Mott transitions in correlated electron systems with orbital degrees of freedom

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Mott metal-insulator transitions in an $M$-fold orbitally degenerate Hubbard model are studied by means of a generalization of the linearized dynamical mean-field theory. The method allows for an efficient and reliable determination of the critical interaction $U_c$ for any integer filling $n$ and different $M$ at zero temperature. For half-filling a linear dependence of $U_c$ on $M$ is found. Inclusion of the (full) Hund’s rule exchange $J$ results in a strong reduction of $U_c$. The transition turns out to change qualitatively from continuous for $J = 0$ to discontinuous for any finite $J$. This result is confirmed using the exact-diagonalization method for $M = 2$.

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I. INTRODUCTION

The Mott metal-insulator transition (MIT) has been the subject of numerous experimental and theoretical investigations over the past decades1–3. As a transition from a paramagnetic metal to a paramagnetic insulator that is driven by the competition between the conduction electrons’ kinetic energy and their mutual Coulomb repulsion, the Mott MIT represents a prime example for a quantum phase transition. Realizations of the Mott transition can be found in different 3d transition-metal oxides1,2 as well as in alkali-doped Fullerides4,5, for example. Its description is a challenging task for many-body theory.

The minimal model to describe the Mott transition is the Hubbard model6 which in its simplest form contains the local Coulomb interaction between the electrons in a conduction band formed by a single orbital only. The investigation of such a single-band model might be justified in some cases, in particular as an effective model for low-energy properties. A consistent description of the experimental observations for the above-mentioned systems, however, certainly requires the use of more realistic models which include orbital degrees of freedom, possible crystal field splittings, etc.

Theoretical advances in the past decade – mainly due to the development of the dynamical mean-field theory (DMFT)7,8 – have led to an increased understanding of multi-orbital Hubbard models. Recent investigations have either concentrated on fundamental questions such as the nature of the Mott MIT as a function of orbital degeneracy9–16 or, within the so-called LDA+DMFT approach17, on a realistic description of the transition.

Nevertheless, there are still no reliable DMFT results for the zero-temperature ($T = 0$) Mott transition in general multi-band models: Numerical methods to solve the DMFT equations are either restricted to fairly large temperatures (quantum Monte-Carlo approach, QMC) or have not yet been extended to the multi-band case (numerical renormalization-group method, NRG). Additionally, due to the sign problem within the QMC approach only a simplified exchange part of the Coulomb interaction can be taken into account13,18. The exact-diagonalization method (ED) is restricted to rather small system sizes, and data are available for $M \leq 2$ orbitals only16. The $M$ dependence of the critical interaction strength $U_c$ for different integer fillings $n$ is particularly interesting. Detailed results for $U_c$ as a function of $M$ and $n$ are available from the Gutzwiller approximation and from a slave-boson approach20,11. Numerical DMFT-QMC calculations have been performed for $M \leq 3$12,13. Remarkably, in the limit of large orbital degeneracy $M \to \infty$ an analytical treatment of the DMFT becomes possible for the MIT15. A scaling $U_c = U_{c,2} \propto M$ for the actual transition is found while $U_{c,1} \propto \sqrt{M}$ is obtained for the critical interaction where the insulating solution breaks down15. This is consistent with the linear dependence for large $M$ found in Refs. 9, 11 and with the square-root dependence reported in Ref. 5. However, there is still a need for a DMFT method which works at $T = 0$ and which allows for an efficient and reliable determination of $U_c$ for arbitrary $n$ and $M$ and for a model including the full exchange interaction.

Here we present and apply a proper multi-band generalization of the so-called linearized DMFT (L-DMFT)19 which is a simple but rather successful technique that originally was developed for the critical regime of the Mott MIT in the single-band model. Within the L-DMFT the lattice problem is mapped onto an Anderson impurity model with a single bath site only by considering a simplified self-consistency condition just at the critical point. The approach can be considered to be the simplest non-trivial variant of the more systematic projective self-consistent method (PSCM) by Moeller et al20. It allows for extremely fast numerical calculations or even analytical results to estimate the critical parameters. Extensions have been considered for the (Mott-Hubbard or charge-transfer) MITs in a $d$-$p$ model21, for a quantum critical point in the periodic Anderson model22, and for...
the $M = 2$ degenerate Hubbard model at half-filling and 
$J = 0$. Furthermore, a general two-site DMFT has 
been developed which reduces to the L-DMFT at the 
MIT but is not restricted to the critical point. An applica-
tion to multi-band systems is likewise possible but has 
not yet been considered.

As concerns effective single-band models, the linearized 
DMFT has been tested extensively by comparing with 
numerical results from the full DMFT. For the standard 
Hubbard model but also for different thin-film and 
semi-infinite surface geometries as well as for the $d-p$ 
multi-band model a remarkable agreement has been found: 
Detailed trends of $U_c$ as a function of electronic model 
parameters and as a function of parameters characterizing 
the lattice geometry are predicted reliably. For quantita-
tive estimates errors of the order of a few percent have 
to be tolerated.

As it is demonstrated here, a straightforward extension of 
the method to multi-band systems is possible. We ex-
pect the L-DMFT to be a fast but reliable tool in this 
case as well since the effective impurity model includes 
the complete atomic part and the self-consistently de-
termined bath with the complete degeneracy (although 
restricted to a single site only). Furthermore, the reliabil-
ity of the L-DMFT for the multi-band case is checked 
by comparing with different fully numerical DMFT tech-
niques whenever results are available. All DMFT calcula-
tions discussed in this paper were performed on a Bethe 
lattice with $t^2 = 1$, corresponding to bandwidth of 4.

II. MODEL AND LINEARIZED DMFT

We consider a multi-band Hubbard-type model $H = 
H_0 + H_1$ including an intra-orbital hopping

$$H_0 = \sum_{ij\sigma} t_{ij} \epsilon_i \epsilon_j c_i^{\dagger} c_j \sigma$$

and the direct and the exchange part of the on-site 
Coulomb interaction:

$$H_1 = \frac{1}{2} \sum_{ia\sigma'} U_{a\sigma'} c_{ia\sigma} \epsilon_i c_{ia\sigma'} ^* c_{ia\sigma'} ^* c_{ia\sigma} + \frac{1}{2} \sum_{ia\sigma' \sigma''} J_{a\sigma'} c_{ia\sigma} \epsilon_i c_{ia\sigma'} ^* c_{ia\sigma''} ^* c_{ia\sigma''}$$

using standard notations. $i$ is a site index, and $\sigma = \uparrow, \downarrow$ 
refers to the spin direction. The different orbitals la-
beled by $\alpha = 1, ..., M$ are considered to be equivalent, 
$M$ is the orbital degeneracy. Exploiting atomic symme-
tries, we have $U_{a\sigma'} = (U + 2J) \delta_{a\sigma'} + U (1 - \delta_{a\sigma'})$ and 
$J_{a\sigma'} = J (1 - \delta_{a\sigma'})$ as usual (see e.g. Ref. 27). Opposed to 
calculations using the QMC method (see, e.g. Refs. 13, 
18), $H_1$ includes the full exchange part and thus preserves 
rotational invariance.

The DMFT assumes the self-energy to be local. Fur-
thermore, the self-energy as well as the Green function 
are diagonal with respect to the orbital index $\alpha$ for the 
model considered here. In case of equivalent orbitals and 
in the absence of any spontaneous symmetry breaking, we 
thus have $\Sigma_{ij\alpha\beta}(\omega) = \delta_{ij} \delta_{\alpha\beta} \Sigma(\omega)$ and likewise for the 
local Green function: $G_{i\alpha\beta}(\omega) = \delta_{\alpha\beta} G(\omega)$. Within the 
framework of the DMFT, the model $H$ is mapped onto 
an impurity model $H' = H_0' + H_1'$ where $H_1'$ is the local 
part of the interaction (2) at a distinguished site $i_0$ and 

$$H_0' = \sum_{\alpha\sigma} t_{i_0\alpha\sigma} \epsilon_{i_0\alpha\sigma} c_{i_0\alpha\sigma} + \sum_{\alpha\sigma, k=2}^n \epsilon_k a_{k\alpha\sigma} ^* a_{k\alpha\sigma} + \sum_{\alpha\sigma, k=2}^n V_k (\epsilon_j \delta_{i_0\alpha\sigma} a_{k\alpha\sigma} + \text{h.c.})$$

with $t_{0} = t_{ij}$. The self-energy $\Sigma(\omega)$ of $H'$ is identified 
with the self-energy of the lattice model $H$ and yields, 
via the lattice Dyson equation, the on-site Green func-
tion $G(\omega)$. The latter determines the one-particle pa-
rameters of $H'$ or, equivalently, the hybridization func-
tion $\Delta(\omega) = \sum_k V_k^2 / (\omega + \mu - e_k)$ via the DMFT self-
consistency condition $\Delta(\omega) = \omega + \mu - \Sigma(\omega) - 1 / G(\omega)$.

Within the full DMFT an infinite number of bath de-
grees of freedom $n_s \rightarrow \infty$ are necessary to fulfill the self-
consistency equation. This implies the need for further 
approximations to treat $H'$. Contrary, a simple two-site 
model is considered here. For $n_s = 2$ the model (3) can 
be solved exactly; however, a simplified self-consistency 
condition must be tolerated.

Our approach rests on the assumption that similar as 
for the single-band model, the MIT is characterized by 
a vanishing weight of a low-energy quasi-particle reso-
nance. Close to the MIT it is then plausible to neglect 
any internal structure of the quasi-particle peak and to 
assume that there is no influence of high-energy features 
on the low-energy part of the excitation spectrum. The 
quasi-particle peak is then approximated by a single pole 
at the Fermi energy ($\epsilon_c \equiv \epsilon_{k_F} = \mu$) which reproduces 
itself in the DMFT self-consistency cycle. One can pro-
cess as for the single-band model and is finally left with 
the following simple algebraic self-consistency equation 
which determines the critical parameters ($V \equiv V_{k=2}$):

$$V^2 = z M_{2}^{(0)}.$$
solving $F$ minimization of the function with results from ED calculations which we have performed with results from numerical methods to solve the full For $U > U_c(n, M)$ and $\mu_- < U < \mu_+$ the system is a Mott insulator with integer filling $n$. On the other hand, for $\mu < \mu_-(U)$ or $\mu > \mu_+(U)$, a non-integer filling is realized, and the system becomes metallic. The chemical potential is a continuous function of the filling $\mu$, and the system becomes metallic for $\mu < \mu_-(U)$ or $\mu > \mu_+(U)$. The chemical potential is fixed to $\mu = (M - \frac{1}{2})U$ due to particle-hole symmetry. Then we find $F = M_2^{(0)}(4M + 2)^2/U^2$ which results in the critical interaction

$$U_c(n = M, M) = (4M + 2)\sqrt{M_2^{(0)}}.$$ \hfill (6)

By solving $F(\mu, U) = 1$ with Eq. (5) for $U > U_c$ we obtain the jump in the chemical potential

$$\Delta \mu \equiv \mu_+ - \mu_- = U \sqrt{B - \sqrt{B^2 - 1 + \frac{U_c^2}{U^2}}} \hfill (7)$$

where $B = 1 + 1/(2(2M + 1)^2(U/U_c)^2)$. The result for $\Delta \mu(U)$ is shown in Fig. 1 for $M = 1, \ldots, 5$. Here and hereafter we set $M_2^{(0)} = 1$. For fixed $M$ and $U \to \infty$, the jump in the chemical potential increases linearly with $U$. For $U \to U_c$, $\Delta \mu(U)$ shows a square-root behaviour.

Qualitatively and even quantitatively, this agrees well with results from numerical methods to solve the full DMFT equations. In particular, we compare with results from ED calculations which we have performed with $n_s = 6$ for $M = 2$ (circles) and with data available for $T = 0$ from the projective self-consistent method (crosses). The non-degenerate case $M = 1$ has already been discussed in Ref. 24. For $M = 2$ and $n_s = 6$ the error due to finite-size effects in the ED is almost negligible. One can state that the agreement of the L-DMFT results with the ED and PSCM data is equally good for both $M = 1$ and $M = 2$.

The generalized L-DMFT predicts $U_c(n = M, M)$ to depend linearly on $M$, see Eq. (6). This agrees with previous results from the Gutzwiller method, the non-crossing approximation for low temperature, and the projective technique for large $M$. This finding can easily be understood by looking at the weight factors in the Lehmann representation of the spectral density of the two-site Anderson model. Expanding into the different configurations, one finds that the ground state is composed of the order of $M$ singlet states in the limit $V \to 0$ and that there are of the order of $M$ excited states coupling to the ground state. Hence, there are of the order of $M^2$ processes to be considered in the perturbation theory which are contributing to the spectral weight near $\omega = 0$. This implies $z \propto M^2V^2/U^2$ and eventually results in $U_c \propto M$.

In Fig. 2 the dependence of $U_c$ on both $n$ and $M$ is shown for all integer fillings and $M \leq 5$. We find $U_c$ to be reduced away from half-filling, consistent with previous results. The fit shows an almost perfect quadratic dependence of $U_c$ on the filling.

Let us now discuss the influence of the Hund’s rule coupling $J$ on the Mott transition at $T = 0$. Contrary to the QMC method, the full exchange part of the onsite Coulomb interaction can easily be included in the L-DMFT calculation. A quantitative influence, i.e. a reduction of $U_c$ due to $J$ has already been observed in Refs. 5, 13. This behavior is also evident from our L-DMFT results, as shown below.

The situation is, however, more complicated: It turns out that a finite $J$ leads to a qualitative change of the nature of the $T = 0$ Mott transition. This is illustrated in Fig. 3 which shows the $U$-dependence of the quasiparticle weight $z$ for various values of $J$ and $n = M = 2$. Note that $z$ is plotted not only close to the transition but also for $U$ far below $U_c$ which means that the self-
FIG. 2. Critical interaction $U_c$ for different fillings $n = 1, 2, ..., 2M - 1$ and different $M$ as a function of $n/M$. Symbols: L-DMFT results. Lines: simple fit function $U_c(n, M) = U_c(n = M, M) - c(1 - (n/M))^2$ and $c = -0.217 + 2.671M$.

consistency relation (4) is used for any $U$. This is reasonable as long as there is a clear separation between the low-energy (quasi-particle) and the high-energy scale (Hubbard bands). Although the approximation becomes questionable for higher $z$ and particularly in the limit $z \to 1$, the trend of $z(U)$ looks rather plausible in whole range.

In the critical regime where $z$ goes to zero continuously, we find $z(U)$ to decrease with decreasing $U$ which seems to be unphysical. In fact, this behavior belongs to a metastable state (dashed line) while for the true ground state (solid line) $z(U)$ is always decreasing with increasing $U$. The two metallic states with finite $z > 0$ but different slope $dz/dU$ are coexisting in a finite range of interaction strengths, namely for $U$ larger than $U_{c,2}$ (given by $z = 0$) and for $U$ smaller than another critical value $U_{c,3}$ where they merge together.

The actual critical interaction strength $U_c$ where the transition to the insulator takes place must be determined by comparing the respective energies with the energy of the insulator. This is shown in Fig. 4 for $J = 0.1$. For any $J > 0$ it is found that $U_{c,2} < U_c < U_{c,3}$. At $U_c$ the energies of the metallic and insulating states are crossing as functions of $U$. Consequently, the transition at $T = 0$ is discontinuous with a jump in physical quantities like the double occupancy $\langle n_\uparrow n_\downarrow \rangle = dE/dU$. Equivalently, one can look at the local magnetic moment at the impurity site $\langle S^2 \rangle$ (see Fig. 5). The local moment increases with $U$, typical for a strongly correlated normal metal. At $U_c$ it jumps to $\langle S^2 \rangle = 2$ for any finite $J$. Similarly, the quasi-particle weight $z$ decreases with increasing $U$ and drops to zero at $U_c$ from a finite value $z > 0$. In Fig. 3 this is indicated by solid vertical lines.

The phase diagram in the $(U, J)$-plane is shown in Fig. 6. The critical interaction for the actual transition is...
plotted together with $U_{c,2}$ and $U_{c,3}$. $U_c$ is found to be very close to $U_{c,3}$ for all $J$-values. Note that already a moderate value of $J \approx 0.3$ leads to a drastic reduction of $U_c$ from $U_c = 10$ to $U_c \approx 5$.

Interestingly, similar results have been obtained beforehand by a rather different method: Applying a generalized Gutzwiller approximation to a degenerate two-band model, Bünemann et al. found the Mott transition to be discontinuous for any finite $J > 0$ and to be continuous for $J = 0$ only. While the Gutzwiller method (for $J = 0$) significantly overestimates the critical interaction strength as compared to the L-DMFT, the reduction of $U_c$ by a finite $J$ is stronger. Common to both methods, however, is the qualitative change of the Mott transition for a finite $J$.

We have additionally checked the nature of the Mott transition by a fully numerical evaluation of the DMFT equations using the ED method with $n_s = 6$ sites for $n = M = 2$. The results for $J = 0$ and $J = 0.1$ are shown in Fig. 7. The critical interaction $U_c$ is found to be somewhat smaller as compared to the L-DMFT result as shown in Fig. 6. More important, however, the ED calculations show the same qualitative trend: The slope of $z(U)$ approaches $-\infty$ at a critical value $U = U_{c,3}$. This clearly indicates a second metastable metallic solution and a discontinuous transition and thus corroborates our results discussed above. Fig. 7 also shows the $U$-dependence of the local magnetic moment where the effect is even more pronounced. Note that in the insulator ($S^2$) is close but not equal to the prediction of the linearized DMFT ($S^2 = 2$ (cf. Fig. 5). This is due to residual local fluctuations which are neglected by the L-DMFT for the insulating phase.

The limit $J \rightarrow 0$ which appears to be somewhat exceptional can be analyzed easily. From Fig. 3 it is obvious that the critical interaction $U_{c,2}$ at which $z \rightarrow 0$ in the metastable phase shows a discontinuous jump from a value $U_{c,2} \approx 5 < U_{c,3}$ for $J > 0$ to $U_{c,2} = 10 = U_{c,3}$ for $J = 0$. To find the physical reason for this discontinuity of $U_{c,2}$, one can again look at the Lehmann representation of the spectral density for $V \rightarrow 0$. The main difference between the case $J = 0$ and the case $J > 0$ consists in the fact that for $J > 0$ only those configurations with a triplet on the impurity site contribute to the (singlet) ground state in the limit $V \rightarrow 0$ (see Fig. 5). As compared with $J = 0$ this is about a factor of 2 configurations less. Thus, the discontinuous jump of $U_{c,2}$ is caused by a strong suppression of orbital fluctuations due to a finite $J$ (a similar argument in a slightly different context has been put forward in Ref. 16). On the other hand, it should be stressed that the actual critical interaction $U_c$ remains continuous in the limit $J \rightarrow 0$ as $U_c \rightarrow U_{c,3}$. Note that to uncover this subtle distinction it has been necessary to extend the theory beyond the $z \rightarrow 0$ limit.

### IV. CONCLUSION

To summarize, we have presented an extension of the linearized dynamical mean-field theory (L-DMFT) to multi-orbital Hubbard models including the full exchange term. This allows a reliable calculation of the critical interaction strength $U_c$ for the zero-temperature Mott transition analytically for all $M$ at half-filling and numerically for all integer fillings $n \neq M$ and $M \leq 5$. We find a linear dependence of $U_c$ on the number of orbitals. Remarkably, a qualitative change of the nature of the $T = 0$ Mott transition is observed for any finite $J$: the transition becomes discontinuous with a finite jump of the quasi-particle weight at $U_c$ close to $U_{c,3}$.

For finite temperatures one might speculate that a finite $J$ increases the tendency for a first-order Mott-transition. This could be of experimental relevance for the Mott-transition in transition-metal oxides.

The L-DMFT for correlated electron systems with orbital degrees of freedom is a handy and trustable approach with a large potential for future applications. One might consider, for instance, the multi-band Hubbard...
model investigated in the context of the LDA+DMFT approach for the transition-metal oxide V$_2$O$_3$. The L-DMFT can be easily extended to answer questions about the importance of the $e_{g}$-bands (neglected in Ref. 18) on the Mott transition, or the influence of the full exchange term. We have already observed a significant quantitative change of $U_c$ when excluding the spin-dependent part from the exchange term in (2). This should have consequences for a realistic description of the MIT in transition-metal oxides which are worth to be studied.

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29. Taking into account additionally the insulating ($z=0$) solution for $U > U_{c,1}$ ($U_{c,1} < U_{c,2}$), this amounts to three coexisting solutions of the mean-field equations in the above interval.