Dichotomy between Large Local and Small Ordered Magnetic Moments in Iron-Based Superconductors

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We study a four-band model for iron-based superconductors within the local density approximation combined with dynamical mean-field theory (LDA + DMFT). This successfully reproduces the results of models which take As p degrees of freedom explicitly into account and has several physical advantages over the standard five d-band model. Our findings reveal that the new superconductors are more strongly correlated than their single-particle properties suggest. Two-particle correlation functions unveil the dichotomy between local and ordered magnetic moments in these systems, calling for further experiments to better resolve the short time scale spin dynamics.

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In the recently discovered iron-based superconductors (iron SC) [1], the role of electronic correlation is still highly unclear. Strongly correlated materials are characterized by the presence of large local magnetic moments, which typically order if temperatures are sufficiently low. In the proximity of such magnetic phases, superconductivity is also often observed, with the close-by magnetic fluctuations usually cited as evidence for unconventional (not phonon-mediated) superconductivity. If the local magnetic moment is small and the system is metallic, weak-coupling theories like local spin-density approximation (LSDA) [2] can be applied. On the other hand, if the local magnetic moment is large and the exchange coupling between neighboring spins is the dominant interaction, not only the electronic states around the Fermi level but also the higher energy excitations (on the scale of the local Hubbard interaction U) are expected to play a role in the superconducting pairing mechanism. For instance, the latter is definitely the case for cuprates, which are Mott insulating in the absence of carrier doping. Iron pnictides are instead metallic and undergo a spin-density wave transition below $T \approx 150$ K. The characteristics of this transition are still under debate. Understanding the nature of the magnetic properties can therefore also help to clarify the origin of superconductivity in these materials.

Experimentally it has been clarified that the different members of the pnictide family have quite different ordered magnetic moments [3], ranging from $0.3\mu_B$ (or $0.6\mu_B$ [4]) in LaFeAsO to $2.2\mu_B$ in FeTe. The band structures of these compounds, however, do not show distinctive differences, and indeed, LSDA always yields an ordered moment of $\sim 2.0\mu_B$, for the experimental crystal structures [5,6]. Interestingly, while LSDA usually underestimates the size of the ordered moment, here the opposite happens. Even more important than the size of the ordered moment is the fact that in these systems the magnetic properties are extremely sensitive to the choice of the exchange-correlation functional or of the crystal structure [5]. In this situation, and also with the small magnetic moment indicating the proximity to a quantum-critical point, quantum fluctuations strongly influence the physics and the moments of iron SC. It is therefore crucial to treat dynamical quantum fluctuations beyond LSDA. These can indeed explain the presence of large local magnetic moments which form because of local Coulomb and exchange interaction but only give rise to a much smaller ordered moment at lower temperatures. There have been many attempts to go beyond LSDA taking electronic correlations more accurately into account. Among these, dynamical mean-field theory (DMFT) is one of the most promising, particularly when combined with *ab initio* band structure calculations [7]. However, the results of such local density approximation plus dynamical mean-field theory (LDA + DMFT) calculations for iron SC [8–14] strongly depend on which orbitals are included in the *ab initio* one-particle Hamiltonian and on the values of the interaction parameters used. As a consequence, DMFT calculations have been employed by different groups in fairly different ways, namely, to support that iron pnictides are strongly, intermediately, or weakly correlated, respectively. The majority of these studies focused on single-particle spectra and on the comparison with photoemission (PES) experiments, except for Refs. [13–15], where the spin susceptibility has also been calculated. In this Letter we focus on the dynamics of the local magnetic moment, which we argue is a key indicator for understanding the physics of iron SC. In particular, we conclude that in the single-particle spectral function correlation effects are hardly visible, while, at the same time, the spin-spin correlation function reveals the existence of a large local

magnetic moment. This turns out to be crucial for the explanation of some controversial experimental results in these systems.

For iron SC, two classes of models have been proposed [16,17]: One is a d-only model which takes the Fe $3d$ degrees of freedom into account, while the others are dp or dpp models considering pnictogen or chalcogen p and O $2p$ electrons explicitly. Here, we take the d model as a starting point. The d models considered hitherto, however, pose some physical and technical problems: Each Wannier function of d character has a fairly different spread in real space, due to the orbital-dependent hybridization between p and d [16]. Thus the interaction parameters strongly depend on the orbital [18,19]. In our coordinate system, in which x and y axes point to the pnictogen or chalcogen atom, the $3z^2$-r^2 orbital, e.g., has a small p-d hybridization. On the other hand, the x^2-y^2 orbital has long tails in the direction of the pnictogen or chalcogen. Such an orbital dependence causes problems when including electronic correlations: In order to avoid a double counting of correlation effects already considered within LDA, one would have to introduce an *ad hoc* orbital-dependent level shift in the many-body calculation. This shift turns out to be particularly important for the $3z^2$-r^2 orbital. In calculations based on the so-called fluctuation-exchange (FLEX) approximation [20], it has been shown that the $3z^2$-r^2 level becomes higher in energy and makes a large Fermi surface not present in LDA. A similar tendency is also seen in DMFT calculations [13], namely, the $3z^2$-r^2 occupancy gets dramatically smaller than in LDA depending on the strength of interaction parameters, contrary to what happens in LDA + DMFT calculations for dp and dpp models [10–12]. In order to overcome this problem, some authors added a constant part to the self-energy [20] or constrained the zero frequency value of the self-energy to get the appropriate orbital shift [21].

We take another route and assume the $3z^2$-r^2 to be fully occupied; therefore, we do not include it in our low-energy Hamiltonian for LaFeAsO [22]. This approximation is justified by the fact that the band with mainly $3z^2$-r^2 character lies below the Fermi level. We argue that the results of the four orbital model compare to the dpp model much better than the five-band one. It has been already shown [22] that the Fermi surface of the four-band model is almost exactly the same as that of the five-band model in LDA, and does not change so drastically even after the inclusion of many-body effects.

The four and the five d-band models have, of course, different values of the interaction parameters. Constrained random phase approximation calculations for the five-band model give an intraorbital Coulomb interaction U of about 2.2–3.3 eV and a Hund's rule coupling of about 0.3–0.6 eV [18]. The Hund's coupling J is quite robust as it is not screened. Therefore we take $J = 0.45$ eV, close to the average of [18]. For an appropriate choice of U, we have to consider (i) the screening effects of the $3z^2$-r^2 states and (ii) the slightly larger spread of the four-band Wannier functions which have to account for the $3z^2$-r^2 tails. Both effects reduce the value of U considerably. A further requirement is the agreement with the dpp model and thus with experiments. The best choice in this respect turned out to be $U = 1.8$ eV. Numerical limitations necessitate a Hund's exchange J of Ising type. Below, we will show, however, a comparison of results between Ising and full SU(2) symmetric interaction, for a model with fewer orbitals. The differences turn out not to be relevant for the present discussion.

In Fig. 1, we show the spectral functions of the four-band model for noninteracting electrons (dashed lines) and for $U = 1.8$ eV, $J = 0.45$ eV, and temperature $T = 460$ K ($\beta = 25$ eV^{-1}). Including U and J renormalizes (shrinks the width of) the minimum structure around the Fermi level. At the same time, some of the spectral weight is shifted to Hubbard-like shoulders at higher energies, in a way such that the overall bandwidth and spectrum remains close to the noninteracting one. The values of the quasiparticle weights are $Z \sim 0.51$, 0.45, and 0.60 for yz (xz), x^2-y^2, and xy bands, respectively, in agreement with PES [23,24]. As anticipated above, the results of the four orbital model resembles very closely the dp and the dpp models [11,12]. For instance, in our xy band the peak around $\sim$1 eV shifts towards lower energies, very similarly to the corresponding spectral function in Refs. [11,12] (there denoted as x^2-y^2 orbital as their coordinate system is rotated by 45°). On the other hand, our x^2-y^2 has a structure around $\sim$1 eV coming form the hybridization to the

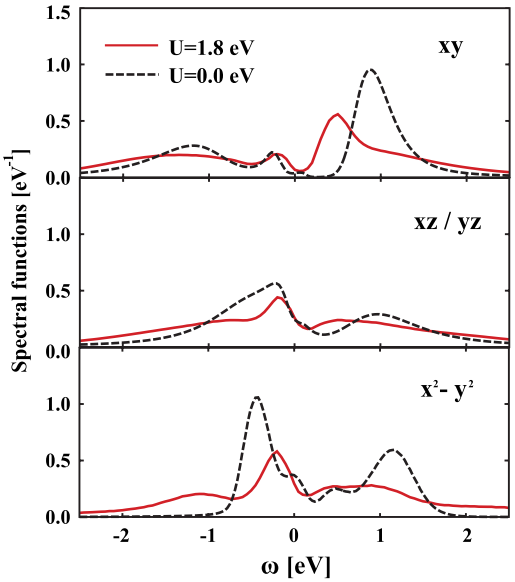

FIG. 1 (color online). Orbital-resolved spectral function of the four-band model for LaFeAsO at $T = 460$ K, for $U = 1.8$ eV, $J = 0.45$ eV (solid lines) and $U = J = 0$ (dashed lines). Including electronic correlations does not change the spectrum drastically.

$3z^2$-r^2 and the pnictogen or chalcogen p, which are included effectively in our four-band Hamiltonian. We can therefore conclude that the four-band model is reliable, with the clear advantage of (i) having a smaller number of parameters and (ii) yielding a set of d orbitals with much more similar spatial spread (and double-counting correction).

The picture arising from merely analyzing the spectral function hence suggests that iron SC are quite far from being standard strongly correlated materials, such as cuprates or other transition-metal compounds. On the other hand, calculations based on the FLEX approximation that one would expect to work for weakly correlated materials, here fail to reproduce the correct stripe pattern of antiferromagnetic spin fluctuations [22]. This indicates that iron SC cannot be categorized as weakly correlated systems, at least not with respect to their two-particle correlation functions. These considerations naturally lead to the question: Are iron SC more correlated than their single-particle quantities such as the PES spectra suggest? To answer this question we calculated the local spin susceptibility of LaFeAsO within LDA + DMFT and study whether or not this indicates the existence of a large local magnetic moment in these compounds.

In Fig. 2, we plot the (dynamical) local spin-spin correlation function $\chi_{lm}(\tau) \equiv \langle S_l^z(\tau) S_m^z(0)\rangle$ for (imaginary) time τ. Resolved are its intraorbital ($l = m$) and interorbital [$\sum_{l\neq m}\chi_{lm}(\tau)$] contribution. Similar to the case of one-particle properties, the U-driven intraorbital spin correlation is only slightly enhanced in comparison to the non-interacting value which for equal times ($\tau = 0$) is $0.5\mu_B^2$ [25]. In stark contrast, the interorbital contribution which vanishes without interaction is strongly enhanced. This

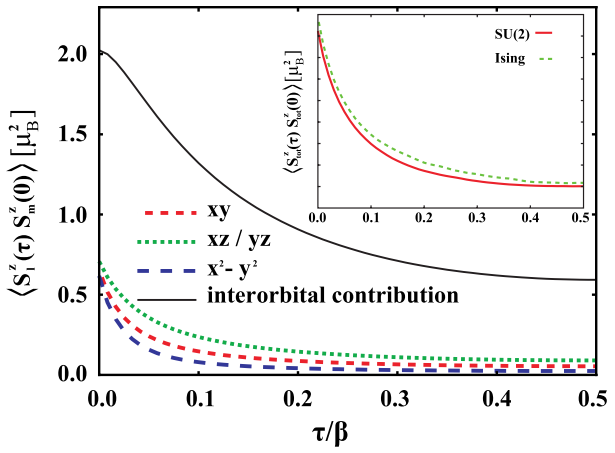

FIG. 2 (color online). Spin-spin correlation function for $J = 0.45$ eV and $\beta = 25$ eV^{-1}. We plot the different intraorbital contributions and the sum of all interorbital contributions. This orbital-resolved presentation clearly shows that the J-induced interorbital correlation is particularly large. Inset: Comparison between Ising- and SU(2)-symmetric Hund's exchange for a related two-band model, showing only quite small differences.

reflects the strong tendency of the system to align spins between different orbitals. It can be understood by noting that, since the crystal field splitting is small (~0.2 eV) [22], even an intermediate value of the Hund's rule exchange J is very effective. Hence, the large interorbital spin correlation function points to the important role J plays for inducing electronic correlations in iron SC.

In the inset of Fig. 2 we compare the total $\chi(\tau)$ for a half filled two-band Hubbard model with a semielliptic density of states with bandwidth 4 eV, $U = 2.5$ eV, $J = 0.5$ eV, and $\beta = 20$ eV^{-1}, with a very similar Z as in our four-band realistic calculation. As already mentioned, this allows us to exclude that the Ising approximation for the J term has a big influence in the relevant parameter regime. This is in line with Refs. [11,13], where a non-negligible difference between Ising and SU(2)-symmetric coupling is found only for values of the mass renormalization larger than in LaFeAsO.

The central result of our study is therefore that LDA + DMFT gives a strong enhancement of the total $\chi(\tau)$ (see Fig. 3) with respect to the noninteracting case. $\chi(\tau = 0)$ yields the bare local moment $m_{\mathrm{loc}} = \sqrt{\chi(0)}$, which corresponds to the responses on short time (or high energy) scales. The values of m_{loc} are quite large, i.e., 2.16 and $2.45\mu_B$ for $J = 0.45$ and 0.5 eV, respectively. Note that such variations of J are realistic when going from LaFeAsO to FeTe. While the $J = 0.45$ eV value of m_{loc} is only slightly T dependent ($m_{\mathrm{loc}} = 2.36\mu_B$ at $\beta = 10$ eV^{-1}), at $J = 0.5$ eV an even larger moment is formed at higher temperature ($m_{\mathrm{loc}} = 3.44\mu_B$ at $\beta = 10$ eV^{-1}).

While this local moment is large and indicates strong correlations, it is not the one that was hitherto measured experimentally. Experiments such as magnetic susceptibility measurements, nuclear magnetic resonance, Mössbauer spectroscopy, and muon relaxation are slow compared to the electronic dynamics on the femtosecond time scale. Hence, these experiments correspond to larger τ's or the

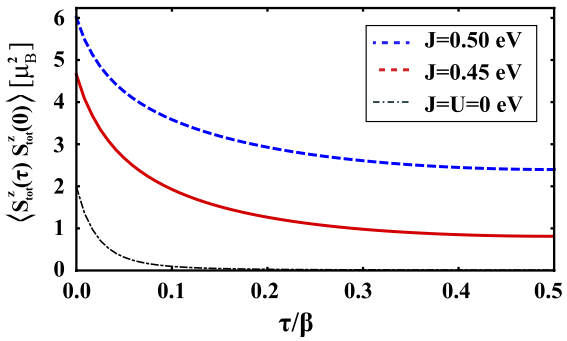

FIG. 3 (color online). Total spin-spin correlation function for LaFeAsO at two different values of J and $\beta = 25$ eV^{-1}, compared to the noninteracting $U = J = 0$ case. The short-time ($\tau = 0$) LDA + DMFT local moment for $J = 0.45$ eV is $m_{\mathrm{loc}} = 2.16\mu_B$, comparably large as in LSDA; whereas the long-time moment is screened to only $m \approx 0.7\mu_B$ at $T = 50$ K; see text.

integrated (static) susceptibility $\chi(\omega = 0) = \int_0^\beta d\tau \chi(\tau)$. A central result of our calculation is that this long-time susceptibility or a corresponding magnetic moment, which one can define through $\chi(\omega = 0) = m^2/T$, is strongly reduced (screened) compared to the instantaneous m_{loc}. Already at $\beta = 25\ \mathrm{eV}^{-1}$ ($\beta = 10\ \mathrm{eV}^{-1}$) the dynamic screening leads to strongly reduced moments of $m = 1.2$ (1.9) and 1.8 (3.4) for $J = 0.45$ and 0.5 eV, respectively. And for lower temperatures these values are much further reduced because of screening. At $T = 50$ K, i.e., in the temperature range where such magnetic moments were experimentally measured, an extrapolation of our data yields a crude (overestimated) approximation of $m \approx 0.7\mu_B$ [26,27]. Since the spin-density wave phase of iron SC is also very itinerant for both spin species, we expect that the dynamical screening identified here as the origin of the smallness of the (long-time) magnetic moment also survives (to a large extent) in the magnetic phase.

In conclusion, LDA + DMFT predicts that the local magnetic moment in iron SC is, in the paramagnetic phase, comparable to the ordered moment of LSDA. This moment is formed due to a local Hund's rule spin alignment. However, there is a dichotomy between this local magnetic moment and the dynamically screened moment, which is much smaller and beyond LSDA. Experiments performed hitherto measured the low-energy (or long-time) moment, i.e., the dynamically screened one. For measuring the (bare) local moment, experimental measurements on the time scale of femtoseconds are needed. A possibility to this end is to integrate neutron scattering measurements over $\mathbf{Q}$ and ω. For such an experiment, our calculations predict an intermediate-to-large value of the local magnetic moment. Similarly, x-ray spectroscopy is a very promising technique for measuring the size of the local magnetic moment, but so far this has been mainly used to estimate the strength of the interaction parameter, such as in Ref. [28]. Such experiments, if performed, will clarify whether our idea of iron SC being more strongly correlated than what is naively expected from PES experiments is correct. This can eventually settle the role electronic correlations play in the new class of iron SC.

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Note added.—Recently, we became aware of an x-ray study of LaFeAsO by Kroll *et al.* [29], where the authors find a high spin configuration in agreement with our conclusions.

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[25] This stems from the fact that if there is a spin σ in orbital l at time 0, looking at time $\tau = 0$ again it is still there.
[26] For $J = 0.45$ eV, we have extrapolated $\chi(\tau)$ to larger τ's via a polynominal χ^2 fit (fourth order in $1/\tau$) in the τ interval [2,6]. Supplementing the calculated $\chi(\tau)$ by this fit for $\tau = 6, \ldots, 1/(2T)$ yields the crude estimate quoted. Note, this overestimates m since it assumes $\chi(\tau)$ not to change upon lowering T, while it is (slightly) decreasing.
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