Heavily electron-doped electronic structure and isotropic superconducting gap in $A_x Fe_2 Se_2$ ($A$=K,Cs)

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The low energy band structure and Fermi surface of the newly discovered superconductor, $A_x Fe_2 Se_2$ ($A$=K,Cs), have been studied by angle-resolved photoemission spectroscopy. Compared with iron pnictide superconductors, $A_x Fe_2 Se_2$ ($A$=K,Cs) is the most heavily electron-doped with $T_c \sim 30$ K. Only electron pockets are observed with an almost isotropic superconducting gap of $\sim 10.3$ meV, while there is no hole Fermi surface near the zone center, which indicates the inter-pocket hopping or Fermi surface nesting is not a necessary ingredient for the unconventional superconductivity in iron-based superconductors. Thus, the sign changed s± pairing symmetry, a leading candidate proposed for iron-based superconductors, becomes conceptually irrelevant in describing the superconducting state here. A more conventional s-wave pairing is a better description.

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Since the discovery of iron-based high temperature superconductors in 2008 [1, 2], the pairing mechanism of this new class of materials has been extensively studied [3–9]. Among various aspects that were suggested to be essential for the unconventional superconductivity, the multi-band nature of the electronic structure arguably received the most attention [10]. In particular, in the weak coupling approach, it was suggested that the superconductivity could be boosted by the nesting or hopping between the electron-like Fermi surfaces near the zone corner and the hole-like Fermi surfaces near the zone center [8], and the sign change of the superconducting order parameters between the hole and electron Fermi surfaces stems from the inter-pocket scattering [8]. Moreover, the gap symmetry and gap anisotropy (e.g. the appearance of nodes) are sensitive to the presence of certain Fermi surface and the interactions among the bands [11]. However, in the strong coupling approach, the superconducting pairing is dominated by the intra-orbital pairing and relatively robust against the change of band structures [4]. The dominating pairing symmetry is an $A_{1g}$ s-wave. Experimentally, both nodal and nodeless gap distributions have been reported by different techniques in various systems. Although the scanning tunneling spectroscopy (STS) data on Fe(Se,Te) suggest a nodeless s±-wave gap [12], the STS data on FeSe indicate a nodal gap [13]. While photoemission data on Ba$_{1-x}$K$_x$Fe$_2$As$_2$ and BaFe$_2−x$Co$_x$As$_2$ fit the s±-wave gap function well [6, 9], the thermal conductivity measurements suggest gap in heavily hole doped KFe$_2$As$_2$ to be a nodal type [14]. A conclusion on the pairing behaviors seems to be still far-fetched for the iron based superconductors.

Recently, a new series of iron-based superconductors, $A_x Fe_2 Se_2$ ($A$=K, Cs), has been discovered with relatively high transition temperature of $\sim 30$ K [15–18]. Judging from their chemical formula, these compounds would be the most heavily electron-doped amongst the iron-based superconductors. More importantly, it provides an opportunity to examine the common aspects of the electronic structure and pairing mechanism in iron-based systems from the most electron-doped end. In this paper, we report the angle resolved photoemission spectroscopy (ARPES) study of $A_x Fe_2 Se_2$ ($A$=K, Cs). We found that they are indeed the most heavily electron-doped iron-based superconductor. Only electron pockets are observed, while there is no hole Fermi surface near the zone center. The superconducting gap around the electron pockets is isotropic, and about 10.3 meV, i.e. $\sim 4k_BT_c$, for both compounds. Our result indicates the inter-pocket hopping or Fermi surface nesting is not a necessary ingredient for the unconventional superconductivity in iron-based superconductors. Thus the sign change in s±-wave pairing, a promising pairing symmetry candidate suggested earlier, is not a fundamental property of iron-based superconductors. Rather, the isotropic gap structure is better to be considered as a more conventional s-wave.

$A_0,8 Fe_2 Se_2$ ($A$=K, Cs, nominal composition) single crystals were synthesized by self-flux method as described elsewhere in detail [18], which show flat shiny surfaces with dark black color. Fitting the X-ray diffraction data assuming the $I4/ mm$ symmetry gave $a = 3.8912$ Å, $c = 14.1390$ Å for K$_{0.8} Fe_2 Se_2$, and $a = 3.9618$ Å, $c = 15.285$ Å for Cs$_{0.8} Fe_2 Se_2$. K$_{0.8} Fe_2 Se_2$ shows the onset superconducting transition temperature ($T_c$) of 31.7 K, and it reaches zero resistivity at 31.2 K; while Cs$_{0.8} Fe_2 Se_2$ shows the onset $T_c$ of 30 K with a transition width of 2 K in the resistivity data. The magnetic sus-
ceptibility measurements showed almost 100% shielding fraction, indicating the bulk superconducting nature and good quality of the crystals. ARPES measurements were performed with SPECS UVLS discharge lamp (21.2 eV He-I α light) and a Scienta R4000 electron analyzer. The overall energy resolution was set to 9 or 12 meV, and the angular resolution was 0.3 °. The sample was cleaved in situ, and measured under ultra-high-vacuum of 5 × 10^{-11} torr. The actual chemical compositions were determined by energy dispersive X-ray (EDX) spectroscopy on the cleaved surface after the photoemission measurements, which gave K : Fe : Se = 0.94 : 1.98 : 2, and Cs : Fe : Se = 0.92 : 1.99 : 2. For simplicity, they are called by the nominal compositions throughout the text.

Figure 1 shows the photoemission intensity maps at the Fermi energy ($E_F$) for (a) Cs$_{0.8}$Fe$_2$Se$_2$, and (b) K$_{0.8}$Fe$_2$Se$_2$. The intensity was integrated over a window of [$E_F$-15 meV, $E_F$+15 meV].

The Fermi surface topology is similar in both systems. There is an electron-like Fermi pocket surrounding the zone corner, and some spectral weight is located at the zone center.

The sizes of the electron pocket are about the same for both K and Cs compounds, and their band structures are very much alike as well. Figure 2 further reveals the band structure of K$_{0.8}$Fe$_2$Se$_2$. Around the zone center [Figs. 2(a) and 2(b)], there is a small electron-like feature, the $\kappa$ band. The spectral weight of this band is rather weak, indicating that it might be a tail of certain band slightly above $E_F$. This is consistent with a recent band structure calculation that shows an electron pocket in the zone center for K$_{x}$Fe$_2$Se$_2$ at certain doping [19]. At -0.1 eV and below, the fast dispersive features are most likely the $\alpha$ and $\beta$ bands that form the hole-like pockets in Fe(Te,Se) [20] and iron pnictides. Moreover, the $\omega$ band is observed around 0.35 eV below $E_F$ near $\Gamma$, and similar band is observed in iron pnictides and known to be made of the $d_{z^2}$ orbitals [10]. Around the zone corner [Figs. 2(c) and 2(d)], an electron-like band, $\delta$, is observed together with the $\beta$ band from the zone center. Theoretically, two electron-like bands around M were predicted for K$_{0.8}$Fe$_2$Se$_2$ [19], just like for the iron pnictides. However, at certain experimental geometry, only one electron-like band is often observed due to the matrix element of the $3d$ orbitals [10]. It is also predicted that the Fermi crossings of these two bands are almost-degenerate, and the bottom of the other electron like band would coincide with the $\beta$ band at M [10, 19]. In general, the bands around $\Gamma$ resemble those observed in Fe(Te,Se) and iron pnictides, except that the chemical potential is shifted up via electron doping here. Take BaFe$_{1.85}$Co$_{0.15}$As$_2$ for a comparison, although it is op-
The superconducting gap, we find it to be of 4 meV at 10 K. Taking the coherent peak position as \[ \text{Fig. 3(b)}. \] One can clearly identify a leading edge gap. The coherent peak feature grows with decreasing temperature in this region is rather flat. The temperature dependence of the spectrum illustrates that the spectrum integrated over the momentum region indicated by the black bar in panel a, and (c) its stacked symmetrized version. Energy resolution was set to 9 meV in the gap measurements.

Eventually, electron-doped with a \( T_c \) of about 25 K, there are still several hole-like pockets around the zone center. Moreover, the binding energy of the \( \omega \) band near \( \Gamma \) is about 200 meV larger in \( \text{K}_{0.8}\text{Fe}_2\text{Se}_2 \) than that in \( \text{BaFe}_1.85\text{Co}_{0.15}\text{As}_2 \) due to the higher electron doping in the former compound. However, it is interesting to note that the \( \delta \) band around M is rather flat and shallow in \( \text{K}_{0.8}\text{Fe}_2\text{Se}_2 \), indicative of a non-rigid band behavior.

Assuming the electronic structure of \( \text{A}_{0.8}\text{Fe}_2\text{Se}_2 \) to be two dimensional and two almost-degenerate electron pockets around M, in the ionic picture, we could obtain \( 3d_\delta^{6.18} \) per Fe ion for \( \text{A}_{0.8}\text{Fe}_2\text{Se}_2 \) by the measured Fermi surface volume. This cannot account for all the electrons in the \( 3d_\delta^{45} \) configuration calculated directly from their actual chemical compositions. Therefore, the inconsistency may suggest possible \( k_z \) dispersions of the \( \delta \) and \( \kappa \) bands. To fully reveal the Fermi surface topology, more detailed \( k_z \) and polarization dependence studies are required with variable photon energies at a synchrotron facility. In any case, the measured electronic structure clearly shows that \( \text{A}_{0.8}\text{Fe}_2\text{Se}_2 \) is indeed the most heavily electron-doped iron-based superconductor by far.

To study the superconducting gap of \( \text{K}_{0.8}\text{Fe}_2\text{Se}_2 \), high resolution data taken above and below \( T_c \) along the cut #2 are compared in Fig. 3. As shown in Fig. 3(a), there is no gap at \( E_F \) near the Fermi momentum in the normal state, while a clear gap shows up at low temperatures in the superconducting state. The temperature dependence of the spectrum illustrates that the spectral weight near the Fermi energy is depleted and a coherent peak feature grows with decreasing temperature [Fig. 3(b)]. One can clearly identify a leading edge gap of 4 meV at 10 K. Taking the coherent peak position as the superconducting gap, we find it to be \( \sim 10.3 \) meV, i.e., \( \sim 4k_B T_c \). This ratio between gap and \( T_c \) falls into the same regime as other iron-based superconductors.\[ \text{Fig. 3(c)} \]

The symmetrized spectra in Fig. 3(c) further indicate that the gap disappears when the temperature is above \( T_c \). We note that the spectra are integrated over a small region around the normal state Fermi momentum to compensate the relatively weak signal, but the estimated gap amplitude would not be affected much, as the gap feature in this region is rather flat [Fig. 3(a)].

By examining the symmetrized EDC’s in the superconducting state at various Fermi crossings of both the \( \kappa \) and \( \delta \) bands, the momentum distribution of superconducting gap is deduced in Fig. 4. For both K and Cs compounds, the gap of the \( \delta \) band around the M point is of the isotropic \( s \)-wave type within the experimental uncertainty, which averagely is about 10.3 meV; while the gap is 4 meV for the \( \kappa \) band at \( \Gamma \). The smaller gap at \( \Gamma \) than those around M certainly violates the simple gap function of \( \cos k_x \cos k_y \) for \( s \)-wave order parameter, and indicates the gaps may be orbital dependent. More-
over, since the spectral weight near the zone center is minimal, its contribution to the superconductivity would be rather negligible with such a small gap. Therefore, the inter-pocket scattering previously suggested can not be the essential force driving the superconductivity.

In summary, our data show that the rather robust superconductivity in such a highly electron-doped iron-based superconductor could mainly rely on the electron Fermi surfaces near M. The rather unique electronic structure in $A_0.8\text{Fe}_2\text{Se}_2$ ($A = \text{K, Cs}$) further highlights the diversity of the iron-based superconductors, and suggests that the superconductivity is very robust against the change of Fermi surfaces. Our data also strongly suggest that the inter-band hopping might not be so substantial as previous data suggested. Thus, the promising candidate, the so called $s_\pm$ wave characterized by the sign change of the superconducting orders between electron and hole pockets, is not a proper description of the superconducting state in $A_0.8\text{Fe}_2\text{Se}_2$. Instead, the more conventional $s$-wave type is a more proper and general description for the iron-based superconductors. Our result can also be viewed as a support to the picture derived from the strong coupling approach suggesting the pairing being the intra-orbital pairing caused by local electron-electron correlation.

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