(π, π)-electronic order in iron arsenide superconductors.

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The distribution of valence electrons in metals usually follows the symmetry of an ionic lattice. Modulations of this distribution often occur when those electrons are not stable with respect to a new electronic order, such as spin or charge density waves. Electron density waves have been observed in many families of superconductors[1, 2, 3], and are often considered to be essential for superconductivity to exist[4]. Recent measurements[5, 6, 7, 8, 9] seem to show that the properties of the iron pnictides[10, 11] are in good agreement with band structure calculations that do not include additional ordering, implying no relation between density waves and superconductivity in those materials[12, 13, 14, 15]. Here we report that the electronic structure of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ is in sharp disagreement with those band structure calculations[12, 13, 14, 15], instead revealing a reconstruction characterized by a (π, π) wave vector. This electronic order coexists with superconductivity and persists up to room temperature.

Calculations of the electronic structure of the new pnictide superconductors unanimously predict a Fermi surface (FS) consisting of hole-like pocket in the centre (Γ point) of the Brillouin zone (BZ) and electron-like ones at the corners (X point) of the BZ. A shift by the (π, π) vector would result in a significant overlap of these FSs. Such an electronic structure is highly unstable since any interaction allowing an electron to gain a (π, π) momentum would favour a density wave order, which then results in a concomitant opening of the gaps, thus strongly reducing the electronic kinetic energy. It is surprising that ARPES data are reported to be in general, and sometimes in very detailed[9], agreement with the calculations giving a potentially unstable solution[5, 6, 7]. Even in the parent compound, where the spin density wave transition is clearly seen by other techniques[16, 17], no evidence for the expected energy gap has been detected by photoemission experiments[7, 8]. In fact, no consensus exists regarding the overall FS topology. According to Refs. 6 and 5, there is a single electron-like FS pocket around the X point, while Ref. 18 reports two intensity spots without any discernible signature for the electron pocket in the normal state. Intensity spots near the X point can also be found in Refs. 6, 7, and 9, but those are interpreted as parts of electron-like pockets. Obviously, such substantial variations in the photoemission signal preclude unambiguous assignment of the observed features to the calculated FS, leaving the electronic structure of the arsenides unclear.

In Fig.1 we show experimental FS map of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ (BKFA) measured in superconducting state. To eliminate possible effects of photoemission matrix elements, as well as to cut the electronic structure at different $k_z$ values, we have done measurements at several excitation energies (Fig. 1a–b) and polarizations (Fig. 1c–d). Although there are obvious changes in the intensities of the features, no signatures indicating $k_z$ dispersion can be concluded. With this in mind, the apparently different intensity distributions at neighboring Γ points appear unusual. While in the first BZ the two concentric contours are broadly consistent with band structure calculations, the “design wheels” in the second BZ are at variance with predicted hole-like circles. The major discrepancy with theoretical calculations and ARPES data[5, 6, 7, 8, 9, 18] is observed near the X point, where, according to the calculations, one expects a sizeable double-walled electron

Fig. 1. Fermi surface topology of Ba$_{1-x}$K$_x$Fe$_2$As$_2$. The colour plots display the photoelectron intensity distribution as a function of quasimomentum $k_z$ integrated in a small energy window of 15 meV around the Fermi level. a–b, Fermi Surface maps of Ba$_{1-x}$K$_x$Fe$_2$As$_2$, measured using excitation energy $h\nu = 80$ and 50 eV respectively at $T = 14$ K. Images c and d, measured with $h\nu = 80$ eV, demonstrate strong effect of light polarization on the photoemission from the four small FS’s surrounding the X point. The component of electric vector parallel to the sample surface is shown by a double-headed arrow. Experimental details concerning the sample preparation can be found in Supplementary Information.
To examine the topology of the five aforementioned pockets, we look at momentum distribution of intensity below and above the Fermi level (FL). As can be seen in Fig. 2a–c, the size of the X-centered pocket clearly increases when cutting the electronic structure above the FL, and decreases for the cut below the FL, which proves its electron-like topology. On the other hand, the behaviour of the blades surrounding the X point is opposite, which shows that they are hole-like. In Fig. 2e–i we analyze the band dispersions along the cuts given in Fig. 2d, once again supporting these conclusions (see caption and Supplementary Information). Therefore, the observed topology of the Fermi surface is different from that predicted by band structure calculations. However, in the following we will argue that the results of the calculations can be reconciled with the experimental data, provided the system reacts to the predicted nesting instability and an ordered state develops.

In Fig. 3 we collate the ARPES data taken at low temperatures for the parent compound with the spectra of the doped superconductor, measured using the same light polarization as in Fig. 1d. The photoemission intensity distributions shown in panels (a) and (b) are comparable, when taking into account the difference in the charge carrier concentrations. Moreover, the momentum-energy cuts (c)–(e) and (f)–(h) show that there is one-to-one correspondence between the underlying band dispersions in the parent and superconducting samples. Similar locations in the momentum-energy space, as well as the characteristic, polarization induced intensity variations clearly suggest that the blades are also present in BaFe$_2$As$_2$ (BFA). Remarkably, the distance between the centers of opposite blades tracks the size of the $\Gamma$-FS. Closer inspection of all existing ARPES data on arsenides confirms an intriguing universality of this observation[6, 7, 8, 9, 18].

The origin of the unexpected FS topology near the X point is elucidated by panels (i) and (j) where momentum-energy cuts separated by the ($\pi$, $\pi$) vector are shown: the blades in the parent compound are created by the interaction of the ($\pi$, $\pi$)-replica of the $\Gamma$-centered FS with the X-centered electron-like FS predicted in the calculations. Such a folding implies the presence of additional ordering that sets in to relax the nesting instability. The formation of the blades, within this scenario, is illustrated by simple sketch in Fig. 3k. The intensity of the $\Gamma$-derived band in panel (j) is lower than that shown in panel (i) due to the weakness of the scattering potential introduced by the new order as compared to the original crystalline potential[19, 20]. This is also the case for the FS map in panel (a), where the ARPES intensities near $\Gamma$ and X points still differ. Similarly, in the superconducting BKFA this effect prevents immediate detection of the ordered state alluded by the blades. Though, recovering the band dispersion in the vicinity of the Fermi level as shown in Fig. 3l–m, the presence of new BZ boundaries (white lines) can be identified.

An immediate interpretation of the observed similarity in the electronic structures of $x$=0.0 and $x$=0.3 compounds as shown in Fig. 3 would be the persistence of magnetic order in the superconducting case, which does not contradict the phase diagram suggested in the Ref. 21. However, the temperature evolution of the photoemission intensity of the blades presented in Fig. 4b rules out any direct connection between the observed FS topology and the static magnetic order. Despite a noticeably lower spectral weight of the blades as compared to the $\Gamma$-FS sheets, these hole-like structures clearly persist to the room temperature, which is above the structural, magnetic and superconducting transitions in arsenides. Such temperature behaviour also rules out the possibility that the observed FS topology near the X points is a consequence of a pronounced deformation of the calculated band structure, which would destroy the FS nesting. In order to account for the observed hole-like structures, one would have to shift some of bands at the X point.
by an unlikely 250 meV[5, 13, 14, 15]. In addition, the potential candidates to form the hole and electron pockets at the X point do not interact due to symmetry reasons and thus would not be able to account for the picture shown in Fig. 3k (see Supplementary Information).

Our ARPES data manifest the presence of electronic order of a special kind. This order sets in already at high temperatures and is obviously dictated by the nesting instability predicted in the calculations. Most likely it is this electronic order which results in the structural transition at lower temperatures in the parent compound, since in the case of LaOFeAs the static magnetic order develops only after the structural transition and magnetic moments are much smaller than theoretically expected[16, 17]. On the other hand, presence of the $(\pi, \pi)$ excitations ($q=1.15$ Å$^{-1}$) above the superconducting transition is indeed implied by the inelastic neutron scattering data [22] and their transformation into a resonance below $T_c$ could indicate their crucial role in the mechanism of superconductivity. If these excitations correspond to a fluctuating stripe-like order, the propeller structure can result from a superposition

![Fig. 3](image_url)

**Fig. 3.** $(\pi, \pi)$-reconstruction of the electronic structure. **a-b**, FS maps for the BFA and BKFA respectively, $hv = 80 \text{ eV}$. **c-e, f-h**, Several typical energy-momentum cuts, normalized to integrated intensity, showing similar band dispersion for BFA and BKFA. The cuts positions in momentum space are indicated by the arrows in panels a and b. **i-j**, Parallel cuts through the electronic structure of BFA set apart by the $(\pi, \pi)$ vector. **k**, Simplest model showing the result of folding of single hole- and electron-like bands. **l-m**, Parallel cuts through the $\Gamma$ and X points in electronic structure of BKFA set apart by the $(\pi, \pi)$ vector. Red curves are the MDCs integrated within the 8 meV showing the symmetric behaviour about the new BZ boundaries (white lines) due to $(\pi, \pi)$-folding of the original structure.

![Fig. 4](image_url)

**Fig. 4.** Blade structure at the X point. **a**, FS map measured at $T=300 \text{ K}$ ($hv = 50 \text{ eV}$) demonstrating persistence of the blades related intensity to high temperatures. **b**, Temperature dependence of the blades intensity as compared to the $\Gamma$ barrel. The plot represents the intensity along the cut schematically shown in panel a and was integrated in the 100 meV window centered at the FL.
of two pairs of blades originating from folding associated with \((\pi, \pi)\) and \((\pi, -\pi)\) wavevectors.

The observed order is not a conventional charge-density-wave, since the atoms respond only at considerably lower temperatures\([16, 17]\), but since the low-lying electronic structure is formed exclusively by the d-electrons, it may well be related to a more complex order parameter as in the case of the hidden order scenario suggested for the cuprates \([4]\). Observation of the reconstruction below \(T_c\) implies the coexistence of the \((\pi, \pi)\) order and the superconductivity, which is confirmed by opening of the superconducting gaps on the “propeller” FS\([23]\).

The order weakens with doping, which probably explains that it is no longer strong enough to cause the structural transition and/or static magnetism, but is sufficient to open smaller \((\pi, \pi)\)-gaps near the X point, thus providing a high density of states at the Fermi level that might be necessary for superconductivity \([24]\).

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