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High temperature superconductivity from realistic Coulomb and Fröhlich interactions

A. S. Alexandrov1,2,∗ J. H. Samson2,† and G. Sica2,3

1 Instituto de Física “Gleb Wataghin”, Universidade Estadual de Campinas, UNICAMP 13083-970, Campinas, São Paulo, Brasil
2 Department of Physics, Loughborough University, Loughborough LE11 3TU, UK, EU
3 Dipartimento di Fisica “E.R. Caianiello”, Università degli Studi di Salerno, I-84084 Fisciano (SA), Italy, EU

In recent years ample experimental evidence has shown that charge carriers in high-temperature superconductors are strongly correlated but also coupled with lattice vibrations (phonons), signalling that the true origin of high-Tc superconductivity can only be found in a proper combination of Coulomb and electron-phonon interactions. On this basis, we propose and study a model for high-Tc superconductivity which accounts for realistic Coulomb repulsion, strong electron-phonon (Fröhlich) interaction and residual on-site (Hubbard U) correlations without any ad-hoc assumptions on their relative strength and interaction range. In the framework of this model, which exhibits a phase transition to a superconducting state with a critical temperature Tc well in excess of 100K, we emphasize the role of U as the driving parameter for a BEC/BCS crossover. Our model lays a microscopic foundation for the polaron-bipolaron theory of superconductivity. We argue that the high-Tc phenomenon originates in competing Coulomb and Fröhlich interactions beyond the conventional BCS description.

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I. INTRODUCTION

Unconventional symmetries of the order parameter allowed some researchers to maintain that a purely repulsive interaction between electrons (Hubbard U) accounts for superconductivity without phonons in a number of high-temperature superconductors [1]. However, recent analytical [2] and numerical (Monte-Carlo) [3, 4] studies shed doubts on the possibility of high temperature superconductivity from repulsive interactions only.

Also, a growing number of experimental and theoretical results suggest that strong electron correlations and significant electron-phonon interaction (EPI) are the unavoidable features for a microscopic theory of high Tc superconductivity 5 and 6. In particular, the doping-dependent oxygen-isotope effects on the critical temperature Tc of high-Tc materials as the polaron ab-
been introduced as input parameters not directly related to the material. Different from those studies an analytical multi-polaron model of high-temperature superconductivity in highly polarisable ionic lattices has been recently proposed [39] and numerically studied (for two-particle states) [40] with generic (bare) Coulomb and Fröhlich interactions avoiding any ad-hoc assumptions on their range and relative magnitude. It has been shown that the generic Hamiltonian comprising any-range Coulomb repulsion and the Fröhlich EPI can be reduced to a short-range $t-J_p$ model at very large lattice dielectric constant, $\epsilon_0 \rightarrow \infty$, for moderate and strong EPI. In this limit the bare static Coulomb repulsion and Fröhlich EPI negate each other giving rise to a novel physics described by the polaronic bare static Coulomb repulsion and EPI negate each other giving rise to a novel physics described by the polaronic

The cancellation of the bare Coulomb repulsion by the Fröhlich EPI is accurate up to $1/\epsilon_0$ corrections. At finite $\epsilon_0$ a residual on-site repulsion of polarons, $\tilde{U}$, could be substantial if the size of the Wannier (atomic) orbitals is small enough. Here we study the effect of this on-site repulsion on the ground state of the extended $t$-$J_p$-$\tilde{U}$ model accounting for all essential correlations in high-temperature superconductors. It is worth emphasizing that the effect of the on-site $\tilde{U}$ does not follows as a mere generalization of the $t$-$J_p$ model. The residual Hubbard $\tilde{U}$ in fact leads not only to the suppression of on-site pairs but also to the reduction of the exchange interaction and to the Bose-Einstein condensation (BEC) to BCS (BEC/BCS) crossover.

II. BARE HAMILTONIAN

Keeping major terms in both interactions, diagonal with respect to sites, yield our generic Hamiltonian in the site representation,

$$
H = \sum_{i,j} (T_{ij}\delta_{\sigma\sigma'} + \mu\delta_{ij})c_i^\dagger c_j + \frac{\alpha}{2\epsilon_0} \sum_{i\neq j} \tilde{n}_i\tilde{n}_j + \sum_{q,\nu,i} \hbar\omega_q\tilde{n}_i |u_i(q)d_q + H.c.] + H_{ph}.
$$

Here $T_{ij}$ is the bare hopping integral, if $\mathbf{m} \neq \mathbf{n}$, or the site energy, if $\mathbf{m} = \mathbf{n}$, $\mu$ is the chemical potential, $i = (\mathbf{m}, \sigma)$ and $j = (\mathbf{n}, \sigma')$ include both site $(\mathbf{m}, \mathbf{n})$ and spin $(\sigma, \sigma')$ quantum numbers, $c_i, d_q$ are electron and phonon operators respectively, $\tilde{n}_i = c_i^\dagger c_i$ is a site occupation operator, $\alpha = e^2/4\pi\epsilon_0 \epsilon_\infty$ (approx. $8.85 \times 10^{-12}$ F/m is the vacuum permittivity), and $H_{ph} = \sum_q \hbar\omega_q (d_q^\dagger d_q + 1/2)$ with the phonon frequency $\omega_q$.

The EPI matrix element is

$$
|u_i(q)| = (2N)^{-1/2} \gamma(q) \exp(i\mathbf{q} \cdot \mathbf{m})
$$

with the dimensionless EPI coupling, $\gamma(q)$ ($N$ is the number of unit cells). Deriving the generic Hamiltonian in the site representation [39] we approximate the Wannier or-

Quantitative calculations of the EPI matrix elements in semiconductors and metals have to be performed numerically from pseudopotentials. Fortunately one can parametrize EPI rather than compute it in many physically important cases [41]. EPI in ionic lattices such as the cuprates is dominated by coupling with polar optical phonons. This dipole interaction is much stronger than the deformation potential coupling to acoustic phonons and other multipole EPIs. While the EPI matrix elements are ill-defined in metals, they are well defined in doped insulators, which have their parent dielectric compounds with well-defined phonon frequencies $\omega_q$ and the electron band dispersion.

To parameterize EPI one can calculate the lowest order two-particle vertex function comprising the direct Coulomb repulsion and a phonon exchange [41],

$$
\Gamma(q, \Omega_n) = \frac{4\pi\alpha}{\epsilon_\infty \sqrt{\hbar q^2}} + |\gamma(q)|^2 (\hbar\omega_q)^2 D(q, \Omega_n).
$$

Here $q = k'_1 - k_1$, $\Omega_n = \omega_{n+1} - \omega_{n+1}$ are the momentum and energy transfer in a scattering process of two carriers with the initial momenta and the Matsubara frequencies $k_{1,2}$ and $\omega_{n,1,2}$, respectively, and $D(q, \Omega_n) = -(h\omega_q/\Omega_n^2 + (h\omega_q)^2)$ is the propagator of a phonon of frequency $\omega_q$, and $V_q$ is the unit cell volume. In the static limit, $\Omega_n = 0$, Eq.(3) yields the Fourier component of the particle-particle interaction as

$$
\Gamma(q, 0) = \frac{4\pi\alpha}{\epsilon_\infty \sqrt{\hbar q^2}} - |\gamma(q)|^2 h\omega_q.
$$

On the other hand, two static carriers localised on sites $\mathbf{m}$ and $\mathbf{n}$ in the ionic lattice repel each other with the Coulomb potential

$$
v_{ij} = \frac{\alpha}{\epsilon_0 |\mathbf{m} - \mathbf{n}|},
$$

where the static dielectric constant, $\epsilon_0$ accounts for the screening by both core electrons and ions. Comparing Eq.(4) and Eq.(5) we find

$$
|\gamma(q)|^2 \hbar\omega_q = \frac{\alpha}{\kappa} \sum_{\mathbf{m}} \frac{e^{i\mathbf{q} \cdot \mathbf{m}}}{m} \approx \frac{4\pi\alpha}{\sqrt{\hbar q^2}},
$$

at relatively small $q \lesssim 1/a$. Here $a$ is the lattice constant and $\kappa = \epsilon_0/\epsilon_\infty/(\epsilon_0 - \epsilon_\infty$) with the high-frequency dielectric constant $\epsilon_\infty$. The static dielectric constant $\epsilon_0$ and the high-frequency dielectric constant $\epsilon_\infty$ are readily measured by putting the parent insulator in a capacitor and as the square of the refractive index of the insulator, respectively. Hence, different from many models of
high-temperature superconductors proposed so far, our
generic Hamiltonian with the bare Coulomb and Fröhlich
interactions is defined through the measurable material
parameters.

III. \(t - J_p\) AND \(t - J_p - \bar{U}\) MODELS

Using the Lang-Firsov (LF) canonical transformation
[42] one can integrate out most of both interactions in the
transformed Hamiltonian [39],

\[
\hat{H} = -\sum_{i,j} (\bar{\sigma}_{ij} \delta_{\sigma\sigma'} + \tilde{\mu}_{ij}) \epsilon_i^c \epsilon_j^c + H_{ph} + \frac{1}{2} \sum_{i \neq j} v_{ij} n_i n_j,
\]

since the residual repulsion, \(v_{ij}\), is substantially dimin-
ished by the large dielectric constant of the polar lattice
[see Eq.(5)]. Here

\[
\bar{\sigma}_{ij} = T_{ij} \hat{X}_i \hat{X}_j
\]

is the renormalised hopping integral involving multi-
phonon transitions with \(\hat{X}_i = \exp \left[ \sum_{q} u_i(q)d_q - H.c. \right] \)
and \(\tilde{\mu}\) is the chemical potential shifted by the polaron
binding energy.

Then, using the Schrieffer-Wolf (SW) canonical transfor-
mation [43] and neglecting \(v_{ij}\), the transformed Hamil-
tonian Eq.(7) is reduced to the \(t - J_p\) Hamiltonian as [39]

\[
\mathcal{H}_{tJ_p} = -\sum_{i,j} t_{ij} \bar{\delta}_{\sigma\sigma'} \epsilon_i^c \epsilon_j^c + \frac{2}{3} \sum_{m \neq n} J_p(m - n) \left( \mathbf{S}_m \cdot \mathbf{S}_n + \frac{1}{4} n_m n_n \right).
\]

Here the sum over \(n \neq m\) counts each pair once only,
\(\mathbf{S}_n = (1/2) \sum_{\sigma,\sigma'} \epsilon_n^\sigma \sigma'^\sigma \sigma^\sigma^\sigma\sigma^\sigma\sigma\sigma\sigma\sigma\)
the spin 1/2 operator (\(\bar{T}\) are the Pauli matrices), \(n_n = n_{m^\uparrow} + n_{m^\downarrow}\), and
\(n_{m^\uparrow\downarrow} = (\epsilon_{m^\uparrow}^c \epsilon_{m^\downarrow} + \epsilon_{m^\downarrow}^c \epsilon_{m^\uparrow})\) are site occupation operators.

All quantities in the polaronic \(t-J_p\) Hamiltonian (9)
are defined through the material parameters, in par-
ticular the polaron hopping integral, \(t_{ij} = T(m - n)\exp[-g^2(m - n)]\) with the polaron band-narrowing ex-
ponent

\[
g^2(m) = \frac{2\pi e^2}{\kappa \hbar \omega_L N V_0} \sum_q \frac{1 - \cos(q \cdot m)}{q^2},
\]

and

\[
J_p(m) = T^2(m)/2g^2(m)\hbar \omega_L.
\]

It has been proposed that the \(t-J_p\) Hamiltonian,
Eq.(9), has a high-\(T_c\) superconducting ground state
protected from clustering [39]. The polaronic exchange \(J_p\)
is attractive for polarons in the singlet channel and re-
pulsive for polarons in the triplet channel. The origin of
this exchange attraction is illustrated in Fig.1.

If two polarons with opposite spins occupy nearest-
neighbour sites, they can exchange sites without any po-
tential barrier between them, which lowers their energy
by \(J_p\) proportional to the unrenormalized hopping inte-
gral squared.

Importantly the LF transformation Eq.(7) is exact, and
the SW transformation is accurate for intermediate and
strong EPI coupling, \(\lambda \geq 1/\sqrt{2}\), where \(\lambda\) is the BCS
coupling constant and \(z\) is the lattice coordination
number as discussed in details in Ref. 39. The residual
repulsion of polarons, \(v_{ij}\), in the transformed Hamil-
tonian, Eq.(7), is small compared with the exchange inter-
site polaron attraction \(J_p\) and the short-range bipolaron-
bipolaron repulsion of about the same magnitude, as long
as \(\epsilon_0 \gg \alpha/a J_p\). With the typical parameters of the
cuprates \(J_p\) is about 1 eV and \(\alpha/a \approx 4\) eV, so that
the residual inter-site repulsion \(v_{mn}\) is small if \(\epsilon_0 \gg 1\), which
will be well satisfied in all relevant compounds [44].

Nevertheless the on-site term in \(v_{mm}\), Eq.(5), \(\bar{U}\) could
be substantial, if the size of the Wannier orbitals is small
enough \(a_0 \ll a\). This renormalised \(\bar{U}\) is strongly di-
ninished by the lattice polarization with respect to the
bare on-site repulsion. We have emphasised in Refs.39 and
40 that our model describes carriers doped into the
charge-transfer Mott-Hubbard (or any polar) insulator,
rather than the insulator itself, different from the con-
ventional Hubbard \(U\) or \(t-J\) models. The bare Hubbard-
\(U\) on the oxygen orbitals (where doped holes reside) in
a rigid cuprate lattice is of the same order of magnitude as
the on-site attraction induced by the Fröhlich EPI \((\approx 1 - 2\) eV [44]), so that the residual Hubbard \(\bar{U}\) could be as
large as a few hundred meV. We now take it into account
in the energy of a virtual double occupied state \(|p\rangle\) with
two opposite spins on the same site,

\[
E_p - E_n = \bar{U} + \sum_{n_q \neq 0} \hbar \omega_q n_q.
\]

Then performing the SW transformation the exchange
attraction is found as

\[
J_p(u, m - n) = \frac{\hbar^2}{\omega_L} \sum_{k=1}^\infty \frac{(2g^2(m - n))^k}{k!(k + u)},
\]

where \(u = \bar{U}/\hbar \omega_L\). The reduction with respect to
\(J_p(0, m)\) is moderate as long as the relative \(u\) is less
than \(2g^2\), but becomes substantial for \(u > 2g^2\), Fig.2,
which puts the characteristic bipolaron binding energy
in the range of a hundred meV comparable with the double pseudogap in the cuprates [5]. Importantly \( J_p(u) \) remains large or comparable with the polaron hopping integral \( t = T(a) \exp(-g^2(a)) \) since the spin exchange of the \( t - J_p \) model , Eq.(11), does not contain the small polaron narrowing exponent \( \exp(-g^2) \).

Hence our extended \( t-J_p(u)-\tilde{U} \) model including major correlations effects reads as follows:

\[
\mathcal{H} = -\sum_{i,j} t_{ij} \delta_{\sigma\sigma'} c^\dagger_i c_j + \tilde{U} \sum_{\mathbf{m}} n_{\mathbf{m}\uparrow} n_{\mathbf{m}\downarrow} + 2 \sum_{\mathbf{m} \neq \mathbf{n}} J_p(u, \mathbf{m} - \mathbf{n}) \left( \mathbf{S}_\mathbf{m} \cdot \mathbf{S}_\mathbf{n} + \frac{1}{4} n_{\mathbf{m}\uparrow} n_{\mathbf{n}\downarrow} \right) \tag{14}
\]

IV. LOW-DENSITY LIMIT AND HIGH Tc.

As in refs 39 and 40 we adopt here the strong-coupling approach to the multi-polaron problem described by the Hamiltonian, Eq.(14), solving first a two-particle problem and then projecting the Hamiltonian on the repulsive Bose gas of small inter-site bipolarons. Such projection allows for a reliable estimate of the superconducting critical temperature for low carrier density as long as bipolarons remain small.

If we neglect the polaronic hopping taking \( t = 0 \), then the ground and the highest energy states are bipolaronic spin-singlet and spin-triplet, respectively, made up of two polarons on neighbouring sites. The zero-energy states [in the nearest-neighbour (NN) approximation] are pairs of polarons separated by more than one lattice spacing. The on-site bipolaron has energy \( \tilde{U} > 0 \).

For \( t \neq 0 \) our exact diagonalization (ED) results on finite clusters show that the probability to find NN bipolarons falls as we increase the hopping or the strength of the on-site repulsion \( \tilde{U} \) as shown in Fig.3 for a 100 \( \times \) 100 square lattice. Consistently, the bipolaron size increases but remains on the order of the lattice spacing in a wide domain of the parameters (see Fig.4). Importantly, although the small bipolaronic configuration persists for any values of the hopping at \( \tilde{U} = 0 \), for \( \tilde{U} \neq 0 \) and large values of \( t \) up to a critical value \( t_c = \tilde{U} J_p(u)/(2\tilde{U} - 8J_p(u)) \), the presence of a finite on-site interaction leads to the crossover from a small to a large bipolaronic configuration. Finally, for further increasing \( t \) the system undergoes a phase transition to an unbound state at \( t = t_c \). The crossover from a small to a large bipolaronic configuration is also confirmed by the calculation of the bipolaron to polaron effective mass ratio with \( m^{*\ast} = 2m^* \) in the large bipolaron regime, as shown in Fig.5.

In the small bipolaron regime, the kinetic energy operator in Eq.(14) connects singlet configurations in the first and higher orders with respect to the polaronic hopping.
integrals. Taking into account only the singlet bipolaron and discarding all other configurations one can map the $t-J_p(U)$ model on the square lattice for low carrier density $n_{\text{h}} = 0.05/a^2$ with $\hbar\omega_0 = 0.08$eV, $g^2 = 1.24$ and $J_p(0) = 1.81$eV.

As shown in Fig.6, for a physical choice of the parameters ($\hbar\omega_0 = 0.08$eV, $g^2 = 1.24$, $T = 0.6$eV, giving $J_p(0) = 1.81$eV from Eq. 11) the critical temperature is found to be well in excess of 100K, despite a low carrier density. Increasing $U$ decreases $J_p(u)$, as shown in Fig. 2, and $T_c$.

It is worth noting that, unlike in other theories, the strength of the on-site interaction term reduces $T_c^*$.

V. CONCLUSIONS

In conclusion, we have introduced and studied the polaronic $t-J_p(U)$ model, defined through the bare material parameters. The model, being an essential generalization of the $t-J_p$ model [39], includes all electron-electron and electron-phonon correlations providing a microscopic explanation of the high-$T_c$ phenomenon without any ad-hoc approximations. We show that the inclusion of the residual on-site interaction $\tilde{U}$ (neglected in the $t-J_p$ model [39, 40]), drives the system to a BEC/BCS crossover that reconciles the polaron-bipolaron theory of superconductivity with the observation of a large Fermi surface in overdoped cuprate superconductors. We offer an explanation, on microscopic grounds, of the high-$T_c$ phenomenon as a consequence of competing Coulomb and Fröhlich interactions in highly polarizable ionic lattices beyond the conventional BCS description.

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