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Characteristic two-dimensional Fermi surface topology of high- T_c iron-based superconductors

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Unconventional Cooper pairing originating from spin or orbital fluctuations has been proposed for iron-based superconductors. Such pairing may be enhanced by quasi-nesting of two-dimensional electron and hole-like Fermi surfaces (FS), which is considered an important ingredient for superconductivity at high critical temperatures (high- T_c). However, the dimensionality of the FS varies for hole and electron-doped systems, so the precise importance of this feature for high- T_c materials remains unclear. Here we demonstrate a phase of electron-doped CaFe_2As_2 (La and P co-doped CaFe_2As_2) with $T_c = 45$ K, which is the highest T_c found for the AEFe_2As_2 bulk superconductors (122-type; AE = Alkaline Earth), possesses only cylindrical hole- and electron-like FSs. This result indicates that FS topology consisting only of two-dimensional sheets is characteristic of both hole- and electron-doped 122-type high- T_c superconductors.

Iron-based superconductors are considered to be important for understanding high temperature superconductivity from a new perspective due to the presence of a high superconducting critical temperature (T_c) that cannot be explained within the framework of conventional phonon-mediated BCS superconductivity. Moreover, the electronic states of the parent compounds and the superconducting symmetry in these systems are different from those for high- T_c cuprate superconductors^{1–3}. Spin fluctuation-mediated superconductivity, as proposed for cuprate and heavy fermion systems, and orbital fluctuation-mediated superconductivity have been proposed as mechanisms for superconductivity^{4–7}. Such fluctuations are believed to be derived from the quasi-nesting between the hole- and electron-like Fermi surfaces (FSs) (Fig. 1a). There is much discussion surrounding the relevance of the FS topology to iron-based superconductivity.

In order to clarify the relevance of the Fermi surface to the mechanism of superconductivity in iron-based superconductors from an experimental perspective, numerous angle-resolved photoemission spectroscopy (ARPES) measurements, which are a direct measurement of the Fermi surface, have been conducted on iron-based superconductors^{8–27}. A number of substances with $T_c > 50$ K have been discovered for REFeAsO (1111-type; RE = Rare Earth) superconductors²⁸, which has drawn corresponding interest in the electronic structures that manifest such high values of T_c . However, few ARPES measurements have been performed in these materials, due to the difficulty in fabricating single crystals of large dimensions^{8–10}. In addition, bulk band structures are difficult to determine due to substantial surface effects originating from the existence of the REO layers¹⁰. As a result, little is known about the Fermi surface topology in the 1111-type materials. In contrast, numerous ARPES measurements have been performed for AEFe_2As_2 (122-type; AE = Alkaline Earth) iron-based superconductors^{17–27}, which do not possess REO layers. ARPES measurements are more feasible for this system due to the availability of larger single crystals and the lack of surface effects associated with REO layers. ARPES experiments in the 122 system are thus believed to more closely reflect bulk electronic structure. ARPES studies in the 122 system reveal the presence of hole-like and electron-like Fermi surfaces at the center of the Brillouin zone and the

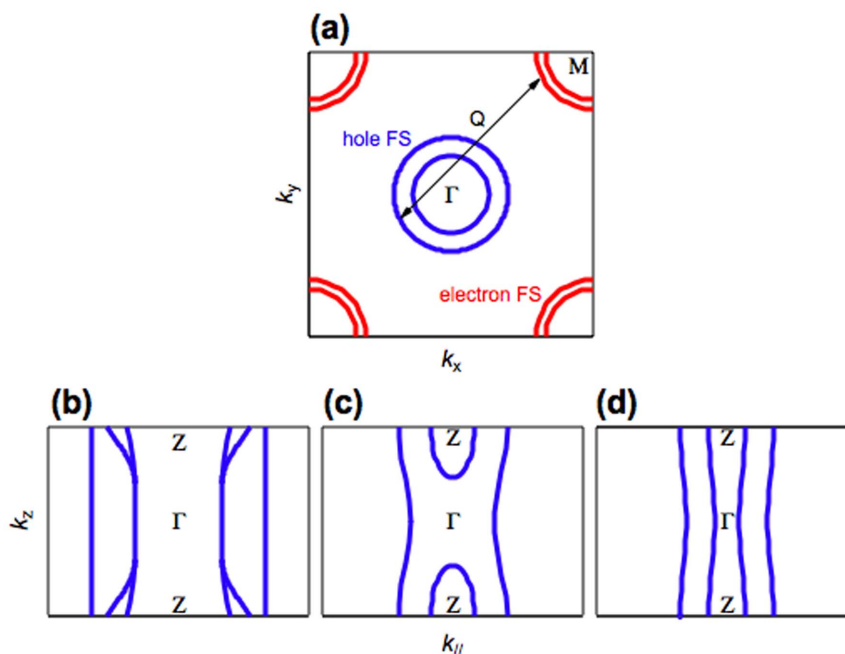


Figure 1 | Schematic Fermi surface topology. (a) Sketch of the band calculation results of Refs. 4 and 5, illustrating the Fermi surface (FS) and the nesting with nesting vector Q in the k_x - k_y plane for iron-based superconductors. The red and blue curved lines indicate electron- and hole-like FSs, respectively. (b) and (c) Sketch of the ARPES results of Refs. 24 and 26, illustrating the hole-like FSs around the zone center in the k_z - $k_{||}$ plane for $Ba_{1-x}K_xFe_2As_2$ and $Ba(Fe_{1-x}Co_x)_2As_2$. (d) Sketch of the present ARPES results for $Ca_{0.82}La_{0.18}Fe_2(As_{0.94}P_{0.06})_2$.

corner, respectively, and the cylindrical shape of the electron-like Fermi surfaces at the zone corner are common features of the electronic structure in high- T_c 122-type superconductors.

However, as shown in Fig. 1b and 1c, the shape of the Fermi surface at the zone center varies depending on the particular compound. The k_z dispersion is weak for the hole-doped 122-type $Ba_{1-x}K_xFe_2As_2$ ($T_c = 38$ K) and so the FSs is nearly two-dimensional^{24,25}. However, FSs with strongly three-dimensional oval shapes exist in the vicinity of the zone center for $Ba(Fe_{1-x}Co_x)_2As_2$ ($T_c = 25$ K)^{26,27}. Two-dimensional FSs, which are more conducive to nesting, are considered to be linked to the emergence of high- T_c in iron-based superconductors. However, since the FS dimensionality varies for these two iron-based superconductors with high- T_c , the importance of a two-dimensional Fermi surface topology is unclear.

An electron-doped $CaFe_2As_2$ superconductor derived by co-doping La and P was recently discovered with $T_c = 45$ K²⁹. The T_c of this new iron-based superconductor is the highest among the 122-type bulk superconductors that have ever been studied with ARPES. Thus, it is important to study the electronic structure using ARPES to clarify the FS topology of the iron-based superconductors at such a high T_c .

In our study, we revealed the Fermi surface of the electron-doped $CaFe_2As_2$. ARPES measurements were conducted with three photon polarizations to resolve multiple bands and tunable excitation photon energy to observe the three-dimensional shape of the Fermi surfaces (k_z dispersion of Fermi surfaces). All Fermi surfaces of this new superconductor had a weak k_z dispersion. Since this characteristic is common to $Ba_{1-x}K_xFe_2As_2$, which has the maximum T_c among hole-doped 122-type superconductors, the two-dimensional topology of the Fermi surface is relevant to the presence of high T_c in the iron-based superconductors and does not rely on the character of doped carriers.

Results

Polarization and photon energy dependent ARPES. In Fig. 2a–f, we show polarization dependent ARPES intensity plots (for the

geometrical measurement configuration, see Supplementary Fig. S1) along the cuts passing through the Γ point ($h\nu = 31$ eV) and the Z point ($h\nu = 19$ eV), together with peak positions determined from energy distribution curve (EDC) and momentum distribution curve (MDC) analyses (See, Supplementary Fig. S2 and S3 for corresponding intensity plots, intensity plots normalized with the Fermi-Dirac distribution function, and selected MDCs and EDCs.). At both points, ARPES intensity plots exhibit marked polarization dependence, which enables us to distinguish the dispersions of the three bands. At the Γ point, using ARPES data for s -polarized (s -pole) light (Fig. 2b, S2e–h), we found an intense hole-like dispersion with an energy maximum around 25 meV (α_1). The multiple structure of MDC at E_F (and its detailed line shape analysis) (Fig. S2g) also indicates the existence of E_F crossings of another band (α_2). In ARPES data for circular-polarized (c -pole) light (Fig. 2a, S2a–d), we found that the hole-like band (α_2) approaches and crosses E_F , as evident from multiple structures in the MDC at E_F (and its detailed line shape analysis) (Fig. S2c) and the parabolic dispersion in the EDCs (Fig. S2d). In addition, we found a hole-like feature (β) crossing E_F outside of the α_2 band in ARPES data for p -polarized (p -pole) light (Fig. 2c, S2i–k), which is more easily observed in MDCs for a higher binding energy region (Fig. S2k). At the Z point, while the intense hole-like α_1 band with an energy maximum around 10 meV is similarly found in s -pole data (Fig. 2e, S3d–f), the β band is observed in c - and p -pole data (Fig. 2d and 2f, S3a–c and S3g–h). Although intensities of the α_2 band are weaker than that of the β band at the Z point ($h\nu = 19$ eV), the existence of k_{FS} can be confirmed by the multiple structure in the MDC at E_F (and its detailed line shape analysis) taken with $h\nu = 82$ eV, which corresponds to another Z point (Supplementary Figs. S3i–k). These data and analyses confirm the existence of three bands consistent with band structure calculations^{4,5}.

Having established the existence of three bands, we now examine the k_z dispersion of the bands from photon energy-dependent ARPES. The peak positions of the normal emission spectra in Fig. 2h, corresponding to α_1 , measured with various photon energies

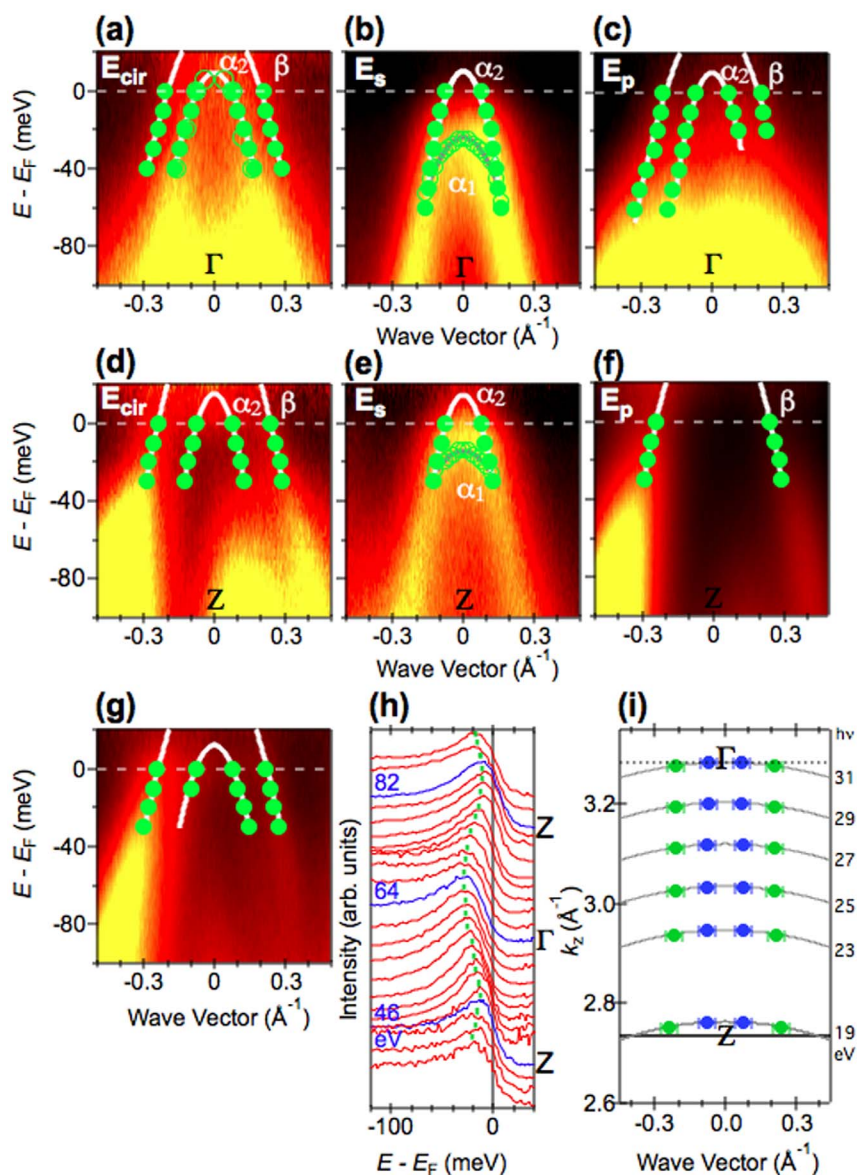


Figure 2 | Polarization and photon energy dependent ARPES data for $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ near the zone center. (a)–(c) ARPES intensity plots taken at $h\nu = 31$ eV ($k_z \sim \Gamma$) with circular (E_{clr}), s (E_s) and p (E_p) polarizations. (d)–(f) are the same as (a)–(c) but taken at $h\nu = 19$ eV ($k_z \sim Z$). (g) ARPES intensity plot taken at $h\nu = 23$ eV with circular polarization. In (a)–(g), intensities are divided by the Fermi-Dirac function. Filled and open circles denote peak positions determined from analyses of the MDCs and EDCs, respectively. (h) The EDCs divided by the Fermi-Dirac function at $k_{\parallel} = 0$ measured with various photon energies. (i) The k_{F} s determined from the MDC analysis shown in Supplementary Figure S5.

show a periodic variation consistent with the periodicity of the bulk Brillouin zone. More importantly, the peak positions are located below E_{F} for all k_z s measured, indicating that this band does not contribute to the FS. For α_2 and β , we found that hole-like Fermi surfaces are formed at both Γ and Z points. ARPES data at $h\nu = 23$ eV, corresponding to k_z between Γ and Z , also shows E_{F} crossings of these bands (Fig. 2g and S4a–d). We found that photon energy-dependent MDCs at E_{F} for the $[100]$ direction near the zone center can be well reproduced with four Lorentzian functions (Supplementary Fig. S5). We plotted the k_{F} s determined from fitting the MDCs at E_{F} in Fig. 2i. We now find that the α_2 and β bands form a nearly cylindrical small and large hole-like FS, respectively.

In Fig. 3a and 3b, ARPES intensity plots at E_{F} as functions of the two-dimensional wave vectors (k_x, k_y) around the M and A point, respectively, together with the positions of k_{F} (filled circles) obtained from fits to the MDC are shown. We observed two electron-like FSs. These FSs originate from the inner ε and outer δ electron-like bands,

as shown in Fig. 3c and 3d. We identified these bands from the detailed analysis of the MDCs (Supplementary Fig. S6a and S6b). We separate the two electron-like bands at the A point by performing polarization dependent ARPES taken at $h\nu = 24$ eV, which corresponds to another A point (Supplementary Fig. S6c and S6d). The elliptical shape of the intersections of the inner ε and outer δ electron-like FSs rotates 90° from M to A, which is consistent with the shape of the boundary of the body-centered tetragonal Brillouin zone. In Fig. 3e, we show the results of the FS mapping in the k_{\parallel} - k_z plane near the zone corner. The direction of k_{\parallel} is the same as the cuts in Fig. 3a and 3b. The electron-like FSs show a sizeable undulation along the k_z direction (Fig. 3e), reflecting the elliptical shape of these FSs and the shape of the zone boundary, as described above.

The Fermi surface topology determined by ARPES. From these ARPES measurements, we draw the shape of the FSs corresponding to the observed bands crossing E_{F} ($\alpha_2, \beta, \varepsilon, \delta$), as shown in Fig. 4a



and 4b. We now find four FSs: two around the zone center derived from the hole-like bands (α_2 , β) and two around the zone corner derived from the electron-like bands (ε , δ). All the FSs around the zone center and the corner are found to be nearly cylindrical with a small undulation. Here, hole-like FSs shifted by the AFM wave vector (π/a , π/a , $2\pi/c$) of CaFe_2As_2 ^{30,31}, where $a = 3.914 \text{ \AA}$ and $c = 11.48 \text{ \AA}$ are the in-plane and out-of-plane lattice constants of $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$, are shown with broken blue and sky blue lines in Fig. 4a. Some values for k_F of the inner ε electron-like FS overlap with those of the shifted β FS. Note that the back folding of the bands, which has been observed in the 122-type parent compounds³² and $\text{Ca}_{0.83}\text{La}_{0.17}\text{Fe}_2\text{As}_2$ with trace superconductivity³³, is absent in the present compound, consistent with the absence of AFM ordering. The total hole and electron count from the observed FS yields a hole volume of $6 \pm 2\%$ and an electron volume of $18 \pm 5\%$. Here, we assume that the hole-like parts of the FS are circular. The deduced total carrier number of 0.12 ± 0.07 electrons per Fe is consistent with the value of 0.09 electrons per Fe expected from the chemical composition, indicating that our measurements reflect the observation of the bulk electronic structure of La and P co-doped CaFe_2As_2 .

Discussion

We discuss the implication of the present ARPES results for iron-based superconductivity. As shown in Fig. 1b and 1d, all hole-like FSs in hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ and electron-doped $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ have a nearly two-dimensional shape^{24,25}. We find that a two-dimensional FS topology, which favors electron pair scattering between

quasi-nested FSs, is universal for high- T_c superconductivity regardless of the type of doped carrier. This observation supports the exotic pairing mechanisms proposed for iron-pnictide superconductors. Here it is noted that the quasi-nesting between hole- and electron-like FSs would not explain the high T_c of $\text{K}_x\text{Fe}_{1-y}\text{Se}_2$ superconductors where hole-like FSs are absent^{14–16}.

One of the most important questions is the origin of high- T_c superconductivity in the electron-doped superconductor $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ ($T_c = 45 \text{ K}$). We shall discuss the FS topology in three-dimensional momentum space. As described above, the size of the β hole-like FS is nearly the same as that of the ε electron-like FS and both FSs have a weak k_z dispersion, giving rise to a partial overlapping of k_F s. These observations are similar to that for electron-doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ ($T_c = 25 \text{ K}$)²². An observable difference between $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ and $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ in terms of FS topology is the dimensionality of the inner hole-like FS, as shown Fig. 1c and 1d. For $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, the inner hole-like FS shows a strong k_z dispersion and is closed near the Γ point^{25,26}. In contrast, the inner hole-like FS (α_2) shows a cylindrical shape and survives near the Γ point in $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$. These results suggest that an enhancement of the two-dimensionality of the inner hole-like FS induces a large T_c difference in the two materials, possibly due to an increased

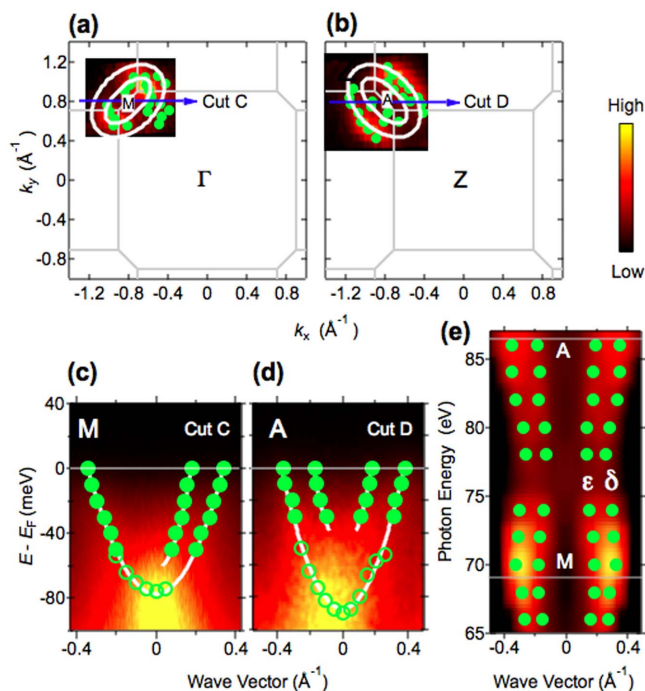


Figure 3 | Photon energy-dependent ARPES data for $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ around the zone corner. (a),(b) ARPES intensity plots at E_F as functions of two-dimensional wave vectors taken at $h\nu = 69 \text{ eV}$ and $h\nu = 86 \text{ eV}$, respectively, around M and A. (c),(d) ARPES intensity plots along cuts C and D, respectively. Cuts C and D are shown by blue arrows in (a) and (b). In these plots, $k_{||} = 0$ corresponds to the (π, π) point. Filled and open circles indicate the peak position of the MDCs and EDCs, respectively. (e) ARPES intensity plot at E_F as a function of photon energy, together with k_{FS} (green dots) determined from the MDC analysis. The direction of $k_{||}$ is the same as (c) and (d) ($[100]$ direction). The intensities are symmetrized about the $k_{||} = 0$. In this plot, $k_{||} = 0$ also corresponds to (π, π) point.

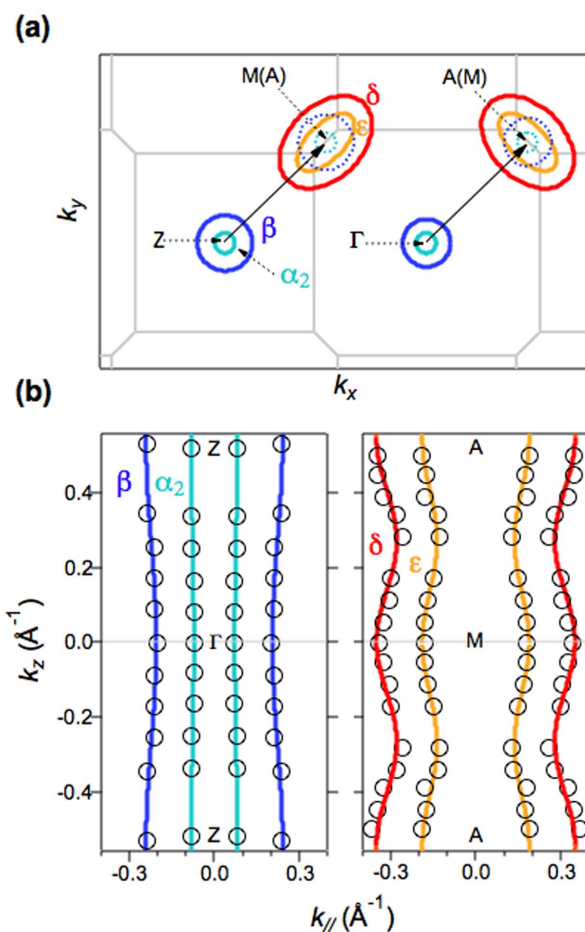


Figure 4 | Fermi surface shape for $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ determined by ARPES. The shapes of Fermi surfaces (FSs) in the (a) k_x - k_y and (b) k_z - $k_{||}$ planes are drawn with lines. Dotted blue and sky blue lines in (a) are two hole-like FSs around the zone center shifted by the antiferromagnetic vector (black arrows). In (b), black open circles represent experimentally determined k_{FS} from photon energy-dependent ARPES. The positions of k_F have been symmetrized with respect to the symmetry lines.



tendency toward quasi-nesting between α_2 and ε parts of the FS. Regarding the orbital character of the bands in $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$, the polarization dependence of the odd symmetry with respect to the mirror plane and k_z dispersion of α_1 , enable us to suggest a d_{YZ} character for α_1 , where X and Y refer to the direction rotated by 45 degrees from the Fe-Fe direction and Z is normal to the XY plane. The observable ARPES intensity of the α_2 band with both *s*-pole and *p*-pole light (Fig. 2a–c) suggests a mixed orbital character of even and odd symmetry. The mixed orbital character of bands near E_F (d_{XY} and $d_{X^2-Y^2}$) were reported from polarization dependent ARPES of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ and LiFeAs ^{11,19}. The β band has even symmetry. The lifting of the hole-like band degeneracy at the Γ point does not agree with previous calculations^{4,5}, where bands with d_{XZ} and d_{YZ} orbital characters are degenerate. However, recent calculations in Refs. 34 and 35 taking into account the change of the Fe-As-Fe bond angle and the change of the positive charge in the blocking layer predicted the lift of the degeneracy, in agreement with the present study.

In iron-based superconductors, band structure calculations predict that the shapes of the hole-like FSs become more three-dimensional with a reduction in pnictogen height³⁶. This theoretical prediction holds for $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ and $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, as the pnictogen height of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ is shorter than that of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ^{37,38}. For $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$, the pnictogen height is not available thus far. As for the *c* parameter, that of $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ is the shortest among the three compounds. However, this does not necessarily mean a shorter pnictogen height in $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$, due to the difference in alkaline earths. In order to verify this relationship, detailed structural studies of $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ are indispensable.

In summary, we have investigated the three-dimensional electronic structure near E_F in electron-doped $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ ($T_c = 45$ K). The observed FS topology is nearly two-dimensional and similar to that of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, indicating the existence of universality in the FS topology for realizing high- T_c in 122-type superconductors.

Methods

High quality $\text{Ca}_{0.82}\text{La}_{0.18}\text{Fe}_2(\text{As}_{0.94}\text{P}_{0.06})_2$ single crystals were grown as described elsewhere²⁵. Polarization dependent ARPES measurements ($h\nu = 19\text{--}31$ eV) were carried out at BL-9A of Hiroshima Synchrotron Radiation Center (HSRC). ARPES measurements using circularly polarized light ($h\nu = 40\text{--}86$ eV) were carried out at BL-28A of the Photon Factory. The total energy resolution was set to 10–30 meV. Clean surfaces were obtained by *in situ* cleaving of the crystal in a working vacuum better than 3×10^{-8} Pa and measured at 60 K (above T_c). The inner potential was determined to be 14 eV from photon energy-dependent ARPES studies as described above. Calibration of E_F of the sample was achieved using a gold reference.

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Author contributions

M.S., T.I., K.T., T.J. and J.S. performed ARPES experiments. K.O., H.K., M.A., K.S., H.N. and M.T. supported Synchrotron ARPES experiments. M.S. analyzed data. M.S. and T.Y. designed experiments. K.I., K.K. and M.N. synthesized single crystals. M.S., K.K., M.N., T.M., T.W., Y.M. and T.Y. discussed the results and M.S. and T.Y. wrote the manuscript.

Additional information

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