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Effects of orbital degeneracy on the Mott transition in infinite dimensions

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We investigate the Mott transition in infinite dimensions in the orbitally degenerate Hubbard model. The qualitative features of the Mott transition found in the one-band model are also present in the orbitally degenerate case. We show that the quantitative aspects of the low-energy behavior near the Mott insulating state with one electron per site are not very sensitive to orbital degeneracy, justifying the quantitative success of the one-band model which had been previously applied to orbitally degenerate systems. We contrast this with quantities that have a sizable dependence on the orbital degeneracy and comment on the role of the intra-atomic exchange *J*. [S0163-1829(96)51740-6]

The Mott transition in transition metal oxides has received renewed theoretical and experimental attention. On the experimental side, new compounds have been synthesized[1] and older compounds such as V_2O_3 and $Ni_xSe_{1-x}S$ have been studied with higher resolution[2]. From the theoretical point of view new insights have been obtained from studying the one band Hubbard model in the limit of large lattice coordination.[3,4] These studies allowed quantitative comparisons of this model against three-dimensional transition-metal oxides. The doping dependence of the electronic specific heat in $La_xSr_{1-x}TiO_3$ can be described by the one-band Hubbard model without adjustable parameters after the values of U and D have been determined from the photoemission data.[5] The single band Hubbard model can describe the temperature dependence of the optical and the dc conductivity of V_2O_3[2] and $Sr_{1-x}Ca_xVO_3$.[6]

The degree of quantitative agreement between the mean-field theory and the experimental data is surprisingly good, if one considers the fact that in three-dimensional materials such as $La_{1-x}Sr_xTiO_3$ the d^1 electron has a quasithreefold degenerate level while the simplest description of V_2O_3 involves one electron in a twofold degenerate level.[7]

In this publication we report the main conclusions of our study of the *degenerate* Hubbard model in the limit of infinite dimensions: (a) All the *qualitative* features found in earlier studies of the Mott transition in the single-band Hubbard model persist in the degenerate case. (b) The quantitative values of certain low-energy quantities such as the effective mass and the optical gap, near the Mott insulating state with *one electron per site* depend rather weakly on the band degeneracy. This justifies, *a posteriori*, the quantitative success of the one-band Hubbard model when applied to orbitally degenerate systems with carrier density between 0 and 1. (c) We contrast the previous point with the high-energy behavior of the spectral functions that have a sizable dependence on the band degeneracy and comment on the role of the exchange parameter J, which only appears in the orbitally degenerate case.

We studied the Hubbard Hamiltonian for $l_b=2$ bands given by

$$H=-\frac{t}{\sqrt{d}}\sum_{\langle ij\rangle,m\sigma}c^{\dagger}_{im\sigma}c_{jm\sigma}+\frac{U_1}{2}\sum_{i,m\sigma}n_{im\sigma}n_{im\bar{\sigma}}+\frac{U_2}{2}\sum_{i,m\sigma}n_{im\sigma}n_{i\bar{m}\bar{\sigma}}+\frac{U_3}{2}\sum_{i,m\sigma}n_{im\sigma}n_{i\bar{m}\bar{\sigma}}, \qquad (1)$$

where $m=1,2$ ($\sigma=\pm1$) denotes the band (spin) index. A bar over m or σ means the complementary value, i.e., $\bar{m}=2$ if $m=1$ and $\bar{m}=1$ if $m=2$ ($\bar{\sigma}=-1$ if $\sigma=1$ and $\bar{\sigma}=1$ if $\sigma=-1$).

We introduced three interaction parameters U_i, which we parametrize in terms of U and J, $U_1=U$, $U_2=U_1-2J$, $U_3=U_2-J$.[8] We study the model on a Bethe lattice with

coordination d in the limit of infinite d. When not stated explicitly we measure energies in units where the half bandwidth $D=2t=1$.

The lattice model in infinite dimensions (in the paramagnetic phase) is mapped onto an impurity model describing an orbital $f_{m\sigma}$ with band index m and spin σ coupled to a bath of conduction electrons ($c_{km\sigma}$):

$$H_{\rm imp}=\epsilon_f\sum_{m\sigma} f^\dagger_{m\sigma}f_{m\sigma}+\sum_{k,m\sigma}\epsilon_k c^\dagger_{km\sigma}c_{km\sigma} + \sum_{k,m\sigma} V_k(c^\dagger_{km\sigma}f_{m\sigma}+f^\dagger_{m\sigma}c_{km\sigma})+\frac{U_1}{2}\sum_{m\sigma} n^{(f)}_{m\sigma}n^{(f)}_{m\bar\sigma} + \frac{U_2}{2}\sum_{m\sigma} n^{(f)}_{m\sigma}n^{(f)}_{m\sigma}+\frac{U_3}{2}\sum_{m\sigma} n^{(f)}_{m\sigma}n^{(f)}_{m\bar\sigma}, \quad (2)$$

where the hybridization function $\Delta(\omega):=\Sigma_k V_k^2/\omega-\epsilon_k-i\eta$ fulfills the self-consistency condition:[9] $\Delta(\omega)=t^2G(\omega)$.

To analyze the qualitative behavior of these equations we apply the projective self-consistent method.[10,11] This technique exploits the separation of scales between the quantities describing high-energy features of the spectral function (labeled by the superscript H) and the quantities describing low-energy features (labeled by L). We restrict ourselves to the $J=0$, Su($2l_b$) symmetric case. The local description of the Mott insulating state with a given integer occupancy is a degenerate Anderson impurity model in an insulating bath:

$$\mathcal{H}_a=\frac{U}{2}\sum_{(m\sigma)\neq(m'\sigma')} n_{m\sigma}n_{m'\sigma'}+\sum_{\alpha,k\in H} V_k^H(f^\dagger_\alpha c^H_{k\sigma}+\text{H.c.}) + \sum_{\alpha,k\in H}\epsilon^H_k(c^H_{k\alpha})^\dagger c^H_{k\alpha}. \quad (3)$$

$\mathcal{H}_a$ has d degenerate ground states carrying a representation of SU($2l_b$) [$d=2l_b$ for $n_{\rm tot}=1$ and $d=l_b(2l_b-1)$ for $n_{\rm tot}=2$] . We label the ground states of $\mathcal{H}_a$ (for a given value of chemical potential corresponding to integer occupancy $n_{\rm tot}$) as $|l\rangle$ and define Hubbard operators $X_{ll'}=|l\rangle\langle l'|$ acting on this low energy impurity manifold.

From Hamiltonian (3) describing the insulator and its ground states one extracts the matrix$J^{\mu\alpha}_{ll'}$

$$J^{\mu\alpha}_{ll'}=\left\langle l\left| f^\dagger_\mu\frac{1}{H-E_{\rm gs}}f_\alpha\right|l'\right\rangle-\left\langle l\left| f^\dagger_\alpha\frac{1}{H-E_{\rm gs}}f_\mu\right|l'\right\rangle. \quad (4)$$

For $n_{\rm tot}=1$ the indices μ, μ', and α in Eq. (4) denote the $2l_b$ spin-orbital combinations ($m\sigma$). For $n_{\rm tot}=2$ and $l_b=2$, however, the indices l, l', p, and p' are double indices, e.g., $l=(l_1,l_2)$, describing one of six possible pairs $(m\sigma,m'\sigma')$, which denote the quantum numbers of the ground states of Eq. (3).

Because of SU($2l_b$) invariance this matrix is characterized by two independent coupling constants. If $n_{\rm tot}=1$ ($l_b=1$ or $l_b=2$), the $J^{\mu\alpha}_{ll'}$ are of the form $J^{\mu\alpha}_{ll'}=a_1\delta_{ll'}\delta_{\mu\alpha}+b_1\delta_{l'\alpha}\delta_{l\mu}$, while for $n_{\rm tot}=2$ ($l_b=2$) one has $J^{\mu\alpha}_{ll'}=a_2\delta_{l_1l'_1}\delta_{l_2l'_2}\delta_{\mu\alpha}-b_2\epsilon^{\rho\alpha l_1l_2}\epsilon^{\rho\mu l'_1l'_2}$. We shall express the coefficients a and b in terms of a potential scattering interaction J_p and a spin spin interaction J_s below.

The equations which determine the critical value of chemical potential and interaction strength at which the metallic solution disappears can be written in a physically transparent fashion if one introduces an operator describing the normalized operators of the local low-energy bath electrons at the impurity site, $c_\alpha:=1/\sqrt{w}\Sigma_{k\in L}V^L_k c^L_{k\alpha}$, the low-energy part of the operators f_α given by $F^{LL}_\alpha=\sqrt{w}X_{ll'}J^{\mu\alpha}_{ll'}c_\mu$, and a low-energy impurity Hamiltonian describing the interaction of the degenerate ground-state manifold $|l\rangle$ and the low-energy bath electrons:

$$\mathcal{H}^L_{\rm eff}=wD^2(J_sO_s+J_pO_p)+\sum_{\alpha,k\in L}\epsilon_k{}^L c^{L\dagger}_{k\alpha}c^L{}_{k\alpha}. \quad (5)$$

Here O_s and O_p are the SU($2l_b$) generalizations of the spin-spin interaction and the potential scattering between the impurity states and the low-energy part of the bath of conduction electrons, respectively. For $n_{\rm tot}=1$ and SU($N=2l_b$), O_s has the form $O_s=(X_{\mu\mu'}c_\mu c^\dagger_{\mu'}-(1/N)X_{\mu\mu}c_{\mu'}c^\dagger_{\mu'})$, which for $l_b=1$ (i.e., $N=2$) reduces to $O_s=4\vec{S}c^\dagger_\sigma\vec{\sigma}_{\sigma\sigma'}c_{\sigma'}$. For $l_b=2$ and $n_{\rm tot}=2$, O_s is given by $O_s=(\frac{1}{2}X_{pp}c_\mu c^\dagger_\mu-\epsilon^{\rho\mu p'_1p'_2}\epsilon^{\rho\mu' p_1p_2}X_{pp'}c_\mu c^\dagger_{\mu'})$. The potential scattering term is $O_p=n_{\rm tot}X_{l'l'}c^\dagger_\mu c_\mu$.

The parameters V^L_k $\epsilon_k{}^L$ solve the low-energy self-consistency condition $t^2G^{LL}(\omega)=\Sigma_{k\in L}(V^L_k)^2/(\omega-\epsilon^L_k)$ where $G^{LL}(t-t')=-\langle TF^{LL}_\alpha(t)[F^{LL}_\alpha(t')]^\dagger\rangle_{\mathcal{H}^L_{\rm eff}}$.

The condition for the destruction of the metallic solution is obtained by expanding the previous equation to lowest order in w (Refs. 10 and 11) to obtain

$$\frac{1}{t^2}=[i(n_{\rm tot},l_b)-\langle O_s\rangle]J_s^2+J_p^2. \quad (6)$$

As shown in Ref. 11 the conditions for the destruction of the insulating solution is given by an equation similar to Eq. (6) but with $\langle O_s\rangle=0$:

$$\frac{1}{t^2}=i(n_{\rm tot},l_b)J_s^2+J_p^2. \quad (7)$$

Equations (6) and (7) allow us to discuss the trends in the values of critical couplings as the filling and band degeneracy is varied. Here $\langle\ldots\rangle$ indicates the expectation value with respect to the effective low-energy Hamiltonian $\mathcal{H}^L_{\rm eff}$ and $i(n_{\rm tot},l_b)$ are rational numbers which depend only on the group, i.e, on the band degeneracy l_b, and on the group representation which is determined by the number of electrons per site.

We notice that, as in the one band case,[10] the interaction Hamiltonian favors the formation of an SU($2l_b$) singlet between the conduction electrons and the degenerate SU($2l_b$) manifold of degenerate ground states $|l\rangle$. This implies that $\langle O_s\rangle<0$. Since the spin exchange constant decreases with increasing U, the critical value of the interaction at which a metal disappears U_{c2} is strictly larger than the critical value of the interaction at which the insulator breaks down U_{c1}. There is a region in the $\mu-U$ plane where a metallic and an insulating solution coexist. The metallic solution is lowest in

energy. Therefore the *qualitative structure* of the Mott transition in infinite dimensions is unaffected by the band degeneracy.

To calculate the phase diagram in the paramagnetic phase, one needs to solve a system of functional equations for the parameters ϵ_k, V_k which is necessary for the exact determination of $\langle O_s \rangle$, a task which is left for future work. Here we focus on the *trends* as degeneracy and occupation is changed, to illuminate the qualitative aspects of the problem. For this purpose it is sufficient to estimate $\langle O_s \rangle$ by half of its strong coupling value (denoted by a subscript sc) , which was shown to be a good approximation in the one band case. Their values are: $\langle O_s \rangle_{\rm sc} = -\frac{3}{2}$ for $n_{\rm tot}=1$ and $l_b=1$, $\langle O_s \rangle_{\rm sc} = -5/4$ for $n_{\rm tot}=1$ and $l_b=1$ and $\langle O_s \rangle_{\rm sc} = -5$ for $n_{\rm tot}=2$ and $l_b=2$. We have evaluated some of the corresponding values of $i(n_{\rm tot}, l_b)$, in Eqs. (6) and (7) using group theoretical methods and found $i(n_{\rm tot}=1, l_b=1)=\frac{3}{4}$, $i(n_{\rm tot}=1, l_b=2)=\frac{15}{16}$, and $i(n_{\rm tot}=2, l_b=2)=\frac{5}{4}$.

From Eqs. (6) and (7) it is clear that $\langle O_s \rangle / i(n_{\rm tot}, l_b)$ is a measure of the separation between U_{c1} and U_{c2}, which also determines the width of the coexistence region and the strength of the first-order transition. The $n_{\rm tot}=2$, $l_b=2$ transition has a much larger coexistence region than the $n_{\rm tot}=1$, $l_b=1$, which is slightly larger than the $n_{\rm tot}=1$, $l_b=2$ transition.

To estimate the trends in the critical values of the interactions, the insulating gaps, and the jump in the chemical potential we need also estimates of the exchange constants. It turns out that the qualitative behavior of the ratios of critical interactions already appear if we evaluate Eq. (4) to lowest order in t/U. We have for $n_{\rm tot}=1$ and $l_b=1$ or 2, $J_s = b_1$, and $J_p = a_1 + b_1/2l_b$ with $a_1 = (1/\mu - U)$ and $b_1 = 1/\mu + 1/(U-\mu)$. For $n_{\rm tot}=2$ and $l_b=2$, $J_s = b_2$ and $J_p = a_2 - b_2/2$ with $a_2 = 1/(\mu - U)$ and $b_2 = 1/(\mu - U) + 1/(2U-\mu)$. Inserting these results in Eq. (7) we can estimate U_{c1}, the critical value of the interaction when the insulator ceases to exist, as well as the Mott insulating gap. In the particle hole symmetric situations $\mu = U/2$ for $l_b=1$ and $U=\frac{3}{2}$ for $l_b=2$ the result is very simple since $U_{c1} \propto i[n_{\rm tot}=(l_b/2), l_b]$, so $U_{c_1}(l_b=2, n_{\rm tot}=2)/U_{c_1}(l_b=1, n_{\rm tot}=1) = 1.4$. Contrast this with the weak dependence of U_{c1} on the degeneracy for $n=1$ where $U_{c_1}(l_b=2, n_{\rm tot}=1)/U_{c_1}(l_b=1, n_{\rm tot}=1) \approx 1.0$ to lowest order in t/U.

Similar trends are found in the values of U_{c2} using half of the strong coupling value as an estimate of $\langle O_s \rangle$ in Eq. (6) yielding $U_{c_2}(l_b=2, n_{\rm tot}=2)/U_{c_2}(l_b=1, n_{\rm tot}=1) \approx 1.6$. and $U_{c_2}(l_b=2, n_{\rm tot}=1)/U_{c_2}(l_b=1, n_{\rm tot}=1) \approx 1.0$ Notice once again a very similar trend while the critical interaction for $l_b=2$ and $n_{\rm tot}=2$ is clearly larger than for $n_{\rm tot}=1$.

To obtain quantitative results we use a generalization of iterative perturbation theory (IPT) to particle-hole asymmetric problems[12]. Our goal here is to demonstrate that the low-energy behavior between densities zero and one is rather insensitive to the value of l_b and contrast this with the high-energy features of the photomission and optical spectroscopy where, in the same density range, band degeneracy introduces changes of order unity. We focus our quantitative comparison in the density range $0 \leqslant n_{\rm tot} \leqslant 1$ because, on one hand, the most controlled experiments have been done in systems with d^0 and d^1 configurations and because in this regime the reliability of the IPT has been tested extensively.

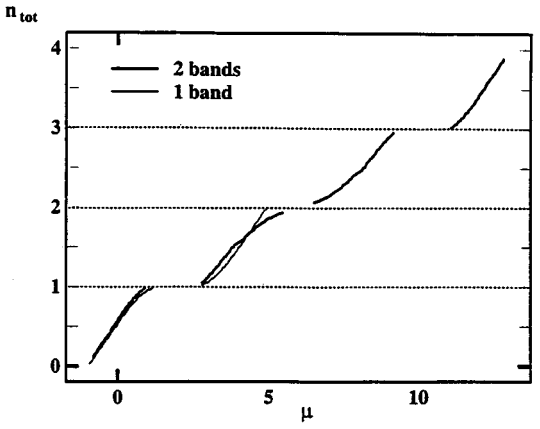

FIG. 1. Particle density of the two-band Hubbard model as a function of μ for $U=4$ (thick line) For comparison the thin line indicates a similar plot for the one-band Hubbard model with the same interaction strength.

Figure 1 displays the total particle density per lattice site as a function of the chemical potential for $U=4$. There is a Mott transition at $n_{\rm tot}=1$, 2, and 3. The gap around the $n_{\rm tot}=1$ transition is larger than the gap around the $n_{\rm tot}=2$ transition. The transitions at $n_{\rm tot}=1$ and $n_{\rm tot}=3$ are equivalent because of particle-hole symmetry. Notice that the curves for $l_b=1$ and $l_b=2$ are very close for densities not very far from unity. Notice the smaller value of the gap around the $n_{\rm tot}=2$ transition, which reflects the larger value of $U_{c2}(l_b=2)$.

The Mott transition as a function of filling is driven by the collapse of an energy scale, the renormalized Fermi energy, and the consequent divergence of the effective mass. The control parameter is either the interaction U or the chemical potential μ. We calculated the quasiparticle residue vs interaction strength curves at fixed doping and the quasiparticle residue vs doping curves at fixed interaction strength for densities between zero and one for $l_b=1$ and 2, the IPT scheme. Our conclusions are that the band degeneracy introduces changes which are at most of the order of

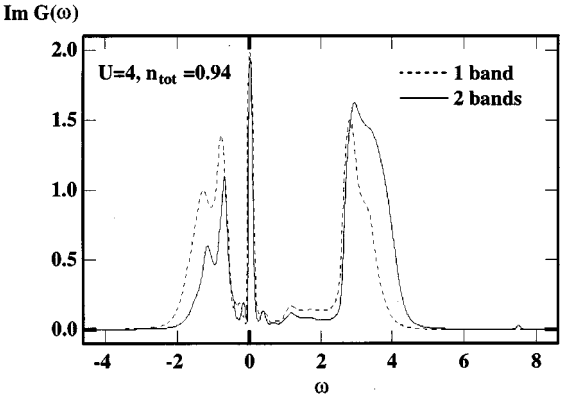

FIG. 2. Comparison of the spectral functions in the one- and the two-band model for $U=4$: metallic state for small doping ($n_{\rm tot}=0.94$).

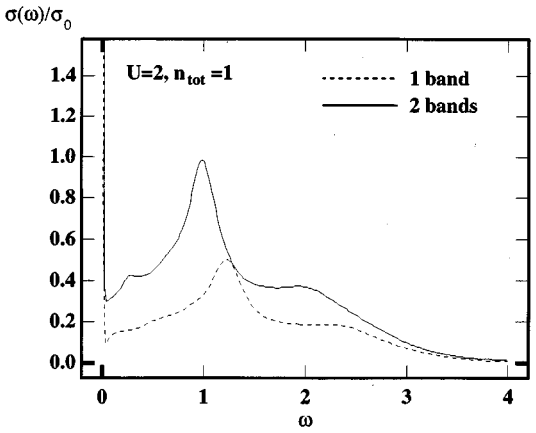

FIG. 3. Comparison of the optical conductivity for the one- and the two-band model at $U=2$ and $n_{\mathrm{tot}}=1$.

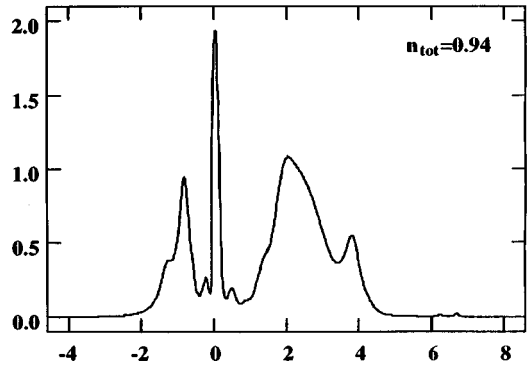

FIG. 4. Spectral function for $U=4$ and $J=0.5$ for density $n_{\mathrm{tot}}=0.89$.

10%. This surprising result should be contrasted with the strong dependence of the high-energy features on orbital degeneracy.

To illustrate this point we display (Fig. 2) the spectral functions in the one-band and two-band model for $U=4$. Notice the differences in the weights and widths of the Hubbard bands which should be readily observed in photoemission experiments. At $n_{\mathrm{tot}}=1$ the weights of both Hubbard bands are equal (i.e., $\frac{1}{2}$) in the single-band model, while in the two-band problem the weights are $\frac{1}{4}$ and $\frac{3}{4}$, respectively. Contrast this with the width of the low-energy resonance which is very similar in the one- and two-band model, since it is given by the quasiparticle residue, which depends weakly on band degeneracy near the $n=1$ Mott transition.

Figure 3 shows a comparison of the *finite frequency* optical conductivity for $U=2$ and $n=1$. The intensity has a clear dependence on the band degeneracy since it involves the low-energy resonance, the lower and the upper Hubbard band.

An important parameter with no analogue in the one-band model is the intraorbital exchange J, which originates in Hund's rule. This parameter does not affect the low-energy features significantly but affects the high-energy features of the photoemission spectra as shown in Fig. 4. The low-energy feature is very similar to what was found previously in the one-band model but there is now a splitting of the upper Hubbard band due to the finite value of J which results in three different interaction parameters ($U_1=U$, $U_2=U-2J$, and $U_3=U-3J$), causing a splitting of the upper Hubbard band.

The interatomic exchange has also a profound influence on the phase diagram. This can be shown analytically in the regime of large interaction strength by noticing that in infinite dimensions one can neglect the effects of quantum fluctuations after an effective spin orbital Hamiltonian is derived. As expected the ground state is ferromagnetic, but the difference in energy per spin with a Néel state is given by the energy scale (t^2J/U^2), which is small since J tends to be much smaller than U. The small energy differences between different configurations of spin pairs which describe the possible magnetic orderings are the ultimate justification for the stability of the paramagnetic solution in realistic systems. This magnetic degeneracy becomes even more pronounced when one considers the superexchange via ligand oxygens and will be discussed elsewhere.[13]

Recently the degenerate Hubbard model has been investigated with the Gutzwiller approximation,[14] and a novel extension of the slave boson method.[15] The qualitative trends in the low-energy quantities that we found are in good agreement with these works. The critical values of the finite-temperature Mott transition in the degenerate Hubbard model in infinite dimensions have been evaluated using the quantum Monte Carlo method (Ref. 16). The trends found are in excellent agreement with our work.

The prospects of understanding realistic strongly correlated materials using dynamical mean-field methods look very promising. In particular, it would be interesting to incorporate more realistic band structures together with a realistic orbital degeneracy (threefold degeneracy for the earlier transition metal oxide), to eliminate some of the limitations of our work. A second important direction is the estimation of the corrections due to finite dimensionality.

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