Unconventional pairing symmetry of layered superconductors caused by acoustic phonons

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An inevitable anisotropy of sound velocity in crystals makes the phonon-mediated attraction of electrons nonlocal in space providing unconventional Cooper pairs with a nonzero orbital momentum. As a result of this anisotropy, quasi-two-dimensional charge carriers weakly coupled with acoustic phonons undergo a quantum phase transition from a conventional *s*-wave to an unconventional *d*-wave superconducting state with less carriers per unit cell. In the opposite strong-coupling regime, rotational symmetry breaking appears as a result of a reduced Coulomb repulsion between unconventional bipolarons dismissing thereby some constraints on unconventional pairing in the Bose-Einstein condensation limit. The conventional acoustic phonons, and not superexchange, are shown to be responsible for the *d*-wave symmetry of cuprate superconductors, where the on-site Coulomb repulsion is large.

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A great number of observations, in particular, phase-sensitive experiments,[1] point to the unconventional *d*-wave symmetry of cuprate and some other superconductors (for a review, see Ref. 2). It has been thought for a long while that Cooper pairs in the Bardeen-Cooper-Schrieffer (BCS) theory with the conventional electron-phonon interaction (EPI) are singlets and their wave function is isotropic (*s* wave).[3] This interaction has been thought to be local in space, so it could not lead to a higher angular-momentum pairing. Thus, it has gone unquestioned that the unconventional pairing requires unconventional electron-phonon interactions with specific optical phonons and poor screening,[4–8] sometimes combined with antiferromagnetic fluctuations[9] and vertex corrections,[10] or nonphononic mechanisms of pairing (e.g., superexchange[11]), and a specific shape of the Fermi surface.

The pairing symmetry breaking is a many-body effect in accordance with a well-known quantum mechanics theorem,[12] which states that the coordinate wave function of two particles does not become zero (or has no nodes) in the ground state. Hence, any superconductor should seem to be *s* wave in the strong-coupling limit,[13] where pairs are individual (e.g., bipolarons[14]) rather than overlapping Cooper pairs.

Here, the symmetry of the superconducting state mediated by conventional acoustic phonons is revised. The sound-speed anisotropy leads to a double surprise: (a) the BCS state of layered crystals is *d* wave in a wide range of carrier densities; (b) the strong-coupling BEC state can break the rotational symmetry as well. The anisotropic EPI with acoustic phonons is proposed as the origin of the unconventional pairing owing to a giant sound-speed anisotropy in layered cuprate superconductors.

In the framework of the BCS theory, the symmetry of the order parameter $\Delta(\mathbf{k})$ and the critical temperature T_c are found by solving the linearized "master" equation,[3]

$$\Delta(\mathbf{k}) = -\sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')\frac{\Delta(\mathbf{k}')}{2\xi_{\mathbf{k}'}}\tanh\left(\frac{\xi_{\mathbf{k}'}}{2k_BT_c}\right). \quad (1)$$

The interaction $V(\mathbf{k},\mathbf{k}')$ comprises the attraction $-V_{ph}(\mathbf{q})$, mediated by phonons, and the Coulomb repulsion $V_c(\mathbf{q})$ as,

$$V(\mathbf{k},\mathbf{k}') = -V_{ph}(\mathbf{q})\Theta(\omega_D - |\xi_{\mathbf{k}}|)\Theta(\omega_D - |\xi_{\mathbf{k}'}|) + V_c(\mathbf{q})\Theta(\omega_p - |\xi_{\mathbf{k}}|)\Theta(\omega_p - |\xi_{\mathbf{k}'}|), \quad (2)$$

where $V_{ph}(\mathbf{q}) = C^2/NMc_l^2$ is the square of the matrix element of the electron-phonon interaction,[15] divided by the square of the acoustic-phonon frequency, $\omega_{\mathbf{q}} = c_l q$, c_l is sound velocity, M is the ion mass, N is the number of unit cells in the crystal, and $\xi_{\mathbf{k}}$ is the electron energy relative to the Fermi energy. The deformation potential matrix element C is nearly q independent near the Γ point of the Brillouin zone in conventional metals[15] and near extremum points of valence and conduction bands in doped semiconductors.[16] While the validity of this approximation for cuprate superconductors has never been discussed, it affects none of our qualitative conclusions. The magnitude of C is roughly the electron bandwidth in rigid metallic[15] or semiconducting[16] lattices. The electron momentum transfer $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ or its in-plane component has the magnitude $q = 2^{1/2}k_F[1-\cos\psi]^{1/2}$ for the spherical or cylindrical Fermi surface, respectively, where ψ is the angle between $\mathbf{k}$ and $\mathbf{k}'$ and $\hbar k_F$ is the Fermi momentum. Theta functions in Eq. (2) [$\Theta(x)=1$ for positive x and zero otherwise] account for a difference in frequency scales of the electron-phonon interaction ω_D and the Coulomb repulsion $\omega_p \gg \omega_D$, where ω_D and ω_p are the maximum phonon and plasmon energies, respectively.

If one neglects anisotropic effects,[3] replacing $V_{ph}(\mathbf{q})$ and $V_c(\mathbf{q})$ by their Fermi-surface averages, $V_{ph}(\mathbf{q}) \Rightarrow V_{ph}$, $V_c(\mathbf{q}) \Rightarrow V_c$, then there is only an *s*-wave solution of Eq. (1), Δ_s, independent of $\mathbf{k}$. The sound-speed anisotropy actually changes the symmetry of the BCS state. While c_l is a constant in the isotropic medium, it depends on the direction of $\mathbf{q}$ in any crystal. The anisotropy is particulary large in layered crystals such as cuprate superconductors, where an elastic stiffness constant in the *a*-*b* plane is substantially greater than in the *c* direction (see Refs. 17 and 18 and references therein). As an example, the measured velocity of longitudinal ultrasonic waves along *a*-*b* plane, $c_{\parallel} = 4370\ \mathrm{ms}^{-1}$, is almost twice larger than that along *c* axis, $c_{\perp} = 2670\ \mathrm{ms}^{-1}$, in $Bi_2Sr_2CaCu_2O_{8+y}$.[18] It makes $V_{ph}(\mathbf{q})$ anisotropic,

$$V_{ph}(\mathbf{q}) = \frac{C^2}{NMc_\perp^2(1+\alpha q_\parallel^2/q^2)}, \quad (3)$$

where $\alpha=(c_\parallel^2-c_\perp^2)/c_\perp^2$ is the anisotropy coefficient, which is about 2 in cuprates. Also, the Coulomb repulsion is q dependent, $V_c(\mathbf{q})=4\pi e^2/V\epsilon_0(q^2+q_s^2)$. 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, ϵ_0 is the (*in-plane*) static dielectric constant of the host cuprate lattice of the volume V.

Solving the master equation [Eq. (1)] with two-dimensional (2D) electron spectrum, one can expand $\Delta(\mathbf{k})=\Sigma_m\Delta_m\exp(im\,\phi)$ and $V_{ph,c}(\mathbf{q})=\Sigma_m V_{ph,c}(q_\perp,m)\exp[im(\phi-\phi')]$ in series of the eigenfunctions of the c-axis component of the orbital angular momentum, where ϕ and ϕ' are polar angles of the in-plane momenta, $\mathbf{k}_\parallel$ and $\mathbf{k}'_\parallel$, respectively.

The solution for the m component of the order parameter $(m=0,\pm1,\pm2,\ldots)$ is found in the form $\Delta_m=\Delta_m^{(1)}\Theta(\omega_D-|\xi_\mathbf{k}|)+\Delta_m^{(2)}\Theta(\omega_p-|\xi_\mathbf{k}|)\Theta(|\xi_\mathbf{k}|-\omega_D)$ with different values of $\Delta_m^{(1)}$ and $\Delta_m^{(2)}$ below and above the cutoff energy ω_D, respectively. Integrating in Eq. (1) over $\xi_{\mathbf{k}'}$, ϕ' (using the integral $\int_0^{2\pi}d\psi\cos(m\psi)/(1-p\cos\psi)=2\pi[1-(1-p^2)^{1/2}]^m/p^m(1-p^2)^{1/2}$), and finally over $q_\perp$ yields the following pair of equations:

$$\Delta_m^{(1)}\left[1-(\lambda_m-\mu_m)\ln\frac{1.14\omega_D}{k_BT_c}\right]+\Delta_m^{(2)}\mu_m\ln\frac{\omega_p}{\omega_D}=0, \quad (4)$$

$$\Delta_m^{(2)}\left[1+\mu_m\ln\frac{\omega_p}{\omega_D}\right]+\Delta_m^{(1)}\mu_m\ln\frac{1.14\omega_D}{k_BT_c}=0. \quad (5)$$

Here, λ_m and μ_m are the phonon-mediated attraction and the Coulomb pseudopotential in the m-pairing channel, given, respectively, by

$$\frac{\lambda_m}{\lambda}=\delta_{m,0}+\frac{\alpha}{2\sqrt{\gamma}}\int_0^\gamma\frac{dx[x+1-\sqrt{x(x+2)}]^m}{\sqrt{x+2}} \quad (6)$$

and

$$\frac{\mu_m}{\mu_c}=\frac{\sqrt{\tilde\gamma}}{2}\int_0^{\tilde\gamma}\frac{dx[x+\beta+1-\sqrt{(x+\beta)(x+\beta+2)}]^m}{\sqrt{x(x+\beta)(x+\beta+2)}}, \quad (7)$$

where $\lambda=N(0)C^2/NMc_\parallel^2$, $\gamma=\pi^2/2d^2k_F^2(1+\alpha)$, d is the interlayer distance, $\tilde\gamma=\gamma(1+\alpha)$, $\mu_c=4e^2d^2N(0)/\pi V\epsilon_0$, and $\beta=q_s^2/2k_F^2$ [note that λ, μ_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 nonzero orbital momentum turns out finite at any finite anisotropy, $\alpha\neq0$, but numerically smaller than in the s-channel [Fig. 1 (inset)], as is also seen from its analytical expressions for s-wave pairing, $m=0$ (λ_s), p-wave pairing, $m=1$ (λ_p), and for d-wave pairing, $m=2$ (λ_d), obtained by integrating in Eq. (6). When the interlayer distance is much larger than the wavelength of electrons, $\gamma\ll1$, one obtains

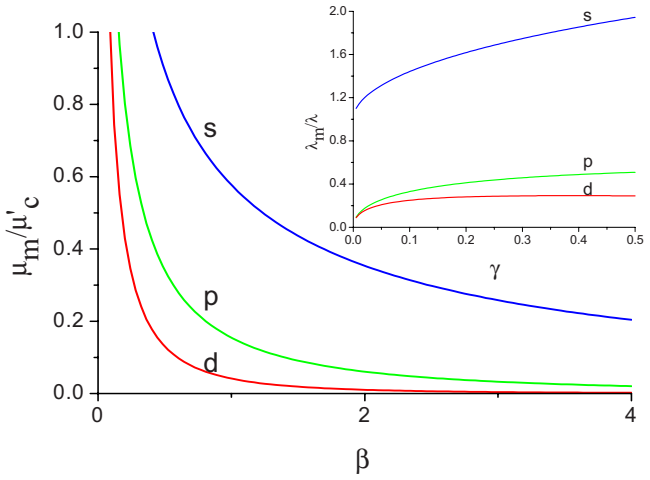

FIG. 1. (Color online) The Coulomb repulsion μ_m as a function of the ratio of the electron wavelength to the screening length squared ($\beta=q_S^2/2k_F^2$), and the electron-phonon coupling constant λ_m as a function of the ratio of the electron wavelength to the interplane distance squared, $\gamma=\pi^2/2d^2k_F^2(1+\alpha)$ for $\alpha=4$ (inset) in s, p, and d pairing channels. Here, $\mu_c'=\mu_c\tilde\gamma$.

$\lambda_s\approx\lambda$, $\lambda_p\approx\lambda\alpha(\gamma/2)^{1/2}/2$, and $\lambda_d\approx\lambda_p$. In the opposite limit, $\gamma\gg1$, one finds $\lambda_s\approx\lambda(1+\alpha)$, $\lambda_p\approx\lambda\alpha(2/\gamma)^{1/2}/3$, and $\lambda_d\approx\lambda_p/5$.

The Coulomb repulsion is much smaller in the unconventional pairing states than in the conventional s-wave state (Fig. 1), which is also seen from the analytical expression for μ_m [Eq. (7)]. If $\tilde\gamma\lesssim\beta$, the repulsion constant μ_m drops as $1/\beta^{m+1}$ in the m channel at strong screening, when $\beta\gg1$. It provides a wide region with unconventional pairs in the "γ-β" parameter space, in spite of the lower values of their electron-phonon coupling constants (Fig. 2). Indeed, the critical temperature for m-Cooper pairing is

$$T_{cm}=1.14\omega_D\exp[-1/(\lambda_m-\mu_m^*)], \quad (8)$$

where $\mu_m^*=\mu_m/[1+\mu_m\ln(\omega_p/\omega_D)]$, as found from Eqs. (4) and (5). For a fixed set of material parameters (which define λ and μ_c), the physically realized superconducting instability appears in the angular-momentum channel with the highest T_{cm}. A minimum (i.e., critical) ratio λ/μ_c for the existence of superconductivity in the m channel is determined by the condition $\lambda_m=\mu_m^*$ as the function of the parameters γ and β. Naturally, the m-pairing state with the lowest value of the critical ratio has the highest T_c. The critical d-wave surface, $\lambda/\mu_c=S(\gamma,\beta)$, defined using $\lambda_m=\mu_m^*$, is found below s-wave and p-wave surfaces, if $\beta\gtrsim1$, so that the d-wave state is physically realized in this region of parameters, as seen from Fig. 2.

Higher-momentum states, $m\geqslant3$, have even a smaller Coulomb repulsion at large β [Eq. (7)], so that they can be realized as well, if γ is so small, that λ_m in Eq. (6) is almost m independent for $m\geqslant1$. On the other hand, an in-plane anisotropy of the sound velocity, compatible with the symmetry of the perovskite lattice, makes d-wave state more stable compared with the higher-momentum states. Naturally, if the sound speed is enhanced along the diagonal di-

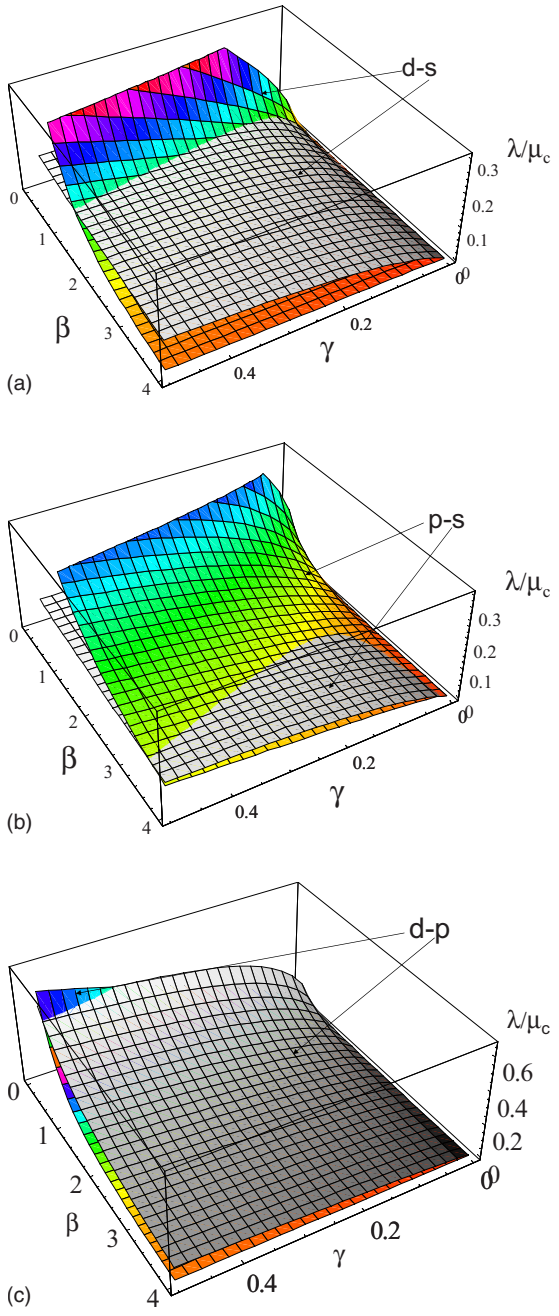

FIG. 2. (Color online) The critical values of the electron-phonon coupling λ for *d*-wave (bright colors) versus *s*-wave (gray color) (d-s), *p*-wave (bright colors) versus *s*-wave (gray color) (p-s), and *d*-wave (bright colors) versus *p*-wave (gray color) Cooper pairs, as functions of the parameters β and γ. The Tolmachev-Morel-Anderson logarithm is set here at $\mu_c \ln(\omega_p/\omega_D)=3$ and $\alpha=3$.

rections of the in-plane primitive cell, the *d*-wave order parameter would be zero along diagonals of the 2D Brillouin zone, where it changes its sign.

Using the simplest parabolic approximation for a 2D-electron energy spectrum, we can draw some conclusions on the carrier-density evolution of the order-parameter symmetry. Within this approximation, $k_F^2=2\pi dn$ and $N(0)=mV/2\pi d\hbar^2$, where $n=2x/\Omega$ is the carrier density and x is the doping level as in $La_{2-x}Sr_xCuO_4$ with the unit cell volume Ω. The ratio of the parameters $\beta=me^2\Omega/2\pi\hbar^2 d^2\epsilon_0 x$ and $\tilde{\gamma}=\pi\Omega/8d^3x\approx 0.044/x$ is independent of the carrier density, $\beta/\tilde{\gamma}=4me^2d/\pi^2\hbar^2\epsilon_0$, 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 μ_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. 3), 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. 3). 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 $\alpha<\alpha_c$ (see Fig. 3).

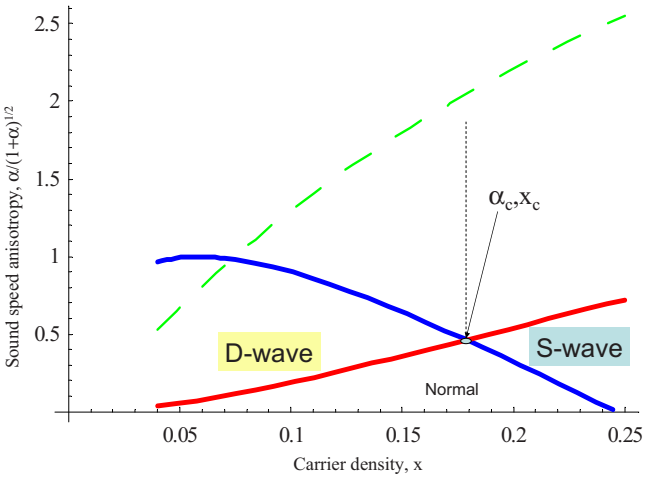

FIG. 3. (Color online) Critical sound-speed anisotropy, $\alpha/(1+\alpha)^{1/2}=(c_\parallel^2-c_\perp^2)/c_\parallel c_\perp$, as a function of doping, x, for $\lambda=\mu_c/12$ (solid lines correspond to *d* and *s* states, and dashed line to *p* state). With increasing carrier density, there is a 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 $\alpha<\alpha_c$.

In the strong-coupling regime, $\lambda\gtrsim 1$, the pairing is individual,[14] in contrast with the collective Cooper pairs. While the Bose condensate of individual bipolarons could break the symmetry on a discreet lattice,[19,20] here I propose a symmetry breaking mechanism, which works even in a continuum model, where the ground state, it would seem, be s wave[13] to satisfy the theorem.[12]

The unscreened Fröhlich EPI with optical phonons in layered ionic lattices such as cuprates has been suggested by us as the key for pairing.[14] 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. (3)] to bind carriers into real-space bipolarons, if $\lambda \geqslant 0.5$.[14] The corresponding potential, $V(\mathbf{r})=-\Sigma_{\mathbf{q}} V_{ph}(\mathbf{q})\exp(i\mathbf{q}\cdot r)$, is nonlocal in real space,

$$V(\mathbf{r}) = -V_{ph}\Omega\left[\frac{\delta(\mathbf{r})}{d} + \frac{\alpha}{4\pi(1+\alpha)^{1/2}r^3}\right], \tag{9}$$

falling as $1/r^3$ at the distance $r \gg d$ between two carriers in the plane, where $V_{ph}=C^2/Mc_{\perp}^2$. While its local part $[\propto \delta(\mathbf{r})=\delta(x)\delta(y)]$ is negated by the strong on-site repulsion U, the second nonlocal part provides bound pairs of different symmetries with the binding energies $\Delta_s > \Delta_p > \Delta_d > \cdots$, in agreement with the theorem.

However, there is the residual Coulomb repulsion between bipolarons $v_c(R)$, significantly reduced by optical phonons. If we approximate the bipolaron as a point charge $2e$, then $v_c(R) \approx 4e^2/\epsilon_0 R$. Since bipolarons have a finite extension ξ, 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,[12] $v_c(R)=4e^2(1 \pm \eta\xi^2/R^2)/\epsilon_0 R$, where η is a number of the order of 1, and plus and/or minus signs correspond to bipolarons in the same or different planes, respectively. The dielectric screening, ϵ_0 is highly anisotropic in cuprates, where the in-plane dielectric constant $\epsilon_{0\|}$ is much larger than the out-of-plane one $\epsilon_{0\perp}$.[21] Hence, the interplane repulsion provides the major contribution to the condensation energy. Since $\xi^2 \propto 1/\Delta$, the repulsion of unconventional bipolarons with smaller binding energies, Δ_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.

Several authors[11] have remarked that superexchange, and not phonons, is responsible for the symmetry breaking in unconventional superconductors such as doped cuprates. Here, I arrive at the opposite conclusion. Indeed, superexchange interaction J is proportional to the electron hopping integral t^2 divided by the on-site Coulomb repulsion (Hubbard U), $J=4t^2/U$, estimated as $J \approx 0.15$ eV in cuprates.[11] 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[15] or the bandwidth squared divided by the *ion-ion* interaction energy of the order of the *nearest-neighbor* Coulomb repulsion, $Mc_l^2 \approx V_c$ in a doped insulator.[16] 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[17,18] favor conventional EPI as the origin of the unconventional pairing both in underdoped cuprates, where the pairing is individual,[14] and in overdoped samples apparently with Cooper pairs.[3]

Nowadays, compelling evidence for a strong EPI has arrived from isotope effects,[22] more recent high resolution angle resolved photoemission spectroscopies,[23] and a number of earlier optical,[24–27] neutron-scattering,[28] and recent inelastic scattering measurements[29] in cuprates. Whereas calculations based on the local spin-density approximation (LSDA) often predict negligible EPI, the inclusion of Hubbard U in the LSDA+U calculations greatly enhances its strength.[30] While the coupling with particular phonon modes is quite different,[23,29,30] EPI with conventional acoustic phonons and the substantial sound-speed anisotropy explain alone the unconventional symmetry of cuprate superconductors.

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