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# High-resolution angle-resolved photoemission study of Pb-substituted Bi2201

T. Sato<sup>a,\*</sup>, Y. Naitoh<sup>a</sup>, T. Kamiyama<sup>a</sup>, T. Takahashi<sup>a</sup>, T. Yokoya<sup>b</sup>, J. Mesot<sup>c</sup>,  
A. Kaminski<sup>d</sup>, H. Fretwell<sup>d</sup>, J.C. Campuzano<sup>c,d</sup>, H. Ding<sup>e</sup>, I. Chong<sup>f</sup>, T. Terashima<sup>f</sup>,  
M. Takano<sup>f</sup>, K. Kadowaki<sup>g</sup>

<sup>a</sup>Department of Physics, Tohoku University, Sendai 980-8578, Japan

<sup>b</sup>Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan

<sup>c</sup>Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA

<sup>d</sup>Department of Physics, University of Illinois at Chicago, Chicago, IL 60607, USA

<sup>e</sup>Department of Physics, Boston College, Chestnut Hill, MA 02467, USA

<sup>f</sup>Institute for Chemical Research, Kyoto University, Uji 611-0011, Japan

<sup>g</sup>Institute of Materials Science, University of Tsukuba, Ibaraki 305-3573, Japan

## Abstract

We have performed a systematic doping-dependent angle-resolved photoemission spectroscopy (ARPES) of Pb-substituted Bi2201 ( $\text{Bi}_{1.80}\text{Pb}_{0.38}\text{Sr}_{2.01}\text{CuO}_{6-\delta}$ ) with high energy (7–11 meV) and momentum ( $0.01 \text{ \AA}^{-1}$ ) resolutions. We found that all the samples show a large hole-like Fermi surface centered at  $(\pi, \pi)$ , whose volume changes systematically as a function of doping. The superconducting gap exhibits a  $d_{x^2-y^2}$ -like anisotropy with a typical gap value of 10–15 meV near  $(\pi, 0)$ . By comparing with  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Bi2212), we found that the size of superconducting gap is well scaled with the maximum  $T_c$  ( $T_c^{\text{max}}$ ). Doping-dependence of spectral line shape at  $(\pi, 0)$  is very similar between Bi2201 and Bi2212, while its energy scale is different by a factor of 4, corresponding to the ratio of  $T_c^{\text{max}}$  between the two compounds. These indicate that the superconducting and pseudogap properties are characterized by  $T_c^{\text{max}}$  irrespective of the number of  $\text{CuO}_2$  layers or the BiO superstructure. © 2000 Elsevier Science Ltd. All rights reserved.

**Keywords:** A. Oxides; C. Photoelectron spectroscopy; D. Superconductivity

## 1. Introduction

One of the most important features in cuprate high- $T_c$  superconductors (HTSCs) revealed by angle-resolved photoemission spectroscopy (ARPES) is the  $d_{x^2-y^2}$ -like superconducting order parameter [1–3]. Intensive temperature-dependent ARPES studies in underdoped region have revealed the existence of pseudogap above the superconducting transition temperature ( $T_c$ ) [4–7]. Detailed analysis of the ARPES spectra has elucidated the interaction of electrons with a collective mode of wave vector  $(\pi, \pi)$  [8]. These findings have been regarded as key features of the HTSCs to approach the high  $T_c$  mechanism. On the other hand, a recent ARPES study on Bi-system HTSCs using higher photon

energies has raised a question to the so-far believed hole-like Fermi surface (FS) in HTSCs, proposing an electron-like FS centered at  $(0, 0)$  point [9]. It is well known that  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Bi2212) has double  $\text{CuO}_2$  layers per unit cell and also a strong incommensurate modulation in the BiO layer. It is still unknown how the double  $\text{CuO}_2$  layer affects the electronic structure and consequently the superconducting properties. Further, the incommensurate superstructure in the BiO layer complicates the interpretation of ARPES spectra by creating additional spurious “ghost bands” [9–11]. In contrast to Bi2212,  $\text{Bi}_2\text{Sr}_2\text{CuO}_6$  (Bi2201) has a single  $\text{CuO}_2$  layer per unit cell though a slight modulation in the BiO layer still remains. It has been found by a recent structural study that substitution of Bi with Pb completely erases the superstructure in the Bi(Pb)O layer [12]. It is thus very important to perform ARPES measurements on Pb-substituted Bi2201 to check the universality of

\* Corresponding author.

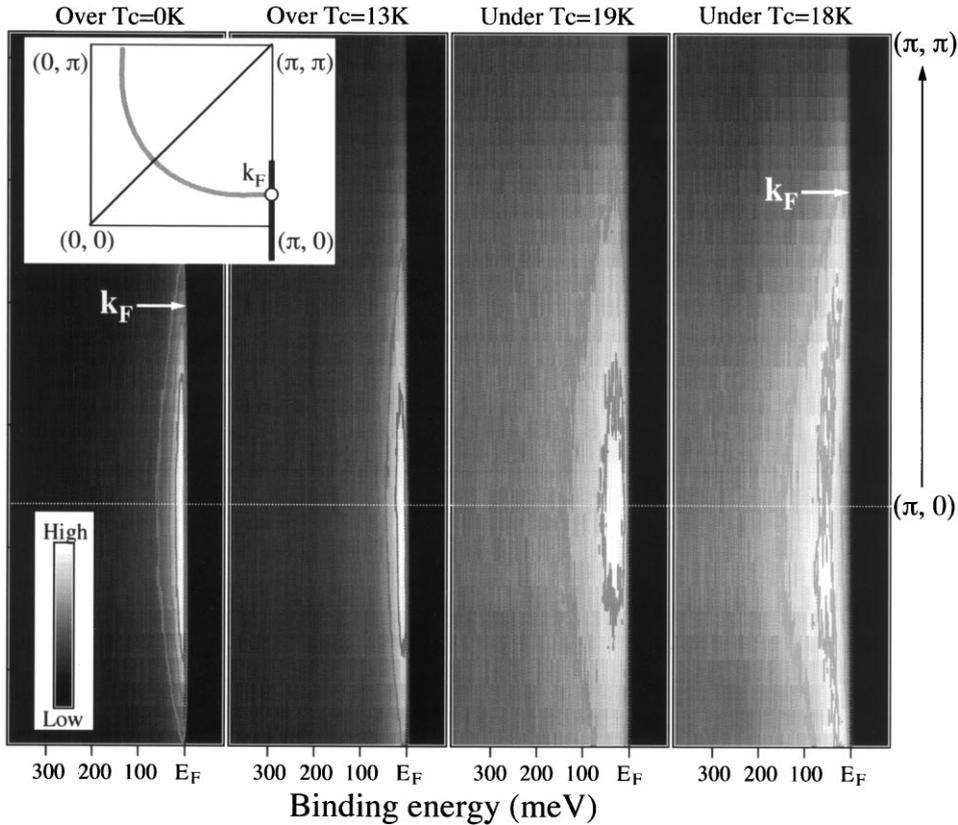


Fig. 1. ARPES intensity map of  $\text{Bi}_{1.80}\text{Pb}_{0.38}\text{Sr}_{2.01}\text{CuO}_{6-\delta}$  for four samples with different dopings (overdope  $T_c = 0, 13$  K to underdope  $T_c = 18, 19$  K). Bright area corresponds to the high ARPES intensity. Vertical axis corresponds to the momentum along  $(\pi, 0)$ – $(\pi, \pi)$  cut while the abscissa shows the binding energy relative to  $E_F$ . The Fermi momentum ( $k_F$ ) determined by the  $|\nabla \cdot \mathbf{k} \cdot n(\mathbf{k})|$  method [16] is shown by arrows for OD 0 K ( $k_F = 0.1\pi$ ) and UD 18 K ( $k_F = 0.15\pi$ ). Inset shows the Brillouin zone with a schematic Fermi surface obtained from the present measurement (gray line) and a part of  $(\pi, 0)$ – $(\pi, \pi)$  cut where ARPES measurements were done is shown with black line.

the key features observed so far in Bi2212 as well as to obtain an insight to the Fermi-surface topology free from the structural modulation.

In this paper, we report a systematic ultrahigh-resolution ARPES study on  $\text{Bi}_{1.80}\text{Pb}_{0.38}\text{Sr}_{2.01}\text{CuO}_{6-\delta}$  with various dopings (underdope (UD)  $T_c = 18, 19$  K to overdope (OD)  $T_c = 13, 0$  K). We found that the FS retains its hole-like character for all the doping levels from UD 18 K to OD 0 K. We have succeeded in observing the superconducting gap as well as the pseudogap above  $T_c$ . By comparing the present experimental results on Bi2201 with those on Bi2212, we found that the key superconducting properties observed so far in Bi2212 are universal in Bi-system HTSCs irrespective of the number of  $\text{CuO}_2$  layers or the structural modulation in the BiO layer, and their characteristic energies are well scaled with the maximum  $T_c$  of each compound.

## 2. Experimental

Single crystals of Pb-substituted Bi2201 ( $\text{Bi}_{1.80}\text{Pb}_{0.38}\text{Sr}_{2.01}\text{CuO}_{6-\delta}$ ) were grown by the traveling-solvent floating-zone

method [12]. We have confirmed by the transmission electron microscopy that the crystals are free from any structural modulations [12]. The doping level was controlled by annealing under vacuum at high temperature of 500–600°C. The  $T_c$  of samples was determined by the magnetic susceptibility measurement and is UD  $T_c = 18, 19$  K and OD  $T_c = 13, 0$  K, where 0 K means that the sample does not show any signature of superconductivity down to 4 K. 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 I $\alpha$  resonance line (21.218 eV) to excite photoelectrons. The energy and angular (momentum) resolutions were set at 7–11 meV and 0.25° (0.01 Å<sup>-1</sup>), respectively. The Fermi level ( $E_F$ ) of the sample was referenced to a gold film evaporated onto the sample substrate and its accuracy is estimated to be better than 0.4 meV.

## 3. Results and discussion

Fig. 1 shows the photoelectron intensity map along the

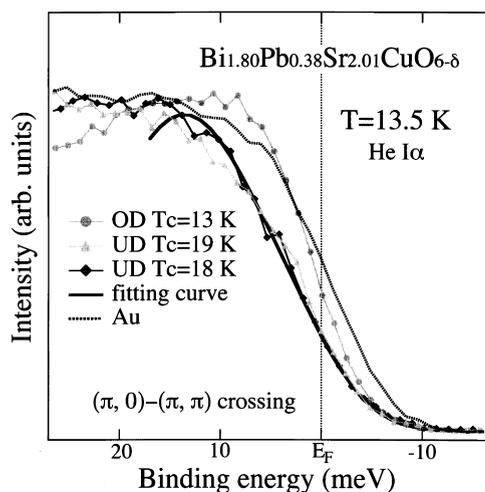


Fig. 2. ARPES spectra in the very vicinity of  $E_F$  at 13.5 K measured at each Fermi momentum ( $k_F$ ) along the  $(\pi, 0)$ – $(\pi, \pi)$  cut for superconducting compounds (UD 18, 19 K and OD 13 K), compared with a gold reference. A black line is the result of numerical simulation to estimate the superconducting gap.

$(\pi, 0)$ – $(\pi, \pi)$  cut at 13.5 K for four samples with different dopings. It is clear from Fig. 1 that there is a dispersive feature near  $E_F$  centered at  $(\pi, 0)$  which seems to cross  $E_F$  midway between  $(\pi, 0)$  and  $(\pi, \pi)$  for all the samples. This suggests the existence of a hole-like FS centered at  $(\pi, \pi)$  as shown in the inset, in good agreement with the previous reports [13–15]. It is noted here that there is no additional intensity due to a possible superstructure in the BiO layer in contrast with Bi2212 [10,11], showing that the present Pb-substituted Bi2201 crystals are actually free from the structural modulation. The Fermi momentum ( $k_F$ ) determined by the  $|\nabla \cdot \mathbf{k} \cdot \mathbf{n}(k)|$  method [16] is shown by arrows for OD 0 K ( $k_F = 0.1\pi$ ) and UD 18 K ( $k_F = 0.15\pi$ ).<sup>1</sup> We find that the Fermi momentum is gradually shifted toward  $(\pi, \pi)$  point when the doping level is decreased. The present result thus clearly shows that the volume of FS systematically changes with doping but the topology retains its hole-like character for all the doping levels from OD 0 K to UD 18 K.

Fig. 2 shows ARPES spectra in the very vicinity of  $E_F$  measured at each  $k_F$  point at 13.5 K for superconducting samples (UD 18, 19 K and OD 13 K), compared with that of gold reference. As seen in Fig. 2, the leading edge of ARPES spectrum is systematically shifted with doping. The leading edge of OD 13 K sample seems to almost coincide with that of gold with a very small positive (toward higher binding energy) shift of 1 meV. Since the measurement temperature (13.5 K) is almost the same as the  $T_c$  of the sample (13 K), this suggests that the superconducting gap is

almost closed around  $T_c$  or a small pseudogap may open above  $T_c$ . In contrast to OD 13 K, the ARPES spectra of UD 18 and 19 K show a remarkable leading-edge shift relative to the gold reference. Referring to the temperature dependence of the superconducting gap of Bi2212 [17], it is expected that an almost full superconducting gap opens at 13.5 K in these Bi2201 samples ( $T_c = 18$ –19 K) because an almost full gap opens at 60 K in Bi2212 sample with  $T_c = 90$  K [17].

In order to estimate the size of superconducting gap in the  $(\pi, 0)$ – $(\pi, \pi)$  cut, we have simulated the spectra with the BCS spectral function by taking into account the finite energy resolution and the finite temperature effect (Fermi–Dirac function at 13.5 K). The result is shown with black line in Fig. 2. We find that the leading edge of the ARPES spectrum is well fitted by the simulation, while the higher-binding energy region is not well reproduced probably because the incoherent part and the background in ARPES spectrum are not included in the simulation. The estimated gap size of UD 18 (19) K sample is  $14 \pm 1.5$  meV. This value is relatively small compared with a typical value for Bi2212 with the same hole concentration (40–45 meV) [18,19],<sup>2</sup> reflecting the difference in the  $T_c$ , namely the strength of pairing interaction. It is remarked here that the ratio of the gap size (40–45 meV/14 meV = 2.9–3.2) is similar to the ratio of the maximum  $T_c$  ( $T_c^{\max}$ ) for each compound (90 K (Bi2212)/23 K (Bi2201)  $\sim$  3.9). This suggests a certain scaling for the characteristic energy of the superconducting property.

Fig. 3 shows the doping dependence of ARPES spectrum at  $(\pi, 0)$  for Bi2201 and Bi2212 [8]. Detailed analysis of the ARPES spectra of Bi2212 in this momentum region has revealed the interaction of electrons with a collective mode. For Bi2212, we clearly find a “hump–dip–peak” structure produced through this interaction in all the spectra from UD 55 K to OD 72 K. We find a small hump-like structure around 50 meV in the spectra of UD 18 K and UD 19 K in Bi2201 as denoted by arrows. This hump-like structure may correspond to that around 200 meV in Bi2212. Further a quasiparticle-like peak appears at about 20 meV in the spectrum of UD 19 K as shown by a broken bar. It is thus inferred that the overall ARPES spectral feature near  $E_F$  and its doping dependence are quite similar between Bi2212 and Bi2201, although the energy scale is distinctly different between the two. The absence of a clear “hump–dip–peak” structure in Bi2201 may be due to (1) the relatively small energy scale in Bi2201 compared with that of Bi2212 and (2) the relatively “high” measurement temperature (13.5 K) compared with the  $T_c$  (18–19 K). When we compare the energy scale between Bi2212 and Bi2201 from the energy position of the hump-like structure in each ARPES spectrum (Bi2201 UD 19 K and Bi2212 UD 75 K), we obtain the ratio of 3.8 (= 190/50). Surprisingly,

<sup>1</sup> We have checked that the determined  $k_{FS}$  in the superconducting state are the same as the  $k$  points at which the leading edge shows a smallest gap, known as a minimum gap locus [7].

<sup>2</sup> We assumed a similar  $T_c$ -doping curve for Bi2212 and Bi2201 to estimate the hole concentration from the  $T_c$ s [18].

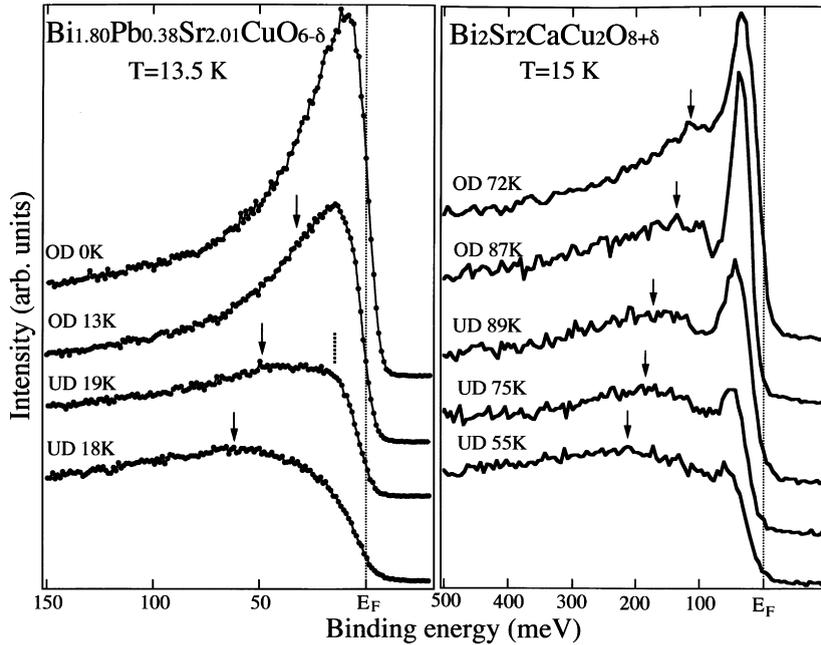


Fig. 3. Comparison of doping dependence of ARPES spectrum at  $(\pi,0)$  point between Bi2201 and Bi2212 [8]. Rough energy position of the hump structure is marked by arrows. Broken bar in the ARPES spectrum of Bi2201 UD 19 K shows a possible position of the quasiparticle peak.

this value is almost the same as that of the  $T_c^{\max}$  (90 K (Bi2212)/23 K (Bi2201) = 3.9), suggesting an energy scaling in the ARPES spectrum near  $E_F$  between Bi2201 and Bi2212.

We have measured the temperature dependence of ARPES spectrum and found that the leading-edge shift relative to  $E_F$  still remains even above 20 K in underdoped samples [20]. This suggests that a pseudogap opens in Bi2201 as in Bi2212 [4–7]. Although we could not observe the accurate temperature at which the pseudogap closes ( $T^*$ ) because of a relatively fast degradation of the sample surface at high temperatures, it is speculated that a similar  $T^*$ -doping relation stands in Bi2212 and Bi2201. In fact, the in-plane resistivity of optimally doped Bi2201 shows a deviation from  $T$ -linear behavior at 42 K [12] which is 1.9 times higher than the  $T_c$ , while that of optimally doped Bi2212 is 150 K which is 1.7 times higher than the  $T_c$  [21]. We find that these two values (1.9 and 1.7) are more or less the same. Since the in-plane resistivity is closely related to opening of the pseudogap, these experimental results suggest a similar  $T^*$ -doping phase diagram for both Bi2201 and Bi2212. Here we also find that the ratio of  $T^*$  deduced from the in-plane resistivity between Bi2212 and Bi2201 (150 K (Bi2212)/42 K (Bi2201)  $\sim$  3.6) is almost equal to that of  $T_c^{\max}$  ( $\sim$ 3.9). This suggests that the energy of pseudogap properties is also scaled with  $T_c^{\max}$ . This is supported by the experimental fact that the pseudogap is smoothly connected to the superconducting gap and is expected to have the same origin [6,15].

Fig. 4 summarizes the comparison of characteristic energies of the superconducting and pseudogap properties between Bi2201 and Bi2212. A straight line in the figure shows the ratio of  $T_c^{\max}$  of each group (90 K (Bi2212)/23 K (Bi2201) = 3.9). As seen in Fig. 4, the size of superconducting gap, the energy position of hump structure in ARPES spectrum, and  $T^*$  at which the

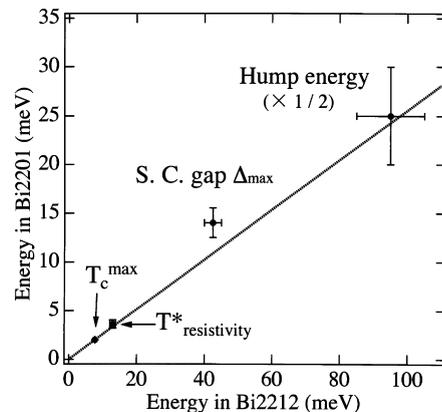


Fig. 4. Comparison of representative characteristic energies of superconducting and pseudogap properties (size of superconducting gap, energy position of hump structure in ARPES spectrum, and  $T^*$  at which pseudogap closes) between Bi2201 [12] and Bi2212 [19,21]. Straight lines show the ratio of  $T_c^{\max}$  of each compound (90 K (Bi2212)/23 K (Bi2201) = 3.9).

pseudogap closes are well scaled with  $T_c^{\max}$ . This clearly indicates that the superconducting and pseudogap properties are essentially the same in both Bi2212 and Bi2201 irrespective of the number of CuO<sub>2</sub> layers or the structural modulation in the BiO layer, and their characteristic energies are well scaled with the maximum  $T_c$ , namely the strength of pairing interaction.

#### 4. Conclusions

We have performed an ultrahigh-resolution ARPES study on superstructure-free Pb-substituted Bi2201 with various dopings. We found that the Fermi surface retains its hole-like character for all the doping levels. Superconducting gap with the size of 10–15 meV is observed in UD samples. The ARPES spectral feature near  $E_F$  (hump–dip–peak structure) and its doping dependence of Bi2201 are quite similar to those of Bi2212 while the energy scale is remarkably different. We found that the superconducting gap and pseudogap properties are well scaled with the maximum  $T_c$  of the respective compounds. All these indicate that key superconducting properties observed so far in Bi2212 are universal in Bi-system HTSCs irrespective of the number of CuO<sub>2</sub> layers or the structural modulation in the BiO layer.

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