Fine structure in the tunneling spectra of electron-doped cuprates: No coupling to magnetic resonance mode

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We analyze high-resolution scanning tunneling spectra of the electron-doped cuprate Pr$_{0.85}$LaCe$_{0.15}$CuO$_4$ ($T_c = 24$ K). We find that the spectral fine structure below 35 meV is consistent with strong coupling to a bosonic mode at 16 meV, in quantitative agreement with early tunneling spectra of Nd$_{1.85}$Ce$_{0.15}$CuO$_4$. Since the energy of the bosonic mode is significantly higher than that (9.5-11 meV) of the magnetic resonance-like mode observed by inelastic neutron scattering, the coupling feature at 16 meV cannot arise from strong coupling to the magnetic mode. The present work thus demonstrates that the magnetic resonance-like mode cannot be the origin of high-temperature superconductivity in electron-doped cuprates.

The microscopic pairing mechanism for high-temperature superconductivity in cuprates remains elusive despite tremendous efforts for over twenty years. The most central issue is the origin of the bosonic modes mediating the electron pairing. Most workers believe that magnetic resonance modes, which have been observed in various hole-doped double-layer cuprate systems, predominantly mediate the electron pairing. Recent observation of a magnetic resonance-like mode at 9.5-11.0 meV in two electron-doped cuprates \cite{1 2} seems to suggest that the magnetic resonance is a universal property of all cuprate systems and thus essential to the pairing mechanism of high-temperature superconductivity. However, this speculated magnetic pairing mechanism is seriously undermined by recent optical experiments \cite{3} which showed that the electron-boson spectral function $\alpha^2(\omega)F(\omega)$ is independent of magnetic field, in contradiction with the theoretical prediction based on the magnetic pairing mechanism (see Fig. 9 of Ref. \cite{2}). In contrast, extensive studies of various unconventional oxygen-isotope effects in hole-doped cuprates have clearly shown strong electron-phonon interactions and the existence of polarons \cite{4 5 6 7 8 9 10 11 12}. Neutron scattering \cite{13}, angle-resolved photoemission (ARPES) \cite{14 15}, and Raman scattering \cite{16} experiments have also demonstrated strong electron-phonon coupling. Further, ARPES data \cite{17} and tunneling spectra \cite{18 19 20 21 22} have consistently provided direct evidence for strong coupling to multiple-phonon modes in hole-doped cuprates. Therefore, electron-phonon coupling in hole-doped cuprates should play an important role in the pairing mechanism.

On the other hand, the role of electron-phonon coupling in the pairing mechanism of electron-doped cuprates has not been clearly demonstrated. Early tunneling spectra in Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ (NCCO) suggested predominantly phonon-mediated pairing \cite{23} while the oxygen-isotope exponent $\alpha_O$ in Pr$_{1.85}$Ce$_{0.15}$CuO$_4$ was found to be 0.08±0.01 (Ref. \cite{24}), which is significantly below 0.5, expected for the phonon-mediated mechanism. Moreover, surface-sensitive ARPES experiment showed very weak electron-phonon coupling \cite{25}, which may support an alternative mechanism where the 10 meV magnetic resonance mode is mainly responsible for the pairing. If this were the case, the strong coupling to the 10 meV magnetic excitation would show up in single-particle tunneling microscopy that has a much higher energy resolution than ARPES. However, the early tunneling spectra \cite{22} do not seem to show this coupling feature at about 10 meV. One might argue that the absence of this coupling feature in the early data could be due to a low experimental resolution and poor sample quality. Therefore, it is essential to obtain reproducible high-resolution single-particle tunneling spectra and analyze the spectra in a correct way to unambiguously address this issue.

Here we re-analyze high-resolution single-particle tunneling spectra of the electron-doped cuprate Pr$_{0.85}$LaCe$_{0.15}$CuO$_4$ ($T_c = 24$ K) \cite{26}. The $d^2I/dV^2$ spectra reveal one dip and two peak features below $V = 35$ meV, where $I$ is the tunneling current and $V$ is the bias voltage. We find that these fine features are consistent with strong coupling to a bosonic mode at 16 meV, in quantitative agreement with early tunneling spectra \cite{22} of Nd$_{1.85}$Ce$_{0.15}$CuO$_4$. Since the energy of the bosonic mode is significantly higher than that (9.5-11 meV) of the magnetic resonance-like mode observed by inelastic neutron scattering \cite{1 2}, this coupling feature cannot arise from strong coupling to the magnetic mode. The present work thus demonstrates that the magnetic resonance-like mode cannot be the origin of high-temperature superconductivity in electron-doped cuprates.

For conventional superconductors, the energies of the phonon modes coupled to electrons can be precisely determined from the second derivative tunneling spectra $d^2I/dV^2$. Measured from the isotropic $s$-wave superconducting gap $\Delta$, the energy positions of the dips (minima) in $d^2I/dV^2$ correspond to those of the peaks in the electron-phonon spectral function $\alpha^2(\omega)F(\omega)$ (see Fig. 1 below and also Refs. \cite{27 28}). In a recent article \cite{29} attempting to show an important role of phonons in the electron pairing, the authors assign the energy
(52 meV) of a peak position in $d^2I/dV^2$ spectra of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+δ}$ to the energy of a phonon mode. Such an assignment is incorrect because the energies of phonon modes are equal to the energies of dip positions rather than peak positions in $d^2I/dV^2$ (see Fig. 1 below and also Refs. [21, 22]). The same mistake occurs in a more recent article [26] where the authors also assign the energy (10.5 meV) of a peak position in $d^2I/dV^2$ spectra of $\text{Pr}_{0.88}\text{LaCe}_{0.12}\text{CuO}_4$ (PLCCO) to the energy of a bosonic mode. Since this mistakenly assigned mode energy (10.5 meV) is very close to the energy (9.5-11 meV) of the magnetic resonance-like mode measured by inelastic neutron scattering [1, 2], the authors [26] conclude that the magnetic resonance mode mediates electron pairing in electron-doped cuprates.

Figure 1 shows the normalized second derivative $(d^2I/dV^2)_S/(dI/dV)_N$ for the conventional superconductor Pb along with the electron-phonon spectral function $\alpha^2(\omega)F(\omega)$ (where $S$ represents the superconducting state and $N$ the normal state). The figure is reproduced from Ref. [27]. It is apparent that the dip features in $(d^2I/dV^2)_S$ match precisely with the peak features in $\alpha^2(\omega)F(\omega)$ (see the vertical solid lines).

In Fig. 2a, we show the tunneling conductance $(dI/dV)_S$ in the superconducting state for the electron-doped $\text{Pr}_{0.88}\text{LaCe}_{0.12}\text{CuO}_4$ (PLCCO) crystal (open circles). The data are digitized from Ref. [26]. The normal-state tunneling conductance $(dI/dV)_N$ is approximated by a straight line, which is obtained with conservation of states. $b$) Normalized conductance $(dI/dV)_S/(dI/dV)_N$. The solid line is the numerically calculated curve in terms of an anisotropic $s$-wave gap symmetry and the dashed line is the numerically calculated curve in terms of $d$-wave gap symmetry.

$$\frac{dI}{dV} \propto \int_{0}^{2\pi} p(\theta - \theta_0) \text{Re} \left[ \frac{eV - i\Gamma}{\sqrt{(eV - i\Gamma)^2 - \Delta^2(\theta)}} \right] N(\theta) d\theta,$$

where $N(\theta)$ represents the anisotropy of the band dispersion, $\Gamma$ is the life-time broadening parameter of an electron, $p(\theta - \theta_0)$ is the angle dependence of the tunneling probability and equal to $\exp[-\beta \sin^2(\theta - \theta_0)]$, and $\theta_0$ is the angle of the tunneling barrier direction measured from the Cu-O bonding direction. For simplicity, we assume a cylindrical Fermi surface so that both $N(\theta)$...
and $\beta$ are independent of the angle. The solid line is the numerically calculated curve using $\Gamma = 0.73$ meV, $\beta = 6$, $\theta_0 = 0.18\pi$, and an s-wave gap function: $\Delta = 4.4(1.0 - 0.6 \sin \theta)$ meV. The dashed line is the numerically calculated curve using $\Gamma = 0.40$ meV, $\beta = 7$, $\theta_0 = \pi/4$, and a simple d-wave gap function: $\Delta = 7.2 \cos 2\theta$ meV. It is interesting that the calculated curves for the anisotropic s-wave and d-wave gaps are all in good agreement with the data. We further find that isotropic s-wave gap is inconsistent with the data. Therefore, the tunneling spectrum alone rules out isotropic s-wave symmetry.

In Fig. 3a, we show the second derivative spectrum $(d^2I/dV^2)_S$ together with the first derivative spectrum $(dI/dV)_S$ for the Pr$_{0.88}$La$_{0.12}$CuO$_4$ crystal. The figure is reproduced from Ref. [26]. In the $(d^2I/dV^2)_S$ spectrum, there are two peak features at $E_{p1} = 17.8$ meV and $E_{p2} = 29.0$ meV, and one dip feature at $E_{d1} = 23.4$ meV, as indicated in the figure. It is clear that the dip feature is just half-way between the two peak features. The energy position $\Delta_R$ of the peak in the $(dI/dV)_S$ spectrum is about 7.0 meV. Following the result of Fig. 1 for Pb, the energy of the bosonic mode coupled strongly to electrons is $\Omega_1 = E_{d1} - \Delta_R = 16.4$ meV, which is slightly lower than the energy of a very strong coupling feature observed in hole-doped cuprates (e.g., 20 meV in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ [20, 21, 31] and 18 meV in La$_{2-x}$Sr$_x$CuO$_4$ [22]).

Figure 3b shows a histogram of the occurrences of $\Delta_R$ (red) and the energies $E_{p1}$ and $E_{p2}$ (blue) for a map of tunneling spectra on a 64 Å×64 Å area of the sample. According to the result in Fig. 3a, the mid-point between $E_{p1}$ and $E_{p2}$ should mark $E_{d1}$. Then, the difference between $E_{d1}$ and $\Delta_R$ is found to be 15.9 meV, that is, $\Omega_1 = 15.9$ meV. More bosonic modes would be revealed if these spectra were extended to higher energies.

Figure 4 shows electron-boson spectral functions for two Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ samples with $T_c = 22\pm 2$ K. The figure is reproduced from Ref. [23]. The energies of the lowest bosonic modes in the spectral functions of the two crystals are 15.2 meV and 17.2 meV, respectively (see arrows in the figure). A simple average of the mode energies of the two NCCO samples is 16.2 meV, which is in quantitative agreement with the mode energy (15.9 meV) averaged from thousands of tunneling spectra of the PLCCO crystal [26]. This quantitative agreement clearly indicates that the strong coupling feature at about 16 meV in the tunneling spectra of electron-doped cuprates is intrinsic.

Having established the bosonic mode energy and statistics, we now discuss the nature of this mode. The measured mode energy of 16 meV rules out its connection to the magnetic resonance mode which has energy of 9.5 meV in NCCO (Ref. [2]) and 11 meV in PLCCO (Ref. [1]). Alternatively, this 16 meV bosonic
mode should be associated with unscreened c-axis polar phonons (transverse optical phonons). Both ARPES data [33] of hole-doped Bi$_2$Sr$_2$CuO$_4$ and a theoretical study [34] indicate that the transverse optical (TO) phonons are strongly coupled to electrons due to the unscreened long-range interaction along the c-axis and play a predominant role in electron pairing. This long-range electron-phonon interaction should be present in any layered system, but is often ignored when one theoretically calculates electron-phonon coupling. For electron-doped Pr$_{1.85}$Ce$_{0.15}$CuO$_4$, the lowest TO modes with energies of 15.6 meV ($E_a$ symmetry) and 17.0 meV ($A_{2u}$) were identified by infrared reflectivity measurements [32]. Since these two TO modes have energies very close to the energy of the bosonic mode seen in the tunneling spectra, it is natural that the 16 meV coupling feature is associated with strong coupling of these TO phonon modes to electrons.

In summary, we have analyzed the high-resolution tunneling spectra of the electron-doped Pr$_{0.89}$LaCe$_{0.12}$CuO$_4$. We find that the spectral fine structure below 35 meV is consistent with strong coupling to a bosonic mode at 16 meV, in qualitative agreement with early tunneling spectra [23] of Nd$_{1.85}$Ce$_{0.15}$CuO$_4$. Since the energy of the bosonic mode is significantly higher than that (9.5-11 meV) of the magnetic resonance-like mode observed by inelastic neutron scattering, the coupling feature at 16 meV cannot arise from strong coupling to the magnetic mode. The present work thus demonstrates that the magnetic resonance-like mode cannot be the origin of high-temperature superconductivity in electron-doped cuprates.

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