Key pairing interaction in layered doped ionic insulators

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A controversial issue on whether the electron-phonon interaction (EPI) is crucial for high-temperature superconductivity or it is weak and inessential has remained one of the most challenging problems of contemporary condensed matter physics. We employ a continuum RPA approximation for the dielectric response function allowing for a selfconsistent semi-analytical evaluation of the EPI strength, electron-electron attractions, and the carrier mass renormalisation in layered high-temperature superconductors. We show that the Fröhlich EPI with high-frequency optical phonons in doped ionic lattices is the key pairing interaction, which is beyond the BCS-Migdal-Eliashberg approximation in underdoped superconductors, and it remains a significant player in overdoped compounds.

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For a long time, a basic question concerning the key pairing interaction in cuprate and other high-temperature superconductors has remained open. Some density functional (DFT) calculations\textsuperscript{[1, 2]} found small EPI insufficient to explain high critical temperatures, $T_c$, while some other first-principles studies found large EPI in cuprates\textsuperscript{[3]} and in recently discovered iron-based compounds\textsuperscript{[4]}. It is a common place that DFT underestimates the role of the Coulomb correlations, predicting an anisotropy of electron-response functions much smaller than that experimentally observed in the layered high-$T_c$ superconductors. The inclusion of a short-range repulsion (Hubbard $U$) via the LDA+$U$ algorithm\textsuperscript{[5]} and/or nonadiabatic effects\textsuperscript{[6]} significantly enhances the EPI strength due to a poor screening of some particular phonons. Also substantial isotope effects on the carrier mass and a number of other independent observations (see e.g.\textsuperscript{[7]} and references therein) unambiguously show that lattice vibrations play a significant although unconventional role in high-temperature superconductors. Overall, it seems plausible that the true origin of high-temperature superconductivity could be found in a proper combination of strong electron-electron correlations with a significant EPI\textsuperscript{[8]}.

Here, we calculate the EPI strength with optical phonons, the phonon-induced electron-electron attraction, and the carrier mass renormalisation in layered superconductors at different doping using a continuum approximation for the renormalised carrier energy spectrum and the RPA dielectric response function. The Fröhlich EPI with high-frequency optical phonons turns out the key pairing interaction in underdoped highly polarizable ionic lattices and remains significant player in overdoped compounds.

We start with a parent insulator as La$_2$CuO$_4$, where the magnitude of the Fröhlich EPI is unambiguously estimated using the static, $\epsilon_s$ and high-frequency, $\epsilon_\infty$ dielectric constants\textsuperscript{[9, 10]}. To assess its strength, one can apply an expression for the polaron binding energy (polaronic level shift) $E_p$, which depends only on the measured $\epsilon_s$ and $\epsilon_\infty$,

$$E_p = \frac{\epsilon^2}{2\epsilon_0\kappa} \int_{BZ} \frac{d^3q}{(2\pi)^3 q^2}$$

Here, the integration goes over the Brillouin zone (BZ), $\epsilon_0 \approx 8.85 \times 10^{-12}$ F/m is the vacuum permittivity, and $\kappa = \epsilon_s\epsilon_\infty/(\epsilon_s - \epsilon_\infty)$. In the parent insulator, the Fröhlich interaction alone provides the binding energy of two holes, $2E_p$, an order of magnitude larger than any magnetic interaction ($E_p = 0.647$ eV in La$_2$CuO$_4$\textsuperscript{[9]}). Actually, Eq.\textsuperscript{(1)} underestimates the polaron binding energy, since the deformation potential and/or molecular-type (e.g. Jahn-Teller\textsuperscript{[10]}) EPIs are not included.

It has been argued earlier\textsuperscript{[8]} that the interaction with $c$-axis polarized phonons in cuprates remains strong also at finite doping due to a poor screening of high-frequency electric forces as confirmed in some pump-probe\textsuperscript{[11, 12]} and photoemission\textsuperscript{[13, 14]} experiments. However, a quantitative analysis of the doping dependent EPI has remained elusive because the dynamic dielectric response function, $\epsilon(\omega, \mathbf{q})$, has been unknown.

Recent observations of the quantum magnetic oscillations in some underdoped\textsuperscript{[15]} and overdoped\textsuperscript{[16]} cuprate superconductors are opening up a possibility for a quantitative assessment of EPI in these and related doped ionic lattices with the quasi two-dimensional (2D) carrier energy spectrum. The oscillations revealed cylindrical Fermi surfaces, enhanced effective masses of carriers (ranging from $2m_e$ to $6m_e$) and the astonishingly low Fermi energy, $E_F$, which appears to be well below 40 meV in underdoped Y-Ba-Cu-O\textsuperscript{[16]} and less or about 400 meV in heavily overdoped Tl2201\textsuperscript{[17]}. Such low Fermi energies\textsuperscript{[15]} make the Migdal-Eliashberg (ME) adiabatic approach to EPI\textsuperscript{[18]} inapplicable in these compounds. Indeed, the ME non-crossing approximation breaks down at $\lambda\omega_0/E_F > 1$ when the crossing diagrams become important. The characteristic oxygen vibration energy is about $\hbar\omega_0 = 80$ meV in oxides\textsuperscript{[20, 21]}, so that the ME theory cannot be applied even for a weak EPI with the coupling constant $\lambda < 0.5$. In the strong coupling
regime, $\lambda \gtrsim 0.5$, the effective parameter $\lambda\hbar\omega_0/E_F$ becomes large irrespective of the adiabatic ratio, $\hbar\omega_0/E_F$, because the Fermi energy shrinks exponentially due to the polaron narrowing of the band \cite{22}. Since carriers in cuprates are in the non-adiabatic (underdoped) or near-adiabatic (overdoped) regimes, $E_F \lesssim \hbar\omega_0$, their energy spectrum renormalised by EPI can be found with the familiar small-polaron canonical transformation at any coupling $\lambda$ \cite{22}.

The matrix element of the screened electron-phonon (Fröhlich) interaction is found as \cite{24}:

$$
\gamma(\mathbf{q}) = \frac{\gamma_0(\mathbf{q})}{\epsilon(\omega_0, \mathbf{q})},
$$

(2)

where $\gamma_0(\mathbf{q})$ is the bare (unscreened) vertex in the parent insulator, Fig.1a. In our selfconsistent approach $\epsilon(\omega, \mathbf{q})$ is calculated in the loop (RPA) approximation \cite{23} with the exact (polaronic) carrier propagators taking into account the phonon “dressing” of carriers:

$$
\epsilon(\omega, \mathbf{q}) = 1 + \frac{2e^2}{\epsilon_0\epsilon_{\infty}q^2\Omega} \sum_k f_k f_{k+\mathbf{q}/2} - f_k f_{k-\mathbf{q}/2} h(\omega + i/\tau) - \epsilon_{\mathbf{k}+\mathbf{q}/2} + \epsilon_{\mathbf{k}-\mathbf{q}/2}.
$$

(3)

Here, $\epsilon_k$ is the polaron band dispersion, $\tau$ is the relaxation time, $\Omega$ is the normalization volume, and $f_k$ the Fermi-Dirac distribution function. The effect of collisions cannot be always taken into account merely by replacing $\omega$ by $\omega + i/\tau$, in the collision-less Lindhard dielectric function, Eq.(3), in particular at low frequencies or in the static limit \cite{20,27}, where ladder-type vertex corrections are important \cite{28}. However, in our case the relevant frequency is the (renormalised) optical phonon frequency, so that the vertex corrections are negligible as $1/\omega_0\tau \ll 1$.

Since the Fermi surfaces measured in the quantum oscillation experiments \cite{16,17} are almost perfect cylinders, one can apply the continuum (parabolic) approximation for the quasi-2D polaron energy spectrum, $\epsilon_k = h^2(k^2 + k_0^2)/2m^*$, where the polaron effective mass, $m^*$ has to be found self-consistently as a function of EPI. Calculating the sum in Eq.(3) yields the following dielectric function that is used to determine the quasi-2D energy and collisions

$$
\epsilon(\omega, \mathbf{q}) = 1 + \frac{Ne^2}{\epsilon_0\epsilon_{\infty}q^2m^*v_F} \chi_1(\omega, q) + i\chi_2(\omega, q)].
$$

(4)

Here and below $q^2 = q_x^2 + q_y^2$ is the square of the phonon momentum, $N = k_F^2/2\pi c$ is the carrier density, $a$ and $c$ are in-plane and c-axis (chemical) unit cell constants, respectively, and $v_F = \hbar k_F/m^*$ is the Fermi velocity. The real and imaginary parts of susceptibility are found as \cite{30,31}:

$$
\chi_1(\omega, q) = 2 - |R(z, z^2 - u, \beta) + R(z, z^2 + u, \beta)|/z^2, \quad \chi_2(\omega, q) = |I(z, z^2 - u, \beta) - I(z, z^2 + u, \beta)|/z^2.
$$

The polaron level shift, $E_p = V(0)/2$, the carrier attraction induced by EPI, $-V(r)$, the in-plane polaron mass and the mass renormalisation exponent, $g^2$, are found as \cite{32,33}:

$$
E_p = \frac{e^2}{2\epsilon_0\kappa(2\pi)^3} \int_{BZ} \frac{d^3q}{q^2|\epsilon(\omega_0, \mathbf{q})|^2}.
$$

(6)
and neutron, $V\beta$ phonon frequencies, $V\alpha$ respectively, where $x$ Fröhlich EPI for different doping 

Holes in cuprates reside on oxygen, so that the nearest-neighbor hopping distances is $j = a/\sqrt{2}$. The BCS coupling constant with phonons is defined as $\lambda = E_p ma^2/\pi \hbar^2$ in the case of 2D carriers with a constant density of states ($ma^2/2\pi \hbar^2$ per spin), where $m$ is the bare band mass in a rigid lattice. Using $E_p \approx 0.6eV$ and $m = 2m_e$ places cuprates in the strong-coupling regime, $\lambda \gtrsim 0.86$.

Approximating the Brillouin zone as a cylinder of the volume $2\pi^2 q^2/\epsilon$ with the Debye momentum $q_D = 2\sqrt{\pi}/a$ and integrating over azimuthal (in-plane) angle yield

$$\frac{V(r)}{4E_c} = \int_0^1 \int_0^1 \frac{dt dy J_0(\sqrt{2\pi r t}) t^5 (y^2 + \eta t^2)}{[t^2(y^2 + \eta t^2) + \zeta(t^2 - R)]^2 + \zeta'^2}$$  \hspace{1cm} (10)

where $R = R(k, t)$, $t^2/2 - \tilde{u}, k^2\tilde{\beta}) + R(k, t^2/2 + \tilde{u}, k^2\tilde{\beta})$, $\zeta = I(k, t^2/2 - \tilde{u}, k^2\tilde{\beta}) - I(k, t^2/2 + \tilde{u}, k^2\tilde{\beta})$, $E_c = e^2 c/(2\pi^2 k_0^2 a^2) \approx 0.71eV$ (with $\epsilon_\infty = 5$ and $\epsilon_s = 30$ of La$_2$CuO$_4$ [1]). The on-site, the nearest-neighbor and the next-nearest-neighbor attractions correspond to $r = 0, 1, \sqrt{2}$, respectively. $J_0(x)$ is the Bessel function, $k = k_F/d$ is the dimensionless Fermi momentum, $\tilde{u} = \omega_0 m^* a^2/4\pi \hbar \approx 0.015(m^*/m_e)$ (for $\omega_0 = 80 meV$), $\tilde{\zeta} = e^2 c m^*/(\epsilon_0 \kappa \pi^2 \hbar^2) \approx 1.23(m^*/m_e)$, and $\eta = 4c^2/\pi a^2 \approx 3.93$. If one assumes that carriers are scattered off impurities with the density equal to the carrier density, as in La$_{2-x}$Sr$_x$CuO$_4$, then in the Born approximation $\tilde{\beta} = \beta_0(m^*/m_e)^2$ with $\beta_0$, independent of the carrier mass and density. Using $k_F l \approx 20$ and $m^* = 2m_e$ as found in YBa$_2$Cu$_{3}O_{7-\delta}$ [3] yields $\beta_0 \approx 0.0125$.

We can now evaluate the doping ($x$) dependence of $V(r)$ and $m^*$ by just changing the dimensionless Fermi momentum, $k = (x/2)^{1/2}$, in Eq.(10) and solving Eqs [10,11] self-consistently with any bare band mass (we choose $m = m_e$ in our numerical calculations). At very
low doping the on-site and nearest-neighbor inter-site attractions are enormous, $V(0) \approx 1.25$ eV and $V(1) \approx 0.87$ eV, respectively, and carriers are rather heavy, $m^*/m \approx 10$. Such heavy polarons are readily localised by disorder accounting for the Mott variable range hopping, which explains conduction of lightly doped cuprates.

With doping, the attraction and the polaron mass drop, Figs. [24], respectively. However, the on-site ($r = 0$) and the inter-site ($r = 1$) attractions are well above the superexchange (magnetic) interaction $J$ (about 100 meV) in underdoped and optimally doped compounds since the non-adiabatic carriers cannot perfectly screen high-frequency electric fields. Both attractions and the mass renormalization remain also substantial at overdoping. The polaron mass, Fig. [3] agrees reasonably well with the experimental masses [16, 17]. Increasing the phonon frequency enhances the attraction and lowers the polaron mass in underdoped compounds with little effect on both quantities at overdoping, Figs. [23]. Decreasing (increasing) the band mass makes polarons lighter (heavier).

In conclusion, we have quantified the carrier-carrier attraction and mass renormalisation induced by EPI in layered doped ionic lattices. The Fröhlich EPI with high-frequency optical phonons turns out to be the key pairing interaction in underdoped cuprates and remains essential at overdoping. What is more surprising is that EPI is clearly beyond the BCS-ME approximation since its magnitude is larger or comparable with the Fermi energy and the carriers are in the non-adiabatic or near-adiabatic regimes depending on doping. Both conditions point to a crossover from the bipolaronic to polaronic superconductivity [22] with doping.

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