Orbital-dependent electron correlation effect on the two- and three-dimensional Fermi surfaces in KFe$_2$As$_2$ revealed by angle-resolved photoemission spectroscopy

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We have performed an angle-resolved photoemission study of the hole-overdoped iron pnictide superconductor KFe$_2$As$_2$, which shows a low $T_c$ of $\sim 4$ K. Most of the observed Fermi surfaces show nearly two-dimensional shapes, while a band near the Fermi level shows a strong dispersion along the $k_x$ direction and forms a small three-dimensional hole pocket centered at the $Z$ point, as predicted by band-structure calculation. However, hole Fermi surfaces of $yz$ and $xz$ orbital character centered at the $\Gamma$ point of the two-dimensional Brillouin zone are smaller than those predicted by the calculation while the other hole Fermi surfaces of $xy$ orbital character is much larger. Clover-shaped hole Fermi surfaces around the corner of the 2D BZ are also larger than those predicted by the calculation. These observations are consistent with the de Haas-van Alphen measurement and indicate orbital-dependent electron correlation effects. The effective masses of the energy bands show moderate to strong enhancement, partly due to electron correlation and partly due to energy shifts from the calculated band structure.

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I. INTRODUCTION

In contrast to the $d$-wave superconducting gaps in the high-$T_c$ cuprate superconductors, experimental results on most of the iron-pnictide superconductors have indicated that superconducting gaps are nodeless and on the entire Fermi surfaces (FSs)\cite{1,2}. However, some of the iron pnictide superconductors show signatures of the nodes in the superconducting gaps. For example, thermal conductivity measurements of isovalent substituted system BaFe$_2$(As$_{1-x}$P$_x$)$_2$\cite{2} and the electron doped systems Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ in the conducting state have shown signature of line nodes. According to the theories of spin fluctuation-mediated superconductivity, line nodes may appear when the nontight height becomes small\cite{3},\cite{4}, the hole FS of $d_{xy}$ character around the zone center disappears and nesting between hole and electron FSs becomes weakened. (Here, $x$ and $y$ are referred to the direction of the nearest neighbor Fe atoms.) The hole FSs of these systems exhibit strong three-dimensionality\cite{5},\cite{6}, resulting in poor nesting between the hole and electron FSs.

In the K-doped BaFe$_2$As$_2$ (Ba122) system, a full superconducting gaps opens and a high $T_c$ of $\sim 37$ K is achieved in the optimally doped region\cite{7}, while the existence of line nodes in the superconducting gap has been suggested for the end member compound KFe$_2$As$_2$, which shows a low $T_c$ of $\sim 4$ K from penetration depth\cite{8},\cite{9}, thermal conductivity\cite{10},\cite{11}, and nuclear quadrupole resonance (NQR) measurements\cite{12}. The electronic specific heat coefficient $\gamma$ of KFe$_2$As$_2$ is as large as $\sim 70$ mJ/K$^2$mol\cite{13}, indicating strong electron mass renormalization due to electron correlation. A previous angle-resolved photoemission study (ARPES) of KFe$_2$As$_2$ has revealed that there are hole FSs ($\alpha$ and $\beta$) around the zone center while the electron pockets around the zone corner disappear and change to small hole pockets ($\epsilon$) surrounding the zone corner in a clover shape\cite{12}. Band-structure calculation has predicted the third hole FS around the zone center, namely, the $\zeta$ hole FS which has a similar size to the $\alpha$ FS, but it has not been resolved in the previous ARPES study. Recently, a de Haas-van Alphen (dHvA) study has revealed that the $\zeta$ FS exists around the zone center\cite{14}. Furthermore, the dHvA study has indicated the shrinkage of the $\alpha$ and $\zeta$ FSs and the enhancement of the electron masses compared to those predicted by the band-structure calculation. In the present study, in order to reveal the shapes of the FSs in three-dimensional momentum space, we have performed an ARPES study of KFe$_2$As$_2$ using high-quality single crystals and various photon energies. We have determined the orbital character of the FSs by polarization dependent measurements and have revealed strongly orbital dependent correlation effects.

II. EXPERIMENT AND BAND-STRUCTURE CALCULATION

ARPES measurements were performed at beamline 5-4 of Stanford Synchrotron Radiation Lightsource (SSRL) and at BL10.0.1 of Advanced Light Source (ALS). Scienta SES-R4000 electron analyzers and linearly polarized...
III. RESULTS AND DISCUSSION

Band dispersions for a cut along the diagonal of the two-dimensional Brillouin zone (BZ) taken with $h\nu=25$ eV and 30eV are shown in Figs. 1(a) and 1(b). All the energy bands predicted by the calculation [Fig. 1(c)] are observed. Particularly, we have clearly observed the $\zeta$ band near the $\alpha$ band around $E_F$ consistent with the dHvA results. In addition to these bands, we find that another hole-like band crossing $E_F$ exists near the zone center. Since this is not predicted by the bulk band-structure calculation, we attributed this band to surface states. While the $\zeta$ band is nearly degenerated with the $\alpha$ band for $h\nu=25$ eV, these bands are separated for $h\nu=30$ eV, indicating three-dimensionality of the band dispersions.

FS mapping in the $k_X$-$k_Y$ plane is shown in Figs. 2 (a) and 2(b). By assuming the inner potential $V_0=13.0$ eV, panels (a) and (b) approximately represent the $k_X$-$k_Y$ planes including the $\Gamma$ and the $Z$ point, respectively. The overall FS shapes nearly agree with those observed in the previous study. Small hole FSs also appear around the BZ corner due to heavy hole doping. However, several new observations should be remarked. One is that all the three hole FSs around the center of the 2D BZ have been clearly resolved. The middle hole FS is ascribed to the $\zeta$ FS reported in the dHvA study. Another point is that a small FS around the zone center has been observed. Since this FS is not predicted by the band-structure calculation and has nearly two-dimensional dispersions as indicated below, it can be ascribed to surface states. The dispersions of the surface states form ridge-like structures extending to the $k_X$ and $k_Y$ directions, resulting in the peculiar cross-like intensity distribution at $E_F$.

In Fig 3 we compare the FSs obtained by ARPES with the band-structure calculation. As seen in panels (a) and (b), the sizes of the observed $\alpha$ and $\beta$ FSs do not show appreciable change with $k_z$. On the other hand, the shape of the $\zeta$ FS significantly changes between $k_z$ and $Z$. While the $\zeta$ FS has a diamond-like cross-section.
FIG. 3. (Color online) Comparison of FSs obtained by ARPES and those predicted by band-structure calculation. (a), (b) FSs determined by ARPES. $k_F$ positions in Fig. 2 have been symmetrized in the first BZ. (c), (d) FSs given by the band-structure calculation. A small FS around the $Z$ point comes from a three-dimensional $d_{x^2-y^2}$ band shown in Fig. 4 (c).

for $k_z \sim \Gamma$ and is nearly degenerate with the $\alpha$ FS in the zone diagonal direction, it has a circular cross-section for $k_z \sim Z$ point.

Since the Ba122 system in general has three-dimensional FSs as predicted by band-structure calculations and confirmed by ARPES, we have investigated the electronic band dispersions in the $k_z$ direction by changing the excitation photon energy.

Figure 4(a) shows spectral weight mapping at $E_F$ in the $k_{||}-k_z$ plane, where the direction $k_{||}$ in the $k_X-k_Y$ plane is the $\Gamma-X$ direction. $k_z$ has been determined by assuming the inner potential of $V_0 = 13.0$ eV, so that the dispersion of the $d_{x^2-y^2}$ band along $k_z$ has the correct periodicity and phase as mentioned below. From Fig. 4(a), one can see that the $\alpha$ and $\beta$ FSs are nearly two-dimensional, while the $\zeta$ FS has some three dimensionality and becomes large near the $Z$ point compared to the $\Gamma$ point. This behavior has also been observed in the ARPES spectrum of K-doped Ba122. In normal emission spectra [Fig 4 (b)] the $d_{x^2-y^2}$ band clearly shows a parabolic dispersion along the $k_z$ direction and slightly crosses $E_F$ in the vicinity of the $Z$ point forming a small three-dimensional hole pocket around the $Z$ point. With a mass enhancement factor $m^*/m_b \sim 3$, this band is also well explained by the band-structure calculation.

In order to determine the orbital character of the FSs, we have investigated the polarization dependence of the ARPES intensity as shown in Fig. 5 FS mapping shown in panels (a) and (b) indicates clear polarization dependence in the intensity distribution for each FS. We have simulated the intensity distribution by using the following assumptions. Based on the result of the band-structure calculation, we assume that three orbitals $xy$, $yz$ and $xz$ constitute the FSs. We refer to the three band as $xy$, $yz$ and $xz$ band according to the orbital character of the band with momentum in the zone diagonal $k_z$ ($\parallel k_X+k_Y$) direction. Using the angle $\theta$ around the $\Gamma$ point, the orbital character of the $xy$, $yz$ and $xz$ band can be approximately expressed by $|xy>$, $\cos \theta |xz>$, $\sin \theta |yz>$ and $-\sin \theta |xz>$, $\cos \theta |yz>$, respectively. By assuming the dipole approximation of the transition matrix element $|<i|\varepsilon \cdot r|f>|^2$, where $|i>$, $|f>$, and $\varepsilon$ are the initial state, the final state, and the polarization vector, respectively, one can predict the intensity distribution. For example, when $|i>=|xy>$, $\varepsilon/|x|$ and $|f>$ is a wave function of a free electron, the transition matrix element $|<i|\varepsilon \cdot r|f>|^2$ is proportional to $k_x^2$ in the lowest order in $k$.

Figures 5(c) and 5(d) are the results of the intensity simulations. Here, we assign the inner, middle, and outer FSs to the $yz$, $xz$, and $xy$-band, respectively, so that we can reproduce the experimental intensity distribution. This assignment of the orbital character is difference from the band-structure calculation where the inner, middle, and outer FSs have $xz$, $xy$ and $yz$ orbital character, respectively. However, present ARPES result is consistent with the previous ARPES result of Co-Ba122 and the theoretical prediction of LDA+DMFT, which indicate the energy inversion of the $xy$ and $yz/zx$ bands due to orbital-dependent correlation effect. That is, the $xy$ band in most strongly affected by electron correlation and is shifted upward relative to the other bands.

Another discrepancy from the band-structure calculation is the inversion of the $yz$ and $xz$ bands in the $k_Y$ direction. In the result of the band-structure calculation,
FIG. 5. (Color online) Polarization dependence of the FS mapping for KFe$_2$As$_2$. (a),(b) Measured ARPES intensity at $E_F$ in the $k_X$-$k_Y$ plane taken at $h\nu$=40 eV ($k_z \sim \Gamma$). Electric vectors are shown by arrows. (c),(d) Simulation of the ARPES intensity distribution corresponding to panels (a) and (b). Shapes of FSs have been taken from Fig. 2. By assuming certain orbital character for each FS, intensity distribution has been simulated and is shown by thickness of the curves. (For details, see the text.)

the inner FS has $xz$ character (in the $k_X$ direction) with rounded-square shape around the $\Gamma$ point and becomes circular around the $Z$ point because of hybridization with the $z^2$ orbital. In the present ARPES result, such a character has been observed in the middle $\zeta$ FS. According to the angular-dependent magnetoresistance oscillations, such a rounded-square FS is also bigger than a circular hole FS$^{21}$. The observed inversion of the $xz$, $yz$ bands is consistent with the ARPES result of Co-Ba122$^{21,22}$ and the LDA+DMFT calculation for KFe$_2$As$_2$.\textsuperscript{20}

In the dHvA study, the size of the hole FSs are found to be smaller than those predicted by band-structure calculations.\textsuperscript{2} We have determined the cross-sectional area of the FSs as listed in Table I together with those of the dHvA measurements and the band-structure calculation. The cross-sectional areas for the $\alpha$ and $\zeta$ FSs observed by ARPES are close to those obtained by the dHvA result and are smaller than the band-structure calculation. On the other hand, the area of the $\beta$ and $\epsilon$ FSs determined by ARPES are much larger than the calculation results. The total hole count from the observed FSs yields the hole FS volume of 61 $\%$ of the BZ, indicating a deviation from the value of 50% expected from the chemical composition, because most of the FSs observed by ARPES are nearly 10-20 $\%$ larger than those observed by dHvA. The deviation of the FS volume implies that there are excess hole doping of 0.11 per Fe atom at the sample surface. Nevertheless the surface effect is not so serious as those in 1111 system where excess 0.5-0.6 holes per Fe are doped, and one can still discuss mass renormalization from the present result.

The effective masses determined by ARPES are compared with those derived from the dHvA measurements and the band-structure calculation in Table I. For the $\alpha$ and $\zeta$ FSs, the effective mass ratio $m^*/m_e$ of the free electron mass, determined by ARPES is in good agreement with those obtained by dHvA, except for the $k_z=Z$ data for the $\zeta$ FS. These values give mass enhancement factor, $m^*/m_b$ of ~3-4, where $m_b$ is a band mass. As for the $\beta$ FS, $m^*/m_e$ of ~16-18, corresponding to $m^*/m_b$ ~6 is much larger than other FSs. This strong mass enhancement for the $\beta$ band is partly due to the fact that the band bottom is closer to the Fermi level than that of the calculation. Another origin of the enhancement is electron correlation due to the $xy$ orbital character. According to the LDA+DMFT calculation,\textsuperscript{20} a larger mass renormalization is expected in the $xy$ band than those of the $yz/xz$ bands. Thus, the observed mass enhancement factors indicate moderate to strong electron correlation. From the effective masses $m^*$ listed in Table I, the electronic specific heat coefficient $\gamma$ is calculated to be $\gamma \sim$84 mJ/mol K$^2$, which is close to $\gamma=70$ mJ/mol K$^2$ estimated from specific heat measurements.\textsuperscript{12} Since in 2D material $\gamma$ is proportional to the sum of the $m^*$s of all the FSs, the $\beta$ band has a large contribution to the enhancement of $\gamma$.

The penetration depth\textsuperscript{14} and thermal conductivity\textsuperscript{13} measurements of KFe$_2$As$_2$ suggest that line nodes exist in the superconducting gap. Because the small hole FSs around the zone corner are too small to account for the linear temperature dependence of the superfluid density, the node should be on the zone-centered hole FSs. According to spin-fluctuation mediated pairing mechanism, $s$-wave and $d$-wave pairings are found to be in close competition in KFe$_2$As$_2$,\textsuperscript{22} and they might show crossover

| FS  | $k_z$ | Area (unit) | $m^*/m_e$ (unit) | $m^*/m_b$ (unit) |
|-----|------|------------|-----------------|-----------------|
| $\alpha$ | $\Gamma$ | 9.1  | 8.2  | 20.8 | 5.1 (2.0) | 6.0 (2.3) | 2.6 |
| $\Gamma$ | 9.8  | 8.6  | 21.6 | 6.6 (2.3) | 6.5 (2.2) | 2.9 |
| $\zeta$ | $\Gamma$ | 12.2 | 10.3 | 12.2 | 11.0 (7.9) | 8.5 (6.1) | 1.4 |
| $\Gamma$ | 17.0 | 15.7 | 13.8 | 9.6 (4.0) | 18 (7.5) | 2.4 |
| $\beta$ | $\Gamma$ | 27.3 | 16.7 | 16.3 (6.3) | 2.6 |
| $\Gamma$ | 30.0 | 17.4 | 17.9 (6.9) | 2.6 |
| $\epsilon$ | $\Gamma$ | 2.1  | 0.86 | 0.11 | 5.6 (18.7) | 6.0 (20) | 0.3 |
| $\Gamma$ | 1.29 | 0.36 | 7.2 (24) | 0.3 |

Table I. Cross-sectional areas and effective masses of FSs of KFe$_2$As$_2$ determined by ARPES and dHvA experiment. The areas are expressed as a percentage of the area of the 2D BZ. $m_e$ and $m_b$ are the free-electron and band masses, respectively.
with doping. For the $s$-wave pairing, there are two possibilities: horizontal nodes exist on the $xz/yz/z^2$ FS or the gap is small the entire $xy$ FS. In the present results, the $\beta$ hole FS which has $xy$ orbital character is much larger than that predicted by LDA. This implies that, if $s\pm$ paring is realized, the $\beta$ FS lays close to the nodal line of the $s\pm$ wave order parameter, resulting in a small superconducting gap or nodes.

Alternatively, horizontal nodes may be realized in the strongly warped region of the FS around the $Z$ point, where a significant amount of $3z^2$-$r^2$ character is hybridized. The small-angle neutron scattering measurement on KFe$_2$As$_2$ has suggested the existence of a horizontal node. If this is the case, the three-dimensionality of the $\zeta$ FS revealed by the present work may favor a horizontal node. In order to make a conclusive remark on the line nodes, ARPES measurement of the superconducting gap with higher energy resolution is necessary in future studies.

IV. CONCLUSION

We have studied FSs of KFe$_2$As$_2$ in three-dimensional momentum space by ARPES. All FSs except for the surface states are qualitatively consistent with the band-structure calculation. Particularly, some three-dimensionality in the $\zeta$ FS has been identified. The sizes of the FSs nearly accord with the dHvA observation: the $\alpha$ and $\epsilon$ FSs are smaller than those in the band-structure calculation while the $\beta$ and $\epsilon$ FSs are larger, which may be attributed to orbital-dependent electron-electron correlation effects.

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