Dichotomy between large local and small ordered magnetic moment in Iron-based superconductors

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We study a four band model for iron-based superconductors within local density approximation + 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 the role of electronic correlation is still highly unclear. Strongly correlated materials are characterized by the presence of large local magnetic moments, which typically long-range order if temperatures are sufficiently low. In the proximity of such magnetic phases, also superconductivity is 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) 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 instead are metallic and undergo a spin-density wave (SDW) transition below \( T \approx 150 \text{K} \), whose characteristics 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, ranging from 0.3\( \mu_B \) (or 0.6\( \mu_B \)) in \( \text{LaFeAsO} \) (1111 compound) to 2.2\( \mu_B \) in \( \text{FeTe} \) (11 compound). The band structures of these compounds however do not show distinctive differences, and indeed, LSDA always yields an ordered moment of \( \approx 2.0 \mu_B \), for the experimental crystal structures. The failure of density functional theory (DFT) in capturing the correct ground-state properties of pnictides is hence a very important point which needs to be carefully analyzed in order to understand the physics of these compounds: While usually LSDA 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. 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-based superconductors. It is therefore natural to conclude that dynamical quantum fluctuations, not included in LSDA, are crucial for these systems. In particular they can 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 \textit{ab-initio} band structure calculations. However, the results of such LDA+DMFT calculations for iron-based superconductors strongly depend on which orbitals are included in the \textit{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 experiments, except for Refs. and where also the spin susceptibility has been calculated. In this paper we focus on the dynamics of the local magnetic moment, which we argue is a key indicator for understanding the physics of iron-based superconductors. 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-based superconductors, two classes of models have been proposed [13,18]: One is a $d$ only model which takes the Fe 3$d$ degrees of freedom into account, while the others are $dp$ or $dpp$ models considering pnictogen/chalcogen $p$ and O 2$p$ electrons explicitly. In this work, 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 $d$ [19]. Thus the interaction parameters strongly depend on the orbital [17,18]. In our coordinate system, in which $x$- and $y$-axes point to the pnictogen/chalcogen atom, the $3z^2-r^2$ orbital, for example, has a small $p$-$d$ hybridization, so that it is well localized. On the other hand, the $x^2-y^2$ orbital has long tails in the direction of the pnictogen or chalcogen sites. Such an orbital dependence causes problems when calculating the self-energy due to electron correlation: 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 level shift has been realized to be particularly important for the $3z^2-r^2$ orbital. In calculations based on the so-called FLEX approximation [19], 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 [10], or constrained the zero frequency value of the self-energy to get the appropriate orbital shift [20].

Here we take another route and assume the $3z^2-r^2$ to be fully occupied. We therefore do not include it in our low-energy Hamiltonian for LaFeAsO and are left with a model with merely four equally correlated orbitals [21], which allows us to circumvent an orbital dependent double-counting adjustment. 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 [21] 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 (RPA) calculations for the five band model give an intra-orbital Coulomb interaction $U$ of about $2.2-3.3$ eV and a Hund’s rule coupling of about $0.3-0.6$ eV [17]. In our case we have to consider the screening effect of $3z^2-r^2$ orbital as well as the slightly more extended tails of the Wannier functions which both reduce the value of $U$. For this reason, hereafter we use $U = 1.8$ eV. Numerical limitations necessitate a Hund’s exchange $J$ of Ising-type for four orbitals. 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 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.
shifts towards lower energies, very similarly to the corresponding spectral function in Ref. [12] and Ref. [11] (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 \(3z^2-r^2\) and the pnictogen/chalcogen \(p\), which are included effectively in our four band Hamiltonian. We can therefore conclude that the four band model reliably reproduces the results of photoemission experiments with the clear advantage of (i) having a smaller number of parameters and (ii) of 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-based superconductors are quite far from being standard strongly correlated materials, such as cuprates or other transition-metal compounds. On the other hand, calculations based on FLEX which one would expect to work for weakly correlated materials, here fail to reproduce the correct stripe pattern of antiferromagnetic spin fluctuations [21]. This indicates that iron-based superconductors cannot be categorized as weakly correlated systems, at least not with respect to their two-particle correlation functions.

These considerations naturally leads to the question: Are iron-based superconductors more correlated than their single particle quantities such as the photoemission 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 \(\chi_{lm}(\tau)\equiv \langle S_\tau^l S_{\mu\nu}^m(0)\rangle\) for (imaginary) time \(\tau\). Resolved are its intra-orbital \((l=m)\) and inter-orbital \(\sum_{l\neq m} \chi_{lm}(\tau)\) contribution. Similar to the case of one-particle properties, the \(U\)-driven intra-orbital spin correlation is only slightly enhanced in comparison to the non-interacting value which for equal times \((\tau=0)\) is \(0.5\mu_B^2\) [24]. In stark contrast, the inter-orbital contribution which vanishes without interaction is strongly enhanced. This 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 \((\sim 0.2\)eV) [21], even an intermediate value of the Hund’s rule exchange \(J\) is very effective. Hence, the large inter-orbital spin correlation function points to the importance \(J\) plays for inducing electronic correlations in iron-based superconductors.

Let us note in passing the inset of Fig. 2 which compares the total \(\chi(\tau)\) for a half-filled two-band Hubbard model with a semi-elliptic density of states with bandwidth 4eV, \(U = 2.5\)eV, \(J = 0.5\)eV and \(\beta = 20\)eV\(^{-1}\), a choice that gives renormalization factors very similar to our four-band realistic calculation. As already mentioned, the close agreement of the two curves allows us to exclude that the Ising approximation for the \(J\) term has a big influence in the relevant parameter regime.

Turning to the central result of our paper, the total \(\chi(\tau)\) in Fig. 3 we see a much larger susceptibility than in the non-interacting case. This enhancement of \(\chi(\tau)\) is a measure for the correlations in iron-based superconductors. At \(\tau = 0\), the susceptibility \(\chi(\tau)\) gives us a direct measure of the bare local moment \(m_{\text{loc}} = \sqrt{\chi(0)}\). This local moment corresponds to the responses on short time (or high energy) scales and is quite large, i.e., \(m_{\text{loc}} = 2.16\mu_B\) and \(2.45\mu_B\) for \(J = 0.45\)eV and 0.5eV, respectively. Note that such variations of \(J\) are realistic when

![Fig. 2: Spin-spin correlation function for \(J = 0.45\)eV and \(\beta = 25\)eV\(^{-1}\). We plot the different intra-orbital contributions and the sum of all inter-orbital contribution. This orbital-resolved presentation clearly shows that the \(J\)-induced inter-orbital 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.]

![Fig. 3: Total spin-spin correlation function for LaFeAsO at two different values of \(J\) and \(\beta = 25\)eV\(^{-1}\), compared to the non-interacting \(U = J = 0\) case. The short-time \((\tau = 0)\) LDA+DMFT local moment for \(J = 0.45\)eV is \(m_{\text{loc}} = 2.16\mu_B\), 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.]

### Footnotes

1. Ising model
2. SU(2) symmetry
3. Local moment
4. Hund’s rule
5. LDA+DMFT approximation

going from LaFeAsO to FeTe. While the \( J = 0.45 \text{ eV} \) value of \( m_\text{loc} \) is only slightly \( T \)-dependent (\( m_\text{loc} = 2.36 \mu_B \) at \( \beta = 10 \text{ eV}^{-1} \)), at \( J = 0.5 \text{ eV} \) an even larger moment is formed at higher temperature (\( m_\text{loc} = 3.44 \mu_B \) at \( \beta = 10 \text{ eV}^{-1} \)).

While this local moment is large and indicates strong correlations, it is not the one which 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 \( fs \) time scale. Hence, these experiments correspond to larger \( \tau \)'s or the integrated (static) susceptibility \( \chi(\omega = 0) = \int_0^\beta \text{d}\tau \chi(\tau) \). A central result of our calculation is that this long-time susceptibility or a corresponding magnetic moment, which one can define by resolving \( \chi(\omega = 0) = m^2 / T \) for \( m \), is strongly reduced (screened) compared to the instantaneous local magnetic moment \( m_\text{loc} \). Already at \( \beta = 25 \text{ eV}^{-1} (\beta = 10 \text{ eV}^{-1}) \) the dynamic screening leads to strongly reduced moments of \( m = 1.2(1.9) \) and \( 1.8(3.4) \) for \( J = 0.45 \text{ eV} \) and \( 0.5 \text{ eV} \), respectively. And for lower temperatures these values are much further reduced because of the screening. At \( T = 50 \text{ K} \), i.e., in the temperature range where such magnetic moments were experimentally measured, an extrapolation of our data yields a crude (overestimated) approximation of \( m \approx 0.7 \mu_B \) [23, 24]. Since also the SDW phase of iron-based superconductors is 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 survive (to a large extent) also in the magnetic phase.

In conclusion, LDA+DMFT predicts that the local magnetic moment in iron-based superconductor 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. The latter is actually much smaller in LDA+DMFT. 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 \( fs \) are needed. A possibility to this end is to integrate neutron scattering measurements over \( Q \) and \( \omega \). 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, like, e.g., in Ref. [27]. Such experiments, if performed, will clarify whether our idea of iron-based superconductors being more strongly correlated than what is naively expected from photoemission experiments, is correct. This can eventually settle the role electronic correlations play in the new class of iron-based superconductors.

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[24] This stems from the fact if there is a spin \( \sigma \) in orbital \( l \) at time 0, looking at time \( \tau = 0 \) again it is still there.
[25] For \( J = 0.45 \text{ eV} \), we have extrapolated \( \chi(\tau) \) to larger \( \tau \)'s via a polynomial \( \chi^2 \)-fit (4th order in \( 1/\tau \)) in the interval \( \tau = 10 \ldots 30 \). Supplemeting the calculated \( \chi(\tau) \) by this fit for \( \tau = 30 \ldots 1/\tau \) yields the crude estimate quoted. Note, this overestimated \( m \) since it assumes \( \chi(\tau) \) not to change upon lowering \( \tau \), while it is (slightly) decreasing.
[26] Note, frustration effects can further reduce the size of the ordered moment and may be relevant for the physics of these compounds. Indeed \( J_1-J_2 \) type of models have been proposed and are currently being extensively studied; see, e.g., T. Yildirim, Physica C 469 125 (2009); I. I. Mazin and J. Schmalian, Physica C 469, 614 (2009).
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