

Orbital-Selective Mott Transition out of Band Degeneracy Lifting

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We outline a general mechanism for orbital-selective Mott transition, the coexistence of both itinerant and localized conduction electrons, and show how it can take place in a wide range of realistic situations, even for bands of identical width and correlation, provided a crystal field splits the energy levels in manifolds with different degeneracies and the exchange coupling is large enough to reduce orbital fluctuations. The mechanism relies on the different kinetic energy in manifolds with different degeneracy. This phase has Curie-Weiss susceptibility and non-Fermi-liquid behavior, which disappear at a critical doping, all of which is reminiscent of the physics of the pnictides.

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The localization of electrons in partially filled bands due to their mutual Coulomb repulsion, or Mott metal-insulator transition, is perhaps the most striking effect of electron-electron interaction and it escapes a proper description in the standard one-body theory of solids. Only the development of many-body methods like slave-particle mean fields and dynamical mean-field theory [1] (DMFT) allowed to draw a coherent picture at least for the single-band Hubbard model, which accounts for many properties of three-dimensional oxides [2].

In multiband systems the Mott physics is richer and still lacks a complete understanding. The orbital-selective mott transition (OSMT) (i.e., the possibility of localization of part of the conduction electrons while the rest remains itinerant) is a most interesting and unique feature. OSMT has been evoked [3] as a possible explanation of the seemingly mixed itinerant (metallic resistance) and localized (Curie-Weiss magnetic susceptibility) character of the conduction bands in the triplet-superconductor $\text{Sr}_{2-x}\text{Ca}_x\text{RuO}_4$. This idea has attracted considerable attention and sparked a good deal of theoretical investigation [4–7], uncovering a rich physics in which the Hund's coupling plays an important role [5,6] and non-Fermi-liquid behavior is found in the metallic component [8]. Even if the occurrence of an OSMT in $\text{Sr}_{2-x}\text{Ca}_x\text{RuO}_4$ is still under debate [9–13], it is suggestive that other anomalous superconductors like the *K*-BEDT organics and the iron oxypnictides [14] have been considered as possible realization of this physics [15], and that a *k*-space selectivity has been evoked even for the high- T_c cuprates [8,16].

Several distinct mechanisms that can lead to an OSMT have been identified. The original and most obvious is that the conduction electrons lie in separate nonhybridized correlated bands of different bandwidths [3], or that bands of

similar bandwidth have different intraband Coulomb repulsion [17]. In Ref. [18] the crystal-field splitting of two bands of equal bandwidth has been identified as a source of the OSMT. By lightly doping the Mott insulator, electrons populate the lower band, while the higher one will remain half-filled and insulating, leading to an OSMT driven by doping at incommensurate fillings. Unfortunately all these situations, in which the OSMT arises, are quite specific.

In this Letter we identify and study a new mechanism for the OSMT which brings it from a rare to a much more plausible phenomenon. We consider a system in which a crystal-field splitting divides the original degenerate manifold in subsets with different degeneracy, the simplest example being three levels split so that one level has different energy with respect to the other two, that remain degenerate (or nearly degenerate). In this case an OSMT can take place even in the case of equal bandwidth and Coulomb repulsion for all bands, and for commensurate fillings. This phenomenon can be understood on the basis of our knowledge about Mott transitions for degenerate bands. The critical interaction strength U_c for the Mott transition is indeed larger for manifolds of bands with larger degeneracy due to their increased kinetic energy [19,20]. For example, in the $SU(N)$ -orbital Hubbard model U_c scales with N at large N , while, for fixed number of bands, a Mott transition occurs at any integer filling [21,22] and U_c is largest at half-filling and decreases moving away from it. Therefore if the split manifolds were composed by different number of levels and completely decoupled they would definitely undergo Mott transitions at distinct values of U . Here we show how this survives when the manifolds are coupled by an interband Coulomb repulsion and a spin-spin exchange interaction, identifying the role of the latter and of the crystal field to suppress orbital fluctuations, leading to an effective orbital decoupling.

The situation we consider is indeed very general, making our OSMT probably the most common in nature. It is well known that, e.g., a cubic crystal field splits the five d orbitals in two groups, t_{2g} and e_g , respectively, originating three and two bands. Further lowering of the symmetry can induce further splittings.

We investigate the simplest realization of such a mechanism, namely, a system of three bands (the minimal situation in order to have manifolds of different degeneracy after the splitting, i.e., two degenerate bands and one lifted by the crystal field) of equal bandwidth with 4 electrons per site. In absence of the crystal-field splitting each band will be populated by $4/3$ electrons. If we continuously lift one of the bands to higher energy, the electrons will gradually move to the lower levels. Therefore the density of the lifted band will decrease from $4/3$ eventually reaching 1, becoming half filled. Then, if the interaction strength is enough to localize the half-filled band, but it is smaller than the critical value for the remaining three electrons hosted by the lower two bands, we can expect an OSMT.

A first step in this direction has been taken in Ref. [9], where an OSMT has been reported for two wide degenerate bands and a narrower one lifted in energy, with 4 electrons per site. Unfortunately in that model both the difference in bandwidth and the lifted degeneracy are at work and none could be singled out as the driving one.

The electrons in the three bands are coupled via a local $SU(2)$ invariant interaction. The Hamiltonian reads

$$\begin{aligned}
 H = & -t \sum_{\langle ij \rangle, m\sigma} (d_{im\sigma}^\dagger d_{jm\sigma} + \text{H.c.}) + \sum_{i,m\sigma} \epsilon_m d_{im\sigma}^\dagger d_{im\sigma} \\
 & + U \sum_{i,m} n_{im\uparrow} n_{im\downarrow} + \left(U' - \frac{J}{2} \right) \sum_{i,m>m'} n_{im} n_{im'} \\
 & - J \sum_{i,m>m'} [2\mathbf{S}_{im} \cdot \mathbf{S}_{im'} + (d_{im\uparrow}^\dagger d_{im\downarrow}^\dagger d_{im'\downarrow} d_{im'\uparrow} + \text{H.c.})].
 \end{aligned} \tag{1}$$

Here $d_{i,m\sigma}$ is the destruction operator of an electron of spin σ at site i in orbital m , and $n_{im\sigma} \equiv d_{im\sigma}^\dagger d_{im\sigma}$, $n_{im} \equiv \sum_{\sigma} d_{im\sigma}^\dagger d_{im\sigma}$, \mathbf{S}_{im} is the spin operator for orbital m at site i , t is the nearest-neighbor hopping (denoted in the sum by $\langle \rangle$), ϵ_m is the bare energy level in orbital m . U and $U' = U - 2J$ are intra- and interorbital repulsions and J is the Hund's coupling. The densities of states of the three bands are semicircular of half-bandwidth D .

We study this three-band model assuming that two bands have the same energy ($\epsilon_2 = \epsilon_3$) and one is lifted by a crystal-field splitting $\Delta \equiv \epsilon_1 - \epsilon_2 > 0$. The $\Delta < 0$ case, which is believed to be relevant to $\text{Sr}_{2-x}\text{Ca}_x\text{RuO}_4$ has been studied in [10] and does not lead to an OSMT. Yet, it has been recently proposed that a similar mechanism to what we present here applies in $\text{Sr}_{0.2}\text{Ca}_{1.8}\text{RuO}_4$ even if $\Delta < 0$ thanks to a doubling of the unit cell [11].

We use two local mean-field approximations: the faster and computationally inexpensive slave-spin mean-field [5,23] (SSMF) for surveying the phase diagram and

DMFT, solved with exact diagonalization (ED), for more accurate and aimed calculations. In Fig. 1 we show the SSMF phase diagram obtained adjusting Δ to have 1 electron in the lifted band, and 1.5 electrons in each of the degenerate ones. Indeed, an orbitally selective Mott phase (OSMP) is found for a large zone of the parameters U and J . It is worth noting that a finite Hund's coupling is needed to stabilize the OSMP, while for small J a direct transition from a metal to a Mott insulator is found. The indications of SSMF are confirmed by the more accurate DMFT, as shown in Fig. 2, where we plot $Z_\alpha = (1 - \text{Im}\Sigma_\alpha(i\omega_0)/\omega_0)^{-1}$ [$\Sigma_\alpha(\omega)$ being the self-energy for the band α], which measures the low-frequency spectral weight associated with metallic behavior. Z_1 for the lifted band vanishes at a critical U , signaling the localization of this band, while the same quantity is still finite for the two lower bands. We notice that ED calculations suffer from truncation effects. Analyzing these effects we find that the actual U_c will be higher than what shown in the figure and we estimate the DMFT value of $U_c \simeq 2.5D$, for $J/U = 0.25$. Comparison with SSMF confirms the reliability of the latter approach, which only slightly overestimates U_c .

The phase diagram clearly shows that increasing J/U increases the region of the OSMP. We can gain more insight analyzing the orbital fluctuations $\langle n_1 n_2 \rangle - \langle n_1 \rangle \langle n_2 \rangle$. In order to have an OSMT this quantity should be small, signaling a decoupling of the bands which opens the way for a different behavior between them, and the localization of the half-filled one. As shown in the inset of Fig. 2, for $J = U = 0$ the orbitals are uncorrelated. Increasing the two quantities, U initially prevails, leading to an increased orbital correlation. Further increasing U and J makes the electrons more and more localized. In this regime the effect of J becomes predominant [24], and it reduces the orbital correlations. The role of J can be understood in the atomic limit: increasing J enhances the distance between the lowest-lying high-spin state in which

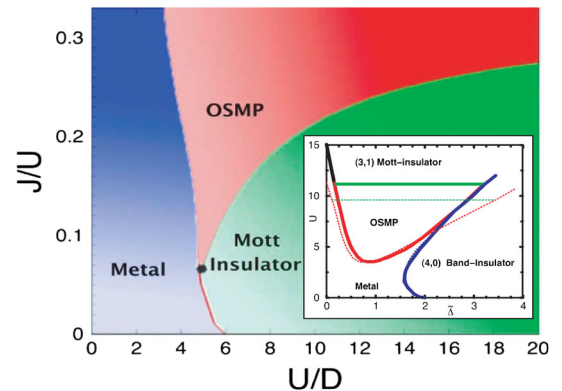


FIG. 1 (color online). Phase diagram for fixed populations $n_m = (1, 1.5, 1.5)$ (obtained by adjusting the crystal field Δ) within Slave-spin mean-field. Inset: phase diagram for fixed total filling $n = 4$ as a function of U and Δ at $J/U = 0.25$. Dashed lines: modification of this diagram under a small splitting ($\sim 0.4/D$) of the two degenerate bands.

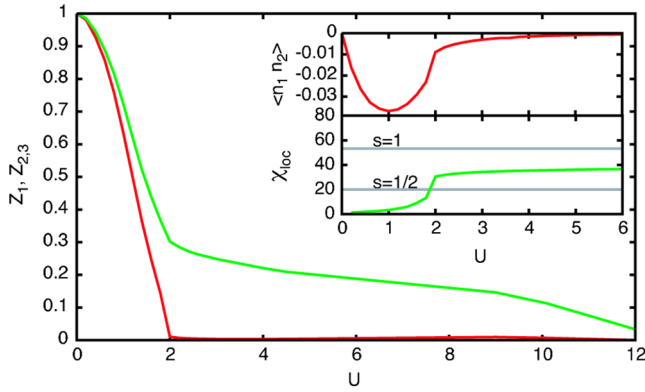


FIG. 2 (color online). Quasiparticle residue as a function of the interaction strength for $J/U = 0.25$ and fixed populations $n = (1, 1.5, 1.5)$ in DMFT with $N_s = 9$. Inset: interorbital correlations (top panel) and local spin susceptibility (bottom panel). The cutoff “temperature” (see text) is $\beta D = 80$.

the orbitals are equally populated, and the multiplets with different orbital population. Thus fluctuations between the two orbitals are suppressed. The effect of J is clearly stronger in the presence of the crystal-field splitting that naturally decouples the orbitals. The U_c for the OSMT slowly approaches the single-band one ($U_c = 3.34D$ in SSMF) as J/U increases. The transition for the other bands instead gets pushed at larger U/D , so that the OSMP zone widens at large J .

It is important, in order to assess the generality of this phase, to show that the OSMT occurs for a finite range of Δ and that no fine-tuning is needed. We have thus fixed only the total population to 4 electrons per site and studied the system as a function of Δ for several values of U at fixed $J/U = 0.25$. The phase diagram obtained in SSMF is plotted in the inset of Fig. 1 and shows that the region of the OSMP is quite large, and it increases with U . The critical Δ assumes reasonable values already for intermediate coupling. We notice that this phase diagram bears resemblance to what found in Ref. [9] for a version of our model in which the bandwidth of the lifted band is half of the other two, suggesting that also in that case the driving mechanism for the OSMT is the different degeneracy and not the bandwidth difference.

In Fig. 3 we show the spectral densities. As previously studied [5,6] for finite J the insulating band shows a full gap, while the other two show spectral weight at the chemical potential. This gives an explanation of the phase diagram in the $\Delta - U$ plane (inset of Fig. 2): there is a large range of Δ for which the chemical potential falls inside the gap of the localized band, leaving it insulating.

A key quantity for understanding the nature of the OSMP is the local spin susceptibility χ_{loc} (DMFT results in the second panel in the inset of Fig. 2). When a band gets localized χ_{loc} goes from a Pauli-like ($\sim \text{const}$) to a Curie-like ($\sim 1/T$) behavior indicating the formation of free moments. At $T = 0$ the susceptibility therefore diverges, but

in the ED calculation the fictive temperature β plays the role of a low-energy cutoff. As a result the saturation value is $\beta S(S+1)/3$. In a one-band Mott insulator $\chi = \beta/4$, signaling $S = 1/2$ [1], while for an N -band half-filled Mott insulator with $J > 0$ the high-spin state $S = N/2$ is selected [25], and one finds $\chi = \beta N/2(N/2 + 1)/3$.

When an OSMT occurs, we expect a two-stage saturation as a function of U , with the lifted band localizing before the complete Mott transition occurs. Therefore, in the OSMP χ_{loc} is expected to show a Curie component due to the magnetic moment of the localized electrons and a contribution of the itinerant electrons. The Hund’s coupling between the different orbitals will actually induce a local moment in the itinerant bands. Thus the total effective moment will be larger than that of the localized component [8]. The DMFT results clearly confirm this, as the moment is intermediate between the spin-1/2 of an independent single-band Mott state and spin-1 (Fig. 2).

Another interesting property of the OSMP is the non-Fermi-liquid nature of the itinerant component. The magnetic moments of the localized electrons act as scattering centers for the itinerant electrons. In the two-band model with different bandwidths, for an $SU(2)$ Hund’s coupling, a logarithmic behavior [8] at low ω or a power-law [26] has been reported for the imaginary part of the self-energy. In the present model the non-Fermi-liquid behavior is confirmed. Indeed as shown in Fig. 3, while in the metallic phase the self-energies have a linear behavior at low ω , in the OSMP the metallic one shows a much faster drop.

We finally investigate the stability of the OSMP by respect to doping. The “standard” Mott insulating phase in the single-band and in degenerate multiband models is destroyed by any finite doping, while, as mentioned in the introduction, in Ref. [18] an OSMT is found doping a half-

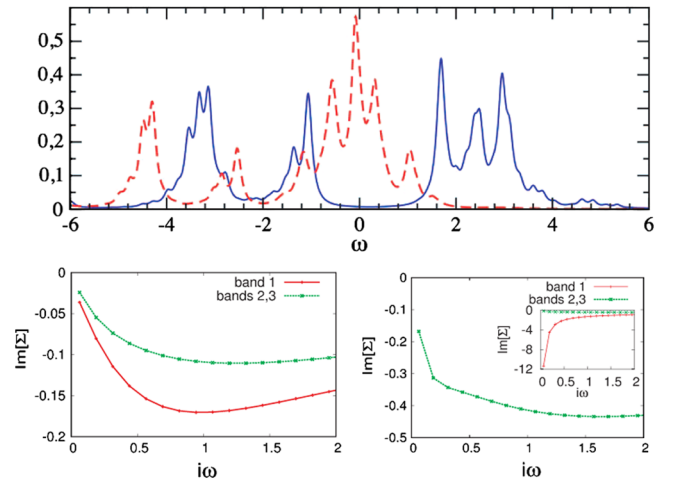


FIG. 3 (color online). Upper panel: local spectral densities for the lifted (solid blue) and the degenerate (dashed red) bands in the Orbital-selective Mott phase. (calculations in DMFT-ED with $N_s = 12$ for $U/D = 4.5$). Bottom panels: self-energies, respectively, in the metallic ($U/D = 1.0$ —left) and orbitally selective ($U/D = 2.4$ —right) phases.

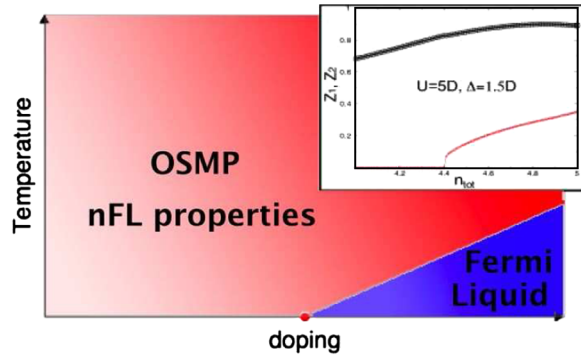


FIG. 4 (color online). Sketch of the general phase diagram of a doped OSMP. Inset: quasiparticle residues as a function of the filling in SSMF for $U = 5$, $J/U = 0.25$ and $\Delta = 1.5$. The OSMP turns into a Fermi liquid at a finite doping.

filled two-band Mott insulator with a Hund's coupling if the two levels are shifted. In our model a similar mechanism applies: in the OSMP the chemical potential can move within the gap of the localized band (See Fig. 3) without doping it, and at the same time dope the gapless metallic bands without spoiling the OSMP. Then at some critical doping we expect the chemical potential to hit the Hubbard band of the localized electrons, leading to a doping also of this band, and to its metallic behavior. Indeed numerical results confirm this scenario that bares substantial analogies with previous calculations on doped OSMP's [4,27]. As we see in the inset of Fig. 4 for a set of parameters not far from the border of the phase, the OSMP is quite robust with respect to doping, then eventually at a *finite critical doping* the OSMP leaves the place to a normal Fermi liquid. It is interesting to notice that the general phase diagram of a doped OSMP is reminiscent of the behavior of the iron-pnictide superconductors (and also of cuprates). Doping leads from a "strange phase" in which metallic and insulating properties coexist, to a more regular Fermi liquid.

From a general perspective, the electronic configuration of the pnictides [28] is ideal for our mechanism to apply: Coulomb correlations and Hund's coupling are sizeable and the population of 6 electrons in 5 bands is even more favorable than the one we studied. Here the starting point in the absence of splittings is $n = 6/5$ for each band, suggesting that a smaller crystal-field splitting may be sufficient to make the highest band half-filled, and therefore localized (or quasilocalized).

In summary we have outlined a new mechanism for the OSMT: a crystal field can split a multiband system in manifolds of different degeneracy, in which correlated electrons have different kinetic energy despite the equal bandwidth. If the crystal field and the Hund's coupling are large enough, the suppression of the orbital fluctuations decouples the manifolds and allows a selective localization of the lifted band at intermediate couplings. Importantly, the result occurs in a wide range of model parameters,

suggesting that this mechanism can be realized in several systems [29]. We have also checked (see Fig. 1) that the OSMP is solid with respect to further degeneracy lifting of the lower bands, at least for small to intermediate splittings. Even if all the electrons come from bands of the same bare dispersion, the OSMP is a highly nonconventional metal resulting from the selective localization: it has a non-Fermi-liquid behavior and a Curie-Weiss magnetic response, and it is generally unstable to a normal Fermi-liquid phase at a finite doping.

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