenomenon. The possible relevance of the results obtained to the high-T_c superconductors is discussed in detail in the framework of a two-stage Fermi-Bose-liquid scenario of superconductivity that is caused by single-particle and pair condensation of attracting bipolarons. [S0163-1829(96)00533-4]

I. INTRODUCTION

As is generally known, the pairing of carriers in superconductors plays an important role in the formation of the superconducting (SC) state. In connection with the discovery of the high-temperature superconductors (HTSC's) various pairing mechanisms under different names (such as phonons, excitons, plasmons, polarons, local pairs, peroxitons, electron-charge and spin bags, solitons, magnons, etc.) have been discussed.[1–8] In many versions of these pairing theories, two limiting cases are considered, namely, the formation of a BCS-like k-space pairing state[1–7] and of a spatially separated bipolaronic real- $(r$-) space pairing one.[1,2,6,7] In both limiting cases the appearance of the SC state is assumed possible. However, a very controversial question in the theory of superconductivity is the relevance to the superconductivity of the pairing of carriers in r space. According to the Landau criterion, an ideal Bose gas (BG) is not superfluid,[8] while the BCS-like k-space pairing may have some relation to the superconductivity. Such pairing of carriers at least is necessary for studying their collective properties, which are very important in the establishment of the following key scenarios of SC phase transitions.[9–11] Therefore, studying the relationship of the above two versions of pairing to each other is of particular interest. The present paper is devoted to discussion of this question. First we consider the possibility of formation of different types of bipolarons in the r-space pairing approximations. The ground state of electrons and holes in solids is the self-trapped (i.e., polaronic and bipolaronic) state. This is discovered in many classes of substances, such as alkali halides, rare-gas solids, oxides, organic molecular crystals, and semiconductors.[7,12–15] In other words, due to strong electron-lattice interaction, the free (F) or itinerant state of carriers becomes unstable and leads to the formation of the r-space large [delocalized (D)] polarons or even the small localized (L) polarons. Further, we consider the pairing of polarons within the k-space approximation.

II. THE PAIRING OF POLARONS IN *r* SPACE

The question of the possibility of formation of large bipolarons (in our context D) was first considered by Pekar.[14] Pekar used a variational method without taking into account the short-range component of the electron-lattice interaction and electron correlation within the continuum model and adiabatic approximation. It was shown that in such approximations the formation of D bipolarons is impossible. Later, a similar approach was used by Vinetskii[15] to consider the formation possibility of two-center D bipolarons. It was found that the formation of such D bipolarons becomes energetically favorable under the condition $x=\varepsilon_\infty/\varepsilon_0 \leqslant 0.05$ (where ε_0 and ε_∞ are the static and high-frequency dielectric constants) and their binding energy $E_{bB}\sim 3$–4% of twice the polaron energy $(2E_P)$. In Ref. 15 (see also Ref. 16) the important role of the short-range electron-lattice interaction in the formation of one-center D bipolarons in the absence of electron correlation was pointed out also. In Ref. 17, in contrast to the above, the possibility of formation of Pekar's bipolarons (or one-center D bipolarons) at $x<0.018$ with binding energy $E_{bB}\simeq 0.04\times 2E_P$ (at $x\to 0$) is assumed. However, the validity of such an assertion is doubtful. Further, according to

Ref. 18 the formation of two-center D bipolarons considered in Ref. 15 is also unlikely. In Ref. 19 (see also Ref. 20) the question of Pekar's bipolaron is considered within a variational method taking into account the electron correlation. There the formation possibility of such D bipolarons in a wide region of values $x \leqslant 0.125$ is shown, with the binding energy reaching up to 22% (at $x \to 0$) of twice the polaron energy.[20] Further, this problem is investigated in Ref. 18, using the path integral method, by Adamowski, who obtained almost the same results. In Ref. 21, using this method and taking into account both the long- and the short-range components of the electron-lattice interaction as well as the electron correlation, the formation possibility of D and L bipolarons is studied. The existence region of the D bipolarons was found to be comparatively narrow (i.e., $0 \leqslant x < 0.079$) and their binding energy was not evaluated. In Ref. 22 a softer criterion for Fröhlich D bipolaron formation at $x < 7/15$ was obtained. However, this is incorrect.[23] The formation of the L bipolarons in the absence of long-range electron-lattice interaction and with incomplete inclusion of electron correlation was studied in Ref. 25. Thus the formation possibility of D and L bipolarons both with and without inclusion of electron correlation and short-range electron-lattice interaction has not been investigated sufficiently yet.

A. The pairing of polarons without electron correlation

The total energy of the two-electron+crystal lattice system in the continuum model and adiabatic approximation is given by the functional form

$$E_B[\Psi,\Delta,\Phi] = \int\int dr_1 dr_2 \Psi(r_1,r_2)\left[-\frac{\hbar^2}{2m_1}\nabla_1^2 - \frac{\hbar^2}{2m_2}\nabla_2^2 + \frac{e^2}{\varepsilon_\infty |r_1 - r_2|}\right]\Psi(r_1,r_2) + \int dr_1 dr_2 [E_d\Delta(r_1) + E_d\Delta(r_2)]\Psi^2(r_1,r_2) + \int dr_1 dr_2[-e\Phi(r_1) - e\Phi(r_2)]\Psi^2(r_1,r_2) + \frac{K}{2}\int dr \Delta(r)^2 + \frac{\tilde{\varepsilon}}{8\pi}\int dr [\Delta\Phi(r)]^2, \tag{2.1}$$

where $\Psi(r_1,r_2) = \Psi(r_1)\Psi(r_2)$ is the electron wave function at the coordinates r_1 and r_2, $\Delta(r)$ is the deformation of the lattice, $\Phi(r)$ is the electrostatic potential due to the ionic displacement polarization, E_d is the deformation potential of the electron, K is an elastic constant, and $\tilde{\varepsilon} = \varepsilon_\infty/(1-x)$ is the effective dielectric constant. The functional (2.1) after minimizing first with respect to Δ and Φ and then with respect to a trial wave function chosen in the form[26] $\Psi(r) = (\alpha\sqrt{2}/a_0)^{1.5}\exp[-\pi(\alpha r/a_0)^2]$ (where α is the dimensionless variational parameter characterizing the degree of localization of the carrier and a_0 is the lattice constant) has the form

$$E_B(\alpha) = 2B(\alpha^2 - g_{s2}\alpha^3 - g_{l2}\alpha), \tag{2.2}$$

where $B = 3\pi\hbar^2/2m^* a_0^2$, $g_{s2} = 2g_{s1}$, $g_{s1} = E_d^2/2Ka_0^3 B$, $g_{l2} = g_{l1}(1-2x)/(1-x)$, and $g_{l1} = e^2/\tilde{\epsilon}a_0 B$. The functional (2.2) depending on the quantities g_{s2}, g_{l2}, and x may have one minimum at $\alpha = \alpha_D < 1$ or two at $\alpha = \alpha_D < 1$ and $\alpha = \alpha_L = 1$, which are separated by the energy barrier

$$E_{aB} = \frac{8B}{27g_{s2}^2}\left[1 - \frac{6y(1-2x)}{(1-x)}\right]^{3/2}. \tag{2.3}$$

The energy of these D and L states of bipolarons is determined from

$$E_B(\alpha_D) = \frac{2B}{27g_{s2}^2}\left[2 - 18y\frac{(1-2x)}{1-x} - 2\left(1 - 6y\frac{(1-2x)}{(1-x)}\right)^{3/2}\right], \tag{2.4}$$

and

$$E_B(\alpha_L = 1) = 2B(1 - g_{s2} - g_{l2}), \tag{2.5}$$

respectively, where $y = g_{s1}g_{l1}$. In Eq. (2.2), if we substitute $g_{s1}/2$, $g_{l0}(1-x)$, and B for g_{s2}, $g_{l0}(1-2x)$, and $2B$, respectively, then we obtain the functional of the polaron total energy $E_p(\alpha)$. The binding energy of the bipolaron is determined by the difference

$$E_{bB} = |E_B - 2E_p| \tag{2.6}$$

where E_p and E_B are the polaron and bipolaron energy, respectively. Then, for the formation criterion of the stable D and metastable L or D (L) states of bipolarons from the inequality $E_B(\alpha_D) - 2E_p(\alpha_D) < 0$ we have (cf. Refs. 15 and 16)

$$9y\frac{1}{1-x} - \left[1 - 6y\frac{1-2x}{1-x}\right]^{3/2} + 4(1-3y)^{3/2} < 3. \tag{2.7}$$

The formation condition of the L bipolaron only has the form

$$x < \left[1 + \frac{y}{g_{s1}^2}\right]^{-1} \tag{2.8}$$

with the additional condition for $x < 0.5$

$$y > \frac{(1-x)}{6(1-2x)}, \tag{2.9}$$

while the formation of the stable L and metastable D or L (D) states of bipolarons is possible at

$$y > g_{l0}x(1-x), \tag{2.10}$$

where $g_{l0} = e^2/\varepsilon_\infty a_0 B$. The dependence of the ratio of D bipolaron binding energy $E_{bB}(D)$ twice the D polaron energy $2E_F(D)$ on x for different values of y is presented in Fig. 1. The phase diagram for stable bipolaronic states in two-dimensional coordinate space (x,y) is shown in Fig. 2.

B. The pairing of polarons with electron correlation

For the calculations of the ground-state energy of a polaron and bipolaron by using the variational method in the

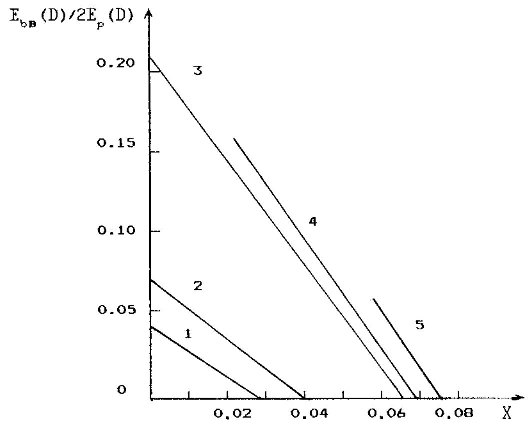

FIG. 1. The dependence of the ratio of the binding energy of the D bipolaron to twice that of the D polaron on x at different y: (1) $y=0.05$, (2) $y=0.1$, (3) $y=1/6$, (4) $y=0.17$, (5) $y=0.178$.

continuum model and adiabatic approximation, the functional of their total energies after minimizing with respect to lattice deformation and polarization can be written in the form

$$E_p\{\psi(r)\}=\frac{\hbar^2}{2m^*}\int[\nabla\psi(r)]^2dr-\frac{e^2}{2\tilde{\varepsilon}}\int\frac{\psi^2(r)\psi^2(r')}{|r-r'|}\,dr\,dr'-\frac{E_d^2}{2K}\int\psi^4(r)dr, \tag{2.11}$$

$$E_B\{\Psi(r_1,r_2)\}=\frac{\hbar^2}{2m^*}\int[|\nabla_1\Psi(r_1,r_2)|^2+|\nabla_2\Psi(r_1,r_2)|^2]dr_1dr_2+\frac{e^2}{\varepsilon_\infty}\int\frac{\Psi^2(r_1,r_2)}{|r_1-r_2|}\,dr_1dr_2-\frac{2e^2}{\tilde{\varepsilon}}\times\int\frac{\Psi^2(r_1,r_2)\Psi^2(r_3,r_4)}{|r_1-r_3|}\,dr_1dr_2dr_3dr_4-\frac{2E_d^2}{K}\int\Psi^2(r_1,r_2)\Psi^2(r_2,r_3)dr_1dr_2dr_3, \tag{2.12}$$

where $\psi(r)$ and $\Psi(_1,r_2)$ are one- and two-electron wave functions, respectively. In order to minimize Eqs. (2.11) and (2.12) with respect to $\psi(r)$ and $\Psi(r_1,r_2)$ the trial functions are chosen in the form[23]

$$\psi(r)=N\exp[-(\sigma r)^2], \tag{2.13}$$

$$\Psi(r_1,r_2)=N\{1+\beta(\sigma r_{12})^2\,\exp[-\sigma^2(r_1^2+r_2^2)]\}, \tag{2.14}$$

where σ and β are the variational parameters characterizing the carrier localization and the correlation between them, respectively, N is the normalization factor, and r_{12} is the distance between the carriers. Substitution of Eqs. (2.13) and (2.14) into Eqs. (2.11) and (2.12) and then calculation of the integrals and introduction of the dimensionless parameters

$$\gamma=\hbar\omega/[(\pi\hbar)^2/2m^*a_0^2],\quad g_l=2e^2/\hbar\omega_0\tilde{\varepsilon}a_0,$$
$$g_s=E_d^2/\hbar\omega_0Ka_0^3,\quad U=2e^2/\hbar\omega_0\varepsilon_\infty a_0 \tag{2.15}$$

give (in units of the Debye energy $\hbar\omega_0$)

$$E_p(\alpha)=\frac{3}{\pi^2\gamma}\,\alpha^2-\frac{g_s}{2\pi^{3/2}}\,\alpha^3-\frac{g_l}{2\pi^{1/2}}\,\alpha, \tag{2.16}$$

$$E_B(\alpha,\beta)=\frac{6C_1(\beta)}{\pi^2\gamma C_2(\beta)}\,\alpha^2-\frac{2g_sC_3(\beta)}{\pi^{3/2}C_2^2(\beta)}\,\alpha^3-\left[\frac{2g_lC_4(\beta)}{\pi^{1/2}C_2(\beta)}-\frac{UC_5(\beta)}{\pi^{1/2}C_2(\beta)}\right]\alpha, \tag{2.17}$$

where $\alpha=\sigma a_0$ and

$$C_1(\beta)=1+2\beta+13\beta^2/4,\quad C_2(\beta)=1+3\beta+15\beta^2/4,$$
$$C_3(\beta)=1+9\beta/2+309\beta^2/32+1395\beta^3/128+23\,745\beta^4/4096,$$
$$C_4(\beta)=1+11\beta/2+449\beta^2/32+2301\beta^3/128+43\,545\beta^4/4096,$$
$$C_5(\beta)=1+2\beta+2\beta^2.$$

The mean size (or radius) of a polaron and bipolaron is easily calculated according to the determination of the mean value

$$\langle r^2\rangle=\int r^2\psi^2(r)dr\quad\text{and}\quad\langle r_{12}^2\rangle=\int r_{12}^2\Psi^2(r_1,r_2)dr_1dr_2,$$

from which we find

$$d_p=\sqrt{\langle r^2\rangle}=\sigma^{-1}\sqrt{3/2}\quad\text{and}$$
$$d_B=\sqrt{\langle r_{12}^2\rangle}=\sigma^{-1}\sqrt{1.5C_6(\beta)/C_2(\beta)}, \tag{2.18}$$

where $C_6(\beta)=1+5\beta+35\beta^2/4$. At $g_l=0$ and strong short-range electron-lattice interaction, only small-radius ($d_B\sim a_0$) L bipolarons can be formed. Such bipolarons are also formed

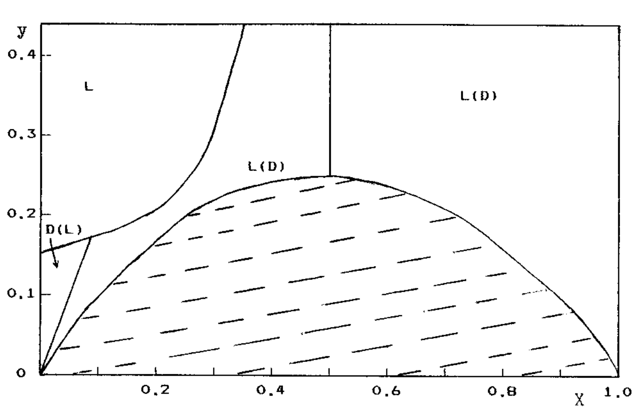

FIG. 2. The phase diagram for the stable D and L states of bipolarons in the coordinates space (x,y).

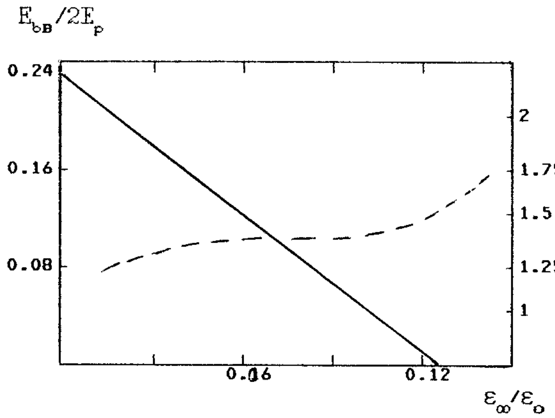

FIG. 3. The dependence of the ratios $E_bB/2E_p$ (solid line) and d_B/d_p (dotted line) on $\varepsilon_\infty/\varepsilon_0$ for D bipolarons at $g_s=0$.

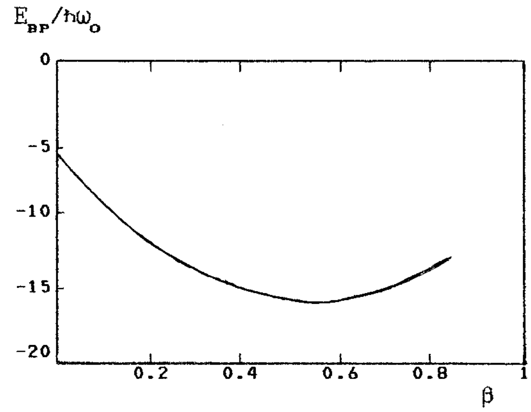

FIG. 4. The dependence of the ground-state energy of the L bipolaron on the parameter β.

at $g_l \neq 0$ and $g_s \neq 0$, and under certain conditions D and L bipolarons can coexist. In the case of the L polaron and bipolaron formation it is usually accepted that $d_p(L)=a_0$ and $d_B(L)=a_0$ The binding energy of D and L bipolarons is determined from Eq. (2.6) after minimizing the functionals (2.16) and (2.17) with respect to α and β.

C. Numerical results and their discussion

For alkali halides and HTSC's the width of the upper valence band is ~ 1 eV,[12,14,27,28] and the Debye energy is $\sim 10^{-2}$ eV. So the condition of the adiabatic approximation $\gamma \sim 10^{-2} \ll 1$ is well satisfied. Then at $\varepsilon_\infty \sim 5$ and $a_0 \sim 5A$ we have $U \sim 100$. Therefore in the calculations of $E_p(\alpha)$ and $E_B(\alpha,\beta)$ we may put g_s, g_l, and $U \sim 0$–120 and $\gamma \sim 0.02$. Below, the results of the numerical calculations for the cases $g_s=0$, $g_l \neq 0$; $g_s \neq 0$, $g_l=0$; and $g_s \neq 0$, $g_l \neq 0$ are presented.

1. Long range electron-lattice interactions and the formation of large-radius delocalized bipolarons

The numerical results show that the role of the correlation between carriers is crucial in D bipolaron formation at $g_s=0$ as was pointed out in Ref. 19. The dependence of the ratio of D bipolaron binding energy $E_{bB}(D)$ to twice the D polaron energy $2E_p(D)$ on $\varepsilon_\infty/\varepsilon_0$ is illustrated in Fig. 3. The dependence of the ratio d_B/d_p on $\varepsilon_\infty/\varepsilon_0$ is also presented. As shown in this figure, the ratio $E_{bB}(D)/2E_p(D)$, calculated by means of trial functions (2.13) and (2.14), is ~ 0.22 at $\varepsilon_\infty/\varepsilon_0 \to 0$ and the formation of a D bipolaron is possible at $\varepsilon_\infty/\varepsilon_0 \leqslant 0.13$. These results are very close to those of Suprun and Moizhes[19] (see also Ref. 20), which were obtained by means of somewhat different trial functions than Eqs. (2.13) and (2.14). From Fig. 3 it is also clear that with the decreasing of the ratio $E_{bB}(D)/2E_p(D)$, d_B/d_p increases, i.e., the size of a D bipolaron increases with increase of $\varepsilon_\infty/\varepsilon_0$ more quickly than the size of a D polaron.

2. Short-range electron-lattice interaction and the formation of continuum small-radius self-trapped bipolarons

In Ref. 23 the electron correlation effect is taken into account in the kinetic and Coulombic repulsive energies of the electrons. Here these effects are taken into account in all terms of the total energy of the considered system. The dependence of the ground-state energy of L bipolarons on the parameter of electron correlation β at $g_l=0$, $\gamma=0.02$, and $U=40$ is presented in Fig. 4. As is evident from this figure, the role of the electron correlation is highly important also in the formation of the continuum small-radius L bipolarons, and the decrease of the total energy of such bipolarons in comparison with the case without electron correlation is three times larger.

3. The combined effect of the short- and long-range electron-lattice interaction on the formation of delocalized and localized bipolarons

Our numerical results show that in the presence of electron correlations and $g_l \neq 0$ the role of a short-range electron-lattice interaction is also essential in the formation of both D and L bipolarons. In the present case the dependence of $E_{bB}(D)/2E_p(D)$ on $\varepsilon_\infty/\varepsilon_0$ has the form shown in Fig. 5. In this figure, for $g_s=70$ and $g_l=20$, the ratio $E_{bB}(D)/2E_p(D)$ reaches up to 0.372 (at $\varepsilon_\infty/\varepsilon_0 \to 0$) which is almost two times larger than 0.22 obtained at $g_s=0$. Further, the formation

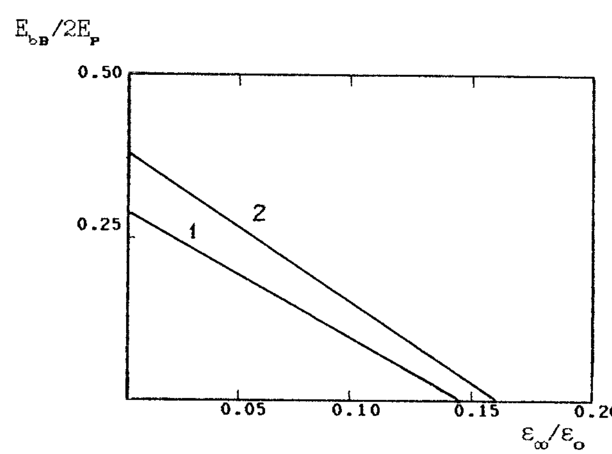

FIG. 5. The dependence of the ratio $E_{bB}/2E_p$ on $\varepsilon_\infty/\varepsilon_0$ for D bipolarons at (1) $g_s=20$, $g_l=50$ and (2) $g_s=70$, $g_l=20$.

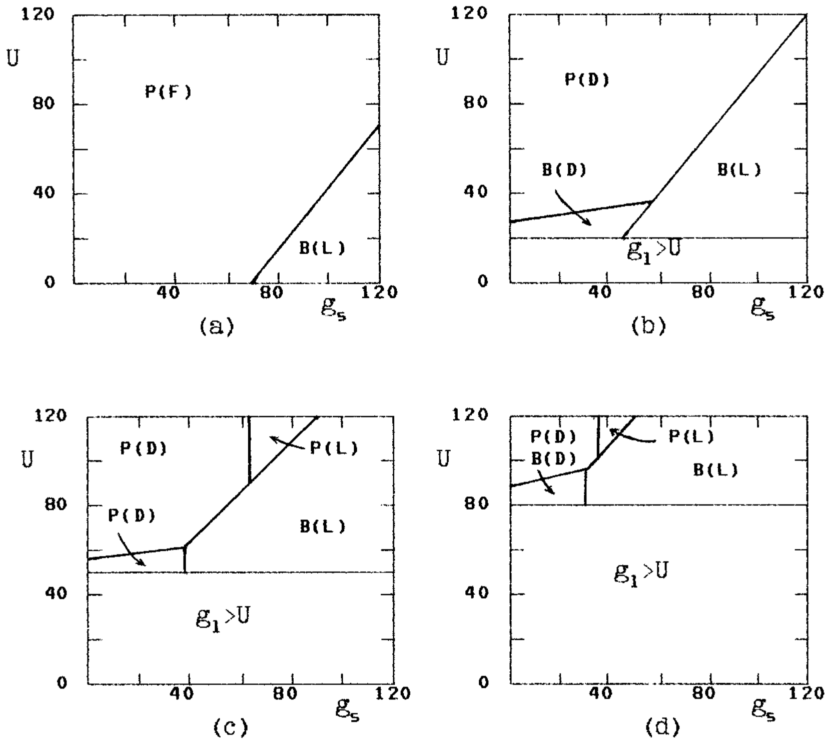

FIG. 6. The phase diagram on the $\{g_s, U\}$ plane of polarons (P) and bipolarons (B) at (a) $g_l=0$, (b) $g_l=20$, (c) $g_l=50$, and (d) $g_l=80$.

of a D bipolaron becomes possible in a wider region of the values $\varepsilon_\infty/\varepsilon_0<0.16$. Now from the relation (2.15) we have

$$\frac{U}{g_l}=\left(1-\frac{\varepsilon_\infty}{\varepsilon_0}\right)^{-1}>1. \tag{2.19}$$

In the two-dimensional space of the parameters (g_s,U), the phase diagrams of the stable states of carriers in the short- and long-range field of the lattice for $\gamma=0.02$ and $g_l=0$, 20, 50, 80 are presented in Fig. 6 (cf. the analogous phase diagram obtained in Ref. 21 with the path integral method). The results obtained by us allow us to make the following conclusions. (a) At $g_l=0$ and comparatively small values of g_s the stable state of carriers is the free (or Bloch) state, and with the increase of g_s this state becomes metastable (at $g_s>g_s^*$ and the value of g_s^* grows with increasing U) and the L bipolaron state is stable [Fig. 6(a)]. (b) At small g_s and g_l the D state of the polarons and bipolarons is stable and at large enough values of g_s the L bipolaron state is stable [Figs. 6(b), 6(c), and 6(d)]. (c) with increasing U, decay of the D and L bipolarons into two D and L polarons occurs.

III. THE PAIRING OF POLARONS IN k SPACE

The above self-trapping scenarios of carriers and their subsequent pairing leads to the formation of spatially separated polarons and bipolarons with zero bandwidth as an ideal Fermi gas and BG, respectively. Hence separate levels of polarons and bipolarons are formed in the energy gap of semiconductors, insulators, and HTSC's. However, with increase of the concentration of polarons their bandwidth becomes nonzero. At a definite bandwidth of the polarons their pairing may be considered in the k space as in BCS-like theory (see also Refs. 5, 7, and 29).

The Hamiltonian of the multicomponent Fermi-gas system with pair interaction between particles has the form[9,10]

$$H=\sum_{nk\sigma}[\varepsilon(k)-\mu_{Fn}]a^\dagger_{nk\sigma}a_{nk\sigma}+\frac{1}{2\Omega}\sum_{nk,lk'}V^F_{nl}(k,k')a^\dagger_{nk\uparrow}a^\dagger_{n-k\downarrow}a_{l-k'\downarrow}a_{lk'\uparrow}, \tag{3.1}$$

where $\varepsilon(k)=\hbar^2k^2/2m_{Fn}, m_{Fn}$ and μ_{Fn} are the mass and chemical potential of the nth polaron, respectively, $a^\dagger_{nk\sigma}$ $(a_{nk\sigma})$ the creation (annihilation) operators of these polarons in state $|nk\sigma\rangle$, k and σ their wave vector and spin indices, respectively, $V^F_{nl}(k,k')$ is the pair interaction potential (which has both an attractive and a repulsive part) between the nth and lth polarons, Ω is the volume of the system, and $n,l=\{1,2,...,\nu\}$, $\nu\leq 4$. The Hamiltonian (3.1) is diagonalized by the standard Bogoliubov transformation of Fermi operators. Then in the excitation spectrum

$$\omega_{Fn}(k)=\sqrt{\tilde{\varepsilon}^2_{Fn}(k)+\Delta^2_{Fn}(k)}$$

there are n energy gaps determined from

$$\Delta_{F_n}(k)=-\frac{1}{\Omega}\sum_{k',l}V^F_{nl}(k,k')\frac{\Delta_{Fl}(k')}{E_{Fl}(k')}\tanh\frac{E_{Fl}(k')}{2T}, \tag{3.2}$$

where $\tilde{\varepsilon}_{Fn}(k)$ is the energy of the nth polaron measured relative to μ_{Fn} and a repulsive Hartree-Fock potential (which can be incorporated and denoted as $\tilde{\mu}_{Fn}$) (see also Ref. 30). For simplicity we consider the case $V^F_{nn}(k,k')\neq 0$ and

$V^F_{nl}(k,k')=0(n\neq l)$. Further, we use the usual BCS-like approximation[8] for $V^F_{nn}(k,k')$:

$$V^F_{nn}(k,k') = \begin{cases} V^F_{Rn}-V^F_{An} & \text{if } \varepsilon(k),\varepsilon(k')<E_{An}=E_{bB}+\hbar\omega_0, \\ V^F_{Rn} & \text{if } E_{An}<\varepsilon(k),\ \varepsilon(k')<E_{Rn}, \\ 0 & \text{if } \varepsilon(k) \text{ or } \varepsilon(k')>E_{Rn}, \end{cases} \tag{3.3}$$

where E_{An} and E_{Rn} are the cutoff parameters for the hybrid attractive V^F_{An} (which has both phonon- and polaron-bag attracting parts) and the repulsive V^F_{Rn} part of the potential $V^F_{nn}(k,k')$, respectively, in the nth polaronic band, $E_{An}\leqslant E_{Rn}$. Then we determine the depairing temperature of the nth bound polaron pairs T_{Fn} and the ratio $g_{Fn}=2\Delta_{Fn}/T_{Fn}$ as

$$T_{Fn}\simeq 1.14(E_{bB}+\hbar\omega_0)\exp(-1/\gamma_{Fn}) \tag{3.4}$$

and

$$g_{Fn}=3.52, \tag{3.5}$$

where $\gamma_{Fn}=\widetilde{V}_{Fn}D_{Fn}$, $\widetilde{V}_{Fn}=V_{An}-V^*_{Rn}$, $V^*_{Rn}=V_{Rn}/[1+D_{Fn}V_{Rn}\ln(E_{Rn}/E_{An})]$ and D_{Fn} are the effective interaction potential and density of states in the nth polaronic band, respectively. Here the combined phonon- and polaron-bag-mediated processes are taken into account as in the case of the combined electron- and phonon-mediated ones in Ref. 4. The expression for the cutoff energy obtained within the spin- and correlation-bag approaches[5,31] is also similar to the results of Ref. 4. So our approach is a modified and generalized variant of the BCS theory. In the absence of static lattice deformation [which occurs in many low-temperature superconductors (LTSC's)] $E_{bB}=0$ and we have a pure BCS picture. In general, a pure BCS pairing is possible only in F states of carries, whereas in their D and L states the combined BCS and non-BCS (i.e., polaron bag) pairing is always realized. Further, when the Fermi energy of the nth polarons $E_{Fn}<(E_{bB}+\hbar\omega_0)$ (which takes place in, e.g.,[32] ^{3}He) the cutoff energy in Eq. (3.3) is replaced by E_{Fn} (see Ref. 33) or the width of the polaronic band.[29] In the case of $0\leqslant V^*_{Rn}\leqslant V_{An}$, $0<\widetilde{V}_{Fn}D_{Fn}\leqslant 1$, and the above modified BCS-like pairing theory of polarons is quite applicable.

IV. DISCUSSION OF THE RESULTS AND THEIR POSSIBLE RELATION TO HIGH-T_c SUPERCONDUCTIVITY

Now we compare the theoretical results obtained with earlier existing ones and experimental situations, as well as the distinctive features of F, D, and L states of pairing carriers responsible for the realization of the different SC states. The ground states of carriers, which depend on the quantity of short- and long-range electron-lattice interactions as well as the Coulomb repulsion between them, are F, D, and L states. It is clear that the carriers in D and L states are static polarons and bipolarons, whereas they in F-states they are dynamic. From the above it follows that not only electron correlation but also the short-range electron-lattice interaction play an important role in the formation of D and L bipolarons. As is clear from Eq. (2.7) the formation of D bipolarons without electron correlation is impossible both for $g_{s1}=0$ and for $g_{l1}=0$ (cf. Refs. 15 and 16), while according to Eq. (2.8) the formation of L bipolarons is impossible only for $g_{s1}=0$ but for $g_{s1}\neq 0$ it becomes possible for any g_{l1} (cf. Ref. 16). So the short-range electron-lattice interaction plays a crucial role in the formation of not only L (cf. Ref. 25) but also D states of bipolarons (cf. Refs. 15 and 16). Unlike the L bipolarons, the formation of D bipolarons is possible without such interaction also. The binding energy of a D bipolaron in the absence of electron correlation may reach up to 21% twice of the polaron energy $2E_p(D)$ (at $x\to 0$) and the existence region of the bipolaron is sufficiently wide, $x\leqslant 0.075$. It should be noted that at $g_{s1}=0$ and $g_{l1}\neq 0$ the inclusion of the electron correlations within the path integral method gives nearly the same result for the existence region of D bipolarons, $x\leqslant 0.079$.[21] Further, some variational calculations indicate that, with the inclusion of electron correlation at $g_{s1}=0$ and $g_{l1}\neq 0$, the ratio $E_{bB}(D)/2E_p(D)\simeq 0.22$.[19,20] It is interesting that inclusion of either the electron correlation (at $g_{s1}=0$) or the short-range electron-lattice interaction (at $g_{l1}\neq 0$) gives nearly the same results, while inclusion of both the short-range electron-lattice interaction and the electron correlation leads to an essential increase of the binding energy of D bipolarons, reaching up to 37% of twice the polaron energy, and to a noticeable broadening of their existence region up to $x\leqslant 0.16$. So the formation possibility of D bipolarons in real materials without and with electron correlation is restricted by the conditions $x\leqslant 0.075$ and $x\leqslant 0.16$, respectively, which are satisfied in some compounds, such as TiO_2 ($\varepsilon_\infty\simeq 6$–$7.2$, $\varepsilon_0=170$),[34] $SiTiO_3$ ($\varepsilon_\infty=5.2$, $\varepsilon_0=2300$),[19] oxide HTSC's ($\varepsilon_\infty=2$–5, $\varepsilon_0=50$–85),[16,35] BaO($\varepsilon_\infty=4$, $\varepsilon_0=34$),[34] metal-ammonia solutions ($\varepsilon_\infty=1.8$, $\varepsilon_0=22$),[17] and probably in others (e.g., organic HTSC's, intermetallic compounds, Ti_4O_4, etc.). The formation conditions (2.8), (2.9), and (2.10) for L bipolarons are satisfied in many compounds. Thus the experimental observation of small bipolarons (possibly above the continuum L bipolarons) and the absence of such evidence for D bipolarons most probably are caused by these circumstances.[18] The observed deep localized states in the energy gap, effective mass of carriers $m^*\simeq(10$–$30)m_e$, and coherence length $\xi\simeq 4$–50 Å in oxide and organic HTSC's indicate the existence there of D and L polarons and bipolarons.[12,13,36–39] It is reasonable to assume that increasing the concentration of D and L polarons and bipolarons having thick local deformation clouds leads to the overlap of their deformation clouds, which later on become thinner but more extended. This leads to decreasing of the lattice deformation or m^* and E_{bB} and broadening of the bandwith of polarons and bipolarons. In such situations the pairing of carriers cannot be considered in r space. The collective effects then lead to modulation of the basic parameters of pairing polarons (see also Ref. 5). In the present case, the k-space approximation is better suited for study of the pairing of polarons.

We now discuss the possible relevance of our results (in particular, the distinctive pairing features of F, D, and L carriers) to the superconductivity mechanism in different superconductors. In the F state of carriers the static lattice deformation is absent. The pairing of F carriers is possible if we consider the dynamic effects of their dressing in the phonon cloud, i.e., their interaction with the virtual phonons. In the present case dynamic F polarons and bipolarons (i.e., Cooper pairs) are formed. The conventional BCS theory of superconductivity (see Refs. 9–11) describes only the Cooper pairing processes of such F polarons by means of phonon exchange. The BCS theory may be modified also to the cases of the pairing of D and L polarons, analogous to the exciton, plasmon, and other nonphonon pairing mechanisms of carriers.[2,4] Here the contribution of both the dynamic and static lattice deformations, i.e., the combined phonon- and polaron-bag-mediated processes, should be taken into account in the pairing of D and L polarons, as for phonon- and electron-bag-mediated processes.[4] Then the depairing temperature of these polarons may be determined by Eq. (3.4).

It is evident that the pairing mechanism of carriers is phonon and polaron mediated at $E_{bB} \ll \hbar\omega_0$ and $E_{bB} \gg \hbar\omega_0$, respectively. The first takes place for D and especially F polarons and the second for L polarons. Indeed, for HTSC's the Fermi energy $E_F \sim 7eV$,[40,41] $B \sim 1$ eV,[7] $K \sim 1.4\times10^{12}$ dyn/cm^2,[42] $a_0 \simeq 4$ Å, $E_d \simeq (2/3)E_F \simeq 4.66$ eV, $x \simeq 0.03$–0.10, $g_{s1} \simeq 0.19$, $E_{bB}(D) \simeq 0.01$–0.04 eV (for $x=0.04$–0.05 and $y=0.16$–0.17), $E_{bB}(L)=0.24$–0.31 eV (for $x=0.04$–0.05 and $y \simeq 0.217$–0.260), and $\hbar\omega_0 \simeq 0.06$ eV.[43] For $x=0.05$ and $y \simeq 0.16$ the mean size of D and L bipolarons is $d_D \simeq a_0/\alpha_D > a_0$ and $\simeq a_0$, respectively, which agree well with the observed values of the coherence length $\xi \simeq 4$–10 Å in HTSC's.[37] The effective mass of D polarons is $m_p^* = \pi^2\hbar^2/2d_p^2E_p(D) \simeq 4.27m_e$ (the mean size of the D polaron $d_D \sim a_D/\alpha_D \geq 2a_0$) and that of L polarons is $\sim \pi^2\hbar^2/2a_0^2E_p(L) \simeq 7.23m_e$. Then, according to Ref. 15 the effective masses of D and L bipolarons are equal to $m_B^* = 4m_p^* \simeq 12m_e$ (for $x=0.05$ and $y=0.16$) and $\simeq 17m_e$ (for $\varepsilon_\infty=2.5$ and $x=0.05$), $=29m_e$ (for $\varepsilon_\infty=3$ and $x=0.05$), which also agree well with the observed values of the effective masses of carriers, $m^* \simeq (3-30)m_e$ and $\simeq (10$–$30)m_e$, respectively, in titanates and oxide HTSC's.[12,13] For the determination of T_F according to Eq. (3.4) it is reasonable to assume that $\gamma_{Fn} \simeq 0.35$ [usually $\gamma_{Fn} \simeq 0.25$–0.50 (Refs. 44 and 45)]. Then for D and L bipolarons with binding energies $E_{bB}(D) \simeq 0.03$ eV and $E_{bB}(L) \simeq 0.24$ eV, $E_{bB}(L) \simeq 0.26$ eV, we find $T_F \simeq 68$, $\cong 227$, and $\simeq 242$K, respectively. Apparently, the disappearance of some order parameters below T_c near $T \simeq 60$–70 K,[46] and phase transitions above T_c at $T \simeq 230$–240 K,[47] in HTSC's are caused by the depairing of such D and L bipolarons. In the Fermi system the pairing of carriers and the formation of their BCS-like state is only a necessary but not a sufficient condition for the appearance of superfluidity[9] (superconductivity) (see also Ref. 2). Indeed, in this system the SC phase transition may be considered as a two-stage process accompanied as a rule, by the formation of composite bosons (F, D, and L bipolarons), with their subsequent transition to the superfluid state by means of single-particle and pair condensation.[10,11] So the appearance of superconductivity is possible with the existence of order parameters of both attracting fermion pairs Δ_F (determined according to the above BCS-like theory) and boson pairs Δ_B (determined according to the single-particle and pair condensation theory of composite bosons[10,11]). According to Refs. 9–11 the critical temperature of the SC transition T_c for the so-called fermion superconductors (FSC's) $[\Delta_F(T=T_F) = \Delta_B(T=T_F)=0]$ and boson superconductors, (BSC's) $[\Delta_B(T=T_B)=0$ and $\Delta_F(T=T_F>T_B)=0]$ is determined as $T_c=T_F=T_B \geq T_{\rm BEC}$ (where $T_{\rm BEC}$ is the Bose-Einstein condensation temperature of an ideal BG) and $T_c=T_B<T_F$, respectively. In FSC's, near $T=T_F$, the number of composite bosons $n_B(T) \ll n_B(0)$ and they may be considered as an ideal BG, while in BSC's $n_B(T) \simeq n_B(0)=$const up to $T=T_B$. For a three-dimensional attracting BG $T_B \simeq T_{\rm BEC}[1+1.427\gamma_B\sqrt{T_{\rm BEC}/\zeta_A}]$,[11] where γ_B is the boson-boson coupling constant, ζ_A is the cutoff parameter for the attractive part of the boson-boson interaction potential, and $\gamma_B \ll 1$, $T_{\rm BEC}/\zeta_A \ll 1$.

We now estimate $T_{\rm BEC}$ and $T_c=T_B$ at $n_B=n_B(0)$ for D and L bipolarons. At $n_B(0) \simeq 10^{21}$ cm^{-1} for $m_B^* \simeq 12m_e$, $17m_e$, and $29m_e$, we find $T_{\rm BEC} \simeq 244$, 172, and 101 K, respectively. Then, assuming $\gamma_B \simeq 0.2$ and $T_{\rm BEC}/\zeta_A \simeq 0.1$ we obtain $T_c=266$, 187, and 110 K, respectively. Indeed, in $YBa_2Cu_3O_{7-x}$ the SC transition is observed in the interval $T_c \simeq 110$–220 K.[48] As is clear for D bipolaron, $T_F \ll T_B$ when $n_B \simeq n_B(0)$. However, at $T \to T_F$ the number of such D bipolarons rapidly decreases and as a result $T_B \to T_{\rm BEC} \to T_F$. For the two types of L bipolarons with $m_B^* \simeq 17m_e$ and $\simeq 29m_e$ the condition $T_F > T_B$ is satisfied. Here the value of $T_c=T_B \simeq 110$–187 K is very close to the observed values of T_c in HTSC's.[48] Consideration of the distinctive features of F, D, and L bipolarons is very important for determination of not only T_F, $T_{\rm BEC}$, T_B, and T_c, but also other SC parameters, such as the London penetration depth λ_L, the critical magnetic fields H_{c1} and H_{c2}, etc. So, for example, at $n_B(0) \simeq 10^{21}$ cm^{-3} in the case of F bipolarons, assuming $m_B^* \simeq 2m_p$, we find $\lambda_L=[m_B^*C^2/4\pi n_B(0)e^2]^{1/2} \simeq 2400$ Å and in the case of D and L bipolarons with $m_B^* \simeq 12m_e$ and $\simeq 29m_e$ we obtain $\lambda_L \simeq 3400$ and $\simeq 8200$ Å, respectively. These values of λ_L are consistent with the observed ones; $\simeq 1400$–$10\,000$ Å in HTSC's.[49,50] Further, coexistence of F, D, and L bipolarons can explain other observed anomalous properties of HTSC's as well as type-II LTSC's, heavy-fermion and organic superconductors. These include, for example, two SC order parameters,[37] mixed states in the interval of magnetic field $H_{c1}<H \leq H_{c2}$ and temperature $T_{c1}<T<T_{c2}$,[51] the fractional Meissner effects in type-III superconductors,[52] the jump of specific heat at $T=T_{c1}<T_c$ and $T=T_c=T_{c2}$,[51,53] etc. Coexistence of two types of order parameters Δ_F and Δ_B allows one to explain the nonzero density of states inside the assumed SC gap $\Delta_{\rm SC}$ $(=\Delta_F)$ in HTSC's and LTSC's (Ref. 43) [including the precursor peak observed[44] outside $\Delta_{\rm SC}$ $(=\Delta_F)$ in some LTSC's] as well as the existence of a pseudogap $\Delta_{\rm SC}$ in the normal state in HTSC's.[54,55] Thus, our alternative more general pairing theory allows us to interpolate between the BCS-like and r-space pairing limits, as is done already in Refs. 56 and 57, but with essential distinctive approaches to the problem. In particular, our approach allows us to find the real applicability boundary (which up to now remains unknown) between BCS-like and r-space pairing regimes. Moreover, unlike Refs. 7, 16, 17, 56, and 57, we show that the r-space pairing state of polarons and the BEC of r-space bipolarons due to their zero bandwidth and immobility are irrelevant to the superconductivity.

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