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Orbital-selective Mott-Hubbard transition in the two-band Hubbard model

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Recent advances in the field of quantum Monte Carlo simulations for impurity problems allow—within dynamical mean field theory—for a more thorough investigation of the two-band Hubbard model with a narrow-wide band and SU(2)-symmetric Hund's exchange. The nature of this transition has been controversial, and we establish that an orbital-selective Mott-Hubbard transition exists. The wide band still shows metallic behavior after the narrow band became insulating—not a pseudogap as for an Ising Hund's exchange. The coexistence of two solutions with a metallic wide band and an insulating *or* metallic narrow band indicates, in general, first-order transitions.

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By virtue of dynamical mean field theory (DMFT),[1,2] our understanding of the Mott-Hubbard transition[3] in the one-band Hubbard model has greatly improved in the last years. The bandwidth-controlled Mott-Hubbard transition is, at least within DMFT,[2,4] of first order at low temperatures (T) and becomes a smooth crossover for temperatures above a critical point, which terminates the first-order line. A further complication arises exactly at zero temperature where two solutions coexist as for low Ts. But at $T=0$, the insulating solution is always higher in energy than the metallic one, i.e., the insulating solution is metastable throughout the whole coexistence region. The DMFT Mott-Hubbard transition is of second order at $T=0$ despite the coexistence of two solutions.

For making contact with experiments, orbital realism has to be taken into account, e.g., within the merger of local density approximation and DMFT (LDA+DMFT approach).[5] In the case of transition metal oxides, typically either the three t_{2g} or the two e_g bands cross the Fermi energy. At the very least, these orbitals should be included. For degenerate orbitals, the situation seems to be clear, at least within DMFT; there is a first-order Mott-Hubbard transition.[6] Most transition metal oxides are, however, noncubic. Hence, the orbital degeneracy is lifted. Take, for example, the unconventional superconductor Sr_2RuO_4 (Ref. 7), which has a wide d_{xy} band and narrow $d_{xz,zy}$ bands[8] and which becomes a Mott-Hubbard insulator upon substituting Sr by Ca.[9]

For such a situation with wide and narrow bands the details of the Mott-Hubbard transitions are so far inconclusive, even within DMFT and even for a simple two-band Hubbard model with Coulomb interaction U and Hund's exchange J between the two bands. Koga *et al.*[10] employed the so-called exact diagonalization (ED) method to solve the DMFT equations and obtain two Mott-Hubbard transitions: first the narrow band becomes insulating, then the wide band. This scenario has been coined orbital-selective Mott-Hubbard transition.[11] In contrast, Liebsch[12] uses quantum Monte Carlo (QMC) simulations and the iterated perturbation theory (IPT) to solve the DMFT equations and finds a single first-order Mott-Hubbard transition with similar changes in both bands. On the insulating side, the wide band has a pseudogap that gradually amplifies to a real gap with increasing U. In principle, the QMC is more suitable for addressing the Mott-Hubbard transition since ED only gives discrete peaks in the spectra, making it difficult to unambiguously identify a gap. However, the QMC simulations are restricted to relatively high temperatures and there is a sign problem[13] if the Hund's exchange coupling is taken into account in full, i.e., not only the Ising but also the spin-flip component. Since the same limitations as in Ref. 12 also apply to all previous LDA+DMFT(QMC) calculations,[5] there is an urgent need to clarify whether and how the details of the Mott-Hubbard transition are affected. Another important aspect is whether two solutions coexist. Liebsch finds two coexisting solutions at a single transition, while Koga *et al.*[10] do not address this question. If there was a first-order transition, two consecutive transitions might even be bridged into a single one.

In this paper, we study this transition by employing the most recent advances in the field of QMC simulations for DMFT. The recently introduced projective QMC (PQMC) method[14] enables us to address $T=0$. Furthermore, the Hubbard-Stratonovich decoupling of Ref. 15 allows for the calculation with the full SU(2)-symmetric Hund's exchange at a still-manageable sign problem, in particular in combination with PQMC.

Model. The starting point is the two-band Hubbard model

$$H = -\sum_{m=1}^{2} t_m \sum_{\langle i,j\rangle\sigma} \hat{c}^{\dagger}_{im\sigma}\hat{c}_{jm\sigma} + U\sum_{im\sigma} \hat{n}_{im\uparrow}\hat{n}_{im\downarrow} + \sum_{i;\sigma<\sigma'} (U' - \delta_{\sigma\sigma'}J)\hat{n}_{i1\sigma}\hat{n}_{i2\sigma'} \underbrace{- \frac{J}{2}\sum_{i\sigma;l\neq m} \hat{c}^{\dagger}_{il\sigma}\hat{c}_{il\bar{\sigma}}\hat{c}^{\dagger}_{im\bar{\sigma}}\hat{c}_{im\sigma} - \frac{J}{2}\sum_{i\sigma;l\neq m} \hat{c}^{\dagger}_{il\sigma}\hat{c}^{\dagger}_{il\bar{\sigma}}\hat{c}_{im\sigma}\hat{c}_{im\bar{\sigma}}}_{\equiv H_2}. \quad (1)$$

Here, $\hat{c}^{\dagger}_{im\sigma}$ and $\hat{c}_{im\sigma}$ are the creation and annihilation operators for electrons on site i within orbital m and with spin σ.

The first line describes the kinetic energy for which we employ the semielliptic noninteracting density of states $N^0(\varepsilon) = 8/(\pi W_m^2)\sqrt{(W_m/2)^2-\varepsilon^2}$ (orbital-dependent hopping amplitudes t_m on a Bethe lattice). For the following calculations, we use different widths for the two bands: $W_1=4$ for the wide and $W_2=2$ for the narrow band as in Refs. 10 and 12. Note that there is no hopping (hybridization) between the two bands. The second line describes the intraorbital (U) and interorbital (U') Coulomb interaction as well as the Ising component of the Hund's exchange J ($U'=U-2J$ by symmetry; we set $J=U/4$ as in Refs. 10 and 12). The third line consists of the spin-flip contribution to the Hund's exchange (yielding together with the second line an SU(2)-symmetric contribution that can also be written as $J\mathbf{S}_{i1}\mathbf{S}_{i2}$, where $\mathbf{S}_{im}$ denotes the spin for orbital m and site i). The last term represents a pair-hopping term of same strength J.

Method. QMC calculations that take the spin-flip component of Hund's exchange term into account have been a challenge. Although a straightforward Hubbard-Stratonovich decoupling, $\exp(J\Delta\tau c_1^\dagger c_2 c_3^\dagger c_4) = (1/2)\Sigma_{s=\pm1}\exp[s\sqrt{J\Delta\tau}(c_1^\dagger c_2 - c_3^\dagger c_4)]$, is possible, it has been recognized that it leads to a serious sign problem.[13] Therefore, it was neglected in almost every DMFT(QMC) calculation so far, including Ref. 12.

To overcome this problem, several attempts have been made.[15–17] Among these, Sakai, Arita, and Aoki proposed a discrete transformation for the spin-flip contribution of the exchange and pair-hopping term:[15]

$$e^{-\Delta\tau H_2} = \frac{1}{2}\sum_{r=\pm1} e^{\lambda r(f_\uparrow - f_\downarrow)} e^{a(N_\uparrow+N_\downarrow)+bN_\uparrow N_\downarrow}. \tag{2}$$

Here, $\lambda \equiv \frac{1}{2}\ln(e^{2J\Delta\tau}+\sqrt{e^{4J\Delta\tau}-1})$, $a \equiv -\ln[\cosh(\lambda)]$, $b \equiv \ln[\cosh(J\Delta\tau)]$, $f_\sigma \equiv c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma}$, $N_\sigma \equiv n_{1\sigma}+n_{2\sigma} - 2n_{1\sigma}n_{2\sigma}$. The advantage of this decoupling is that the auxiliary field r is real in contrast to that of Ref. 16. Hence, it is expected to yield better statistics in general.[15]

However, even with this decoupling, we note that the usual Hirsch-Fye QMC algorithm[18] does not work very well for DMFT calculation in the strong coupling regime or at low temperatures. For instance, for Hamiltonian (1) and $J=U/4$, we found it to be infeasible to obtain a self-consistent DMFT solution for $U>2.2$ when $\beta(=1/T)>50$ because the Green's function $G(\tau)$ has a large statistical error at $\tau \sim \beta/2$. Therefore, it is difficult to clarify without ambiguity whether an orbital selective Mott transition indeed occurs in multiorbital systems at low T by means of finite-temperature Hirsch-Fye QMC calculations; see also Ref. 19.

Another recent advancement was the development of a projective QMC (PQMC) algorithm by Feldbacher, Held, and Assaad.[14] In this algorithm, ground state expectation values $\langle\Psi_0|\mathcal{O}|\Psi_0\rangle/\langle\Psi_0|\Psi_0\rangle$ of an arbitrary operator $\mathcal{O}$ are calculated as

$$\langle\mathcal{O}\rangle_0 = \lim_{\theta\to\infty}\lim_{\tilde{\beta}\to\infty}\frac{\mathrm{Tr}[e^{-\tilde{\beta}H_T}e^{-\theta H/2}\mathcal{O}e^{-\theta H/2}]}{\mathrm{Tr}[e^{-\tilde{\beta}H_T}e^{-\theta H}]}, \tag{3}$$

where H_T is an auxiliary Hamiltonian [its ground state $|\Psi_T\rangle$ is the trial wave function that is assumed to be nonorthogonal to the ground state $|\Psi_0\rangle$ of H (Ref. 14)].

For H_T, it is convenient to take the one-body part of the Hamiltonian, because the limit $\tilde{\beta}\to\infty$ can be taken analytically in this case. Then, the starting point is the zero-temperature noninteracting Green's function $G_0(\tau,\tau')$ truncated to $0\leqslant\tau,\tau'\leqslant\theta$ and discretized as an $L\times L$ matrix ($L=\theta/\Delta\tau$). From this $G_0(\tau,\tau')$, the zero-temperature interacting Green's function $G(\tau,\tau')$ is obtained via the same updating equations for the auxiliary Hubbard-Stratonovich fields as for the finite-temperature Hirsch-Fye algorithm.

While PQMC gives $G(\tau)$ only for a limited number of (not too large) τ points, we need $G(i\omega)$ to solve the DMFT self-consistency cycle. To this end, the maximum entropy method (MEM) is employed to yield the spectral function $A(\omega)$ that allows for calculating $G(i\omega_n)=\int d\omega[A(\omega)/(i\omega_n-\omega)]$ at any frequency $i\omega_n$. This makes a crucial difference to finite-temperature calculations. The large statistical errors occurring at $\tau\sim\beta/2$ for finite temperatures now occur for rather large τ's. But even if there is a large statistical error for larger τ's, the maximum entropy method can extract sufficient information from the first τ points, discarding the larger τ's with excessive statistical error.

One of the main advantages of the PQMC method is that the convergence with respect to θ is much faster than that with respect to β in the Hirsch-Fye algorithm.[14] (Note that the calculation time increases cubically for θ and β.) Hence, we take in the following PQMC calculations a finite $\theta=20$ ($L=64$), which should be sufficiently close to the $T=0$ result: quantitatively small deviations are expected for larger θ's; qualitatively the behavior should not change anymore as in Ref. 14. Similarly as in Ref. 14, the central $\mathcal{L}=20$ are for measurement and $\mathcal{P}=(L-\mathcal{L})/2=22$ time slices on the right and left side of the measuring interval for projection. Typically, we performed 7×10^6 to 3×10^7 QMC sweeps.

Results. An indicator for the Mott-Hubbard transition is the quasiparticle weight Z, which is plotted in Fig. 1(a). We clearly see that for the narrow band $Z=0$ for $U\geqslant2.6$, while Z is still finite for the wide band. This means that there is a first Mott-Hubbard transition in which only the narrow band becomes insulating at $U\approx2.5$. This is consistent with the result of the DMFT(ED) calculation of Ref. 10, in which the critical U_c is estimated to be about 2.6. In contrast, there is a single first-order Mott-Hubbard transition at a smaller value $U_c\approx2.1$ if only the Ising component of Hund's exchange is taken into account (at $T=0.03$; between $U_c\approx1.8$ and 2.1 there are two coexisting solutions (hysteresis)).[12] In our DMFT(PQMC) results, the wide band is still metallic at $U=2.7$. But eventually, also the wide band has to become insulating at larger Coulomb interactions, since in the atomic limit both bands are insulating. (The calculation for larger Coulomb interactions unfortunately became computationally too expensive as even in the PQMC the statistical error brought about by the spin flip term of Hund's exchange increases dramatically.) Nonetheless, we can conclude from the data available that there are two different Mott-Hubbard transitions in which first the narrow and then the wide band become insulating. We have an orbital-selective Mott-Hubbard transition.

In Fig. 1(b), the double occupancy $D=\langle n_\uparrow n_\downarrow\rangle$ for the two different bands is plotted as a function of U. We see that for

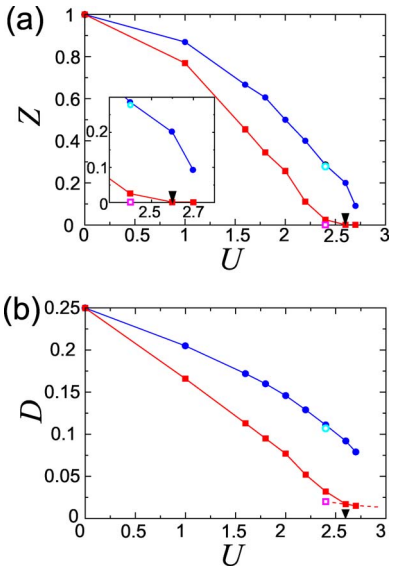

FIG. 1. (Color online) (a) Quasiparticle weight Z and (b) double occupancy D as a function of $U(J=U/4)$. Red (blue) squares (circles) are the data for the narrow (wide) band. For $U=2.4$, two solutions are found: The wide band is metallic for both solutions whereas the narrow band is metallic (closed symbols) *or* insulating (open symbols). The solid triangle in (a) and (b) is the U_c estimate from DMFT(ED) (Ref. 10); the inset enlarges the behavior around the transition.

the narrow band $D\approx 0.01$ for $U\geqslant 2.6$. A similar value of $D\approx 0.01$ was reported [4] for the one-band Hubbard model above the Mott-Hubbard transition, i.e., for $U/W\geqslant 5.9/4$. This suggests a Mott-Hubbard transition very similar to the one-band Hubbard model, albeit only for the narrow band.

Final evidence for the orbital-selective Mott-Hubbard transition is obtained from the spectral functions shown in Fig. 2: We can unambiguously say that the wide band is still metallic at $U=2.6$, whereas the narrow band is already insulating with a pronounced gap. While the wide band shows a pseudogap for an Ising type of Hund's exchange,[12] our SU(2) symmetric result reveals a peak in Fig. 2.

Let us now study the possibility of first-order Mott-Hubbard transitions. The first question is whether at $U=2.6$ (where we find a metallic wide and insulating narrow band) a second solution in which both bands are insulating (co)exists. Starting the DMFT self-consistency cycle with an insulating self-energy for the wide band, we obtain, however, the

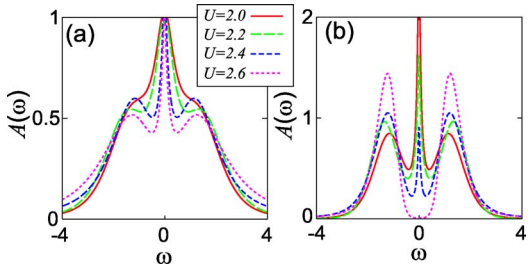

FIG. 2. (Color online) Spectral functions $A(\omega)$ for (a) the wide band and (b) the narrow band. For $U=2.6$, the narrow band is insulating while the wide band is metallic.

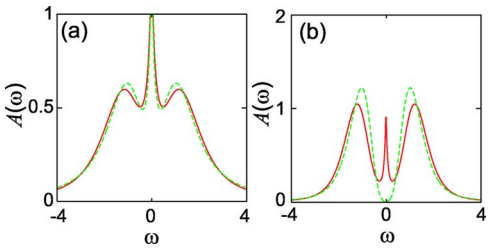

FIG. 3. (Color online) Spectral functions $A(\omega)$ for (a) the wide and (b) the narrow band at $U=2.4$. Two solutions with insulating or metallic narrow bands coexist.

very same (single) solution as in Figs. 1 and 2. This demonstrates that the orbital-selective Mott-Hubbard transition is not merged into a single first-order transition. There are two distinct Mott-Hubbard transitions.

The second question is, are the orbital-selective Mott-Hubbard transitions (generally) of first order? In this case, two solutions should coexist for $U\lesssim 2.6$: one with a metallic *and* one with an insulating narrow band. Special care is necessary for insulating solutions in the PQMC with a very narrow charge gap. For such small charge gaps, θ might not be sufficiently large to project, via $e^{-\theta H/2}$, from the metallic trial wave function onto the insulating ground state solution. We then note systematic errors even for intermediate τ's, and substantial noise appears in the charge gap of the spectrum calculated with the maximum entropy method. This makes the stabilization of a small-gap insulating solution delicate. This problem can be mitigated, however, by doing the maximum entropy calculation with a reduced number of τ points. Therefore, we used τ points up to $\tau_c\sim 3.2$ and ~ 1.6 for the following results.

For almost all values of U, only a metallic or only an insulating solution is obtained for both $\tau_c\sim 3.2$ and ~ 1.6. However, for $U=2.4$, we find both a solution with a metallic and with an insulating narrow band (the wide band is metallic in both solutions with only minor differences). In Fig. 3, the spectral function of these two solutions are shown; the value of Z and D for the insulating solution is plotted in Fig. 1 as open circles and squares. The DMFT(PQMC) data suggest that two solutions with metallic and insulating narrow band coexists for $U\sim 2.4$, so that the Mott-Hubbard transition in which the narrow band becomes insulating (and in which the wide band stays metallic) is in general of first order. Possibly, the insulating solution is metastable at $T=0$.

Discussion. For understanding the DMFT results it is instructive to remind ourselves of what is known for the two-orbital Anderson impurity model (AIM). If $J>T_K$ (the AIM Kondo temperature) the impurity spins of the two orbitals form a steadfast spin-1 (triplet). For such an AIM and inequivalent orbitals it is known that this spin-1 is screened in two stages: first only by one orbital to a spin-$\frac{1}{2}$ at T_K^1, and then by the second orbital to a spin-0 at T_K^2.[20] Within DMFT we now have to solve AIMs self-consistently: The AIM's T_K's of one DMFT iteration (crudely $T_K\approx ZW$) sets the hybridization strength for the next DMFT iteration. Hence, we can interpret our DMFT results as following: Given the two inequivalent Kondo scales of the AIM, there is a U interval where only the hybridization strength (and T_K) of the narrow

orbital is renormalized to zero by the DMFT iterations. Only the narrow band is insulating.

If only the Ising component of Hund's exchange is taken into account, the behavior of the AIM is completely different. Instead of a triplet, the impurity spins align to $S_Z=\pm 1$ (no $S_Z=0$ component). For $J>T_K$ ($J\approx 0.5$ and $T_K\approx ZW \approx 0.4$ at the Mott-Hubbard transition of Ref. 12), there is no spin Kondo effect any more since it requires a spin flip of the conduction electrons and, hence, a *change* of S_Z by ± 1. As soon as one orbital becomes insulating, there is also no orbital Kondo effect anymore: the whole system is unscreened, i.e., insulating. It is certainly interesting to study whether this kind of physics is relevant for magnetically anisotropic materials.

Taking the full SU(2) symmetry of Hund's exchange into account in the PQMC calculation, we conclude that there are two consecutive Mott-Hubbard transitions, whereby—at least around the first transition—two solutions coexist. By clarifying the theoretical side, we hope to stimulate further experiments on the orbital-selective Mott-Hubbard transition, e.g., in Sr_2RuO_4 where results were so-far negative in this respect [21].

Note added. Recently, we learned about several related studies.[19,22]

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