Origin of superconductivity in Ni-doped SrFe$_2$As$_2$, an Insight from DMFT

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I describe the issues of the competing orders in normal state of a parent and Ni-doped iron pnictide superconductor, SrFe$_2$As$_2$, using LMTO band structure calculation plus multi orbital dynamical mean-field theory. Strong, electronic correlations along with minimal antiferromagnetic instability create a partially gapped Fermi surface, in qualitative agreement with earlier experiments. A good semiquantitative similarity in both normal and ordered state with the earlier experimental transport data is observed along with suppression of magnetic ordering and explained within a correlated, multiorbital viewpoint. These results suggest that soft electronic fluctuation mediate superconductivity in broad class of parent and underdoped 122 pnictides with suppression of magnetic ordering.

I. INTRODUCTION

After two decades of the discovery of cuprate high temperature superconductivity (SC)\textsuperscript{1}, a new type of high-temperature superconductors has been launched in layered iron compounds in early 2008\textsuperscript{2}. Addition of these iron compounds to the list of ill-understood strongly correlated electronic system (SCES) provided a new direction of searching high-$T_c$ superconductor other than cuprate based systems. Interplay of various interesting effects are almost common in SCES. Magnetism, superconductivity, and interplay of multiorbital interaction and its structure is a novel topic of research in different SCES till date. Despite intense efforts a complete understanding of different properties of SCES is still debatable.

Discovery of superconductivity at 26 K in fluorine-doped LaFeAsO\textsuperscript{2} has triggrued a global research to explain the origin of SC in this novel compounds. Iron based superconductor has always been surprising because of strong local magnetic moment of iron though iron itself is a superconductor under high pressure (20 Gpa, 1.8 K)\textsuperscript{3}. Currently, there is an increased interest in 122 pnictides i.e. AF$_2$As$_2$ (A=Ba, Ca, Sr, Eu) due to pressure induced SC in this compounds\textsuperscript{4,5}. Among them particularly SrFe$_2$As$_2$ shows external pressure induced SC at relatively high temperature with $T_c = 21$ K\textsuperscript{6}. In pressure temperature phase diagram of SrFe$_2$As$_2$ the antiferromagnetic phase transition is found at about 198 K and alongwith applied pressure SC appears suppressing magnetic ordering\textsuperscript{7}. How much strong is the Coulomb interaction in iron pnictides is another important query in the field\textsuperscript{8,9} with two distinct debated studies: the first proposes that the iron pnictides are weakly correlated i.e. Coulomb interaction is small compared to the bandwidth while the inelastic neutron scattering study posits strong correlation view with the observation of drastic reduction of sublattice magnetization\textsuperscript{10,11}. If it is weakly correlated a standard Fermi liquid response in the normal (metallic) state is expected where the spin density wave (SDW) is destroyed and dc resistivity should show a $T^2$ dependence with a very low residual resistivity at low temperature but experimental data exhibits different $T$ dependence\textsuperscript{12}. While weak correlation should result in a clean Drude peak with Fermi liquid behavior in optical conductivity but perusal of experiments show a pseudogap in optical conductivity\textsuperscript{13} with quasilinear temperature dependence of resistivity concluding presence of strong correlation in SrFe$_2$As$_2$. All these experimental data point out that SC arises in this iron pnictide as an instability of non-Fermi liquid metallic state. Earlier theoretical studies also predict incoherent metallic behavior close to a Mott inslator in other pnictides (LaFeAsO, LaFePO etc.)\textsuperscript{14,15}. Such type of theoretical study is not available in Ni doped SrFe$_2$As$_2$. Though many rich theories have been discovered, the mechanism of superconducting transition is remaining as a controversial subject. How from an incoherent metal the SC can arise in parent material with iso electronic substitution will be aim of this article.

Theoretically explaining mechanism of high $T_c$ superconductivity lies on identifying features that are directly tied to high $T_c$, among complexity of different physical phenomena. So in this paper it is shown that the 122 iron pnictides in doped or parent phase can be a new class of materials to test the superconductivity. I have compared the electronic properties of Ni doped and parent SrFe$_2$As$_2$ obtained from DMFT with earlier experiment and established the idea of electronic correlation behind different transition in its doping temperature phase diagram. Since parent 122 pnictide is doped with Ni, the magnetic interactions become weak. Electronic structure near Fermi level is mainly from $t_{2g}$ orbitals, which form electron pockets in Fermi surface and superconductivity emerges as a consequence of weak of magnetic order.

In following sections, I calculated the DMFT spectral DOS and optical conductivity of parent and Ni doped SrFe$_2$As$_2$. I will show how orbital selective Mott transition underpins the properties of this 122 pnictide. In particular I will present specific non-FL feature behind phase transition of the system.

II. METHOD

The one electron band structure of SrFe$_2$As$_2$ was calculated using the linear muffin-tin orbital (LMTO) scheme\textsuperscript{16} in the atomic sphere approximation (ASA) as a function of doping concentration in the orthorhombic
as well as tetragonal phases. Lattice parameter inputs of LMTO are taken from earlier experimentally determined $a$, $b$, $c$, and $z_{As}$ as a function of doping and temperature. In SrFe$_2$As$_2$ the effective electronic states include the carriers in the FeAs layers. In Fig. 11 show the orbital dependent DOS of five Fe-d bands which have significant weight at Fermi energy ($E_F$) in both parent and doped Sr122 system. The density of states (DOS) shows that among the five d-orbitals $xy$ and $z^2$ band is almost gapped at the Fermi energy and other bands have large weight at the $E_F$. This noninteracting DOS in doped SrFe$_2$As$_2$ changes at the Fermi energy due to redistribution of electrons with doping. Theoretically the system is doped using supercell approach and paramagnetic calculations are carried out with $H/mmm$ space group symmetry (No. 139) in tetragonal phase and $Fmmm$ space group (No. 69) in antiferromagnetic orthorhombic phase respectively. The noninteracting part of the Hamiltonian is given by

$$H_0 = \sum_{k,a,\sigma} \epsilon_{k,a} c_{k,a,\sigma}^\dagger c_{k,a,\sigma}$$

where $\epsilon_{k,a}$ is band energy for the five Fe-d bands. The crystal field with $S_4$ symmetry creates inter-orbital splitting in iron-pnictides (Fe-pn). As discussed earlier the strong coupling view agrees well with quasi-local spin fluctuations associated with Mott physics and subsequent ARPES and STM studies at normal state. Theoretically it is well established that in a strongly correlated system LDA (local density approximation) is unable to describe excited states while LDA+DMFT (Dynamical mean field theory) has been successful in describing physical properties of various correlated electronic structures. The multi orbital iterated perturbation theory (MO-IPT) is used as an impurity solver in DMFT: though not exact, it is a computationally fast and effective solver, and has been proven to work very well in real multi-band systems throughout all temperature range. I choose $U=0.5-0.7$ eV and $U'=0.2$ eV as intra and inter-orbital Coulomb interaction as appropriate (for good description of earlier experiments) parameters and $J_H$ is determined from the relation $U'=U-2J_H$. The interaction part of the Hamiltonian is given by,

$$H_{int} = U \sum_{i,a} n_{ia\uparrow} n_{ia\downarrow} + U' \sum_{i,a,b,\sigma,\sigma'} n_{ia\sigma} n_{ib\sigma'} - J_H \sum_{i,a,b} S_{ia} S_{ib}$$

Further in pnictides the relevant phonon mode which couples electrons is given by $H_{el-l} = g \sum_i ( A_i + A_i^\dagger) c_i^\dagger c_i + h.c.$ (g is taken 0.01 after checking all possible realistic values to get a good description of experiments). Given these I have followed earlier procedure to incorporate interaction effects in the five-band model above within the DMFT.

III. DMFT RESULTS

In Fig.2a I show DMFT local density of states for different temperature. A significant change is noticed in the sharpness of the DMFT spectral function for the bands which are very close to the Fermi energy as the temperature crosses 200K. The dynamical correlations lead to change in spectral weight over an higher energy scale. At 200 K spectral DOS reveals finite energy gap as shown in Fig.2a. Thus these result strongly suggest that AFM correlation sets in orbital selective Mott transition (OSMT) in 122 pnictide (See supplementary information for smaller range near Fermi energy). The DMFT studies also suggest that the selective gaps will also appear in band structure at strong limit of the AFM correlations, while a Mott transition is caused by strong Coulomb interaction notwithstanding the magnitude of magnetic correlations. In parent SrFe$_2$As$_2$ the OSMT coincides with the AFM transition which thus establishes strong magnetic order in the system. Further there is no sign of FL quasiparticle signature in the low energy spectra, which drive orbital selective Mott transition in the d-band. Fig.2b presents spectral functions of SrFe$_{1.86}$Ni$_{0.14}$As$_2$ as a function of temperature. With increasing x, a and c-axis lattice parameters are changed. Increasing Ni concentration change the spectral DOS in a significant way. The DMFT spectral function becomes more coherent which indicates the reduced correlation. Finite spectral weight at the Fermi energy represents the metallic nature of the doped 122 pnictide, as the weight decreases with increasing $T$ the system evolves to incoherent metal, the self energy also shows the deviation from $\omega^2$ behaviour. As the doping is increased from $x=0.14$ to $x=0.2$ a small pseudogap is formed in one of the d-band (Fig.2c) which also predicts to set an ordering at low temperature where the electronic correlation is suppressed and incoherent fluctuations are supposed...
In the parent Sr-122 system, conductivity ($\sigma$) to a very good approximation. Fig. 3 shows the optical conductivity results in a gap-like structure. Further, a shoulder appears near 0.5 eV and the Drude response shrinks with decreasing $T$. The formation of gap in optical conductivity has been observed earlier also. These results conclude that Fermi surface is gapped at few areas of the Brillouin zone due to the AFM ordering. The DMFT optical conductivity with two different Ni doping level is presented in Fig. 3b and Fig. 3c. The spectral weight transfer with cooling is still discernible in $\sigma(\omega)$ but a large reduction is observed at 100 K, rather than below 200 K as in parent Sr122 which may be related to magnetic ordering transition. This result corresponds to the decrease in ordering temperature due to Ni doping. However, with doping the shoulder like feature is found below 100 K in the energy range 0.1 eV which is smaller than parent Sr122. Increase in the Ni doping $\sigma(\omega)$ follows the same trend overall. The spectral weight transfer becomes smaller with narrower Drude response and the shoulder like feature also become insignificant throughout the temperature range except 10 K. $\sigma(\omega)$ is identified almost flat in every temperature. Based on these considerations and the optical conductivity results, it can be inferred that magnetic ordering transition is suppressed due to Ni doping and the electron lattice interaction plays a significant role in suppression of magnetic ordering.

Fig. 4a presents the electrical resistivity $\rho(T)$ of parent and doped SrFe$_2$As$_2$. The dc resistivity is computed here using the Kubo formalism in DMFT. Interestingly $\rho(T)$ for Sr122 manifests metallic behavior, decreasing with $T$ from 300 K and linear in $T$ till around 200 K. At around 200 K, $\rho(T)$ manifests a sharp kink, which is exactly the temperature where AFM ordering sets in. With Ni doping, the anomaly in resistivity becomes less distinct and is defined by change in the slope of resistivity. Finally in x=0.2, no such anomaly is found. The sharp reduction of $\rho(T)$ in parent material is changing behavior with increased Ni concentration as it is shifted to lower temperature in full accord with experimental data. This type of behavior in resistivity has already been observed in other doped 122 pnictides. Since this anomaly is associated with a magneto structural transition so it can be due to reduced incoherent scattering at the onset of magnetic ordering and structural transition associated changes in carrier concentration. Also, this concludes the role of electronic scattering in resistivity. To find out actual $T$ dependence of resistivity if $\rho(T)$ is fitted with the expression $\rho(T) = \rho_0 + AT^n$ for the parent Sr122. In the AFM ordered region it is found that the value of $n$ remains around 2.7, this fact is rather unusual and it represents quantum fluctuation around that region.
To investigate the effect of doping on the magnetic order and fluctuating moments, I computed the dynamic magnetic susceptibility $\chi(\omega)$, which estimates the temporal and spatial dependence of fluctuating magnetic moments. In Fig. 4b the $\chi(\omega)$ varying with real frequency is presented for both undoped and doped phases. The Matsubara frequency dependent local dynamic susceptibility is calculated from DMFT first and then it is used to get $\chi(\omega)$ as a function of real frequency using maximum entropy method. Sharp peak is noticed in $\chi(\omega)$ at low energy which dies out with increasing energy. The peak here evidents large fluctuating moment which is very obvious in the high temperature-phase. Whereas a substantial drop in local moment in the low temperature ordered phase makes the susceptibility diminishing at high energy. Similarly in the doped phase also the peak height shrinks which establishes reduction of local moment oscillation in the ordered and doped 122 pnictide.

Now to further investigate about the ordering in the doped phase I calculated the degree of correlation. To know about that I will present effective mass ($m^* = 1/Z$), where $1/Z = (1/\delta \Sigma/\delta \omega)_{\omega=0}$. In a Landau Fermi liquid $Z$ is denoted as the quasiparticle weight, for a non-interacting system $Z$ becomes 1, and for a strongly correlated system it is very much smaller than 1 and near about zero for an insulator. I have computed effective mass for all the five d orbitals and plotted them with change in doping in Fig. 4a inset. A substantial drop in the $m^*$ for all Fe-d orbitals is found while varying doping in Sr122. It is found that the $d_{xz}$, $d_{yz}$ orbitals are less correlated and the $t_{2g}$ orbitals ($d_{yz}$, and $d_{xy}$) are more correlated in parent Sr122. The effect of doping on effective mass is observed to undergo a large change in the $d_{xz}$ orbital. This iron pnictide in undoped phase exhibit AFM ordering which is due to strong effective NNN (next nearest neighbour) exchange coupling between two iron atoms. This NNN coupling must originate from coupling between iron $t_{2g}$ d-orbitals and the anion p-orbitals.
and superconducting pairing with extended s-wave symmetry is expected to stem from this AFM interaction. The electronic structure of $t_{2g}$ orbitals near Fermi level generates the AFM interaction which is expected to be the key of phase transition in the 122 pnictide. The change in effective mass due to doping in $t_{2g}$ d-orbitals puts another proof of development of superconductivity with suppressed magnetic ordering.

Here, I presented a theoretical study of parent and doped SrFe$_2$As$_2$ to investigate the magnetism, superconductivity following suppressed magnetic ordering and the relation of crystal structure and charge doping to the different phases. Started with LMTO electronic structure calculations the study is centred on doping dependent changes in transport studies and photoemission spectras for Sr-122. Since the electronic structure is quite similar to the other pnictide families so the physical properties can be predicted and this approach provides insight into many questions. Since accurate description of the interaction of the iron arsenide layer and interlayer distance from geometry optimized density functional calculation is still debatable I have taken experimentally determined lattice parameters as function of temperature and doping. Electronic structure calculation essentially represent the signature of structural and magnetic order related changes with increased doping or temperature but the presence of correlation in the system is treated within DMFT. It is found that the structural transition in the 122 compounds is intrinsically connected with antiferromagnetic transition. I noticed suppression of the magnetic ordering with Ni doping which is in excellent accord with earlier experiments. Moreover theoretical ARPES studies (See Supplementary Information) suggest FS is gapped only in few direction. Doping dependence of the parent Sr122 in this theory follows correct trends as in experiments but a quantitative comparison will call for a quantitative agreement with an entire range of spectral portions of Brillouin zone corresponds to an divergence of effective mass. Thus, altogether these strongly supports the proposal of unconventional SC phase from an extended s-wave pairing. Similar results can be expected for same parent structure.

IV. ACKNOWLEDGEMENT

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1. J.G. Bednorz, K.A. Muller, Z. Phys. B- Condensed Matter 64, 189 (1986).
2. Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. 130, 3296 (2008).
3. K. Shimizu, T. Kimura, S. Furomoto, K. Takeda, K. Kontani, Y. Omukai, and K. Amaya, Nature 412, 316 (2001).
4. M. S. Torikachvili, S. L. Budko, N. Ni, and P. C. Canfield, Phys. Rev. Lett. 101, 057006 (2008); M. S. Torikachvili,
S. L. Budko, N. Ni, and P. C. Canfield, *Phys. Rev. B* **78**, 104527 (2008).

P. L. Alireza, Y. T. C. Ko, J. Gillett, C. M. Petrone, J. M. Cole, G. G. Lonzarich, and S. E. Sebastian, *J. Phys.:Condens. Matter* **21**, 012208 (2008).

H. Kotegawa, H. Sugawara, and H. Tou, *J. Phys. Soc. Jpn.* **78**, 013709 (2009); H. Kotegawa, T. Kawazoe, H. Sugawara, K. Murata, and H. Tou, *J. Phys. Soc. Jpn.* **78**, 083702 (2009).

K. Igawa, H. Okada, H. Takahashi, S. Matsuishi, Y. Kamihara, M. Hirano, H. Hosono, K. Matsuabayashi, and Y. Uwatoko: *J. Phys. Soc. Jpn.* **78**, 025001 (2009).

H. Takahashi, H. Okada, K. Igawa, K. Arrii, Y. Kamihara, S. Matsuishi, M. Hirano, H. Hosono, K. Matsuabayashi, and Y. Uwatoko, *J. Phys. Soc. Jpn.* **77**, 08 (2008).

J. J. Wu et al., *Scientific reports* **4**, 3685 (2014).

I.J. Mazin, M.D. Johannes, L. Boeri, K. Koepernik, D.J. Singh, *Phys. Rev. B* **78**, 085104 (2008).

C. de la Cruz et al., *Nature (London)* **453**, 890 (2008).

A.V. Boris, N.N. Kovaleva, S.S.A. Seo, J.S. Kim, P. Popovich, Y. Matiks, R.K. Kremer, B. Keimer, *Phys. Rev. Lett.* **102**, 027001 (2008).

O.K. Andersen, *Phys. Rev. B* **12**, 3060 (1975).

M. Yi et al., *Proceedings of the National Academy of Sciences* **108**, 6878 (2011).

H. Z. Arham et al., *Phys. Rev. B* **85**, 214515 (2012).

A. Taraphder, S. Koley, N.S. Vidhyadhiraja, and M.S. Laad, *Physical Review Letters* **106**, 236405 (2011).

S. Koley, M.S. Laad, N.S. Vidhyadhiraja, and A. Taraphder, *Physical Review B*, **90**, 115146 (2014).

S. Koley, N. Mohanta, and A. Taraphder, *Journal of Physics: Condensed Matter* **27**, 185601 (2015).

J.C. Loudon et al., *Phys. Rev. B* **81**, 214111 (2010).

S.R. Saha et al., *Phys. Rev. B* **79**, 214519 (2009).

J.M. Tomczak, and S. Biermann, *Phys. Rev. B* **80**, 085117 (2009).

W. Z. Hu et al., *Phys. Rev. Lett.* **101**, 257005 (2005).

D. Wu et al., *Phys. Rev. B* **81**, 100512(R) (2010).

H. Okamura et al., *J. Phys. Soc. Jpn.* **82**, 074720 (2013).

A. Leithe-Jasper, W. Schnelle, C. Geibel, and H. Rosner, *Phys. Rev. Lett.* **101**, 207004 (2008).

P. C. Canfield, S. L. Budko, N. Ni, J. Q. Yan, A. Kracher, *Phys. Rev. B* **80**, 060501(R) (2009).

V. SUPPLEMENTARY INFORMATION

A. Angle resolved photoemission within DMFT (IPT)

It is needed to inquire the microscopic origin of this ordering. Below $T = 200 K$ three d-bands are Mott localized due to interaction (but there is lack of clean gap due to inequal changes in Fermi surface, which completely remove the idea of FS nesting induced order). If this theory is to be credible, then other observations also must go hand in hand with experiments. In Fig.s1 and Fig.s2 I show DMFT one particle spectral function $A(k, \omega) = -ImG(k, \omega)/\pi$ which also reflects renormalized band dispersion $E_{k,a} = \epsilon_{k,a} + Re\Sigma_{k,a}(\epsilon)$.

![Image](image_url)

In-detail ARPES study is not available in literature for Ni doping but photoemission spectra for parent Sr122 can be compared with earlier results. While LDA with static Hartree Fock can show agreement with band dispersion, simultaneous explanation of ARPES will test the theory. For a high temperature dynamically fluctuating liquid ARPES is presumed to reveal broad continuum like features without Landau Fermi liquid quasiparticle peaks. However the DMFT results here provide a good description of extant ARPES dispersion upto high energies. Particularly they unveil the gap features in the ordered state. Fig.s2 shows the normal state map of the photoemission intensity from $\Gamma$ to $M$ direction. Fig.s1 illustrate the photoemission intensity (PES) energy distribution curves (EDC) across $\Gamma$ to $M$ direction in the antiferromagnetic ordered state. At low $T$ ($T = 5 K$, Fig.s1a-s1d) two bands can be identified to cross Fermi Energy providing two holelike Fermi surface portion around $\Gamma$ point of the Brillouin zone, in accord with earlier band structure calculations. Additionally there is a feature at around -0.2 eV, which can be identified as the valence band backfolded and hybridize with hole-like bands to open energy gaps near Fermi energy. Fig.s1e-Fig.s1h and Fig.s2 shows the ARPES intensity at two higher temperatures 200 K and 300 K. The EDCs are dominated by a strong feature dispersing towards M point. EDCs at 200 K reveals that the hole like bands moves towards $E_F$. From the ARPES at 200 K a new peak can be identified near $E_F$ originating from coupling to phonons which disappears with increasing temperature however at high $T$ an extra low energy feature is resolved which remains almost dispersionless. Fig.s3 and Fig.s4 show ARPES intensity along $\Gamma - M$ direction of Ni doped Sr122. The calculated EDCs are very similar in two doping state. However details of the electronic structure around $E_F$ for doped and undoped state were sensitive to this parameter. At high temperatures the dispersive features of the EDCs near $E_F$ is considerably suppressed due to orbital and spin fluctuations in the paramagnetic metal phase. As identified from the EDC plot at low temperature the spectral weight near Fermi energy is increased in the ordered state in comparison to paramagnetic phase. This finding infers that due to magnetic ordering transition the intensity suppression...
caused by the orbital and spin fluctuations is eliminated.
FIG. 5: (Color Online) DMFT ARPES Intensity of parent $SrFe_2As_2$ at (a)-(d) 5 K and (e)-(h) 200 K for $xy$, $x^2 - y^2$, $z^2$ and $yz$ bands along $\Gamma - M$ direction.
FIG. 6: (Color Online)(a)-(d) DMFT ARPES Intensity of parent $SrFe_2As_2$ at 300 K for $xy, x^2 - y^2, z^2$ and $yz$ bands along $\Gamma - M$ direction.
FIG. 7: (Color Online) DMFT ARPES Intensity of $SrFe_{1.86}Ni_{0.14}As_2$ at (a)-(d) 150 K and (e)-(h) 300 K for $xy, x^2 - y^2, z^2$ and $yz$ bands along $\Gamma - M$ direction.
FIG. 8: (Color Online) DMFT ARPES Intensity of $SrFe_{1.8}Ni_{0.2}As_2$ at (a)-(d) 150 K and (e)-(h) 300 K for $xy, x^2 - y^2, z^2$ and $yz$ bands along $\Gamma - M$ direction.
FIG. 9: (Color Online) DMFT density of states of parent $SrFe_2As_2$ at different temperature. In upper panel $yz$, $x^2 - y^2$ and in lower panel $xy$ and $z^2$ DOS is shown. Clear gap at $E_F$ manifests orbital selective Mott localization.