Complex Fermiology of $CaFe_2As_2$

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ABSTRACT

The investigation of Fe-based compounds reveals the coexistence of multiple phases and complex electronic properties. Through a detailed study of the Fermi surfaces using ARPES (Angle-Resolved Photoemission Spectroscopy) and high photon energies, the presence of a hidden cT (collapsed tetragonal) phase within the normal structural phase is discovered. The sample quality significantly influences the emergence of the kz-dependence of the Fermi surface, with better quality samples exhibiting more distinct δ and κ bands. Comparison of experimental results and calculated energy bands suggests that the δ and κ bands correspond to the cT phase. The crystal structure is found to be sensitive to small perturbations such as pressure and preparation conditions.

Keywords: Angle-Resolved Photoemission Spectroscopy, Crystal Structure, Multiple Phases

INTRODUCTION

Paramagnetic metals and undergo structural & magnetic transitions revealing a spin density wave (SDW) state as the magnetic ground state [1, 2]; Fe atoms with a magnetic moment close to a Bohr magneton [2, 6] are the parent compounds of Fe-based superconductors. These materials display a wide range of anomalous behaviours, such as superconductivity in a pressure-induced non-magnetic phase [7, 8], coexistence of magnetic order and superconductivity [7, 8], etc. Spin-fluctuation theory of superconductivity is called into question due to the discovery of superconductivity under pressure [10, 11]. $CaFe_2As_2$ serves as a paradigm case study for this type of research. At ambient temperature, it possesses a para- magnetic tetragonal structure, but below 170 K, it changes to an orthorhombic antiferromagnetic (AFM) phase [2, 3, 6]. Collapsed tetragonal (cT) phase, which does not display magnetic order, is formed when a small pressure (> 0.35 GPa) is applied to the system. Numerous research have been conducted on this system, with varying results [13, 14]. Doped $CaFe_2As_2$ under pressure has been found to exhibit Tc as high as 45 K [15], with the cT phase being responsible for the superconductivity [16]. The spin-fluctuation explanation of superconductivity is supported by studies that fail to detect superconductivity in the cT phase. Superconductivity, structural phase, and spin fluctuations are all yet open questions. To shed light on the mysterious exoticness of these materials, we investigate the Fermiology of $CaFe_2As_2$ across a range of temperatures and pressures, where we find indications of a cT phase concealed within the structural phase at atmospheric pressure.

RESULTS

High temperature solution growth with Sn as an aux was used to produce $CaFe_2As_2$ single crystals . Energy dispersive x-ray analysis (EDAX), x-ray photoemission, and x-ray diffraction (XRD) were used to confirm the material's composition and structure. The sharp Laue Pattern guaranteed the crystals were all single crystals. Using a Scienta R4000 WAL electron analyzer with an energy resolution of 15 meV and an angle resolution of $\approx 0.3^{\circ}$., angle resolved photoemission (ARPES) measurements were taken at Elettra, Trieste, Italy and TIFR, Mumbai. A pristine, mirror-like surface was achieved by cleaving the materials on-site along the ab plane at a pressure of 4×10^{-11} Torr. For the electronic structure computations, the Wien2k programme was used to capture the full potential linearized augmented plane wave method (FLAPW) with $10\times10\times10$ k-points within the rst Brillouin zone, and $39\times39\times10$ k-points were used for the Fermi surface calculations. In order to determine the Fermi surfaces, Xcrysden was used.

It has been noted that the sample preparation procedures have a significant impact on the electrical characteristics of Fe-based compounds. For instance, $SrFe_2As_2$ exhibits both superconductivity and ferromagnetism simultaneously; the volume proportion of each phase is determined by the preparation circumstances, i.e. the degree of strain in the system . We looked at a variety of single crystalline $CaFe_2As_2$ samples, all of which showed magnetic and structural

changes at 170 K, which is consistent with the reported results. The photoelectric effect forms the basis of this method, as it is a rapid process that allows for instant approximation to capture the important elements of the experimental results. As a result, ARPES is able to expose the signature of phases that are obscured by other experimental approaches utilising slower time scales, and it can capture the local electronic structure at a fast time scale.

Figure 1.1 displays ARPES data, which reveals multiple energy bands and some very cool evolutions. In Fig. 1.1(a), the energy bands generated from ab initio density functional theory are superimposed on the data at 200 K (paramagnetic tetragonal phase). This band narrowing is commonly observed to occur as a result of electron correlation generated effects, and it manifests correlated behaviour of Fe 3d electrons. The energy scale of the estimated data is compressed by around 40% to match the bandwidth of the experiments.

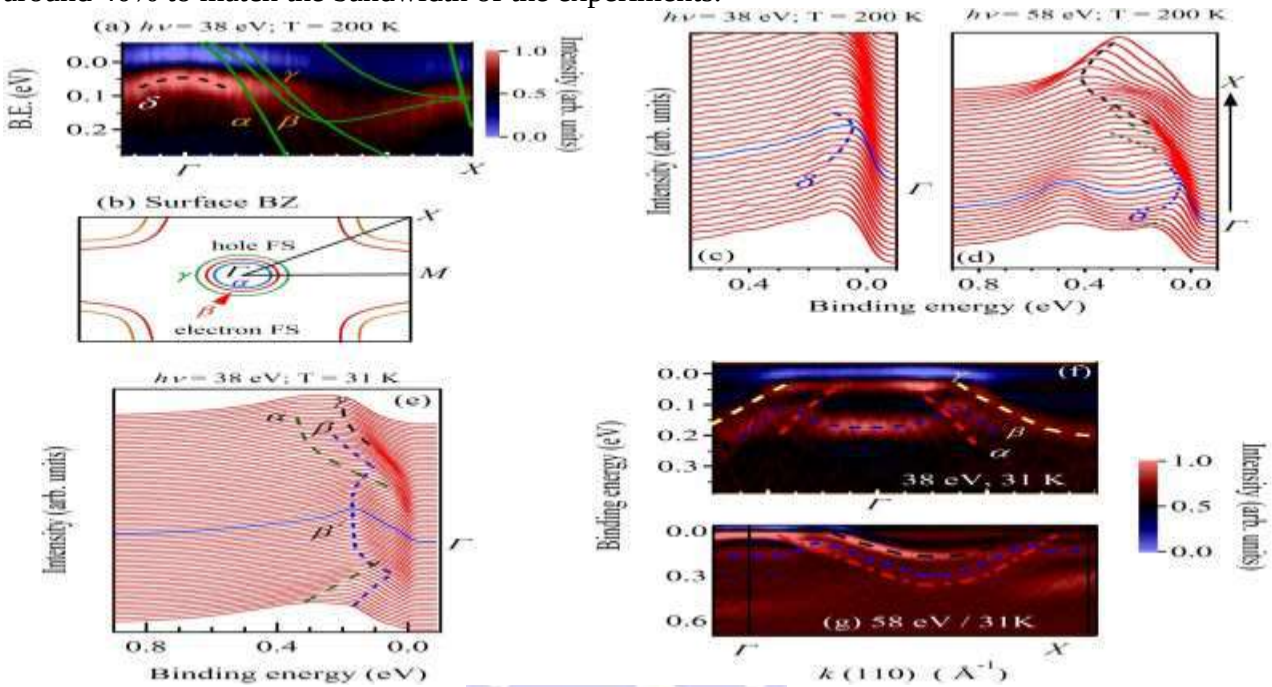

Fig. 1.1: ARPES results of $CaFe_2As_2$. (a) The second derivative of the ARPES data at 200 K ($h\nu$ = 38 eV) exhibiting three energy bands (α, β and γ). The lines represent the *ab initio* results for the tetragonal structure after 40% compression of the energy scale. There exists one additional filled band denoted by δ around Γ-point. (b) Schematic of the surface Brillouin zone (BZ). (c) EDCs at 200 K with $h\nu$ = (c) 38 eV and (d) 58 eV exhibiting distinct signature of the δ band. (e) EDCs at 31 K ($h\nu$ = 38 eV) exhibit α and γ bands crossing the Fermi level. The folded β in the SDW state is denoted by β'. Second derivative of the ARPES data at 31 K for $h\nu$ = (f) 38 eV and (g) 58 eV demonstrating again the band folding scenario with clarity.

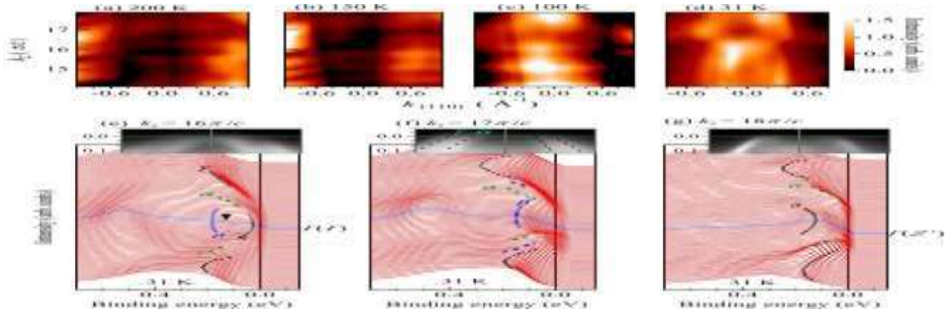

Fig. 1.2: Fermi surface in the kx- kz plane at (a) 200 K, (b) 150 K, (c) 100 K and (d) 31 K. The Fermi surface topology is two dimensional at 200 K and 150 K. A kz-dependence of the

Fermi surface is observed at 100 K and 31 K. EDCs at 31 K at kz = (e) 16π/c, (f) 17π/c and (g) 18π/c; the insets are the same ARPES data in color plot. α and γ bands appear to cross ϵF at all kz values. β′ band is the folded β band in the SDW state. In (e), the down arrow indicates a shoulder due to δ band, which is distinctly visible in (g). The band below ϵF at Γ is denoted by κ.

Precursor effect linked with magnetic transitions likely accounts for the low 200 K J band intensities seen in many systems .In the (00kz) spectra, when temperatures rise, the δ band disappears and the band shifts towards ϵ_F (the spacing between J and δ band increases). The δ band may be clearly seen at 200 K and over the entire temperature range investigated. We calculated the energy bands of $CaFe_2As_2$ in the cT phase, keeping the lattice constants at their experimental values under pressure (P = 0.63 GPa), in order to understand where these extra bands (and) came from. Figure 1.3(c) displays the outcomes for the value of zAs = 0.372 (tetragonal phase). Below ϵF, two of the three Fe 3d bands arise and become degenerate; they are indicated by the notation. The third, labelled as, crosses ϵ_F almost exactly at the moment where F becomes a hole. The Fermi surface map shows that the matching hole pocket has a weak k_z dependency, providing evidence for this conclusion. Even the hole pocket associated with the -band shifts below F at point if the experimentally observed value of zAs (= 0.3663 at P = 0.63 GPa) is utilised in the calculations, which equates to a (lower pnictogen height). It is clear that zAs is a crucial factor in determining whether or not the Fermi surfaces persist, which in turn causes charge fluctuations that significantly affect the system's physics. Although the Fe-moments are still finite, the loss of magnetic order due to the absence of hole pockets corresponding to the -band surrounding is a favourable situation for quantum spin fluctuations. The theoretically observed energy bands corresponding to the cT phase with 3D topology of the Fermi surfaces are strikingly similar to the and bands in the experimental observations.

It is evident from the provided results that the structural alterations at 170 K are distinct from the three-dimensionality of the Fermi surface at low temperatures. The structural change may be taking place throughout a broad temperature range, as observed in other systems . In this case, the electronic structure is derived from the features corresponding to the dominant structure, and the structural transition temperature represents the temperature at which the system transitions from the structural phase dominated by one structure to the one dominated by the other structure. Curiously, a different scenario is suggested by the data in Fig. 1.5.

The results of our investigation of the ARPES data from sample 2 (s2), which we refer to as the second sample in our investigation, are displayed in Fig. 1.4(a).

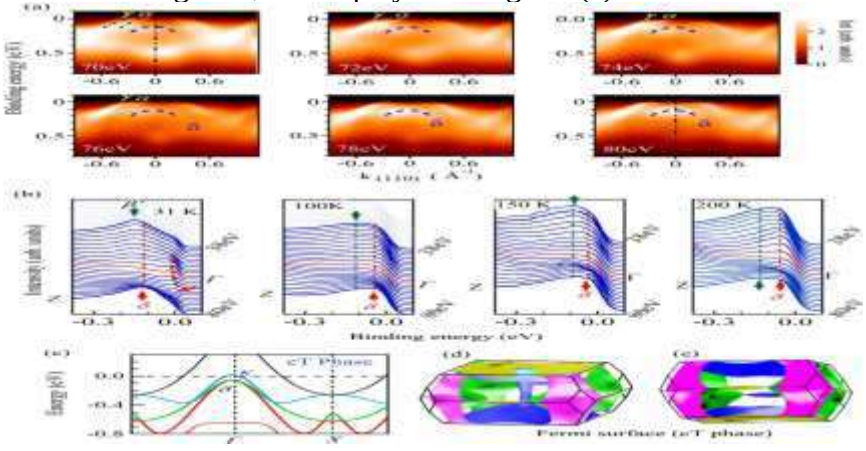

Fig. 1.3: (a) Energy bands at 31 K probed with different photon energies exhibiting distinct signature of α and γ bands crossing the Fermi level, and one additional band, δ. (b) Normal emission EDCs at 31 K, 100 K, 150 K and 200 K as a function of photon energies. The red line in each plot correspond to the Γ point in the k_z axis. Signatures of δ and β' bands are shown. (c) Calculated energy band dispersions of

$CaFe_2As_2$ in the cT phase with z_{As} = 0.372 (the value in tetragonal phase). Calculated Fermi surfaces in the cT phase with (d) z_{As} = 0.372 and (e) z_{As} = 0.3663 (experimental value at P = 0.63 GPa).

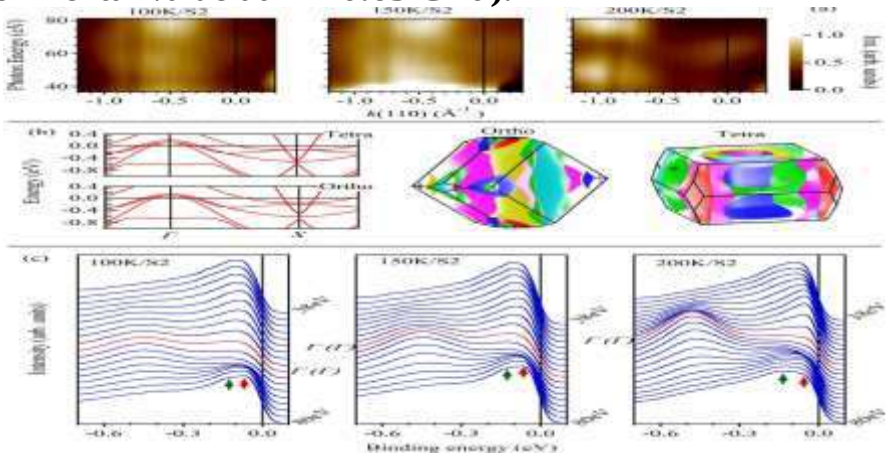

Fig. 1.4: (a) Fermi surface of sample 2 in kz - kx plane at 100 K, 150 K and 200 K. Here the two dimensional topology of the Fermi surface is preserved down to 100 K. (b) Calculated energy bands and the Fermi surfaces for tetragonal and orthorhombic structures in the paramagnetic phase exhibiting similar energy band dispersions and Fermi surface topologies. (c) EDCs at Γ point at 100 K, 150 K and 200 K, as a function of photon energies (kz). The red line in each plot correspond to the Γ point in the kz axis. Two up arrows exhibit signature of two features (β′ and δ bands).

As predicted , the Fermi surface topology is found to be two-dimensional at 200 K, and it maintains its similarity all the way down to 100 K, suggesting that the temperature for the shift in Fermiology is even lower; the 31 K results from different samples are comparable, therefore they are not displayed here. The volume of the Fermi surface in the orthorhombic phase decreases slightly both above and below the transition temperature of 170 K. Two-dimensional Fermi surfaces are observed in our tests on a wide variety of samples between 100 K and 200 K. Coherent characteristic (quasiparticle part of spectral function appearing at Fermi level) is well captured by density functional theory (DFT). Therefore, we used DFT formalism to explore the electronic structure shift caused by the structural modification, while keeping the system's magnetic intact. Figure 1.4(b) depicts the predicted electronic band structure for the tetragonal and orthorhombic structures. In accordance with the data from experiments, the tetragonal structure has three hole pockets centred on the point. There is no loss of any of the three hole pockets in the orthorhombic phase; instead, the Fermi surfaces shrink somewhat, increasing the distance between the Fermi sheets that are centred on the and X points. The or- thorhombic structure has lattice constants that are quite similar to those in the tetragonal phase, so the shift of 1% is not surprising. Both the tetragonal and orthorhombic Fermi surfaces estimated agree well with the experimental scenario. In Fig. 1.4(c), we display the spectral functions from s^2 acquired as a function of kz; each spectrum corresponds to a (0,0,kz) point gathered by adjusting the photon energies, which may be used to learn more about the development of Fermiology. Fe $3d_{z2}$ orbitals make up the bulk of the energy band near the 0.45 eV binding energy, and this band is constant across all temperatures. The comparison of the photoemission cross-section of the various electronic states constituting the valence bands reveals the presence of two additional peaks near the Fermi level, indicated by the arrows in the figure; the features are most distinct in the spectra at high photon energies near 80 eV, indicating dominant contribution by Fe 3d states. However, the signature of the features remains at all the photon energies and does not appear to breach the Fermi level in the entire kz-range investigated, despite the fact that the overall intensity of the patterns is dramatically reduced in the photon energy range 50 - 60 eV due to the decrease in photoemission cross-section.

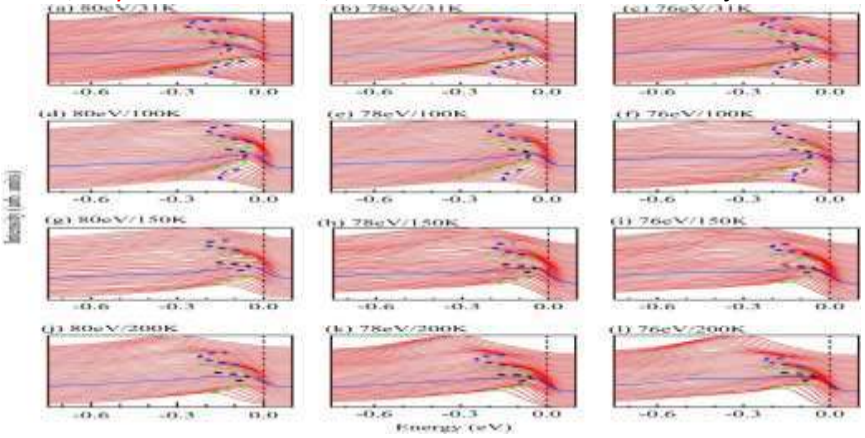

Fig. 1.5: EDCs with different photon energies and different temperatures as shown in the legends. The vertical column show the evolution of the energy bands as a function of temperature and the horizontal panels show the evolution with probing photon energy at the same temperature. It appears that *δ* band (black dash) survives even at 200 K and shifts to higher binding energy with the decrease in temperature.

In Fig. 1.5, we examine the δ band's temperature change by plotting the ARPES data of s1 along the ΓX axis. Its effective two-dimensional topology is limited to the $k_x - k_y$ plane, as shown by the evolution of the bands with photon energy in the horizontal panels, and by the presence of a discrete band with the same binding energy. As the temperature is raised from 31 K, the band moves closer and closer to the Fermi level. Surprisingly, this δ band persists all the way up to 200 K, well above the magnetic and structural transition temperature of 170 K. This suggests that the relevant phase is stable over the whole temperature range investigated.

DISCUSSIONS

While magnetic and structural transitions are generally consistent, the appearance of a kz-dependent Fermi surface is an intriguing outlier. More distinct and bands can be seen in a high-quality sample with brilliant, sharp, and well-defined Laue spots in the x-ray Laue diffraction pattern. These bands can be seen in other samples, but their clarity is reduced due to the effects of disorder and scattered strain. Energy band calculations for various structural phases agree with experiment, showing that the cT phase corresponds to the and bands. Antiferromagnetically ordered orthorhombic $CaFe_2As_2$ has the lowest computed ground state energy, which is very similar to the ground state energy of the nonmagnetic cT phase . Therefore, it is anticipated that even a slight change, such as the application of pressure and/or a change in the preparation condition, will have a considerable impact on the crystal structure. This is because the strain in the samples is amplified by thermal compression, which also explains why the and bands move as the samples cool.

Non-infinite electron-electron Band narrowing and local electron character brought on by strong Coulomb repulsion between Fe 3d electrons boost spin fluctuation and also have an effect on charge excitations. However, strong Hund's coupling among the 3d electrons seeks to align their spin moments and boosts spin fluctuations without significantly impacting charge excitations. Ground state Fe atoms have magnetic moments near to a Bohr magneton due to the effects of competing interactions, which cause the Fe-moment to be significantly reduced from its atomic . These systems achieve superconductivity through the suppression of magnetic order, which is thought to be caused by spin fluctuations. The inherent tensions clearly push this material in the direction of quantum fluctuations. Extreme charge and spin fluctuations are produced close to the vanishing -point by the nesting of vanishing Fermi surfaces.

CONCLUSION

There are likely many phases in Fe-based compounds that cannot be directly observed by any single experimental probe, leading to baffling electrical characteristics. By carefully studying the

Fermi surfaces at intermediate temperatures and utilising high photon energies, we are able to use ARPES to uncover signatures of the cT phase buried in the conventional structural phase. It is possible that the varied exotic electronic properties of these materials can be attributed to the enhancement of quantum fluctuations brought about by the coexistence of the cT phase within the ambient phases.

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