Band Structure of the Heavily-Electron-Doped FeAs-Based $Ba(Fe,Co)_2As_2$ Superconductor Suppresses Antiferromagnetic Correlations

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In the heavily-electron-doped regime of the $Ba(Fe,Co)_2As_2$ superconductor, three hole bands at the zone center are observed and two of them reach the Fermi level. The larger hole pocket at the zone center is apparently nested with the smaller electron pocket around the zone corner. However, the $(\pi,0)$ Fermi surface reconstruction reported for the hole-doped case is absent in the heavily-electron-doped case. This observation shows that the apparent Fermi surface nesting alone is not enough to enhance the antiferromagnetic correlation as well as the superconducting transition temperature.

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The discoveries of the superconductivity in the layered FeP [1] and FeAs [2] systems have ignited tremendous research activities for understanding of the superconductivity in Fe pnictides. The superconducting transition temperature T_c of electron-doped $LaFeAsO_{1-x}F_x$ reaches 43 K under pressure [3], and $NdFeAsO_{1-x}F_x$ and $SmFeAsO_{1-x}F_x$ show T_c higher than 40 K at ambient pressure [4,5]. $BaFe_2As_2$ also shows superconductivity by hole doping with the highest T_c of 38 K in $(Ba,K)Fe_2As_2$ [6–8] and by electron doping with the highest T_c of 25 K in $Ba(Fe,Co)_2As_2$ [9–11]. These Fe pnictides commonly have the FeAs layers, where the Fe ions form a square lattice and each Fe ion is tetrahedrally coordinated by four As ions.

The geometry of Fermi surfaces and the symmetry of the superconducting gap are key issues to understand the nature of the superconductivity. The angle-resolved photoemission spectroscopy (ARPES) technique has been playing important roles to observe the band dispersions, the Fermi surfaces, and the superconducting gap in various superconductors including the high-T_c cuprates. As for the Fe pnictides, ARPES studies on hole-doped $BaFe_2As_2$ [12–14] show that the two hole pockets are located around the Brillouin zone center (Γ point) and the electron pockets at the zone corners (M points). As for electron-doped $BaFe_2As_2$, it has been reported that one hole band is located below the Fermi level (E_F) and one hole pocket remains at the zone center [15,16]. The band-structure calculations predict that three hole bands should cross E_F or, at least, should be located near E_F around the zone center [17–19]. Therefore, in the ARPES studies of hole-doped and electron-doped $BaFe_2As_2$, one hole band is missing compared to the prediction by the band-structure calculations. Such disagreement in the FeAs systems is in sharp contrast to the agreement in the FeP system where an ARPES study by Lu *et al.* has successfully identified all hole and electron bands near E_F as predicted by band-structure calculations [20]. The ARPES studies also report that the Fermi surface reconstruction due to the antiferromagnetism coexists with the superconductivity in hole-doped systems [13,14]. Also, it has been pointed out that the superconducting gap in one of the hole pockets is enhanced if the hole pocket satisfies the nesting condition with one of the electron pockets at the zone corner [12,15]. However, since all of the hole bands expected from the band-structure calculations are not identified yet in the ARPES studies of hole-doped and electron-doped $BaFe_2As_2$, it is still difficult to make a conclusive remark on the relationship between the Fermi surface geometry and the superconductivity.

Here, we report on an ARPES study of $Ba(Fe,Co)_2As_2$ in the heavily-electron-doped regime. In the present ARPES results, three hole bands are successfully identified at the zone center, and two of them are found to cross E_F. The Fermi surface geometry is qualitatively consistent with the prediction of the band-structure calculations. The $(\pi,0)$ Fermi surface reconstruction observed in the hole-doped systems [13] is absent in the heavily-electron-doped system although one of the hole pockets at the zone center apparently satisfies the nesting condition with one of the electron pockets at the zone corner.

Single crystals of $BaFe_{1.7}Co_{0.3}As_2$ ($T_c = 7$ K) were grown by the Bridgman method with FeAs flux [21]. ARPES measurements were performed at beam line 9A, Hiroshima Synchrotron Radiation Center (HSRC) using a SCIENTA R4000 analyzer with circularly polarized light

($h\nu = 17$ and 23 eV). Total energy resolutions were set to 18 and 14 meV for $h\nu = 23$ and 17 eV, respectively. We cleaved the single crystals at 30 K under ultrahigh vacuum and took ARPES data at 30 K within 4 h after the cleavage.

Figure 1(a) shows a second derivative plot of the ARPES data taken at $h\nu = 23$ eV around the zone center (Γ point) of the two-dimensional square-shaped Brillouin zone. The in-plane momentum is swept along the in-plane Fe-Fe direction. In the second derivative plot, three parabolic features A, B, and C are observed, and A and B reach E_F at the zone center. The blue, green, and red dots in Fig. 1 indicate the band locations for parabolas A, B, and C, respectively, which are determined by fitting the momentum distribution curve (MDC) in Fig. 1(b) to model Lorentzian functions. The obtained band locations are in good agreement with the energy distribution curve as shown in Fig. 1(c) indicating that the fitted result is reliable. In Fig. 1(d), the fitted results are decomposed into the components for parabolas A, B, and C and are compared with the experimental results in Fig. 1(d). Parabola C does not reach E_F, and its maximum point is located at $\sim$40 meV below E_F. The residual central peak at $k = 0$ over the top of parabola C is due to the broadening caused by the finite energy resolution and the correlation effect of the electron-electron and/or electron-phonon interactions.

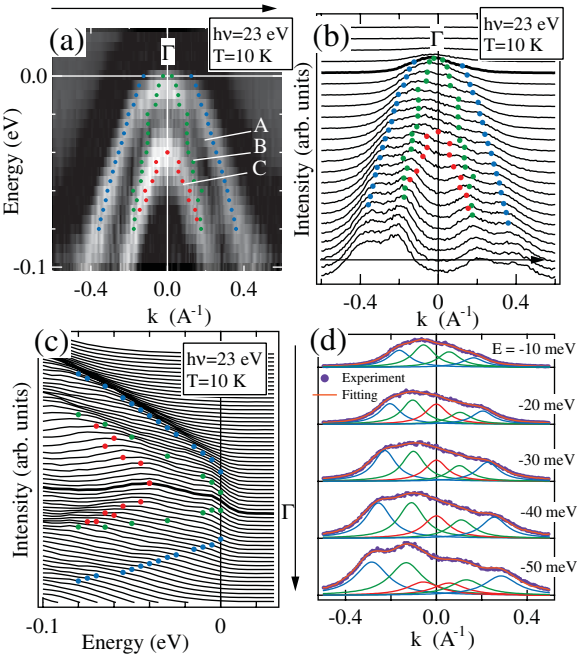

FIG. 1 (color). (a) Second derivative plot, (b) momentum distribution curves, and (c) energy distribution curves for $Ba(Fe,Co)_2As_2$ at 10 K along the Γ-M direction around the zone center. The ARPES data are taken at $h\nu = 23$ eV. The blue, green, and red dots indicate the band locations determined by fitting the momentum distribution curves to Lorentzian functions. (d) The fitted results are decomposed into the components for the three bands (parabolas A, B, and C). The $k = 0$ component for $E = -20$ and -30 meV can be assigned to the tail of parabola C above its band maximum and is not shown in (a)–(c).

Figure 2 shows a second derivative plot of the ARPES data taken at $h\nu = 17$ eV around the zone center and the zone corner (M point) of the two-dimensional square-shaped Brillouin zone. Around the zone corner, the two bands are found to cross E_F and to form the two electron pockets. The crossing point of the inner electron band is close to the zone corner while that of the outer electron band is relatively far from the zone corner. As for the zone center, two parabolic features are observed. The outer parabola reaches E_F and corresponds to parabola A observed at $h\nu = 23$ eV. The inner parabola is asymmetric between the right-hand side and the left-hand side of the zone center. The asymmetry indicates that the inner parabola is made up from two bands that are parabolas B and C observed at $h\nu = 23$ eV. Parabola A is very flat around the zone center. The spectral weight at E_F on the inside of the hole pocket is substantial, indicating that the maximum point of the hole band is very close to E_F. In contrast to the flatness of the hole band at the zone center, the spectral weight at E_F on the inside of the electron pockets is very small and the minimum points of the inner and outer electron bands are well below E_F and become very broad due to electron-electron and/or electron-phonon scatterings.

The zone centers at $h\nu = 23$ eV (17 eV) approximately correspond to the Γ point (the midpoint of the Γ and Z points) in the three-dimensional Brillouin zone for $BaFe_2As_2$. Also, the zone corner at $h\nu = 17$ eV is located around the X point in the three-dimensional Brillouin zone for $BaFe_2As_2$ [19]. As reported by Vilmercati *et al.*, the hole pockets at the zone center depend on the photon energy, namely, the momentum perpendicular to the FeAs plane k_z [22]. The comparison between the ARPES data taken at $h\nu = 23$ and 17 eV shows that the areas of the hole pockets increase in going from the Γ point to the Z

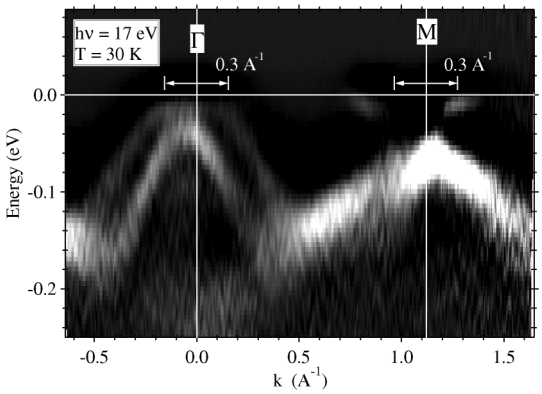

FIG. 2. Second derivative plot for $Ba(Fe,Co)_2As_2$ at 30 K along the Γ-M direction. The ARPES data are taken at $h\nu = 17$ eV. The Γ-M direction is parallel to the in-plane Fe-Fe bond, and the M point is located at the zone corner.

point of the three-dimensional Brillouin zone. This observation agrees with the reports by Vilmercati *et al.* [22] The lowest hole band or parabola C is located well below E_F along the Γ-Z direction of the three-dimensional Brillouin zone.

The observation of the three hole bands (parabolas A, B, and C) around the zone center is qualitatively consistent with the band-structure calculations for $BaFe_2As_2$ [19]. However, the observed energy separation between the top of parabola C and those of parabolas A and B is reduced from the prediction of the band-structure calculation. On the other hand, in the hole-doped system, the energy separation observed in the ARPES study [23] agrees with that of the band-structure calculation. The contrast between the electron-doped and hole-doped systems suggests that the correlation effect of the electron-electron and/or electron-phonon interactions strongly depends on the doping level. The maximum points of parabolas A and B are almost degenerate in energy. This degeneracy also agrees with the band-structure calculation. The outer parabola seen at $h\nu = 17$ eV or parabola A seen at $h\nu = 23$ eV becomes very flat near the zone center, and the spectral weight at E_F is substantial on the inside of the Fermi surface. Therefore, the maximum point of the higher two hole bands or parabolas A and B is expected to be very close to E_F due to the upward chemical potential shift by the electron doping.

Figures 3(a) and 3(b) show MDC plots of the ARPES data taken at 30 K around the zone center and the zone corner, respectively. The momentum is swept along the in-plane Fe-Fe direction with photon energy of $h\nu = 17$ eV. In the present electron-doped system, there is no signature of coexistence of the antiferromagnetism and the superconductivity that is reported in the hole-doped systems by Zhang *et al.* [14] The unusual flatness of the outer hole band is clearly seen in the MDC plot shown in Fig. 3(a). As mentioned in the previous paragraphs, the spectral weight at E_F is substantial even on the inside of the Fermi surface. On the other hand, in the inner and outer electron bands, the spectral weight at E_F rapidly decreases in going from the crossing point on the Fermi surface to the inside of the Fermi surface [see Fig. 3(b)]. The size of the hole pocket at the zone center is similar to that of the smaller electron pocket at the zone corner as indicated in Figs. 3(a) and 3(b).

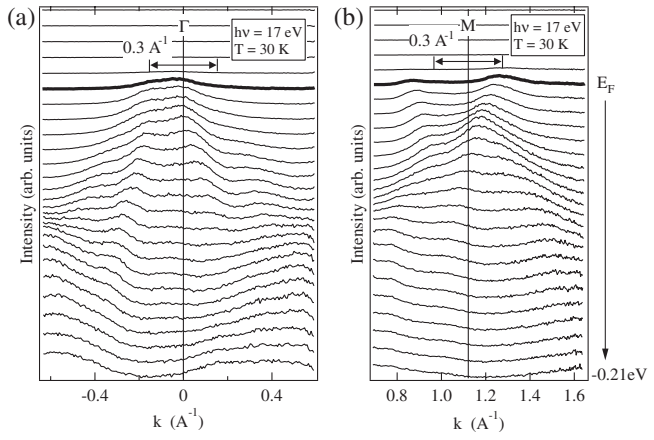

FIG. 3. Momentum distribution curves for $Ba(Fe,Co)_2As_2$ at 30 K along the Γ-M direction (a) around the zone center and (b) around the zone corner. The ARPES data are taken at $h\nu = 17$ eV. The Γ-M direction is parallel to the in-plane Fe-Fe bond, and the M point is located at the zone corner.

The Fermi surface mapping is displayed in Fig. 4 to show the geometries of the hole pockets around the zone center and the electron pockets around the zone corner. The Fermi surface mapping is constructed from the ARPES data taken at 30 K with $h\nu = 17$ eV by integrating the spectral weight within ± 5 meV from E_F. The momentum points where the bands cross E_F can be determined by fitting the momentum distribution curves of Fig. 3 to Lorentzian functions. The momentum points thus determined are indicated by the green dots in Fig. 4. The circular hole pockets around the zone center and the elongated electron pockets around the zone corner are clearly observed. Since the zone center of $h\nu = 17$ eV is approximately located at the midpoint of the Γ and Z points, the hole pocket area obtained at $h\nu = 17$ eV is expected to be close to the average of the hole pocket area that monotonically increases from Γ point to Z point of the three-dimensional Brillouin zone. The total area of the two electron (hole) pockets is $\sim$17% (3%) of the two-dimensional Brillouin, and the doped electron number per unit cell is estimated to be $\sim$0.14, which is roughly consistent with the composition of $BaFe_{1.7}Co_{0.3}As_2$. The magnitude of the k_z dependence of the Fermi surfaces observed in the heavily-electron-doped system is very similar to that observed in the optimally-electron-doped system [22]. The dimension along the diagonal line of the larger hole pocket is ~ 0.3 Å^{-1} and is similar to that of the smaller electron pocket at the zone corner. In Fig. 4, the larger hole pocket at the zone center is shifted by the momentum for $(\pi,0)$ wave vector and is displayed by blue dots. The large portion of the shifted hole pocket overlaps with the smaller electron pocket and that rotated by 90°. Therefore, the pair of hole and electron pockets is apparently nested and may cause antiferromagnetic correlation. Here, considering the three-dimensional Brillouin zone of $Ba(Fe,Co)_2As_2$, the elongated electron pockets observed at $h\nu = 17$ eV are rotated by 90° at different k_z [19]. Therefore, in order to discuss two-dimensional nesting character, the electron pockets rotated by 90° should be considered as has been done by Terashima *et al.* for the optimally-electron-doped system [15].

If the apparent nesting causes the antiferromagnetic correlation characterized by the $(\pi,0)$ wave vector, the Fermi surface would be reconstructed due to band folding and/or partial gap opening even without static antiferro-

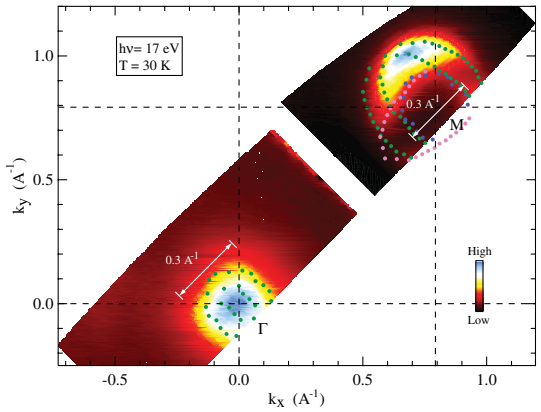

FIG. 4 (color). Fermi surface mapping for $Ba(Fe,Co)_2As_2$ at 30 K (above T_c) as a function of in-plane momentum k_x and k_y. The ARPES data are taken at $h\nu = 17$ eV. The Γ-M direction is parallel to the in-plane Fe-Fe bond, and the M point is located at the zone corner. The green dots indicate the momentum points where the bands cross E_F. The two hole pockets and the two electron pockets are seen around the zone center and the zone corner, respectively. The larger hole pocket is shifted by the momentum for the $(\pi,0)$ wave vector and is shown by the blue dots. The smaller electron pocket is rotated by 90° and is shown by pink dots to discuss two-dimensional nesting character.

magnetic order. The $(\pi,0)$ Fermi surface reconstruction is reported in the optimally hole-doped system [13], and is somewhat similar to the Fermi surface reconstruction due to antiferromagnetic correlation discussed for high-T_c cuprates [24]. On the other hand, such $(\pi,0)$ Fermi surface reconstruction is absent in the present heavily-electron-doped system as well as in the optimally-electron-doped system [15], indicating that the antiferromagnetic correlation is actually weak in the electron-doped case compared to that in the hole-doped one. Here, the $(\pi,0)$ Fermi surface reconstruction corresponds to (π,π) Fermi surface reconstruction of the study by Zabolotnyy *et al.* [13], and the difference of the characteristic wave vector comes from the different definition of two-dimensional Brillouin zone. In addition, it should be noted that the $(\pi,0)$ Fermi surface reconstruction is reported to be weak or absent in other ARPES studies on the hole-doped systems [12]. The degree of Fermi surface nesting in the heavily-electron-doped system is comparable to that of the optimally-electron-doped system [15] although it would be reduced compared to that in the hole-doped systems. While T_c reaches 25 K in the optimally-electron-doped system, the heavily-electron-doped system with similar nested Fermi surface has T_c as low as 7 K, indicating that the nesting effect is not that important to enhance T_c at least in the electron-doped systems.

In summary, the electronic structure of $Ba(Fe,Co)_2As_2$ in the heavily-electron-doped regime has been investigated using ARPES. The ARPES experiments at $h\nu = 17$ and 23 eV have shown that the hole pockets at the zone center and the electron pockets at the zone corner are qualitatively consistent with the prediction of the band-structure calculations and that the effect of antiferromagnetic correlations is very small in the electron-doped system compared to that in the hole-doped systems. The two hole bands crossing E_F are extremely flat near the zone center compared to the electron bands at the zone corner. Even in the heavily hole-doped system with low T_c, the larger hole pocket at the zone center is apparently nested with the smaller electron pocket at the zone corner, indicating that the Fermi surface nesting alone is not enough to enhance T_c.

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