Doubling of the bands in overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$-probable evidence for c-axis bilayer coupling

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We present high resolution ARPES data of the bilayer superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) showing a clear doubling of the near $E_F$ bands. This splitting approaches zero along the (0, 0) $\rightarrow$ ($\pi$, $\pi$) nodal line and is not observed in single layer $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201), suggesting that the splitting is due to the long sought after bilayer splitting effect. The splitting has a magnitude of approximately 75 meV near the middle of the zone, extrapolating to about 100 meV near the (\(\pi, 0\)) point.

One of the central features of the cuprate superconductors is that their physical properties depend strongly on the number of $Cu-O$ planes (\(n\)) per unit cell. For example, increasing \(n\) from 1 to 3 almost universally results in large enhancements in the superconducting $T_c$ [1]. This would seem to argue for the importance of some sort of coupling between the $Cu-O$ planes within a unit cell. Despite this, there have been essentially no direct observations of the intracell coupling between the planes, and the vast majority of theoretical models for the cuprates focus on a single $Cu-O$ plane, i.e. on the limit of zero intra and intercell c-axis coupling.

In this letter, we present the first high energy/momentum resolution angle resolved photoemission (ARPES) data of a cuprate superconductor which shows a clear doubling of the bands in the normal state. The splitting is observed in the double layer compound $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) but is much weaker or absent in the single layer compound $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201). The splitting approaches zero along the (0, 0) $\rightarrow$ ($\pi$, $\pi$) nodal line and is maximal at $\bar{M}$(0, 0), and appears to be larger for overdoped samples. These trends point towards an intracell c-axis coupling between the $Cu-O$ planes (bilayer splitting) as the likely origin of the splitting, although other effects such as microscopic phase separation may play a role. In addition to the new physics of this coupling, our new findings may help resolve the recent controversy about the electron-like [2, 3] and hole-like [4] Fermi surfaces which have recently been measured on Bi2212.

The experiments were carried out at the Stanford Synchrotron Radiation Laboratory (SSRL), Stanford, CA, at the Advanced Light Source (ALS), Berkeley, and at the Synchrotron Radiation Center (SRC), Stoughton, WI. At each facility we used a Scienta 200mm energy analyzer running angle mode to simultaneously collect up to 84 individual Energy Distribution Curves (EDCs) along an $\approx$ 14° angular slice. The angle resolution was about 0.08° along the angular slice (the $\theta$ direction) and about 0.4° in the perpendicular direction (the $\phi$ direction). At the experimental photon energy $h\nu = 24.7 eV$, the Brillouin Zone edge ($\pi/a$) will be $\approx 20°$ and the corresponding momentum resolution will be ($\Delta k_x, \Delta k_y$) $\approx$ (0.008 $a^{-1}$, 0.04 $a^{-1}$) with $a$ the lattice constant. The energy resolution was better than 20 meV determined from the 10 – 90% width of a gold reference which was in electric contact with the sample and was used to determine the Fermi Energy $E_F$. Data is shown from a lightly overdoped Bi2212 sample with a $T_c = 85 K$ and a lightly overdoped Bi2201 samples with $T_c \approx 5 K$. The photon polarization direction was along the $\theta$ direction and the $\Gamma$(0, 0) $\rightarrow$ $Y$($\pi, \pi$) high symmetry line. All data shown here was normalized by using the spectral weight in the high binding energy window ($-0.8 eV$, $-0.6 eV$) only [8].

![Figure 1](image)

**FIG. 1.** (a),(b) ARPES intensity map within $\pm$5 meV of the $E_F$ from the normal state ($T = 100 K$) of an overdoped Bi2212 sample. The photon energy was 24.7 eV and the polarization 40% along the $\theta$ direction and 60% out of plane. An overlay of the data with the Bi2212 Brillouin Zone is shown in (a). The hole-like main FS (labeled "Main") plus superstructure bands (labeled "S.S.") was obtained at 19 eV by Ding et al. [5] (yellow lines). The FS crossings determined from MDC fitting are indicated by white dots.

Figure 1a shows the schematic plot of the commonly accepted Fermi Surface (FS) topology of Bi2212 [2] in...
part of the first Brillouin Zone. The thick yellow lines are the main FS and the thin yellow lines are the FS produced by the crystal distortion (the superstructure, labeled “S.S.”). Figure 1b is the intensity distribution of the ARPES spectral weight at $E_F$ in the normal state of a lightly overdoped Bi2212 sample, with the $\vec{k}$ space location of this panel indicated in figure 1a. Modulo matrix element effects, the high intensity locus at $E_F$ should represent the FS topology of the Fermi energy. In panel b, one can see that the main FS splits into two branches, one produced by band A and the other by B. This splitting behavior can also be seen in the superstructure FS, even though the intensity is much weaker there. This behavior is not expected within the generic FS topology shown by the yellow lines in figure 1a.

In order to examine the splitting of these bands more carefully, we examine the $\vec{k}$-dependence of the states as a function of binding energy. Figure 2a shows data along the blue line ($\phi = 7^\circ$) in figure 1a, with the main band crossing $E_F$ near the center of the window. The so-called “main band”, which shows up as the high intensity feature, disperses from binding energy $\approx -0.12$ eV (around emission angle $\theta = -6^\circ$) towards $E_F$. Around $\theta = -12^\circ$, another weaker feature appears at slightly deeper binding energy. This two-peak structure (labeled “A” and “B” here) can be easily seen in the energy distribution curve (EDC) shown in panel b, which was taken at $\theta = -12^\circ$ (vertical white line in panel a). We emphasize that such two-peak structure in EDC is self-evident without any analysis on the data. However, further quantitative determination of the peak positions, FS crossings, or splitting is difficult from the EDCs, as a detailed understanding of their line shape and background is still unclear. In addition, the Fermi function distorts the line shape near the Fermi energy.

On the other hand, it is now well established that for relatively dispersive states the Momentum Distribution Curves (MDCs, intensity versus emission angle $\theta$ with fixed binding energy) have a simple line shape which can be modeled by a simple Gaussian or Lorentzian function. This is equivalent to saying that the electron self-energy is essentially $\vec{k}$-independent over the narrow $\vec{k}$-range of a MDC peak. A further advantage of the MDC method is that it won’t be affected by the Fermi function cutoff, so in principle one can track ARPES features all the way up to or even above $E_F$. Figure 2c shows the MDCs from the data of 2a over the full $\theta$ range and from 10 meV above $E_F$ to 60 meV below. The MDC at $E_F$ is further shown in panel d (blue dots), along with a fitting (red line) of the main band into two separate Lorentzian peaks A and B and the superstructure band into two corresponding features (green lines), plus a linear background (black dash line). For the fit, the angular width, position, and intensity of each feature was allowed to vary. It was found that the angular width of both branches were essentially identical, and the intensity ratio and the angular splitting between the branches found to be roughly constant as a function of binding energy (see figure 2e). The high quality fits allow an accurate determination of the peak positions for each component. Such fits were performed for each MDC at $E_F$ between the $\phi$ angles of 3$^\circ$ and 8$^\circ$. In this way the white dots of figure 1a were determined which, as expected, perfectly match the high intensity locus.

![FIG. 2. (a) False color plot of $E$ vs $\theta$ for the $\phi = 7^\circ$ cut (blue line in figure 1a). (b) EDC at $\theta = -12^\circ$ from panel a (vertical white dash line). Two distinct features, A and B, can be clearly seen in this EDC. (c) MDCs from panel a from binding energies $+10$ meV to $-60$ meV, with 5 meV step. (d) The MDC at $E_F$ (blue dots), including a deconvolution of the main band (red line shows the fitting result) into two Lorentzian functions A and B (green lines) plus two corresponding features in superstructure band (green lines) on top of a linear background (black dash line). (e) The energy dependence of the $\theta$ value of MDC peaks A (close circles) and B (open circles). The error bar from the fitting is smaller than the symbol size.](image-url)
the possibility that the sample surface was slightly multifaceted, which each facet contributing to a feature at a slightly different angle. This is not the case, as a post-cleave laser reflection plus optical inspection revealed angular deviations no greater than 0.2°, while the angular splitting of the states is on the order of 2°. X-ray Laue and LEED reflections showed sharp spots as well, indicating high quality crystalline of the samples. We also considered the possibility that there may be Bi2201 intergrowths in the Bi2212 samples, which would be very difficult to detect from magnetization or x-ray measurements. In this possibility, it could be imagined that one of the branches was due to Bi2212 and the other to Bi2201. One could possibly imagine this effect causing the splitting on one cleave, but not on the numerous cleaves we have studied. Also, we have cooled a sample down to 35K which is below $T_c$ for Bi2212 and above the $T_c$ for Bi2201. Since the superconducting gap was observed in both branches, it rules out the possibility of the Bi2201 contamination.

Two main intrinsic possibilities exist: (a) the bilayer splitting due to the intracell c-axis coupling, and (b) microscopic phase separation into hole rich and hole poor regions each with distinct bands and Fermi surfaces. We first discuss option (a), which we believe to be much more likely. We first remind the reader that in the bilayer splitting scenario, the electronic states from each of the two CuO planes per unit cell will couple, breaking their degeneracy. One set (antibonding) will increase in energy while the other (bonding) will decrease. Within this picture we make the assignment on the figures as peak A=antibonding and peak B=bonding. Important information is garnered by comparing the spectra to that of the single layer compound Bi2201, which cannot exhibit bilayer splitting. Figure 3a shows an MDC at $E_F$ from the $\phi = 7^\circ$ angular slice of a lightly overdoped Bi2201 sample taken under the identical experimental configuration as that of figure 2. Unlike figure 2d, a single Lorentzian function alone is enough to produce a very good fit to either the main or superstructure band. This indicates that there is no splitting of the bands in Bi2201, at least not one with a similar magnitude as observed in Bi2212. This Bi2201 data also helps rule out other possible origins for the band doubling since the crystal structures for Bi2212 and Bi2201 are very similar. For example, if the additional band in Bi2212 was a Bi $- O$ band or a $Cu_{3d_{xy}z}$ $- O_2$ hybrid then we would expect to see them showing up in Bi2201 as well. The lack of the apparent doubling in Bi2201 seems to argue against microscopic phase separation, however since the mechanism for the phase segregation is not completely conclusive, we hold this as a less likely mechanism.

The $\vec{k}$-space dependence of the splitting of the two bands is also consistent with the bilayer splitting effect. Symmetry arguments dictate that the coupling between the planes should vanish along the $(0,0) \rightarrow (\pi,\pi)$ symmetry line and grow as the $(\pi,0)$ region is approached, exactly as the data in figure 1 shows. More precisely, the bilayer splitting has been parameterized by $\Delta(k) = 0.5 * t_\perp (\cos(k_x a) - \cos(k_y a))^2$ which has a maximum splitting of $2t_\perp$ at $(\pi,0)$ $[15,16]$. Using this equation to parameterize the overdoped data of figure 1 and 2, we find a $t_\perp$ of $\sim 55meV$ and an extrapolated splitting at $(\pi,0)$ of $110meV$. This is small compared to the band structure prediction of nearly $300meV$ $[17]$ implying important correlation effects present in the system, but it is still larger than or comparable to other important energy scales $(T_c, T^*, J)$ meaning that it is an effect which should not be ignored for the physics of these compounds.

Although we have not yet carried out a full doping study, our preliminary data indicates that near-optimal or underdoped samples have a smaller splitting effect. This smaller splitting, as well as the broader peaks associated with less heavy doping, make the detection of the splitting much more difficult, probably explaining why this effect has not been observed previously. We also note that it would be harder to argue for the existence of the splitting from the conventional EDCs because the lineshapes are poorly known. For the MDC’s which have only been used extensively in the past year or so, the lineshape is simple and additional peaks can be more readily seen, even if the peaks are not as sharp or splitting as large as it is in the data shown here. Additionally, much effort has been concentrated along either the $(0,0) \rightarrow (\pi,\pi)$ nodal line where the splitting vanishes, or near the $(\pi,0)$ point where the superstructure bands contaminate the data and give additional peaks. Hence for a clear determination of the bilayer splitting effect it is important to study the middle of the zone, as done here.

Also relevant to why it may not have been previously detected are the matrix element effects, which may
strengthen one of the peaks at the expense of the other. For example, for the data of figure 1 and 2, a large out-of-plane component of the photon polarization was used, which helped to enhance the already-weak B peak. The identical data taken with a more in-plane photon polarization shows a much weaker B peak, as shown in figure 3b and 3c, however the fitting result produces same amount of the splitting in energy (data not shown here). The major difference between the data with less grazing photon incidence angle (case in figure 3) and more grazing angle (case in figure 1 and 2) is the relative intensity of peak A and B which makes the feature B barely visible in figure 3b. Of course, the relative strength of A compared to B can also depend on other parameters, most notably the photon energy, for which oscillations in the relative magnitude of the branches may exist.

Although the bilayer splitting scenario is convincing, we note that there is increasing evidence for microscopic phase separation in BSCCO and related compounds, and such an explanation for the band splitting is hard to completely rule out [8]. However, if this is the explanation a couple of important restrictions can be made from this study: (1) The degree of phase separation should be larger for Bi2212 than Bi2201. (2) There would be metallic regions of two dominant types (possibly with other insulating regions) as the MDCs show two main peaks. (3) The angular width of each MDC peak is on the order of 1° HWHM or 0.04Å⁻¹, giving a mean free path for scattering $l = 1/\Delta k \sim 25A$ which sets the minimal domain size in the sample.

From our new data, we see that the B piece of FS matches the traditional hole-like piece of FS very well [9,10], while the A piece deviates in k-space significantly and approaches $M(\pi, 0)$. While an extrapolation is required, it appears that the A piece probably remains centered around $Y(\pi, \pi)$, i.e. it remains to be hole-like. However, it comes close enough to $(\pi, 0)$ that a very small amount of $k_z$ dispersion of this band could easily push it back and forth across $(\pi, 0)$, making it either electron-like or hole-like. Experimentally, $k_z$ is varied by changing photon energies, which also should affect the intensity ratio of the two components. Assuming a small amount of $k_z$ dispersion, this would give a very natural explanation for the recent controversy over the FS topology, in which the existence of an electron-like portion centered around $(0, 0)$ has been suggested [2,3] as well as denied [11,12].

Such a $k_z$ dispersion would come from a c-axis intercell coupling, and while this has yet to be directly observed, we note that the existence of the intracell coupling shown here makes the non-vanishing intercell coupling more plausible. Recent optical measurements on Tl-based cuprates do suggest the presence of this interlayer coupling [13]. Similar to the intracell coupling, symmetry arguments tell us that the intercell coupling should vanish along the $(0, 0) \rightarrow (\pi, 0)$ nodal line and should be largest near $(\pi, 0)$, precisely where they would need to be to resolve the controversy. However, how to connect above argument and Bi2201 FS topology at 33eV is still not clear to us.

In summary, we have shown that in the normal state of Bi2212 there is a doubling of the band structure. Near the middle of the zone the angular splitting is a little more than 2° and the energy splitting is roughly 75meV. The $k$-dependence of the splitting is consistent with that expected from bilayer splitting, and if we parameterize this, we obtain a intra-cell coupling $t_\perp$ of ~ 55meV. Another less likely origin for the splitting is the microscopic phase separation. Similar results on even more heavily overdoped samples have been independently obtained by the Stanford group as well [20]. We acknowledge sample preparation help from J. Koralek and beamline support from H. Hochst, S. Kellar and D.H. Lu. This work was supported by the NSF Career-DMR-9985492 and the DOE DE-FG03-00ER45809. SSRL and the ALS are operated by the DOE, Office of Basic Energy Sciences and the SRC is supported by the NSF.

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