Abstract

It has gone almost unquestioned that superexchange in the $t - J$ (or Hubbard) model, and not phonons, is responsible for the unconventional ("d-wave") pairing symmetry of cuprate superconductors. However a number of advanced numerical studies have not found superconductivity in the Hubbard (or $t - J$) model. On the other hand compelling experimental evidence for a strong electron-phonon interaction (EPI) has currently arrived. Here I briefly review some phonon-mediated unconventional pairing mechanisms. In particular the anisotropy of sound velocity makes the phonon-mediated attraction of electrons non-local in space providing unconventional Cooper pairs with a nonzero orbital momentum already in the framework of the conventional BCS theory with weak EPI. In the opposite limit of strong EPI rotational symmetry breaking appears as a result of a reduced Coulomb repulsion between unconventional bipolarons. Using the variational Monte-Carlo method we have found that a relatively weak finite-range EPI induces a d-wave BCS state also in doped Mott-Hubbard insulators or strongly-correlated metals. These results tell us that poorly screened EPI with conventional phonons is responsible for the unconventional pairing in cuprate superconductors.

Keywords: electron-phonon interaction, sound speed anisotropy, pairing symmetry, bipolarons, cuprates

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I. INTRODUCTION

It has been thought for a long while that Cooper pairs in the Bardeen-Cooper-Schrieffer (BCS) theory [1] with the conventional electron-(acoustic) phonon interaction are singlets and their wave function is isotropic (s-wave). This interaction has been thought to be local in space, so it could not lead to a higher angular momentum pairing. The pairing symmetry breaking is a many-body effect in accordance with a well-known quantum mechanics theorem, which states that the coordinate wave function of two particles does not become zero (or has no nodes) in the ground state [2]. Hence any superconductor should seem to be s-wave in the strong-coupling limit, where pairs are individual (e.g. bipolarons [3, 4]) rather than overlapping Cooper pairs.

Recently I have revised the symmetry of the superconducting state mediated by conventional acoustic phonons [5]. The sound speed anisotropy leads to a non-local attraction between carriers and unconventional Cooper pairs in the BCS layered superconductors in a wide range of carrier densities. The Bose-Einstein condensate (BEC) can also break the rotational symmetry due to a reduced Coulomb repulsion between unconventional small bipolarons bound by strong EPI.

Earlier we have proposed that a strong departure of the cuprate superconductors from conventional BCS/ Fermi-liquids originates in the Fröhlich EPI of the order of 1 eV [3, 6, 7], routinely neglected in the Hubbard $U$ and $t-J$ models of cuprate superconductors [8]. This interaction with $c$–axis polarized phonons is virtually unscreened because the upper limit for the out-of-plane plasmon frequency ($\lesssim 200 \text{ cm}^{-1}$ [9]) in cuprates is well below the characteristic frequency of optical phonons, $\omega_0 \approx 400 - 1000 \text{ cm}^{-1}$. Since screening is poor, the magnetic interaction remains small compared with the Fröhlich EPI at any doping of cuprates (see also Ref.[10]). Taking into account that the direct Coulomb repulsion is of the same order as the Fröhlich EPI, we have proposed a so-called Coulomb-Fröhlich model (CFM) of cuprate superconductors with the ground state in the form of mobile small bipolarons or polaronic Cooper pairs (depending on doping) [6, 11, 12], which can condense at high temperatures [13]. More recently we have shown that even a weak long-range EPI combined with the Hubbard $U$ provides sizable superconducting order in doped Mott-Hubbard insulators and/or strongly-correlated metals [14].

Now compelling experimental evidence for a strong EPI has arrived from isotope effects
high resolution angle resolved photoemission spectroscopies (ARPES), a number of optical, neutron-scattering and some other spectroscopies of cuprates, in particular from recent pump-probe experiments. These experimental observations and our theoretical findings tell us that EPI with conventional phonons is responsible for the unconventional pairing in cuprate superconductors.

II. UNCONVENTIONAL COOPER PAIRS GLUED BY ACOUSTIC PHONONS

It has gone unquestioned that the unconventional pairing requires unconventional electron-phonon interactions with specific optical phonons, sometimes combined with anti-ferromagnetic fluctuations or vertex corrections, or non-phononic mechanisms of pairing (e.g. superexchange), and a specific shape of the Fermi surface. Here I show that even conventional acoustic phonons can bind carriers into unconventional Cooper pairs due to the sound-speed anisotropy in layered crystals.

In the framework of the BCS theory the symmetry of the order parameter \( \Delta(k) \) and the critical temperature, \( T_c \), are found by solving the linearised "master" equation,

\[
\Delta(k) = -\sum_{k'} V(k, k') \frac{\Delta(k')}{2\xi_{k'}} \tanh \left( \frac{\xi_{k'}}{2k_B T_c} \right).
\]

The interaction \( V(k, k') \) comprises the attraction, \(-V_{ph}(q)\), mediated by phonons, and the Coulomb repulsion, \( V_c(q) \) as \( V(k, k') = -V_{ph}(q)\Theta(|\omega_D - |\xi_k|)\Theta(|\omega_p - |\xi_{k'}|) + V_c(q)\Theta(|\omega_p - |\xi_k|)\Theta(|\omega_p - |\xi_{k'}|) \), where \( V_{ph}(q) = C^2/NC\omega_D^2 \) is the square of the matrix element of the deformation potential, divided by the square of the acoustic phonon frequency, \( \omega_q = c_q q \), \( c_q \) is the sound speed, \( M \) is the ion mass, \( N \) is the number of unit cells in the crystal, and \( \xi_k \) is the electron energy relative to the Fermi energy. The magnitude of \( C \) is roughly the electron bandwidth in rigid metallic or semiconducting lattices. The electron momentum transfer \( q = k - k' \) or its in-plane component has the magnitude \( q = 21/2k_F|1 - \cos \psi|^{1/2} \) for the spherical or cylindrical Fermi surface, respectively, where \( \psi \) is the angle between \( k \) and \( k' \), and \( \hbar k_F \) is the Fermi momentum. Theta functions account for a difference in frequency scales of the electron-phonon interaction, \( \omega_D \), and the Coulomb repulsion, \( \omega_p \gg \omega_D \), where \( \omega_D \) and \( \omega_p \) are the maximum phonon and plasmon energies, respectively.

If one neglects anisotropic effects, replacing \( V_{ph}(q) \) and \( V_c(q) \) by their Fermi-surface averages, \( V_{ph}(q) \Rightarrow V_{ph}, V_c(q) \Rightarrow V_c \), then there is only an s-wave solution of Eq. (1),
\[ \Delta_s, \text{ independent of } k. \] The sound speed anisotropy actually changes the symmetry of the BCS state. While \( c_\parallel \) is a constant in the isotropic medium, it depends on the direction of \( q \) in any crystal. The anisotropy is particularlly large in layered crystals like cuprate superconductors. As an example, the measured velocity of longitudinal ultrasonic waves along \( a-b \) plane, \( c_\parallel = 4370 \text{ ms}^{-1} \) is almost twice larger than that along \( c \) axis, \( c_\perp = 2670 \text{ ms}^{-1} \) in Bi$_2$Sr$_2$CaCu$_2$O$_{8+y}$ [29]. It makes \( V_{ph}(q) \) anisotropic,

\[
V_{ph}(q) = \frac{C^2}{NMc_\perp^2(1 + \alpha q_\parallel^2/q^2)}, \quad (2)
\]

where \( \alpha = (c_\parallel^2 - c_\perp^2)/c_\perp^2 \) is the anisotropy coefficient, which is about 2 in cuprates. The corresponding real-space potential is non-local,

\[
V(r) = -V_{ph}\Omega\left[\frac{\delta(r)}{d} + \frac{\alpha}{4\pi(1 + \alpha)^{1/2}r^3}\right], \quad (3)
\]

falling as \( 1/r^3 \) at the distance \( r \gg d \) between two carriers in the plane, where \( V_{ph} = C^2/Mc_\perp^2 \). Also the Coulomb repulsion is \( q \) dependent, \( V_c(q) = 4\pi e^2/V\epsilon_0(q^2 + q_s^2) \). In the framework of the random phase approximation the inverse screening radius squared is found as \( q_s^2 = 8\pi e^2N(0)/V\epsilon_0 \) with the density of states (per spin), \( N(0) \), at the Fermi surface. Here \( d \) is the inter-layer distance and \( \epsilon_0 \) is the (in-plane) static dielectric constant of the host cuprate lattice of the volume \( V \).

Solving the master equation (1) with 2D electron spectrum one can expand \( \Delta(k) = \sum_m \Delta_m \exp(i m \phi) \) and \( V_{ph,c}(q) = \sum_m V_{ph,c}(q_\perp, m) \exp[i m (\phi - \phi')] \) in series of the eigenfunctions of the \( c \)-axis component of the orbital angular momentum, where \( \phi \) and \( \phi' \) are polar angles of the in-plane momenta, \( k_\parallel \) and \( k'_\parallel \), respectively. The critical temperature of an \( m \)-pairing channel \( (m = 0, \pm 1, \pm 2, ...) \) is found as

\[
T_{cm} = 1.14\omega_D \exp\left[-1/(\lambda_m - \mu_m^*)\right], \quad (4)
\]

where \( \mu_m^* = \mu_m/[1 + \mu_m \ln(\omega_p/\omega_D)] \). Here \( \lambda_m \) and \( \mu_m \) are the phonon-mediated attraction and the Coulomb pseudopotential in the \( m \)-pairing channel, given respectively by

\[
\frac{\lambda_m}{\lambda} = \delta_{m,0} + \frac{\alpha}{2\sqrt{\gamma}} \int_0^{\gamma} dx\left[ x + 1 - \sqrt{x(x+2)} \right]^m, \quad (5)
\]

and

\[
\frac{\mu_m}{\mu_c} = \frac{\sqrt{\gamma}}{2} \int_0^{\gamma} dx\left[ x + \beta + 1 - \sqrt{(x+\beta)(x+\beta+2)} \right]^m, \quad (6)
\]
where \( \lambda = N(0)C^2/NMc_1^2 \), \( \gamma = \pi^2/2d^2k_F^2(1 + \alpha) \), \( \bar{\gamma} = \gamma(1 + \alpha) \), \( \mu_c = 4e^2d^2N(0)/\pi V\epsilon_0 \), and \( \beta = q_s^2/2k_F^2 \) (note that \( \lambda, \mu_c, \) and \( q_s \) do not depend on the carrier density since \( N(0) \) is roughly constant in the quasi-two dimensional Fermi gas).

The effective attraction of two electrons in the Cooper pair with non-zero orbital momentum turns out finite at any finite anisotropy, \( \alpha \neq 0 \), but numerically smaller than in the \( s \)-channel, as seen from its analytical expression for \( s \)-wave, \( m = 0 \), \( p \)-wave, \( m = 1 \), \( d \)-wave, \( m = 2 \), and higher orbital momentum pair states, obtained by integrating in Eq.(5). The Coulomb repulsion turns out much smaller in the unconventional pairing states than in the conventional \( s \)-wave state, which is seen from the analytical expression for \( \mu_m \), Eq.(6) and from Fig.1.

Using the simplest parabolic approximation for the 2D-electron energy spectrum we can draw some conclusions on the carrier-density evolution of the order-parameter symmetry. Within this approximation, \( k_F^2 = 2\pi n \) and \( N(0) = m^*V/2\pi dh^2 \), where \( n = 2x/\Omega \) is the carrier density, \( m^* \) is the effective mass, and \( x \) is the doping level as in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) with the unit cell volume \( \Omega \). The ratio of the parameters \( \beta = m^*e^2\Omega/2\pi h^2 d^2\epsilon_0 x \) and \( \bar{\gamma} = \pi\Omega/8d^3x \approx 0.044/x \) is independent of the carrier density, \( \beta/\bar{\gamma} = 4m^*e^2d/\pi^2 h^2 \epsilon_0 \), which is approximately 5 for the values of \( m^* = 4m_e \) and \( \epsilon_0 = 10 \). Fixing the value of the EPI constant at \( \lambda = \mu_c/12 \) (which corresponds to the weak-coupling BCS regime with \( \lambda \approx 0.1 \) since \( \mu_c \) is of the order of 1) and taking \( \mu_c \ln(\omega_p/\omega_D) = 3 \), we draw the anisotropy-doping phase diagram, Fig.2, with the critical lines for \( s \), \( p \) and \( d \) order parameters, defined by \( \lambda_m = \mu_m^* \). The state with the lowest magnitude of the anisotropy, \( \alpha/(1 + \alpha)^{1/2} \), is physically realized since it has the highest \( T_c \). At substantial doping the screening length becomes larger than the typical wavelength of electrons, \( \beta \to 0 \), so that the \( s \)-wave state is the ground state at a large number of carriers per unit cell for any anisotropy. On the contrary, the Coulomb repulsion is reduced to the local interaction at a low doping, \( \beta \to \infty \), and \( d \)-wave Cooper pairs are the ground state even at very low value of the anisotropy, Fig.2. Interestingly, \( s \)- and \( d \)-states turn out degenerate at some intermediate value of doping, \( x = x_c \). Hence there is a quantum phase transition with increasing doping from \( d \) to \( s \)-superconducting state, if \( \alpha > \alpha_c \), and from \( d \) to the normal state and then to the \( s \)-wave superconductor, if \( \alpha < \alpha_c \), Fig.2.
III. BREAKDOWN OF ROTATIONAL SYMMETRY IN THE STRONG-COUPLING LIMIT

In the strong-coupling regime, $\lambda \gtrsim 1$, the pairing is individual, in contrast with the collective Cooper pairing. While BEC of individual bipolarons can break the symmetry on a discreet lattice, I have proposed a symmetry breaking mechanism, which works even in a continuum model, where the ground state, it would seem, be $s$-wave to satisfy the theorem. The unscreened Fröhlich EPI in layered ionic lattices like cuprates has been suggested by us as the key for pairing. Acting alone it cannot overcome the direct Coulomb repulsion, but almost nullifies it since $\epsilon_0 \gg 1$. That allows the weaker deformation potential, Eq. (2), to bind carriers into real-space bipolarons, if $\lambda \geq 0.5$. While its local part is negated by the strong on-site repulsion $U$, the non-local tail provides bound pairs of different symmetries with the binding energies $\Delta_s > \Delta_p > \Delta_d > ...$ in agreement with the theorem. However, there is the residual Coulomb repulsion between bipolarons, $v_c(R)$. Since bipolarons have a finite extension, $\xi$, there are corrections to the Coulomb law. The bipolaron has no dipole moment, hence the most important correction at large distances between two bipolarons, $R \gg \xi$, comes from the charge-quadrupole interaction,

$$v_c(R) = 4e^2 \frac{1 \pm \eta \xi^2/R^2}{\epsilon_0 R},$$

where $\eta$ is a number of the order of 1, and plus/minus signs correspond to bipolarons in the same or different planes, respectively. The dielectric screening, $\epsilon_0$ is highly anisotropic in cuprates, where the in-plane dielectric constant, $\epsilon_{0||}$, is much larger then the out-of-plane one, $\epsilon_{0\perp}$.[32]. Hence the inter-plane repulsion provides the major contribution to the condensation energy. Since $\xi^2 \propto 1/\Delta$, the repulsion of unconventional bipolarons with smaller binding energies, $\Delta_d, \Delta_p < \Delta_s$, is reduced compared with the repulsion of $s$-wave bipolarons. As a result, with increasing carrier density we anticipate a transition from BEC of $s$-wave bipolarons to BEC of more extended $p-$ and $d$-wave real-space pairs in the strong-coupling limit.

IV. CONCLUSIONS

Several authors have remarked that superexchange, and not phonons is responsible for the symmetry breaking in unconventional superconductors like doped cuprates. Here I
arrive at the opposite conclusion. Indeed, superexchange interaction, \( J \), is proportional to the electron hopping integral, \( t \), squared divided by the on-site Coulomb repulsion (Hubbard \( U \)), \( J = 4t^2/U \), estimated as \( J \approx 0.15 \) eV in cuprates. This should be compared with the acoustic-phonon pairing interaction, \( V_{ph} \), which is roughly the Fermi energy, \( V_{ph} \approx E_F \approx 4t \) in a metal, or the bandwidth squared divided by the ion–ion interaction energy of the order of the nearest-neighbour Coulomb repulsion, \( MC^2_m \approx V_c \) in a doped insulator. The small ratio of two interactions, \( J/V_{ph} \approx t/U \ll 1 \), or \( J/V_{ph} \approx V_c/U \ll 1 \) and the giant sound-speed anisotropy favor conventional EPI as the origin of the unconventional pairing both in underdoped cuprates, where the pairing is individual, and also in overdoped samples apparently with polaronic Cooper pairs, which can coexist with bipolarons.

Moreover recent studies by Aimi and Imada of the Hubbard model, using a sign-problem-free variational Monte Carlo (VMC) algorithm, have shown that previous approximations overestimated the normal state energy and therefore overestimated the condensation energy by several orders of magnitude, so that the Hubbard model does not account for high-temperature superconductivity. This remarkable result is in line with earlier numerical studies using the auxiliary-field quantum (AFQMC) and constrained-path (CPMC) Monte-Carlo methods, none of which found superconductivity in the Hubbard model. On the other hand using VMC method we have found that even a relatively weak finite-range EPI induces the d-wave superconducting state of doped Mott-Hubbard insulators and/or strongly-correlated metals with a sufficient condensation energy.

I conclude that the finite-range electron-phonon interaction is the key to the high and a higher temperature superconductivity.

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FIG. 1: The Coulomb repulsion, $\mu_m$, as a function of the ratio of the electron wavelength to the screening length squared ($\beta = q^2_S/2k_F^2$), and the electron-phonon coupling constant, $\lambda_m$, as a function of the ratio of the electron wavelength to the inter-plane distance squared, $\gamma = \pi^2/2d^2k_F^2(1+\alpha)$ for $\alpha = 4$ (inset) in $s, p$ and $d$ pairing channels. Here $\mu'_m = \mu_c\tilde{\gamma}$. (Reprinted with permission from Ref. [5]. ©2008 by the American Physical Society.)
FIG. 2: Critical sound-speed anisotropy, $\alpha/(1+\alpha)^{1/2} = (c_\parallel^2 - c_\perp^2)/c_\parallel c_\perp$, as a function of doping, $x$, for $\lambda = \mu_c/12$ (solid lines correspond to $d$ and $s$ states, and dashed line to $p$-state). With increasing carrier density there is a quantum phase transition at $x = x_c$ from a d-wave to an s-wave superconductor, when $\alpha > \alpha_c$, and two quantum phase transitions from $d$-wave to the normal state and from the normal state to the $s$-wave state when $\alpha < \alpha_c$. (Reprinted with permission from Ref. [5]. ©2008 by the American Physical Society.)