Band antiferromagnetism in a two-band model for iron pnictides

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Abstract – In this work I investigate a two-band Hubbard model using the Gutzwiller wave function. The tight-binding part of the model was constructed to have a gapless spin-density wave state which leads to Dirac points in the bandstructure, a common feature of many iron-pnictide compounds. For quarter, half and three-quarter fillings I show that Hund’s rule coupling has a large impact on the metal-insulator transition in the paramagnetic phase. For the half-filled model in the antiferromagnetic phase, the magnetism evolves in a Stoner-like behavior and the size of the ordered moment is mainly determined by the Hubbard interaction. As Hund’s coupling plays a minor role in this state, the model does not describe Hund’s metal which is in contrast to more realistic models for iron-pnictide compounds.

The study of (antiferro)magnetism in multi-band Hubbard models has received a strong push by the recently discovered iron-based superconductors. The ground states of many of these materials are characterized by an antiferromagnetic state (AFM) with a small ordered moment. This small magnetic moment cannot be captured by Density Functional Theory (DFT) calculations [1–3].

To understand the discrepancy between the predictions of DFT and experiments, electron-electron correlations are believed to play an important role. This issue was addressed by several groups employing the Dynamical Mean-Field Theory (DMFT) [4–6]. They have shown that the ground state of a multi-band Hubbard model with realistic values of \((U, J)\) is an ordered stripe AFM (metallic), with a magnetic moment that is strongly reduced with respect to the LSDA value \((m_{\text{LDA}} \approx 2.0 \mu_B)\). Especially, the ordered magnetic moment is significantly smaller than the local moment. This particular ground state was named Hund’s metal, to signify that in these correlated multi-band metals, Hund’s coupling plays a more important role than the on-site Coulomb correlation \(U\) [6].

The same questions were also recently studied by employing the Gutzwiller wave function [7–9]. It was shown that the Gutzwiller approximation captures the correct physics, i.e., a small ordered magnetic moment and a large local moment. Additionally, the small ordered moment phase reveals a very simple Stoner (Slater) picture, i.e., the value of the magnetic moment is essentially determined by band physics. The ground state displays the band magnetism of correlated quasi-particles, i.e., an intermediate state between the local moment (Heisenberg) picture and the itinerant (Slater) picture. Both pictures have been proposed as a starting point for the magnetic ground state of iron pnictides [10–12].

Besides magnetism the explanation of superconductivity in iron-pnictide compounds remains an important issue. Unfortunately, the rich bandstructure of realistic models complicates the discussion. Therefore, one tries to reduce the complexity of the models, and a minimal model for the iron pnictides usually contains two bands with \(d_{xz}\) and \(d_{yz}\) orbitals [13–16]. In this study, I address the question how many-particle correlations influence the magnetic properties and the bandstructure of such a two-band model by using the Gutzwiller wave function. In the first part of this work I investigate the paramagnetic phase of the model. I show that the metal-insulator transition depends strongly on the size of Hund’s rule coupling. In the second part of this work I show how antiferromagnetism evolves in the half-filled model. The magnetic moment is determined by an effective energy scale that depends mainly on the Hubbard interaction, and Hund’s rule exchange has only a small impact. Therefore, the investigated two-band model cannot be considered as Hund’s metal which is assumed to be an important property of iron-pnictide compounds.

The investigated two-band model was introduced by Ran et al., and contains essential topological single-particle properties of a large group of the iron
that the model is not particle-hole symmetric. In a previous publication, the model was studied using a slave-rotor approach which leads to a rich phase diagram [18]. In the current study I apply the Gutzwiller wave function to the model. Compared to other methods, the Gutzwiller ansatz is numerically fairly cheap. Therefore, it is possible to scan a large part of the \((U,J)\) parameter space as compared to other methods [19].

I investigate the following model in two dimensions:

\[
\hat{H} = \hat{H}_0 + \hat{H}_C = \sum_{i,j,b,b',\sigma} t_{i,j}^{b,b'} c_{i,b,\sigma}^\dagger c_{j,b',\sigma} + \sum_i \hat{H}_{C,i},
\]

where \(\hat{H}_0\) describes the kinetic energy of the electrons and \(\hat{H}_C\) includes the correlation part. As already mentioned, for the kinetic energy term, I employ the two-band model of Ran et al. [17]. The underlying lattice is quadratic, with \(x\) and \(y\) axes directed along the edges of the square. The two orbitals have \(XZ\) and \(YZ\) symmetry and the total bandwidth of the model is \(W = 12.8\) eV. For the correlated part of the Hamiltonian, I choose

\[
\hat{H}_C = \hat{H}_C^{\text{dens}} + \hat{H}_C^{\text{gg}},
\]

\[
\hat{H}_C^{\text{dens}} = U \sum_b \hat{n}_{b,\uparrow} \hat{n}_{b,\downarrow} + \sum_{\sigma,\sigma'} \bar{U}_{\sigma,\sigma'} \hat{n}_{XZ,\sigma} \hat{n}_{YZ,\sigma'},
\]

\[
\hat{H}_C^{\text{gg}} = J \sum_{\sigma,\sigma'} \hat{c}_{XZ,\sigma}^\dagger \hat{c}_{YZ,\sigma'}^\dagger \hat{c}_{XZ,\sigma'} \hat{c}_{YZ,\sigma} + (J^{\text{gg}} \hat{c}_{XZ,\sigma}^\dagger \hat{c}_{XZ,\sigma'}^\dagger \hat{c}_{YZ,\sigma'} \hat{c}_{YZ,\sigma}).
\]

Here, I dropped the lattice-site indices and introduced the abbreviations \(\uparrow = \downarrow, \downarrow = \uparrow\), and \(\bar{U}_{\sigma,\sigma'} = (U' - \delta_{\sigma,\sigma'} J)\), where \(U, U'\) and \(J\) are the local Coulomb and exchange interactions. For \(t_{2g}\)-orbitals the relation \(U = U' + 2J\) holds. In the current study an equi-spaced grid of \(U, J\) values, for \(U < 20.0\) eV and \(J < 2.0\) eV is explored. To investigate the saturation of the magnetic moment, I push \(U\) and \(J\) up to 30 eV and 5 eV, respectively, for selected isocontours.

The Gutzwiller variational wave function

\[
|\Psi_G\rangle = \hat{P}_G |\Psi_0\rangle = \prod_i \hat{P}_i |\Psi_0\rangle
\]

approximates the true ground state of \(\hat{H}\) in (1). The wave function \(|\Psi_0\rangle\) is a product state of filled Bloch orbitals and is determined self-consistently. The operator \(\hat{P}_i\) is the local Gutzwiller correlator which is defined as

\[
\hat{P}_i = \sum_{\Gamma} \lambda_{\Gamma} |\Gamma_i\rangle \langle \Gamma_i|.
\]

Here, \(|\Gamma_i\rangle\) are the eigenstates of \(\hat{H}_{C,i}\), and for each eigenstate variational parameters \(\lambda_{\Gamma}\) are introduced. The wave function (3) generates the energy functional \(E[|\Psi_G\rangle] = \langle \hat{H} |\Psi_G\rangle\) that has to be minimized with respect to the single-particle product state \(|\Psi_0\rangle\) and the parameters \(\lambda_{\Gamma}\).

In the limit of infinite spatial dimensions, expectation values can be evaluated without further approximations [20,21]. For finite-dimensional systems, I use this energy functional as an approximation (“Gutzwiller approximation”). This approach also provides the Landau-Gutzwiller quasi-particle bandstructure that can be compared with ARPES data [19,22,23].

I begin the discussion with considering the paramagnetic state of the system with average electron numbers \(N = 1, 2, 3\) per site which corresponds to quarter, half and three-quarter fillings. The results of the Gutzwiller calculations are shown in fig. 1. For these calculations, I fixed the \(J/U\)-ratio and fig. 1 shows the quasi-particle renormalization \(Z\) as a function of the Hubbard \(U\). For \(Z\) going to zero, I find a metal insulator transition (MIT) of the Mott-Hubbard type (Brinkman-Rice transition [24]). The critical value of the Hubbard interaction for this MIT, \(U_c\), is typically not well estimated in the Gutzwiller approximation. Nonetheless, the qualitative trend is usually correct. Therefore, the results shown in fig. 1 are consistent with a recent DMFT model study [25,26].

For quarter filling and \(J = 0\), the quasi-particle weight goes to zero at \(U_c \sim 2.5\) W. The critical value for the Hubbard interaction increases with \(J/U\). For \(N = 1\), larger Hund’s rule coupling suppresses the MIT. If the system is half-filled, i.e. \(N = 2\), Hund’s rule coupling has the opposite effect. For \(N = 2\) and \(J = 0\), the critical value for the Hubbard interaction is also \(U_c \sim 2.5\) W. However, the critical value decreases with increasing \(J\), e.g., for \(J/U = 0.15\)
the critical value is reduced to $\sim 1.2 W$. The case of a three-quarter filling of the model, $N = 3$, is similar to quarter filling, \textit{i.e.} $N = 1$. For $N = 3$ and $J = 0$, the MFT occurs at $U_c \sim 1.6 W$, \textit{i.e.}, the system with electron number $N = 3$ is more correlated than the system with a smaller number of electrons. For a finite $J$, the critical $U$-value increases as a function of $J$, as in the quarter-filled case, $N = 1$. The explanation of this observation can be related to the fact that the system is not particle-hole symmetric. Therefore, the trends for $U_c(J)$ are the same for $N = 1$ and $N = 3$ although there are quantitative differences. Similar results have been observed in a previous DMFT study \cite{25}. This is especially interesting since the model from the DMFT study contains three orbitals and is particle-hole symmetric.

To summarize these results, in the paramagnetic case the system shows a strong dependency on the size of Hund’s rule coupling. As already observed in previous studies, Hund’s rule coupling decreases ($N = 1.3$) or increases ($N = 2$) correlation effects, depending on the number of electrons. In contrast, the antiferromagnetic phase of the half-filled two-band model only weakly depends on $J$, as I will show next.

The present two-band model may be considered a minimal model for iron-pnictide superconductors with emphasis on the topological aspects of the bandstructure \cite{17}. In the following discussion of the antiferromagnetic ground state, I focus on the system at half-filling and the corresponding Fermi surface of the model is shown in fig. 2. The Fermi surface —fig. 2(a)— comprises hole pockets around the $\Gamma$ and the $M$ point, and two electron pockets around the $X$ and the $Y$ point, with $XZ$ and $YZ$ orbital characters. This Fermi surface topology shows some similarities in comparison with the Fermi surface topology of more realistic models for iron-based superconductors, \textit{e.g.}, an eight-band model \cite{27}. The main differences are that

the hole pocket at the $\Gamma$ point is much smaller in the more realistic model and, as a result of the more complex bandstructure, two hole pockets are located at the $M$ point.

Nevertheless, in accordance with more realistic models the Fermi surface topology of the present two-band model leads to a strong nesting between the hole and electron pockets with vector $\mathbf{Q} = (0, \pi)$ (the vector $\mathbf{Q} = (\pi, 0)$ is equivalent, but was not used in this work). This corresponds in real space to a \textit{stripe} spin-density wave (SDW) pattern, in which the spins align ferromagnetically along one of the two equivalent directions ($x$ in the following), and antiferromagnetically along the other ($y$). In this SDW state, the bands fold on top of each other along the $1/2 \Gamma-X$ line, bringing the $Y$ point on top of the $\Gamma$ point, and the $M$ point on top of the $X$ point.

The unperturbed bandstructure for the antiferromagnetic irreducible Brillouin zone is shown in black in fig. 3(a) and the Fermi level is chosen to be zero. The bands display four crossings: one along the $1/2 \Gamma-Y-\Gamma$ direction, two along the $\Gamma-X$ direction and one along the $X-1/2 XM$ direction. Comparing these bands with the bands from fig. 3(b) shows the influence of local interactions onto the bandstructure. The black bands in fig. 3(b) result from a Gutzwiller calculation with $U = 7.0 \text{ eV}$ and $J = 1.0 \text{ eV}$, and with $U = 9.0 \text{ eV}$ and $J = 1.0 \text{ eV}$. One observes that exchange gaps in the electronic spectrum open only along the $\Gamma-X$ direction. These are the crossings where the orbital characters of the folded states at the Fermi surface match.

The two remaining crossings result from bands with different orbital character. As a consequence, no exchange gap opens along the $1/2 \Gamma-Y-\Gamma$ and the $X-1/2 XM$ line, but the folded bands form \textit{Dirac} cones in the dispersion. The topological reasons for this behavior are explained in
Fig. 4: (Colour on-line) Paramagnetic (dashed lines) and antiferromagnetic (full lines) results from Gutzwiller calculations for the model for $J = 0$ (black), $J = 0.5$ eV (green), $J = 0.8$ eV (red) and $J = 1.0$ eV (blue); (a) quasi-particle weight $Z$, (b) population of the local charge state $n = 2$.

Fig. 5: (Colour on-line) Gutzwiller calculation results for the model at $N = 2$; (a) magnetization $m$, (b) average local moment $S^2$. Denotation of lines as in fig. 4.

In fig. 4(a) I show the quasi-particle renormalization $Z$, for both the paramagnetic (dashed) and the antiferromagnetic solutions. As already seen in fig. 1, in the paramagnetic case, I find a Brinkman-Rice transition at some critical value $U_c$. This critical value depends on Hund’s rule exchange and for $N = 2$ larger Hund’s rule exchange decreases $U_c$. The situation changes drastically in the presence of antiferromagnetic stripe order. Considering the quasi-particle renormalization $Z$, the antiferromagnetic solution closely follows the paramagnetic one for not too large $U$ ($U \leq 0.5\, W$). As the long-range magnetic order sets in, the values for $Z$ become larger than in the paramagnetic case, i.e., correlations become weaker. Increasing $U$ further leads to a jump in $Z$ towards unity. The reason for such a behavior is the metal insulator transition of Slater type at $U_S$ which was already discussed. One observes that for larger values of Hund’s rule coupling the MIT occurs for smaller values of $U$. As a consequence, the change in the quasi-particle renormalization is less dramatic for larger values of $J$.

In fig. 4(b), I plot the expectation value of the atomic configurations with two electrons ($n(2)$). In the paramagnetic case, $n(2)$ starts from the non-interacting limit $n(2) = 3/8$ and saturates to unity at the MIT, where all charge fluctuations are frozen (Brinkman-Rice localization transition). A comparison of fig. 4(a) and fig. 4(b) shows that the behavior of $n(2)$ follows closely that of the quasi-particle weight $Z$. As in the case of the quasi-particle renormalization $Z$, the behavior of $n(2)$ is strongly influenced by the antiferromagnetic order. Here, $n(2)$ starts by following the paramagnetic curve, but it departs around
the MIT. While in the paramagnetic case with a Mott-Hubbard MIT $n(2)$ goes to unity, in the antiferromagnetic case $n(2)$ only increases slowly. It saturates to unity at values of $U$ much larger than the critical $U_c$ for the paramagnetic MIT. This again shows that the (Slater) band-insulating regime is less correlated than the corresponding paramagnetic regime.

In fig. 5(a) I plot the long-range–ordered magnetic moment $m$ as a function of $U$ for different values of Hund’s rule coupling. The plot shows indeed that the point where the paramagnetic and antiferromagnetic curve of $Z$ ($n(2)$) run apart, is the point where long-range antiferromagnetic order sets in. Moreover, for every curve in fig. 5(a) one can observe a value of the Hubbard interaction with a sudden change of the slope of the $m$ vs. $U$-curve, e.g., at $U \approx 1.5$ W for $J = 0$. These points indicate the Slater MIT in the antiferromagnetic case.

In order to conclude these observations, in the parameter range of this model study only three phases are stable: a paramagnetic metal for small values of $U, J$, an AFM metal for intermediate values and, finally, an AFM band (Slater) insulator for $U \gg W$. This is in stark contrast to the phase diagram obtained by the slave rotor approach for the same model in ref. [18]. The phase diagram obtained by this method is much richer compared to what is observed with the variationally controlled Gutzwiller approach.

Next, I discuss the relationship between the ordered magnetic moment and the local magnetic moment in more detail. Figure 5(b) shows the average value of the local spin operator $\langle S^2 \rangle$, where the dashed lines show the results in the paramagnetic case. For the local moment one has to distinguish the cases $J = 0$ and finite values of $J$. For $J = 0$, the value $\langle S^2 \rangle$ as a function of $U$ increases from the non-interacting limit $\langle S^2 \rangle = 3/4$ only slowly. In contrast to this, for finite $J$, one observes a strong increase to the fully localized value $\langle S^2 \rangle = 2$ at the Mott-Hubbard MIT. Again, the results change substantially when one allows for antiferromagnetic order. Only for $J = 0$ there is a sudden increase of $\langle S^2 \rangle$ at the Slater MIT. For a finite value of $J$, however, the dependence of $\langle S^2 \rangle$ on $U$ is smooth. It is interesting to note that, for $U \gtrsim 15$ eV $\sim 1.2$ W and for finite $J$, the local magnetic moment is larger in the paramagnetic than in the antiferromagnetic phase. This seemingly counter-intuitive result is also found in a recent DMFT study of the single-band Hubbard model [28,29]. In this publication, it is also reported that in the weak-coupling regime the antiferromagnetic order is stabilized by a gain in potential energy, while in the strong-coupling regime a gain in kinetic energy leads to the stabilization of the magnetic phase. The results of the current work agree with this observation.

Finally, I consider the shape of the curves $m(U)$ in fig. 5(a) and the shape of $\langle S^2 \rangle$ as a function of $U$ (fig. 5(b)) in the antiferromagnetic case for finite $J$. All curves strongly resemble each other and, as seen in fig. 4(a), the quasi-particle renormalization is close to unity. This indicates that a band picture for quasi-particles will provide a reasonable description. In order to illustrate this concept, I show the results of a Stoner mean-field calculation for the half-filled two-band model in fig. 6. Here, the magnetization $m$ is shown as a function of the Stoner parameter $I$ (green line). The two quantities are related to each other via the self-consistency condition $\Delta = mI$ with the splitting parameter $\Delta$. The plot shows that the $m(I)$ curve has a continuous and monotonous behavior between $m = 0$ and $m = 2 \mu_B$. The inflection point at $m \sim 1.41 \mu_B$ coincides with the metal-insulator transition, i.e., with the point, where the exchange gap is large enough to eliminate the Dirac points. It corresponds to $I = 3.43$ eV and $\Delta = 4.8$ eV.

In fig. 6 I also plot the ordered magnetic moment from the Gutzwiller calculations as a function of the effective energy scale $I_{\text{eff}} = J + U/3$. The data for small $J$ are anomalous, therefore I just consider results with $J > 0.2$ eV. By choosing the energy scale $I_{\text{eff}}$, all the curves $m(U, J)$ fall on top of each other. The inset of fig. 6 shows the magnetization as a function of the band gap. The results are from the Stoner calculation (green) and from the Gutzwiller calculation with $J = 1.0$ eV (black). In principle, the dependence of the ordered magnetic moment on the bandwidth could be tested experimentally. As these two curves fall onto each other, also from this point of view there is no fundamental difference between the Stoner results and the Gutzwiller results. From these observations one can draw three important conclusions:

1) Except for small $J$, which is anomalous, there is a unique, atomic scale governing magnetism, both for the ordered and local moment. In the present case it is $I_{\text{eff}} = J + U/3$.

2) The value of the magnetic moment, both ordered and local, is set by the band response of the system.
Note in particular, that $U$ is considerably larger than $J$. Therefore, the effective energy scale $I_{\text{eff}}$ is mainly determined by the value of the Hubbard interaction. This is in strong contrast to the recent Gutzwiller study on an eight-band model for LaOFeAs. In that study the effective energy scale for the magnetism was mainly determined by Hund’s coupling $J$ [9]. I consider this as an important point as it discloses qualitative differences between the minimal two-band model and more realistic models.

To summarize, I studied a two-band model for the iron pnictides. This model contains two important properties of this class of materials: the strong nesting that leads to a stripe-ordered spin-density wave and the Dirac points. The magnetism itself is determined by an effective energy scale $I_{\text{eff}} = J + U/3$. Since $I_{\text{eff}}$ is large, of the order of $U$, all results reflect Stoner-type band magnetism. Moreover, as $I_{\text{eff}}$ depends marginally on Hund’s coupling the model does not describe Hund’s metal. This observation is in contrast to studies on more realistic models. For large interactions the model describes an antiferromagnetic band insulator. The influence of the Dirac points is important as they delay the opening of the band gap.

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