Normal-state correlated electronic structure of iron pnictides from first principles

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(Received 23 May 2008; revised manuscript received 17 September 2008; published 9 October 2008)

We describe the *correlated* electronic structure of a prototype Fe-pnictide superconductor, $SmO_{1-x}F_xFeAs$, using local-density approximation plus dynamical mean-field theory. Strong, multiorbital electronic correlations generate a low-energy *pseudogap* in the undistorted phase, giving a bad, incoherent metal in qualitative agreement with observations. Very good semiquantitative agreement with the experimental spectral functions is seen (for $U=4.0$ eV and $J_H=0.7$ eV) and interpreted within a correlated, multiorbital picture. Our results show that Fe pnictides should be understood as low-carrier density, incoherent metals, in resemblance to the underdoped cuprate superconductors.

DOI: 10.1103/PhysRevB.78.134511 PACS number(s): 74.25.Jb, 71.27.+a, 74.70.−b

I. INTRODUCTION

Discovery of high-T_c superconductivity (HTSC) in the Fe-based pnictides[1] 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[2,3] unconventional superconductivity (SC). Existent normal-state data indicate a "bad metal" without Landau Fermi liquid (FL) quasiparticles (QPs) at low energy.[1] These observations in Fe pnictides are reminiscent of cuprate SC. The small carrier density (giving rise to carrier pockets), along with Uemura scaling from μ-SR (Ref. 4) similar to hole-doped cuprates, strongly suggests a SC closer to the Bose condensed rather than a BCS (superconducting coherence length, $\xi \simeq 1000a$, with a the lattice spacing) limit.

How correlated are electronic states in Fe pnictides is an important issue in the field. Studies have spawned the rise of two distinct theoretical view points: the first posits that Fe pnictides are itinerant metals with *weak* electron-electron interactions, i.e., the Hubbard U is small compared to the local-density-approximation (LDA) bandwidth.[5,6] LDA studies show that the relevant electronic states in Fe pnictides are associated with the Fe-*d* states hybridized with As-*p* states; this leads to two holelike and two electronlike pockets.[5] Nesting features between separated electronlike and holelike pockets at $\mathbf{K}=(\pi,\pi)$ lead to SDW order with wave vector $\mathbf{q}=(\pi,0)$ in the *bare* spin susceptibility.[6,7] This SDW order, found within Hartree-Fock–random-phase-approximation (HF-RPA) studies of *effective*, two-, and four-orbital Hubbard models, is in seeming agreement with inelastic neutron-scattering results.[8] In these itinerant theories, the small sublattice *ordered* moment $M_s \simeq 0.3\mu_B$ (where μ_B is the Bohr magneton) is supposed to result from strong itinerance (large LDA bandwidth) in these pictures. It is important to notice that this should not be confused with the *atomic* spin on each Fe site. What is measured in the magnetic fluctuation probes such as neutrons, nuclear magnetic resonance (NMR), μSR, etc. is the quantum-mechanical and thermal *average* value of the sublattice magnetization operator, and in Fe pnictides quasi-itinerance as well as the strong geometric frustration resulting from the LDA hoppings (seen best in the tight-binding fits of Raghu *et al.*[9]). Indeed, Yildirim[7] has shown how this strong frustration translates into a frustrated J_1-J_2 Heisenberg model in the strong-coupling limit of the model, leading to the strong, frustration reduction in the sublattice magnetization, M_s. This shows that having strong correlations is quite compatible with the observation of a drastically reduced M_s measured in inelastic neutron scattering (INS). Consequently, it is not valid to necessarily link the observation of a small M_s with an itinerant model.

Are there other observations which can serve as a pointer toward the degree of correlated behavior in Fe pnictides? A perusal of available experiments at this stage of development of the field already (in our opinion, see also earlier work[10–13]) leads one to discern fingerprints of strong electronic correlations. First, if the Fe pnictides were indeed weakly correlated, one would expect a standard FL response in the "normal" metallic state for the range of dopings ($x>0.1$) where the SDW state is destroyed. In particular, the dc resistivity $\rho_{dc}(T) \simeq T^2$ and one would observe a low residual resistivity, $O(\mu\Omega\ \mathrm{cm})$, at low T. A clean Drude peak with characteristic FL properties should be visible in optics, and the spin susceptibility should be T independent. Perusal of experiments in the doped pnictides, however, shows a very different metallic state emerging for $x>0.1$. Observation of quasilinear temperature dependence of the resistivity, pseudogap in optics,[14] and a spin gap in NMR (Ref. 15) are benchmark features of the relevance of strong, dynamical spin, and charge correlations in the pnictides, and cannot be rationalized within a weakly correlated band (FL) scenario. Clear conclusions in early studies were hampered due to the difficulties associated with interpreting transport measurements on polycrystals. However, very recently, single crystals of AFe_2As_2 with $A=Ba,Sr$ were synthesized: the dc resistivity in doped samples was again found to be bad metallic and quasilinear in T, suggesting that bad-metallic, incoherent transport is an intrinsic feature of Fe pnictides.[16] Similar conclusions can be inferred from optics done in the same study, where the low-energy, "quasicoherent" peak is estimated to carry less than 5% of the total spectral weight, the remainder is distributed across a large energy scale $O(3\ \mathrm{eV})$, in incoherent electronic states. More evidence for the absence of FL-like QPs at low energy in doped pnictides comes from a recent angle-resolved photoemission study (ARPES), where

a low-energy pseudogap without any sharp quasicoherent FL peaks (the low-energy **k**-resolved spectra are anomalously broad) in the "normal" state evolves smoothly into the SC gap below T_c.[17] These observations constrain theoretical modeling: the SC arises as an instability of an incoherent, non-FL metal and the normal state above T_c is not a FL.

In cuprates, such features 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 state than generally thought.[10] Actually, the undoped pnictides show an insulator-like resistivity *without* magnetic order for $T>150$ K.[1] This suggests that, in absence of the tetragonal-orthorhombic (T-O) distortion, the undoped Fe pnictides could have developed into insulators at low T. Onset of bad-metallic behavior, even at low T, well below T_{SDW} for $x=0$, correlates with a structural (T-O) distortion at $T^* \simeq 150$ K, *below* which SDW order sets in. This T-O distortion seemingly generates a small carrier number and this reasoning is consistent with the observation of the high resistivity even at low T. This view has been pioneered by Si and Abrahams,[10] Baskaran,[11] and Seo *et al.*[12] In this context, earlier local-density approximation plus dynamical mean-field theory (LDA+DMFT) studies already indicate the possibility of incoherent metallic behavior[13,18] and, in fact, Haule *et al.*[13] characterize the Fe pnictides (with $U=4.0$ eV) as being close to a Mott insulator (the critical $U=U_c$ being 4.5 eV).

In context of first-principles modeling, extant work still leaves a few important issues open. These are the following: (i) if the system is in fact strongly correlated, one would expect that strong multiorbital (MO) correlations stabilize a high-spin (HS) state *locally* on each Fe site (again, not to be confused with the M_s measured in INS, for example). In the weak correlation scenario, strong itinerance is invoked to explain the strong reduction in the *ordered* spin moment from its atomic (HS) value. Which scenario is correct? (ii) If strong correlations stabilize the HS state locally on Fe, the mechanism for the drastic reduction in the ordered moment as seen in INS work would then involve geometric frustration effects.[7,10] We draw attention to the fact that this effect is intrinsic to the Fe pnictides and is seen in LDA calculations, where the hopping along diagonals is an appreciable fraction of, or even larger than, the nearest-neighbor hopping.[7] Unearthing the spin state of Fe should go a long way toward clinching this issue since in the band picture small U, and hence small J_H, cannot stabilize the HS state in a situation where the LDA bandwidth is appreciable. This point can be reliably accessed within LDA+DMFT but has not hitherto been addressed. To be internally consistent, one also requires that such a calculation also provide a quantitative description of one-particle spectra. We are unaware of earlier work addressing this issue either.

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 LDA+DMFT, incorporating one-electron band-structure aspects. Extant LDA+DMFT works give either a strongly renormalized FL-incoherent metal crossover[13] or an orbital selective, incoherent, pseudogapped metal.[18] 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 studies (XAS) are a reliable tool to study the *correlated* electronic structure of d- and f-band compounds.[19] PES experiments have been performed on Fe pnictides, including $SmO_{1-x}F_xFeAs$ (Ref. 20) with $x=0.15$. Very recently, XAS has also been performed for $SmO_{1-x}F_xFeAs$,[21] in addition to other pnictides. Together, they provide additional evidence of the *incoherent metal* normal state in Fe pnictides. The PES spectra show a kink at low energy, $\Omega=15$ meV,[20] 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 (2 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 SC itself.

II. MODEL AND SOLUTION

Starting with the high-T tetragonal structure with lattice parameters found in Ref. 22, one-electron band-structure calculations were performed for SmOFeAs using the linear muffin-tin orbital (LMTO) (Ref. 23) scheme in the atomic sphere approximation.[24] 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 LDA to DMFT, which enables direct access to the *correlated* spectral functions.[25] The one-electron part for SmOFeAs is $H_0=\Sigma_{\mathbf{k},a,\sigma}\epsilon_a(\mathbf{k})c^\dagger_{\mathbf{k},a,\sigma}c_{\mathbf{k},a,\sigma}$, 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) (Fig. 1) shows that *all* the five d orbitals participate in the bands crossing 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, structural pseudogap at E_F, the x^2-y^2, xz, yz bands have large DOS at E_F. In Fe pnictides, the d^6 configuration of Fe^{2+} dictates that the full, MO Coulomb interactions must be included. These constitute the interaction term, which reads

$$H_{\mathrm{int}} = U\sum_{i,a} n_{ia\uparrow}n_{ia\downarrow} + U'\sum_{i,a\neq b} n_{ia}n_{ib} - J_H\sum_{i,a,b}\mathbf{S}_{ia}\cdot\mathbf{S}_{ib}.$$

We choose parameters employed by Haule *et al.*:[13] $U=4.0$ eV, $U'=2.6$ eV, and $J_H=0.7$ eV, along with the five LDA bands, and solve $H=H_0+H_{\mathrm{int}}$ within LDA+DMFT. To solve the MO-DMFT equations, we use the MO iterated-perturbation theory (IPT) as an impurity solver. Although not quantitatively exact, it has many advantages. It is numerically very efficient, it is valid at $T=0$, in contrast to quantum Monte-Carlo (QMC) , and its self-energies $[\Sigma_a(\omega)]$ can be

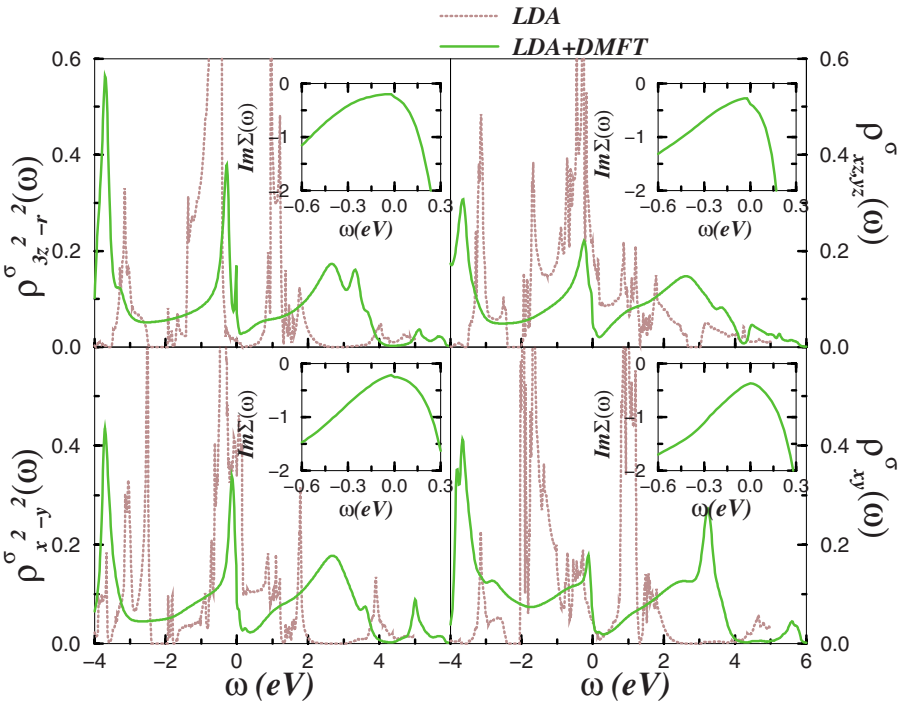

FIG. 1. (Color online) Orbital-resolved DOS: LDA and LDA+DMFT (U=4.0 eV, U'=2.6 eV, and total band filling, n=6.0) for SmOFeAs. The insets show the imaginary part of the corresponding self-energies, showing clear evidence of non-FL behavior.

extracted very easily. These are of particular importance in a complicated, MO situation that obtains in 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.[26] This requires strong U and 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', and 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 Fe pnictides as those above, does an OSMT occur[18] or does it not?[13] 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 large-scale spectral weight redistribution over large energy scales O(5 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,[18] this occurs even *without* a strict OSMT although almost all bands are very close to Mott localization. The orbital-resolved self-energies (inset of Fig. 1) clearly reveal this aspect: strong, interorbital electron-electron correlations produce large scattering between the "more localized" $d_{3z^2-r^2,x^2-y^2}$, the intermediately localized $d_{xz,yz}$, and the "itinerant" d_{xy} electronic states, manifesting large damping at E_F. Given the short quasiparticle lifetime, the FL quasiparticles are not well-defined elementary excitations any more. Instead, an incoherent, pseudogapped, bad-metallic state is realized. This incoherent state has also been found in very recent LDA+DMFT work,[13,18] and, as discussed there, is in qualitative agreement with experimental observations. Here, however, we also show that our results are in very good semiquantitative agreement with key features of the experimental PES and XAS spectra (see Fig. 2).

Analyzing the LDA+DMFT spectra of Fig. 1, we find that the $3z^2-r^2$, x^2-y^2 orbitals continue to be almost degenerate (they are 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, curiously, 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 twofold-degenerate sector. This is an interesting manifestation of the twofold $3z^2-r^2$, x^2-y^2 orbital degeneracy surviving in Fe pnictides and explicitly requires strong, MO correlations.

Using LDA+DMFT, we have also estimated the spin state on Fe sites. With parameters as those above, the quantity $\sqrt{\langle S^2_{z,\text{total}}\rangle} \simeq 1.8$, slightly less than the atomic value of S=2, and consistent with a HS state. Quite interestingly, very recent XAS measurements indeed imply a HS (S=2) state,[27] in excellent agreement with our result. We observe that the large J_H required for the HS state implies even larger Hubbard U and U' from d-shell quantum chemistry, in agreement with the parameter set used here. Here, we emphasize that the XAS work extracts the *atomic* spin moment and not the sublattice magnetization, M_s, measured in INS work. In contrast to the atomic spin state, which is a HS state for appreciable U, U', and J_H, the ordered moment is drastically reduced from this value by various factors: metallicity of

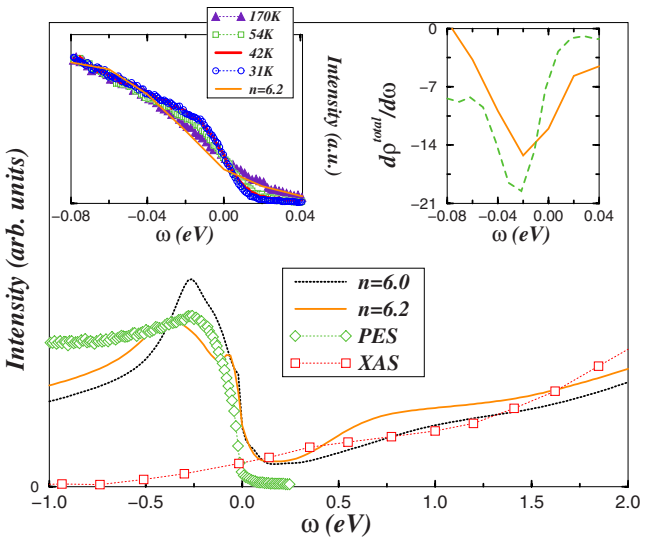

FIG. 2. (Color online) Comparison between theoretical (LDA+DMFT DOS) and experimental (PES and XAS) spectra in the *n*FL metallic phase of $SmO_{1-x}F_xFeAs$, $x=0.15$. Notice the good agreement with experimental PES result up to 0.5 eV binding energy. PES and XAS data are taken, respectively, from Refs. 20 and 21. The insets show the T dependence of the experimental angle-integrated PES (left) at low binding energies for $x=0.15$ and the derivative of the total DOS (for $n=6.2$) shows a sharp peak at $\Omega=20$ meV (solid: theory, dashed: experiment), indicating the kink structure in very good agreement with the experimental value of 15 meV below $T^{\star}$.

carriers, geometric frustration, etc. In Fe pnictides, both strong geometric frustration and quasi-itinerance are operative. The observed suppression of the sublattice magnetization in neutron scattering must therefore be attributed to strong geometric frustration in Fe pnictides, as discussed recently by several authors.[7,10] This is hitherto an open issue: should Fe pnictides be modeled using itinerant or localized $S=1$ or $S=2$ models?[10] Our analysis resolves this issue in favor of $S=2$ modeling. Our results are consistent with the view that the Fe pnictides fall into the *strongly correlated* class. This is because it is not possible to generate a HS state in a weakly correlated model, where U, U', and J_H are appreciably smaller than the LDA bandwidth. This is known from earlier analysis for the classic Mott system V_2O_3,[28] where the atomic spin value of $S=1$ *requires* strong correlations; in fact, observation of this feature in polarized XAS work[29] was one of the factors prompting the reexamination of this classic system. Thus, XAS for correlated materials may well serve as a strong check on the plausible parameter values for use in microscopic models; in our case, the observation of an atomic HS ($S=2$) state on Fe constrains theoretical models to employ large J_H, i.e., it implies that one should describe the Fe pnictides within MO Hubbard-type models with appreciable U, U', and J_H compared to the LDA bandwidths.

III. COMPARISON WITH EXPERIMENT

A LDA+DMFT-experiment comparison, carried out here for Fe pnictides, puts the above features into deeper perspective. In Fig. 2, we compare our results to recent PES and XAS results on $SmO_{1-x}F_xFeAs$ with $x=0.15$. Very good semiquantitative agreement with experiment is clearly visible. In particular, the broad peak at −0.3 eV in PES, as well as that at 0.5 eV in XAS, is in nice agreement with observations. We do not find, nor expect, good agreement with PES/XAS at high energies, where additional O, Sm, and As bands—neglected in the DMFT—will begin to contribute. The good quantitative agreement in a rather wide energy region around E_F, however, shows that only the Fe-d states are important in this range. Furthermore, the transfer of spectral weight from the 2 eV feature to that at 0.5 eV in XAS is also reproduced semiquantitatively in the correlated spectra. This is a characteristic evidence for relevance of strong, dynamical, electronic correlations and is inaccessible within LDA/LDA+U, as extensively documented.[25] Repeating the DMFT calculations with $U=5.0$ eV, as in Ref. 18, we found far worse agreement (not shown) 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 $LaO_{1-x}FeAsF_x$.[30]

Analyzing 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 $d\rho^{\text{total}}(\omega)/d\omega$ at an energy of 20 meV, implying a kink in the DOS at that energy, in excellent semiquantitative agreement with PES results. Furthermore, analyzing the orbital-resolved DOS, this feature is seen to originate from the $3z^2-r^2$, x^2-y^2 (DMFT) spectra. Since this structure is a consequence of near twofold $(3z^2-r^2, x^2-y^2)$ 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^{*} (Ref. 31) 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's functions enter the dynamical, second-order contributions [of the generic form $\int G_a^{(0)}(\omega-\omega_1-\omega_2)G_b^{(0)}(\omega_2)G_a^{(0)}(\omega-\omega_1)d\omega_1 d\omega_2=\int \chi_{ab}^{(0)}(\omega_1)G_a^{(0)}(\omega-\omega_1)d\omega_1$, with $\chi_{ab}^{(0)}(\omega)$ the interorbital 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. Thus, this low-energy kink feature has a clear collective *interorbital* character and will show up in the renormalized low-energy dispersion in the normal state of Fe pnictides. Recent ARPES work[32] indeed finds such a kink feature, exactly in the energy range (15–50 meV) found by our LDA+DMFT calculation. This is again consistent with the strong correlation picture where, in analogy with $SrVO_3$,[33] low-energy kinks characteristically arise in a strongly correlated system close to the Mott metal-insulator transition. Taking this together with the observation of a linear-in-T resistivity

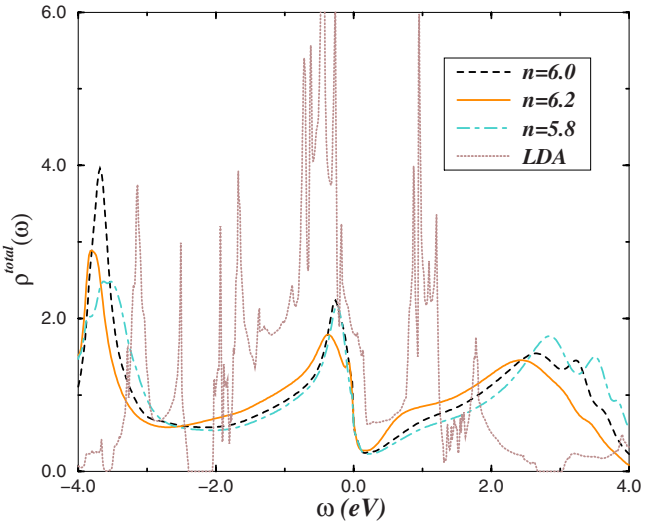

FIG. 3. (Color online) Total LDA+DMFT spectral functions for $SmO_{1-x}F_xFeAs$. Notice the large dynamical SWT upon electron and/or hole doping. LDA result for SmOFeAs is shown for comparison.

immediately above T_c for doped Fe pnictides bares the normal-state *incoherent* behavior, and, in view of the observation of the kinks, the relevance of strong, local correlations. Note that, in this regime ($x \geq 0.1$), SDW spin-wave-like fluctuations *cannot* be invoked to rationalize the kink. Coupling to coherent spin-wave-like excitations of the SDW state *cannot* generate the low-energy kink simply because SDW order has given way already around $x=0.1$. Furthermore, it would not account for the observation of the kink in the normal state above the SC transition in doped Fe pnictides since it is obvious that no SDW order exists in this regime. 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 may show this in future.

IV. COMPARISON WITH EARLIER WORK

Our finding of an incoherent metallic state is in agreement with earlier LDA+DMFT calculations.[13] However, no attempt has been hitherto made to compare these with actual experimental one-particle spectral functions, measured by PES (occupied part) and XAS (unoccupied part). A direct comparison between earlier results and experiment shows that neither of the earlier LDA+DMFT[13,18] results obtain the correct low-energy structures in PES, where a low energy kink ($O(15)$ meV) is clearly resolved. Neither is the XAS lineshape in accord with observations. In contrast, as discussed above, our results give a very good semiquantitative agreement with *both* these features. In addition, we are able to estimate the correct spin state on iron, something not attempted to date. Taken together, we argue that our results describe the basic aspects of the actual electronic structure of Fe pnictides in a quantitative way. We emphasize that the ability to make a quantitative comparison between theory and experiment is one of the main motivations of LDA+DMFT itself, and is of importance when a first-principles understanding of the nature of electronic states in correlated systems is attempted. It is in this light that our results (see below) should be considered as interesting.

The sources of the differences between our LDA+DMFT and earlier work are manifold: there are differences in the LDA DOS to begin with, and these will lead to changes in the correlated lineshapes upon using the LDA as an input in DMFT calculations. Second, the choice of the impurity solver used to solve the DMFT equations will cause some differences. We recall that use of quantum Monte Carlo (QMC) method to solve the impurity problem of DMFT precludes accessing temperatures below $T \simeq O(400)$ K for MO Hubbard-type models. Since the spectral functions within LDA+DMFT are known to be strongly T-dependent, it follows that there will be noticeable differences between MO-IPT (which can be used at low T) and QMC, which can only access high-T regimes. The strength of our approach is that, in addition to finding an incoherent metal at low T, we are also able to resolve the low-energy kinks in detail, and to interpret them in terms of a correlated, MO picture, as discussed above.

In an interesting work, Nakamura *et al.*[34] have carried out a detailed estimation of the orbital-dependent hoppings ($O(0.2–0.3)$ eV) and interaction parameters ($U=2.2–3.3$ eV) for an effective *five* band Hubbard-type model for Fe pnictides. First, we notice that $U \simeq 10t$ implies that Fe pnictides are in the intermediate-to-strong-coupling region of the MO Hubbard model. This *cannot* be interpreted as a weak-coupling scenario, and precludes the use of weak-coupling approaches, which are only valid for $U<t$. Even though we have not used orbital-dependent U,J_H in our LDA+DMFT, the effective interaction felt by electrons in a given orbital will be orbital dependent. This is because of the different orbital (one-particle) energies which already enter in the LDA input in our work. Also, the strongly orbital-dependent hoppings imply that individual d-bands will be anisotropically renormalized by correlations. This is apparent from our LDA+DMFT spectra, which, as discussed before, differ markedly from those computed in the LDA.

More recent LDA+DMFT work of Anisimov *et al.*[35] argues for the weakly correlated description of Fe pnictides. First, we note that it is in disagreement with Haule *et al.*, as well as Nakamura *et al.*, both of whom find $U \simeq W_{LDA}$, the LDA bandwidth. Second, the authors claim agreement with XAS data, while no explicit comparison is made. Looking more carefully at the occupied part of their DMFT spectra, one sees that the low-energy kinks observed in earlier PES work[20] as well as more recent ARPES work[32] are *not* reproduced in their result. Neither is the weakly correlated FL picture able to account for the quasilinear-in-T dependence of the dc resistivity ($\rho(T)$) in the $x \geq 0.1$ doped Fe pnictides: it would predict a quadratic-in-T dependence of $\rho(T)$, which is not what is observed.

V. DISCUSSION

Our LDA+DMFT results are consistent with the view[11,12] that Fe pnictides are strongly correlated, bad metals, lying

perhaps in the proximity to a Mott insulating state. What do our results imply for the nature of the SC in Fe pnictides? We restrict ourselves to a few qualitative remarks.

Since the kink in PES in our LDA+DMFT is related to the T-O distortion, rather than the SDW, it should vanish 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 $n=6$ (dashed line) and $n=6.2$ (solid line). 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.

In view of our finding of the HS ($S=2$) state on Fe in LDA+DMFT, the description of the SDW with $\mathbf{q}=(\pi,0)$ at low T for $x<0.1$ will involve geometric frustration (GF) effects, as postulated by several authors.[7,10,11] This will require extending the present analysis to include short-ranged spatial correlations into the DMFT. This ambitious enterprise is currently beyond numerical capabilities, and we leave its consideration for the future.

Based on this detailed theory-experiment agreement, we discuss the implications of our work on SC setting in beyond $x>0.1$ in Fe pnictides. First, in our LDA+DMFT results, *all* d bands cross E_F. Our finding of a low-energy kink having MO character in the DMFT DOS implies that such a low-energy kink should be observable in ARPES. This has actually been seen in the corresponding energy range $O(15)$ meV in a recent ARPES study.[32] These findings imply that SC in Fe pnictides should involve inducing the SC gap on all FS sheets, i.e., that it should be of the multiband type. This does not necessarily conflict with two-band Hubbard model results,[36] since the multiband SC proximity effect[37] could operate here. Once SC pairing occurs in the $d_{xz,yz}$ manifold, such an effect could induce secondary gaps over the remaining Fermi-surface (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 carrier number (given the proximity to a Mott insulator in DMFT) is consistent with a host of 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[38] up to optimal doping, putting the Fe pnictides into the strongly correlated, unconventional, HTSC category. However, we are unable to presently make any comment on the *symmetry* of the SC gap function: within LDA+DMFT, this involves more work, and is left for future consideration.

VI. CONCLUSION

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. We have argued that using a model with negligible correlations leads to discrepancies when confronted with experiment. As in cuprates, the normal state that becomes unstable to unconventional[2–4] SC is *not* a Fermi liquid. Our LDA+DMFT result gives an incoherent, non-Fermi-liquid state with small carrier density, as was observed.[4] Very good semiquantitative agreement with extant PES/XAS data, including finding of low-energy kinks in the LDA+DMFT spectra, lend strong credence to our view of Fe pnictides as multiorbital, strongly correlated materials close to the itinerant-localized boundary.

ACKNOWLEDGMENTS

L.C. thanks A. Ormeci for discussions. L.C. and H.R. thank the Emmy-Noether Program of the DFG (Grant No. RO-2460) for support. M.S.L. thanks the MPIPKS for financial support. S.L. acknowledges ZIH Dresden for computational time.

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