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 random phase approximation for the dielectric response function allowing for a self-consistent semianalytical evaluation of the EPI strength, electron-electron attractions, and the carrier mass renormalization 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 theory (DFT) calculations [1,2] found small electron-phonon interaction (EPI) insufficient to explain high critical temperatures, T_c, in the framework of the BCS-Migdal-Eliashberg (BCS-ME) approach, while some other first-principles studies found large EPI in cuprates [3] and in recently discovered iron-based compounds [4]. It is a commonplace that DFT underestimates the role of the Coulomb correlations and nonadiabatic effects, predicting an anisotropy of electron-response functions much smaller than that experimentally observed in the layered high-T_c superconductors. The adiabatic DFT calculations could not explain the optical infrared c-axis spectra and the corresponding electron-phonon coupling in the metallic state of the cuprates. On the other hand, these spectra are well described within the nonadiabatic response approach of Ref. [3]. There is a strong nonlocal polar EPI along the c axis in the cuprates together with the optical conductivity as in an ionic insulator even in the well-doped "metallic" state [3]. The inclusion of a short-range repulsion (Hubbard U) via the LDA + U algorithm [5] also significantly enhances the EPI strength due to a poor screening of some particular phonons. Substantial isotope effects on the carrier mass and a number of other independent observations (see, e.g., Ref. [6] 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.

Here, we calculate the EPI strength with optical phonons, the phonon-induced electron-electron attraction, and the carrier mass renormalization in layered superconductors at different doping using a continuum approximation for the renormalized carrier energy spectrum and the random phase approximation (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 a significant player in overdoped compounds.

We start with a parent insulator as La_2CuO_4, where the magnitude of the Fröhlich EPI is unambiguously estimated using the static, ϵ_s and high-frequency, ϵ_∞ dielectric constants [7,8]. 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 ϵ_s and ϵ_∞,

$$E_p = \frac{e^2}{2\epsilon_0 \kappa} \int_{\mathrm{BZ}} \frac{d^3q}{(2\pi)^3 q^2}. \tag{1}$$

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_2CuO_4 [8]). Actually, Eq. (1) underestimates the polaron binding energy, since the deformation potential and/or molecular-type (e.g., Jahn-Teller [9]) EPIs are not included.

It has been argued earlier [7] 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 [10,11] and photoemission [12–14] 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 [15] and overdoped [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 [15] and less or about 400 meV in heavily overdoped Tl2201 [16]. Such low Fermi energies [17] make the Migdal-Eliashberg (ME) adiabatic approach to EPI [18] inapplicable in these compounds. Indeed, the ME noncrossing approximation breaks down at $\lambda\hbar\omega_0/E_F > 1$ when the crossing diagrams become important. The characteristic oxygen vibration energy is about $\hbar\omega_0 = 80$ meV in oxides [19,20], 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 [21]. Since carriers in cuprates are in the nonadiabatic (underdoped) or near-adiabatic (overdoped) regimes, $E_F \lesssim \hbar\omega_0$, their energy spectrum renormalized by EPI can be found with the familiar small-polaron canonical transformation at *any coupling* λ [22].

The matrix element of the screened electron-phonon (Fröhlich) interaction is found as $\gamma(\omega, \mathbf{q}) = \gamma_0(q)/\epsilon(\omega, \mathbf{q})$, where $\gamma_0(q)$ is the bare (unscreened) vertex in a parent insulator [23], Fig. 1(a). In our self-consistent approach $\epsilon(\omega, \mathbf{q})$ is calculated in the loop (RPA) approximation but with the exact (polaronic) carrier propagators taking into account the phonon "dressing" of carries:

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

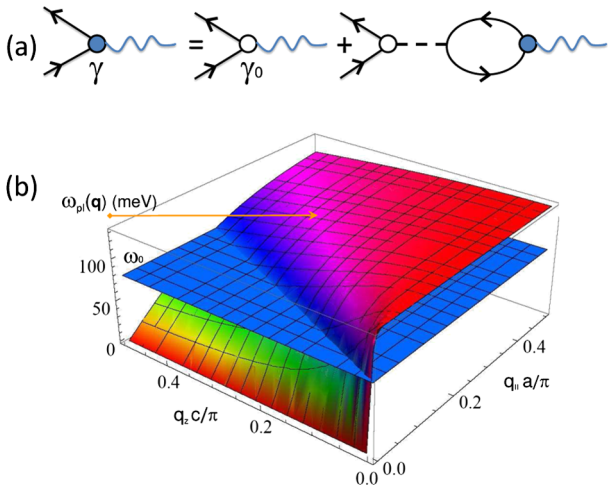

FIG. 1 (color online). Diagrammatic representation of the screened EPI vertex (a): solid lines correspond to polaron propagators, wavy lines are the exact phonon propagator, and the dashed line is the Coulomb repulsion. (b) long-wave dispersion of zeros of the dielectric function of quasi-2D carriers with the 3D Coulomb repulsion. An optical phonon with the energy 80 meV is also shown.

Here, $\epsilon_{\mathbf{k}}$ is the polaron band dispersion, τ is the relaxation time, Ω is the normalization volume, and $f_{\mathbf{k}}$ the Fermi-Dirac distribution function. The effect of collisions cannot be always taken into account merely by replacing ω by $\omega + i/\tau$, in the collisionless Lindhard dielectric function, Eq. (2), in particular, at low frequencies or in the static limit [24,25], where ladder-type vertex corrections are important [26]. In our case the relevant frequency is the (renormalized) optical phonon frequency, $\omega \approx \omega_0$, so that the vertex corrections are negligible as $1/\omega_0\tau \ll 1$. More generally, one can question the appropriateness of the RPA form of the dielectric function, Eq. (2), which neglects local field corrections, especially in correlated electron systems with strong local repulsion (Hubbard U). However, a characteristic wavelength of carriers, $2\pi/k_F$, appears to be 1 order of magnitude larger than the lattice constant, since the measured Fermi wave vector, k_F, is very small in underdoped cuprates [15]. It makes local field corrections irrelevant. At overdoping, where k_F is comparable with the BZ size [16], such corrections might be sizable, but they only enhance EPI in the optimally doped and overdoped cuprates [27].

Since the Fermi surfaces measured in the quantum oscillation experiments [15,16] are almost perfect cylinders, one can apply the continuum (parabolic) approximation for the quasi-2D polaron energy spectrum, $\epsilon_{\mathbf{k}} = \hbar^2(k_x^2 + k_y^2)/2m^*$, where the polaron effective mass, m^* has to be found self-consistently as a function of EPI. Calculating the sum in Eq. (2) yields the following dielectric response function extending the familiar pure 2D collisionless result by Stern [28] to the 3D Coulomb interaction of carriers with the quasi-2D energy spectrum and collisions,

$$\epsilon(\omega, \mathbf{q}) = 1 + \frac{Ne^2}{\epsilon_0\epsilon_\infty q^2 m^* v_F^2}[\chi_1(\omega, q_\parallel) + i\chi_2(\omega, q_\parallel)]. \quad (3)$$

Here and below $q^2 = q_z^2 + q_\parallel^2$ 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 (see also Ref. [29]) $\chi_1(\omega, q_\parallel) = 2 - [R(z, z^2 - u, \beta) + R(z, z^2 + u, \beta)]/z^2$ and $\chi_2(\omega, q_\parallel) = [I(z, z^2 - u, \beta) - I(z, z^2 + u, \beta)]/z^2$, where

$$\frac{R(z, y, \beta)}{\mathrm{sgn}(y)} = \left[\frac{y^2 - z^2 - \beta^2 + \sqrt{(y^2 - z^2 - \beta^2)^2 + 4\beta^2 y^2}}{2}\right]^{1/2}, \quad (4)$$

$$I(z, y, \beta) = \left[\frac{z^2 + \beta^2 - y^2 + \sqrt{(z^2 + \beta^2 - y^2)^2 + 4\beta^2 y^2}}{2}\right]^{1/2}, \quad (5)$$

$z = q_\parallel/2k_F$, $u = \omega/(2k_F v_F)$ and $\beta = 1/(2k_F l)$ ($l = v_F\tau$ is the mean-free path).

As shown in Fig. 1(b), a collective mode, $\omega_{\rm pl}(\mathbf{q})$, defined by $\epsilon(\omega_{\rm pl}, \mathbf{q}) = 0$ appears within the same frequency range as the optical phonon mode, ω_0, facilitating the plasmon-phonon mixing (so-called plasphons [30]). Taking additionally a weak dispersion of the quasiparticles along the c axis into account, i.e., the real three-dimensionality of the material, leads to a strongly enhanced nonadiabatic phonon-plasmon coupling along the c axis [3]. In the long-wave limit we find using Eq. (3), $\omega_{\rm pl}(\mathbf{q}) \approx \omega_p q_\parallel/q$ for collisionless carriers ($\beta = 0$), where $\hbar\omega_p = (e^2E_F/4\pi\epsilon_0\epsilon_\infty c)^{1/2}$ is approximately 132 meV for $E_F = 40$ meV. This mode is softer when it propagates *across* the planes than *along* the planes, Fig. 1(b), due to a low susceptibility of quasi-2D carriers to the electric field applied across the planes.

The polaron level shift, $E_p = V(0)/2$, the carrier attraction induced by EPI, $-V(\mathbf{r})$, the in-plane polaron mass and the mass renormalization exponent, g^2, are found as [31]

$$V(\mathbf{r}) = \frac{e^2}{\epsilon_0\kappa(2\pi)^3}\int_{\rm BZ}\frac{d^3q\cos(\mathbf{r}\cdot\mathbf{q})}{q^2|\epsilon(\omega_0,\mathbf{q})|^2}, \quad (6)$$

$$m^* = m\exp(g^2), \quad (7)$$

and

$$g^2 = \frac{E_p - V(\vec{j})/2}{\hbar\omega_0}, \quad (8)$$

respectively, where $\vec{j}$ connects nearest-neighbor sites. Holes in cuprates reside on oxygen, so that the nearest-neighbor hopping distance 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 $E_p \gtrsim 0.6$ eV 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 $2\pi^2 q_d^2/c$ with the Debye momentum $q_D = 2\sqrt{\pi}/a$ and integrating over azimuthal (in-plane) angle yield

$$\frac{V(r)}{4E_c} = \int_0^1\int_0^1 \frac{dtdyJ_0(\sqrt{2\pi}rt)t^5(y^2+\eta t^2)}{[t^2(y^2+\eta t^2)+\zeta(t^2-\Re)]^2+\zeta^2\Im^2}, \quad (9)$$

where $\Re = R(kt, t^2/2-\tilde{u}, k^2\tilde{\beta}) + R(kt, t^2/2+\tilde{u}, k^2\tilde{\beta})$, $\Im = I(kt, t^2/2-\tilde{u}, k^2\tilde{\beta}) - I(kt, t^2/2+\tilde{u}, k^2\tilde{\beta})$, $E_c = e^2c/(2\pi^2\epsilon_0\kappa a^2) \approx 0.71$ eV (with $\epsilon_\infty = 5$ and $\epsilon_s = 30$ of La_2CuO_4 [8]). 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, $k = k_F/q_D$ is the dimensionless Fermi momentum, $\tilde{u} = \omega_0 m^* a^2/4\pi\hbar \approx 0.015(m^*/m_e)$ (for $\hbar\omega_0 = 80$ meV), $\zeta = e^2cm^*/(\epsilon_0\kappa\pi^3\hbar^2) \approx 1.23(m^*/m_e)$, 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 $La_{2-x}Sr_xCuO_4$, then in the Born approximation $\tilde{\beta} = \beta_0(m^*/m_e)^2$ with β_0 independent of the carrier mass and density. Using $k_F l \approx 20$ and $m^* = 2m_e$ as found in $YBa_2Cu_3O_{6.5}$ [15] 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. (9) and solving Eqs. (9), (7), and (8) 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 intersite 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 localized 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. 2 and 3, respectively. However, the on site ($r = 0$) and the intersite ($r = 1$) attractions are well above the superexchange (magnetic) interaction J (about 100 meV) in underdoped and optimally doped compounds since the nonadiabatic carriers cannot perfectly screen high-frequency electric fields. Both attractions and the mass renormalization

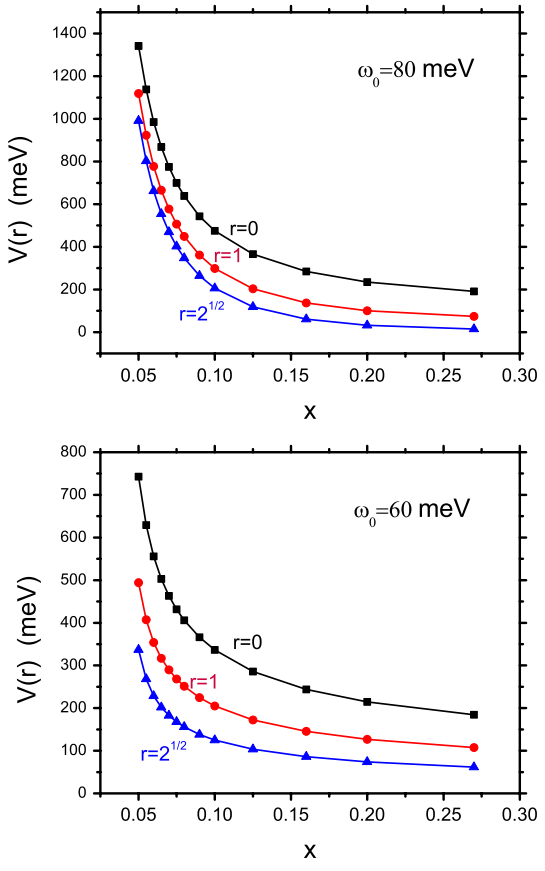

FIG. 2 (color online). The on site, $V(0)$ (upper curve), the nearest-neighbor, $V(1)$ (middle curve), and the next-nearest-neighbor, $V(\sqrt{2})$ (lower curve), attractions induced by the Fröhlich EPI for different doping x and two characteristic phonon frequencies, $\hbar\omega_0 = 80$ meV and $\hbar\omega_0 = 60$ meV.

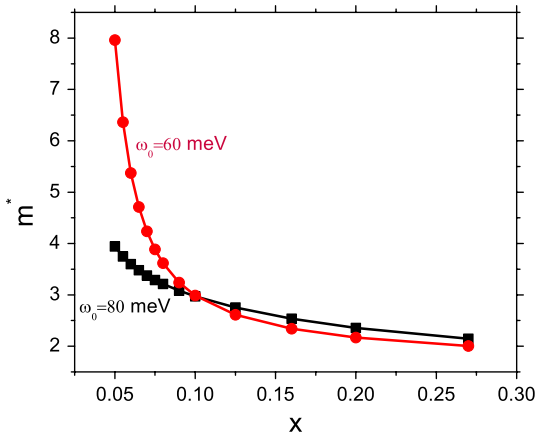

FIG. 3 (color online). The carrier mass (in units of the band mass $m = m_e$) as a function of doping for two characteristic phonon frequencies, $\hbar\omega_0 = 80$ meV and $\hbar\omega_0 = 60$ meV.

remain also substantial at overdoping. The polaron mass, Fig. 3, agrees reasonably well with the experimental masses [15,16]. Increasing the phonon frequency enhances the attraction and lowers the polaron mass in underdoped compounds with little effect on both quantities at overdoping, Figs. 2 and 3. Decreasing (increasing) the band mass makes polarons lighter (heavier).

In conclusion, we have quantified the carrier-carrier attraction and mass renormalization 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 nonadiabatic or near-adiabatic regimes. Together with the deformation potential and Jahn-Teller EPIs, the Fröhlich EPI overcomes the direct Coulomb repulsion at distances compared with the lattice constant even without any retardation [21]. Since EPI is not local in the nonadibatic electron system with poor screening, it can provide the *d*-wave symmetry of the pairing state [32]. All these conditions point to a crossover from the bipolaronic to polaronic superconductivity [21] in the cuprates with doping.

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