# Angle-Resolved Photoemission Study on Strongly Correlated Electron Materials

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Strongly correlated electron materials has attracted more attentions in the last few decades because of their unusual and fascinating properties such as high- $T_c$  superconductivity, giant magnetoresistance, heavy fermion and so on. Those unique properties can offer a route toward the next-generation devices. We investigate the mechanism of the physical properties as well as the electronic structure of those materials by using angle-resolved photoemission spectroscopy (ARPES), a powerful tool in studying the electronic structure of complex materials, based on synchrotron radiation.



Figure 1. The superconducting (SC)-gap distribution on the hole FSs of K-Ba122 ( $x \sim 0.25$ ). (a1)-(b2) The photoemission intensities taken along two momentum cuts near the BZ center with 21 and 32 eV photons. (b) Temperature dependence of ARPES spectra on K-Ba122 ( $x \sim 0.25$ ) of  $k_{\rm F}$ position at the  $\Gamma$  point. (c1), (c2) EDCs of the  $\gamma$ band at  $k_{\rm F}$  position

divided by Fermi-Dirac (FD) distribution function for K-Ba122 ( $x \sim 0.25, 0.3$ ). (d1) and (d2) are the same as in panel (c1) and (c2) of the  $\alpha$  band. The gray circles in (c1)–(d2) are guides to the eyes for the value of the superconducting gap.

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# 1. Suppression of Superconducting Gap on $Ba_{1-x}K_xFe_2As_2$ Observed by Angle-Resolved Photoemission Spectroscopy

Iron-based superconductors have a complex phase diagram with the antiferromagnetic, structural, and superconducting (SC) transition phases as well as high- $T_c$  cuprates superconductors.<sup>1)</sup> Recently, nematicity, defined as broken rotational symmetry [a trigonal  $(C_4)$ -to-orthorhombic  $(C_2)$  structural transition], has shed light on the understanding of the mechanism on the iron-based superconductivity in the underdoped regime. In hole-doped BaFe<sub>2</sub>As<sub>2</sub> (Ba122) system, thermal expansion, specific heat, and neutron diffraction measurements of Ba<sub>1-x</sub>Na<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> and Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> (K- Ba122) at a certain hole concentration have revealed the magnetic order without  $C_4$  symmetry breaking in the underdoped regime.<sup>2)</sup> Besides, these materials show that the SC transition temperature  $(T_c)$  is suppressed between the SC phase and the  $C_4$ -magnetic phase by ~20-50%. Although these intriguing physical properties have been come into the open in the hole-doped Ba122 system universally, the microscopic mechanism of the  $C_4$ -magnetic phase and the  $T_{\rm c}$  suppression has been unclear yet. In this study, we performed ARPES study on underdoped K-Ba122 (x ~ 0.25,  $T_c$  ~ 26 K) with the C<sub>4</sub>-magnetic phase and  $T_c$  suppression, to elucidate the mechanism of superconductivity.

Figures 1(a1) and 1(a2) show the ARPES intensity plot as a function of energy and momentum taken at hv = 21 eV and 32 eV, corresponding to the  $\Gamma$  and Z points, respectively. We observe at least two hole bands for  $\Gamma$  using *s* polarization, indicating that the degenerated  $d_{yz}$ ,  $d_{xz}$  bands and the  $d_{xy}$  band are observed at the  $\Gamma$  point. Hereafter, hole bands at the BZ center are designated as  $\alpha$ ,  $\beta$ , and  $\gamma$  for the inner, middle, and outer hole FSs, respectively.

In order to elucidate the character of the SC gap on the hole FSs, Figure 1(b) shows the temperature dependence of the near- $E_{\rm F}$  ARPES spectrum measured across  $T_{\rm c}$  (~26 K) at Fermi momentum ( $k_{\rm F}$ ) for the  $\alpha$  band. The EDC shows a SC peak in the SC state (12 K) while that disappears in the normal state (50 K). To eliminate the effect of the Fermi-Dirac (FD) distribution function, we have symmetrized the ARPES spectra measured



Figure 2. The SC-gap distribution on the electron FSs of K-Ba122 ( $x \sim 0.25$ ). (a), (b) The photoemission intensities

taken at the momentum cut at the BZ corner with 23 eV photons below and above  $T_c$ , respectively. (c) Raw spectra of the data in (a) enclosed by a dotted line. (d): Temperature dependence of ARPES spectra for  $\varepsilon$  and  $\delta$ bands at  $k_F$ . (e), (f) Raw spectra at  $k_F$  and that divided by the Fermi-Dirac function on the  $\varepsilon$  band taken at 23 eV photons. (g), (h)  $k_z$  dependence of the spectra divided Fermi-Dirac function measured on the  $\varepsilon$  and  $\delta$ electron bands. (i), (j) The expansion of the spectra on the  $\varepsilon$  FS in the vicinity of  $E_F$  and the leading-edge midpoint for each EDC of (i), respectively. Measured  $k_z$ 's are taken at 17 eV–26 eV photons, corresponding to the  $k_z$  of the  $\Gamma$ -Z points shown in (k). The gray circles on the spectra are guides to the eyes for the peak position, reflecting SC gaps.

above and below  $T_c$  as shown in Figure 1(b). On decreasing temperature, the spectral weight near  $E_F$  is transferred to high binding energy below  $T_c$  and the symmetrized EDC shows a peak at ~10 meV, suggesting the opening of a SC gap. FDfunction-divided EDCs obtained from K-Ba122 (x = 0.25 and 0.3) at the  $k_F$ 's of the  $\alpha$  and  $\gamma$  bands are shown in Figures 1(c1)– 2(d2) for comparison. While the SC peak (x = 0.3) is far from the  $E_F$ , indicating that the SC gap is observed on the  $\alpha$  and  $\gamma$  FSs, we found that the SC peak of  $x \sim 0.25$  moves towards  $E_F$ , which indicates the decrease or almost close of SC gaps for the  $\gamma$  band.

Figure 2 gives the detailed SC gap distribution of the electron bands designated as  $\varepsilon$  and  $\delta$  for the inner and outer electron FSs, respectively. To precisely determine the SC gap size and its momentum dependence, we have performed an ARPES study at several  $k_F$  and  $k_z$  points of the  $\varepsilon$  and  $\delta$  bands. For the  $k_z$  direction, while the SC gap for the  $\delta$  band shows an isotropic gap [Figure 2(g)], the EDC peak position reflecting the SC gap for the  $\varepsilon$  band does not seem to be constant, namely, the peak position as shown by a gray circle decreases in going from the A to M points [Figure 2(i)]. This indicates that an anisotropic SC gap is exhibited along the  $k_z$  direction for the  $\varepsilon$  FS.

The present ARPES study indicates that the  $T_c$  suppression of K-Ba122 ( $x \sim 0.25$ ) corresponds to the suppressed SC gap on the  $\gamma$  FS and the  $k_{\tau}$  dependent SC gap on the  $\varepsilon$  FS. According to the previous ARPES study, the SC gap size on hole FSs was almost identical in optimally doped K-Ba122, which was interpreted by the s++-wave SC gap symmetry due to orbital fluctuations. Though SC gaps on the  $\alpha$  and  $\beta$  FSs of K-Ba122 (x ~ 0.25) are identical, that on the  $\gamma$  FS seems not to be explained directly by the orbital fluctuation. The SC gap might be suppressed in the presence of the  $C_4$ -magnetic fluctuation, assuming that the presence of the  $C_4$  symmetry of the electronic structure with disappearance of the orbital order. We also find that the ARPES intensity plots of K-Ba122 (x = 0.21, 0.3) at the BZ center show the band folding of the electron-like band, which is known to be observed below the SDW transition temperature in the underdoped regime, but disappears in  $x \sim$ 0.25 sample (not shown). Probably recently reported antiferroic electronic instability is suppressed around  $x \sim 0.25$  because of the  $C_4$ -magnetic phase fluctuation. Thus, the experimental evidence indicates that the SC gap on the  $\gamma$  FS of K-Ba122 (x ~ (0.25) is strongly influenced by the  $C_4$ - magnetic phase.

# 2. Development of Micro-Focused Beam ARPES

A soft X-ray beamline BL5U has been open for users from 2016 and used as high energy resolution ARPES beamline. By introducing a final focusing mirror close to the sample position (~50 mm), the synchrotron light whose original size was 400 (H) × 120 (V) is successfully focused to 23 (H) × 40 (V)  $\mu$ m. ARPES study on small samples or inhomogeneous samples is now available.

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