Orbital-Selective Mott Transitions in the Degenerate Hubbard Model

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We investigate the Mott transitions in two-band Hubbard models with different bandwidths. Applying dynamical mean field theory, we discuss the stability of itinerant quasiparticle states in each band. We demonstrate that separate Mott transitions occur at different Coulomb interaction strengths in general, which merge to a single transition only under special conditions. This kind of behavior may be relevant for the physics of the single-layer ruthenates, $Ca_{2-x}Sr_xRuO_4$.

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Strongly correlated multiorbital electron systems are among the most active topics in condensed matter physics. In Mott insulators the addition of orbital to localized spin degrees of freedom leads to complex phase diagrams. In itinerant electron systems multiple Fermi surface sheets appear with very distinct properties. Subtleties occur when localized and itinerant electrons coexist, as is well known in the case of itinerant d and localized f electrons which give rise to the rich physics of heavy Fermion systems. In view of their very different character this coexistence is not surprising. We may ask, however, whether the coexistence of itinerant and localized electrons is possible for degenerate nonhybridizing orbitals with small bandwidth differences.

A nearly degenerate d-electron system where multiorbital properties obviously play an important role is the single-layer isovalent ruthenate alloy $Ca_{2-x}Sr_xRuO_4$ [1]. The end-member Sr_2RuO_4 is a well-known unconventional superconductor [2,3], while Ca_2RuO_4 is a Mott-insulating $S = 1$ antiferromagnet [4,5]. The relevant $4d$ orbitals belong to the t_{2g} subshell. The planar structure prevents hybridization between orbitals which have even (d_{xy}) and odd parity (d_{yz}, d_{zx}) under the reflection $z \to -z$. The complex evolution between these different end members has led to various theoretical investigations [5,6], and among others to the proposal that some of the d orbitals display localized spin and orbital degrees of freedom, and others provide itinerant electrons. This orbital-selective Mott transition (OSMT) could explain the experimental observation of a localized spin $S = 1/2$ in the metallic system at $x \sim 0.5$ which is difficult to obtain from the entirely itinerant description [4,6,7].

The concept of the OSMT was recently challenged, in particular, by Liebsch whose dynamical mean field theory (DMFT) calculations suggested that two bands of different width coupled by electron-electron interactions would always undergo a common Mott transition [8]. The aim of this Letter is to revisit this problem and to analyze the Mott transition in the degenerate two-orbital Hubbard model with different bandwidths. Using another scheme of DMFT, we obtain a different result and an OSMT. In addition, we show also that correlations can stabilize a commensurate filling of one band even when the total electron count is fractional.

We consider the following Hubbard Hamiltonian with two orbitals:

$$H = \sum_{\substack{\langle i,j\rangle \\ \alpha,\sigma}} [t_{ij}^{(\alpha)} - \mu_\alpha \delta_{ij}] c_{i\alpha\sigma}^\dagger c_{j\alpha\sigma} + U \sum_{i\alpha} c_{i\alpha\uparrow}^\dagger c_{i\alpha\uparrow} c_{i\alpha\downarrow}^\dagger c_{i\alpha\downarrow} + U' \sum_{i\sigma\sigma'} c_{i1\sigma}^\dagger c_{i1\sigma} c_{i2\sigma'}^\dagger c_{i2\sigma'} - J \sum_{i\sigma\sigma'} c_{i1\sigma}^\dagger c_{i1\sigma'} c_{i2\sigma'}^\dagger c_{i2\sigma} - J \sum_i [c_{i1\uparrow}^\dagger c_{i1\downarrow}^\dagger c_{i2\uparrow} c_{i2\downarrow} + \text{H.c.}], \quad (1)$$

where $c_{i\alpha\sigma}^\dagger (c_{i\alpha\sigma})$ creates (annihilates) an electron with spin $\sigma (=\uparrow, \downarrow)$ and orbital index $\alpha (=1, 2)$ at the ith site. We restrict ourselves to the case of nonhybridizing orbitals, relevant to the ruthenates, and $t_{ij}^{(\alpha)}$ denotes the hopping integral for orbital α, μ the chemical potential, U (U') the intraband (interband) Coulomb interaction, and J the Hund coupling. In the following, we impose the condition $U = U' + 2J$, obtained by symmetry arguments for degenerate orbitals.

We examine the stability of the metallic ground state of this model by means of DMFT which maps the lattice model to the problem of a single-impurity connected dynamically to a "heat bath" [9]. The electron Green's function is obtained via the self-consistent solution of this impurity problem. We represent the two-electron bands by semicircular density of states (DOS), $\rho_\alpha(x) = 2/\pi D_\alpha \sqrt{1 - (x/D_\alpha)^2}$ where $2D_\alpha$ is the bandwidth. For the case of identical hopping integrals for the two bands, $t_{ij}^{(\alpha)} = t_{ij}$ ($D_1 = D_2$), the role of orbital fluctuations has been discussed in Refs. [10,11]. There are various methods to solve the effective impurity problem. The quantum Monte Carlo (QMC) simulations used by Liebsch [8] suffer from sign problems at low temperatures, in particular, if the Hund coupling is included. We apply here the exact diagonalization method proposed by Caffarel and Krauth [12], whose finite-size effects are rather small even for small systems, giving results in good agreement with other numerical methods applied to the

single-impurity model. Additionally, we treat the linearized version of DMFT (two-site DMFT) [13], which allows us to discuss electronic properties well even in the vicinity of the critical point. We restrict our discussions to the paramagnetic case to clarify the nature of the Mott transition. In the following, the width of the narrower band is used as the energy unit.

We first consider the case $\mu_1 = \mu_2 = U/2 + U' - J/2$, i.e., both bands are half filled. The quasiparticle weight Z_α, defined by $Z_\alpha^{-1} = 1 - d\mathrm{Re}[\Sigma_\alpha(\omega)]/d\omega$ in terms of the self-energy $\Sigma_\alpha(\omega)$ of each band, will be used to characterize the stability of the metallic state of the two bands. The results obtained with fixed ratios U'/U and J/U are shown in Fig. 1 for half-filled bands. We first focus on the case of $U = U'$ and $J = 0$ [shown in Fig. 1(a)] with bandwidths $D_1 = 1.0$ and $D_2 = 2.0$. When the Coulomb interaction is turned on, the quasiparticle weights Z_1 and Z_2 decrease from unity in slightly different ways reflecting the difference of the bandwidth. A strong reduction of the quasiparticle weight appears initially in the narrower band. However, when the system approaches the Mott transition, the quasiparticle weights merge again displaying a very similar dependence on U, and eventually reach zero at the same critical point. The inset shows the more extreme case of $D_1 = 1.0$ and $D_2 = 5.0$ (very wide band). The common Mott transition originates from the enlarged symmetry inherent in $U = U'$ and $J = 0$, which will be discussed below. This result is in agreement with the conclusion by Liebsch [8]. For small interaction strengths the quasiparticle weight depends on the effective Coulomb interactions U/D_α which are different for two bands of different width, D_α and yields a distinct behavior of Z_1 and Z_2. In the vicinity of the Mott transition, however, the effect of the bare bandwidth is diminished due to the strong renormalization of the effective quasiparticle bandwidth allowing Z_1 and Z_2 to vanish together [14,15].

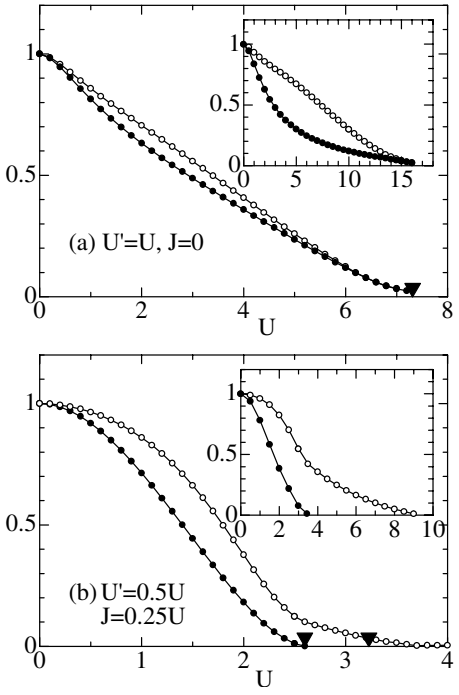

FIG. 1. The quasiparticle weights Z_1 and Z_2 at half filling as a function of the Coulomb interaction U: (a) $U'/U = 1.0$ ($J = 0$) and (b) $U'/U = 0.5$ ($J/U = 0.25$). The bandwidth is set as $D_1 = 1.0$ and $D_2 = 2.0$. Open (closed) circles represent the results for orbital $\alpha = 1(2)$ obtained by solving the DMFT impurity problem by means of the exact diagonalization of a small cluster ($N = 6$). Solid triangles represent the Mott-transition points obtained by the two-site DMFT method, which produce the values quite consistent with those of the numerical diagonalization. Insets show the same plot for bandwidths $D_1 = 1.0$ and $D_2 = 5.0$.

The introduction of a finite Hund coupling J makes $U \neq U'$ and leads to a qualitatively different behavior, as seen in Fig. 1(b). With increasing U keeping the ratio $U'/U = 0.5$ fixed, the quasiparticle weights decrease differently and vanish at different critical points: $U_{c1} \approx 2.6$ for Z_1 and $U_{c2} \approx 3.5$ for Z_2. Therefore, we observe an intermediate phase with one orbital localized and the other itinerant, though strongly renormalized ($Z_2 \ll 1$). The analogous behavior is observed for different choices of the bandwidths, if J takes a finite value [inset of Fig. 1(b)]. Although it is difficult to precisely determine the second critical point U_{c2}, this result certainly suggests the existence of the OSMT with $U_{c2} > U_{c1}$.

In Fig. 2 we show how the quasiparticle states evolve and then disappear inside of the Mott-Hubbard gap. The DOS is computed by the two-site DMFT scheme [13]. In both cases the Mott-Hubbard gap develops as U increases and is accompanied by narrow quasiparticle midgap bands. For case (a) with $J = 0$, these quasiparticle bands disappear simultaneously, whereas for case (b) with finite J, they have different critical points, consistent with the results mentioned above.

Repeating similar DMFT calculations for various choices of the parameters, we derive the ground-state phase diagram shown in Fig. 3, which displays some remarkable features. First, the metallic phase (I) remains stable up to surprisingly large Coulomb interaction U when $U \to U'$ (small J). Here the Mott transitions merge to a single transition. This behavior originates from the high symmetry when $U = U'$ ($J = 0$) with six degenerate two-electron onsite configurations: four spin configurations with one electron in each orbital and two spin singlets with both electrons in one of the two orbitals. The additional symmetry in orbital/spin degrees of freedom enlarges the phase space for charge fluctuations and leads to a decrease of the Mott-Hubbard gap E_g at large U. A rough estimate of E_g can be obtained from the second moment of the hopping Hamiltonian for a state $|a\rangle$ with an extra electron (or hole). In $\langle a|H^2|a\rangle = T^2$ all possible configurations with the same onsite energies are considered as intermediate states [16]. Because all charge excitations mix with each other, there is only one gap. Assuming a staggered spin or orbital configuration as the

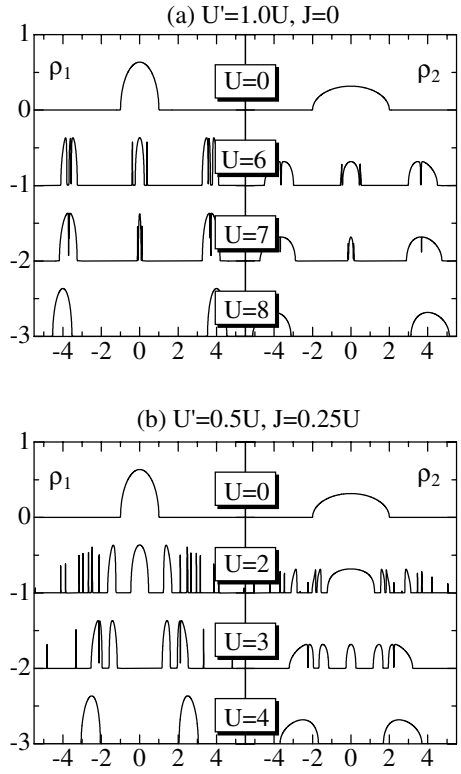

FIG. 2. The density of states $\rho_\alpha(\omega)$ at half filling: left (right) panels for $\alpha = 1$ (2). (a) $U'/U = 1.0$ ($J = 0$) and (b) $U'/U = 0.5$ ($J/U = 0.25$). It is clearly seen that the Mott transitions occur simultaneously in (a), while the orbital selective transitions occur in (b).

most dominant local correlation for neighboring two-electron sites we obtain for the effective hopping matrix element of an extra carrier $T = \sqrt{t_1^2 + t_2^2}$. This allows us to estimate the Mott-Hubbard gap at large U:

$$E_g = U - 2zT = U - 2z\sqrt{t_1^2 + t_2^2}, \qquad (2)$$

where z is the coordination number. We can get a simple estimate of U_c by setting $E_g \to 0$ leading to values $U_c = 2D_1\sqrt{2}$ (for $D_1 = D_2$) and $U_c = 2D_1\sqrt{5}$ (for $2D_1 = D_2$). Both estimates are enhanced relative to the single-band case $U_c = 2D$ [11].

Away from the symmetric limit, i.e., $U > U'$ ($2J = U - U'$) orbital fluctuations are suppressed and the spin sector is reduced by the Hund coupling to three onsite spin triplet components as the lowest multiplet for two-electron sites. Applying the same scheme as above, we recognize that charge excitations with two electrons in one or the other of the orbitals do not mix, since all hopping processes included in (1) preserve orbital configurations in the lowest multiplet sector. The effective hopping for each orbital is now $T_\alpha = t_\alpha/\sqrt{2}$ assuming a staggered spin-1 state at half filling. The reduction compared to the single-band case occurs due to the locking of the spins into an onsite spin triplet. If we consider again the case $2D_1 = D_2$ we find two separate Mott transitions with critical values

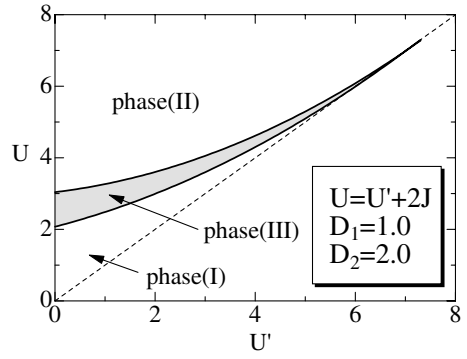

FIG. 3. Phase diagram for the two-orbital Hubbard model with $D_1 = 1$ and $D_2 = 2$. In the phase (I) [phase (II)], both bands are in the metallic (insulating) state. The phase (III) is induced by the orbital-selective Mott transition, where the metallic state coexists with the Mott insulating state. Since we are concerned with the ferromagnetic Hund coupling, $J > 0$, the relevant region in the diagram is $U > U'$.

$$U_{c1} = \frac{1}{\sqrt{2}}D_1 + \frac{U'}{2}, \qquad U_{c2} = \sqrt{2}D_1 + \frac{U'}{2}. \qquad (3)$$

In between the two transitions we find the metallic intermediate phase (III) with one band localized and Mott insulating and one band itinerant. Within our DMFT scheme we have also confirmed that various choices of bandwidths give rise to the qualitatively same structure of the phase diagram as shown in Fig. 3.

Our result for the Mott transitions is different from that of both Anisimov *et al.* and Liebsch [7,8]. The former group derived an OSMT for a special model which includes only the intraband Coulomb repulsion U within the DMFT approach, and drops effects due to coupling between the orbitals. This scheme was criticized by Liebsch, who took into account U, U', and J. He claimed based on a DMFT analysis that only a single Mott transition occurs in the generic case. Liebsch's solution of the single-impurity problem within DMFT is based on QMC and iterative perturbation methods. The former suffers from sign problems which limit its validity at low temperatures, while the latter is an extrapolation from the small-U regime. Therefore this method may obscure the observation of separate Mott transitions for the system with small Hund coupling, where two transition points are very close to each other. Our DMFT analysis which uses the exact diagonalization of the impurity problem on a finite cluster is valid at zero temperature and is not restricted to weak coupling. It shows separate Mott transitions, except for the special case with high symmetry $U = U'$ ($J = 0$), for which the transitions merge irrespective of the different bandwidths.

We have so far treated the case of two individually half-filled bands. We now address the question of what will happen when the electron count is nonstoichiometric. This may be a further key problem to understanding the OSMT in $Ca_{2-x}Sr_xRuO_4$ [4], since all three original metallic bands possess fractional filling. In order to study

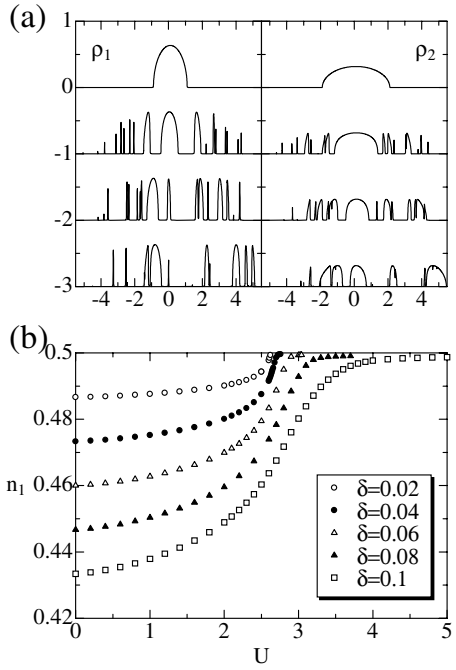

FIG. 4. (a) The density of states $\rho_1(\omega)$ and $\rho_2(\omega)$ for finite hole doping, $\delta = 0.1$. The Coulomb interaction is chosen as $U = 0, 2.0, 3.0$, and 4.0 ($U' = 0.5U$ and $J = 0.25U$) from the top to the bottom. (b) The number of electrons in the orbital ($\alpha = 1$) as a function of U when $U' = 0.5U$ and $J = 0.25U$.

this kind of system, we introduce a finite hole doping, and observe how commensurability can emerge due to interactions.

In Fig. 4(a) we show the DOS which is computed by using the two-site DMFT. With increasing interactions quasiparticle states with large DOS appear around the Fermi energy in both bands. Enhancing the interactions further we drive the first band insulating. This is in contrast to the single-band system, where finite hole doping obscures the Mott transition and always gives metallic behavior. In the two-band system, however, commensurability in one of the bands gradually emerges, as is clearly seen in Fig. 4(b). The electron number for the first band n_1 is plotted here. When $U = 0$, n_1 and $n_2\,(= 1 - \delta - n_1)$ are smaller than 0.5 because of finite hole doping ($\delta = 0.1$). Coulomb interaction causes electron transfer from one orbital to the other, giving rise to one half-filled band at a certain interaction strength, thereby causing an OSMT.

In conclusion, we have discussed the Mott transitions in the degenerate Hubbard model with nonhybridizing orbitals and different bandwidths using DMFT. A single Mott transition occurs when the Hund coupling is absent ($U = U'$), rendering the different bandwidth essentially irrelevant at the transition point, as discussed by Liebsch. In the more generic situation with finite Hund coupling, however, we find the OSMT. This remains true for non-stoichiometric systems. We believe that our study resolves the apparent contradictions on this issue and sheds light on the nature of this kind of Mott transitions. In real materials, Hund coupling between orbitals as well as the Coulomb interactions are present. Therefore, the OSMT should occur depending on the interaction, the band structure and filling, etc. A direct comparison with $Ca_{2-x}Sr_xRuO_4$, however, is inappropriate here, since the number of bands is different from two orbitals discussed in our model. Taking into account three bands in combination with accurate band structure information and realistic parameters (U, U', J), in order to clarify the behavior of this complex material, is under investigation at present.

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