Observation of Bogoliubov Band Hybridization in the Optimally Doped Trilayer $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$

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Using a laser-excited angle-resolved photoemission spectroscopy capable of bulk sensitive and high-energy resolution measurements, we reveal a new phenomenon of superconductors in the optimally doped trilayer $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$. We observe a hybridization of the Bogoliubov bands derived from the inner and outer CuO_2 planes with different magnitudes of energy gaps. Our data clearly exhibit the splitting of coherent peaks and the consequent enhancement of spectral gaps. These features are reproduced by model calculations, which indicate that the gap enhancement extends over a wide range of Fermi surface up to the antinode. The significant modulation of electron pairing uncovered here might be a crucial factor to achieve the highest critical temperature in the trilayer cuprates.

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The critical temperature of superconductivity (T_c) in cuprates is sensitive to the number of CuO_2 layers per unit cell. The magnitude of T_c reaches the maximum in trilayer compounds among the homogulous series [1,2]; notably, the highest T_c in cuprates is obtained in $HgBa_2Ca_2Cu_3O_{8+\delta}$(Hg1223), which is a trilayer compound. While the intriguing relation between T_c and the number of CuO_2 layers was found shortly after the discovery of cuprates, its mechanism has not been understood yet and still been under a huge debate. In solving this long-standing issue, it is particularly significant to clarify the electronic properties distinctive for the trilayer cuprates.

The angle-resolved photoemission spectroscopy (ARPES) is best suited for clarifying the electronic structure in matters, and it has been utilized a great deal for a study of cuprates, especially the Bi-based compounds, which are easy to cleave [3]. The ARPES results for the trilayer $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) are, however, very limited [4–8] due not only to difficulty in growing a crystal but also to speculation that the investigation of Bi2223 by ARPES would not have much superiority over that for other compounds. In fact, earlier works for Bi2223 [4–6] reported that the band structure and the d-wave superconducting gap are similar to those of the double-layer $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212).

Anomalous features in Bi2223 have been uncovered only recently by utilizing the low energy photons ($h\nu = 7$–11 eV) for ARPES, which allow bulk sensitive measurement. This compound exhibits multibands reminiscent of Bi2212. In Bi2223, however, the different bands show different magnitudes of the superconducting gap [7]. Thus the situation differs from that of Bi2212, which consists of bonding and antibonding bands with the same gaps. The multiple bands observed for Bi2223 are attributed to be the inner and outer planes with different carrier densities; Bi2223 is expected to have nonuniform distribution of charges over the triple CuO_2 layers [9–12] in contrast to Bi2212 with evenly doped double layers. The charge nonuniformity is common to the compounds with three or more layers, and it has been also confirmed for the four-layer $Ba_2Ca_3Cu_4O_8F_2$ [13,14], which exhibits superconductivity via interlayer self-doping [15].

The interaction between differently doped (under- and overdoped) layers with a high pairing energy and large phase stiffness has been proposed as a promising ingredient to raise T_c [16,17]. The intensive debate on it has been further stimulated by the recent discovery of the anomalous increase of T_c in Bi2223 induced by pressure above a critical value [18]. To uncover the nature of the interlayer interaction in multilayer systems, the correlation among the multibands should be identified. The previous studies by ARPES, however, have been limited to the investigation of each separated band, and thus the mutual relation among these bands has not been clarified.

The multiple gaps are also observed for other fascinating materials such as a conventional superconductor, MgB_2 [19,20], and unconventional superconductors, iron pnictides [21]. Nonetheless, the role of multiple gaps in achieving high T_c for these materials has not been fully understood. Bi2223 with extremely large gaps and rather simple band topology would provide an ideal platform to address these issues, and, moreover, to pave the way for designing materials with even higher T_c.

In this Letter, we reveal a new phenomenon of multilayer superconductors, Bogoliubov band hybridization, which enhances the spectral gaps in the optimally doped Bi2223. The gap enhancement is further confirmed via the observation of mode coupling in the band dispersion, which appears at an expected energy. Model calculations demonstrate that the hybridization is forbidden in bilayer cuprates and requires nontrivial electronic states with multiple gaps, which are realized in the trilayer cuprates owing to inherent nonuniform distribution of charges over layers. The significant modulation of energy gap induced by the hybridization might be a crucial factor for the particularly high T_c achieved in the trilayer cuprates.

Optimally doped $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ single crystals with $T_c = 108$ K were grown by the conventional floating-zone technique [22]. Magnetic susceptibilities for these crystals show sharp superconducting transitions with ∼4 K in width, indicative of a high quality [see the Supplemental Material, Fig. S1(c) [23]]. ARPES data were accumulated using a laboratory-based system consisting of a Scienta R4000 electron analyzer and a 6.994 eV laser [24]. The overall energy resolution in the ARPES experiment was set to ∼3 meV for all measurements. We have measured several pieces of samples, and confirmed the consistency in our results.

Figure 1(i) shows the Fermi surface mapping for optimally doped Bi2223. Two sheets of Fermi surface derived from the inner and outer CuO_2 layers [Fig. S1(b)] are observed as previously reported for the data measured with a synchrotron source [7,8]. We used the vertical polarization [$\theta = 90°$ in the Supplemental Material, Fig. S2(b) [23]], in which the ARPES intensities of both bands are equally strong. The dispersion maps are plotted in Figs. 1(a)–1(h) along several momentum cuts [arrows in Fig. 1(i)]. As expected for a d-wave superconductor, the energy gap opens off the node ($\phi = 0°$) and its value increases toward the zone edge ($\phi = 45°$). The splitting of bands for double outer layers is not observed, indicating that these two are almost degenerate.

The surprising new finding here is that band hybridization occurs in the superconducting state [see schematics in Figs. 1(j)–1(l)]; the bands with back bending cross with each other, which induces the band hybridization. A clear split-off of spectral intensities [a dashed circle in Fig. 3(c)] is detected, where the two bands cross [Figs. 1(d)–1(g)]. Significantly, this behavior is not observed in Bi2212 [25] with the orthogonal bonding and antibonding bands, likely because the bilayer splitting bands established in the normal state are not coupled further below T_c. The absence of Bogoliubov band hybridization in such a two-layer system is indeed confirmed by model calculations (see Supplemental Material [23]); it is shown that the hybridization could occur only when the gaps for the two split bands are different.

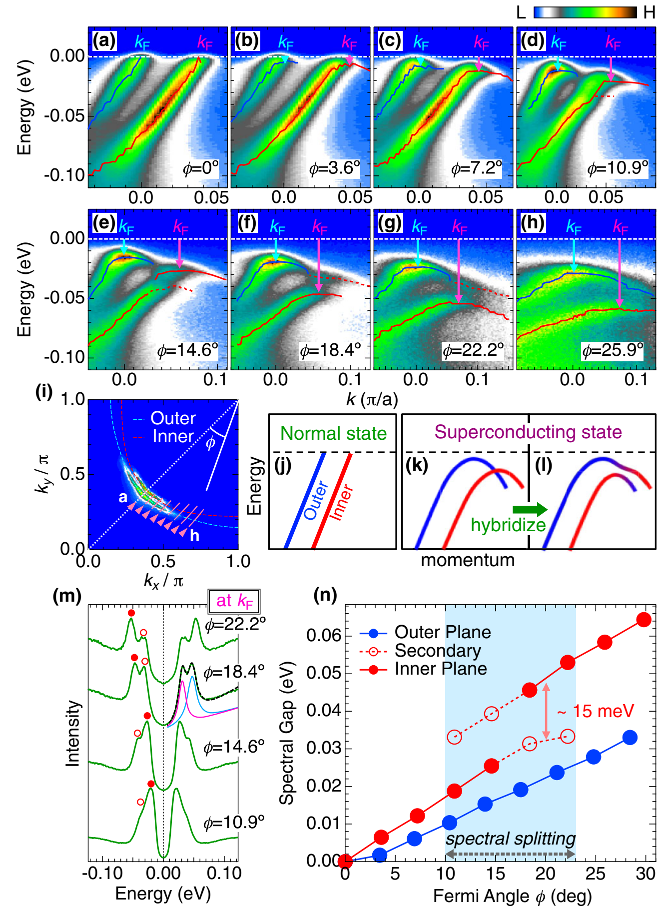

FIG. 1. Superconducting band hybridization and spectral splitting. (a)–(h) ARPES dispersions measured deep below T_c ($T = 10$ K) in directions indicated by arrows in (i). Band dispersions determined from peak energies of spectra (EDCs) are overlapped in each image. Dotted red curves trace the secondary peaks derived from split-off of spectral weight due to the superconducting hybridization. (i) Fermi surface mapping in the superconducting state. (j)–(l) Schematics illustrating superconductivity-induced hybridization: two bands, dispersing alongside in the normal state, cross with each other in the superconducting state, and hybridization occurs. (m) ARPES spectra at k_F of the inner band, extracted from (d)–(g). Double Lorentzian curves added by a slope for the secondary photoelectrons are fit to the spectrum for $\phi = 18.4°$. (n) Spectral gaps for the inner- and outer-plane bands around the node ($\phi = 0°$). Energy positions of the main and secondary coherent peaks, marked in (m), are plotted with open and filled red circles, respectively.

The distinguishing feature of Bi2223 is further presented in Fig. 1(m), where several spectra (energy distribution curves, EDCs) at k_F's for the inner-plane band are extracted from Figs. 1(d)–1(h). Here each curve is symmetrized about E_F to eliminate the Fermi cutoff: the EDC is flipped about E_F and added to the original one. We find that the dominant coherent peak [red filled circles in Fig. 1(m)] switches from one with a smaller gap to the other with a larger gap with increasing Fermi angle ϕ, likely depending on the degree of the hybridization. Consequently, an anomaly appears in the momentum dependence of the spectral gap. In Fig. 1(n), we plot the energies of dominant coherent peaks for the spectra at k_F's [red filled circles, or magenta arrows in Figs. 1(a)–1(h)]. While the gap value smoothly increases with ϕ in the close vicinity of the node ($\phi = 0°$), it abruptly jumps to a higher value around $\phi = 20°$ (hatched region) before further increasing toward the antinode. In Fig. 1(n), the energies of the secondary coherent peaks, marked in Fig. 1(m) by red open circles are also plotted. The hybridization gap between the Bogoliubov bands is estimated to be ∼15 meV [red arrow in Fig. 1(n)].

We emphasize here that the feature of the spectra discussed above clearly differs from the peak-dip-hump structure generated by mode couplings. The hump shape due to mode couplings should be broad with a typical width of more than 100 meV [26]. In Fig. 1(m) we demonstrate, for the spectrum at $\phi = 18.4°$, that two split peaks are both very sharp and have similar widths (14 and 18 meV). This is clearly against the mode-coupling scenario.

The expected kink behaviors due to the mode couplings are actually detected at higher binding energies as demonstrated in Fig. 2(a) for the data at $\phi = 18.4°$. We find, while the energy location for the kink is significantly different between the two dispersions, the bosonic energies (marked by arrows) are estimated to be almost the same (∼40 meV). For comparison, we have also observed the ARPES dispersion for Bi2212 along the similar momentum cut [Fig. 2(b)]. The bosonic energy obtained is very similar to that of Bi2223 [27,28], as previously argued with the antinodal spectra [29]. Our results ensure that the enhancement of energy gaps due to the Bogoliubov band hybridization is intrinsic to Bi2223; if the gap enhancement is not a real effect, the kink structure due to the mode coupling to the bosons of ∼40 meV should appear at a lower binding energy.

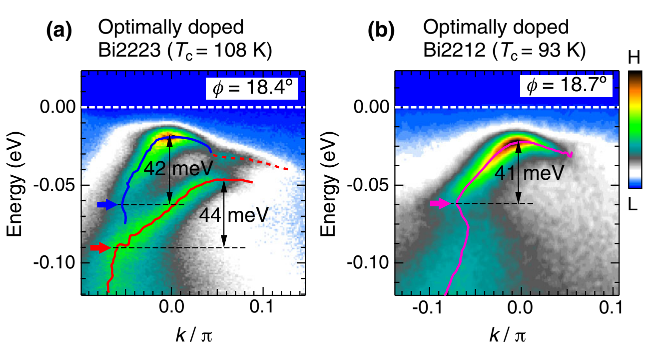

FIG. 2. The signature of mode coupling in the band with the gap enhanced by the superconducting hybridization. (a),(b) The ARPES images and the band dispersion determined from the peak positions of spectra measured at $T = 10$ K for the optimally doped Bi2223 and Bi2212, respectively. The arrows indicate the energy scale of bosons, which couple to the superconducting band dispersions. The comparable values (∼40 meV) are estimated for all bands in Bi2223 and also in Bi2212.

In Fig. 3, we examine the ARPES dispersion at elevated temperatures across T_c, and confirm that the band hybridization is indeed induced by the superconducting transition. The typical data along the momentum cut at $\phi = 17°$ are exhibited in Figs. 3(c) and 3(d), where the ARPES dispersions deep below T_c ($T = 10$ K) and just at T_c are compared. We find that the split-off of spectral intensities due to the band hybridization [a dashed circle in Fig. 3(c)] disappears above T_c, leaving only two parallel dispersions [see Figs. 3(a) and 3(b)]. The spectra extracted at k_F for the inner-plane band [Fig. 3(e)] demonstrates that two split peaks seen at low temperatures merge to one peak around T_c.

Our high quality data reveal that the energy gap close to the node stays open even above T_c [see Figs. 3(e) and 3(f)]. This contrasts to the previous claim with synchrotron data that Fermi arcs (disconnected segments of gapless Fermi

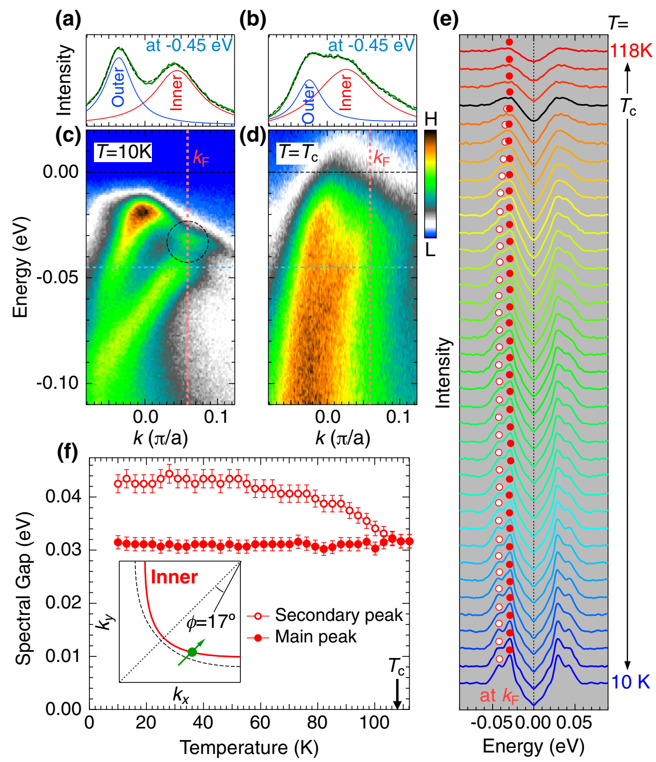

FIG. 3. Temperature evolution of spectral splitting across T_c. (a),(b) Momentum distribution curves at −0.45 eV along light blue dotted lines in (c) and (d), respectively. Two Lorentzian functions are fit to the data. (c),(d) Band dispersion maps measured along a green arrow in the inset of (f) at $T = 10$ K and $T = T_c$, respectively. Dashed circle in (c) marks spectral split-off due to band hybridization. (e) Temperature scan of symmetrized EDCs at k_F of the inner-plane band [magenta dashed lines in (c) and (b)] from 10 up to 118 K with a 3 K step. (f) Temperature evolution of spectral gaps estimated from energy positions of two split peaks: the main and secondary peaks [filled and open red circles in (e), respectively].

surface) abruptly emerge just above T_c [8]. We stress that difficulty in unveiling the properties of energy gap close to the node comes from the small magnitude. The laser ARPES is best suited for overcoming it, owing to an ultrahigh-energy resolution and bulk sensitivity, both of which are required to obtain sharp peaks in the ARPES spectra [30]. To determine the gap closing temperature, however, more careful analysis with spectral weight near E_F would be necessary.

In order to fully understand our ARPES data, we have conducted model calculations for the trilayer systems (Fig. 4). We found that the essential ingredients to develop the Bogoliubov band hybridization are the following two: the interlayer electron hopping and the difference of superconducting order parameters between the inner and outer layers (see the Supplemental Material for more details [23]). The latter is characteristic to three or more layered cuprates, which show a nonuniform carrier distribution.

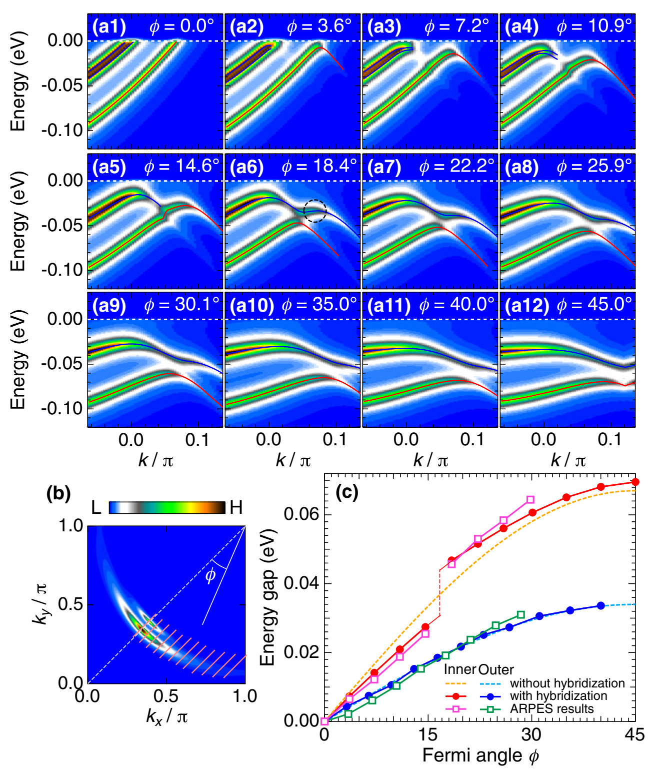

FIG. 4. Model calculations for the band structure of Bi2223, which reproduce the ARPES data exhibiting the effect of superconducting band hybridization. (a1)–(a12) Each image plots the dispersion of spectral intensities extracted along magenta lines marked in (b). The ϕ noted on the panels indicates the Fermi angle for the inner-layer band. The red and blue curves trace band dispersions determined from the spectral peaks. (b) Fermi surface mapping for the calculations. (c) Energy gaps for the bands in (a) determined from the spectral peaks. The original gaps without interlayer hopping are indicated with dashed curves for both the inner- and outer-plane bands. The ARPES results for gaps are also plotted with open squares.

Furthermore, the double outer layers form two bands, thus the back-folded Bogoliubov bands could be considerable in spectral intensity, even though these are quickly diminished off the k_F point. These unusual situations fulfilled in the trilayer cuprates cause significant hybridization.

Figure 4 plots the results of the model calculations (see Supplemental Material for details [23]). Along the node, three split bands are visible. However, the upper two bands are almost degenerate, originating from the two outer layers, thus the splitting is very small in energy. This is compatible with our ARPES data, which shows no indication of the splitting. With leaving the node, the superconducting gap opens and the back folding of dispersions occurs across k_F. While the hybridization among the bands is negligible at small angles ϕ's, it becomes significant toward the antinode, since the back folding of the Bogoliubov band gets enhanced with an increase of gap magnitude. The clear split-off of spectral intensities, similar to the ARPES results, is obtained around $\phi = 18°$ [dotted circle in Fig. 4(a6)]. At the larger ϕ's, the well-defined hybridized bands (red and blue curves) are eventually generated, and the gap enhancement occurs for one of the bands (red curve). As in the ARPES data, the islandlike spectral intensities get unclear with approaching the antinode. However, the simulations indicate that the effect of hybridization persists up to the antinode ($\phi = 45°$), and it causes the enhancement of gap over a wide range of the Fermi surface.

The momentum variation of energy gaps for the hybridized bands are summarized in Fig. 4(c). For comparison, the original gaps before the hybridization and the ARPES results are also plotted. Our model calculations almost perfectly reproduce the ARPES results showing a discontinuous momentum variation of energy gap and the gap enhancement toward the antinode, which thus justify our conclusion. It is also indicated that, while the energy gap close to the node is slightly reduced via the band hybridization, the momentum range for it is more limited than the range where the gap enhancement occurs.

Our results show that the electron pairing of Bi2223 is significantly modulated by the superconducting hybridization, resulting in the enhancement of gaps for one of the bands. It, therefore, would be reasonable to expect its connection with the high T_c of Bi2223. In order to consider this connection, it is interesting to see the case of MgB_2, which shows multiple gaps and the highest T_c among the conventional superconductors. In this compound, only one of two bands (σ band) develops the gap fulfilling the BCS condition ($2\Delta_\pi/k_BT_c = 3.54$), whereas the other band (π band) does not with a smaller gap ($2\Delta_\sigma/k_BT_c = 1.42$). This fact implies that the superconductivity could be triggered by the band with the largest gap among the multiple bands. Likewise, if it is the inner-layer band that dominates the superconductivity in Bi2223, the enhancement of the inner gap will raise T_c. This argument would be supported by the unique phase diagram of Bi2223, which shows negligible reduction of T_c with overdoping [31]; namely, the inner layer is likely protected against a carrier doping, keeping its energy gap large and, consequently, T_c unchanged. To prove this, future works are desired.

In conclusion, we observe the Bogoliubov band hybridization for the first time in the trilayer Bi2223 by using a laser-ARPES. Model calculations demonstrate that the difference in gap magnitude among bands is the requisite condition to develop the hybridization. This situation is inherent to the trilayer compounds, thus the same hybridization is also expected to occur in Hg1223 with the highest T_c among cuprates. Our results indicate that the energy gap is increased extensively in the momentum region up to the antinode by the hybridization. The gap enhancement, while observed only for one band, might be effective to raise the T_c of Bi2223, in analogy with MgB_2, where the magnitude of T_c is likely determined by one of the two bands with the larger gap. The increase of gap due to the hybridization could be ~10 meV, which is comparable to the difference of gap magnitude between the over- and optimally doped Bi2212 with T_c of 72 and 95 K, respectively [32]. Thus, the hybridization effect might be significant enough to explain the higher T_c in Bi2223 ($T_c = 108$) than Bi2212 ($T_c = 95$ K). Our results may also be relevant to the mechanism for the anomalous increase of T_c by pressure recently discovered for Bi2223 [18], since the Bogoliubov band hybridization would be sensitive to the interlayer distance in a crystal.

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