Normal State Correlated Electronic Structure of Iron Pnictides

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We describe the correlated electronic structure of a prototype Fe-pnictide superconductor, SmO$_{1-x}$F$_x$FeAs, using LDA+DMFT. Strong, multi-orbital electronic correlations generate a low-energy pseudogap in the undistorted phase, giving a bad, incoherent metal in qualitative agreement with observations. Very good semi-quantitative agreement with the experimental spectral functions is seen, and interpreted, within a correlated, multi-orbital picture. Our results show that Fe-pnictides should be understood as low-carrier density, incoherent metals, in resemblance to the underdoped cuprate superconductors.

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Discovery of high-$T_c$ superconductivity (HTSC) in the Fe-based pnictides is the latest among a host of other, ill-understood phenomena in $d$-band oxides. HTSC in Fe-pnictides emerges upon doping a bad metal with spin density wave (SDW) order at $\mathbf{q} = (\pi, 0)$. Preliminary experiments indicate unconventional SC. Existent normal state data indicate a “bad metal” without Landau Fermi Liquid (FL) quasiparticles at low energy. These observations in Fe-pnictides are reminiscent of cuprate SC. The small carrier density (giving rise to carrier pockets), along with Uemura scaling from $\mu$-SR similar to hole-doped cuprates strongly suggests a SC closer to the Bose condensed, rather than a BCS ($\xi \sim 1000 \alpha$) limit.

LDA studies show that SC in Fe-pnictides is associated with the Fe-$d$ states hybridized with As-$p$ states: this leads to two hole, and two electron-like pockets. Finding of van-Hove singularities and peaks at $\mathbf{q} = (\pi, 0)$ in the bare spin susceptibility indicates a SDW state, found within Hartree-Fock random phase approximation (RPA) studies of effective, two- and four-orbital Hubbard models, in apparent agreement with inelastic neutron scattering results. The observation of quasi-linear temperature ($T$) dependence of the resistivity, a pseudogap in optics, and a spin gap in NMR, however, is a benchmark feature of the relevance of strong, dynamical spin and charge correlations in the pnictides. In cuprates, these are seen in the underdoped state in the proximity of a Mott insulator (MI), suggesting that the Fe-pnictides might be closer to a MI than generally thought. Actually, the undoped pnictides show an insulator-like resistivity without magnetic order for $T > 150 \text{K}$. Onset of bad metallic behavior correlates with a structural (tetragonal-orthorhombic (T-O)) distortion at $T^* \approx 150 \text{K}$, below which SDW order sets in. The small carrier number apparently generated upon the structural distortion accords with the observed high resistivity, lending further credence to such a view.

The above suggests that one should study a single Fe-As layer with strong electronic correlations to begin with. Here, we study the five-orbital Hubbard model within the local-density-approximation plus dynamical-mean-field-theory (LDA+DMFT) approach, incorporating one-electron band structure aspects. Extant LDA+DMFT works give either a strongly renormalized FL or an orbital selective, incoherent, pseudogapped metal. Apart from the known sensitivity to the value of $J_H$ (Hund coupling), the LDA+DMFT spectra show noticeable qualitative differences at low energy. Are Fe-pnictides then strongly renormalized FL metals, or incoherent non-FLs in their normal state? Comparison with experimental one-particle spectra should go a long way toward resolving this question. We do this in this work.

Photoemission (PES) and X-ray absorption (XAS) studies are a reliable tool to study the correlated electronic structure of $d$- and $f$-band compounds. To date, the only PES experiments have been performed on SmO$_{1-x}$F$_x$FeAs and LaO$_{1-x}$F$_x$FeAs. Very recently, XAS has also been performed for SmO$_{1-x}$F$_x$FeAs. Together, they provide additional evidence for the “incoherent metal” normal state in Fe-pnictides. The PES spectra show a kink at low energy, $\Omega = 15 \text{meV}$, below the T-O distortion followed by SDW order. This kink sharpens with cooling, and evolves, apparently smoothly, across $T_c$. Its microscopic origin is an enigma. Is it related to the T-O distortion, or to the SDW transition? Is it observed only for electron-doped systems? Correspondingly, XAS shows a well-defined peak at 0.5 eV, and a transfer of weight from high energy (2.0 eV) to low energy with F doping, a characteristic feature of correlated systems. Answering these questions within a correlated electronic structure approach provides deeper insight into the underlying correlations in the non-FL metal phase, aiding in the search to identify mechanism(s) of superconductivity itself.

Starting with the high-$T_c$ tetragonal structure with lattice parameters found in Ref. 18, one-electron band structure calculations were performed for SmOFeAs using the linear muffin-tin orbitals (LMTO) scheme in the atomic sphere approximation. LDA provides valuable description of the relevant orbitals on a
single-particle microscopic level, but falls short of describing dynamical correlations in \(d\)- and \(f\)-band compounds. This requires “marrying” \(\text{LDA} \) to \(\text{DMFT}\), which enables direct access to the correlated spectral functions \([21]\). The one-electron part for \(\text{SmFeAs}\) is 
\[
\hat{H}_0 = \sum_{\mathbf{k},a,\sigma} \epsilon_a(\mathbf{k}) \hat{c}_{\mathbf{k},a,\sigma} \hat{c}^\dagger_{\mathbf{k},a,\sigma},
\]
where \(a = x^2-y^2, 3z^2-r^2, xz, yz, xy\) label the diagonalized, five \(d\) bands. The corresponding density-of-states (DOS) \((\text{Fig. 1})\) shows that all the five \(d\)-bands cross the Fermi energy, \(E_F\), but the \(3z^2-r^2\) band is almost gapped at \(E_F\). While the \(xy\)-band has a deep pseudogap at \(E_F\), the \(x^2-y^2, xz, yz\)-bands have large DOS at \(E_F\). In \(\text{Fe-pnictides}\), the $d^5$ configuration of \(\text{Fe}^{2+}\) dictates that the full, multi-orbital (MO) Coulomb interactions must be included. These constitute the interaction term, which reads
\[
\hat{H}_{\text{int}} = U \sum_{i,a} n_{ia\uparrow} n_{ia\downarrow} + U' \sum_{i,a \neq b} n_{ia\uparrow} n_{ib\uparrow} - J_H \sum_{i,a,b} \hat{S}_{ia} \hat{S}_{ib}. 
\]

We choose parameters employed by Haule et al. \([12]\), \(U = 4.0\) eV, \(U' = U - 2J_H = 2.6\) eV, and \(J_H = 0.7\) eV, along with the five LDA bands, and solve \(\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}\) within LDA+DMFT. To solve the MO-DMFT equations, we use the MO iterated-perturbation-theory (IPT) as an impurity solver. Though not quantitatively exact, it has many advantages. It is numerically very efficient, is valid at \(T = 0\), in contrast to QMC, and self-energies \((\Sigma(\omega))\) can be extracted very easily. These are of particular importance in a complicated MO situation that occurs in \(\text{Fe-pnictides}\).

Given orbital-induced anisotropies in the LDA, strong MO correlations renormalize various \(d\)-bands in widely differing ways. Generically, one expects these to partially (``Mott'') localize a subset of \(d\)-bands, leading to orbitally selective Mott transitions (OSMT), and bad metallic states \([22]\). This requires strong \(U, U'\). Within LDA+DMFT, this orbital selective mechanism involves two renormalizations: \(a\) static (MO Hartree) shifts rigidly move various \(d\)-bands relative to each other by amounts depending upon their on-site orbital energies and occupations, and, more importantly, \(b\) dynamical effects of strong \(U, U', J_H\) drive large spectral weight transfer (SWT) over wide energy scales. Upon small changes in bare LDA parameters, large changes in SWT lead to OSMT, as well as to incoherent metallic phases characteristic of a wide variety of correlated systems. With parameters for \(\text{Fe-pnictides}\) as above, does an OSMT occur \([13]\), or does it not \([12]\)? What is the origin of the observed incoherent metallic behavior?

To answer these questions, we now turn to our results. In sharp contrast to LDA, our LDA+DMFT results show drastic modification of the spectral functions. The dynamical correlations lead to dramatic spectral weight redistribution over large energy scales \(O(5.0)\) eV. Most interestingly, we find no FL quasiparticle signatures in the low-energy spectra; instead, the metallic state is totally incoherent. In contrast to earlier work \([13]\), this occurs even without a strict orbital selective Mott localization, though almost all bands are very close to Mott localization. The orbital-resolved self-energies (inset in \(\text{Fig. 1}\)) clearly reveal this aspect: large damping at \(E_F\) destroys the FL quasiparticle, giving an incoherent, pseudogapped, bad metallic state. This incoherent state has also been found in previous LDA+DMFT works \([12, 13]\), and, as discussed there, is in qualitative agreement with experimental observations. Here, however, we also show that our results are in very good semi-quantitative agreement with key features of the experimental PES and XAS spectra, see \(\text{Fig. 2}\).

Analyzing the LDA+DMFT spectra of \(\text{Fig. 1}\) we find that the \(3z^2-r^2, x^2-y^2\) as well as the \(xz, yz\) orbitals continue to be almost degenerate (the first pair is split by 0.06 eV). This is also reflected by the fact that the DMFT spectra of \(3z^2-r^2, x^2-y^2\) bands show noticeable similarities, even as, interestingly, large differences between them exist at level of LDA. Of interest are the sharp, very low-energy \((20\) meV below \(E_F)\) structures in the \(3z^2-r^2, x^2-y^2\) spectra. In the light of their near-degeneracy, we ascribe these peaks to the low-energy orbital fluctuations (coupled to charge fluctuations) in this two-fold degenerate sector. This is an interesting manifestation of the two-fold \(3z^2-r^2, x^2-y^2\) orbital degeneracy surviving in \(\text{Fe-pnictides}\), and explicitly requires strong MO correlations. Using LDA+DMFT, we have also estimated the spin state on \(\text{Fe}\) sites. With parame-
agreement with both PES and XAS data. Most importantly, PES reveals a kink at 15 meV upon cooling the sample below $T^* = 150$ K, where a T-O distortion, followed by the $\mathbf{q} = (\pi,0)$ SDW order, takes place. This feature sharpens with decreasing $T$, and weakens with doping, but does not undergo further change across the SC $T_c$. This appears to be a more generic feature of the electronic structure of Fe-pnictides, as similar evolution of the low energy pseudogap has also been resolved in LaO$_{1-x}$F$_x$FeAs [10]. Analysing our LDA+DMFT spectra for $n=6.2$ ($n$ is the total band filling of the $d$ shell), we find a sharp nonanalytic structure in $\Delta \rho^{\text{total}}(\omega)/d\omega$ at an energy of 20 meV, implying a kink in the DOS at that energy, in very good semi-quantitative agreement with PES results. Further, analyzing the orbital-resolved DOS, this feature is seen to originate from the $3z^2 - r^2, x^2 - y^2$ orbital degeneracy, as discussed above, its appearance below $T^*$ now has an attractive interpretation: it reflects the low energy, coupled charge-orbital fluctuations in this pnictide. It appears only below $T^*$ because the T-O distortion, interpreted as a Jahn-Teller instability, occurs at $T^* > 2\alpha$, and lifts this degeneracy. The itinerant (albeit incoherent) character of the system suppresses the bare $3z^2 - r^2, x^2 - y^2$ splitting to small values (20 meV, as pointed out above). Strong orbital fluctuations in this almost degenerate orbital sector coupled to one-electron Green functions enter the dynamical, second order contributions (of the generic form $\int G^{(0)}_a(\omega - \omega_1 - \omega_2)G^{(0)}_b(\omega_2)G^{(0)}_a(\omega - \omega_1)d\omega_1d\omega_2 = \int \chi^{(0)}_{ab}(\omega_1)G^{(0)}_a(\omega - \omega_1)d\omega_1$, with $\chi^{(0)}_{ab}(\omega)$ the inter-orbital susceptibility describing dynamical orbital correlations) to the self-energies, and hence show up in the LDA+DMFT spectra as sharp, low energy peaks in the orbital-resolved spectral functions, as seen in our results. We have also computed the LDA+DMFT spectra for hole doping ($n = 5.8$, dot-dashed line in Fig. 3). In contrast to electron doping, no noticeable change is observed in the low energy spectra; we predict that PES/XAS on hole-doped Fe-pnictides will show this in future.

Since the kink in PES is now related to the T-O distortion, rather than the SDW, it should be smeared out for $T > T^*$, exactly as observed. Therefore, its survival without apparent modification across $T_c$ is not connected to the destruction of SDW order apparently required for SC to emerge. If this turns out to be generic for Fe-pnictides, it would imply an indirect link, at most, to SC, to the extent that it reflects electronic structure changes (viz. removal of $3z^2 - r^2, x^2 - y^2$ orbital degeneracy) required for the SC instability to emerge from such a normal state. “Melting” of the T-O distortion upon F doping implies rapid suppression of this kink feature in our picture: this is indeed seen in our LDA+DMFT spectra for $n = 6.2$ (orange line) curves. Only a weak remnant of the 15 meV
kink is resolved at \( n = 6.2 \), as shown in the right inset of Fig. 2, indicative of surviving uncorrelated distortions among an “undoped” fraction of Fe-sites, even as long-range distortion melts with doping.

Based on this detailed theory-experiment agreement, we discuss the implications of our work on SC. First, since all \( d \) bands cross \( E_F \), SC should involve inducing the SC gap on all FS sheets. This does not necessarily conflict with two-band Hubbard model results [24], since the multi-band SC proximity effect [25] could operate here. Once SC pairing occurs in the \( d_{xz,yz} \) manifold, such an effect could induce secondary gaps over the remaining FS sheets. Second, our results indicate that having \( 3z^2 - r^2, x^2 - y^2 \) orbital degeneracy drives a Jahn-Teller T-O distortion, to the detriment of SC, as seen. Finally, our finding of a strongly incoherent “normal” state with drastically reduced charge carrier number (given the proximity to a Mott insulator in DMFT) is consistent with observations [1, 2, 4] indicating a non-FL metal with low carrier density SC. Many of these observations are reminiscent of those seen in HTSC cuprates [26] up to optimal doping, putting the Fe-pnictides into the “strongly correlated, unconventional, HTSC” category.

In conclusion, using a correlated electronic structure (LDA+DMFT) approach, we show that strong dynamical correlations are essential to proper understanding of the basic physics of Fe-pnictides. As in cuprates, the normal state that becomes unstable to unconventional SC is not a Fermi liquid. Our LDA+DMFT result gives an incoherent, non-FL state with small carrier density, as observed experimentally [4]. Very good semi-quantitative agreement with extant PES/XAS data lend strong credence to our view of Fe-pnictides as multi-orbital, strongly correlated materials close to the itinerant-localized boundary.

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