Fermi-liquid, non-Fermi-liquid, and Mott phases in iron pnictides and cuprates

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The role of Coulomb correlations in the iron pnictide LaFeAsO is studied by generalizing exact diagonalization dynamical mean field theory to five orbitals. For rotationally invariant Hund’s rule coupling a transition from a paramagnetic Fermi-liquid phase to a non-Fermi-liquid metallic phase exhibiting frozen moments is found at moderate Coulomb energies. For Ising-like exchange, this transition occurs at a considerably lower critical Coulomb energy. The correlation-induced scattering rate as a function of doping relative to half-filling, i.e., \( \delta = n/5 - 1 \), where \( n = 6 \) for the undoped material, is shown to be qualitatively similar to the one in the two-dimensional single-band Hubbard model which is commonly used to study strong correlations in high-\( T_c \) cuprates. In this scenario, the parent Mott insulator of LaFeAsO is the half-filled \( n = 5 \) limit, while the undoped \( n = 6 \) material corresponds to the critical doping region \( \delta_c \approx 0.2 \) in the cuprates, on the verge between the Fermi-liquid phase of the overdoped region and the non-Fermi-liquid pseudogap phase in the underdoped region.

PACS. 71.20.Be Transition metals and alloys - 71.27+a Strongly correlated electron systems

I. INTRODUCTION

The discovery of superconductivity in the iron-based pnictides\textsuperscript{1-4} has recently led to an intense debate on the question whether Coulomb correlations in these materials play a role of similar importance as in the high-\( T_c \) cuprates.\textsuperscript{5-40} Moreover, in contrast to the effective single-band character of the 3\( d_{z^2-r^2} \) states in the cuprates, the number of relevant \( d \) orbitals in the pnictides has also been a subject of discussion. In addition, because of the multi-band character of the Fe 3\( d \) bands, the interplay of Coulomb repulsion and Hund’s rule coupling should be of crucial importance for the pnictides. Finally, while it is generally accepted that the non-Fermi-liquid pseudogap properties in the underdoped regime of the cuprates are associated with the vicinity to the Mott insulating phase, in the pnictides it is not yet clear whether there exists a nearby Mott phase in the range of realistic Coulomb and exchange energies.

To study the effect of Coulomb correlations in iron pnictides, various groups\textsuperscript{5,16-18,20,21,25,32,33,37} have applied single-site dynamical mean field theory (DMFT). Depending on the details of the single-particle Hamiltonian and the magnitude of Coulomb and exchange energies, weakly or strongly correlated solutions were found. In the present work we extend exact diagonalization\textsuperscript{32,37} (ED) DMFT to five orbitals in order to investigate correlations effects in LaFeAsO as a function of Coulomb energy. The single-particle properties are described in terms of a five-band tight-binding Hamiltonian\textsuperscript{24} Each \( d \) orbital hybridizes with two bath levels, giving 15 levels in total. The five baths are coupled indirectly via the interorbital 3\( d \) Coulomb and exchange interactions. Thus, the Hilbert space is extremely large and finite-size effects are effectively reduced. The advantage of this multi-orbital ED/DMFT approach is that it is particularly useful at low temperatures and that it can handle large Coulomb energies and full Hund exchange. As will be discussed below, the latter feature is of special importance in the pnictides since Hund’s rule coupling leads to electronic properties that differ qualitatively from those obtained for the more approximate Ising-like exchange treatment. The sensitive role of exchange interactions in the pnictides was also noted in several previous papers.\textsuperscript{16,23,32}

The main result of this work is the identification of a paramagnetic Fermi-liquid to non-Fermi-liquid transition at moderate Coulomb energies, \( U_c \approx 3 \) eV (\( J = 0.75 \) eV), i.e., well below the overall width of the Fe 3\( d \) bands, \( W \approx 4.5 \) eV. This incoherent metallic phase extends up to rather large values of \( U > 6 \) eV if \( J \) is kept fixed at 0.75 eV). It is associated with the formation of local moments and with substantial low-frequency scattering rates in all 3\( d \) bands. Below this transition, all bands exhibit strong correlation-induced effective mass enhancement. These properties are intimately related to the multi-band nature of LaFeAsO and the Hund’s rule coupling among the Fe 3\( d \) subbands. A similar spin freezing transition had been found recently by Werner \textit{et al}\textsuperscript{44} in a fully degenerate three-band model. Within the present five-band system, a Mott phase is not obtained below \( U = 6 \) eV. Thus, correlation effects in LaFeAsO appear to be related to the formation of local moments within the non-Fermi-liquid phase, and not to the vicinity of a Mott insulating phase.

On the other hand, LaFeAsO readily turns into a Mott insulator at realistic Coulomb energies in the hypothetical limit of one-hole doping, i.e., \( n = 5 \). The 3\( d \) bands then become half-filled and split into lower and upper Hubbard bands. At intermediate hole doping, non-Fermi-liquid behavior dominates, while for electron doping (\( n > 6 \)) the system becomes a normal Fermi liquid. These results suggest a remarkable correspondence between the multi-band compound LaFeAsO and the two-dimensional single-band Hubbard model. Indeed, if the correlation-induced scattering rate is plotted as a func-
tion of doping relative to half-filling, i.e., \( \delta = n/5 - 1 \) (\( n = 6 \) for pure LaFeAsO), both systems exhibit the same sequence of phases for increasing \( \delta \): a Mott insulator at half-filling, a non-Fermi-liquid phase up to a critical doping of the order of \( \delta_c \approx 0.15 \ldots 0.20 \), and a weakly correlated Fermi liquid for \( \delta > \delta_c \). In this scenario, the paramagnetic phase diagrams of iron pnictides and cuprates are strikingly similar, with LaFeAsO (\( \delta = 0.2 \)) located slightly on the overdoped side above critical doping. As a result, the system exhibits a clear asymmetry with respect to doping. Whereas electron doping beyond \( n = 6 \) (\( \delta > 0.2 \)) reinforces Fermi-liquid properties, hole doping \( n < 6 \) (\( \delta < 0.2 \)) enhances bad-metallic behavior. Although the real material is undoubtedly more complex because of doping-dependent single-particle properties and the presence of antiferromagnetism\(^\text{43}\) at \( n = 6 \), we believe that the above picture nevertheless provides a useful new perspective for the role of correlation effects in iron pnictides in comparison with analogous physics in the cuprates.

If exchange interactions among Fe 3d electrons are approximated in terms of Ising-like exchange, i.e., by neglecting spin-flip and pair-exchange processes, the Fermi-liquid to non-Fermi-liquid transition still exists, but the critical value of \( U \) is shifted down to about 2 eV (assuming \( J = U/4 \)) and the transition is more abrupt. Similar qualitative changes from Hund to Ising exchange were found previously also for the Mott transition in two-band Hubbard models.\(^\text{45,46}\)

Because of the strong hybridization between Fe 3d and As 4p and O 2p states, there are indications that an accurate Wannier representation should encompass not only d but also p type basis functions, even if Coulomb correlations are explicitly only included among the 3d orbitals.\(^\text{18,19,21,25,30,32}\) A particular consequence of \( dp \) hybridization is that the effective 3d Coulomb interaction is considerably reduced. Moreover, Coulomb interactions among different d states are differently screened, giving rise to anisotropic intraorbital and interorbital matrix elements.\(^\text{18,19,21,30,32}\) The present approach is general in the sense that these choices only affect the single-particle Hamiltonian and not the evaluation of the d electron self-energy matrix. Nevertheless, in this initial five-orbital ED/DMFT study we use, for simplicity, a purely d electron tight-binding picture\(^\text{24}\) in order to elucidate the nature of the transition from Fermi-liquid to non-Fermi-liquid behavior. A more detailed investigation within a \( dp \) formulation is planned for future work. As in previous papers,\(^\text{5,10,18,20,21,25,32,33,37}\) we focus here on the paramagnetic phase.

The outline of this paper is as follows. In Section II we discuss several theoretical details concerning the single-particle properties of LaFeAsO and the ED/DMFT procedure that is used to evaluate the Fe 3d self-energy components. In Section III we analyze the results, with particular focus on the differences obtained for Hund and Ising exchange. Subsection A discusses the neutral system, whereas the effect of doping and the analogy between pnictides and cuprates are the subject of Subsection B. Section IV contains the summary.

II. MULTI-BAND ED/DMFT

In this section we briefly outline the theoretical details of the multi-band ED/DMFT approach used in this work. The focus is on the role of Coulomb correlations within the Fe 3d subbands of LaFeAsO. The single-particle properties are described in terms of the five-band tight-binding Hamiltonian \( H(\mathbf{k}) \) which was recently derived by Fraser et al.\(^\text{24}\) for a single plane of Fe atoms from an accurate fit to the density functional results by Xiao et al.\(^\text{47}\) The low-energy part of these bands are in excellent agreement with analogous calculations by Singh et al.\(^\text{48}\) The basis functions are \( d_{xz}, d_{yz}, d_{x^2−y^2}, d_{xy}, \) and \( d_{3z^2−r^2} \), where the \( x, y \) axes point along Fe nearest-neighbor directions. The first three of these orbitals comprise the \( t_{2g} \) subset, the remaining two orbitals represent the \( e_g \) subset. Hopping up to fifth nearest neighbors was included in the tight-binding fit. The onsite energies are \( E_{xz,yz} = 0.13 \text{ eV}, \ E_{x^2−y^2} = −0.22 \text{ eV}, E_{xy} = 0.3 \text{ eV}, \) and \( E_2 = −0.211 \text{ eV} \). Thus the \( d_{xz,yz,xy} \) levels lie \( 0.2 \ldots 0.4 \text{ eV} \) above the \( d_{x^2−y^2,3z^2−r^2} \) levels. The hopping parameters are given in the Appendix of Ref.\(^\text{24}\) and the one-electron band structure corresponding to this Hamiltonian is shown in Fig. 5 of this Reference.

The Fe 3d density of states components are shown in Fig. 1. For symmetry reasons, the \( d_{xz} \) and \( d_{yz} \) components are degenerate. The widths of the \( t_{2g} \) and \( e_g \) bands are approximately 3.0 eV and 4.0 eV, respectively. The total band width is about 4.5 eV. All Fe 3d bands exhibit a pronounced bonding-antibonding splitting, with a deep pseudogap at small positive energies, due to the hybridization with the neighboring LaAsO layers. In the absence of correlations, the occupancies of these bands are: \( n_{xz,yz} = 0.58, n_{x^2−y^2} = 0.53, n_{xy} = 0.52, \) and \( n_{3z^2−r^2} = 0.78 \). Note that these occupancies do not reflect the crystal field splitting among the onsite energies because of the complex shape of the density of states components. In particular, the bands of \( d_{xz} \) character are considerably more occupied than those of \( d_{x^2−y^2} \) character, despite the fact that \( E_{xz} > E_{x^2−y^2} \). A similar situation exists in the layer compound \( \text{Na}_2\text{CoO}_2 \), where \( n_{e_g} > n_{a_g} \) although \( E_{e_g} > E_{a_g} \).

Previously, we have used finite-temperature ED/DMFT to investigate correlation effects in \( t_{2g} \) three-band transition metal oxides, such as \( \text{Ca}_2\text{RuO}_4 \)\(^\text{39}\), \( \text{Na}_2\text{CoO}_2 \)\(^\text{49,51}\) \( \text{LaTiO}_3 \)\(^\text{52}\) and \( \text{V}_2\text{O}_5 \)\(^\text{53}\). It was shown that, in these systems, accurate projections of the lattice Green’s function onto a finite cluster consisting of three impurity levels and six bath levels (2 bath levels per \( t_{2g} \) orbital) can be achieved, yielding an overall cluster size \( n_s = 9 \). Since the different baths are indirectly coupled via the 3d interorbital Coulomb and exchange interactions, the spacing between excitation energies is rather small, so that finite-size effects are greatly
Spin-flip and pair-exchange terms are denoted explicitly $J$ which describes the modification of the single-particle $J$ neglected, i.e., $\rho_{im\sigma}$. In the case of isotropic Hund exchange, one has $\rho_{im\sigma} = \delta_{im}\rho_{m\sigma}$. Local Coulomb interactions preserve this symmetry, so that $G_{mn}$ and $\Sigma_{mn}$ are also diagonal. We denote these components by $G_m(\omega_n)$ and $\Sigma_m(\omega_n)$. We point out that, because of the non-diagonal nature of $H(k)$, each $G_m$ component is influenced by all components $\Sigma_m$.

For the purpose of the quantum impurity calculation within DMFT it is necessary to first remove the self-energy from the central site. This step yields the impurity Green’s function

$$G_{0,m}(\omega_n) = [G_m(\omega_n)^{-1} + \Sigma_m(\omega_n)]^{-1}.$$  

Within ED/DMFT the lattice impurity Green’s function $G_0$ is approximated in terms of an Anderson impurity model for a cluster consisting of impurity levels $\varepsilon_m = 0 \ldots 15$ and bath levels $\varepsilon_k = 6 \ldots 15$, which are coupled via hopping matrix elements $V_{mk}$. Thus, $G_{0,m}(\omega_n) \approx G_{0,m}^d(\omega_n)$, where

$$G_{0,m}^d(\omega_n) = \left(\omega_n + \mu - \varepsilon_m - \sum_{k=0}^{15} \frac{|V_{mk}|^2}{\omega_n - \varepsilon_k}\right)^{-1}.$$  

Since $G_{0,m}^d$ is diagonal in orbital indices, each impurity level couples to its own bath containing two levels: Orbital 1 couples to bath levels 6, 7, orbital 2 to bath levels 8, 9, etc. Each of the four independent functions $G_{0,m}^d(\omega_n)$ therefore involves 5 adjustable parameters: one impurity level, two bath levels, and two hopping elements. These parameters are found by using a standard minimization procedure. The quality of these fits using 5 parameters is very good, as shown in several previous works.

As a result of the ED quantum impurity calculation one obtains the finite temperature cluster Green’s function which is also diagonal: $G_{0,m}^d(\omega_n)$. In analogy to Eq. 3 the cluster self-energy is given by

$$\Sigma_m^d(\omega_n) = G_{0,m}^d(\omega_n)^{-1} - G_m^d(\omega_n)^{-1}.$$  

The important physical assumption within DMFT is now that this cluster self-energy provides an adequate representation of the self-energy of the extended solid, i.e., $\Sigma_m^d(\omega_n) \approx \Sigma_m(\omega_n)$, which is then used in Eq. 2 to derive the lattice Green’s function in the next iteration step. Further details can be found in Ref. 33.

Since the cluster Hamiltonian is extremely sparse (typically only 20 to 30 off-diagonal elements per row), the quantum impurity calculation is conveniently carried out...
by using the Arnoldi algorithm. The largest spin sector corresponds to \((n_u, n_d) = (7, 8)\), giving matrix dimension \((15!(7!)^2) = 6435^2\). To reduce storage requirements, we have rewritten our multi-orbital ED/DMFT code so that large basis vectors of size \(2^{2n_s}\) are avoided by keeping only vectors of size \(2^n_s\). Moreover, the Arnoldi scheme is readily parallelized. Thus, using 32 processors the largest Hamiltonian subblock requires less than 1 GB storage. Since the spacing between excited states is very small, at finite temperatures a large number of states may contribute to the cluster Green’s function. To reduce computational time in this first five-band ED study, we perform the DMFT calculation at \(T = 0.01\) eV, but retain only the lowest few states, making sure that ground state degeneracies are properly treated. Using 32 processors, one iteration then takes of the order of one to four hours.

### III. RESULTS AND DISCUSSION

#### A. Undoped LaFeAsO

An important consequence of local Coulomb interactions is the rearrangement of electrons among subbands. Fig. 2 shows the variation of the Fe 3d orbital occupancies with \(U\) at \(n = 6\) total occupancy. The precise values of \(U\) and \(J\) for LaFeAsO depend sensitively on the basis functions used for the 3d Hamiltonian. In the range \(U \leq 3\) eV we chose \(J = U/4\). For illustrative purposes we also show results for larger \(U\). In this range \(J\) is kept constant at 0.75 eV, in order to avoid unrealistically large Hund parameters. A charge flow from \(d_{xz}\) to \(d_{z^2-y^2}\) is seen to take place, thereby reducing the orbital polarization of the uncorrelated bands. The occupancies of the \(d_{xz},d_{y^2}\) orbitals are less strongly affected by correlations. The results shown are for full Hund coupling. Ising exchange yields a similar charge rearrangement predominantly between \(d_{xz}\) and \(d_{z^2-y^2}\) orbitals, with only slightly larger modifications of the \(d_{xz,y^2,xy}\) occupancies than seen in Fig. 2. Near \(U = 2.5\) eV, all 3d occupancies are within about 10% of the average occupancy 0.6.

![Graph showing orbital occupancies](image)

**Fig. 2**: (Color online) Fe 3d orbital occupancies (per spin) as functions of Coulomb interaction for \(n = 6\), with \(J = U/4\) for \(U \leq 3\) eV and \(J = 0.75\) eV for \(U > 3\) eV; Hund exchange.

The origin of the unusual reduced \(d_{xz,x^2-y^2}\) orbital polarization is the complex bonding-antibonding shape of the density of states which yields \(n_{z^2} > n_{x^2-y^2}\) although \(E_{z^2} > E_{x^2-y^2}\). A correlation induced reduction of orbital polarization is also found in Na\(_2\)CoO\(_2\) which exhibits a similar pseudogap in the density of states as a result of the strong \(3d-2p\) hybridization in the planar geometry.

To illustrate the effect of Coulomb correlations on the Fe 3d bands in more detail, we show in Fig. 3 the self-energy components as functions of Matsubara frequency for several Coulomb energies. (a) and (b): Hund coupling; (c) Ising exchange. The color coding is defined in panel (b).

![Graph showing self-energy components](image)

**Fig. 3**: (Color online) Imaginary part of self-energy components \(\Sigma_m(i\omega_n)\) as function of Matsubara frequency for several Coulomb energies. (a) and (b): Hund coupling; (c) Ising exchange. The color coding is defined in panel (b).

A correlation induced reduction of orbital polarization is also found in Na\(_2\)CoO\(_2\) which exhibits a similar pseudogap in the density of states as a result of the strong \(3d-2p\) hybridization in the planar geometry.
lifetime. Since this onset is much larger than what is expected due to finite temperature, it implies a breakdown of Fermi-liquid behavior. The loss of coherence is strongest for $d_{xy}$ and weakest for $d_{z^2}$. Ising exchange also gives rise non-Fermi-liquid behavior, except that the onset occurs at about $U = 2 \text{ eV}$, i.e., at considerably lower Coulomb energy than for Hund exchange.

Fig. 4 shows the orbital dependent quasi-particle weights $Z_m = 1/[1 - \text{Im} \Sigma_m(i\omega)/\omega|_{\omega \to 0}]$ as functions of Coulomb energy for Hund exchange. Despite the different orbital occupancies and different $t_{2g}$ and $e_g$ band widths, all five bands are seen to exhibit a similar reduction of $Z_m$ with increasing $U$. Ising exchange yields a slightly deeper decrease of $Z_m$ up to about $U = 2 \text{ eV}$, beyond which all self-energy components show a finite onset. The transition from coherent to incoherent metallic behavior should also manifest itself in the temperature variation of the Fe 3d self-energy. This has been studied recently for LaFeAsO$_{1-x}$F$_x$ at $x = 0.1$ within continuous-time QMC DMFT for full Hund exchange. The coherence temperature was shown to be a highly sensitive function of $J$, becoming extremely small for $J \approx 0.7 \text{ eV}$, which is close to the value assumed here ($J = 0.75 \text{ eV}$ for $U > 3 \text{ eV}$).

To explore the origin of the Fermi-liquid to non-Fermi-liquid transition we have evaluated the spin-spin correlation function $C_{mz}(\tau) = \langle S_m(\tau)S_{mz}(0) \rangle$, where $\tau$ denotes the imaginary time. Fig. 6 shows these orbital dependent functions for several Coulomb energies and Hund coupling. At low values of $U$, the $C_{mz}(\tau)$ decay to zero, as expected for a Fermi liquid. The orbital components of the spin susceptibility

$$\chi_m \sim \int_0^\beta d\tau \langle S_m(\tau)S_{mz}(0) \rangle \tag{6}$$

are then independent of temperature, indicating Pauli behavior. With increasing $U$, the decay becomes less rapid and finite values are approached at large $\tau$ (for $\tau \ll \beta = 1/T$), demonstrating the formation of local moments $S_m$ simultaneously in all subbands. The susceptibility components are then proportional to $\beta$, so that $\chi_m \sim S_m(S_m + 1)/T$, as expected for Curie-Weiss behavior. As shown in Fig. 7 for Ising coupling the formation of frozen moments sets in at much lower Coulomb energies.

A similar spin-freezing transition was recently found by Werner et al. for a fully degenerate three-band model near $n = 2$ occupancy. Using continuous-time QMC as impurity solver, the paramagnetic $U - n$ phase di-
agram was shown to exhibit Fermi-liquid properties at small $U$. For increasing $U$ and $n > 1.5$, an incoherent metallic phase with local moments appears, which is then replaced by a Mott insulating phases at integer occupancies $n = 2$ and $n = 3$. Beyond the critical value of $U$, the low-frequency limit of the self-energy exhibits a finite onset of similar magnitude as shown here in Fig. 5. The unoccupied states are also shifted closer to $E_F$, but there is no evidence of any upper Hubbard peaks. Note that the lower Hubbard bands are very broad. This is related to large uncorrelated band width and to the multiplet structure induced by Hund’s rule coupling.

In the present five-band study, we find in addition that this transition changes approximately from continuous to first-order when Hund exchange is replaced by Ising-like coupling.

To illustrate the correlation-induced transfer of spectral weight in LaFeAsO we show in Figs. 8(a) and (b) the 3$d$ spectral distributions for two Coulomb energies and Hund coupling. For simplicity we plot here the ED cluster spectra since they do not require analytic continuation to real frequencies. The main effect at $U = 1.75$ eV is the band narrowing both below and above $E_F$. In addition, spectral weight is shifted below the bottom of the 3$d$ bands, indicating the formation of weak lower Hubbard bands. At $U = 3$ eV, spectral weight in the occupied part of the 3$d$ bands is greatly reduced and the Hubbard bands are much more prominent. Since there is little experimental evidence for any significant lower Hubbard bands, the above results imply that $U$ within the present 5-band description should be approximately 2.0...2.5 eV, where the precise value depends on the magnitude of $J$. The unoccupied states are also shifted closer to $E_F$, but there is no evidence of any upper Hubbard peaks. The spectra for Ising exchange shown in panel (c) are qualitatively similar. Many small differences arise because of the absence of spin flip and pair hopping exchange processes. The different mutiplet structures associated with Hund and Ising coupling will be considered in more detail elsewhere.

The above results demonstrate the importance of a proper treatment of exchange interactions. For instance, if because of $pd$ screening realistic values of $U$ and $J$ for LaFeAsO are approximately 2.0...2.5 eV and $J \approx U/4$, respectively, full Hund coupling suggests that the system is moderately strongly correlated, with 3$d$ effective mass
enhancements of the order of $m^*/m = 1/Z_m = 2\ldots 3$.
In contrast, if spin-flip and pair-exchange processes are
gnored (as is usually done in Hirsch-Fye QMC calculations
to avoid sign problems, e.g., in Refs.\textsuperscript{20,21,23}), the same
coulomb and exchange parameters suggest that system
has crossed the boundary towards non-Fermi-liquid behavior,
with strongly reduced lifetimes of electronic states
close to $E_F$. It would be interesting to inquire whether
d_{p} formulation of $H(k)$ yields a similar qualitative difference
between Hund and Ising exchange.

At sufficiently large Coulomb energies, $n = 6$ integer
occupancy should eventually lead to a Mott insulating
phase. We have increased $U$ up to 6 eV while keeping $J = 0.75$ eV constant.
Both for Hund and Ising coupling, the
system evolves towards an orbital selective phase, where
the $d_{xz,yz,xy}$ subset is either in or close to an insulating
phase and the $d_{x^2-y^2,z^2}$ subset remains in the strongly
incoherent metallic state.\textsuperscript{55} Thus, for realistic Coulomb and exchange we conclude that in the present five-orbital
ED/DMFT description the system is far below the $n = 6$
Mott insulating region. Orbital selective phases in five-
band systems at $n = 6$ occupancy, with subbands split
via a crystal field, were also found in Refs.\textsuperscript{27,59}

We close this subsection by pointing out that several
papers have recently discussed the role of coexisting itiner-
ant and localized electrons in iron pnictides\textsuperscript{7,10,26-28,36}. A simple system which exhibits this kind of coexis-
tence is the half-filled two-band model where the narrow
band is Mott localized while the wide band is still metallic.\textsuperscript{46,60-62} Coulomb interactions between these two
types of electrons give rise to bad-metallic behavior in the
itinerant band, with a finite scattering rate and a pseudogap at $E_F$ in the case of Ising exchange\textsuperscript{60} and
marginal Fermi-liquid behavior in the case of full Hund
coupling.\textsuperscript{61,62} As we have seen above, the important fea-
ture of FeAsLaO at $n = 6$ occupancy is that the spin
freezing transition occurs simultaneously in all five $d$
bands, far below the Mott transition. Thus, in the range of
coulomb and exchange energies of interest, all sub-
bands have similar occupancies, effective masses, and
low-frequency scattering rates. The subbands therefore
do not split into itinerant and localized subsets. Only at
much larger $U$, the $d_{xz,yz,xy}$ and $d_{x^2-y^2,z^2}$ bands gradually tend towards 0.5 and 0.75 occupancy.

### B. Doped LaFeAsO: correspondence between
pnictides and cuprates

The results discussed above are for paramagnetic, un-
doped LaFeAsO, with Fe 3$d$ occupancy $n = 6$. To il-
lustrate the effect of electron and hole doping we show in
Fig. 9 the orbital occupancies as functions of chemical
potential at fixed $U = 2.5$ eV. In order to elucidate the doping variation of many-body properties, the
one-electron Hamiltonian is kept unchanged. The doping
range extends from one hole to one electron. (At half-
filling $n = 5$, the chemical potential is $\mu = 4.5U - 10J$
$= 5$ eV.) Evidently, the degree of orbital polarization de-
PENDs strongly on the total occupancy. For $n = 5$, all
$d$ bands become half-filled and some orbital components
Im $\Sigma_m(i\omega_n)$ are proportional to $1/\omega_n$. The spectral dis-
tributions reveal that the system then is a Mott insulator
where all $d$ bands are split into lower and upper Hubbard
peaks.

In the case of electron doping, all orbital occupancies
increase in a similar fashion. At $n = 7$, the self-energies
(not shown) indicate that the system is a weakly corre-
lated Fermi liquid. Even for Ising exchange, the qua-

dispersion weights are in the range $Z_m = 0.26\ldots 0.56$.
These properties differ strikingly from those discussed
above at $n = 6$, where under the same interaction condi-
tions the system is much closer to bad-metallic behavior.
Thus, the Fermi-liquid to non-Fermi-liquid phase bound-

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{spectral_distributions.png}
\caption{(Color online) Spectral distributions of Fe 3$d$
subbands for (a) $U = 1.75$ eV, (b) $U = 3$ eV; Hund coupling;
(c) $U = 2.5$ eV; Ising exchange; with $J = U/4$ and 0.05 eV
broadening.}
\end{figure}
liquid phase boundary. Smaller LaFeAsO lies just below the Fermi-liquid / non-Fermi-liquid phase boundary was also found to shift to larger $U$ as the occupancy moves farther away from half-filling.

On the basis of the above results we obtain the paramagnetic $U - n$ phase diagram shown in Fig. 10 where, for $U = 2.5$ eV and $J = U/4$ Hund exchange, undoped LaFeAsO lies just below the Fermi-liquid / non-Fermi-liquid phase boundary. Smaller $U$ and larger $J$ would move this point farther below this phase boundary. In the limit of one-hole doping, the system is a Mott insulator, whereas, for electron doping, Fermi-liquid properties dominate. For Ising exchange, the phase boundary is shifted to roughly 1 eV lower $U$ values, so that $U$ would have to be less than $\sim 2$ eV to preserve Fermi-liquid behavior.

Of course, in real LaFeAsO, this phase diagram should be more complicated because of the inevitable modification of the one-electron properties with doping and because of the antiferromagnetic phase observed at $n = 6$. Nevertheless, the paramagnetic limit permits to draw an interesting analogy between the present multiband iron pnictide and the single-band two-dimensional Hubbard model that is frequently used to investigate Coulomb correlations in high-$T_c$ cuprates.

Fig. 11(a) shows the orbital components of the low-frequency scattering rate $\gamma_m$ for LaFeAsO as functions of doping relative to half-filling, $\delta = n/5 - 1$ (Hund exchange, $J = U/4$). (b) Cluster components of scattering rate for single-band two-dimensional Hubbard model as functions of hole doping (nearest and next-nearest neighbor hopping $t = 0.25$ eV and $t' = -0.075$ eV, respectively, $U = 2.5$ eV, $T = 0.01$ eV.) The vertical bars denote the approximate doping concentrations of the Fermi-liquid to non-Fermi-liquid transition.

FIG. 9: (Color online) Fe 3$d$ orbital occupancies as functions of chemical potential for $U = 2.5$ eV with $J = 0.625$ eV and Hund coupling. The arrows indicate the values of $\mu$ associated with integer total occupancies.

FIG. 10: (Color online) Schematic phase diagram for doped LaFeAsO. Red curves: boundaries between Fermi-liquid (FL) and non-Fermi-liquid (NFL) phases for Hund and Ising exchange. At half-filling ($n = 5$) a Mott insulating phase exists down to rather small $U$. The undoped material at $n = 6$ with moderate $U \approx 2.5$ eV is a Fermi liquid for Hund coupling, but an incoherent metal for Ising exchange. The $n = 6$ Mott phase is located at $U > 6$ eV.

FIG. 11: (Color online) (a) Orbital-dependent scattering rates $\gamma_m$ for LaFeAsO as functions of doping relative to half-filling, $\delta = n/5 - 1$ (Hund exchange, $J = U/4$). (b) Cluster components of scattering rate for single-band two-dimensional Hubbard model as functions of hole doping (nearest and next-nearest neighbor hopping $t = 0.25$ eV and $t' = -0.075$ eV, respectively, $U = 2.5$ eV, $T = 0.01$ eV.)
ilar to the one found within cluster ED/DMFT for hole doping in the two-dimensional Hubbard model. Analogous results have been obtained by several groups. This model yields a Mott insulator at half-filling and exhibits a non-Fermi-liquid pseudogap phase up to a critical doping $\delta \approx 0.15 \ldots 0.2$. Ordinary metallic behavior is restored in the overdoped region, $\delta > \delta_c$. For electron doping the results are similar, except for a smaller critical doping which marks the onset of bad-metal behavior. In view of the analogy seen in Fig. 11, the Mott phase that is relevant for LaFeAsO is not the one that should eventually appear at $n = 6$ for large $U$, but the one that exists at realistic values of $U$ and $J$ at $n = 5$ occupancy.

The results discussed above demonstrate that the Mott transition in multi-orbital and multi-site Hubbard models is far more complex than the paramagnetic metal to insulator transition obtained in single-band, single-site DMFT calculations. The presence of interactions channels involving orbitals or sites not only affects the overall magnitude of the critical Coulomb energy, but gives rise to a much richer phase diagram. In particular, as we have seen here for LaFeASO, a frozen-moment, non-Fermi-liquid phase emerges between the Fermi-liquid and insulating regions. Conceptually, this non-Fermi-liquid behavior is closely related to the pseudogap phase in hole-doped cuprates which arises from short-range Coulomb correlations.

IV. SUMMARY

Multi-band ED/DMFT has been used to investigate the effect of correlations in the iron pnictide LaFeAsO. Starting from an accurate five-band tight-binding single-particle Hamiltonian, the many-body properties are evaluated by extending single-site ED/DMFT to five orbitals, each hybridizing with two bath levels. This scheme is particularly useful at very low temperatures, large Coulomb energies and for fully rotationally invariant Hund’s rule coupling. It is shown that correlation effects in LaFeAsO give rise to a paramagnetic transition from a Fermi-liquid phase to a non-Fermi-liquid phase at a critical Coulomb energy ($pd$ screened) of about 3 eV. This transition appears to be continuous and is caused by the formation of Fe 3d local moments. Below this transition, the quasi-particle weight of all d orbitals is strongly reduced and orbital polarization is less pronounced than in the uncorrelated density functional band structure. A Mott insulating phase does not appear below $U = 6$ eV. For Ising exchange the Fermi-liquid to non-Fermi-liquid transition also exists, but the critical value of $U$ is shifted to about 2 eV. Moreover, the transition appears to be first order rather than continuous.

Despite the multi-band nature of LaFeAsO and the important role of exchange interactions, this system exhibits an interesting relationship to the single-band two-dimensional Hubbard model if the doping concentration is defined with respect to the half-filled 3d shell. Within this picture, iron pnictides and cuprates exhibit the same sequence of paramagnetic phases with increasing doping: from Mott insulator to bad metal to Fermi liquid. Thus, fluctuations between orbitals or lattice sites lead to similar physics. According to the available experimental evidence, LaFeAsO is located on the weakly to moderately correlated side of the Fermi-liquid to non-Fermi-liquid phase boundary. Thus, iron pnictide materials appear to have two parent compounds: the anti-ferromagnetic semi-metal at $n = 6$ and the Mott insulator at $n = 5$.

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