Low Energy Excitation and Scaling in $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$ ($n = 1$–3): Angle-Resolved Photoemission Spectroscopy

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Angle-resolved photoemission spectroscopy (ARPES) has been performed on the single- to triple-layered Bi-family high-T_c superconductors ($Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$, $n = 1$–3). We found a sharp coherent peak as well as a pseudogap at the Fermi level in the triple-layered compound. Comparison among three compounds has revealed a universal rule that the characteristic energies of superconducting and pseudogap behaviors are scaled with the maximum T_c.

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Over a decade, angle-resolved photoemission spectroscopy (ARPES) has played an important role in developing our understanding of high-T_c superconductors (HTSCs). Recent remarkable progress in energy and momentum resolutions of ARPES enables a direct observation of low energy excitation in HTSCs which dominate the superconductivity. The superfluid density [1,2] is discussed in terms of the temperature and doping dependence of the weight of the coherent peak. ARPES line shape at (π, 0) and a characteristic kink in dispersion have been interpreted as an interaction of electrons with (π, π) magnetic mode [3,4] and that with longitudinal optical (LO) phonon [5] . The lifetime of excitation deduced from ARPES has been found to have a direct relation to the optical conductivity [6] and the electrical resistivity [7] and has been discussed in relation to the quantum critical behavior [7]. However, it is unknown at present whether these characteristic features of the low energy excitation are universal to all HTSCs, since a majority of ARPES studies have been done on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) which has CuO_2 bilayers. It is thus important to perform a systematic ARPES study on HTSCs with different numbers of CuO_2 layers in a unit cell to find universal properties in HTSCs. Although Bi-family HTSCs ($Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$, $n = 1$–3) look the most suitable to meet this aim, triple-layered $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) has been scarcely investigated by ARPES [8,9] because of difficulty in growing a high-quality single crystal.

In this Letter, we report a systematic high-resolution ARPES study on single- (Bi2201), double- (Bi2212), and triple- (Bi2223) layered Bi-family HTSCs to investigate the universality of the low energy exitation as well as to find a possible scaling rule in physical properties. By performing ARPES on Bi2223 and comparing the result with the data of Bi2201 and Bi2212, we now address several important issues about (i) appearance of coherent peak, (ii) interaction of electrons with modes, (iii) origin of pseudogap, and (iv) energy scale which dominates the superconducting and pseudogap properties.

High-quality single crystals of $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ ($T_c = 108$ K, nearly optimally doping) were grown by the traveling solvent floating zone (TSFZ) method with a very slow growth rate and a steep temperature gradient [10]. The oxygen content δ was controlled by the temperature of postannealing and/or the oxygen partial pressure. The sharp x-ray diffraction pattern and the steep superconducting transition ($\Delta T < 3$ K) in the dc susceptibility confirm the high quality of crystal. The typical size of crystal for ARPES measurement is $4 \times 2 \times 0.1$ mm^3. ARPES measurements were performed using a SCIENTA SES-200 spectrometer with a high-flux discharge lamp and a toroidal grating monochromator. We used the He Iα (21.218 eV) resonance line to excite photoelectrons. The energy and angular (momentum) resolutions were set at 9 meV and 0.2° (0.005 Å^{-1}), respectively. Clean surface for ARPES measurement was obtained by *in situ* cleaving in an ultrahigh vacuum of 4×10^{-11} Torr. The Fermi level (E_F) of the sample was referenced to a gold film evaporated onto the sample substrate. ARPES spectra of Bi2201 and Bi2212 were obtained with the same ARPES spectrometer under essentially the same experimental conditions [11].

Figure 1 shows ARPES spectra of Bi2223 ($T_c = 108$ K) measured at 40 K along (π, 0)-(π, π) cut in the Brillouin zone. We clearly find a sharp peak at 40–50 meV assigned to the superconducting coherent peak, together with a broad hump structure at 110–150 meV in the spectra. It is thus evident that the ''hump-dip-peak'' structure exists also in a triple-layered HTSC Bi2223, as has been observed by the previous angle-integrated photoemission spectroscopy (AIPES) by Gu *et al.* [9]. As found in Fig. 1, the hump structure shows a remarkable systematic dispersion and the coherent peak looks to follow it although the magnitude is considerably weak. Similar close correlation between the dispersion of coherent peak and hump is also seen in Bi2212 [4]. However, there are some quantitative differences between Bi2223 and Bi2212. Importantly, the hump-dip-peak structure is clearly resolved even at k_F and the

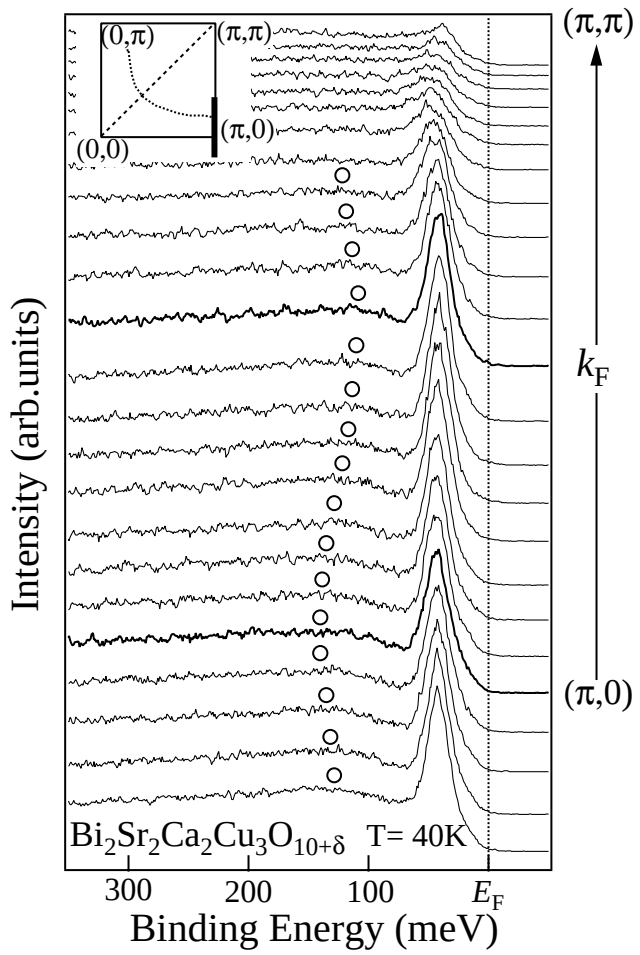

FIG. 1. ARPES spectra near E_F of $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (T_c = 108 K) measured at 40 K along (π, 0)-(π, π) cut shown by a thick line in the inset. The Fermi vector $k_F = (\pi, 0.12\pi)$ has been determined by the minimum-gap-locus method [12]. The energy position of the hump structure is denoted by open circles.

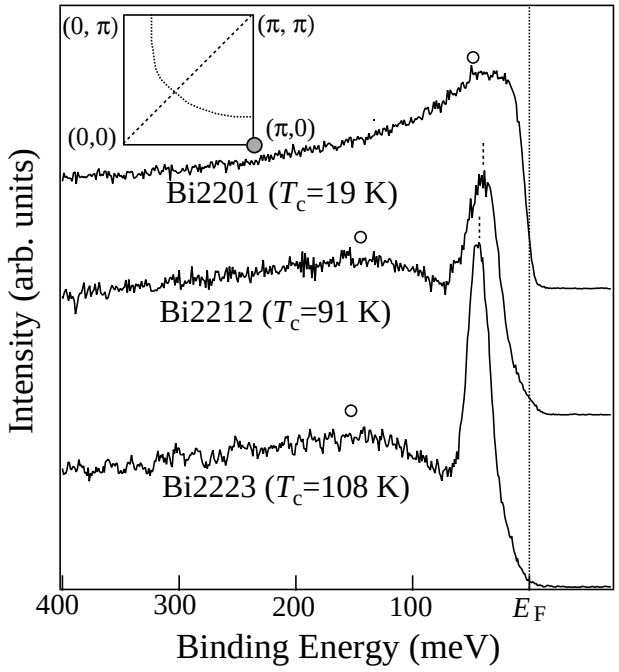

FIG. 2. Comparison of ARPES spectra at the superconducting state of Bi2201 (T_c = 19 K), Bi2212 (T_c = 91 K), and Bi2223 (T_c = 108 K) measured at (π, 0) point. Measurement temperatures are 13.5 K for Bi2201 and 40 K for both Bi2212 and Bi2223. Energy positions of hump and coherent peak are denoted by open circles and dashed lines, respectively.

hump structure disperses back to the high binding energy after passing k_F in Bi2223. This dispersing back behavior has not been clearly observed in optimally and overdoped Bi2212, while similar features are seen in underdoped Bi2212 [3,4]. This suggests that, in the optimally doped region, Bi2223 shows a stronger dispersing back behavior than Bi2212. In a simple model of electrons interacting with modes, a stronger coupling causes a stronger frequency (energy) dependence of electron self-energy [13]. In other words, the broad incoherent feature in ARPES spectrum (namely the hump) gains much intensity at the stronger interaction (coupling constant). Therefore, the present experimental result suggests a stronger interaction of electrons with modes in triple-layered Bi2223 than in bilayered Bi2212. At present there is no clear explanation for this layer-number dependence of interaction strength. A theoretical model to describe the relation of the coupling strength and the number of CuO_2 layers is necessary.

Figure 2 shows comparison of ARPES spectra in superconducting state measured at (π, 0) among 3 Bi-HTSCs at/near optimal doping [14]. The overall line shape of Bi2212 and Bi2223 looks almost identical to each other in the energy position of hump and coherent peak, as well as an intrinsic width of coherent peak (14–16 meV) [17], while that of Bi2201 is apparently different from the others, showing no sharp coherent peak nor the hump-dip-peak structure, although superconducting gap actually opens (leading-edge clearly shifts to high binding energy). This anomalous behavior, the absence of a sharp coherent peak in the single-layered Bi2201, may be explained in terms of a possible small c-axis superfluid density in Bi2201. However, the difference in the superfluid density between Bi2201 and Bi2212 observed by the microwave experiment [18] seems too small to account for the difference in the ARPES spectral line shape, suggesting the other mechanism to suppress the coherent peak.

Next, we discuss the temperature dependence of the coherent peak, superconducting gap and pseudogap. ARPES intensities of Bi2223 (T_c = 108 K) along (π, 0)-(π, π) cut at T = 40–125 K are plotted in Fig. 3(a). A pronounced narrow structure at 40 K [Fig. 3(a) bottom] is assigned to the coherent peak. With increasing temperature, the peak gradually reduces its intensity without changing its position, and finally vanishes around 115 K. This is more clearly seen in the ARPES spectra at k_F

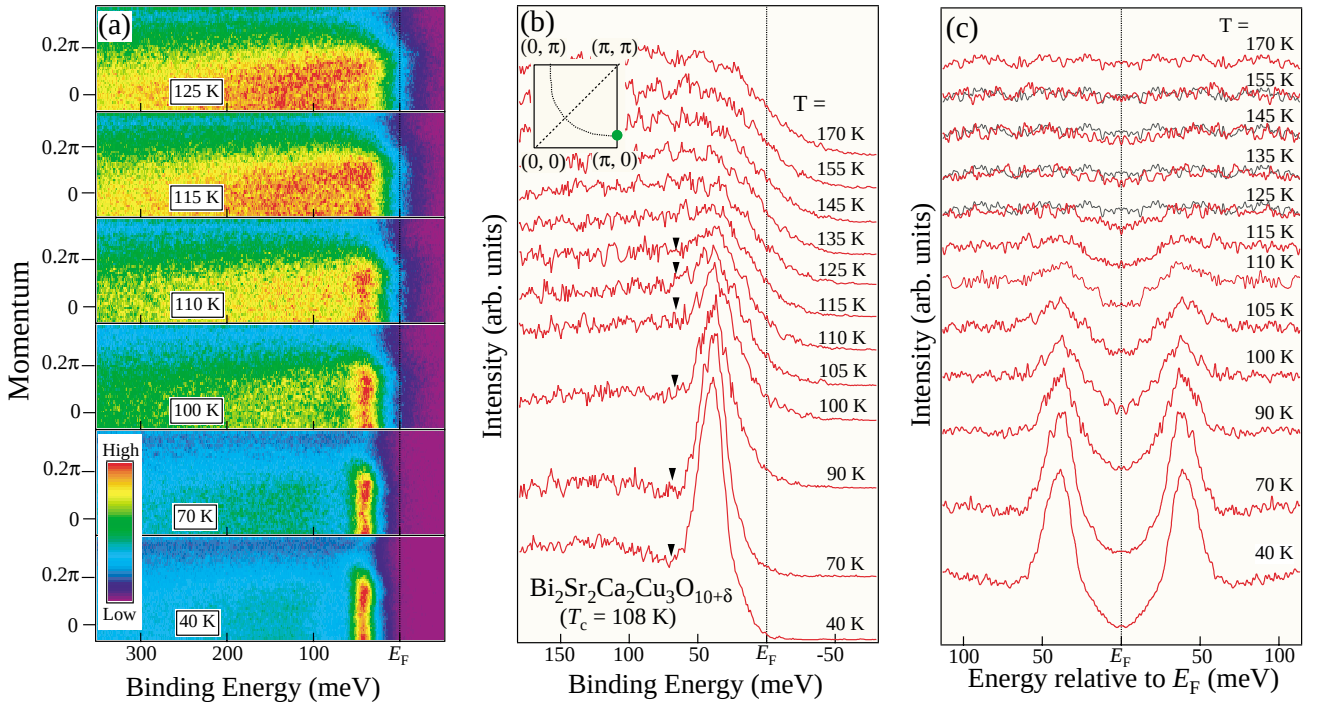

FIG. 3 (color). (a) Temperature dependence of ARPES intensity along (π, 0)-(π, π) cut in Bi2223. Vertical axis corresponds to the momentum along (π, 0)-(π, π) cut while the abscissa shows the binding energy relative to E_F. Intensity is normalized to the peak maximum at each temperature. (b) Temperature dependence of ARPES spectra of Bi2223 at (π, 0)-(π, π) crossing. Intensity of spectra is normalized to the area under the curve. The energy position of spectral break is indicated by arrows. (c) Symmetrized ARPES spectra of Bi2223 at (π, 0)-(π, π) crossing. The 170-K spectrum (black line) is superimposed on each spectrum for comparison.

[Fig. 3(b)], where a spectral break (indicated by triangles) seen in the 110-K spectrum disappears in the 115-K spectrum. The survival of the peak at temperatures slightly above T_c is understood in terms of superconducting fluctuation, as in Bi2212 ($T_c = 91$ K) where the peak at (π, 0) vanishes around 103 K [19]. We thus conclude that the temperature-dependent evolution of the peak is generic to double- and triple-layered HTSCs.

A definite evidence for opening of a pseudogap above T_c in Bi2223 is demonstrated in Fig. 3(c), where ARPES spectra at (π, 0)-(π, π) crossing [Fig. 3(b)] are symmetrized with respect to E_F [12]. We find that the superconducting gap smoothly evolves into the pseudogap above T_c (108 K). It is also found that the energy scale of pseudogap is same as that of superconducting gap, and the pseudogap gradually "fills in" rather than "closes" with increasing temperature. These behaviors are similar to that observed in Bi2212 [19–21], indicating that pseudogaps in Bi2212 and Bi2223 have a same origin, most likely a precursor pairing. In order to estimate T^* at which the pseudogap closes, we have superimposed the 170-K spectrum on the 125–155 K spectra. We find that the spectra at 125–145 K clearly show a deviation from the 170-K spectrum near E_F, while the spectrum at 155 K appears to coincide well with that at 170 K. This suggests that the pseudogap closes around $T^* \sim 150$ K, consistent with the inplane resistivity which shows a deviation from the T-linear dependence around 140–190 K [22].

In Fig. 4, we show key superconducting- and pseudogap energy scales as a function of maximum T_c (T_c at optimal doping) or the number of CuO_2 layers in a unit cell. The hump energy is regarded as a measure of a large pseudogap [3] and the values of T^* are from the ARPES and the in-plane-resistivity measurements [22–25]. The energy posi-

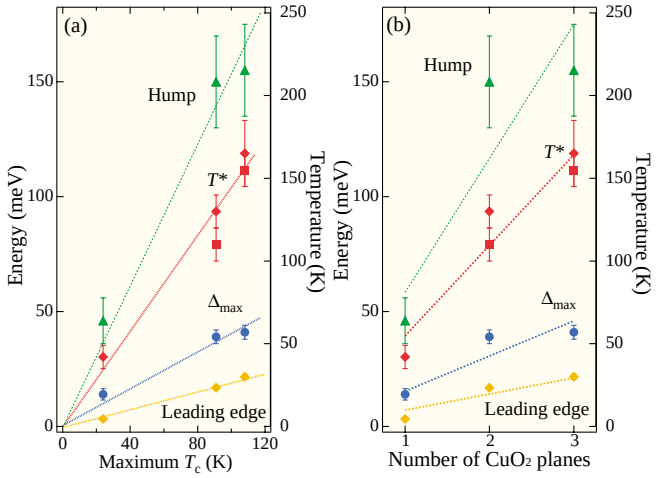

FIG. 4 (color). (a) Size of superconducting gap Δ_{max}, leading-edge shift at superconducting state, the binding energy of hump at (π, 0), and T^* as a function of maximum T_c. Straight lines, obtained by least-square fit, are guides for the eyes. (b) Same as a function of the number of CuO_2 layers in a unit cell.

tion of the hump has been determined by fitting using single Lorentzian around the peak top. $\Delta_{\max}$ is estimated from the fitting by the BCS spectral function multiplied by the Fermi-Dirac function and convoluted with the energy resolution [11,26]. We find that all the energies are well on a straight line with respect to T_c, but not to the number of CuO_2 layers (n). This clearly indicates that the key superconducting and pseudogap properties of HTSCs are well scaled with the maximum T_c. Furthermore, the scaling between the superconducting gap and pseudogap energy suggests that both are closely related. We note that this result contradicts with previous tunneling and AIPES works [9,27] which reports that the superconducting gap size scales with n. The reason is unclear at present but we speculate that the contradiction in part originates in the difference of the levels of information between ARPES and AIPES (or density of states). It would be necessary to solve this issue in future.

We now discuss the implications for the scaling rule. NMR study by Julien *et al.* has suggested a higher value of effective super exchange interaction J_{eff} in triple-layered $HgBa_2Ca_2Cu_3O_{8+\delta}$ (HBCO, $T_c = 121$ K) than in bilayer Y123, from their analysis of hyperfine coupling constant B [28]. Recent scanning tunneling spectroscopy and Raman spectroscopy have shown that the ratio of superconducting gap as well as T^* between single-layered $La_{2-x}Sr_xCuO_4$ (LSCO) and bilayer Bi2212 is same as that of maximum T_c (scaling factor $91/38 \sim 2.4$), and the scaling factor shows a striking agreement with the ratio of J_{eff} (measured by uniform magnetic susceptability) between LSCO and Bi2212 [29]. Considering these experimental results together with the scaling behavior found in the present ARPES study, we conclude that the key superconducting and pseudogap properties are closely related to the antiferromagnetism in HTSCs and the superconductivity is most likely due to spins in CuO_2 layers.

In conclusion, we have performed angle-resolved photoemission spectroscopy of triple-layered high-T_c superconductor $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ ($T_c = 108$ K). We have found (i) a sharp coherent peak which disappears slightly above T_c (115 K), (ii) a hump-dip-peak structure at $(\pi, 0)$ in superconducting state, and (iii) a pseudogap above T_c ($T^* = 150$ K) which smoothly evolves into superconducting gap. By a comprehensive comparison of Bi-family high-T_c superconductors ($Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$, $n =$ 1–3), we found a universal rule that key energies which dominate superconducting- and pseudogap properties are well scaled with maximum T_c.

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During the preparation of this manuscript, three papers have come to our attention, in Refs. [30, 31, 32].

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[1] D. L. Feng *et al.*, Science **289**, 277 (2000).
[2] H. Ding *et al.*, Phys. Rev. Lett. **87**, 227001 (2001).
[3] J. C. Campuzano *et al.*, Phys. Rev. Lett. **83**, 3709 (1999).
[4] A. Kaminski *et al.*, Phys. Rev. Lett. **86**, 1070 (2001).
[5] A. Lanzara *et al.*, Nature (London) **412**, 510 (2001).
[6] A. Kaminski *et al.*, Phys. Rev. Lett. **84**, 1788 (2000).
[7] T. Valla *et al.*, Science **285**, 2110 (1999).
[8] C. Janowitz *et al.*, Physica (Amsterdam) **259B–261B**, 1134 (1999).
[9] C. Gu *et al.*, Phys. Rev. B **51**, 1397 (1995).
[10] T. Fujii, T. Watanabe, and A. Matsuda, J. Cryst. Growth **223**, 175 (2001).
[11] T. Sato *et al.*, Phys. Rev. B **63**, 132502 (2001).
[12] M. R. Norman *et al.*, Nature (London) **392**, 157 (1998).
[13] R. D. Parks, *Superconductivity* (Dekker, New York, 1969).
[14] Doping levels of Bi2201 (Pb substituted) and Bi2212 are estimated to be $x = 0.12$ and 0.16, respectively (Ref. [15]). We note that discussion of the maximum T_c on Pb-substituted Bi2201 is still lively, since recent work by Jianwu *et al.* (Ref. [16]) reports a maximum T_c which exceeds 40 K while the present work uses the sample with that of 22–23 K. To settle this issue would be a future work.
[15] M. R. Pressland *et al.*, Physica (Amsterdam) **176C**, 95 (1991).
[16] Z. Jianwu *et al.*, Supercond. Sci. Tech. **14**, 599 (2001).
[17] The difference in the intensity of the coherent peak between Bi2223 and Bi2212 may be accounted for by the difference in the magnitude of c-axis superfluid density.
[18] M. B. Gaifullin *et al.*, Phys. Rev. Lett. **83**, 3928 (1999).
[19] A. V. Fedorov *et al.*, Phys. Rev. Lett. **82**, 2179 (1999).
[20] A. G. Loeser *et al.*, Science **273**, 325 (1996).
[21] H. Ding *et al.*, Nature (London) **382**, 51 (1996).
[22] T. Fujii, T. Watanabe, and A. Matsuda, cond-mat/0205121.
[23] I. Chong *et al.*, Physica (Amsterdam) **290C**, 57 (1997).
[24] T. Watanabe, T. Fujii, and A. Matsuda, Phys. Rev. Lett. **79**, 2113 (1997).
[25] T. Sato *et al.*, Physica (Amsterdam) **341C**, 815 (2000).
[26] H. Ding *et al.*, Phys. Rev. Lett. **74**, 2784 (1995).
[27] Y. G. Ponomarev *et al.*, Conf. Ser. Inst. Phys. **167**, 2 (2000).
[28] M.-H. Julien *et al.*, Phys. Rev. Lett. **76**, 4238 (1996).
[29] T. Matsuzaki *et al.*, J. Phys. Chem. Solids **62**, 29 (2001).
[30] C. Janowitz *et al.*, Physica (Amsterdam) **364C–365C**, 600 (2001).
[31] R. Müller *et al.*, J. Supercond. **15**, 147 (2002).
[32] D. L. Feng *et al.*, Phys. Rev. Lett. **88**, 107001 (2002).