Possible origin of the reduced ordered moment in iron pnictides: a Dynamical Mean Field Theory study

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We investigate the phase diagram of a two-band frustrated Hubbard model in the framework of dynamical mean field theory. While a first-order phase transition occurs from a paramagnetic (PM) metal to an antiferromagnetic (AF) insulator when both bands are equally frustrated, an intermediate AF metallic phase appears in each band at different $U_c$ values if only one of the two bands is frustrated, resulting in continuous orbital-selective phase transitions from PM metal to AF metal and AF metal to AF insulator, regardless of the strength of the Ising Hund’s coupling. We argue that our minimal model calculations capture the frustration behavior in the undoped iron-pnictide superconductors as well as local quantum fluctuation effects and that the intermediate phases observed in our results are possibly related to the puzzling AF metallic state with small staggered magnetization observed in these systems as well as to the pseudogap features observed in optical experiments.

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The subtle interplay among magnetism, superconductivity, multiorbital effects and structure is a major subject of debate in the recently discovered iron pnictide superconductors \cite{1}. While, similar to high-$T_c$ cuprate superconductors, magnetically mediated pairing was proposed to dominate the superconducting state \cite{2}, the nature of magnetism in undoped iron pnictides is still unclear \cite{3}. The experimentally observed iron ordered moment in the antiferromagnetic phase is too small, compared to that obtained from density functional theory (DFT) calculations. Various DFT studies have shown that this value is strongly dependent on the details of the calculations and on the lattice structure \cite{4,3}. Very recently, a local density approximation (LDA)+U calculation explained the small magnetic moment in terms of large magnetic multipoles without analyzing the nature of the phase transition \cite{4}. A few alternative proposals are based on a localized picture where a frustrated one-band Heisenberg model is considered \cite{6,9}. However, the multi-band and itinerant nature of iron pnictides are overlooked in such approaches. Furthermore, existing dynamical mean field theory (DMFT) and LDA + DMFT studies \cite{10,14} for iron pnictides were performed in the paramagnetic state and did not consider the magnetic ordering. Therefore, a proper microscopic theory for the magnetism in iron pnictides is still missing.

Analysis of recent Fe 3$d$ transfer integrals obtained from downfolding of the bandstructure of a few iron-based superconductors \cite{16} always shows the existence of weakly frustrated (like $d_{xy}$) and highly frustrated (like $d_{yz}/d_{zx}$) orbitals for all cases due to the hopping mediated by a pnictogen or chalcogen ion. Such behavior suggests that a minimal model for exploring the role of frustration on the magnetism of the iron pnictides should be a two-band model with one unfrustrated and one frustrated band. The question to be posed is whether an AF metallic state with small ordered magnetic moment can emerge out of the interplay between frustrated and unfrustrated bands.

In order to investigate this issue, we consider in the present work a two-band half-filled Hubbard model with different degrees of band frustration. We will demonstrate that while the AF metallic state is absent when both bands are equally frustrated, an AF metallic state with small magnetization is present when the frustration in one of the bands is turned off. Moreover, we identify a pseudogap region and show that it originates from the small AF moment which is due to the interplay between frustrated and unfrustrated bands.

The Hamiltonian we study is

$$
H = - \sum_{\langle ij \rangle \sigma} t_{ij} c_{i \sigma}^\dagger c_{j \sigma} - \sum_{\langle ij \rangle \sigma} t'_{ij} c_{i \sigma}^\dagger c_{j' \sigma} + U \sum_{im} n_{i \sigma} n_{i \bar{\sigma}} + \sum_{im \sigma \bar{\sigma}} (U' - \delta_{\sigma \bar{\sigma}} J_z) n_{i \sigma} n_{i \bar{\sigma}},
$$

where $c_{i \sigma} (c_{i \sigma}^\dagger)$ is the annihilation (creation) operator of an electron with spin $\sigma$ at site $i$ and band $m$. $t_{ij}$ ($t'_{ij}$) is the hopping matrix element between site $i$ and nearest-neighbor (NN) site $j$ (next nearest-neighbor (NNN) site $j'$). $t'_m = 0$ for the unfrustrated band. For simplification, we neglect inter-band hybridizations. $U$ and $U'$ are, respectively, intra-band and inter-band Coulomb interaction integrals and $J_z n_{i \sigma} n_{i \bar{\sigma}}$ is the Ising-type Hund’s coupling term. In our calculations we set $U' = \frac{U}{2}$ and $J_z = \frac{U}{4}$ and ignore the spin-flip and pair-hopping processes. For the solution of this model we employ DMFT \cite{17} which includes the local quantum fluctuation effects and we perform the calculations on the Bethe lattice. The DMFT self-consistency equations with inclu-
section of the Néel state are given as \[ G^{-1}_{0, A, \sigma} = i\omega_n + \mu - t_m^2 G_{B, \sigma} - t_m^2 G_{A, \sigma}, \] \[ G^{-1}_{0, B, \sigma} = i\omega_n + \mu - t_m^2 G_{A, \sigma} - t_m^2 G_{B, \sigma}, \] where \( \mu \) is the chemical potential, \( \omega_n \) is the Matsubara frequency and magnetizations of \( A \) and \( B \) sublattices are in opposite directions. As impurity solver, a weak-coupling continuous-time quantum Monte Carlo algorithm was employed \[ 18 \text{ - 19}. \]

We first consider the one-band Hubbard model with magnetic frustration in both bands at half-filling. Previous DMFT calculations done on the frustrated one-band Hubbard model \[ 15 \] with frustration strength \( t'/t = 0.58 \), showed the existence of a first-order phase transition from paramagnetic (PM) metal to AF insulator. For comparison with this one-band case, we set in our two-band model \( t_m = 1 \) and \( t'_m = 0.58 \) for \( m = 1, 2 \). The bandwidth \( W = 4.624 \) is determined as \( W = 4\sqrt{t^2 + t'^2} \).

In Fig. 1 we present the results for the staggered magnetization \( m_s \) as a function of \( U/t \) for \( T/t = 1/16 \) and \( T/t = 1/32 \) (Fig. 1(a)) and the density of states (DOS) at \( U/t = 2.2 \) and \( U/t = 2.6 \) for \( T/t = 1/32 \) (Fig. 1(b)). Below \( U/t = 2.4 \) in Fig. 1(a) the staggered magnetization for both temperatures is negligibly small, indicating a PM state. As the interaction \( U/t \) is increased, for both temperatures a jump is detected around the critical value of \( U_c/t = 2.4 \) and the system goes into an AF state. The discontinuous behavior suggests a first-order phase transition. These results are very similar to those obtained for the one-band Hubbard model with frustration \[ 15 \]. In order to analyze the metal-insulator transition, we present in Fig. 2(b) the DOS close to the critical \( U_c/t \) where we employed the maximum entropy method for analytic continuation. In the PM state at \( U/t = 2.2 \), the observed finite DOS at the Fermi level \( (\omega = 0) \) indicates a metal. In the AF state at \( U/t = 2.6 \), the spin-up and spin-down DOS on the same sublattice become unequal and the spin-up (spin-down) DOS on sublattice \( A \) and the spin-down (spin-up) DOS on \( B \) are pairwise equal due to the development of the AF moments. Due to the Coulomb interaction strength \( U/t \) and the appearance of AF ordering, the system shows insulating behavior with opening of a small gap at the Fermi level \( (\omega = 0) \). Comparing the results of the magnetically frustrated one-band Hubbard model with the two-band model where an orbital degree of freedom is involved, we find that the phase diagrams of both models are qualitatively the same. Such a model cannot reproduce the magnetic behavior of the Fe pnictides and also should not be relevant for the phase diagram of the Mott insulator \( V_2O_3 \) \[ 15 \text{ - 21} \].

Now let us consider the two-band system in which frustration is turned off for one of the bands. This model should mimic the observed behavior in downfolding calculations \[ 16 \] for the Fe pnictides. We set \( t_1 = 1 \) and \( t'_1 = 0 \) for the unfrustrated band and \( t_2 = 1 \) and \( t'_2 = 0.65 \) for the frustrated one. The bandwidths for unfrustrated and frustrated bands are \( W_1 = 4.0 \) and \( W_2 = 4.77 \), respectively. In Fig. 2(a) we show the behavior of the staggered magnetization \( m_s \) as a function of tempera-
find a smooth increase of the magnetization with interaction strength. We show the staggered magnetization as a function of interger magnetization increases more rapidly in the unfrustrated band, and the small staggered magnetization is not sufficient for opening a full gap; this is what we investigate next.

To analyze the metal-to-insulator transition, we present in Fig. 3 the spin-up DOS on the A site for four representative values of $U/t$ at a fixed temperature of $T/t = 1/32$. The PM metallic state in both bands is present in the unfrustrated band ($t$ band). At this value of $U/t$, the small moment only opens a pseudogap. When we increase the interaction, an orbital selective metal-to-insulator transition occurs, and at $U/t = 2.2$ (see Fig. 3 (c)), an AF metal in the frustrated band coexists with an AF insulator in the unfrustrated band. Finally, in the strong-coupling region at $U/t = 2.8$ (see Fig. 3 (d)), both bands are in AF insulating states.

In Fig. 4 we plot the phase diagram $T/t$ versus $U/t$ for the Hamiltonian (1). The PM metal, AF metal and AF insulator phases are present in both bands, but the critical values $U_c/t$ of the unfrustrated band are smaller than those of the frustrated one. The Néel temperature increases as a function of $U/t$. The intermediate AF metals show pseudogap behavior in the DOS as a precursor of gap opening (see e.g. Fig. 3 (b) for the unfrustrated band) due to the continuous phase transitions induced by a continuous change of magnetization (see Fig. 2). This is in contrast to a first-order Mott transition dominated by strong correlations where an abrupt gap opening is observed. The pseudogap features obtained here could account for the experimentally observed optical conductivity behavior of the new Fe-based superconductors [21].

We visualize the mechanism of the appearance of an AF metallic phase with small antiferromagnetic ordered moment in Fig. 5. Without coupling between frustrated and unfrustrated bands, the ground state of the unfrustrated band on a square lattice shows AF insulating behavior with high magnetic ordered moment as soon as the interaction $U > 0$ due to perfect nesting while that of the frustrated band exhibits nonmagnetic metallic behavior below a critical interaction $U_c/t$ as frustration prevents perfect nesting (see Fig. 5 (a)). As the Hund's rule
the pseudogap behavior in the AF metal state is closely
between frustrated and unfrustrated bands. Furthermore,
antiferromagnetic phase may result from an interplay be-
ordered magnetic moments observed in the stripe-type
of the new iron-based superconductors where the small
new phases may be directly relevant for the magnetism
while the unfrustrated one is still an AF metal. These
PM metal, or the frustrated band is already AF insulat-
bond is an AF metal while the unfrustrated one is still a
AF metal to an AF insulator. This leads to new phases
first from a PM metal to an AF metal and then from an
selective continuous phase transitions occur in both bands
order phase transition separating a PM metal from an
phases (see Fig. 4).

density of states, explaining the additional AF metallic
Such an interplay between frustrated and unfrustrated
ant band and become more delocalized (see Fig. 5 (b)).
they follow the spin arrangement in the frustrated itiner-
the Hund’s coupling to the frustrated band. Therefore,
AF unfrustrated band are affected by frustration due to
Hund’s rule. On the other hand, the spins in the
AF unfrustrated band are affected by frustration due to
the Hunad’s coupling to the frustrated band. Therefore,
they follow the spin arrangement in the frustrated itiner-
band and become more delocalized (see Fig. 5 (b)).
Such an interplay between frustrated and unfrustrated
bands results in a reduction of the antiferromagnetic or-
dered moments and therefore of the gap amplitude in the
density of states, explaining the additional AF metallic
phases (see Fig. 4).

In summary, we have studied the frustrated two-band
Hubbard model at half-filling and have shown that a first-
order phase transition separating a PM metal from an
AF insulator occurs if both bands are equally frustrated.
On the other hand, by considering one band frustrated
and turning off frustration in the second band, orbital se-
selective continuous phase transitions occur in both bands
first from a PM metal to an AF metal and then from an
AF metal to an AF insulator. This leads to new phases
where either both bands are AF metals, or the frustrated
band is an AF metal while the unfrustrated one is still a
PM metal, or the frustrated band is already AF insulat-
ing while the unfrustrated one is still an AF metal. These
new phases may be directly relevant for the magnetism
of the new iron-based superconductors where the small
ordered magnetic moments observed in the stripe-type
antiferromagnetic phase may result from an interplay be-
tween frustrated and unfrustrated bands. Furthermore,
the pseudogap behavior in the AF metal state is closely
related to the optical conductivity features of iron-based
superconductors [21].

The new phases involving AF metallic states appear in
a wide range of interaction parameters, indicating that
our model can be applied to a large family of iron-based
superconductors with different interaction strengths. In
the present work we showed the case of one unfrustrated
band coupled with one frustrated band with $t_2/t_1 = 0.65$
buts we have checked a few more cases at $T/t = 1/16$
by tuning to stronger frustrations in the frustrated band
($t_2/t_2 = 0.8$) or even by changing the unfrustrated band
to be weakly frustrated ($t_1/t_2 = 0.2$). In both cases we
find solutions of AF metals, underlining the relevance of
the investigated model for the new iron-based supercon-
ductors. Furthermore, we have checked that AF metallic
states also exist at both $J_z = U/8$ and $J_z = 7U/24$ with
the constraint of $U = U’ + 2J_z$ in addition to the value of
$J_z = U/4$ we present in this work. Our model calcula-
tions show that it is the coupling of strongly frustrated
with weakly frustrated bands which induces a reduced
antiferromagnetic ordered moment, and this should be
applicable to many (more than two) bands with different
degrees of frustration as is the case in the iron pnictides.

While we believe that our model calculations qualita-
tively capture the central physics of AF metal with
small ordered magnetic moment observed experimentally
in undoped iron-based superconductors as well as the na-
ture of the phase transitions, further investigations have
to be done by including all five Fe 3d orbitals with re-
alistic inter-band, intra-band hybridizations and various
fillings on the frustrated square lattice in order to allow
for quantitative comparisons between experiments and
theory.

Our results on the model with two equally frustrated
bands also show that this model is insufficient for ex-
plaining the physics of $V_2O_3$ contrary to previous sug-
gestions [18], and inclusion of other degrees of freedom
like phonons may be necessary.

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