

High-spin to low-spin and orbital polarization transitions in multiorbital Mott systems

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(Dated: March 31, 2007)

We study the interplay of crystal field splitting and Hund coupling in a two-orbital model which captures the essential physics of systems with two holes in the e_g shell such as LaNiO_3 or La_2NiO_4 . We use single site dynamical mean field theory with a newly developed impurity solver which is able to access strong couplings and low temperatures. The fillings of the orbitals and the location of phase boundaries are computed as a function of Coulomb repulsion, exchange coupling and crystal field splitting. We find that the Hund coupling can drive the system into a novel Mott insulating phase with vanishing orbital susceptibility. Away from half-filling, a change in crystal field splitting can induce an orbital selective Mott transition.

PACS numbers: 71.10.Fd, 71.10.Fd, 71.28.+d, 71.30.+h

The Mott metal-insulator transition plays a fundamental role in electronic condensed matter physics [1]. However, most attention has focused on the one-orbital case, in part because of its presumed relevance to high temperature superconductivity [2] and in part because appropriate theoretical tools for the multiorbital case have until recently not been available. This focus on the one-band case is limiting: in most Mott systems more than one orbital is relevant and the orbitals are occupied by more than one electron or hole. Intuition gained from studies of single-orbital models may not necessarily carry over to the multi-orbital case [3, 4, 5]. A particularly interesting topic is “orbital disproportionation”, i.e. the response of a system to a crystal field splitting which acts to lift an orbital degeneracy. This is of interest in the context of orbital ordering [6], of the “orbital selective Mott transition” [7] and of the response of materials to lattice distortions [3, 5]. Weak coupling methods [3] have been used to show that exchange and pair hopping interactions act to suppress the response to a crystal field splitting, and some authors have studied the model without exchange and pair hopping terms [5], but a reliable extension of these results to the rotationally invariant model in the strong coupling regime has until recently not been possible. On a technical level, multiorbital models are more difficult to study both because of the larger number of degrees of freedom, and because the physically important exchange and pair-hopping terms are not easy to treat by standard Hubbard-Stratonovich methods [8].

Dynamical mean field theory (DMFT) provides a non-perturbative and computationally tractable framework to study correlation effects and has allowed insights into the Mott metal-insulator transition [9]. In its single site version, DMFT ignores the momentum dependence of the self-energy and reduces the original lattice problem to the self-consistent solution of a quantum impurity model given by the Hamiltonian $H_{\text{QI}} = H_{\text{loc}} + H_{\text{hyb}} + H_{\text{bath}}$. For multi-orbital models $H_{\text{loc}} = \sum_m \epsilon_m c_m^\dagger c_m + \sum_{j,k,l,m} U^{jklm} c_j^\dagger c_k^\dagger c_l^\dagger c_m$, where $m = (i, \sigma)$ denotes both orbital and spin indices, and U^{jklm} some

general four-fermion interaction. H_{hyb} and H_{bath} are the impurity-bath mixing and bath Hamiltonians, respectively. While the DMFT approximation simplifies the problem enormously (replacing a $3 + 1$ dimensional field theory by a quantum impurity model plus a self consistency condition), the extra complications associated with exchange couplings in multiorbital systems have until recently prohibited extensive numerical work. In Refs. [10, 11] we have introduced a continuous-time impurity solver which can handle the general interactions in H_{loc} . The method is based on a diagrammatic expansion of the partition function in the impurity-bath hybridization term H_{hyb} . In the models investigated so far (Kondo lattice, two- and three orbital models, 4-site cluster), this simulation approach does not encounter a sign problem and leads to lower perturbation orders at stronger interactions. Furthermore, because the local problem is solved exactly, the formalism of Ref. [11] can easily treat spin-exchange or pair-hopping terms and yields information about the relevance of each eigenstate.

In this paper we use the new method to investigate the interplay of crystal field splitting and exchange couplings in orbitally degenerate systems. We consider a two orbital model with identical band widths, which captures the essential physics of systems such as LaNiO_3 with two holes in the e_g band. The local Hamiltonian is

$$\begin{aligned} H_{\text{loc}} = & - \sum_{\alpha=1,2} \sum_{\sigma} \mu n_{\alpha,\sigma} + \sum_{\sigma} \Delta (n_{1,\sigma} - n_{2,\sigma}) \\ & + \sum_{\alpha=1,2} U n_{\alpha,\uparrow} n_{\alpha,\downarrow} + \sum_{\sigma} U' n_{1,\sigma} n_{2,-\sigma} \\ & + \sum_{\sigma} (U' - J) n_{1,\sigma} n_{2,\sigma} \\ & - J (\psi_{1,\downarrow}^\dagger \psi_{2,\uparrow}^\dagger \psi_{2,\downarrow} \psi_{1,\uparrow} + \psi_{2,\uparrow}^\dagger \psi_{2,\downarrow}^\dagger \psi_{1,\uparrow} \psi_{1,\downarrow} + h.c.), \quad (1) \end{aligned}$$

with μ the chemical potential, Δ the crystal field splitting, U the intra-orbital and U' the inter-orbital Coulomb interaction, and J the coefficient of the Hund coupling. We adopt the conventional choice of parameters, $U' = U - 2J$, which follows from symmetry considerations for

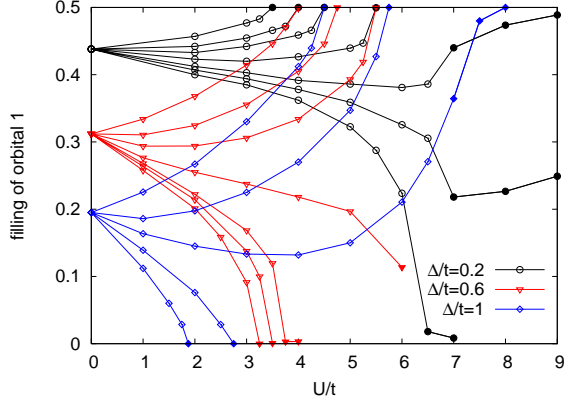


FIG. 1: Filling of orbital 1 as a function of U for $\Delta/t = 0.2, 0.6, 1$ and several values of J/U . The different curves for given Δ correspond (from bottom to top) to $J/U = 0, (0.01), (0.02), (0.05), (0.1), (0.15), (0.25)$, respectively. Open (full) symbols correspond to metallic (insulating) solutions. The metal-insulator transition is characterized by a jump in filling and a coexistence region where both insulating and metallic solutions exist. Our data show the region of stability of the metallic phase.

d -orbitals in free space and is also assumed to hold in solids. With this choice the Hamiltonian (1) is rotationally invariant in orbital space and the condition for half-filling becomes $\mu = \mu_{1/2} \equiv \frac{3}{2}U - \frac{5}{2}J$. In the DMFT self-consistency loop we use a semi-circular density of states of bandwidth $4t$ (Bethe lattice). The temperature, unless otherwise noted, is $T/t = 0.02$ and we suppress magnetic order by averaging over spin up and down in each orbital.

The main result is shown in Fig. 1, which for several values of Δ and J/U plots the filling per spin, n_1 , of orbital 1 as a function of interaction strength. The half filling, non-magnetic condition implies $n_2 = 1 - n_1$. In the $T \rightarrow 0$ limit, three phases are found: a metallic phase (which may have any value of n_1 between 0 and 0.5), an orbitally polarized insulator favored by large Δ and small J , and a Mott insulator (with $n_1 = 0.5 = n_2$) favored by large U , small Δ and large J . If U is increased from zero to a small value, the orbital splitting either increases (small J/U) or decreases (large J/U), consistent with the findings of Ref. [3]. As interaction strength is further increased, one of several things may happen: at very small J/U , n_1 continues to decrease, and the system eventually undergoes a transition to an orbitally polarized insulator (for large Δ essentially a band insulator). For somewhat larger J/U , the occupancy n_1 , after an initial decrease, goes through a minimum and begins to increase. At even stronger interactions, one then observes a transition either to an orbitally polarized insulator (where n_1 may take a range of values) or into a special type of insulator with $n_1 = 0.5$.

Figure 2 shows the metal-insulator phase diagram in

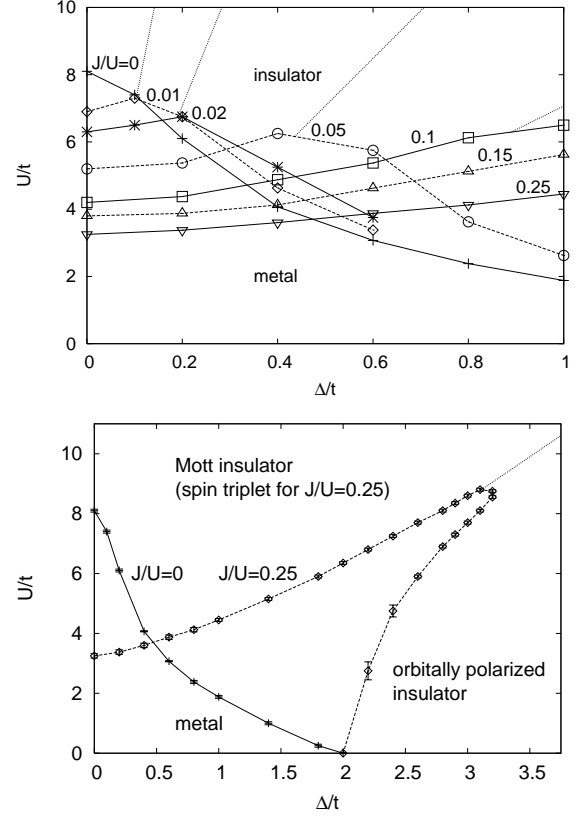


FIG. 2: Phase diagram in the plane of crystal field splitting Δ and intraorbital Coulomb repulsion U for indicated values of J/U . For $J = 0$ the phase boundary is a monotonic function of Δ , whereas for $J/U > 0$ it peaks near $\Delta = \sqrt{2}J$ (indicated by the dotted lines). The insulating state in the region $\Delta \lesssim \sqrt{2}J$ is characterized by a vanishing orbital susceptibility.

the space of crystal field splitting and Coulomb repulsion for several values of J/U . In the absence of a crystal field splitting ($\Delta = 0$), we observe a metal-insulator transition at a strongly J -dependent critical U . This finding is consistent with data presented in Ref. [12]. As Δ is increased, the critical U changes. For $J = 0$ and fixed U , n_1 decreases until the band is emptied and a metal-insulator transition occurs. The monotonic decrease of the critical U with Δ at $J = 0$ is a special case. For $J > 0$, the first effect of a small Δ is to stabilize the metallic phase. Then, at larger Δ , a reentrant insulating phase occurs. We shall show below that this behavior arises from the unusual nature of the insulating state at $J > 0$ and small Δ , which is characterized at $T = 0$ by a strictly vanishing orbital susceptibility. If Δ is increased at large U , this state makes a transition to an orbitally polarized insulator at $\Delta \approx \sqrt{2}J$. We therefore plot in Fig. 2 the curves $\Delta = \sqrt{2}J$ as dotted lines, and suggest that they correspond to the $T = 0$ phase boundary between two distinct insulating states.

Figure 3 plots the filling of orbital 1 as a function of

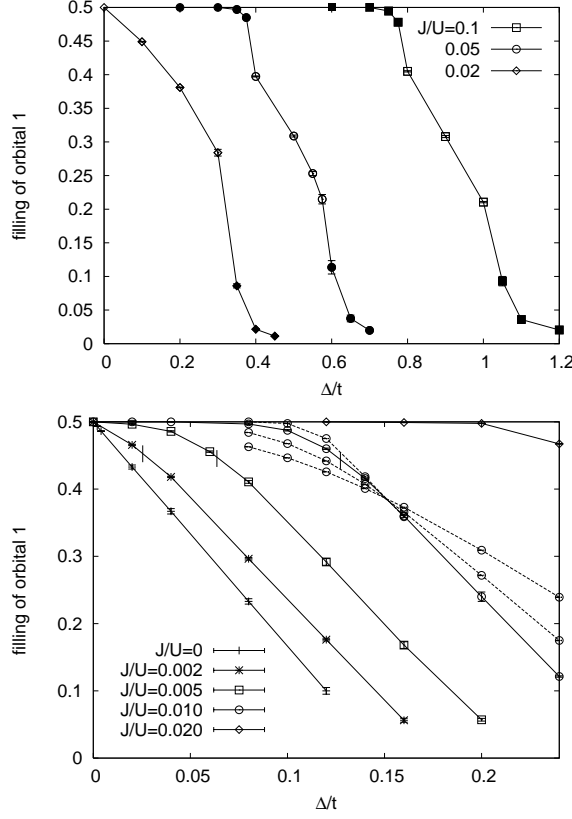


FIG. 3: Filling of orbital 1 as a function of Δ for fixed U and indicated values of J/U . Top panel: $U/t = 6$. Open (full) symbols correspond to metallic (insulating) solutions. Bottom panel: $U/t = 9$. Here, all solutions are insulating. For crystal field splittings smaller than $\Delta_c = \sqrt{2}J$ (indicated by a vertical line) the orbital susceptibility in the $T \rightarrow 0$ limit is completely suppressed. Solid lines are for $\beta t = 50$, dotted lines show results for $\beta t = 12.5, 25$ and 100 , respectively.

crystal field splitting for fixed U/t and several values of J/U . The leftmost curve in the upper panel shows the density variation for $J/U = 0.02$. At $\Delta = 0$, the model is metallic. The rapid variation of n_1 with Δ reflects the large, but finite orbital susceptibility of the metal, which for this small value of J is strongly enhanced by U . At $\Delta/t \approx 0.325 > \sqrt{2}J$, an apparently first order transition occurs to the orbitally polarized insulating state, which then evolves smoothly (as Δ is increased) to the band insulator ($n_1 \rightarrow 0$). The two larger J values reveal a different behavior. For $\Delta < \Delta_c \approx \sqrt{2}J$ the insulating state is characterized by an orbital occupancy which is independent of crystal field splitting. Then, an apparently discontinuous transition occurs to a metallic state with a large orbital susceptibility, which at even larger Δ exhibits a first order transition to the orbitally polarized insulating state. The lower panel of Fig. 3 shows the behavior for larger U , where the model is always insulating. Our data for $J = 0$ exhibit a rapid variation of n_1 with

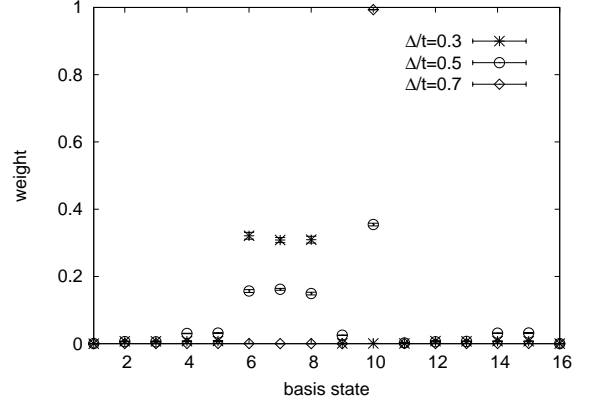


FIG. 4: Weight of the different eigenstates of H_{loc} for $U/t = 6$, $J/U = 0.05$ and $\Delta/t = 0.3, 0.5$ and 0.7 . The smallest crystal field splitting corresponds to an insulating state with suppressed orbital susceptibility, the intermediate value to a metallic state and the largest splitting to a “band insulator” (see Fig. 3).

Δ . The slope is set by the inverse of the Kugel-Khomskii superexchange $\sim t^2/U$; thermal effects are unimportant at $\beta t = 50$. For $J > 0$ and small Δ , the model is insulating, with a vanishing orbital susceptibility, then (near $\Delta = \sqrt{2}J$) makes a transition to the orbitally polarized insulating phase with a differential susceptibility $\partial n_1 / \partial \Delta$ determined by Kugel-Khomskii physics. Note that the transition between the two insulators is sharp only at $T = 0$; for $T > 0$ a rapid (but smooth) crossover occurs.

To gain insight into these phenomena, we look at the contribution to the partition function from the different eigenstates of the local Hamiltonian. H_{loc} has 16 eigenstates, which we number essentially as in Table II of Ref. [11]. For the following discussion it is important to note that $|6\rangle$, $|7\rangle$ and $|8\rangle$ are the three spin triplet states (with energy $U - 3J - 2\mu$), while $|10\rangle$ and $|11\rangle$ are linear combinations of the states $|\uparrow\downarrow, 0\rangle$ and $|0, \uparrow\downarrow\rangle$ with two electrons in one orbital and none in the other. The latter two states are coupled by the pair hopping and affected by the crystal field splitting. Here, we choose them to be eigenstates of H_{loc} corresponding to the eigenenergies $U \pm \sqrt{J^2 + 4\Delta^2} - 2\mu$: $(1 + \alpha_{\pm}^2)^{-1/2}(|\uparrow\downarrow, 0\rangle + \alpha_{\pm}|0, \uparrow\downarrow\rangle)$, $\alpha_{\pm} = \pm(\sqrt{J^2 + 4\Delta^2} \mp 2\Delta)/J$. In particular, we choose $|10\rangle$ to be the eigenstate with lower energy.

Figure 4 shows the weights of these states for the three phases found at $U/t = 6$, $J/U = 0.05$ (see Fig. 3). In the small- Δ phase, the triplet states are occupied, with small excursions into states with occupancy 1 or 3. We therefore call this phase the triplet Mott insulator. In the metallic phase, a large number of states is visited, while in the orbitally polarized insulator, the dominant local state (whose weight increases continuously with Δ) is a singlet ($|10\rangle$). The triplet states are almost completely suppressed in the orbitally polarized phase. The large- U

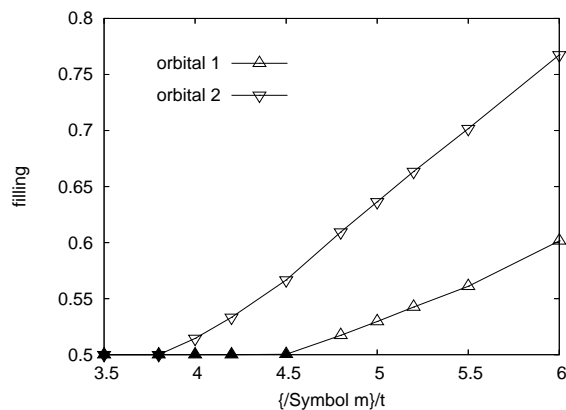


FIG. 5: Filling $n_1(\mu)$, $n_2(\mu)$ for $U/t = 4$, $J/U = 0.25$ and $\Delta/t = 0.4$. Full (open) symbols correspond to insulating (metallic) solutions. At half-filling ($\mu/t = 3.5$), the system is in a triplet Mott insulating state, for $3.9 \lesssim \mu/t \lesssim 4.6$ in an orbital selective Mott state, and for $\mu/t \gtrsim 4.6$ metallic in both bands.

insulator-insulator transition exhibits the same features, but without the intermediate metallic phase, and is therefore also a transition between high and low spin states. Comparison of the eigenenergies of the spin triplet states and $|10\rangle$ show that these levels cross at $\Delta = \sqrt{2}J$. Thus, the transition from triplet Mott insulator to orbitally polarized insulator occurs at $\Delta_c = \sqrt{2}J$, consistent with our numerical data. We also note that the wave-function of state $|10\rangle$ depends on the ratio J/Δ , leading in the large- Δ limit to $n_1(\Delta) \approx (J/4\Delta)^2$. In the low spin phase, the orbital susceptibility has therefore two contributions: one originating from Kugel-Khomskii physics and one of order J^2/Δ^3 from H_{loc} . The latter explains the roundings seen in the right most curve of Fig. 3.

We briefly address the issue of the orbital selective Mott transition, which provides a mechanism for local moment formation in correlated materials, and has been the subject of much recent debate [7]. Previous studies focused on two-orbital models with different band-widths and integer number of electrons. We find that in the presence of a crystal field splitting, shifting the chemical potential can drive the system into an orbital selective Mott state, even if the band-widths are equal. Figure 5 shows the filling per spin in both orbitals as a function of μ , for $U/t = 4$, $J/U = 0.25$ and $\Delta/t = 0.4$. Doping occurs first in one of the bands, leaving the other in a Mott state with a magnetic moment. Further change of the chemical potential drives the second band metallic.

In conclusion, we have shown that multiorbital impurity models with realistic couplings can be efficiently simulated with the method of Ref. [11]. We have presented numerical evidence, based on single site DMFT calculations, for the existence of two distinct Mott insulating phases in a half-filled two-orbital model with Hund cou-

pling and crystal field splitting. At strong interactions and $J < \sqrt{2}\Delta$, the system, in the $T \rightarrow 0$ limit, is in a phase characterized by a vanishing orbital susceptibility, and a spin 1 moment on each site. For $J > \sqrt{2}\Delta$ an orbitally polarized insulator is found. The exchange terms promote insulating behavior at $\Delta = 0$ but can stabilize a metallic phase at values of Δ for which the non-interacting model is a band-insulator.

It is interesting to compare our results to recent work on the bilayer Hubbard model [13, 14]. The model which these authors study is equivalent to our model with $U = U' = J$, and Δ replaced by the interlayer hopping. In the low energy sector of this model, only four states (essentially our three triplets and the pair hopping state $|10\rangle$) are relevant, and what these authors describe as the Mott insulator to band insulator crossover corresponds to our transition (apparently sharp at $T = 0$) between triplet Mott insulator and orbitally polarized insulator.

The existence of two distinct insulating phases raises many interesting questions including the theory of an insulator with strictly vanishing orbital susceptibility and the nature and properties of the different metal-insulator transitions. Our results may have important implications for the interpretation of properties of the pseudocubic and layered family of compounds based on LaNiO_3 .

The calculations have been performed on the Hreidar beowulf cluster at ETH Zürich, using the ALPS-library [15]. We thank M. Troyer for the generous allocation of computer time, A. Georges and A. Poteryaev for stimulating discussions and NSF-DMR-040135 for support.

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