Electronic correlations in multiorbital systems and iron superconductors: Hund vs Mott

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We analyze the electronic correlations in multiorbital systems as a function of intraorbital interaction $U$, Hund’s coupling $J_H$ and electronic filling $n$ and clarify the relationship between Hund’s coupling and Mott physics. We show that correlations are strongly dependent on filling, but not only via the average orbital occupancy. As in half-filled Mott systems the small $Z$ values originate in the suppression of electronic configurations which reduce the atomic magnetic moment, specially the double occupancy of a given orbital, what results in spin fluctuations $C_S$ almost saturated in the Hund metal and connects the suppression of the quasiparticle weight $Z$ at moderate interactions to the Mott insulating state at half-filling. However, contrary to what happens in Mott correlated states the reduction of $Z$ with $J_H$ can happen on spite of increasing charge fluctuations. We also discuss the different behavior found at large $U$ and $J_H$ which includes the increase of $Z$ with $J_H$.

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The Mott transition is one of the most dramatic manifestations of correlations between electrons\cite{1,2}. In the single orbital Hubbard model at half-filling the system becomes insulating at a critical intraorbital interaction $U_c$ to avoid the cost of doubly occupying the orbital. Away from half-filling metallicity is recovered. Nevertheless atomic configurations involving double occupancy are avoided and the quasiparticle weight $Z$ decreases. The reduction of $Z$ with $U$ comes with the suppression of charge correlations, which vanish at the transition, and the enhancement of spin fluctuations. In cuprates superconductivity appears when a Mott insulator is doped.

To describe iron superconductors, ruthenates and many oxides requires the inclusion of several orbitals. In multiorbital systems the Mott transition happens not only at half-filling but at all integer fillings\cite{3}. The crucial role of Hund’s coupling $J_H$ on electronic correlations has been recognized only recently\cite{4,5}. $J_H$ modifies $U_c$ in a doping dependent way\cite{6,7} and promotes bad metallic behavior in a wide range of parameters\cite{8,9}. The term Hund metal was coined\cite{11} in the context of iron superconductors to name the correlated metallic state induced by Hund’s coupling at moderate intraorbital interactions.

Hund metals were described as strongly correlated but itinerant systems which are not in close proximity to a Mott insulating state and have physical properties distinctly different from doped Mott insulators\cite{7}. The suppression of coherence by $J_H$ was related to the reduction of the atomic ground-state degeneracy\cite{10} and the large $U$ limit discussed using Kondo impurity models\cite{7,12,13}.

On the other hand, a number of authors\cite{16,17,18,19,20,21,22} including one of us, have described iron superconductors as doped Mott insulators. Undoped iron superconductors accommodate 6 electrons in 5 orbitals, with average orbital filling $x = 1.2$. In this description correlations increase with hole doping as electronic filling approaches half-filling\cite{14,22}. There is some experimental evidence of an enhancement of correlations with hole-doping\cite{21,23,24}.

It urges to clarify the nature of correlations in Hund metals and its relationship with Mott physics. In this paper we analyze the electronic correlations in multiorbital systems ($N = 2,3,..5$ orbitals) as a function of interactions and electronic filling $n$. We find that the suppres-
sion of the quasiparticle weight $Z$ with $J_H$ at moderate interactions is strongly dependent on doping and it is directly connected to the Mott transition at half-filling. However, contrary to what happens in Mott correlated states the reduction of $Z$ with $J_H$ does not necessary imply a suppression of charge fluctuations. The spin fluctuations increase and are almost saturated in the Hund metal. We show that this behavior can be traced back to the energy cost for hopping in the different channels. The small $Z$ values and the concomitant enhancement of spin fluctuations are due to the suppression of atomic configurations with reduced magnetic moment, specially those involving double occupancy of a given orbital. The dependence of electronic correlations changes at large $U$ and $J_H$. Therefore the large $U$ behavior should not be used to discuss the correlations at moderate interactions.

To avoid the complications due to inequivalent orbitals and to interorbital hybridization, typical of iron superconductors and ruthenates, we consider degenerate multiorbital systems with hopping $t$ restricted to the same orbital and to nearest neighbors. Interactions are local and given by the Kanamori Hamiltonian $H_c[3, 20].$ The system, assumed two-dimensional, satisfies $U'' = U - 2J_H$, with $U''$ the interorbital interaction, as found in rotationally invariant systems $[29].$ The model is particle-hole symmetric with respect to half-filling. We treat the interactions using a $Z_2$ slave spin representation $[27, 28].$ Only density-density are considered, thus the hopping does not enter into the calculation $[21, 22].$

Fig. 1(a) shows in color plot the quasiparticle weight $Z$ as a function of $U$ and $J_H$ for a five-orbital system with six electrons, the filling of undoped iron superconductors. Three regions can be distinguished: a metallic state with moderate correlations in yellow-orange color; an insulating Mott state at large $U$ in black, and a strongly correlated metallic state with reduced coherence in violet. The critical $U_c$ at which the Mott transition sets it depends non-monotonously on $J_H$ $[9].$ At large values of $J_H$ the system remains metallic even for large $U$ $[10].$

The correlated metallic state, in the following Hund metal, appears at finite $J_H$ in a wide range of parameters, including $U$ smaller than the bare bandwidth $W = 8t.$ The way in which this region depends on the interactions reveals the crucial role played by $J_H$ on inducing the strong correlations which seem unrelated to the $n = 6$ Mott insulating state. Similar phase diagrams are found in other cases, e.g. 2 electrons in 3-orbitals in Fig. 2(d) and 2 and 3 electrons in 4 orbitals and 3 electrons in 5 orbital, Fig. 1 in Suppl. Mat.. Contour lines in 2 electron diagrams depend less on $J_H$ for $U < 2W,$ see below.

Phase diagrams with a similar shape have been previously found in multiorbital systems with inequivalent orbitals $[6, 27]$ including models for iron superconductors $[19, 30, 31].$ Our results show that orbital differentiation is not a requisite to observe such a phase diagram, but that it is a generic feature of multi-orbital systems with intermediate and commensurate filling $n \neq 1, 2N - 1, N.$

The dependence of $Z$ on $J_H$ for the system in Fig. 1(a) and selected values of the intraorbital interaction $U$ is plotted in Fig. 1(b). At small $J_H$, $Z$ shows a very slight, but robust, increase with $J_H$ followed by a sharp drop at intermediate $J_H$ driving the system to the strongly correlated state, and even to a Mott insulator with $Z = 0$ at large $U.$ Two different behaviors can be distinguished when $J_H$ keeps growing: $Z$ continues decreasing slowly at moderate $U$, while for larger interactions it changes behavior and increases with $J_H.$ The large $U - J_H$ region with positive derivative $dZ/dJ_H$ extends to interactions smaller than the minimum $U_c$, see also Suppl. Mat.

In Fig. 1(c) we plot $Z$ as a function of the electronic filling $n$ and $J_H$ for $U = W,$ far from the $n = 6$ Mott transition. The strength of correlations show a clear asymmetry with electronic filling around $n = 6.$ No special feature is observed at this filling for this value of $U$ what confirms that the $n = 6$ Mott transition is not responsible for the strong suppression of $Z.$ On the other hand the entrance to the strongly correlated Hund metal appears at smaller $J_H$ as $n$ approaches $n = 5.$ The connection with the Mott insulating state at half-filling is evident.

A clear doping dependence of correlations is also ob-
served in 3-orbital systems, Fig. 2. An extended region of parameters with small quasiparticle weight, in violet, is found only for fillings relatively close to half filling \( n = 3 \). For smaller fillings \( Z \) depends weakly on \( J_H \). The dependence of the Hund metal region on the interaction parameters for filling \( n = 2.5 \) in Fig. 2(e), closely follows the \( n = 3 \) Mott insulating state, in black in Fig. 2(f).

The interaction \( J_H(U) \) at which the system enters into the Hund metal can be defined empirically as the value of \( J_H \) with the strongest suppression of \( Z \), i.e. \( dZ/dJ_H \) most negative, after which \( Z \) stays finite. Fig. 3(a) shows \( J_H(U) \) obtained from the numerical calculations and different multiorbital systems. \( J_H \) decreases with \( U \). In most cases, at a given \( U \), \( J_H \) is smaller for average orbital filling \( x = n/N \) closer to half-filling, as could be expected for doped Mott insulators. However this tendency is reversed when 2 electrons in 3 orbitals \( (x = 0.66) \) and 3 electrons in 5 orbitals \( (x = 0.60) \) are compared.

One of the hallmarks of Mott physics is the suppression of charge fluctuations \( C_{n_T} \) which comes along with the reduction of \( Z \) and evidences the increasing localization of the electrons. Fig. 4(a) shows the evolution of \( C_{n_T} \) with \( J_H \) and compares it with that of \( Z \), both quantities being normalized to the corresponding \( J_H = 0 \) value. Here \( C_{n_T} = < n_T^2 > - < n_T >^2 \) with \( n_T = \sum_{a=1,...,N}(n_{a\uparrow} \pm n_{a\downarrow}) \), where \( a \) is the orbital index, \( \uparrow \) and \( \downarrow \) the spin, \( n_{a\uparrow} \) and \( n_{a\downarrow} \) the electron occupancy of a given orbital with spin \( \uparrow \) or \( \downarrow \) and \( < n_T > \) is the average electron density.

Interestingly, for the system with 2 electrons in 3 orbitals \( C_{n_T} \) increases with \( J_H \). The suppression of \( Z \) happens on spite of an increase of metallicty, contrary to what happens in Mott systems. In the 6 electrons in 5 orbitals case the strong reduction of \( Z \) at \( J_H \) comes along with a reduction of \( C_{n_T} \). However at larger \( J_H \) deviations from Mott physics reappear as \( Z \) continues decreasing, even if more weakly, while \( C_{n_T} \) increases. The regions with increasing and decreasing \( Z \) and \( C_{n_T} \) with \( J_H \) for both cases are compared in the Suppl. Mat., Figs. S2 and S3. Fig. 4(b) show the evolution of \( C_{n_T} \) vs \( J_H \) with electronic filling. \( Z \) continuously decreases with \( J_H \) for all values in this figure and vanishes in the Mott state at \( n = 5 \) (not shown). The enhancement of \( C_{n_T} \) is reduced as half-filling \( (n = 5) \) is approached.

The dependence of \( Z \) and \( C_{n_T} \) on \( J_H \) can be understood from the energy of the hopping processes. We consider two \( N \)-orbital atoms with \( n \) electrons \( (n \leq N) \). We assume that inside each atom the electron spins are parallel to satisfy Hund’s rule. An electron which hops from one atom onto the other one can end into (i) an empty orbital with spin parallel to that of the occupied orbitals. The interaction energy cost is \( E^{\uparrow\uparrow} = U - 3J_H \) and the hopping promoted by \( J_H \); (ii) an empty orbital with spin antiparallel to that of the occupied orbitals with \( E^{\uparrow\downarrow} = U + (n - 3)J_H \). (iii) an occupied orbital with energy \( E^{\uparrow\downarrow} = U + (n - 1)J_H \). At half-filling, \( n = N \), processes (i) and (ii) are blocked by Pauli exclusion principle and process (iii) controls the critical \( U_c(J_H) \) for the Mott transition. \( U_c(J_H) \) strongly decreases with \( J_H \), see Fig. 4(f). For other integer fillings and large \( J_H \) the Mott transition is controlled by process (i) and \( U_c(J_H) \) increases with \( J_H \), but processes (ii) and (iii) are blocked at smaller interactions.

In the metallic state process (i) is allowed and promoted by \( J_H \). We ascribe \( J_H \) to avoiding process (iii).
process gives $J_{H}^{\text{intrNN}} \propto (\tilde{W} - U)/(n-1)$ with $\tilde{W}$ some characteristic scale for the kinetic energy, dependent to some extent on $n$, $U$ and $J_H$ and increasing with $N$ due to larger number of hopping channels. As it happens for the critical Mott $U_C$ at half-filling, due to the small ratios $J_{H}/\tilde{W}$, the value of $J_{H}^{\text{CP}}$ is mostly controlled by $\tilde{W}/(n-1)$ not reaching the limit $J_{H}^{\text{CP}} \propto -U/(n-1)$ in the physical range $J_{H}/U \leq 0.33$.

Process (ii) can enhance or reduce the correlations. It is suppressed by $J_{H}$ for $n > 3$ and promoted for $n < 3$ (consequence of $U' = U - 2J_{H}$) making correlations dependent on the absolute value of $n$ and $N$ and not only on the average orbital filling $x = n/N$. This process is behind a smoother suppression of $Z$ and stronger increase of $C_{\text{nv}}$ with $J_{H}$ found for 2 electrons in 3 or 4 orbitals, Fig. 2 and Suppl. Mat., as compared to that of 6 electrons in 5 orbitals, Fig. 1(a). This process is promoted by $J_{H}$ in the first case and suppressed in the later.

The different behavior of $Z$ and $C_{\text{nv}}$ can also be related to the suppression or promotion of the different hopping processes (i) to (iii) by $J_{H}$ assuming that their effect is weighted differently in $Z$ and $C_{\text{nv}}$. When $n$ approaches half-filling in Fig. 4 the enhancement of $C_{\text{nv}}$ is reduced as process (iii) responsible for it is less likely to happen due to Pauli exclusion principle.

We now focus on the spin fluctuations: $C_{S} = < S^2 > - < S >^2$ with $S = \sum_{a=1,...,N}(n_{a\uparrow} - n_{a\downarrow})$ and $< S > = 0$. Hund’s coupling polarizes the spin locally. Therefore $C_{S}$ is enhanced with $J_{H}^{\text{CP}}$. If the strong suppression of $Z$ is due to avoiding double occupying an orbital a jump in $C_{S}$ should be expected at $J_{H}^{\text{CP}}$ and, as shown in Fig. 5(b), it is indeed observed. The enhancement of $C_{S}$ is smoother for $n = 2$. The clear correlation between the suppression of $Z$ and the enhancement of $C_{S}$ confirms our hypothesis. Above $J_{H}^{\text{CP}}$, $C_{S}$ reaches a value close to that of the Mott insulator at this filling. In the Hund metal state each atom is instantaneously highly spin polarized.

The transition between the Hund metal and the Mott insulator is barely felt by $C_{S}$, Fig. 5(b). As in single-orbital systems $C_{S}$, in general, increases with $U$. However, as shown in the inset and Suppl. Mat., at large $J_{H}/U$ the dependence is weakly non-monotonic. This effect found in a region of parameters, similar to that with $dZ/dJ_{H} > 0$, reflects that the spin fluctuations in the metallic state are larger than in the insulator. At large $U$ $C_{S}$ decreases with $U$ as it promotes localization.

In conclusion, we have shown that the correlations in Hund metals originate in the suppression of atomic configurations which reduce the magnetic moment while the hopping of electrons with spin parallel to the locally spin polarized atoms is allowed. The strongest reduction of $Z$ at $J_{H}$ in Hund metals and the Mott transition at half-filling are both caused by the suppression of transport processes which involve the double occupancy of a given orbital. However, as Hund’s coupling favors hopping in other transport channels, in some cases, the suppression of $Z$ with $J_{H}$ comes along with an enhancement of charge fluctuations. Finally, we note that the dependence of $Z$ on $J_{H}$ and the spin fluctuations on $U$ allow to identify two different behaviors in the strongly correlated metal, thus care has to be taken in data analysis.

Our calculations include density-density interactions at a slave spin level. Comparison of this approach with more elaborate techniques produces good qualitative and semi-quantitative agreement. Crossovers could be less sharp and $Z$ values slightly different in other approaches as DMFT, but on general terms we expect the physics reported here to be robust and serve as the starting point to describe multi-orbital materials as iron superconductors or ruthenates. Nevertheless the physics of these materials will be strongly influenced by the inequivalency of the orbitals specific for each material, and not included here.

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FIG. S1: (Color online) Quasiparticle weight $Z$ as a function of intra-orbital interaction $U$ and Hund’s coupling $J_H$ for (a) 4-orbital system with $n = 2$ electrons, (b) 4-orbital system with $n = 3$ electrons, (c) 5-orbital system with $n = 3$ electrons. $U$ and $J_H$ are respectively given in units of the non-renormalized bandwidth $W$ and of $U$. The system shows particle hole symmetry and the results are also valid for electronic filling $2N - n$. In (a) as in Fig.2(d) both with 2 electrons the $Z$ contour lines for $U < 2W$ are almost vertical, i.e. weakly dependent on $J_H$. These arises from the different effect of $J_H$ on the energy of the transport processes in which one electron hops into an empty (process enhanced) or occupied (process suppressed) orbital with opposite spin to that the atom, see text.

FIG. S2: (Color online) Color representation for the sign of the derivative of the quasiparticle weight with Hund’s coupling $dZ/dJ_H$ for (a) 5-orbital system with 6 or 4 electrons (b) 3- orbital system with 2 or 4 electrons, (c) 3-orbital system with 2.5 or 3.5 electrons. Yellow is used for positive derivative, Red for negative derivative. In black, the Mott region with zero derivative. At intermediates values of $U$ and $J_H$ $Z$ is suppressed by $J_H$. At large $U$ and $J_H$, Hund’s coupling promotes metallicity and $Z$ increases with $J_H$. The increase of $Z$ with $J_H$ found for small $U$ and $J_H$ is always very weak.
FIG. S3: (Color online) Color representation for the sign of the derivative of the charge correlations with Hund’s coupling \( \frac{dC_{\eta_{\uparrow}}}{dJ_H} \) for (a) 5-orbital system with 6 or 4 electrons (b) 3-orbital system with 2 or 4 electrons, and the derivative of the spin correlation with \( U \) for (c) 3-orbital system with 2 electrons. Yellow is used for positive derivative, red for negative derivative. In black, the Mott region with zero derivative. In (a) there is a region of parameters with \( \frac{dC_{\eta_{\uparrow}}}{dJ_H} < 0 \) which coincides with the region in Fig. S2(a) where the suppression of \( Z \) with \( J_H \) is strongest. On the contrary, in the case of 2 electrons in 3 orbitals in (b) charge correlations always increase with increasing Hund’s coupling, even for those interaction values with \( \frac{dZ}{dJ_H} < 0 \). At large \( U \) and \( J_H \), the spin correlations decrease with \( U \) as it promotes localization. The region where this effect is found is similar to that shown in Fig. S2(b) with positive \( \frac{dZ}{dJ_H} \).