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# Possible origin of the reduced ordered magnetic 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.[1] While, similar to high-$T_c$ cuprate superconductors, magnetically mediated pairing was proposed to dominate the superconducting state,[2] the nature of magnetism in undoped iron pnictides is still unclear.[3] The experimentally observed iron ordered moment in the antiferromagnetic (AF) 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.[3,4] 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.[5] A few alternative proposals are based on a localized picture where a frustrated one-band Heisenberg model is considered.[6–9] However, the multiband and itinerant nature of iron pnictides are overlooked in such approaches. Furthermore, existing dynamical mean-field theory (DMFT) and LDA+DMFT studies[10–14] for iron pnictides were performed in the paramagnetic (PM) 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 the band structure of a few iron-based superconductors[15] always shows the existence of weakly frustrated (such as $d_{xy}$) and highly frustrated (such as $d_{yz}/d_{zx}$) 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

$$H = -\sum_{\langle ij\rangle m\sigma} t_m c^{\dagger}_{im\sigma} c_{jm\sigma} - \sum_{\langle ij'\rangle m\sigma} t'_m c^{\dagger}_{im\sigma} c_{j'm\sigma} + U\sum_{im} n_{im\uparrow} n_{im\downarrow} + \sum_{i\sigma\sigma'} (U' - \delta_{\sigma\sigma'} J_z) n_{i1\sigma} n_{i2\sigma'}, \quad (1)$$

where $c_{im\sigma}(c^{\dagger}_{im\sigma})$ is the annihilation (creation) operator of an electron with spin $\sigma$ at site $i$ and band $m$. $t_m$ ($t'_m$) is the hopping matrix element between site $i$ and nearest-neighbor site $j$ (next-nearest-neighbor site $j'$). $t'_m=0$ for the unfrustrated band. For simplification, we neglect interband hybridizations. $U$ and $U'$ are, respectively, intraband and interband Coulomb-interaction integrals and $J_z n_{i1\sigma} n_{i2\sigma}$ 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 (Ref. 16) which includes the local quantum fluctuation effects and we perform the calculations on the Bethe lattice. The DMFT self-consistency equations with inclusion of the Néel state are given as[16,17]

$$G^{-1}_{0,A,\sigma} = i\omega_n + \mu - t_m^2 G_{B,\sigma} - t_m'^2 G_{A,\sigma}, \quad (2)$$

$$G^{-1}_{0,B,\sigma} = i\omega_n + \mu - t_m^2 G_{A,\sigma} - t_m'^2 G_{B,\sigma}, \quad (3)$$

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.[18,19]

We first consider the two-band Hubbard model with magnetic frustration in both bands at half filling. Previous DMFT

 

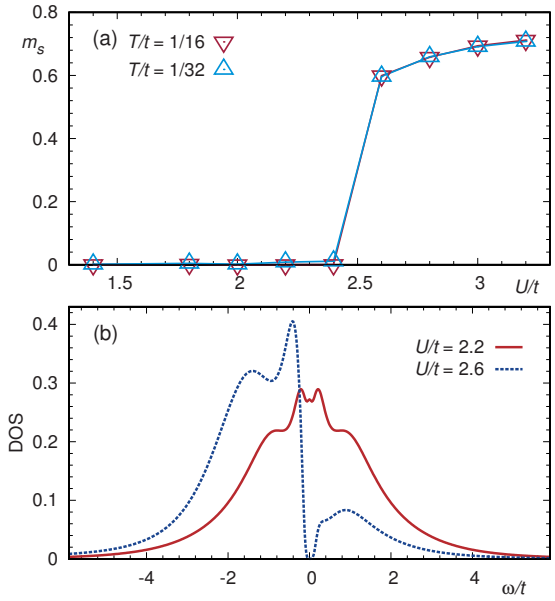


FIG. 1. (Color online) (a) Staggered magnetization $m_s$ as a function of $U/t$ for $T/t=1/16$ and 1/32. A first-order PM to AF transition, recognizable from a jump of $m_s$, is present at the critical interaction strength $U_c/t=2.4$. (b) DOS for the spin-up species on an A lattice site as a function of frequency for $U/t=2.2$ and 2.6 and $T/t=1/32$.

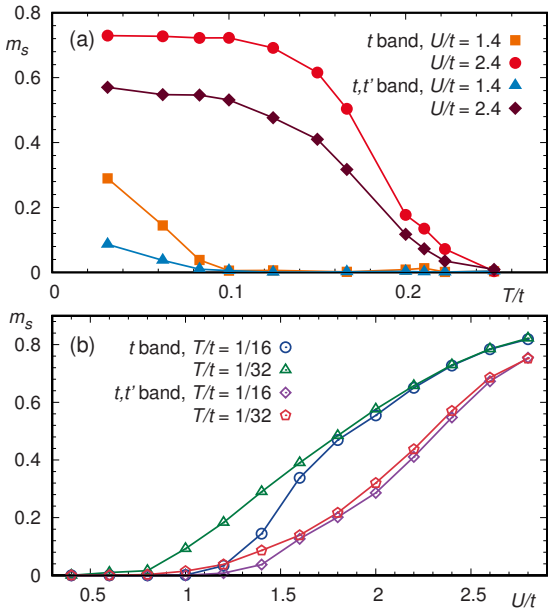


FIG. 2. (Color online) (a) Staggered magnetization $m_s$ for the unfrustrated band ($t$ band) and the frustrated band ($t,t'$ band) (a) as a function of $T/t$ for $U/t=1.4$ and 2.4 and (b) as a function of $U/t$ for $T/t=1/16$ and 1/32. A continuous transition with a smooth increase in $m_s$ is observed as a function of $U/t$.

calculations done on the frustrated one-band Hubbard model[17] with frustration strength $t'/t=0.58$ showed the existence of a first-order phase transition from 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.[17] In order to analyze the metal-insulator transition, we present in Fig. 1(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 DOSs 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 $V_2O_3$.[17,20]

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[15] 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 temperature $T/t$ for two different interaction strengths $U/t$. At $U/t=1.4$ ($U/t=2.4$), the staggered magnetization $m_s>0$ for both frustrated and unfrustrated bands is detected as temperature decreases below the Néel temperature around $T_N/t\simeq 0.1$ ($T_N/t\simeq 0.22$) where the system undergoes a PM-AF phase transition. The staggered magnetization increases more rapidly in the unfrustrated band than in the frustrated one. In Fig. 2(b) we show the staggered magnetization as a function of interaction strength $U/t$ for two temperature values. We find a smooth increase in the magnetization with $U/t$ for both bands and for both temperatures. Unlike the case of magnetic frustration in both bands where a first-order phase transition was observed [Fig. 1(a)], this smoothly increasing behavior of the staggered magnetization $m_s$ is a strong evidence of the existence of continuous phase transitions. It also suggests the existence of an AF metal where 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

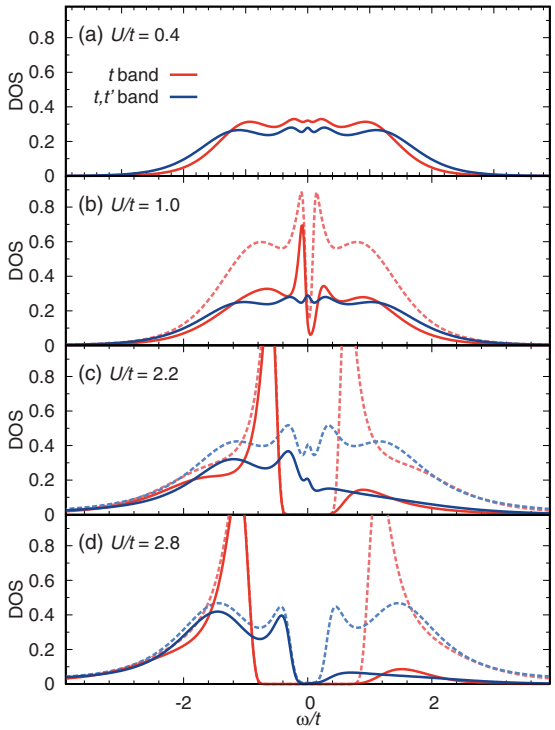


FIG. 3. (Color online) Spin-up DOS on sublattice A (solid lines) as a function of frequency analyzed at $T/t=1/32$ for (a) $U/t=0.4$, (b) $U/t=1.0$, (c) $U/t=2.2$, and (d) $U/t=2.8$. Also shown (dotted lines) is the total spin-up DOS when AF order occurs.

metallic state in both bands is observed at $U/t=0.4$ [see Fig. 3(a)]. As the interaction is increased to $U/t=1.0$ [see Fig. 3(b)], a magnetic transition occurs where the frustrated band $(t,t'$ band) remains in a PM state while an AF metallic state 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 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 in 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

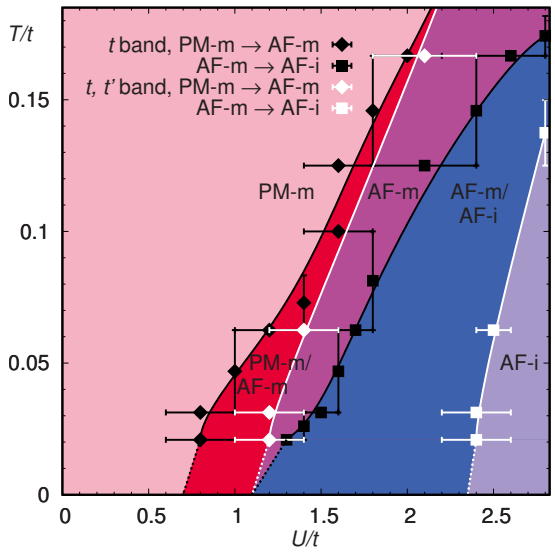


FIG. 4. (Color online) Magnetic phase diagram for the two-band Hubbard model where an unfrustrated band ($t$ band, $t=1$, $t'=0$) and a frustrated band [$(t,t')$ band, $t=1$, $t'=0.65$] coexist. The phase boundaries' error bars are also shown.

a square lattice shows AF insulating behavior with high ordered magnetic 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 coupling ($J_z$), which favors ferromagnetic arrangement of spins on the same site, is switched on between these two bands, the spins in the nonmagnetic frustrated band tend to order antiferromagnetically as in the unfrustrated band, and the itinerant electrons have the tendency to localize since hopping between nearest-neighbor sites violates Hund's rule. On the other hand, the spins in the AF unfrustrated band are affected by frustration due to the Hund's coupling to the frustrated band. Therefore, they follow the spin arrangement in the frustrated itinerant band and

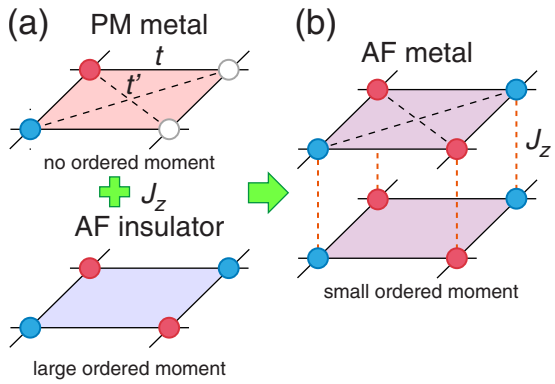


FIG. 5. (Color online) Schematic picture for the mechanism of the appearance of an AF metal with small-ordered moment from the coupling of a frustrated band (PM metal) with an unfrustrated band (AF insulator). The idea is sketched on a square lattice with regard to iron-based superconductors. Red (dark gray) and blue (light gray) sites suggest spin configurations, with white sites representing frustration.

become more delocalized [see Fig. 5(b)]. Such an interplay between frustrated and unfrustrated bands results in a reduction in the antiferromagnetic ordered 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-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 insulating 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 between 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 $t_2'/t_2=0.65$ but 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 ($t_1'/t_1=0.2$). In both cases we find solutions of AF metals, underlining the relevance of the investigated model for the new iron-based superconductors. 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 calculations 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 qualitatively capture the central physics of AF metal with small-ordered magnetic moment observed experimentally in undoped iron-based superconductors as well as the nature of the phase transitions, further investigations have to be done by including all five Fe $3d$ orbitals with realistic interband, intraband 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 explaining the physics of $V_2O_3$ contrary to previous suggestions,[17] and inclusion of other degrees of freedom like phonons may be necessary.

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