Theory of high-temperature superconductivity in doped polar insulators

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received 21 April 2011; accepted in final form 31 May 2011
published online 28 June 2011

PACS 71.38.-k – Polaron and electron-phonon interactions
PACS 72.15.Jf – Thermoelectric and thermomagnetic effects
PACS 74.72.-h – Cuprate superconductors

Abstract – Many high-temperature superconductors are highly polarizable ionic lattices where the Fröhlich electron-phonon interaction (EPI) with longitudinal optical phonons creates an effective attraction of doped carriers virtually equal to their Coulomb repulsion. The general multi-polaron theory is given with both interactions being strong compared with the carrier kinetic energy so that the conventional BCS-Eliashberg approximation is inapplicable. The many-electron system is described by the polaronic $t$-$J_p$ Hamiltonian with reduced hopping integral, $t$, allowed double on-site occupancy, large phonon-induced antiferromagnetic exchange, $J_p \gg t$, and a high-temperature superconducting state of small superlight bipolarons protected from clustering.

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Introduction. – It seems plausible that the true origin of high-temperature superconductivity is found in a proper combination of the finite-range Coulomb repulsion with a significant finite-range EPI as suggested by a growing number of experimental and theoretical studies [1]. In highly polarizable ionic lattices like cuprate superconductors both interactions are quite strong (of the order of 1 eV) compared with the low Fermi energy of doped carriers because of a poor screening by non- or near-adiabatic carriers [2]. In those conditions the BCS-Eliashberg theory [3] breaks down because of the polaronic collapse of the electron bandwidth [4].

The many-body theory for polarons has been developed for extremely weak and strong EPI. In the weak-coupling limit this problem is reduced to the study of a structure factor of the uniform large polaron gas [5]. For strong coupling the problem is reduced to on-site [6] or inter-site [7,8] small bipolarons on a lattice. A strong enhancement of $T_c$ was predicted in the crossover region from the BCS-like polaronic to BEC-like bipolaronic superconductivity due to a sharp increase of the density of states in a narrow polaronic band [4], which is missing in the so-called negative Hubbard $U$ model. Nevertheless the theory of dense polaronic systems in the intermediate coupling regime remains highly cumbersome, in particular, when EPI competes with strong electron correlations.

Corresponding microscopic models with the on-site Hubbard repulsion and the short-range Holstein EPI have been studied using powerful numerical techniques [9,10].

In most analytical and numerical studies mentioned above and many others both interactions are introduced as input parameters not directly related to the material. Quantitative calculations of the interaction matrix elements can be performed from pseudopotentials using the density functional theory (DFT) [11]. On the other hand, one can express the bare Coulomb repulsion and EPI through material parameters rather than computing them from first principles in many physically important cases [12]. In particular, for a polar coupling to longitudinal optical phonons (the Fröhlich EPI), which is the major EPI in polar crystals, both the momentum dependence of the matrix element, $M(q)$, and its magnitude are well known, $|M(q)| = \gamma(q)\hbar\omega_0/\sqrt{2N}$ with a dimensionless $\gamma(q) = \sqrt{4\pi e^2 / \kappa \Omega \hbar \omega_0 q^2}$, where $\Omega$ is a unit cell volume, $N$ is the number of unit cells in a crystal, $\omega_0$ is the optical phonon frequency, and $\kappa = \epsilon_\infty \epsilon_0 / (\epsilon_0 - \epsilon_\infty)$. The high-frequency, $\epsilon_\infty$ and the static, $\epsilon_0$ dielectric constants are both measurable in a parent polar insulator. As is well known, a two-particle bound state exists even in the weak-coupling regime, $\lambda < 0.5$, due to a quantum (exchange) interaction between two large polarons forming a large bipolaron [1] ($\lambda$ is the familiar EPI constant of the BCS-Eliashberg theory). These weakly coupled large pairs overlap in dense systems, so that their many-particle

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ground state is a BCS-like superconductor with Cooper pairs (see below).

Here the analytical multi-polaron theory is given in the strong-coupling regime for highly polarizable lattices with \( \epsilon_0 \gg 1 \).

**Generic Hamiltonian and its canonical transformation.** – The dielectric response function of strongly correlated electrons is a priori unknown. Hence one has to start with a generic Hamiltonian including unscreened Coulomb and Fröhlich interactions operating on the same scale since any ad hoc assumption on their range and relative magnitude might fail,

\[
H = -\sum_{i,j} (T_{ij} \delta_{ss'} + \mu \delta_{ij}) c_i^\dagger c_j + \frac{1}{2} \sum_{ij,q} e^2 \sum_{n,m} e^{i(q \cdot (m-n))} \tilde{n}_i \tilde{n}_j \\
+ \sum_{q,i} \int_\text{d}q \rho_{\text{q}} u(m, q)d_q + \text{H.c.} + H_{ph}.
\]

Here \( T_{ij} \equiv T(m-n) \) is the bare hopping integral, \( \mu \) is the chemical potential, \( i, j = m, n \) and \( s, s' \) include both site \( (m, n) \) and spin \( (s, s') \) states, \( u(m, q) = (2N)^{-1/2} \) is the electron phonon operators, respectively, \( \tilde{n}_i = c_i^\dagger c_i \) is a site occupation operator, and \( H_{ph} = \sum_q \omega_{q} \rho_{\text{q}} d_q^\dagger d_q + 1/2 \) is the polar vibration energy.

In highly polarizable lattices with \( \epsilon_0 \to \infty \) the familiar Lang-Firsov (LF) [13] canonical transformation \( e^{-S} \) is particulary instrumental with \( S = -\sum_{i,j} \int_\text{d}q \rho_{\text{q}} u(m, q)d_q - \text{H.c.} \). It shifts the ions to new equilibrium positions changing the phonon vacuum, and removes most of both interactions from the transformed Hamiltonian, \( H = e^{S} H e^{-S} \),

\[
\tilde{H} = -\sum_{i,j} (\sigma_{ij} \delta_{ss'} + \tilde{\mu} \delta_{ij}) c_i^\dagger c_j + H_{ph},
\]

where \( \sigma_{ij} = T(m-n) X_i^\dagger X_j \) is the renormalised hopping integral involving the multi-phonon transitions described with \( X_i = \exp(\sum_q u(m, q)d_q - \text{H.c.}) \), and \( \tilde{\mu} = \mu + E_p \) is the chemical potential shifted by the polaron level shift,

\[
E_p = \frac{2\pi e^2}{\kappa} \int_{\text{BZ}} \frac{d^3q}{(2\pi)^3} q^2.
\]

Here, the integration goes over the Brillouin zone (BZ) and \( E_p = 0.647 \) eV in La$_2$CuO$_4$ [2]. The electron-phonon coupling constant is defined as \( \lambda = 2E_p N(0) \). In the case of 2D carriers with a constant bare density of states, \( N(0) = ma^2/2\pi h^2 \) per spin, eq. (3) places cuprates in the strong-coupling regime, \( \lambda \geq 0.5 \), if the bare band mass \( m > m_c \) (here \( \sigma \) is the in-plane lattice constant).

**Weak-coupling regime.** – For comparison, let us first consider the weak-coupling limit, where not only \( \lambda < 0.5 \) but also the number of phonons dressing the carrier is small, \( E_p/\hbar \omega_0 \ll 1 \). In this limit one can expand \( \tilde{X}_i \) in eq. (2) in powers of \( \gamma(q) \) keeping just single-phonon transitions so that (in the momentum representation)

\[
\tilde{H} \approx \sum_{k,s} \xi_k c_{k,s}^\dagger c_{k,s} + H_{ph} \\
+ \sum_{q,k,s} M(k, q)c_{k+q, s}^\dagger c_{k, s} (d_q - d_{-q})^\dagger,
\]

where \( \xi_k = E(k) - \mu \), \( E(k) = -\sum_m T(m) \exp(i m \cdot k) \) is the bare band dispersion, and \( M(k, q) = \gamma(q) |E(k) - E(k')|/\sqrt{2N} \) is the transformed EPI matrix element, renormalised by the Coulomb repulsion. There are no other interactions left in the transformed Hamiltonian since the bare Coulomb repulsion is nullified by the Fröhlich EPI.

Applying the BCS-Eliashberg formalism [3] yields the master equation for the superconducting order parameter, \( \Delta(\omega_n, k) \),

\[
\Delta(\omega_n, k) = k_B T \sum_{k', \omega_n'} \frac{\tilde{M}(k, k-k')^2 D(\omega_n - \omega_n') \Delta(\omega_n', k')} {\omega_n' + \xi_k + [\Delta(\omega_n', k')]^2},
\]

where \( D(\omega_n, \omega_n') = -\hbar \omega_n |(\omega_n - \omega_n')^2 + \gamma^2| \) is the phonon propagator and \( \omega_n = \pi k_B T (2n + 1) \) are the Matsubara frequencies \( (n = 0, \pm 1, \pm 2, \pm 3, \ldots) \). Depending on a particular shape of the band dispersion, eq. (5) allows for different symmetries of the order parameter since EPI is not local [14]. Here we confine our analysis to a simple estimate of \( T_c \) by assuming a \( k \)-independent gap function, \( \Delta(\omega_n) \). Then factorizing the kernel in eq. (5) on the “mass shell”, \( E(k') - E(k) = \omega_n - \omega_n' \) and linearizing eq. (5) with respect to the gap function one obtains the familiar estimate of the critical temperature, \( k_B T_c \approx \hbar \omega_n \exp[-1/(\lambda - \mu^*_n)] \), where \( \mu^*_n = \lambda/(1 + \lambda L) \) is the Coulomb pseudopotential. In our case the weak-coupling BCS superconductivity with \( k_B T_c \ll \hbar \omega_0 \) exists exclusively due to the “Tolmachev-Morel-Anderson” logarithm \( L = \ln(\mu/\hbar \omega_0) > 1 \), if the EPI is retarded (i.e. \( \hbar \omega_0 > \mu \)).

**Strong-coupling regime.** – Actually the number of virtual phonons in the polaron cloud is large in oxides and some other polar lattices, \( E_p/\hbar \omega_0 \gg 1 \) with the characteristic (oxygen) optical phonon frequency \( \hbar \omega_0 \lesssim 80 \text{ meV} \), so that multi-phonon vertexes are essential in the expansion of the hopping operator \( \tilde{\sigma}_{ij} \). To deal with this challenging problem let us single out the coherent hopping in eq. (2) averaging \( \tilde{\sigma}_{ij} \) with respect to the phonon vacuum, and consider the remaining terms as perturbation, \( \tilde{H} = H_0 + H_{p-ph} \), Here

\[
H_0 = -\sum_{i,j} (t_{ij} \delta_{ss'} + \tilde{\mu} \delta_{ij}) c_i^\dagger c_j + H_{ph}
\]

describes free phonons and polarons coherently propagating in a narrow band with the exponentially diminished hopping integral, \( t_{ij} = T(m-n) \exp[-y^2(m-n)] \),

\[
y^2(m) = \frac{1}{2N} \sum_q \gamma(q)^2 |1 - \cos(q \cdot m)|.
\]
is the residual polaron–multi-phonon interaction, which is a perturbation at large $\lambda$. In the diagrammatic technique the corresponding vertices have any number of phonon lines as shown in Fig. 1 for the second-order in $H_{p-ph}$ polaron self-energy ($\Sigma_p \approx -E_p/2z\lambda^2$) and the phonon self-energy ($\Sigma_{ph} \approx -x\hbar\omega_0/z^2$) [15], where $x$ is the lattice coordination number and $z$ is the atomic density of carriers. Hence the perturbation expansion in $1/\lambda$ is applied if $\lambda \gg 1/\sqrt{2x}$ [15,16]. Importantly there is no structural instability in the strong-coupling regime since $[\Sigma_{ph}] \ll \hbar\omega_0$ [15].

The LF transformation, eq. (2) is exact for any adiabatic ratio $\hbar\omega_0/T(a)$. However, if the perturbation expansion in $1/\lambda$ is restricted by lowest orders, then it significantly overestimates polaron masses in the adiabatic regime, $\hbar\omega_0/T(a) < 1$, for the case of the short-range (Holstein) EPI [1] (here $T(a)$ is the nearest-neighbor bare hopping integral). The polaronic band narrowing factor, $\exp(-g^2)$ becomes very small for this EPI in the strong-coupling regime, which would eliminate any possibility of high temperature superconductivity and even metallicity of the small Hosltein polarons.

In our case of the long-range (Fröhlich) EPI, Quantum Monte Carlo simulations [17] show that the LF transformation provides numerically accurate polaron masses already in the zero order of the inverse-coupling expansion both in the adiabatic regime as well as in the non-adiabatic one for any strength of the Fröhlich EPI. Moreover, such small polarons [17] and small bipolarons [7] are perfectly mobile in the relevant range of the coupling and the adiabatic ratio [1].

The perturbation $H_{p-ph}$ has no diagonal matrix elements with respect to phonon occupation numbers. Hence it can be removed from the Hamiltonian in the first order using a second canonical transformation $H = e^{S_2}He^{-S_2}$ with $(S_2)_{n,n'} = \sum_{i,j}(\delta_{ij} - t_{ij})c^\dagger_ic_j/(E_n - E_{n'})$, where $E_n$ and $E_n'$ are the energy levels and the eigenstates of $H_0$, respectively. Taking into account that the polaron Fermi energy is small compared with the phonon energy at strong coupling and/or sufficiently low doping [2], one can neglect its contribution to $E_n - E_n' \approx \hbar\omega_0 \sum_q n_q$ and project the second-order in $1/\lambda$ Hamiltonian $H$ onto the phonon vacuum $|0\rangle$ with the following result:

$$H = -\sum_{i,j}(t_{ij}\delta_{ss'} + \tilde{\mu}\delta_{ss'})c^\dagger_ic_j$$

$$- \sum_{mnm'n',ss'}V_{mn}^{m'n'}c^\dagger_mc_s^\dagger_{n's'}c^\dagger_{n's'},$$

fig. 1

where

$$V_{mn}^{m'n'} = it_{ij}T_{ij}^f\int_0^\infty dt_0 e^{-\delta t_0}[\hat{X}_i(t)\hat{X}_j(t)$$

$$- e^{-\sigma(m-n)}\hat{X}_i^\dagger(0)\hat{X}_j^\dagger(0),$$

and $\hat{X}_i(t)$ is the Heisenberg multi-phonon operator obtained by replacing $d_q$ in $\hat{X}_i$ with $d_q \exp(\omega_q t)$. Calculating the integral, eq. (10), with $\delta \to +0$ yields

$$V_{mn}^{m'n'} = \frac{t_{ij}t_{ij}^f}{\hbar\omega_0} \sum_{k,k'=1}^{\infty} \frac{f(m,m',n')}{kk'},$$

where $f(m,m',n') = (1/2N)\sum_q \gamma(q^2)[\cos(q \cdot (m - n')) + \cos(q \cdot (m - m')) - \cos(q \cdot (n - n'))].$

Polaronic $t-J_p$ Hamiltonian. – All matrix elements, eq. (11), of the polaron-polaron interaction are small compared with the polaron kinetic energy except the exchange interaction, $J_p(m - n) \equiv V_{mn}^{m'n'}$ such that $f(m,m',n') = g^2(m - m')$. Using $\sum_{k=1}^{\infty} k'/kk' = -C - \ln(y) + Ei^*(y)$ with $C \approx 0.577$ and $Ei^*(y) \approx e^y/y$ (for large $y$) one obtains a substantial $J_p(m) = T^2(m)/(2g^2(m)\hbar\omega)$, which is much larger than the polaron hopping integral, $t/J_p \propto 2\hbar\omega_0 g^2e^{-s/\lambda}/T(a) \ll 1$ in the strong-coupling limit. Here $t$ is the nearest-neighbor polaron hopping integrals. Keeping only this exchange we finally arrive with the polaronic “$t-J_p$” Hamiltonian,

$$H = -\sum_{i,j}(t_{ij}\delta_{ss'} + \tilde{\mu}\delta_{ss'})c^\dagger_ic_j$$

$$+ 2\sum_{m \neq n} J_p(m - n) \left( \tilde{S}_m \cdot \tilde{S}_n + \frac{1}{4} n_m n_n \right),$$

where $\tilde{S}_m = (1/2)\sum_{s,s'} c^\dagger_{m's'}\vec{c}_{m's'}$ is the spin-1/2 operator ($\vec{c}$ are the Pauli matrices), $n_m = \sum_i n_i$, and $\tilde{\mu} = \tilde{\mu} + \sum_m J_p(m)$ is the chemical potential further renormalized by $H_{p-ph}$.

There is a striking difference between this polaronic $t-J_p$ Hamiltonian and the familiar $t-J$ model derived from the repulsive Hubbard $U$ Hamiltonian in the limit $U \gg t$ omitting the so-called three-site hoppings and EPI [18]. The latter model acts in a projected Hilbert space constrained.
to no double occupancy. Within this standard $t$-$J$ model the bare transfer amplitude of electrons ($t$) sets the energy scale for incoherent transport, while the Heisenberg interaction ($J \propto t^2/U$) allows for spin flips leading to coherent hole motion with an effective bandwidth determined by $J \ll t$. Using the Gutzwiller-type approximation to remove the constraint results in an unconstrained $t$-$J$ model also containing a band narrowing, but purely electronic rather than phononic origin [19]. On the contrary in our polaronic $t$-$J_p$ Hamiltonian, eq. (12), there is no constraint on the double on-site occupancy since the Coulomb repulsion is negated by the Fröhlich EPI. The polaronic hopping integral $t$ leads to the coherent (bi)polaron band and the antiferromagnetic exchange of purely phononic origin $J_p$ bounds polarons into small superlight inter-site bipolarons. Last but not least the difference is in the “$+$” sign in the last term of eq. (12) proportional to $\tilde{n}_m\tilde{n}_n$, which protects the ground superconducting state from the bipolaron clustering, in contrast with the “$-$” sign in the similar term of the standard $t$-$J$ model, where the phase separation is expected at sufficiently large $J$ [20].

The cancellation of the bare Coulomb repulsion by the Fröhlich EPI is accurate up to a $1/e_0$ correction. This correction produces a long-range residual repulsion of (bi)polarons in the transformed Hamiltonian, eq. (12), which is small as soon as $e_0 > e^2/a J_p$. The residual repulsion results in some screening of the Coulomb interactions responsible for the doping dependence of the bipolaron binding energy and of the (bi)polaron mass [2]. In layered polar insulators the static dielectric constant could be anisotropic, which together with local field corrections might result in different EPI matrix elements for in-plane and out-of-plane polarised optical phonons, respectively. The difference is not considered here.

Projection onto bipolaronic Hamiltonian and high $T_c$ — The polaronic $t$-$J_p$ Hamiltonian, eq. (12), is analytically solvable in the limit of sufficiently low atomic density of carriers, $x \ll 1$. Neglecting the first term in $\mathcal{H}$, which is the polaron kinetic energy proportional to $t \ll J_p$, one can readily diagonalise the remaining spin-exchange part of the Hamiltonian. Its ground state is an ensemble of inter-site singlet bipolarons with the binding energy $\Delta_b = J_p$ localised on nearest neighbor sites. Such small bipolarons repel each other and single polarons via a short-range repulsion of about $J_p$.

The kinetic energy operator in eq. (12) connects singlet configurations in the first and higher orders with respect to the polaronic hopping integrals. Taking into account only the lowest-energy degenerate singlet configurations and discarding all other configurations one can project the $t$-$J_p$ Hamiltonian onto the inter-site bipolaronic Hamiltonian using the bipolaron annihilation operators $B_m = \sum_n C_{n,\alpha} b^\dagger_{n,\alpha} + C_{n,\alpha}^\dagger b_{n,\alpha} + C_{m,\alpha} b_{m,\alpha} + C_{m,\alpha}^\dagger b^\dagger_{m,\alpha}$, where $\alpha$ connects nearest neighbors [7]. These operators are similar to the bond-order operators introduced later by Newns and Tsuei [21], which are weakly coupled in their model with the single-plane lattice vibrations via a nonlinear (two-phonon) EPI (eq. (9) in ref. [21]). Actually it is well known that the nonlinear anharmonic corrections to EPI are small compared with the linear Fröhlich interaction in real three-dimensional solids, which makes the two-dimensional model of ref. [21] of cuprate superconductors unfeasible.

Our strong-coupling projection is illustrated using a polar lattice, sketched in fig. 2(a), of anion-cation triangular planes (the in-plane lattice constant is $a$ and the nearest-neighbor hopping distance is $a/2$) separated by the out-of-plane lattice constant $c$. For a zig-zag ladder fragment of the lattice, fig. 2(b), the projected bipolaronic Hamiltonian in the nearest-neighbor hopping approximation is

$$H_b = -t \sum_n B_n^\dagger A_n + D_n^\dagger B_n + D_n^\dagger A_n + C_n^\dagger B_n + A_{n+1}^\dagger C_n$$

$$+ A_{n+1}^\dagger B_n + B_{n-1}^\dagger A_n + C_{n-1}^\dagger A_n + H.c.,$$

with the center-of-mass momentum $hK$. Expanding in powers of $K$ one obtains the effective mass of these small singlets, $m^* = 10 m^*$, where $m^* = 2h^2/5t(a/2)^2$ is the polaron mass. A similar Hamiltonian can be derived also for a square lattice, if next-nearest-neighbor hopping integrals are taken into account [22]. Small bipolarons are hard-core bosons with the short-range repulsion of the radius $r = a/2$ and a huge anisotropy of their effective mass since their inter-plane

![Fig. 2: (Color online) A plane of the 3D polar lattice of anions (circles) and cations (crosses) (a) with doped carriers on anions bound by the polaronic exchange into four degenerate singlet bipolarons $A, B, C$ and $D$ (b).](image-url)
hopping is possible only in the second order of $t$ [23]. The occurrence of superconductivity in bipolaronic strong-coupling systems is not controlled by the pairing strength, but by the phase coherence among the electron pairs [6]. While in two-dimensions Bose condensation does not occur in either the ideal or the interacting system, there is a phase transition to a superfluid state at $T_c = 2\pi n_b h^2/k_B m^{**} \ln[\ln(1/n_b r^2)]$ in the dilute Bose gas [24,25] (here $n_b$ is the boson density per unit area). Using eqs. (3), (7) we obtain $E_p \approx 0.4 E_c$ and $g^2 \approx 0.18 E_p/h \omega_0$, allowing for a quantitative estimate of $T_c$ (here $E_c \approx 2e^2c/\pi \kappa a^2$). With typical values of $a = 0.4 \text{nm}$, $c = 1.2 \text{nm}$, $\kappa = 5$, the bare band mass $m = m_e$, $\hbar \omega_0 = 80 \text{meV}$ and the moderate atomic density of polarons, $x = 0.1$ (avoiding an overlap of bipolarons) one obtains $E_p \approx 0.55 \text{eV}$, $g^2 \approx 1.24$, and $T_c \approx 205 \text{K}$. Importantly, the projection procedure of reducing eq. (12) to eq. (13) is well justified since the ratio $t/J_p \approx 0.1$ is small and $K_B T_c \ll J_p$, so that only the lowest singlet configurations can be included while discarding the others.

In conclusion, it seems very likely that a peculiar cancellation of the long-range Coulomb repulsion by the long-range Fröhlich EPI can help much in producing high-temperature superconductivity in doped polar insulators such as cuprates and other oxides, for instance BaKBO$_3$. The polaronic $t$-$J_p$ Hamiltonian, eq. (12) derived here from the bare long-range Coulomb interactions could provide a novel avenue for analytical and computational studies of superconductivity in complex ionic lattices since the repulsive Hubbard $U$ model and its strong-coupling $t$-$J$ projection do not explain high $T_c$ [26].

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The author thanks A. Bratkovsky, J. Bonča, J. Devreese, H. Fehske, V. Kabanov, D. Mihailović, P. Prelovsek, J. Samson and G. Sica for helpful discussions. This work was partially supported by the Royal Society (London).

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