Consistent explanations of tunneling and photoemission data in cuprate superconductors: No evidence for magnetic pairing

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We have analyzed scanning tunneling spectra of two electron-doped cuprates Pr$_{0.88}$La$_{0.12}$CuO$_4$ ($T_c = 21$ K and 24 K) and compared them with tunneling spectrum of hole-doped La$_{1.88}$Sr$_{0.12}$CuO$_4$ and effective electron-boson spectral function of hole-doped La$_{1.97}$Sr$_{0.03}$CuO$_4$ (extracted from angle-resolved photoemission spectrum). We have also analyzed tunneling spectra and angle-resolved photoemission spectra for hole-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$. These results unambiguously rule out magnetic pairing mechanism in both electron- and hole-doped cuprates and support polaronic/bipolaronic superconductivity in hole-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$.

Developing the microscopic theory for high-$T_c$ superconductivity is one of the most challenging problems in condensed matter physics. Twenty-seven years after the discovery of high-$T_c$ superconductivity by Bednorz and Müller [1], no consensus on the microscopic pairing mechanism has been reached despite tremendous experimental and theoretical efforts. There are essentially two opposite views about the pairing mechanism. Many researchers believe that antiferromagnetic fluctuations predominantly mediate the electron pairing [2]. In contrast, other researchers insist that strong electron-phonon coupling is mainly responsible for high-temperature superconductivity in cuprates. The polaron-bipolaron theory of superconductivity [3], which is based on strong electron correlation and significant electron-phonon interaction (EPI), has gained strong support from various experimental results. In particular, extensive studies of unconventional oxygen-isotope effects in hole-doped cuprates have clearly shown strong EPI and the existence of polarons/bipolarons in both normal and superconducting states [4–13]. Neutron scattering [14–16], angle-resolved photoemission (ARPES) [17,18], pump-probe [19], and optical spectroscopies [20,21] have also demonstrated strong electron-phonon coupling. Further, ARPES data [24] and tunneling spectra [25,26] have consistently provided direct evidence for strong coupling to multiple-phonon modes in hole-doped cuprates.

On the other hand, many researchers still maintain the $d$-wave magnetic pairing mechanism, allegedly supported by some highly publicized experimental papers [30,32]. In one of the papers [31], the authors have used an unrealistic parameter, which overestimates the magnetic coupling constant by two orders of magnitude [32]. Other two papers [30,32] reported the combined neutron and tunneling data for two electron-doped Pr$_{0.88}$La$_{0.12}$CuO$_4$–$y$ (PLCCO) crystals with different superconducting transition temperatures (21 and 24 K). These data seemingly suggest that the energies of the magnetic resonance modes are the same as those of the bosonic modes revealed in the second derivative $(d^2I/dV^2)$ of electron tunneling current ($I$) with respect to bias voltage ($V$). They thus conclude that the magnetic resonance modes rather than phonons mediate electron pairing in electron-doped cuprates [30,32]. However, one of us (GMZ) [34] has already pointed out a basic mistake in the data analyses of Ref. [30] and shown that the combined neutron and tunneling data actually disprove this magnetic pairing mechanism. Despite citing Ref. [24], the same basic mistake has been repeated in Ref. [32]. Based on the repeated incorrect analyses, these authors concluded that their data support $d$-wave magnetic pairing mechanism.

Here we re-analyze scanning tunneling spectra of two electron-doped cuprates Pr$_{0.88}$La$_{0.12}$CuO$_4$ ($T_c = 21$ K and 24 K) and compare them with tunneling spectrum of hole-doped La$_{1.88}$Sr$_{0.12}$CuO$_4$ and effective electron-boson spectral function $\alpha^2(\omega)F(\omega)$ of hole-doped La$_{1.97}$Sr$_{0.03}$CuO$_4$. Our data analysis along with other independent tunneling and ARPES data of hole-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$ (BSCCO) consistently rules out magnetic pairing mechanism in both electron- and hole-doped cuprates and supports polaronic/bipolaronic superconductivity in hole-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$.

It has been well established [26,28,29,32,33] that the energies of the dip positions (rather than the peak positions) in $d^2I/dV^2$ correspond to the energies of the modes strongly coupled to electrons. In Fig. 1, we adopt this well-established protocol to identify the mode energies from the $d^2I/dV^2$ spectra of two electron-doped samples. For the electron-doped sample with $T_c = 21$ K, the mode energies identified from the $d^2I/dV^2$ spectrum below 35 meV are 6.0 meV and 16.7 meV (indicated by the arrows). For the electron-doped sample with $T_c = 24$ K, the mode energy identified from the $d^2I/dV^2$ spectrum between 7 and 28 meV is 16.5 meV. It is apparent that the mode energies revealed in the $d^2I/dV^2$ spectra of the two electron-doped cuprates are nearly the same (16.7 and 16.5 meV) and significantly different from the magnetic resonance energies (9.0 or 10.5 meV) revealed by inelastic neutron scattering experiments [31,32]. The mode energy of about 16.5 meV, which is independent of doping and $T_c$, agrees with the energies of the two transverse optical (TO) phonon modes (15.6 meV for the $E_u$ mode and 17.0 meV for the $A_{2u}$)
of the modes. The similar mode energies across different single-layer hole-doped cuprates. This is also consistent with both ARPES data and theoretical studies, which show that the optical phonons are strongly coupled to electrons due to the unscreened long-range interaction along the $c$-axis. It is interesting that in the structurally similar single-layer hole-doped La$_{1.84}$Sr$_{0.16}$CuO$_4$ (LSCO) sample, there is a similar mode with energy of 17.5 meV (see Fig. 2a). The same mode with energy of 16.8 meV is independently revealed in the effective electron-boson spectral function $\alpha(\omega)F(\omega)$ of hole-doped La$_{1.97}$Sr$_{0.03}$CuO$_4$ sample (see Fig. 2b). It is striking that the mode energies (5.9 meV, 17.5 meV, and 27.3 meV) inferred from the dip positions of the tunneling spectrum match the peak positions (6.0 meV, 16.8 meV, and 26.9 meV) in the effective electron-boson spectral function independently determined from ARPES data. This further justifies the correctness of our mode assignment.

Since the magnetic-resonance mode energy was found to be proportional to $T_c$, insensitivity of the mode energies to $T_c$ further argues against the magnetic origin of the modes. The similar mode energies across different doping levels (Fig. 2) and across different single-layer structures (PLCCO and LSCO) are consistent with the phonon mode assignment and contradict the magnetic mode assignment.

For double-layered hole-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$, strong coupling features below 35 meV are also seen in the $d^2I/dV^2$ spectrum (see Fig. 3a). It is striking that the energies of all the dip features in the $d^2I/dV^2$ spectrum precisely match the energies of the peak features in the phonon density of states obtained from high-resolution inelastic neutron scattering. This suggests that these bosonic modes strongly coupled to electrons should be the phonon modes. Therefore, strong coupling to multiple phonon modes is universally seen in single-layered electron- and hole-doped cuprates and in double-layered hole-doped cuprates.

It is worth noting that the bosonic mode at about 6.0 meV seen in the overdoped $n$-type cuprate (Fig. 1a) and in the $p$-type La$_{1.84}$Sr$_{0.16}$CuO$_4$ (Fig. 2a) and La$_{1.97}$Sr$_{0.03}$CuO$_4$ (Fig. 2b) is also seen in the tunneling spectrum of Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Fig. 3a) and in the tunneling spectrum of YBa$_2$Cu$_3$O$_7$ (Ref. [28]), as well as in the ARPES data of Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Ref. [44–46]). This

![Fig. 1: a) $d^2I/dV^2$ spectrum for an electron-doped PLCCO sample with $T_c = 21$ K, which is reproduced from Ref. [32]. b) $d^2I/dV^2$ spectrum for an electron-doped PLCCO sample with $T_c = 24$ K, which is reproduced from Ref. [30]. The energy positions of the $d^2I/dV^2$ spectra are measured from the superconducting gap.](image1)

![Fig. 2: a) $d^2I/dV^2$ spectrum at 7.2 K for hole-doped La$_{1.84}$Sr$_{0.16}$CuO$_4$, which is reproduced from Fig. 4 of Ref. [24]. The energy position of the $d^2I/dV^2$ spectrum is measured from the superconducting gap. b) Effective electron-boson spectral function $\alpha(\omega)F(\omega)$ for the hole-doped La$_{1.97}$Sr$_{0.03}$CuO$_4$, which is extracted from high-resolution ARPES data. The curve is reproduced from Ref. [24].](image2)
numerically taking the derivative of the dI/dV spectrum (postive bias) of Ref. [42] after the spectrum is smoothened by cubic spline. The energy position of the d²I/dV² spectrum is measured from the superconducting gap (Δ = 31.0 meV). b) Phonon density of states for slightly overdoped Bi₂Sr₂CaCu₂O₈. The curve is reproduced from Ref. [43].

![Graph](image)

**FIG. 3:** a) d²I/dV² spectrum of double-layered hole-doped Bi₂Sr₂CaCu₂O₈. The d²I/dV² spectrum is obtained by numerically taking the derivative of the dI/dV spectrum (positive bias) of Ref. [42] after the spectrum is smoothened by cubic spline. The energy position of the d²I/dV² spectrum is measured from the superconducting gap (Δ = 31.0 meV). b) Phonon density of states for slightly overdoped Bi₂Sr₂CaCu₂O₈. The curve is reproduced from Ref. [43].

indicates that this mode is universal for the cuprate systems and cannot originate from antiferromagnetic fluctuations.

The phenomenological spin-fermion model of Abanov and Chubukov [47] showed that strong coupling to the magnetic resonance mode yields a peak-dip-hump (PDH) structure in ARPES spectra along the antinodal direction [47]. This theory is based on the one-loop correction to the t − t' − J mean-field theory. This PDH structure was also predicted to be present in dI/dV tunneling spectra based on the conventional strong coupling theory [48]. Within both approaches, the energy separation between the dip and peak features is exactly equal to the energy Eₜ of the magnetic resonance mode [47, 48], that is, Eₕp = Eₜ. Since Eₜ was found [41] to be proportional to Tₖ, Eₕp - Eₜ should also be proportional to Tₖ. This is in sharp contrast to the experimental results shown in Fig. 4. The Eₕp - Eₜ values obtained from ARPES (Fig. 4a) and tunneling spectra (Fig. 4b) are nearly independent of doping or Tₖ, in contradiction with the the d-wave magnetic pairing mechanisms based on the spin-fermion model [47] and on the conventional approach [48]. Therefore, the results shown in Fig. 4 rule out the magnetic pairing mechanism based on the phenomenological spin-fermion model and on the conventional approach.

More quantitative approach to the t − t' − J model is the slave-boson mean-field theory developed by Brinckmann and Lee [50]. The pairing interaction is the antiferromagnetic exchange energy J. This mean-field theory also predicts the PDH structure in ARPES spectra along the antinodal direction [51]. The predicted Eₕp - Eₜ is nearly independent of doping and in the range of 0.32-0.46J depending on the energy resolution [51]. With J = 130 meV, one yields Eₕp - Eₜ = 41.6-59.8 meV, which is in reasonable agreement with the data shown in Fig. 4a. Thus, the result shown in Fig. 4a alone cannot rule out the magnetic pairing mechanism based on the t − J model.

On the other hand, the t − t' − J model predicts a doping independent Eₜ = 0.54J = 70.2 meV in the overdoped range [51]. This prediction is in contradiction with neutron data [52], which show that Eₜ is equal to 5.4kBTₖ in the overdoped range. For overdoped Y₀.₈₅Ca₀.₁₅Ba₂Cu₃O₇ with Tₖ = 75 K, Eₜ was found to 34 meV (Ref. [53]), which is only half the predicted value.
of 70 meV. Therefore, the neutron data rule out the magnetic pairing mechanism based on the \( t - t' - J \) model.

The second important prediction of the \( t - t' - J \) model is the existence of the pronounced hump feature at energy close to \( J \) (about 130 meV) at all the doping levels \([51]\). This is in contradiction with the ARPES data shown in Fig. 5. The energy position of the hump feature increases rapidly with the decrease of doping and reaches a value of about 230 meV at a doping level of 0.089. Therefore, the ARPES data further rule out the magnetic pairing mechanism based on the \( t - t' - J \) model.

Alternatively, the peak-dip-hump features consistently observed in scanning tunneling, break-junction tunneling, and angle-resolved photoemission spectra can be quantitatively explained by the polaron-bipolaron theory of superconductivity, where the hump is the incoherent broad feature that reflects the local hopping of electrons in various frozen lattice configurations \([55]\). This theory \([55]\) predicts that \( E_{hp} - E_{pk} \) in break-junction (superconductor-insulator-superconductor) tunneling spectra is close to the energy \( \Omega \) of the phonon modes in polaronic cloud when the coupling strength \( g^2 \) is not too large. Fig. 6a shows the theoretical prediction of \( E_{hp} - E_{pk} \) with varying \( g^2 \) and a fixed \( \Omega = 72 \) meV. The numerically calculated break-junction tunneling spectra are demonstrated in Fig. 1 of the Supplemental Material \([56]\). It is apparent that \( E_{hp} - E_{pk} \) is close to \( \Omega \) when \( g^2 \) is below 1.5 and close to \( 2\Omega \) when \( g^2 \) is above 1.5. This theoretical prediction is in quantitative agreement with the data shown in Fig. 6b. The data are consistent with \( g^2 < 1.5 \) for optimally and overdoped BSCCO and with \( 1.5 \leq g^2 \leq 2.0 \) for underdoped BSCCO.

The 72 meV phonon mode should be mainly associated with oxygen vibration so that the whole broad hump feature should shift down by about 72(1-\(\sqrt{18/16}\)) meV = 4.3 meV upon replacing \(^{16}O\) by \(^{18}O\) (Ref. \([49]\)). Since the peak position in \( d^2I/dV^2 \) simply corresponds to the steepest point of the hump feature in \( dI/dV \), the oxygen isotope shift of the steepest point at 52 meV implies that the hump feature is also shifted down by about 3.7±0.8 meV upon replacing \(^{16}O\) by \(^{18}O\). Therefore, the observed oxygen-isotope shift of the steepest point of the hump feature in \( dI/dV \) provides further proof that PDH arises from the lattice polaronic effect.

In summary, our analyses of tunneling and ARPES spectra in both electron and hole doped cuprates have unambiguously ruled out magnetic pairing mechanism and support polaronic/bipolaronic superconductivity in hole-doped Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8\). For detailed discussions about the polaron-bipolaron theory and the intrinsic pairing symmetry in cuprate superconductors, see the Supplemental Material \([56]\).
