Unusual Layer-Dependent Charge Distribution, Collective Mode Coupling, and Superconductivity in Multilayer Cuprate $\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_8\text{F}_2$

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Low energy ultrahigh momentum resolution angle resolved photoemission spectroscopy study on four-layer self-doped high $T_c$ superconductor $\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_8\text{F}_2$ (F0234) revealed fine structure in the band dispersion, identifying the unconventional association of hole and electron doping with the inner and outer CuO$_2$ layers, respectively. For the states originating from two inequivalent CuO$_2$ layers, different energy scales are observed in dispersion kinks associated with the collective mode coupling, with the larger energy scale found in the electron ($n$-) doped state which also has stronger coupling strength. Given the earlier finding that the superconducting gap is substantially larger along the $n$-type Fermi surface, our observations connect the mode coupling energy and strength with magnitude of the pairing gap.

In the search for the mechanism of high-$T_c$ superconductivity, the discovery of electron-boson coupling in the form of a dispersion anomaly with sharp energy scales has attracted significant attention [1–6]. Their universal presence in high-$T_c$ superconductor families makes them a possible common origin of pairing. The novel four-layer CuO$_2$-doping FSs, which positively identifies the association of pairing gap magnitude in the same sample within a single material under identical experimental conditions.

ARPES experiments were performed at two facilities, with the low photon energy (8.5–10 eV) measurements at beam line 9 A of Hiroshima Synchrotron Radiation Center (HiSOR) and medium energy (22.55 eV) measurements at beam line 10.0.1.1 of the Advanced Light Source (ALS). For the F0234 sample, we utilized 8.5–9.5 eV/55 eV circularly or linearly polarized light at HiSOR/ALS, and the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (Bi2212) sample was measured at ALS with 22 eV linearly polarized light to manifest the bilayer splitting [11]. At both facilities, measurement pressure was kept $\leq 4 \times 10^{-11}$ Torr, and data were recorded by Scienta R4000 analyzers at 20 K sample temperature. The total convolved energy and angle resolution is 10 meV/16 meV and 0.2°/0.2° at HiSOR/ALS.

In Fig. 1, we show photon energy dependent ARPES measurements along the nodal direction that reveal the fine structure in band dispersions, suggesting an unconventional, layer-dependent charge distribution.

Figure 1(a) is the measurement with a photon energy of 9.5 eV. Compared to earlier measurements [7], we find that the $p$ and $n$ bands are much better separated due to the
The dramatic variation of the energy, although both bands have comparable intensity at the node, $n$-band gains weight quickly when moving away from the node. In all three measurements, the simultaneous presence of $p$ and $n$ bands is self-evident, except now the fine structure of $p$ band cannot be resolved due to $k$-resolution limitations [10]. It is obvious from the plots...
that there exists a clear energy scale indicated by strong \( n \)-band dispersion kink (\( \sim 85 \) meV), which becomes clearer from panels (a) to (c), where \( n \) band becomes more dominant. Further, the renormalization of \( n \) band also becomes stronger, especially in panel (c), where the \( n \)-band kink is so prominent that it appears to intersect into the \( p \)-band dispersion. This clearly shows a band-dependent renormalization effect.

This band (or FS) dependence of the mode coupling is different from other high \( T_c \) superconductors that possess multiple FSs such as Bi2212. For comparison, we illustrate in panels (d)–(f) three measurements for overdoped (\( T_c = 70 \) K) Bi2212 sample at the same \( k \) space loci. Unlike the large splitting between \( p \) and \( n \) bands even along nodal direction in F0234 [panel (a)], from (d) one sees that the antibonding (AB) and bonding (B) bands of Bi2212 are almost degenerate. The comparison is similar between panels (b) and (e). In (f), although the AB and B bands are now separated, the two dispersions are nearly parallel with no noticeable renormalization difference, which resembles the \( p1 \) and \( p2 \) bands in Fig. 1 but contrasts obviously to panel (c).

To better compare the electron-bosonic mode coupling strength of both bands, we show in Fig. 3(a) fit dispersions of four fine measurements around the nodal region. The fitting function used is two Lorentzian peaks convolved with a Gaussian resolution function. As seen in Fig. 2(c), in the region far away from the node, the \( n \) band gets too dominant to permit reliable MDC fitting for the \( p \) band. From Fig. 3(a), it is clear that for both \( p \) band (warmer color) and \( n \) band (cooler color), although the high binding energy dispersions are almost parallel, at low binding energy, the electron velocity decreases clearly from cut (1) to (4). This shows that the electron-bosonic coupling is anisotropic in \( k \) space for both bands. To quantify the different strength of the renormalization, we calculate the velocity reduction factor \( \lambda' \) from each dispersion curve [see Fig. 3(b)], where \( \lambda' \) is defined by \( \frac{dE}{dk_{E_b > 100 \text{ meV}}} = \left( 1 + \lambda' \right) \frac{dE}{dk_{E_b < 60 \text{ meV}}} \). The 100 and 60 meV are chosen to avoid the possible uncertainty of kink positions within this region. While \( \lambda' \) is not exactly the coupling constant \( \lambda \), it is proportional to \( \lambda \) and thus is also appropriate for comparing the renormalization strength [3]. Figure 3(b) confirms that the mode coupling strength is band dependent and anisotropic in \( k \) space within each band. Given that the superconducting gap magnitude for \( n \) band is much larger than that of \( p \) band [7], the stronger coupling indicated by larger \( \lambda' \) correlates with larger pairing strength.

Furthermore, in Fig. 3(a), the sharp kink of \( n \) band reveals an energy scale of \( E_b = 85 \) meV (the \( p \)-band dispersion kink seems blurred due to the presence of strong \( n \) band in vicinity and limited \( k \) space resolution), which obviously differs from the energy scale of \( p \) band [67 meV, see Fig. 1]. This difference can clearly be seen by overlapping the \( p \)- and \( n \)-band nodal dispersions in Fig. 3(c).

FIG. 2 (color). ARPES measurements on F0234 and Bi2212 parallel to the nodal direction (\( k \) space loci in insets). (a)–(c) Spectral intensity plots of F0234 [with fitted \( p \) and \( n \) dispersions superimposed in (a),(b)]. White arrows at \( E_b = 85 \) meV indicate the binding energy of the \( n \)-band dispersion kink. (d)–(f) Measurements at the same \( k \) space loci of Bi2212. Fitted dispersions for bonding (B) and antibonding (AB) bands are superimposed on all three plots (see text).

FIG. 3 (color). (a) Dispersions from four sequential cuts with the \( k \) space separation between adjacent cuts as 0.05\( \pi/a \) (see inset). Warm and cool colors are used for \( p \)- and \( n \)-band dispersions, respectively. (b) Velocity reduction factor \( \lambda' \) (see text) extracted from the dispersions in panel (a) and in Fig. 1. (c) Overlapped dispersions of \( p \) and \( n \) bands at nodal direction. The black dashed line is a guide to the eye that shows the coincided dispersions at high binding energy.
The unconventional charge distribution between inequivalent CuO\(_2\) layers, together with the interplay between the electron-boson coupling strength and the energy gaps of \(p\) and \(n\) bands also provide excellent tests for theory. The fact that there are two oppositely doped superconducting subsystems in a same material enables a comparison of results from different subsystems under identical experimental and material conditions. The unusual charge distribution indicates an unusual chemical environment in this material and the positive correlation between the gap size and kink strength raises the intriguing question of whether the difference in the local superconducting gap size is linked to the difference of local electron-boson coupling strength.

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[10] Assuming \(\delta k\) and \(\delta\Delta\) represent the \(k\) space and angle resolution, given the photoelectron energy \(E\) and mass \(m_e\), 
     \[ \delta k = \delta\Delta / \sqrt{2m_eE} \] (nonrelativistic case). Thus for the angle resolution \(\delta\Delta\) (fixed for a given analyzer), the \(k\) space resolution \(\delta k\) becomes smaller (better) with lower \(E\).
[25] Given the breadth of the kink and the previous finding of the presence of multiple modes [X.J. Zhou, Phys. Rev. Lett. 95, 117001 (2005)], these two mode energies are almost identical (even though their absolute values may vary with gap criteria).