Non-Fermi Liquid Behavior and Double-Exchange Physics in Orbital-Selective Mott Systems

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We study a multi-band Hubbard model in its orbital selective Mott phase, in which localized electrons in a narrow band coexist with itinerant electrons in a wide band. The low-energy physics of this phase is shown to be given by a generalized double-exchange model. The high-temperature disordered phase thus differs from a Fermi liquid, and displays a finite scattering rate of the conduction electrons at the Fermi level, which depends continuously on the spin anisotropy.

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The Mott phenomenon, localization of electrons by their mutual interactions, is a key phenomenon in correlated electron materials. Recently, there has been great interest in the possibility of an “orbital-selective Mott phase” (OSMP), in which a Mott transition occurs within a restricted subset of quasi-degenerate orbitals, so that a narrow band of localised electrons coexists with a wider band of itinerant electrons. Originally proposed [1] for the ruthenate compound Ca₃Sr₂RuO₆, this possibility has been intensively discussed since then [2, 3, 4, 5, 6, 7].

In this letter, we clarify the nature of such an orbital-selective Mott phase. We show that for finite Hund’s coupling the orbital-selective Mott transition is accompanied by a breakdown of Fermi liquid theory, which can be understood from a mapping onto an effective double-exchange model at low energy. In the high-temperature disordered phase, the electrons in the conduction band acquire a finite lifetime, due to the scattering on the localised spins. This behavior is eventually cut-off by long-range ordering at low temperature. Detailed calculations within dynamical mean-field theory (DMFT) are performed in order to demonstrate these properties.

We consider the two-orbital Hubbard model for a broad and a narrow band, described by creation operators c_iσ and d_iσ respectively, with hamiltonian:

\[ H = -\sum_{ij\sigma} \left( t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + t_{ij} d_{i\sigma}^\dagger d_{j\sigma} \right) \]

\[ + \sum_i \left( U \sum_{m=c,d} \bar{n}_{i\sigma}^m \bar{n}_{i\sigma}^m + U' \sum_{\sigma\sigma'} \bar{n}_{i\sigma}^c \bar{n}_{i\sigma'}^d - J \sum_{\sigma} \bar{n}_{i\sigma}^c \bar{n}_{i\sigma}^d \right) \]

\[ + \alpha \left[ H_{\text{flip}} + H_{\text{pair}} \right] \]

In this expression, U and U’ are Coulomb interaction parameters and J is the Hund’s rule coupling. We have used the notation \( \bar{n}_{i\sigma}^m = c_{i\sigma}^\dagger c_{i\sigma} - 1/2 \) (similarly, \( \bar{n}_{i\sigma}^d \)) and written the hamiltonian in the manifestly particle-hole symmetric case corresponding to two electrons per site (\( n^c = n^d = 1 \)), on which we shall focus in the following. The spin-flip and pairing hopping terms of the hamiltonian read: \( H_{\text{flip}} = -J \sum_i [c_i^\dagger c_i d_i^\dagger d_i + \text{h.c.}] \) and \( H_{\text{pair}} = -J \sum_i [c_i^\dagger c_i d_i^\dagger d_i + \text{h.c.}] \). A variable parameter \( \alpha \) has been introduced in front of these terms, for further use. For a cubic environment and unbroken spin symmetry, the relations \( U' = U - 2J \) and \( \alpha = 1 \) hold. This model has been the subject of several recent studies using dynamical mean-field theory (or equivalently, in the limit of infinite lattice connectivity). For anisotropic interactions \( U' < U \) and finite \( J \) an orbital selective Mott phase is found even at moderate values of the ratio of the two bandwidths \( \tilde{D} \). As shown recently [1, 4, 5], for large differences in the bandwidths the OSMP can be induced even in the case of SU(4)-symmetric interactions \( U = U', J = 0 \).

First, we consider this model for \( \alpha = 0 \), i.e with density-density interactions only, and make a comparative study of the nature of the paramagnetic OSMP in the absence \( (J = 0) \) or in the presence \( (J \neq 0) \) of the Hund’s coupling. The model is solved using DMFT, for two semi-elliptical densities of states (infinite connectivity Bethe lattice) \( \rho_{\text{H}}(\epsilon) = 2[1 - (\epsilon/D_c)^2]^{1/2} / (\pi D_c) \) (similarly \( \rho_{\text{dd}} \)), with a bandwidth ratio \( D_d/D_c = 0.1 \). We use \( D_c = 1 \) as our unit of energy, choose \( U = 0.8 \), \( U' = U - 2J \), and compare \( J = 0 \) to \( J = 0.2 \). In both cases, the self-energy of the narrow band (not shown) diverges at low frequency, while the self-energy of the broad band does not, indicating that the narrow band is Mott-localized while the broad band is not (OSMP regime). However, the low-frequency behavior of the self-energy is very different in each case, as shown on Fig. 1 which displays our DMFT results obtained with the QMC algorithm of Hirsch and Fye. It is seen that, for \( J = 0 \), the self-energy extrapolates to zero at low frequency \( \lim_{\omega \to 0} \text{Im} \Sigma_c(\omega) \sim (1 - 1/Z_c) \omega + \cdots \) in a Fermi-liquid manner, while for \( J \neq 0 \), it extrapolates to a finite value \( \lim_{\omega \to 0} \text{Im} \Sigma_c(\omega) \neq 0 \). Hence, a finite lifetime is found at the Fermi level for \( J \neq 0 \), so that no well-defined Fermi-liquid quasiparticles exist. Correspondingly, this implies a violation of the Luttinger sum-rule \( \rho_c(\omega) = \rho_{\text{dd}}(0) \) for the spectral function \( \rho_c(\omega) \equiv -\text{Im} G_c(\omega + i\epsilon) / \pi \). The local Green’s function being related to the self-energy by: \( G_c(\epsilon) = \int d\epsilon' \rho_c(\epsilon') / (\omega - \Sigma_c(\epsilon') - \epsilon) \), a non-zero lifetime implies: \( \rho_c(0) = \int d\omega \rho_{\text{dd}}(\epsilon) (\pi(\epsilon^2 + \Gamma_c^2)) < \rho_{\text{dd}}(0) \).
On Fig. 2, we display the spectral functions of the narrow and broad bands, obtained by a maximum-entropy continuation of the QMC data. Pinning at the Luttinger theorem value \( \rho_c(0) = \rho_{\beta D}(0) = 2/(\pi D_c) \) holds for \( J = 0 \) but not for \( J \neq 0 \). This is consistent with Fig.3 in [4]. Note that the spectral function of the (localised) narrow band has a sharp Mott gap in the latter case, but displays rather a pseudogap in the former (as previously discussed in [4, 5]). The Green’s functions obtained for \( J = 0.2 \) using QMC and using an exact diagonalisation (ED) solution of the DMFT equations (inset of Fig. 1) are in perfect agreement.

![Graph showing spectral functions](image)

**FIG. 1**: Broad band self-energy \( \text{Im} \Sigma_c(\omega_n) \) for different temperatures (\( \beta D_c = 40, 60, 80, 120 \) correspond to circles, empty and filled squares and diamonds respectively). In the \( J = 0 \) case (online: black) \( \text{Im} \Sigma_c(\omega_n) \) extrapolates linearly to zero (Fermi liquid behavior), whereas for \( J = 0.2 \) (online: red) a finite lifetime is found. Inset: \( \text{Im} G_c(\omega_n) \) and \( \text{Im} G_c(\omega_n) \) for \( U = 0.8 D_c \) and \( J = 0.2 D_c \). QMC results (symbols) correspond to finite temperatures (\( \beta D_c = 40, 60, 80, 120 \)) given by circles, squares, triangles up and down respectively, while the ED data (lines) are at zero temperature.

In order to understand the origin of this finite-lifetime, non-Fermi liquid behavior, we now derive an effective hamiltonian valid at low-energy in the orbital-selective regime. There, it is expected to be legitimate to neglect entirely the hopping term of the narrow band (which renormalizes to zero at low-energy). Furthermore, since we are interested in low-energy physics, the states with a hole or a double-occupancy in the narrow orbital can be eliminated. Setting \( t^d = 0 \) and \( \tilde{n}^d_c = \pm 1 \) in (2), a straightforward calculation leads to the effective hamiltonian:

\[
\begin{align*}
H_{\text{eff}} &= -\sum_{i,j,\sigma} t^\sigma_{ij} c^\dagger_i c_j + U \sum_i \tilde{n}^c_i c^\dagger_i \tilde{n}^c_i - 2J \sum_i \left[ S^x_{id} S^z_{ic} + \alpha (S^x_{id} S^x_{ic} + S^y_{id} S^y_{ic}) \right] \\
&\quad - 2J \sum_i \left[ S^x_{ic} S^z_{id} + \alpha (S^x_{ic} S^x_{id} + S^y_{ic} S^y_{id}) \right]
\end{align*}
\]

(2)

In this expression, \( S^x_{id}, S^y_{id}, S^z_{id} \) are the components of the spin-1/2 operator acting on the 2 local moment states \( \uparrow \rangle_d \rangle, \downarrow \rangle_d \rangle at each site, while \( S^x_{ic} \equiv c^\dagger_{ic} c_{ic} \), \( S^y_{ic} \equiv c^\dagger_{ic} c_{ic} \), \( S^z_{ic} \equiv c^\dagger_{ic} c_{ic} \) are the components of the itinerant electron spin operator. Note that the inter-orbital Coulomb interaction \( U' \) entirely disappears from this effective hamiltonian (as well as the pair-hopping term, which cannot act in the low-energy subspace of sites with single-occupancy of the \( d \)-band). Hence the low-energy effective hamiltonian in the orbital-selective phase is a ferromagnetic Kondo lattice, with an additional Hubbard interaction in the (broad) itinerant band. The orbital-selective Mott localisation of a single orbital component generates an effective double-exchange model in which the itinerant orbital interacts with the “core-spin” of the localised orbital through the Hund’s coupling. This observation has two important consequences, to be explored in more detail below: (i) it explains the observed non-Fermi liquid behavior, and its dependence on the coupling to spin-flip terms \( \alpha \) and (ii) it suggests that ultimately the orbital-selective phase undergoes long-range ordering as temperature is lowered.

When the interactions are of the density-density form (\( \alpha = 0 \)), the itinerant orbital couples to the core-spin through Ising terms only. The effective hamiltonian [4]
then essentially reduces to a Falicov-Kimball model for each spin species, in which the conduction electrons interact with a given configuration of the Ising variables $S_i^\sigma$, which are conserved quantities. In addition, the two spin components of the itinerant band are coupled by the intra-orbital Hubbard interaction. In order to solve this model, one must consider a given configuration of the Ising spins, solve the one-band model of conduction electrons in that configuration, and average over all Ising spin configurations with appropriate Boltzmann weight. This process can be achieved within DMFT (infinite coordination) by mapping the problem onto an effective single-site action:

$$ S_{\text{eff}}^{\alpha=0} = -\int d\tau \int d\tau' \sum_{\sigma} (c_\sigma(\tau) G_{0}^{-1}(\tau - \tau') c_\sigma(\tau')) $$

$$ + \int d\tau [U \bar{n}_\tau \bar{n}_\tau - JS_z (n_\tau - n_\tau)] $$

subject to the self-consistency condition $G_{0}^{-1}(i\omega_n) = i\omega_n - t^2 G_c(i\omega_n)$. Because $S^\sigma$ is a conserved quantity, each sector $S^\sigma = \pm 1/2$ can be considered independently. In each sector, the effective action is that of an effective Anderson impurity model in a local magnetic field $\pm J$. The itinerant electron Green’s function is obtained as: $G_c = 1/2 [G_{\uparrow \uparrow} + G_{\downarrow \downarrow}]$, where $G_{\sigma \sigma}$ denotes the Green’s function $-\langle T c_{\sigma}(\tau) c_{\sigma}^\dagger(\tau') \rangle$ in the effective action with $S^\sigma = \pm 1/2$, and the obvious symmetry property $G_{\sigma \downarrow} = G_{\sigma \uparrow}$ holds. In writing these expressions, a paramagnetic state has been assumed and particle-hole symmetry corresponding to half-filling has been used. In Fig. 3 we compare the Green’s function obtained by solving this effective model to the full solution of the original model in the OSMP. The two results are indistinguishable for all practical purposes, hence firmly establishing the validity of our low-energy effective model in this phase.

For a given orientation of the core spin (say, $S^z = +1/2$), the Green’s functions of the itinerant electrons for each spin component correspond to those of an impurity model in a local magnetic field, and hence read: $G_{\uparrow \uparrow}(i\omega_n)^{-1} = G_0^{-1} + J/2 - \Sigma_{\uparrow \uparrow}(i\omega_n)$, $G_{\downarrow \downarrow}(i\omega_n)^{-1} = G_0^{-1} - J/2 - \Sigma_{\downarrow \downarrow}(i\omega_n)$, with the symmetry relation (at half-filling): $\Sigma_{\uparrow \uparrow}(i\omega_n) = -\Sigma_{\downarrow \downarrow}(-i\omega_n)$. In these expressions, $\Sigma_{\sigma \sigma}$ is the self-energy originating from the intra-orbital Hubbard interaction. In the itinerant (OSMP) phase for the conduction electrons, these spin-resolved self-energies for a given core spin orientation do have a Fermi-liquid form at low energy: $\Sigma_{\uparrow \uparrow}(i\omega_n) = \Sigma_0 + (1 - 1/Z)i\omega_n + \cdots$, with $\Sigma_0$ a real quantity. $\tilde{J} = J - 2\Sigma_0$ can be viewed as the effective exchange coupling renormalised by the on-site Hubbard interaction. Inserting these expressions into $G_c = 1/2 [G_{\uparrow \uparrow} + G_{\downarrow \downarrow}]$, and taking into account the self-consistency condition $G_0^{-1} = i\omega_n - t^2 G_c$ allows us to obtain the low-frequency form of the full self-energy $\Sigma_c$ in the form:

$$ \Sigma_c(\omega + i0^+) = -i\Gamma_c + (1 - 1/Z_{\text{eff}})\omega + \cdots $$

$$ \Gamma_c = \frac{\tilde{J}^2}{2(4t_1^2 - t_2^2)^{1/2}}, Z_{\text{eff}}^{-1} = Z^{-1} \frac{(\omega - \tilde{J})^2}{(4t_1^2 - t_2^2)} $$

Hence, a finite lifetime at zero energy emerges, due to the scattering of the itinerant electrons onto the core spin associated with the localised orbital. Because the core $S^z$ is a conserved quantity in the absence of spin-flip ($\alpha = 0$), the physics is closely related to that of the Falicov-Kimball model, in which this kind of non-Fermi liquid behavior was first emphasized by Si et al. [10], and later by Furukawa [12] for double-exchange models in the context of manganites (see also [13, 14, 15, 16]).

An obvious question is whether this finite lifetime is a robust feature when spin-flip terms ($\alpha \neq 0$) are reintroduced. In that case, we have to deal with a ferromagnetic Kondo lattice model, with full $SU(2)$ symmetry at $\alpha = 1$ and Ising anisotropy at $\alpha < 1$. Within DMFT, this model maps onto a ferromagnetic Kondo impurity problem, with a self-consistent conduction electron bath. The renormalisation flow of the ferromagnetic Kondo problem is such that the spin-flip component of the interaction is irrelevant as long as $\alpha < 1$, with a continuous family of infra-red fixed points corresponding to X-ray edge physics. Hence, we expect a finite lifetime to survive as long as the $SU(2)$ symmetry is broken (i.e for all $\alpha < 1$). We have solved the DMFT equations for the original 2-orbital model in the presence of spin-flip (and pair-hopping) terms for a range of values of $\alpha$, in the orbital-selective phase. The results are displayed in Fig. 4 and indeed fully support this expectation, with a violation of Luttinger’s sum rule gradually decreasing as $\alpha$ is increased. In the fully symmetric case $\alpha = 1$, the lifetime does extrapolate to zero (Fig. 4) as $\omega \to 0$, but conventional Fermi liquid behavior is not recovered. Indeed, single-impurity Kondo scaling suggests a singular
behavior $\text{Im}\Sigma \sim -1/\ln(\omega)^2$, consistent with the fit of our numerical results displayed in Fig. 4.

![FIG. 4: Low-energy part of $\text{Im}\Sigma_c(\omega)$, obtained by ED at $T = 0$ for $U = 1.2D_c$, $J = 0.15U$, and different values of $\alpha$ (0, 0.3, 0.5, 0.7, 1 from bottom to top). It extrapolates to zero at $\omega = 0$ only in the case of fully symmetric interactions ($\alpha = 1$, red solid curve). Inset: Zero-frequency extrapolation of the Green’s function as a function of $J$ and for different values of $\alpha = 1, 0.7, 0.5, 0$ (from bottom to top). In the case of a fully $SU(2)$ symmetric interaction vertex ($\alpha = 1$) the Luttinger pinning condition $\text{Im}G(0) = -2/D_c$ is fulfilled. As soon as this symmetry is broken ($\alpha < 1$), the pinning condition is lost.](image)

The calculations described above are restricted to the paramagnetic phase, spin symmetry being enforced on the Green’s functions. Clearly, at low temperatures, a phase with long-range order is more favorable. The competition between ferromagnetism, antiferromagnetism and phase separation has been studied in detail for double exchange models relevant to manganites (see e.g. [12, 14, 18, 19]). Fewer studies have taken into account the effect of an on-site Hubbard interaction, which increases the tendency to ferromagnetism [18]. Such a detailed study for the present model is beyond the scope of this paper. We have however performed some calculations without spin-symmetrisation and do find a ferromagnetic solution to be stable below a temperature of order $D_c/50$. The inset of Fig. 4 demonstrates how a ferromagnetic state, by suppressing spin disorder, restores a scattering rate which decreases as temperature is lowered and as the magnetic polarisation increases.

In conclusion, we have shown that the low-energy physics of the orbital-selective Mott phase is given by a double exchange model (with an additional Hubbard repulsion). As a result, in the high-temperature disordered phase a non-Fermi liquid state with a finite lifetime of the conduction electrons is found. At lower temperature, long-range spin ordering and possibly phase separation sets in. By introducing a finite crystal field splitting, it is clearly seen that the double-exchange physics and that of the OSMP are continuously connected, the former corresponding to a large crystal field splitting and a core spin being formed in the lower crystal field level, while the latter corresponds to a small crystal field and Mott localisation of the narrow band. Conversely, the OSMP is a useful way of simulating the double exchange model with fully quantum spins using DMFT techniques. Finally, it is tempting to speculate that a selective localisation of electrons in momentum space may lead to short lifetimes over part of the Fermi surface, as observed in the cuprates.

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