Unusual Fermi surface nesting in parent compounds of iron arsenic high temperature superconductors revealed by Angle Resolved Photoemission Spectroscopy

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We use angle resolved photoemission spectroscopy (ARPES) to study the band structure of BaFe$_2$As$_2$ and CaFe$_2$As$_2$, two of the parent compounds of the iron arsenic high temperature superconductors. We find clear evidence for band back folding and hybridization demonstrating that conduction electrons are strongly affected by the emergence of magnetic order. Our high quality data revealed that although the Fermi surface is strongly three-dimensional, it does indeed have long parallel segments along the $k_z$ direction that can lead to the emergence of magnetic order. More interestingly, we find very unusual incommensurate nesting of the Fermi surface in the $a-b$ plane that is present only at low temperatures. We speculate that this is a signature of a failed Charge Density Wave (CDW) state that was predicted by renormalization group studies.

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Iron arsenic superconductors display a fascinating interplay between magnetism and superconductivity. The influence of the magnetism on the electronic properties and the role it plays in high temperature superconductivity of these materials is a subject of a lively debate within the condensed matter physics community. Neutron scattering experiments reported antiferromagnetic (AF) ordering and 3D character of magnetic interactions in non-doped parent compounds of iron arsenides. A number of previous studies attributed the magnetic ordering to itinerant Spin Density Wave (SDW) or $q_z$ nesting, while others favored magnetic order due to localized moments. The AF ordering is reminiscent of that of the cuprate high temperature superconductors, which occurs in the un-doped materials. The AF ordering introduces a new zone boundary (antiferromagnetic zone boundary - AFZB) leading to band back folding effects. The main difference between the iron arsenides and cuprates is that the parent (undoped) phase in the former is not insulating but metallic. The $k_z$ direction spanning nearly half of the Brillouin zone that may play an essential role in the emergence of magnetic order in these materials. More interestingly, we discovered long parallel segments of the Fermi surface within the $a-b$ plane. This new nesting vector is significantly shorter than the $(\pi/a, \pi/a)$ vector related to the magnetic order. We speculate that these nested parts of the Fermi surface may be a precursor of a failed CDW order predicted by renormalization group studies.

Single crystals of BaFe$_2$As$_2$ and CaFe$_2$As$_2$ were grown out of a FeAs flux as well as Sn flux using conventional high-temperature solution growth techniques. BaFe$_2$As$_2$ and CaFe$_2$As$_2$ undergo a tetragonal to orthorhombic structural transition simultaneously with a paramagnetic to antiferromagnetic transition below $T_S = T_N \approx 130$K and 170K, respectively. ARPES data were measured at the SIS beamline of Swiss Light Source, Switzerland, the beamlines 7.0.1 of the Advanced Light Source (ALS), USA, using a Scienta R4000 analyzer and Ames Laboratory using a Scienta SES2002
with a Gammadata VUV5010 photon source. Energy and angular resolutions were 10 – 30meV and ~0.1°, respectively. The high symmetry points X and Z for both two phases are defined to be \((\pi/a, \pi/a(b), 0)\) and \((0, 0, 2\pi/c)\), respectively, with \(k_x (k_{100})\) and \(k_y (k_{010})\) axes along the Fe-As bonds.

Band calculations predict two types of nearly circular FSs in the paramagnetic state: hole-like FSs centered at \(\Gamma (Z)\) and electron-like FSs centered at \(X\)\textsuperscript{13} \textsuperscript{19}. Below the magnetic ordering temperature one would expect back folding and hybridization of these bands leading to a reconstruction of the FS\textsuperscript{19}. Figure 1 (a) shows a typical FS map from BaFe\textsubscript{2}As\textsubscript{2} sample measured with 105 eV photons deep in the magnetically ordered state. Dark areas indicate the locations of the Fermi surfaces (FSs), which indeed display a more complicated structure than the one predicted in the paramagnetic state. To examine the character of the FSs, we show in Fig.1 (c2) an ARPES intensity along a diagonal direction (blue arrow in Fig.1 (a)) measured with 35eV photons. In addition to hole-like bands that are characteristic of the paramagnetic state, we observe electron-like bands that

![Image 93x633 to 187x727](image1.png)

![Image 93x472 to 194x581](image2.png)

**FIG. 1:** (Color online) (a-c) BaFe\textsubscript{2}As\textsubscript{2} and (d) CaFe\textsubscript{2}As\textsubscript{2} data. FS map (a) along \(a-b\) and (b) along \(k_{100}\) and \(k_{[100]}\) (pink arrow in (a)). (Inner potential: \(V_0 \equiv 1.3\text{eV}\)) Blue arrow indicates the parallel segment. (c1) MDC of (c2) at Fermi energy (black curve) and the fitting curve (yellow). (c2) Band dispersion map along a diagonal cut (blue arrow in (a)), (c3) EDCs of (c2, a white arrow range). Colored circles follow hole-bands (\(\alpha_1\) and \(\alpha_2\)) and electron-bands (\(\beta_1\) and \(\beta_2\)). (d1-d2) MDC at Fermi level of (a3-b3). Band dispersion map along a diagonal cut (d3) below and (d4) above \(T_N\).

**FIG. 2:** (Color online) (a) Model calculation, (b-d) BaFe\textsubscript{2}As\textsubscript{2}, and (e) CaFe\textsubscript{2}As\textsubscript{2} data. (a1) Model FSs in non-magnetic phase. Reconstructed FSs for \(\Delta_{AF}\) of (a2) 0.01 eV, (a3) 0.03 eV, and (a4) 0.05 eV. ARPES intensity about the Fermi energy and \(E = -20\text{meV}\) obtained at (b1-b2) \(h\nu = 49\text{eV}\) (near \(\Gamma\)) and (c1-c2) \(h\nu = 35\text{eV}\) (near \(Z\)). (d1) FS and (d2) energy contour plots at \(E = -20\text{meV}\) for \(h\nu = 49\text{eV}\) data. FS map close to the zone center along \(a-b\) plane (e1) below and (e2) above \(T_N\). Dimension arrows indicate nesting vector \(q(\alpha_1) = 0.08(\pi/a, \pi/a)\) and \(q(\alpha_2) = 0.32(\pi/a, \pi/a)\) for hole-band \(\alpha_1\) and \(\alpha_2\), respectively.
are back folded from X-points about the antiferromagnetic zone boundary (AFZB, marked as red dashed lines in Fig.1(a)). The presence of multiple Fermi crossings is further verified by fitting of the momentum distribution curve (MDC) at Fermi level acquired along the same direction as shown in Fig. 1 (c1). The two hole-like bands ($\alpha_1$ and $\alpha_2$) and two electron-like bands ($\beta_1$ and $\beta_2$) can be also identified from the peak positions of energy distribution curves (EDCs) as marked with color circles in Fig.1 (c3). As shown in Fig. 2 (c1-d1), these bands hybridize leading to energy gaps (disconnected parts in the Fermi surface) and reconstruction of the Fermi surface. In order to compare the band structure between the magnetic and non-magnetic phase, we measured the ARPES data for CaFe$_2$As$_2$ deep below $T_N$ ($T = 12$K) and slightly above $T_N$ ($T = 180$K). The Fermi surface map near the zone center and the band dispersion map measured along a diagonal cut (blue arrow in Fig.1 (a)) are plotted in Fig. 2(e1) and Fig. 1(d3) for below $T_N$ and Fig. 2(e2) and Fig. 1(d4) for above $T_N$. The data were measured with 21.2 eV photons. The electron band ($\beta$) is observed only below $T_N$ (Fig. 1 (d3)), and it is absent above $T_N$ (Fig. 1 (d2)). The associated “flower-like” shape of Fermi surface changes to nearly circular as expected from band calculations in non-magnetic phase.

We demonstrate that a simple model with the effect of band back folding about the AFZB reproduces many aspects of the data. Figure 2 (a1) shows a model of two hole- and electron-pockets surrounding the zone center and the corner, respectively, which are based on the band calculation for non-magnetic phase [20]. We assume that below $T_N$ the states around $\Gamma$ (or Z) are coupled to the states around X by an interband staggered mean field of magnitude $\Delta_{AF}$ via the mean field term in the Hamiltonian $\sum_{k,\sigma} \sigma \Delta_{AF} (d_{k,\sigma,\Gamma}^\dagger d_{k+Q,\sigma,X} + h.c.)$, where $\sigma$ refers to the spin and $Q$ is the magnetic ordering vector. Upon increase of $\Delta_{AF}$ from 10meV to 50meV, the conduction bands are increasingly folded back about the AFZB and the Fermi surfaces are reconstructed as shown in Fig.2(a2-a4) including twinning effects[21]. The degree of the reconstruction and hybridization gets stronger with increasing $\Delta_{AF}$. Eventually the FS shape becomes “flower-like”. We plot the enlarged ARPES FS map of BaFe$_2$As$_2$ close to $\Gamma$ and Z in Fig.2 (b1) and (c1), respectively, and the extracted Fermi crossing points for the latter in Fig.2 (d1). The experimentally observed “flower-like” FS is well reproduced by the simple model for $\Delta_{AF}$ of 50meV (Fig. 2 (a4)). The reconstruction does not occur for $k_s$ where the interband coupling via the momentum $Q$ is not efficient (Fig. 2(b1)). Figure 2 (b2) and (c2) shows the ARPES intensity close to $\Gamma$ and Z, respectively, slightly below the Fermi energy ($E = -20$meV). The band positions for the latter are plotted in Fig. 2 (d2). While the effect of band hybridization (“flower-like” shape) is clearly seen even in this energy range around Z (Fig. 2(c2)), it is absent around $\Gamma$ (Fig. 2(b2)) with a round shape of the energy contour. The coupling between charge carriers and magnetism is therefore strongest whenever efficient low energy scattering between the two sets of FS is kinematically possible, demonstrating the role of interband magnetic scattering in the iron pnictides. Previous ARPES studies [22, 23].

FIG. 3: (Color online) CaFe$_2$As$_2$ data at $T = 40$K. (a) Schematic FSs along $k_{(001)}$. (b) FS map along a-b plane. (c1)-(f1) MDC of (c2)-(f2) at the Fermi level. (c2)-(f2) Energy dispersion map measured along a red arrow in (c3)-(f3). ARPES intensity about (c3)-(f3) Fermi level, (c4)-(f4) -60meV, (c5)-(f5) -90meV, and (c6)-(f6) -120meV. (g1)-(g5) FS and energy contour plots extracted from (c-f). Only stronger peaks between $k_F(\alpha_1)$ and $k_F(\alpha_2)$ are plotted. Dimension arrows indicate a nesting vector $q(\alpha_1)=0.07(\pi/a, \pi/a)$ and $q(\alpha_2)=0.31(\pi/a, \pi/a)$ of hole-band $\alpha_1$ and $\alpha_2$, respectively.
demonstrated strong dispersion along the \( k_z \) direction. This raised a question about the origin of magnetic order and pairing, since the presence of strong \( k_z \) dispersion would significantly limit in-plane \((\pi/a, \pi/a)\) nesting. By acquiring high quality data (Fig. 1(b)), we could see that, despite strong 3D-like dispersion, the Fermi surface has a long (\( \sim 1.7\pi/c \)) parallel segments along the \( k_z \). The strong FS reconstruction discussed above is most prominent in this \( k_z \) range. It might be essential for nesting conditions leading to the emergence of the \((\pi/a, \pi/a)\) magnetic order and superconductivity.

Perhaps the most interesting experimental observation in our data is an incommensurate FS nesting (long parallel segments of FS in the \( a-b \) plane seen in Fig. 2 (d1) for BaFe\(_2\)As\(_2\) and (e1) for CaFe\(_2\)As\(_2\)), which doesn’t appear in the result of the model calculation (Fig. 2 (a4)). The nesting vector \( q(a_1) \approx 0.1(\pi/a, \pi/a) \) and \( q(a_2) \approx 0.3(\pi/a, \pi/a) \) for hole-band \( a_1 \) and \( a_2 \), respectively, does not correspond to any previously reported density wave states. Therefore, we speculate that the gapless, nested Fermi surface may be a precursor of a failed density wave order such as the CDW predicted by renormalization group studies \([16, 17]\). The nesting vector \( q(a_2) \approx 0.3(\pi/a, \pi/a) \) are clearly observed in Fig. 3 (c3)-(b3), we plot FS maps focused on the zone center \((\Gamma(Z))\) measured at various \( h\nu\). The \( k_z \)s corresponding to these photon energies are indicated in Fig. 3 (a) with the 1st Brillouin zone and schematic FSs of hole-bands \((a_1, a_2)\). We note that the intensity of several bands \((a_1, a_2, \text{and } \gamma)\) dramatically changes with photon energy, and the electron bands \((\beta_1 \text{ and } \beta_2)\) are absent due to matrix element effect. These are confirmed in the MDCs at the Fermi level shown in Fig. 3 (c1-f1) - the \( a_2 \) is most intense at 105 eV, while the \( a_1 \) is for 87eV, 81eV and 57eV photons. We also plot energy contour intensities in (c4)-(f4), (c5)-(f5), and (c6)-(f6) \((E=-30\text{meV}, -60\text{meV}, -90\text{meV} \text{ and } -120\text{meV}, \text{respectively})\), which help us to understand how these bands behave with the binding energy. We found that the signature of band reconstruction is absent at specific \( k_z \) where the band doesn’t cross \( E_F \) (Fig.3 (f1)-(f7)), and quickly evolved when the Fermi pockets starts appearing. This indicates that the band hybridization in the magnetic phase occurs only when the Fermi pockets overlap with folding-back bands and the gap opens with the consequent energy gain at the overlapped segments (seen as disconnected parts in Fig.1(c1) and (d1)). The parallel segments in the energy contours are significant up to \( E \approx 100\text{meV} \). This binding energy seems to agree with the energy bottom of electron-band back folded from the zone corner (see Fig. 1 (d3)). This indicates that the failed density wave order is tied to the existence of magnetic long range order, and suggest a strong coupling between the staggered magnetization and other density wave order parameters in the iron pnictides.

In conclusion, we find strong band back folding and hybridization effects that lead to a reconstruction of the Fermi surface (FS) below the magnetic ordering temperature \((T_N)\). The round hole-pockets observed above \( T_N \) evolve into a square shape surrounded by “flower petal” electron pockets below \( T_N \). The FS data can be reasonably understood within a simple model of magnetic ordering. More interestingly, we find long parallel segments of the Fermi surface in the \( a-b \) plane, which is not expected by the simple model. We speculate that these nested parts of the Fermi surface may be a precursor of a failed density wave order such as the CDW predicted by renormalization group studies \([16, 17]\). These findings demonstrate the complexity of these newly discovered materials and form the foundation of understanding the electronic properties that lead to high temperature superconductivity in the carrier-doped phase.

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