Strong nodeless pairing on separate electron Fermi surface sheets in (Tl, K)Fe$_{1.78}$Se$_2$ probed by ARPES

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Abstract – We performed a high-resolution angle-resolved photoemission spectroscopy study of the Tl$_{0.63}$K$_{0.37}$Fe$_{1.78}$Se$_2$ superconductor ($T_c = 29$ K). We show the existence of two electron-like bands at the $M(\pi,0)$-point which cross the Fermi level at similar Fermi wave vectors to form nearly circular electron-like Fermi surface pockets. We observe a nearly isotropic $\sim 8.5$ meV superconducting gap ($2\Delta /k_BT_c \sim 7$) on these Fermi surfaces. Our analysis of the band structure around the Brillouin zone centre reveals two additional electron-like Fermi surfaces: a very small one and a larger one with $k_F$ comparable to the Fermi surfaces at $M$. Interestingly, a superconducting gap with a magnitude of $\sim 8$ meV also develops along the latter Fermi surface. Our observations are consistent with the $s$-wave strong-coupling scenario.

The amplitude and symmetry of the superconducting (SC) gap of a material are determined by its band structure, its Fermi surface (FS) topology and the pairing mechanism itself. The experimental observation of enhanced gap amplitude on hole-like and electron-like FS pockets quasi-nested by the antiferromagnetic (AF) wave vector in iron-based superconductors [1–5] has been widely considered as suggestive of the importance of AF interband scattering in these materials. In particular, the quasi-nesting model is consistent with the strong suppression of superconductivity in heavily hole-doped [6] and heavily electron-doped [7] BaFe$_2$As$_2$ compounds, for which the FS quasi-nesting conditions vanish. Recently, this model faced a serious challenge with the discovery of superconductivity above 30 K in heavily electron-doped K$_{0.8}$Fe$_{2-x}$Se$_2$ and (Tl,K)Fe$_{2-x}$Se$_2$ [8,9]. Indeed, previous angle-resolved photoemission spectroscopy (ARPES) measurements revealed only electron-like FS pockets [10,11].

In this letter, we report high-energy resolution ARPES measurements on the Tl$_{0.63}$K$_{0.37}$Fe$_{1.78}$Se$_2$ superconductor ($T_c = 29$ K). We observed two electron-like $M(\pi,0)$-centred FS pockets that develop a nearly isotropic SC gap below $T_c$ with a magnitude of $\sim 8.5$ meV, leading to a $2\Delta /k_BT_c \sim 7$. In addition, a weak electron-like FS pocket with a similar size and a tiny electron-like pocket are also observed at the $\Gamma(0,0)$-point. The former one also exhibits a SC gap size of about 8 meV. In addition, a high-energy ($\sim 0.8$ eV) incoherent peak undergoes a significant energy shift of $\sim 100$ meV through the metal-nonmetal crossover around 70 K, while the low-energy valence band shows little change. We discuss the possible implications of the SC gap symmetry and the FS topology for the SC pairing mechanism in this unusual iron-based superconductor.

Single crystals of Tl$_{0.63}$K$_{0.37}$Fe$_{1.78}$Se$_2$ ($T_c^{\text{onset}} = 29.1$ K; $T_c^{\text{mid}} = 28.6$ K; $T_c^{\text{zero}} = 27.5$ K) were grown by the Bridgeman method [9]. The precise composition was determined using an energy dispersive X-ray spectrometer (EDXS). The lattice parameters $a = 3.85$ Å and $c = 14.05$ Å were obtained by fitting XRD data. We performed ARPES measurements at the Institute of Physics, Chinese Academy of Sciences, using the He I resonance line ($h\nu = 21.218$ eV). The angular resolution was set to 0.2° while the energy resolution was set to 4–7 meV for high-resolution measurements. Samples with

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a typical size of \( \sim 2 \times 2 \) mm\(^2\) were cleaved in situ and measured between 10 and 35 K in a working vacuum better than \(5 \times 10^{-11}\) torr. The Fermi energy \(E_F\) of the samples was referenced to that of a gold film evaporated onto the sample holder. For convenience, we describe all the results using the 1 Fe site/unit (or unfolded) cell notation.

In fig. 1(a), we show a momentum-resolved photoemission intensity mapping of Tl\(_{0.63}\)K\(_{0.37}\)Fe\(_{1.78}\)Se\(_2\) recorded in the normal state (35 K) and integrated over a 10 meV window centred at \(E_F\). The small red circles indicate the FS obtained from the momentum distribution curve (MDC) peak position at \(E_F\). (b) EDCs recorded along cut1 from panel (a). The blue line indicates the spectrum at the \(M\)-point. (c) EDC at \(k_F\) recorded at 10 and 35 K. (d) Same as (c) but after division by the Fermi-Dirac function.

The raw EDCs at \(k_F\) are given in fig. 2(a). Following a standard procedure in ARPES, we take advantage of the particle-hole symmetry at \(k_F\) by symmetrizing the spectra to approximately remove the Fermi-Dirac function. The symmetrized EDCs are shown in fig. 2(b). The SC gap size \(\Delta\) is approximated by half the distance between the two peaks. Obviously, the peak position does not vary much with momentum within experimental uncertainties. We report the gap size extracted for different samples mounted with different orientations in fig. 2(c). We find that the SC gap size averages at \(\sim 8.5\) meV with little room for anisotropy and even less for nodes, at least at the particular \(k_z\) value corresponding to \(\hbar \nu = 21.218\) eV, in agreement with a previous report on A\(_x\)Fe\(_2\)Se\(_2\) (A = K, Cs) [10]. This gap size leads to a \(2\Delta/k_B T_c\) ratio of \(\sim 7\), indicating that the SC pairing in this material is in the strong-coupling regime. The temperature dependence of spectra recorded at a single \(k_F\)-point are displayed in fig. 2(d), and the corresponding symmetrized EDCs are shown in fig. 2(e). Interestingly, the temperature dependence of the SC gap given in fig. 2(d) indicates that the SC gap size may not close with increasing temperature, but rather fill up gradually to \(T_c\). Such behaviour has been reported recently in NaFe\(_{0.95}\)Co\(_{0.05}\)As [12].

The FS pockets at the \(M\)-point encloses an area corresponding to 4.5% of the Brillouin zone (1 Fe site/unit cell description). This leads to a much smaller electron doping than expected from the Luttinger theorem. However, previous ARPES measurements on other iron-based superconductors indicate that there are two electron-like FS pockets with similar \(k_F\)'s centred at the
Fig. 2: (Colour on-line) (a) Spectra recorded in the SC state (10 K) at different locations around the M-centred electron-like FS. The colours of the spectra correspond to their angular locations, which are given in panel (c) (sample 1; filled symbols). (b) Corresponding symmetrized EDCs. (c) Polar distribution of the SC gap size along the FS for 3 different samples. (d) Temperature dependence of EDC spectra at $k_F$. The inset show the momentum location of the EDC. (e) Corresponding symmetrized EDCs.

M-point [1,3,4,7,12]. To confirm that this observation holds for Tl$_{0.63}$K$_{0.37}$Fe$_{1.78}$Se$_2$, we display the ARPES intensity plot along cut 2 (see fig. 1(a)) in fig. 3(a), and the corresponding intensity plot of second derivative along energy in fig. 3(b). The data suggest that there are two distinct bands with bottoms around 40 and 60 meV, respectively. Within our experimental resolution, the bands have approximately the same $k_F$ values.

The approximate 2-fold degeneracy of the electron-like FS pocket at M leads to an electron counting of 18% electron per Fe, which is still not enough to afford for the 31.5% electron per Fe expected from the Luttinger theorem for this material. We now turn our attention to the band structure around the Γ-point. Figure 3(c) and fig. 3(d) give the ARPES intensity plot along cut 3 (see fig. 1(a)) and the corresponding second-derivative intensity plot along energy, respectively. The strongest feature is a hole-like band topping around 50 meV below $E_F$. We also found a tiny electron-like feature with a bottom around 10 meV below $E_F$ that has been reported previously in A$_x$Fe$_2$Se$_2$ (A = K, Cs) [10]. In addition, we observe a large electron-like band at Γ with a $k_F$ similar to that of the electron bands at M. The inclusion of the electron-like bands at the Γ-point raises the electron counting to 32% electrons per Fe, which is in good agreement with the nominal composition. As shown in fig. 3(e), where the EDC spectra at $k_F$ are presented as a function of temperature, a SC gap that vanishes above $T_c$ is found along the large electron-like FS at Γ. Interestingly, the SC gap size (∼8 meV) is almost the same as the one we find at the M-point.

Another major difference we observed in K$_{0.8}$Fe$_{1.7}$Se$_2$ compared to other iron-based superconductors is the presence of a large incoherent peak at ∼0.8 eV [11]. This incoherent peak is also observed in Tl$_{0.63}$K$_{0.37}$Fe$_{1.78}$Se$_2$, as shown in fig. 4(a). While this broad peak is found to be dispersionless in $k$-space, it has a drastic temperature dependence between 35 K and 150 K, as shown in fig. 4(a). The energy shift at this temperature range is about 100 meV, with much of the shift occurring below 100 K, as indicated in fig. 4(b). This shift corresponds well to the metal-nonmetal crossover around 74 K observed in the resistivity of this material, which is shown in fig. 4(c). Optical data show broad spectral features between 0.56 to 0.74 eV with similar energy shift when going through this metal-nonmetal crossover [13]. This can be interpreted in terms of incoherent features within the Mott picture [14]. Interestingly, the low-energy valence band below 0.3 eV is relatively insensitive to this crossover. This unusual dichotomy of energy shift between the high-energy incoherent feature and the low-energy dispersion could be consistent with the doped Mott insulator picture where quasiparticles emerge from a gapped incoherent background. These quasiparticles with renormalized effective mass and coherence residue form the observed low-energy...
In conclusion, our high-resolution ARPES measurements of the highly electron-doped Tl$_{0.63}$K$_{0.37}$Fe$_{1.78}$Se$_2$ superconductor reveal nearly isotropic superconducting gaps on the two nearly degenerated electron FS sheets at the $M$-point. The SC gap with an amplitude of $\sim$8.5 meV closes above $T_c$, resulting in a pairing strength $(2\Delta/k_BT_c$ of $\sim$7) twice stronger than the weak-coupling BCS value. In addition, an unexpected electron Fermi surface with similar $k_F$ and $\Delta$ is observed around the zone centre, along with a hole-like band sinking $\sim$50 meV below the Fermi energy. On the larger energy scale, the dispersionless incoherent peak at 0.7–0.8 eV shows a significant energy shift of $\sim$100 meV when going through the metal-nonmetal crossover around 74 K, while the low-energy band structure is relatively unaffected.

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