Hund’s metal regimes and orbital selective Mott transitions in three band systems

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Abstract
We analyze the electronic properties of interacting crystal field split three band systems. Using a rotationally invariant slave boson approach we analyze the behavior of the electronic mass renormalization as a function of the intralevel repulsion $U$, the Hund’s coupling $J$, the crystal field splitting, and the number of electrons per site $n$. We first focus on the case in which two of the bands are identical and the number of the third one is shifted by $\Delta > 0$ with respect to the former. We find an increasing quasiparticle mass differentiation between the bands, for system away from half-filling ($n = 3$), as the Hubbard interaction $U$ is increased. This leads to orbital selective Mott transitions where either the higher energy band (for $4 > n > 3$) or the lower energy degenerate bands ($2 < n < 3$) become insulating for $U$ larger than a critical interaction $U_c(n)$. Away from the half-filled case $|n - 3| \gtrsim 0.3$ there is a wide range of parameters for $U < U_c(n)$ where the system presents a Hund’s metal phase with the physics dominated by the local high spin multiplets. Finally, we study the fate of the $n = 2$ Hund’s metal as the energy splitting between orbitals is increased for different possible crystal distortions. We find a strong sensitivity of the Hund’s metal regime to crystal fields due to the opposing effects of $J$ and the crystal field splittings on the charge distribution between the bands.

Keywords: Hund’s metal, Hubbard model, Mott transition

(Some figures may appear in colour only in the online journal)

1. Introduction

The importance of the Hund’s rule coupling $J$ in the electronic properties of a variety of materials has been recognized in recent years (Haule and Kotliar 2009, Georges et al. 2013). An essential role of $J$, early noticed in van der Marel and Sawatzky (1988), is that in atomic shells with a half-filled configuration, such as $\Delta^j$, $J$ acts to enhance the Mott gap for charge excitations, and thus favours the Mott insulating phase, while for configurations different from half-filling, such as $\Delta^c$, it has the opposite effect, thus promoting metallicity. In addition, it has been understood that in the latter class of systems, even in metallic materials with Coulomb interactions well below the critical Mott transition values, $J$ enhances the electronic correlations as measured, for instance, with the electronic effective mass (de’ Medici et al 2011, deMedici and Capone 2017, Stadler et al 2018). Systems in this regime have been dubbed ‘Hund’s metals’ and notable examples that seem to fit into this class can be found in different material families, like Ru based oxides (Mravlje et al 2011) and Fe based superconductors (Haule and Kotliar 2009, Lee et al 2018). While these families include systems in which the transition metal atom has very different crystal environments, a shared property is that due to the oxidation state (Ru$^{4+}$ or Fe$^{2+}$), the electronic density in the relevant low-energy manifold of electronic bands is different from half-filling, and thus, $J$ significantly increases the critical Coulomb interaction of the metal to Mott insulator transition. In this regime, the electronic mass can still be significantly renormalized due to the suppression of hopping processes involving atomic configurations of spin different than maximum caused by $J$.

Crystal field splittings compete with the Hund’s coupling inducing charge imbalances between the different orbitals that can produce a differentiation of the quasiparticle masses...
between the associated electron bands, even leading to orbital selective Mott transitions (OSMTs) in which only a subset of the bands becomes insulating, Fe$^{2+}$ based materials experimentally found to be Mott insulators like La$_2$O$_3$Fe$_2$Se$_2$ (Zhu et al 2010) and BaFe$_2$S$_3$ (Takahashi et al 2015, Yamauchi et al 2015, Takubo et al 2017, Materne et al 2019), have been theoretically predicted to present OSMTs upon doping or application of hydrostatic pressure (Giovannetti et al 2015, Craco and Leoni 2018, Patell et al 2018). In ruthenates, the interest in OSMTs was sparked by the intriguing experimental observations (Nakatsuji and Maeno 2000a, 2000b) and the prediction of coexistence of metallic and insulating behavior in Sr$_2$−$\epsilon$Ca$_2$RuO$_4$ (Anisimov et al 2002). OSMTs have since been predicted to occur in a variety of physical situations including systems with asymmetric bands (Anisimov et al 2002, Rüegg et al 2005) (different bandwidths or densities of states in the non interacting limit), crystal field split bands, deMedici et al (2005), Vojta (2010), Song et al (2015), and even in momentum space on single band systems (Ferrero et al 2009a, 2009b).

We revisit this problem within a three band model expected to capture the essential physics of $t_{2g}$ shells in transition metal oxides as the ruthenates, vanadates, and titanates (Werner et al 2009) and to serve as a guide for understanding systems with a higher number of relevant orbitals (Stadler et al 2009) and to serve as a guide for understanding systems. In Facio et al (2017) we analyzed in detail the limit of vanishing crystal fields, addressing the question of how interorbital interactions affect the Mott metal-insulator transition. In section 3.1 and 3.2 we analyze for arbitrary electronic densities the electronic correlations in the presence of a distortion that shifts the energy of one orbital while keeping the others degenerated. In section 3.3 we analyze the evolution of the $n = 2$ Hund’s metal under the different possible distortions for a three-orbital system. Finally, in section 4 we present our conclusions.

**2. Model and methods**

We consider the Slater–Kanamori Hamiltonian for a three band system

\[
H_K = \sum_{i,j,m,\sigma} \psi_{ijm\sigma}^\dagger d_{im\sigma}^\dagger d_{jm\sigma} + \sum_{i,m,\sigma} (\varepsilon_m - \mu) n_{im\sigma}
+ U \sum_{i,m} n_{im\uparrow} n_{im\downarrow} + U' \sum_{i,m \neq m'} n_{im\uparrow} n_{im'\downarrow}
+ (U' - J) \sum_{i,m < m'} n_{im\sigma} n_{im'\sigma}
\]

\[
- J \sum_{i,m \neq m'} d_{im\uparrow}^\dagger d_{im\downarrow} d_{im'\downarrow}^\dagger d_{im'\uparrow}
+ J \sum_{i,m \neq m'} d_{im\uparrow}^\dagger d_{im\downarrow} d_{im'\uparrow}^\dagger d_{im'\downarrow},
\]

where $\psi_{ijm}$ is a hopping term between orbital $m$ on site $i$ and orbital $m'$ on site $j$, $\varepsilon_m$ is the crystal field energy, $\mu$ is the chemical potential, $J$ is the Hund’s rule coupling, and $U$ and $U'$ are the intraorbital and interorbital interactions, respectively. We consider, for simplicity, a semicircular density of states for each orbital:

\[
D(\varepsilon) = \frac{2}{\pi D} \sqrt{1 - (\varepsilon/D)^2},
\]

where $D$ is the half-bandwidth of the conduction electron band in the absence of interactions. The interlevel interaction $U'$ in a rotational symmetry (atom in free space) reads $U' = U - 2J$, which as expected satisfies $U' < U$. As it is customarily done, we will use this expression in what follows.

In the DMFT approximation, the interacting problem in the lattice is mapped to an auxiliary quantum impurity problem (QIP) plus a self-consistency condition. In this mapping, the local interacting terms are kept exactly as in the lattice while the terms describing processes that contribute to the electron delocalization are accounted by the coupling between the local multiplets in the impurity to a non-interacting fermionic bath. The self-consistency condition requires that the local Green’s function in the lattice, $G_\alpha(i\omega_n)$, matches the Green’s function obtained by solving the QIP, $G^{\text{imp}}_\alpha(i\omega_n)$. In the cases studied in this manuscript, this condition reads:

\[
G_\alpha(i\omega_n) = \int d\varepsilon \frac{D(\varepsilon)}{\varepsilon + \varepsilon - \Sigma_\alpha(i\omega_n)} = G^{\text{imp}}_\alpha(i\omega_n),
\]

where $\Sigma_\alpha(i\omega_n)$ is the local self-energy obtained in the QIP.

We use the RISB method as an impurity solver for DMFT (Ferrero et al 2009a, 2009b, Huang et al 2012, Hallberg et al 2015, Facio et al 2017, Facio et al 2018). In the RISB method, the local Hilbert space is described with an enlarged space which includes auxiliary bosonic and fermionic degrees of freedom. The fermionic operators $d = [d_\alpha]^T$, where $\alpha$ is a spin-orbital index, are represented as

\[
d = R[[\phi_{AB}]^T],
\]

where $f = [f_\alpha]^T$ are auxiliary fermionic operators and $\{\phi_{AB}\}$ is a set of auxiliary bosonic operators, the so-called slave-bosons. The indexes $A$ and $B$ refer to local multiplets, defined as eigenstates of the on-site Hamiltonian. The interacting terms in the Hamiltonian are represented as a quadratic form
in the slave-bosons which, in the saddle-point approximation, are replaced by their mean values. In this approximation, the free energy associated with the auxiliary QIP reads

\[ \Omega = -T \text{Tr} \ln[-G^\scriptscriptstyle{f}^{-1}(\omega_n)] - \lambda_0 + \sum_{ACD} \phi_{ACD} \phi_{\Lambda 0} + \delta_{CD} \lambda_0 \]

\[ + \delta_{CD} E_A - \mu \text{Tr} \{f^\dagger A f D \} \phi_{AC}. \tag{6} \]

Here, \( E_A \) is the eigenenergy of the multiplet \( |A\rangle \), \( A \) is a set of Lagrange multipliers introduced to remove unphysical states from the enlarged Hilbert space and \( G^\scriptscriptstyle{f}(\omega_n) \) is the Green’s function associated with the auxiliary fermionic operators

\[ G^\scriptscriptstyle{imp}(\omega_n) = [i \omega_n + \mu - \Delta \Lambda (\omega_n) R]^{-1}, \tag{7} \]

where \( \Delta (\omega_n) \) is a function that describes the hybridization of the local multiplets with the non-interacting fermionic bath and is determined in each step of the DMFT cycle. The local self-energy obtained with the RISB method after solving the QIP reads

\[ \Sigma(\omega_n) = (1 - (RR^\dagger)^{-1})(i \omega_n + \mu) + R^\dagger AR^{-1} - \phi_0. \tag{8} \]

This frequency dependence corresponds to that of a Fermi liquid at low energy showing that, in the saddle-point approximation, the fermionic operators provide a description of low-energy quasiparticles whose effective inter-site hoppings and crystal field energies are renormalized by the interactions. In particular, if \( m \) is the bare electron mass, the effective mass associated with the orbital \( \alpha, m^*_\alpha \), can be obtained as

\[ m = \frac{m^*_\alpha}{Z_\alpha} = (RR^\dagger)_\alpha. \tag{9} \]

A detailed analysis of the Hamiltonian equation (1) using DMFT with continuous time quantum Monte Carlo as the impurity solver was presented in Werner et al (2009) but no information on the quasiparticle renormalization was provided there. In figure 1 we present the phase diagram in the absence of crystal fields in the space of parameters \( \mu - U \) for \( J / U = 0.167 \). The black triangles indicate the boundary between metal and insulating phases as obtained with CTQMC in Werner et al (2009), while the color-scale shows the calculated quasiparticle weight as obtained by the RISB method. The white regions correspond to insulating phases of integer occupancy (explicitly indicated for each case). The shape of the Mott insulating lobes can be qualitatively understood as follows. Increasing \( \mu \) from \( \mu = 0 \) for a fixed and large \( U/D \) value, e.g. 8, the density increases and the quasiparticle weight decreases until the system reaches the boundary of Mott insulating phase of occupancy \( n = 1 \). Further increasing \( \mu \) does not affect the density while \( \mu \) remains within the Mott-driven gap. When this limit is achieved, the system becomes metallic and as \( \mu \) increases \( Z \) first is enhanced and then reduced until the system reaches the insulating phase corresponding to \( n = 2 \).

The same evolution is repeated as the system goes through the insulating lobes of increasing density. The strong asymmetry between the insulating lobe of \( n = 3 \) and the others is due to the Hund’s coupling: for \( n = 3 \), \( J \) increases the gap for charge excitations, which makes the lobe wider in the \( \mu \)-direction, and it then reduces the critical value of \( U \) that drives the

![Figure 1. Zero crystal field splitting phase diagram in the chemical potential \( \mu \) versus local interaction \( U \) space for Hund’s coupling \( J = 0.167U \). The quasiparticle weight as obtained from RISB is presented using a color scale and white areas correspond to insulating phases (\( Z = 0 \)). The integer numbers in these areas indicate the occupancy of the corresponding insulating phase. The black triangles correspond to the metal-insulating phase boundaries as obtained with DMFT using CT-QMC, and are taken from Werner et al (2009).](image)

3. Results

We analyze first the effects of a tetragonal distortion which leads to \( \epsilon_1 = \epsilon_2 = 0 \) and \( \epsilon_3 = \Delta \). In section 3.1 we study the half-filled case while in section 3.2 we analyze arbitrary electronic densities. Last, in section 3.3 we study the \( n = 2 \) Hund’s metal under different crystals distortions.

3.1. Half-filled

We first consider the half-filled case with an orbital occupancy of 3 electrons per site. In the \( \Delta = 0 \) case the three orbitals have the same occupancy and the main effect of \( J > 0 \) is to shift the critical interaction \( U_c \) to lower values as the charge excitation gap increases to \( U + 2J \) (Haule and Kotliar 2009, de’Medici et al 2011, deMedici and Capone 2017, Isidori et al 2018). The transition is of the first order type with a finite jump in \( Z \) at the transition (see figure 2) (Faciò et al 2017). A finite \( J \) breaks the degeneracy of the atomic ground state favoring a \( S = 3/2 \) configuration with an occupancy of a single electron per orbital. The crystal field splitting \( \Delta > 0 \) makes it energetically unfavorable to occupy the higher energy orbital and produces (in the metallic phase) a charge transfer between the orbitals. The charge redistribution between orbitals is dominated by the ratio \( \Delta / J \). The crystal field splitting competes with \( J \) that favors an even distribution of the charge between the orbitals. This competition manifests itself in the behavior of the critical interaction \( U_c \) with \( J \) and \( \Delta \). While increasing
J leads to a larger charge excitation gap and to a reduction of $U_c$, increasing $\Delta$ for a fixed $J/\Delta$ produces an enhancement of $U_c$. A non-zero $\Delta$ does not change the nature of the transition but makes it concomitant with a sudden charge redistribution such that each orbital has an occupancy of one electron in the insulating phase. Although the charging of the different orbitals can be quite different close to the Mott transition, the quasiparticle weight does not show a significant orbital differentiation for the wide range of parameters analyzed.

### 3.2. Doped system

Huang et al (2012) analyzed the site occupation $n = 4$ and found a rich phase diagram as a function of the level splitting and the Coulomb interaction $U$, including Mott and orbital selective Mott phases. We extend these results to general doping levels and analyze the effect of the crystal field on the Hund’s metal phase.

To stabilize an orbital selective Mott phase we may introduce a hole or electron doping to the system. As it was shown for a two orbital system with nonzero Hund’s coupling and crystal field splitting (Werner and Millis 2007), for a range of doping values, the charge is incorporated to one of the orbitals making it metallic while the other remain insulating. This behavior is also observed for a three orbital system as it is shown in figure 3. The parameters are such that the system is in the Mott phase for the undoped ($n = 3$) case ($U = 1.86D$, $J = 0.25U$, and $\Delta = 0.25$, see figure 2). Electron doping increases the charge in orbitals 1 and 2 which become metallic, while the charge and the insulating nature of orbital 3 remain unchanged for a finite range of dopings, as it can be seen form the behavior of the corresponding quasiparticle weights (see figure 3(a)). On the contrary, upon hole doping orbital 3 decreases its charge and becomes metallic while orbitals 1 and 2 remain insulating with an occupancy $n_1 = n_2 = 1$. For large enough electron or hole doping, the three orbitals become metallic through an orbital selective Mott transition. As it can be seen in figure 3(c), the range of doping in which the orbital selective Mott phase is observed increases with increasing $U$.

The critical interaction for the OSMT is dominated by the occupancy of the different orbitals and at least one of the orbitals must have an integer occupancy in the OSMP. As in the half-filling case, the charge distribution between the orbitals depends on $\Delta$ and $J$. We analyze below the role of these parameters on the OSMT. We first focus on the electron doped case with an occupancy of 3.5 electrons per site. In this case, for $J \neq 0$, orbital 3 becomes insulating for $U > U_c$ while orbitals 1 and 2 remain metallic. In figure 4 we present the quasiparticle weight and the occupancy of orbital 3 as a function of $U$. For the values of $\Delta < 2D$ presented in the figure, there is a partial occupancy of orbital 3 even in the non-interacting limit as the bandwidth is larger than the energy shift between the band associated with orbital 3 and the bands associated with orbitals 1 and 2. As the interaction is increased the quasiparticle bandwidths ($Z_\alpha D$) decrease and $\Delta$ becomes more effective polarizing the charge. An increasing $U$ also leads to an increase in $J = 0.05U$ which favors an even distribution of the charge between orbitals. For $U \sim 0$ ($J \ll \Delta$) the former effect dominates the physics and $n_3$ decreases. For larger values of $U$ we obtain two different behaviors depending on the value of $\Delta$. For $\Delta \ll D$ there is a wide range of interaction parameters where the system presents a Hund’s metal behavior with the physics dominated by the $S = 3/2$ and $S = 1$ multiplets (see lower panels in figure 4). This regime is marked by an increasing effective mass differentiation between the orbitals with an increasing interaction $U$. For larger values of $\Delta$, the range of parameters where the
system is in this regime is reduced as the critical interaction for the OSMT decreases.

For hole doping, orbitals 1 and 2 become insulating at a critical interaction, while orbital 3 remains metallic. This is illustrated in figure 5 for an occupancy \( n = 2.5 \) and the other parameters as in figure 4. The overall behavior as a function of \( U \) is similar to the \( n = 3.5 \) case, the main difference being that the behavior of the quasiparticle masses is interchanged with \( Z_1 = Z_2 \) lower than \( Z_3 \) and vanishing at the OSMT.

The main results of this section are presented in figure 6 which shows the quasiparticle weight for orbital 3 and orbitals 1 and 2 in the regions of the \( n \) versus \( U/D \) plane where the system is in the metallic phase. The white area corresponds to an OSMP where only one of the bands is insulating (\( n < 3 \)), two bands are insulating (\( n > 3 \)) or to a Mott insulator where the three bands are insulating (integer fillings \( n = 2, n = 3, \) and \( n = 4 \)). Away from the \( |n - 3| \lesssim 0.3 \) cases the system presents a wide range of Coulomb interactions where one (or two) of the bands is strongly correlated. This regime correspond to the Hund’s metal phase where the physics is dominated by the high spin multiplets \( S = 3/2 \) and \( S = 1 \).

3.3. Hund’s metal versus crystal field distortions

Here we analyze how the Hund’s metal phase evolves as different crystal field distortions are enabled. We focus on the case \( n = 2 \) and consider the three possible distortions in a three-band model: one orbital energy pushed above or below the energy of a double-degenerated set of orbitals or the three orbitals split in energy. In the context of \( t_{2g} \) shells of transition metal oxides, the first two cases correspond to a tetragonal distortion whereas the third one to the orthorhombic case.

Figure 4. Upper panels: quasiparticle weight as a function of \( U/D \) for different crystal field splittings \( \Delta \) indicated in the figure, \( J/U = 0.05 \), and \( n = 3.5 \). Middle panels: orbital 3 occupancy \( n_3 \) versus \( U/D \). Lower panels: statistical weight of different atomic multiplets versus \( U/D \).

Figure 5. Upper panels: quasiparticle weight as a function of \( U/D \) for different crystal field splittings \( \Delta \) indicated in the figure, \( J/U = 0.05 \), and \( n = 2.5 \). Middle panels: orbital 1 occupancy \( n_1 \) versus \( U/D \). Lower panels: statistical weight of different atomic multiplets versus \( U/D \).

Figure 6. Top and middle panels: quasiparticle weights as function of \( U/D \) and \( n \). Lower panel: occupancy of orbital 3 relative to its value at zero crystal field. The other parameters are \( J/U = 0.25, \beta D = 400, \) and \( \Delta/D = 0.25 \).
In figure 7 we present the quasiparticle mass enhancement and the orbital occupancies for a three orbital system with a tetragonal distortion and two values of the $J/U$ ratio. For low $J$ values ($J/U = 0.05$), increasing the crystal field splitting $\Delta$ leads to a decrease in the critical interaction and to an increase in the mass enhancement differentiation. In the high $J/U = 0.25$ regime, the system has a much larger critical interaction $U_c(\Delta = 0)$ in the $\Delta = 0$ case and presents a wide range of Coulomb interactions $U$ where the quasiparticle mass is strongly renormalized even though $U \ll U_c(\Delta = 0)$. In this regime the system is strongly sensitive to changes in the crystal field splitting which leads to a strong orbital differentiation in the quasiparticle mass enhancement and to a significant reduction of the critical Coulomb interaction.

We now fix the interaction parameters to $U = 4J = 4D$, values which in the absence of crystal fields place the system in the Hund’s metal phase, and study the evolution of the system for different possible crystalline distortions presented above. Figures 8(a) and (b) shows for the different distortions the quasiparticle weight as a function of crystal field. In the three cases, as the energy splitting between orbitals is increased the quasiparticle weights differentiate becoming smaller for the orbitals whose occupancy approach 1. In the tetragonal case having as lowest energy orbitals the double-degenerated set (positive $\Delta$ in figure 8(a)), as $\Delta$ is increased the Hund’s metal evolves to a Mott insulator in which the third orbital has a vanishing occupancy. For the two other distortions the lowest energy orbital has only the spin-degeneracy and the Hund’s metal evolves first to an OSMP in which the lowest energy orbital becomes insulating while the other two remain metallic.

4. Summary and conclusions

In summary, we have analyzed the electronic properties of a three band Hubbard system under a crystal field that partially lifts the orbital degeneracies. We focused on the low energy properties and characterized the electronic correlations through the quasiparticle mass renormalization $Z_\alpha$ for each band. Away from half-filling ($|n - 3| \gtrsim 0.3$) the system presents Hund’s metal behavior, with the physics dominated by local high spin multiplets, for a wide range of interaction parameters.

We found a strong sensitivity of the Hund’s metal to crystal fields that lift the degeneracies of the bands. In this regime, the crystal fields produce a strong quasiparticle mass differentiation between the bands and reduce significantly the range of parameters in which the Hund’s metal regime is stable driving the system into a OSMP.

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