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Enhancement of the Superconducting Gap by Nesting in CaKFe$_4$As$_4$: A New High Temperature Superconductor

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We use high resolution angle resolved photoemission spectroscopy and density functional theory with measured crystal structure parameters to study the electronic properties of CaKFe$_4$As$_4$. In contrast to the related CaFe$_2$As$_2$ compounds, CaKFe$_4$As$_4$ has a high $T_c$ of 35 K at stochiometric composition. This presents a unique opportunity to study the properties of high temperature superconductivity in the iron arsenides in the absence of doping or substitution. The Fermi surface consists of several hole and electron pockets that have a range of diameters. We find that the values of the superconducting gap are nearly isotropic (within the explored portions of the Brillouin zone), but are significantly different for each of the Fermi surface (FS) sheets. Most importantly, we find that the momentum dependence of the gap magnitude plotted across the entire Brillouin zone displays a strong deviation from the simple $\cos(k_x)\cos(k_y)$ functional form of the gap function, proposed by the scenario of Cooper pairing driven by a short range antiferromagnetic interaction exchange. Instead, the maximum value of the gap is observed on FS sheets that are closest to the ideal nesting condition, in contrast to previous observations in other ferropnictides. These results provide strong support for the multiband character of superconductivity in CaKFe$_4$As$_4$, in which Cooper pairing forms on the electron and the hole bands interacting via a dominant interband repulsive interaction, enhanced by band nesting.

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The superconducting mechanism in iron-based, high temperature superconductors is an important topic in condensed matter physics. One key question is whether the system should be described within a weak coupling BCS-type approach with a key role played by the interband repulsion between electron and hole bands, separated by the large momentum transfer or by a strong coupling approach with dominant short-range antiferromagnetic (AF) fluctuations, described by the local exchange interaction [1–7]. The former scenario seemed consistent with experimental results from a number of iron pnictide superconductors [8], but was later challenged by the discovery of iron chalcogenide-based superconductors [9,10]. Theoretical progress in this field was inspired by the discovery of new materials in this family [11], from iron chalcogenide [12] to single-layer FeSe films [13]. Materials with different crystal or electronic structures are extremely useful and provide new insights for constructing global models of high temperature superconductivity in iron-based materials.

Recently, a new iron-based class of superconductors, $AeA$Fe$_4$As$_4$ ($Ae = Ca$, Sr, Eu and $A = K$, Rb, Cs), were reported (generically referred to as $Ae$A1144) [14,15]. Although the chemical composition of $Ae$A1144 is the same as the intensively studied (Ba,K)Fe$_2$As$_2$ system, it has a different crystal structure type with $Ae$ and $A$ layers alternatively stacked between Fe$_2$As$_2$ layers. The crystallographically inequivalent position of the $Ae$ and $A$ atoms changes the space group from $I4/mmm$ to $P4/mmm$. A high transition temperature ($T_c = 31–36$ K) and stochiometric composition makes the $Ae$A1144 family an ideal new platform to test existing theories and inspire new ones. Measurements of its electronic structure and the momentum dependence of its superconducting gap are of critical importance.

In this Letter, we investigate the band structure and momentum dependence of the superconducting gap of CaKFe$_4$As$_4$, with high resolution Angle Resolved Photoemission Spectroscopy (ARPES) and Density Functional Theory (DFT) using experimentally obtained crystal structure parameters. Unlike most other iron-based superconductors, CaKFe$_4$As$_4$ has a high $T_c$ of 35 K at stoichiometric composition, which allows for the study of iron-based high temperature superconductivity in the absence of disorder caused by substitution. The Fermi surface consists of three hole pockets at the $\Gamma$ point and two electron pockets at the $M$ point. The hole pockets have significantly different diameters, which allow us to measure the superconducting gap for different values of the total momentum $k$. We find that the superconducting gap is...
nearly isotropic on each Fermi surface (FS) sheet, but has a significantly different value for each of the FS sheets. Indeed, the largest superconducting gap is found for pairs of hole and electron pockets that have a similar diameter, while other pockets have a smaller value of the superconducting gap. This observation is in stark contrast to the situation in some other ferropnictides. For example, in LiFeAs, which is another stoichiometric pnictide superconductor with multiple FS sheets, the largest superconducting gap is found on the smallest hole pocket, located near the zone center [16,17]. Therefore, our results on CaKFe$_4$As$_4$ provide a new important ingredient that must be included in the superconducting mechanism of iron-based superconductors.

CaKFe$_4$As$_4$ single crystals were grown using the flux method and extensively characterized by thermodynamic and transport measurements [15,18]. The experimental structure parameters were obtained from single-crystal x-ray diffraction. Technical details are provided in the Supplemental Material [19]. Single-phase samples were cleaved in situ at a base pressure of lower than $8 \times 10^{-11}$ Torr. ARPES measurements were performed using a tunable vacuum ultraviolet laser spectrometer [27] ($h\nu = 6.7$ eV, $\Delta E = 4$ meV) and helium microwave plasma spectrometer ($h\nu = 21.2$ eV, $\Delta E = 8$ meV). Assuming that CaKFe$_4$As$_4$ has the same inner potential ($\sim 12$ eV) as (Ba,K)Fe$_2$As$_2$, then 6.7 eV and 21.2 eV light sources measure the electronic structure around $k_z = \pi$ and $k_z = 0$, respectively [28,29]. The energy corresponding to the chemical potential was determined from the Fermi edge of a polycrystalline Au reference in electrical contact with the sample. The consistency of the data was confirmed by measuring six different samples.

The measured FS and band dispersion are shown in Fig. 1. In data collected using a photon energy of 6.7 eV, three hole pockets are observed at the center of the zone [$\alpha, \beta$, and $\gamma$, shown in panels (a)–(d)]. Whereas the $\alpha$ pocket is fairly round, the shapes of the $\beta$ and $\gamma$ pockets are more squarish. The approximate diameters of these three pockets are $\sim 0.2\pi/a$, $\sim 0.4\pi/a$, and $\sim 0.8\pi/a$, respectively. The FS and band structure data, measured using a photon energy of 21.2 eV, are shown in Fig. 1(e)–1(h). At this photon energy, only the largest hole pocket around $\Gamma$ is observed, and its diameter is $\sim 0.45\pi/a$. The $\beta$ pockets are not visible, most likely due to unfavorable matrix elements. The band responsible for $\alpha$ hole pocket is located 30 meV below $E_F$ for this value of $k_z$ [Fig. 1(g)]. This is consistent with the 3D character of most inner hole pockets in 122 systems [28,30]. We note that this change is not due to an increased sensitivity at low photon energies—in such a situation, two copies of the $\alpha$ band would be observed, one below and one above $E_F$, which is not the case. At the zone corner, a shallow electron FS pocket ($\delta$) is observed [Fig. 2(g)–2(h)].

The Fermi surface and orbitally resolved band dispersion were calculated using DFT and a Local Density Approximation (LDA) combination with experimental lattice constants and atomic positions (obtained from single-crystal x-ray diffraction measurements and shown in Table II of the Supplemental Material [19]). The resulting FS and band structure are shown in Fig. 2. The

![Image](315x118 to 432x221)

![Image](456x660 to 532x736)

**FIG. 1.** Measured electronic structure of CaKFe$_4$As$_4$. (a) Fermi surface (FS) intensity acquired using photon energy of 6.7 eV at $T = 40$ K. (b) Sketch of the FS based on data in (a). (c) Measured ARPES intensity along a cut through the $\Gamma$ point. Cut position is indicated in panel (b). (d) Sketch of the band structure based on data in (c). (e)–(h) Same as (a)–(d), but measured using a photon energy of 21.2 eV. Dashed lines in (b) and (d) mark the expected parts of the bands and FS that are not observed due to matrix elements and limited access to Brillouin zone at low photon energy.

![Image](367x459 to 393x508)

**FIG. 2.** (a) Calculated 3D Fermi surface of CaKFe$_4$As$_4$. (b) Band dispersion along the key symmetry directions with respective orbital contributions marked by color-coded outlines.
calculation predicts six hole pockets—slightly deformed, quasi 2D cylinders centered at the Γ point of the Brillouin zone, yet the ARPES data in Fig. 1 show only three Fermi sheets around Γ. Most likely, the intensity of the β sheet is due to three closely located bands that cannot be resolved experimentally. As in many other iron based superconductors, there are several 3d orbitals contributing to the states near the Fermi level. Figure 2(b) shows the band dispersion at the FS pockets, there is also a strong admixture of contributions to the FS pockets, respectively. kF positions are marked in (l). (j) Symmetrized EDCs at kF for lowest measured temperature from (f)–(i). Red, vertical dashed lines mark the energy of largest gap (β FS sheet). The black lines are the fits using phenomenological model [33]. (k) Temperature dependence of the superconducting gaps on the FS sheets, as summarized in Fig. 4, to elucidate the symmetry of the order parameter in CaFeKAs4. For qualitative analysis, we extract the EDCs at different kF on the β and δ FS sheets and symmetrize them in Figs. 4(a) and 4(b). All symmetrized EDCs show a clear dip structure at EF, and the energy positions of the quasiparticle peaks do not show much variation with the FS angle. In Fig. 4(c), we plot the extracted values of the superconducting gap as a function of FS angle. The gap sizes on these two FS pockets have no clear nodes and are roughly isotropic, which directly excludes the possibility of d-wave paring symmetry in CaFeKAs4 superconductor. In order to check the kz dependence of the superconducting

FIG. 3. (a)–(d) Measured electronic structure at four selected temperatures along the cut indicated in (l). The data is divided by the Fermi-Dirac function. (e) Energy Distributed Curves (EDCs) at kF for β FS pocket. (f) The data from panel (e) after symmetrization. (g)–(i) Symmetrized EDCs at kF of α, γ, and δ FS pockets, respectively, kF positions are marked in (l). (j) Symmetrized EDCs at kF for lowest measured temperature from (f)–(i). Red, vertical dashed lines mark the energy of largest gap (β FS sheet). The black lines are the fits using phenomenological model [33]. (k) Temperature dependence of the superconducting gaps for all four pockets. Solid lines are BCS predictions for Δ0 of 10.5 meV, 13 meV, 8 meV, and 12 meV. (l) Sketch of the FS with indication of the cut position and kF positions.
In many of the other iron-based superconductors, $xz/yz$ orbitals contribute mostly to the hole and electron FSs near the $\Gamma$ and $M$ points, respectively. Within the $s^\pm$ scenario, the superconducting gaps appear to be maximal on these orbitals due to strong intraorbital (interband) nesting of the electron and hole pockets. There are, however, scenarios (including the $s^{++}$-wave mechanism [44]) in which nesting does not play an important role and which also predict the largest superconducting gaps to be on the $xz/yz$ orbitals. One of our most important findings is that in the 1144 compound, there are not just $xz/yz$ orbitals, but also $z^2$ and $x^2-y^2$, which participate in the intraorbital interband nesting. This provides strong support in favor of the $s^\pm$
scenario of superconductivity, driven by nesting of the electron and hole bands.

In conclusion, we measured the electronic structure and values of the superconducting gap of a new member of iron arsenic high temperature superconductor—CaKFe$_2$As$_4$. We find the superconducting gap is nearly isotropic within the explored region of the BZ. The largest gap is observed on the $\beta$ hole and $\delta$ electron sheets, which have very similar diameters, whereas the $\alpha$ and $\gamma$ sheets have smaller values of the superconducting gap, as they have no electron counterparts of similar diameter. This strongly supports the multiband character of the superconducting gap in which the Cooper pairing forms on the electron and hole bands with strong interband repulsive interaction, enhanced by the nesting of the electron and hole bands.

Raw data from this manuscript are available through the link in Ref. [45].

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