

# Nodal $s_{\pm}$ Pairing Symmetry in an Iron-Based Superconductor with only Hole Pockets

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The origin of the high temperature superconductivity in the iron-based superconductors is still not understood and determination of the pairing symmetry is essential in understanding the superconductivity mechanism. In the iron-based superconductors that have hole pockets around the Brillouin zone center and electron pockets around the zone corners, the pairing symmetry is generally considered to be  $s_{\pm}$ , which indicates a sign change in the superconducting gap between the hole and electron pockets. For the iron-based superconductors with only hole pockets, however, a couple of pairing scenarios have been proposed but the exact symmetry is still controversial. Here we determine that the pairing symmetry in  $\text{KFe}_2\text{As}_2$  — which is a prototypical iron-based superconductor with hole pockets both around the zone center and around the zone corners — is also of the  $s_{\pm}$  type. Our laser-based angle-resolved photoemission measurements have determined the superconducting gap distribution and identified the locations of the gap nodes on all the Fermi surface around the zone center and the zone corners. These results unify the pairing symmetry in the hole-doped iron-based superconductors and point to the spin fluctuation as the pairing glue in generating superconductivity.

The superconductivity mechanism of the iron-based superconductors, discovered more than a decade ago[1–5], remains an outstanding issue in condensed matter physics[6–9]. The identification of the pairing symmetry and the revelation of the pairing medium are two key prerequisites in understanding the superconductivity mechanism. As a typical multiorbital system, a number of pairing symmetries have been proposed in iron-based superconductors that are associated with different pairing glues[6–9]. In most iron-based superconductors with the hole-like Fermi surface around the zone center and the electron-like Fermi surface around the zone corners, an  $s_{\pm}$  pairing symmetry is expected from spin fluctuations based on weak coupling Fermi surface nesting picture[10, 11] or strong coupling local magnetic picture[12, 13]. In some iron-based superconductors with only electron-like Fermi surface[14–18], the  $d$ -wave pairing is found to be dominant from electron scatterings between the zone corners by spin fluctuations[11, 19, 20] and a transition from  $d$ -wave to  $s$ -wave was proposed with increasing band hybridization[21]. For the iron-based superconductors with only hole-like Fermi surface, there is still a controversy about whether the pairing is nodal or

nodeless[22–32] and whether it is  $d$  wave or  $s$  wave[23–42]. Direct determination of the gap structure in the purely hole-doped iron-based superconductors is important to ascertain its pairing symmetry and establish a unified picture of the pairing mechanism in the iron-based superconductors.

As the end member of the hole-doped  $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ ,  $\text{KFe}_2\text{As}_2$  is a prototypical system for studying superconductivity mechanism in the purely hole-doped iron-based superconductors. It has unique electronic structure that consists of only hole pockets[43–45]. It has been found that  $\text{KFe}_2\text{As}_2$  has a large Sommerfeld coefficient which behaves similarly to heavy Fermion materials[44, 46–51], indicating the presence of strong electron correlation due to its proximity to the putative Mott insulating state[52]. However, the electronic origin of its heavy-electron behaviors remains elusive. More importantly, although extensive studies have been carried out to probe the pairing symmetry of  $\text{KFe}_2\text{As}_2$ , the pairing nature has not been pinned down and remain controversial[22–26, 28–32, 38–42].

In order to determine the superconducting gap structure of  $\text{KFe}_2\text{As}_2$  and other unconventional superconductors with low  $T_C$  and small superconducting gap, we have developed a laser ARPES system with ultra-low temperature ( $\sim 0.8$  K) and super-high energy resolution ( $< 0.5$  meV) (see Methods). We also grew high quality  $\text{KFe}_2\text{As}_2$  single crystals with a  $T_C$  at 3.7 K and a sharp superconducting transition ( $\sim 0.2$  K) (see Methods and Fig. S1 in Supplementary Materials). We have clearly resolved the complete Fermi surface topology and band structures of  $\text{KFe}_2\text{As}_2$ , particularly those near the Brillouin zone corners by taking the advantage of surface reconstruction. We have precisely determined the momentum-dependent superconducting gap on each Fermi surface and identified the locations of the gap nodes. These results unravel the electronic origin of the huge mass enhancement and strong electron correlation in  $\text{KFe}_2\text{As}_2$ . They also provide strong evidence on the  $s_{\pm}$  pairing symmetry with a sign reversal between the zone center and zone corners in the system with only hole pockets.

Figure 1 shows the measured Fermi surface and band structures of  $\text{KFe}_2\text{As}_2$ . As a typical multi-orbital system, we find that strong photoemission matrix element effects are involved in the ARPES measurements of  $\text{KFe}_2\text{As}_2$ . Therefore, in order to fully resolve the Fermi surface and band structures, we carried out ARPES measurements using different light polarizations (Fig. S2 in Supplementary Materials). Combining the Fermi surface mapping

(Fig. 1a) and the associated band structures (Fig. 1c-f and Fig. S3 in Supplementary Materials) measured under various polarization geometries, we have obtained the complete Fermi surface picture of  $\text{KFe}_2\text{As}_2$ , as shown in Fig. 1b. It consists of three main Fermi surface sheets around  $\Gamma$  ( $\alpha$ ,  $\beta$  and  $\gamma$  in Fig. 1b), a small surface state sheet around  $\Gamma$  (SS in Fig. 1b) and four hole pockets around M ( $\varepsilon$  in Fig. 1b). The orbital characters of the Fermi surface are analyzed and shown in Fig. S4 and Tables S1-S2, as described in Supplementary Materials. Our more precise measurements of the Fermi surface (Fig. 1b) indicate that it is rather similar to the Fermi surface at  $k_z \sim 0$  from the previous three-dimensional ARPES measurements[45], indicating that the measured momentum region is close to  $\Gamma$  point ( $k_z \sim 0$ ) in our ARPES results using 7-eV photons. We find that the  $\beta$  Fermi surface further splits into two sheets ( $\beta_1$  and  $\beta_2$  in Fig. 1b) with the maximum splitting along  $\Gamma$ -X, as seen from the band structure in Fig. 1f. Our further synchrotron-based ARPES measurements indicate that the  $\beta_1$  band is from the bulk state while the  $\beta_2$  band can be attributed to a surface state (Fig. S5 and Fig. S6 in Supplementary Materials).

Although laser ARPES is advantageous in taking high quality data with superhigh resolution, it has a limitation in the covered momentum space due to its lower photon energy[26, 53]. Particularly for the iron-based superconductors, laser ARPES can cover mainly the zone center region but cannot reach the zone corner M points[26, 53]. It is interesting and significant that in our present study we can observe the Fermi surface and band structures around M that are folded to the region around  $\Gamma$ . Under a proper polarization geometry, four Fermi pockets can be observed around  $\Gamma$  which are marked by dashed pink lines and labeled as  $\varepsilon'$  in Fig. 1a. The corresponding band structures ( $\varepsilon'_L$  and  $\varepsilon'_R$  in Fig. 1c and 1d) can be clearly resolved and their spectral intensity is comparable to that of  $\alpha$ ,  $\beta$  and  $\gamma$  bands. These four  $\varepsilon'$  pockets can be well attributed to the four  $\varepsilon$  hole pockets around M that are folded to the  $\Gamma$  point by the  $\sqrt{2} \times \sqrt{2}$  surface reconstruction (Fig. S7, Fig. S8, Fig. S9 and Fig. S10 in Supplementary Materials). Such a band folding makes it possible to study the electronic structure and superconducting gap of  $\text{KFe}_2\text{As}_2$  in the full momentum space by laser ARPES.

Figure 2 highlights the band structures and photoemission spectra measured in the normal and superconducting states. Clear quasiparticle peaks are observed in the photoemission spectra (energy distribution curves, EDCs) measured along all the Fermi surface sheets at 4.3 K in the normal state (red curves in left panels of Fig. 2e-1). Superconducting coherence

peaks develop in some EDCs in the superconducting state at 0.9 K (blue curve in left panels of Fig. 2e,f,k). The observed peaks are rather sharp with a width (full width at half maximum, FWHM) of 1~2 meV. In order to visualize the gap opening, band symmetrization or EDC symmetrization with respect to the Fermi level is usually carried out to remove the Fermi distribution function[54]. In this case, the gap opening corresponds to the spectral weight suppression at the Fermi level. As visualized in Fig. 2b,c, in the superconducting state at 0.9 K, the spectral weight at the Fermi level for the  $\alpha$ ,  $\beta$ ,  $\varepsilon'_L$  and SS bands is clearly suppressed indicating the opening of the superconducting gap. In the symmetrized EDCs measured at 0.9 K (blue curves in the right panels of Fig. 2e-l), the spectral weight suppression at the Fermi level is observed for P1 on  $\alpha$  band (Fig. 2e), P2 on  $\beta$  band (Fig. 2f) and P7 on  $\varepsilon'_L$  band (Fig. 2k), signaling the superconducting gap opening (the location of the Fermi momentum points P1-P8 is shown in Fig. 2d). The gap size can be determined from the energy difference between the two peaks in the symmetrized EDCs. On the other hand, the symmetrized EDCs in the superconducting state exhibit a peak at the Fermi level at other momentum points (blue curves in Fig. 2g,h,i,j,l) indicating that no superconducting gap opening is detected within our experimental precision. In all the symmetrized EDCs measured at 4.3 K in the normal state (red curves in right panels of Fig. 2e-l), there is no signature of gap opening observed, excluding the formation of pseudogap in  $\text{KFe}_2\text{As}_2$ .

It was found that  $\text{KFe}_2\text{As}_2$  exhibits heavy Fermion behaviours manifested by its large Sommerfeld coefficient and heavy effective mass[44, 46–50]. Our present high resolution band structure measurements provide key information in understanding its origin. Fig. 3a shows a detailed band structure measured at 0.9 K along a momentum cut that is nearly parallel to  $\Gamma$ -M direction. To directly visualize the gap opening and remove the effect of the Fermi cutoff, the image is obtained by symmetrizing the original data with respect to the Fermi level. The corresponding EDCs and symmetrized EDCs are shown in Fig. 3c and 3d, respectively. The band structure of  $\varepsilon'$  in the superconducting state,  $E_k$ , can be directly extracted from the symmetrized EDCs (Fig. 3d) and plotted in Fig. 3e as pink circles. The superconducting gap can also be determined directly from the symmetrized EDCs (Fig. 3d) which are 0.8 meV and 0 for the left and right Fermi momenta of the  $\varepsilon'$  band ( $k_F^{\varepsilon'_L}$  and  $k_F^{\varepsilon'_R}$ ), respectively. Assuming a linear variation of the superconducting gap between the two Fermi momenta ( $\Delta_k$ ), we obtain the normal state  $\varepsilon'$  band structure ( $e_k$ ) by using the BCS relation  $E_k^2 = e_k^2 + \Delta_k^2$ , as plotted by the blue line in Fig. 3e. The normal state  $\varepsilon'$  band is

strikingly flat and its band top is only  $\sim 1$  meV above the Fermi level. The effective mass obtained from the left side and the right side of the  $\varepsilon'$  band is  $3.2m_e$  and  $13.8m_e$ , respectively, with  $m_e$  being the free electron mass. Such a dramatic anisotropy of the effective mass is understandable because the two sides of the  $\varepsilon'$  band come from different orbitals as seen in Fig. 3f. In particular, when compared with the band structure calculations (Fig. 3f)[46], the effective mass for the left side and right side  $\varepsilon$  band reaches  $16m_b$  and  $8.1m_b$  with  $m_b$  being the corresponding band masses. The Fermi energy of the  $\varepsilon$  band shrinks dramatically from the calculated  $\sim 20$  meV to the measured  $\sim 1$  meV. Such a huge band renormalization of the  $\varepsilon$  band is among the strongest that have been observed in the iron-based superconductors[55].

Our detailed band structure measurements indicate that the large specific heat and the associated heavy Fermion behaviours discovered in  $\text{KFe}_2\text{As}_2$ [44, 46–51] originate from the strong band renormalizations, particularly the  $\varepsilon$  band. Similar to the  $\varepsilon'$  band in Fig. 3e,f, we carried out band structure analysis of the  $\alpha$ ,  $\beta$  and  $\gamma$  bands along different directions (Fig. S11 in Supplementary Materials) to extract the effective mass and the Sommerfeld coefficient  $\gamma$ , as shown in Fig. S11e,f and listed in Table S3 in Supplementary Materials. All the bands exhibit strongly enhanced effective mass with the  $\gamma$  band reaching over 20 times of  $m_e$  (Fig. S11e and Table S3 in Supplementary Materials). When compared with the calculated band structures, the  $\varepsilon$  band shows the strongest renormalization with  $m^*/m_b$  over 15 (Fig. S11f and Table S3 in Supplementary Materials). The anomalously large specific heat found in  $\text{KFe}_2\text{As}_2$  comes from the combined contributions of all these renormalized bands. In spite of its small Fermi surface area, the  $\varepsilon$  band plays a significant role in contributing to the specific heat; nearly half of the Sommerfeld coefficient is contributed by the  $\varepsilon$  band (Table S3 in Supplementary Materials).

Now we come to the determination of the superconducting gap in  $\text{KFe}_2\text{As}_2$ . Since  $\text{KFe}_2\text{As}_2$  is a typical multiorbital system and strong matrix element effects are involved in the ARPES measurements (Fig. 1), in order to extract the superconducting gap along all the Fermi surface sheets, we carried out ARPES measurements along different momentum cuts under different polarization geometries and repeated the measurements on many  $\text{KFe}_2\text{As}_2$  samples (Fig. S12-S17 in Supplementary Materials for the  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\varepsilon'$  and SS, respectively). Fig. 4a-e shows the symmetrized EDCs along the  $\alpha$ ,  $\beta_1$ ,  $\beta_2$ ,  $\gamma$  and  $\varepsilon'$  Fermi surface sheets. The gap size is determined by the peak position in the symmetrized EDCs[54] (for detailed discussion of the gap determination, see Fig. S18, S19 and S20 in Supplementary Materials).

The measured superconducting gap along the  $\alpha$ ,  $\beta_1$ ,  $\beta_2$  and  $\gamma$  Fermi surface is presented in Fig. 4f and the gap along the  $\varepsilon$  Fermi surface is shown in Fig. 4g. The superconducting gap along the  $\varepsilon$  Fermi surface was not measured in the previous ARPES measurement[26] and, for the first time, it has been measured here. Fig. 4i shows a three-dimensional plot of the measured superconducting gap structure in  $\text{KFe}_2\text{As}_2$ . As seen in Fig. 4f, the  $\alpha$  Fermi surface shows a large superconducting gap ( $\sim 1.0$  meV) which is nearly isotropic with a small variation of  $\pm 0.1$  meV. The superconducting gap of the  $\beta$  Fermi surface ( $\beta_1$  and  $\beta_2$ ) is highly anisotropic. It is nearly zero along the  $\Gamma$ -X direction and reaches the maximum ( $\sim 1.0$  meV) along the  $\Gamma$ -M direction. Along the entire  $\gamma$  Fermi surface, the gap is nearly zero within our detection limit. We note that our results of the superconducting gap along the  $\alpha$ ,  $\beta$  and  $\gamma$  Fermi surface around  $\Gamma$  (Fig. 4f) show significant difference from those reported before[26] (the reason of the discrepancy is discussed in Supplementary Materials). As seen in Fig. 4g, the superconducting gap on the small  $\varepsilon$  pocket is highly anisotropic. Along the  $\Gamma$ -M direction, it is the maximum near the tip close to M while it approaches zero on the other side away from M. The superconducting gap structure in  $\text{KFe}_2\text{As}_2$  is quite distinct along different Fermi surface sheets.

Our precise determination of the electronic structure and superconducting gap structure, particularly for the  $\varepsilon$  Fermi surface around M, provides a new picture of the pairing symmetry in  $\text{KFe}_2\text{As}_2$ . We find that our results are more consistent with the  $s_{\pm}$  pairing symmetry with the sign reversal between the  $\Gamma$  and M points based on the following observations. First, the superconducting gap size along all the Fermi surface sheets is qualitatively consistent with the simple  $s_{\pm}$  form:  $|\Delta| = \Delta_0 |\cos k_x + \cos k_y|$ , as shown in Fig. 5a. In the  $s_{\pm}$  pairing symmetry with the simplest form, the gap size is expected to decrease with the increasing distance of the momentum point from  $\Gamma$  or M (thick black line and thick red line in Fig. 5a). The measured superconducting gap of  $\text{KFe}_2\text{As}_2$ , extracted from many data points on different Fermi surface sheets, is found to also decrease with the increasing distance of the momentum point from  $\Gamma$  or M (data points in Fig. 5a). Second, precise nesting vectors along  $\Gamma$ -M can be determined from our measured Fermi surface (Fig. 5b). They are consistent with the previous ARPES measurements[45] and show rather weak  $k_z$  dependence[45]. If the nestings connect the  $\alpha$  and  $\beta$  Fermi surface around  $\Gamma$  as proposed before[35, 36, 42], the corresponding nesting vectors obtained from Fig. 5b are  $0.61 \pi/a$  and  $2.21 \pi/a$  along the  $\Gamma$ -M direction which significantly deviate from the two spin resonance wavevectors of  $0.85 \pi/a$  and  $1.98 \pi/a$

from neutron scattering measurements[42] (Fig. S21 in Supplementary Materials). On the other hand, when the two vectors connect the best nested portion of the  $\varepsilon$  Fermi surface around M and the  $\alpha/\beta$  Fermi surface around  $\Gamma$ , as we identified in Fig. 5b, the obtained wavevectors of  $\mathbf{Q}_1=0.87\pi/a$  and  $\mathbf{Q}_2=1.96\pi/a$  have a perfect match to the two spin resonance wavevectors[42]. This indicates that the two regions connected by the nesting vectors have the reversed sign of the superconducting gap. This is consistent with the  $s_{\pm}$  form  $\Delta = \Delta_0(\cos k_x + \cos k_y)$ , as depicted in Fig. 5b. Neutron scattering measurements found that the spin resonance peak in  $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$  gets weaker with increasing doping in the overdoped region due to the increasing mismatch between the hole pocket around  $\Gamma$  and electron pocket around M[56, 57]. In  $\text{KFe}_2\text{As}_2$  where the  $\varepsilon$  hole pockets emerge around M, the spin resonance peak is observed but is much weaker than that in optimally-doped  $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ [42]. This is consistent with the Fermi surface nesting but with a relatively small nested area in  $\text{KFe}_2\text{As}_2$ . Third, the nesting vectors connect portions of the two Fermi surface that have the maximum superconducting gap (black solid circles in Fig. 5b). It has been found that the energy of the resonance mode  $E_R$  and the superconducting gap  $\Delta_{tot}$  follow a universal relation of  $E_R/\Delta_{tot} \approx 0.64$  in the iron-based superconductors with  $\Delta_{tot}$  being the total superconducting gap summed on the two Fermi surfaces linked by the nesting vectors[58]. The total gap size  $\Delta_{tot} = 2.0\text{ meV}$  where  $1.0\text{ meV}$  is from the  $\alpha/\beta$  Fermi surface while the other  $1.0\text{ meV}$  is from the  $\varepsilon$  Fermi surface. The energy of the resonance mode in  $\text{KFe}_2\text{As}_2$  is then estimated to be  $1.28\text{ meV}$  which is in a good agreement with that measured in neutron scattering ( $1.2\text{ meV}$ )[42]. Fourth, from the measured superconducting gap structure, we have identified the nearly zero gap points on the  $\beta$ ,  $\gamma$  and  $\varepsilon$  Fermi surface, as marked by the white circles in Fig. 5b. In the  $s_{\pm}$  gap symmetry, there are zero gap nodal regions along  $(0, \pm\pi)$ - $(\pm\pi, 0)$  lines (Fig. 5b). It is interesting to note that the measured momentum region with nearly zero gap on the  $\beta$  and  $\varepsilon$  Fermi surface is located closest to the zero gap lines of the  $s_{\pm}$  form. In particular, the  $\gamma$  Fermi surface lies very close to the  $s_{\pm}$  nodal lines and the superconducting gap is nearly zero along the entire Fermi surface. These results indicate that the sign reversal  $s_{\pm}$  pairing symmetry can be realized in the system with only hole pockets. Here the sign change occurs between the hole pockets around  $\Gamma$  and those around M. Previously the  $s_{\pm}$  pairing symmetry was proposed and tested in the superconductors with hole pockets around  $\Gamma$  and electron pockets around M[10, 11, 13, 59, 60]. Neutron scatterings have observed spin fluctuations in  $\text{KFe}_2\text{As}_2$ [42, 61, 62] and spin fluctuations exchange always

leads to a repulsive interaction and can only realize sign-changing superconducting states[6]. Therefore, our identification of the nodal  $s_{\pm}$  pairing symmetry in  $\text{KFe}_2\text{As}_2$  indicates that the spin fluctuations may play a dominant role in generating superconductivity even in a system with only hole pockets.

The determination of the pairing symmetry is essential in understanding the superconductivity mechanism of iron-based superconductors. Our present work has significant implications on the pairing symmetry in  $\text{KFe}_2\text{As}_2$ . The pairing symmetry in heavily hole-doped iron-based superconductors with only hole pockets has been extensively studied theoretically and experimentally. Various possibilities have been proposed and debated, including the competition of  $s_{\pm}$  ( $\Gamma$ -M) and  $d$ [37], pure extended  $d$ [33, 63], pure  $s_{\pm}$  ( $\Gamma$ -M)[64], the competition between  $s_{\pm}$  (two hole pockets around  $\Gamma$ ) and  $d$ [34, 65], and pure  $s_{\pm}$  (two hole pockets around around the zone center)[35, 36]. In particular, based on the previous ARPES results on the superconducting gap of  $\text{KFe}_2\text{As}_2$ [26], the pure  $s_{\pm}$  with the sign reversal between the two hole pockets ( $h_1$  and  $h_2$ ) around the zone center has been focused [35, 36]. The neutron scattering measurements on  $\text{KFe}_2\text{As}_2$  are considered to be consistent with this  $s_{\pm}$  ( $h_1$ - $h_2$ ) symmetry[42]. Our present results have corrected the previous ARPES results[26], pinned down on a new pairing picture in  $\text{KFe}_2\text{As}_2$  and provided key information in theoretically understanding the pairing symmetry in iron-based superconductors with only hole pockets. Furthermore, our results have provided a unified picture of the pairing symmetry in iron-based superconductors with distinct Fermi surface topologies. As a typical multiband system, the pairing symmetry in iron-based superconductors is usually considered to be closely related to the Fermi surface topology. When there are both hole-like Fermi surface around  $\Gamma$  and electron-like Fermi surface around M, the pairing symmetry is generally believed to be  $s_{\pm}$  with the sign change occurring between  $\Gamma$  and M[6–13, 66]. When there are only hole pockets in heavily hole-doped iron-based superconductors, the pairing symmetry is highly controversial [33–37, 63–65] and a pairing symmetry transition is proposed with increasing hole doping[33, 34, 41, 63, 65]. Our finding of a common  $s_{\pm}$  ( $\Gamma$ -M) pairing symmetry indicates that the system with both the electron and hole pockets and the system with only hole pockets may share similar pairing mechanism that can be described by a unified theoretical framework.

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### **Author Contributions**

X.J.Z., L.Z. and D.S.W. proposed and designed the research. D.S.W., W.S.H., Y.J.S, H.Q.L and S.L.L. contributed in sample growth and the magnetic and resistivity measurements. J.J.J., J.G.Y., H.T.Y, H.T.R, P.A., X.Z., C.H.Y., T.M.M., C.L.L., S.J.Z., F.F.Z., F.Y., Z.M.W., N.Z., L.J.L., R.K.L., X.Y.W., Q.J.P., H.Q.M., G.D.L., Z.Y.X., L.Z. and X.J.Z. contributed to the development and maintenance of the ARPES systems and related software development. D.S.W. carried out the ARPES experiment with J.J.J., J.G.Y., J.Y.L., H.K.C., Y.H.Y., C.P., Y.L.C., L.Z. and X.J.Z.. D.S.W., L.Z. and X.J.Z. analyzed the data. X.X.W. contributed to the band structure calculations. X.J.Z., L.Z. and D.S.W. wrote the paper. All authors participated in discussion and comment on the paper.

### **Competing Interests Statement**

The authors declare that they have no competing financial interests.

## Figure Captions

FIG. 1. **Fermi surface and band structures of  $\text{KFe}_2\text{As}_2$  measured at 0.9 K.** **a**, Fermi surface mapping near the zone center  $\Gamma$  point. It is obtained by integrating the spectral intensity within  $\pm 1$  meV with respect to the Fermi level and symmetrized assuming fourfold symmetry. **b**, Measured Fermi surface obtained by analyzing the Fermi surface mappings and band structures measured under different polarization geometries (Fig. S2). The four  $\varepsilon'$  pockets around  $\Gamma$  are produced from folding the four  $\varepsilon$  pockets around M due to  $\sqrt{2} \times \sqrt{2}$  surface reconstruction. **c-f**, Typical band structures measured along different momentum cuts under different polarization geometries. The location of the momentum cuts is marked in **b**. The observed bands are marked by arrows with different colors.

**FIG. 2. Superconducting gap opening along different Fermi surface sheets by measuring photoemission spectra in the normal and superconducting states.** **a-c**, Band structures measured along the same momentum cut at different temperatures under different polarization geometries. The location of the momentum cut (Cut1) is marked by the black line in **d**. **a** is measured in the normal state at 4.3 K under the s polarization. **b** is the same as **a** but measured at 0.9 K in the superconducting state. **c** is the same as **b** but measured under the p polarization. To directly visualize the gap opening, these images are obtained by symmetrizing the original data with respect to the Fermi level. **d**, Fermi surface of  $\text{KFe}_2\text{As}_2$  marked with the momentum cut used in **a-c** and the Fermi momentum points P1-P8 used in **e-l**. **e**, EDCs (left panel) and the corresponding symmetrized EDCs (right panel) measured at the Fermi momentum P1 along the  $\alpha$  Fermi surface in the normal (4.3 K) and superconducting (0.9 K) states. The location of the P1 point is marked in **d**. **f-h**, Same as **e** but measured at the Fermi momenta P2 (**f**), P3 (**g**) and P4 (**h**) along the  $\beta$  Fermi surface sheets ( $\beta_1$  and  $\beta_2$ ). **i-j**, Same as **e** but measured at the Fermi momenta P5 (**i**) and P6 (**j**) along the  $\gamma$  Fermi surface sheet. **k-l**, Same as **e** but measured at the Fermi momenta P7 (**k**) and P8 (**l**) along the  $\varepsilon'$  Fermi surface sheet. To facilitate a comparison, in **k**, the two EDCs measured at 0.9 K under s and p polarizations are normalized in intensity near the Fermi level.

**FIG. 3. Detailed analysis of the  $\varepsilon$  band in the superconducting state.** **a**, Band structure measured along a momentum cut that is nearly parallel to the  $\Gamma$ -M direction. The location of the momentum cut (Cut1) is marked by the black line in **b**. The image is obtained by symmetrizing the original data with respect to the Fermi level. The observed bands,  $\alpha$ ,  $\beta_{1,2}$  and  $\varepsilon'$ , are marked by black, purple and pink lines, respectively. **b**, Fermi surface of  $\text{KFe}_2\text{As}_2$  marked with the momentum cut (Cut1) used in **a**. **c**, Original EDCs from the original data in **a** before symmetrization in the momentum range marked by the black line with an arrow on top of **a**. The EDCs at the Fermi momenta of the  $\alpha$ ,  $\beta_{1,2}$  and  $\varepsilon'$  bands are marked by colors. **d**, The corresponding symmetrized EDCs obtained from **c**. The EDC peaks corresponding to  $\varepsilon'$  band and its Bogoliubov band are marked by pink ticks. **e**, Quantitative determination of the  $\varepsilon'$  band dispersion (pink circles) in the superconducting state from **a** and **d**, which is described by continuous pink lines ( $E_k$ ). Since the gap size is zero at the right Fermi momentum ( $k_F^{\varepsilon'R}$ ) and 0.8 meV at the left Fermi momentum ( $k_F^{\varepsilon'L}$ ), a linear variation of the gap,  $\Delta_k$ , between the Fermi momenta is assumed (brown line). The dispersion in the normal state  $\varepsilon_k$  (blue curve) can then be derived using the equation  $E_k^2 = e_k^2 + \Delta_k^2$  from BCS theory[67]. **f**, The calculated band structure shows where the measured  $\varepsilon$  band is located with a black dashed frame. The measured  $\varepsilon$  band is also plotted (blue dashed line) for a direct comparison.

FIG. 4. **Momentum-dependent superconducting gaps along all the Fermi surface sheets of  $\text{KFe}_2\text{As}_2$ .** **a-e**, Symmetrized EDCs along the  $\alpha$  (**a**),  $\beta_1$  (**b**),  $\beta_2$  (**c**),  $\gamma$  (**d**) and  $\varepsilon'$  (**e**) Fermi surface sheets, respectively. The Fermi momentum positions are marked by empty circles in the inset Fermi surface. The details of extracting these symmetrized EDCs are described in Fig. S13-S17 in Supplementary Materials. **f-g**, Momentum-dependent superconducting gap as a function of the Fermi surface angle  $\varphi$  for the  $\alpha$ ,  $\beta_1$ ,  $\beta_2$  and  $\gamma$  Fermi surface in **f** and  $\theta$  for the  $\varepsilon$  Fermi surface in **g**. The Fermi surface angles  $\varphi$  and  $\theta$  are defined in **h**. The solid symbols are derived from the symmetrized EDCs shown in **a-e** while the open symbols are obtained by symmetrization taking into account the fourfold symmetry. The uncertainties are  $\pm 0.2$  meV as marked by error bars. The thick lines are the fitted curves of the measured gaps. **i**, Three-dimensional plot of the superconducting gaps in  $\text{KFe}_2\text{As}_2$ . The corresponding Fermi surface is shown at the bottom.

FIG. 5. **Gap symmetry of  $\text{KFe}_2\text{As}_2$ .** **a**, The superconducting gap as a function of the distance between the Fermi momentum and the  $\Gamma$  point on all the Fermi surface sheets. The uncertainties are  $\pm 0.2$  meV as marked by error bars. The expected gap size variation from the  $s_{\pm}$  symmetry with the gap form  $|\cos k_x + \cos k_y|$ [[68]] is plotted as a thick black line along  $\Gamma$ -M direction and as a thick red line along  $\Gamma$ -X direction. **b**, Gap nodes and local gap maxima on the Fermi surface of  $\text{KFe}_2\text{As}_2$ . The Fermi surface are plotted on top of a two-dimensional image showing the gap function  $\cos k_x + \cos k_y$  in the  $s_{\pm}$  pairing symmetry where the positive gap sign, negative gap sign and the gap nodes are represented by red, blue and white colors, respectively. The measured gap nodes are marked by white circles while the gap maxima are marked by black circles. Two interband scattering wave vectors ( $\mathbf{Q}_1$  and  $\mathbf{Q}_2$ ) are marked by double-arrow purple lines.  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  connect the local gap maxima between the  $\varepsilon$  band and  $(\alpha, \beta)$  bands.

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## Methods

High-quality single crystals of  $\text{KFe}_2\text{As}_2$  were grown by the KAs flux method[69]. The samples were characterized by magnetic susceptibility measurements (Fig. S1a in Supplementary Materials) and the measured  $T_C$  is 3.7 K with a narrow transition width of  $\sim 0.2$  K. The residual resistivity rate ( $R_{300K}/R_{0K}$ ) of near 1000 is obtained from the resistivity measurement (Fig. S1b in Supplementary Materials). Both measurements indicate that our  $\text{KFe}_2\text{As}_2$  single crystal samples have very high quality.

High-resolution angle-resolved photoemission measurements were carried out by using our laboratory-based ARPES system. It is equipped with a vacuum-ultraviolet (VUV) laser with a photon energy of  $h\nu = 6.994$  eV and a hemispherical electron energy analyzer R8000 (Scienta-Omicron). The light polarization can be varied to get linear polarization along different directions and circular polarization. This system uses the  $^3\text{He}$  pumping technology which can cool the sample down to the lowest temperature of 0.8 K. The energy resolution was set at 0.5 meV and the angular resolution was  $\sim 0.3^\circ$  corresponding to  $0.004 \text{ \AA}^{-1}$  momentum resolution at the photon energy of 6.994 eV. All the samples were cleaved *in situ* at low temperature and measured in ultrahigh vacuum with a base pressure better than  $1 \times 10^{-10}$  mbar. The Fermi level is carefully referenced by measuring polycrystalline gold which is well connected with the sample.

The photon-energy-dependent ARPES measurements were performed at the Bloch beamline of MAX IV, equipped with a hemispherical electron energy analyzer DA30-L (Scienta-Omicron). The angular resolution was  $\leq 0.2^\circ$  and the overall energy resolution was  $\leq 10$  meV. The single crystals were cleaved *in situ* at low temperature and measured in ultrahigh vacuum with a base pressure better than  $1 \times 10^{-10}$  mbar.

## Data availability

All raw data generated during the study are available from the corresponding author upon request.

## Code availability

The codes used for the fitting and simulation process in this study are available from the corresponding authors upon request.

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