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# Electronic structure of heavily electron-doped $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ studied by angle-resolved photoemission 

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#### Abstract

We have performed high-resolution angle-resolved photoemission spectroscopy on heavily electron-doped non-superconducting (SC) $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$. We find that the two hole Fermi surface pockets at the Brillouin zone center observed in the hole-doped superconducting $\mathrm{Ba}_{0.6} \mathrm{~K}_{0.4} \mathrm{Fe}_{2} \mathrm{As}_{2}$ are absent or very small in this compound, while the two electron pockets at the zone corner significantly expand due to electron doping by the Co substitution. Comparison of the Fermi surface between non-SC and SC samples indicates that the coexistence of hole and electron pockets connected via the antiferromagnetic wave vector is essential in realizing the mechanism of superconductivity in the iron-based superconductors.


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## 1. Introduction

The discovery of superconductivity in FeAs-based superconductors [1, 2] has attracted considerable interest since their transition temperatures ( $T_{\mathrm{c}}$; maximally $\sim 55 \mathrm{~K}[3,4]$ ) are the highest among the known superconductors except cuprates. Parent compounds of iron-based superconductors commonly show a collinear antiferromagnetic (AF) spin density wave [5] with distinct anomalies in the transport and thermodynamic properties. Doping holes or electrons into the parent compounds gives rise to superconductivity with $T_{\mathrm{c}}$ values typically above 20 K [6, 7]. For instance, in $A \mathrm{Fe}_{2} \mathrm{As}_{2}$ ( $A$ : alkali metals and alkali-earth metals), the so-called 122 system, holes are doped by the chemical substitution of $A^{2+}$ ions by potassium ions ( $\mathrm{K}^{+}$), whereas electrons are doped by the replacement of divalent iron atoms with trivalent cobalt ( Co ) or tetravalent nickel $(\mathrm{Ni})$ ions $[7,8]$. On the other hand, in $R \mathrm{FeAsO}$ compounds ( $R$ : rare earth atoms; the 1111 system), doping of hole carriers appears to be difficult [1]. Therefore, the 122 system provides a precious opportunity to explore the role of doping with both types of carriers with the same crystal structure. To elucidate the electronic states relevant to the occurrence of superconductivity in the 122 series, angle-resolved photoemission spectroscopy (ARPES) has been performed on the hole-doped $\mathrm{Ba}_{1-x} \mathrm{~K}_{x} \mathrm{Fe}_{2} \mathrm{As}_{2}$ [9]-[12] and its parent compound $\mathrm{Ba}($ or Sr$) \mathrm{Fe}_{2} \mathrm{As}_{2}[13,14]$. ARPES studies of the SC samples revealed the presence of multiple Fermi surfaces (FSs) derived from the Fe 3d orbitals, as well as FS-sheet-dependent SC gaps and many-body interactions [9, 15]. On the other hand, few such experiments have been carried out in the electron-doped counterpart, although this point is crucial despite its importance in examining the possibility of electron-hole symmetry or asymmetry. As demonstrated by electrical resistivity measurements, the $T_{\mathrm{c}}$ value of electron-doped $\mathrm{BaFe}_{2-x} \mathrm{Co}_{x} \mathrm{As}_{2}$ shows a maximum upon doping at around $x=0.15-0.2$ ( $T_{\mathrm{c}}^{\text {mid }}=25.5 \mathrm{~K}$ ), and finally disappears at $\sim 0.3$ [16]. Clarifying the microscopic origin of the characteristic SC phase diagram in the electron-doped iron-based superconductor would be essential in fully understanding the SC mechanism of the iron-based superconductors. It is thus of particular importance to investigate the band structure and the FS of the electron-doped 122 compounds by performing ARPES measurements on these materials and also to directly elucidate the doping evolution of their electronic states from the SC to the metallic region.

In this paper, we report high-resolution ARPES results on metallic $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ ( $T_{\mathrm{c}}=0 \mathrm{~K}$ ). We have determined the band structure near $E_{\mathrm{F}}$ and the FS topology, and compared with the results obtained on superconducting $\mathrm{BaFe}_{1.85} \mathrm{Co}_{0.15} \mathrm{As}_{2}\left(T_{\mathrm{c}}=25.5 \mathrm{~K}\right)$. We demonstrate
that, unlike $\mathrm{BaFe}_{1.85} \mathrm{Co}_{0.15} \mathrm{As}_{2}$, the interband scattering condition via the AF wave vector is not satisfied in $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$. We discuss the implications of our results in comparison with the hole-doped system.

## 2. Experiments

The high-quality single crystals of $\mathrm{BaFe}_{2-x} \mathrm{Co}_{x} \mathrm{As}_{2}$ used in this study were grown by the selfflux method, the same growth method as for $\mathrm{BaFe}_{2-x} \mathrm{Ni}_{x} \mathrm{As}_{2}$ [17]. Co content was determined by energy-dispersive x-ray spectroscopy. The starting materials (nominal compositions) for $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ and $\mathrm{BaFe}_{1.85} \mathrm{Co}_{0.15} \mathrm{As}_{2}$ are $\mathrm{BaFe}_{1.6} \mathrm{Co}_{0.4} \mathrm{As}_{2}$ and $\mathrm{BaFe}_{1.8} \mathrm{Co}_{0.2} \mathrm{As}_{2}$, respectively. The electrical resistivity of $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ does not show any sign of superconductivity down to 2 K . High-resolution ARPES measurements were performed using a VG-SCIENTA SES2002 spectrometer with a high-flux discharge lamp and a toroidal grating monochromator. We used the He $\mathrm{I} \alpha$ resonance line ( $h \nu=21.218 \mathrm{eV}$ ) to excite photoelectrons. The energy and angular (momentum) resolutions were set at $4-10 \mathrm{meV}$ and $0.2^{\circ}\left(0.007 \AA^{-1}\right)$, respectively. Clean surfaces for ARPES measurements were obtained by in situ cleaving of crystals in a working vacuum better than $5 \times 10^{-11}$ Torr. The Fermi level $\left(E_{\mathrm{F}}\right)$ of the samples was referenced to that of a gold film evaporated onto the sample substrate. Mirror-like sample surfaces were found to be stable without obvious degradation for the measurement period of 3 days.

## 3. Results and discussion

### 3.1. Electronic structure of $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$

Figures 1 (a) and (b) show energy distribution curves (EDCs) of $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$, called here the $\mathrm{Co}_{0.3}$ sample, in a relatively wide energy region with respect to $E_{\mathrm{F}}$ measured at 15 K with the $\mathrm{He} \mathrm{I} \alpha$ line $(h v=21.218 \mathrm{eV}$ ) along two high-symmetry lines (a) $\Gamma \mathrm{X}$ and (b) $\Gamma \mathrm{M}$. In the $\Gamma \mathrm{X}$ cut (figure 1(a)), we find a band showing a hole-like dispersion centered at the $\Gamma$ point, which approaches $E_{\mathrm{F}}$ around the $\Gamma$ point. In contrast to the hole-doped $\mathrm{Ba}_{0.6} \mathrm{~K}_{0.4} \mathrm{Fe}_{2} \mathrm{As}_{2}$, we do not find apparent crossing of a hole-like band in this energy range. In the $\Gamma \mathrm{M}$ cut (figure 1(b)), we identify an electron-like band crossing $E_{\mathrm{F}}$ midway between the $\Gamma$ and M points. To see more clearly the dispersive bands, we have mapped out the ARPES intensity as a function of wave vector and binding energy and we show the results in figures 1(c) and (d) for the $\Gamma \mathrm{X}$ and $\Gamma \mathrm{M}$ directions, respectively. We also plot the results of first-principle band-structure calculations at $k_{z}=0$ and $\pi$ (blue and red curves, respectively) [18]. The calculated bands for $\mathrm{BaFe}_{2} \mathrm{As}_{2}$ are shifted downward by 90 meV , and then divided by a renormalization factor of 2 . As seen in figures 1(c) and (d), although some portions of the experimentally determined band structure show rough agreement with the renormalized band calculations, most bands show noticeable discrepancies. For example, the higher energy bands observed at the $\Gamma$ point around 0.2 eV in the experiment are found between 0.4 and 0.6 eV in the calculations, and the bottom of the electron-like band at the M point in the experiment is measured at $\sim 0.1 \mathrm{eV}$ whereas calculations predict $\sim 0.3 \mathrm{eV}$. Moreover, fine structures in the calculations, such as the complicated band dispersion around the M point and the appearance of a small hole pocket near the X point, are not well reproduced in the experiment.

Figure 2(a) displays the ARPES intensity plot at $E_{\mathrm{F}}$ of the $\mathrm{Co}_{0.3}$ sample as a function of the in-plane wave vector measured at 15 K . We identify two bright spots centered at the $\Gamma$ and


Figure 1. EDCs near $E_{\mathrm{F}}$ of non- $\mathrm{SC} \mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ measured at 15 K with the $\mathrm{He} \mathrm{I} \alpha$ line ( $h \nu=21.218 \mathrm{eV}$ ) along two high symmetry lines (a) $\Gamma \mathrm{X}$ and (b) ГМ. (c) and (d) ARPES intensity plots as a function of wave vector and binding energy along the $\Gamma \mathrm{X}$ and $\Gamma \mathrm{M}$ lines, together with the band dispersion from the band calculations for $k_{z}=0$ and $\pi$ (blue and red curves, respectively). Calculated bands for $\mathrm{BaFe}_{2} \mathrm{As}_{2}$ [18] were shifted downward by 90 meV and then renormalized by a factor of 2 .

M points. The intensity centered at the M point originates in the electron-like band as seen in figure 1(b), whereas that at the $\Gamma$ point is produced by the spectral weight at/near the top of the hole-like bands. To examine the character of the FSs in more detail, we plot in figure 2(b) the EDCs in the vicinity of $E_{\mathrm{F}}$ as well as its intensity plot (figure 2(c)) along cut 1 ( $\mathrm{\Gamma}$ direction), measured with a higher energy resolution. As shown in figure 2(d) where the second derivative of the EDCs is plotted as a function of binding energy and wave vector, two hole-like bands can be recognized. One has a top of dispersion around 50 meV at the $\Gamma$ point, whereas the other is located at lower binding energy with a top of dispersion at the $\Gamma$ point at less than 10 meV . According to a previous ARPES study of $\mathrm{Ba}_{0.6} \mathrm{~K}_{0.4} \mathrm{Fe}_{2} \mathrm{As}_{2}$ [9], the inner and outer hole-like bands are attributed to the $\alpha$ and $\beta$ bands, respectively. Judging from the presence of a Fermiedge cutoff around the $\Gamma$ point, the $\beta$ band may touch $E_{\mathrm{F}}$, although it is hard to distinguish


Figure 2. (a) ARPES intensity plot at $E_{\mathrm{F}}$ of $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ as a function of the two-dimensional wave vector measured at 15 K . The intensity at $E_{\mathrm{F}}$ is obtained by integrating the spectra within $\pm 5 \mathrm{meV}$ with respect to $E_{\mathrm{F}}$. The location of the $k_{\mathrm{F}}$ points (open circles) has been determined by tracing the peak position of the second derivative of the MDCs at $E_{\mathrm{F}}$, and then the $k_{\mathrm{F}}$ points were folded by assuming mirror symmetry with respect to the $\Gamma \mathrm{M}$ line ( $k_{x}$-axis). The dashed lines were extracted by fitting the location of the $k_{\mathrm{F}}$ points by using the tightbinding formula [19, 23]. (b) Representative EDCs in the vicinity of $E_{\mathrm{F}}$ and (c) its intensity plot as a function of binding energy and wave vector, measured along cut 1 indicated by a green line in (a). (d) Intensity of the negative part of the second derivative of EDCs along cut 1. It is noted that the second-derivative intensity of the $\beta$ band near $E_{\mathrm{F}}$ is affected by the Fermi-edge cut-off. (e) and (f), same as (b) and (c) but measured along cut 2. (g) Intensity of the negative part of the second derivative of MDCs along cut 2 .
the crossing point with the present experimental accuracy. In any case, the FS produced by the $\beta$ band would be negligibly small as compared to the electron pocket at the M point, in sharp contrast to the largest size of the $\beta$ FS in the hole-doped $\mathrm{Ba}_{0.6} \mathrm{~K}_{0.4} \mathrm{Fe}_{2} \mathrm{As}_{2}$ [9, 19]. Besides one electron-like FS/dispersion, which is prominent around the M point as seen in figures 2(a) and (e), we find clear evidence for another electron pocket. As illustrated in the second-derivative plot of the momentum distribution curves (MDCs) measured along cut 2 (figure 2(g)), two nearly parallel electron-like bands crossing $E_{\mathrm{F}}$ are clearly distinguished. By comparing with the hole-doped $\mathrm{Ba}_{0.6} \mathrm{~K}_{0.4} \mathrm{Fe}_{2} \mathrm{As}_{2}$ [9], [18]-[22], these inner and outer FSs are attributed to the $\gamma$ and $\delta$ FSs, respectively. As in the case of the hole-doped system [19], the existence of these two electron pockets in the $\mathrm{Co}_{0.3}$ sample is basically explained by the presence of two ellipsoidal FSs elongated along the two $\Gamma \mathrm{M}$ directions (along $k_{x}$ - and $k_{y}$-axes) as indicated by dashed lines that represent the tight-binding [19, 23] fits of the determined $k_{\mathrm{F}}$ (Fermi vector) points


Figure 3. (a) Comparison of experimentally determined $k_{\mathrm{F}}$ points between non$\mathrm{SC} \mathrm{BaFe} \mathrm{B}_{1 .} \mathrm{Co}_{0.3} \mathrm{As}_{2}\left(\mathrm{Co}_{0.3}\right)$ and $\mathrm{SC} \mathrm{BaFe}{ }_{1.85} \mathrm{Co}_{0.15} \mathrm{As}_{2}\left(\mathrm{Co}_{0.15}\right)$. The $k_{\mathrm{F}}$ points are symmetrized by assuming fourfold symmetry with respect to the $\Gamma$ and M points. The inset shows the experimental band dispersion in the vicinity of $E_{\mathrm{F}}$ around the $\Gamma$ point determined by tracing the peak position of background-subtracted EDCs divided by the Fermi-Dirac distribution function. An ARPES spectrum away from the $k_{\mathrm{F}}$ points was chosen as a background. The chemical potential of the $\mathrm{Co}_{0.3}$ sample is shifted upward by 20 meV with respect to that of the $\mathrm{Co}_{0.15}$ sample. The peak position of the $\beta$ band near $E_{\mathrm{F}}$ for the $\mathrm{Co}_{0.3}$ sample does not coincide exactly with the second-derivative intensity in figure 2(d), which is affected by a Fermi-edge cut-off.
(open circles) in figure 2(a). It is noted here that, to determine the energy positions of bands from the second-derivative plot, it is better to differentiate spectral intensity perpendicularly to the band dispersion. Hence, we have selected the EDCs method when the top or bottom of bands is included in the energy range of interest, as in the case of figures 2(b)-(d). On the other hand, the MDCs method is more reliable for figures 2(e)-(g) since the bottom of bands is away from $E_{\mathrm{F}}(\sim 80 \mathrm{meV})$ and the band dispersion is steep around $E_{\mathrm{F}}$.
3.2. Comparison of electronic structure between metallic and superconducting $\mathrm{BaFe}_{2-x} \mathrm{Co}_{x} A s_{2}$ ( $x=0.3$ and 0.15)

Figure 3 displays a direct comparison of the momentum location of $k_{\mathrm{F}}$ points between the metallic non- $\mathrm{SC} \mathrm{Co}_{0.3}$ and the $\mathrm{SC} \mathrm{Co}_{0.15}$ samples. Upon electron doping, the hole-like $\beta \mathrm{FS}$, as observed in the $\mathrm{Co}_{0.15}$ sample [24], disappears or becomes indistinguishably small in the $\mathrm{Co}_{0.3}$ sample. Simultaneously, the size of the two electron pockets at the M point significantly expands with electron doping. Indeed, the estimated volume of the $\beta$ FS with respect to the first unfolded


Figure 4. Comparison of energy bands between the $\mathrm{Co}_{0.15}$ and $\mathrm{Co}_{0.3}$ samples. The interband scattering is dramatically suppressed in the non- $\mathrm{SC} \mathrm{Co}_{0.3}$ sample since the holelike $\alpha$ and $\beta$ bands at the $\Gamma$ point are basically occupied.

Brillouin zone for the $\mathrm{Co}_{0.3}$ sample is $0-0.5 \%$, much smaller than that for $\mathrm{Co}_{0.15}(1.5 \pm 0.5 \%)$, and the volume of the electron-like FS is larger in the $\mathrm{Co}_{0.3}$ sample ( $6.5 \pm 0.5 \%$ ) as compared to the $\mathrm{Co}_{0.15}$ sample ( $3.5 \pm 0.5 \%$ ). By taking into account the volume of all FS sheets including the two electron pockets, the total electron concentration of the $\mathrm{Co}_{0.3}$ sample is estimated to be $0.13 \pm 0.01$ electrons $\mathrm{Fe}^{-1}$, where the error in the FS volume and carrier concentration originates in the experimental uncertainties in the determination of the location of the $k_{\mathrm{F}}$ points. This value is close to the expected value ( 0.15 electrons $\mathrm{Fe}^{-1}$ ). We emphasize here that the FSs at the $\Gamma$ point of the SC and non-SC samples are drastically different since the $\beta$ hole pocket observed in the $\mathrm{Co}_{0.15}$ sample is absent or fairly small in the $\mathrm{Co}_{0.3}$ sample. This marked difference is reasonably explained by a chemical potential shift due to electron doping. As shown in the inset of figure 3 , the energy positions of bands in the vicinity of $E_{\mathrm{F}}$ near the $\Gamma$ point for the $\mathrm{Co}_{0.3}$ and $\mathrm{Co}_{0.15}$ samples quantitatively match when we shift down the bands of $\mathrm{Co}_{0.15}$ by 20 meV , suggesting the basic applicability of a rigid-band model. A simple extrapolation using a tightbinding formula suggests that the hole-like $\beta$ band in the $\mathrm{Co}_{0.15}$ sample tops at less than 20 meV above $E_{\mathrm{F}}$ [24]. The $\beta$ band for the $\mathrm{Co}_{0.3}$ sample would be almost completely occupied if we assume the observed chemical potential shift of 20 meV .

Now we discuss the mechanism of $T_{\mathrm{c}}$ reduction and resultant disappearance of superconductivity in the overdoped region. As seen in the schematic band structure in figure 4, the $\beta$ FS and $\gamma \mathrm{FS}$ of the $\mathrm{SC} \mathrm{Co}_{0.15}$ sample are well connected by the AF wave vector $Q_{\mathrm{AF}}=(\pi, 0)$ [24]. Therefore, the enhanced interband scattering via the $Q_{\mathrm{AF}}$ vector would promote the pair scattering between these two FSs by low-energy fluctuations, leading to an increase of the pairing amplitude [25]-[27]. On the other hand, in the non-SC $\mathrm{Co}_{0.3}$ sample, this interband scattering would decrease since the $\beta$ band is in the occupied side and the size of the $\gamma$ and $\delta$ pockets expands. This suggests that the interband scattering between the hole and electron pockets is essential to achieve high $T_{\mathrm{c}}$ values. The absence (deterioration) of this scattering condition in the $\mathrm{Co}_{0.3}$ sample, likely assisted by the Co-substitution-induced disorder, would completely kill superconductivity. It is also remarked here that a similar mechanism is also at work in the hole-doped $\mathrm{Ba}_{1-x} \mathrm{~K}_{x} \mathrm{Fe}_{2} \mathrm{As}_{2}$ where the disappearance of the electron-like $\gamma$ band and resultant ill-defined interband scattering condition plays a critical role in the $T_{\mathrm{c}}$-reduction mechanism in the overdoped region [20]. All these experimental results suggest
a possible electron-hole symmetry of the interband scattering and the pairing mechanism in the iron-based superconductors.

## 4. Summary

In summary, we have reported ARPES results on $\mathrm{BaFe}_{1.7} \mathrm{Co}_{0.3} \mathrm{As}_{2}$ and determined the band dispersion near $E_{\mathrm{F}}$ and the FS topology. The experimentally determined FS consists of two electron pockets centered at the M point. The $\beta$ hole pocket seen in the superconducting $\mathrm{BaFe}_{1.85} \mathrm{Co}_{0.15} \mathrm{As}_{2}$ sample is absent or very small, resulting in the suppression of the $Q_{\mathrm{AF}}=$ $(\pi, 0)$ interband scattering through the electron pocket at the M point, which is likely responsible for the disappearance of $T_{\mathrm{c}}$ at this heavy electron doping level.

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