Eur. Phys. J. B **2**, 457–461 (1998)

THE EUROPEAN PHYSICAL JOURNAL B

A scenario for the electronic state in the manganese perovskites: the orbital correlated metal

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Received: 3 October 1997 / Revised: 9 December 1997 / Accepted: 12 January 1998

Abstract. We propose a novel scenario for the electronic state in the manganese perovskites. We argue that, at low temperatures and within the ferromagnetic state, the physics of these colossal magnetoresistance compounds may be characterized by a correlated metallic state near a metal insulator transition where the orbital degrees of freedom play the main role. This follows from the observation that a two-band degenerate Hubbard model under a strong magnetic field can be mapped onto a para-orbital single band model. We solve the model numerically using the quantum Monte-Carlo technique within a dynamical mean field theory which is exact in the limit of large lattice connectivity. We argue that the proposed scenario may allow for the qualitative interpretation of a variety of experiments which were also observed in other (early) transition metal oxides.

PACS. 75.50.Cc Other ferromagnetic metals and alloys – 72.10.Fk Scattering by point defects, dislocations, surfaces, and other imperfections (including Kondo effect) – 71.20.Be Transition metals and alloys

There is a great current interest in transition metal compounds displaying colossal magnetoresistance (CMR). This effect is a strong dependence of resistivity with the applied magnetic field and is observed experimentally in compounds such as $La_{1-x}A_xMnO_3$, with $A = Sr, Ca, Pr$.

From the point of view of the electronic structure, these systems have 3 electrons in a t_{2g}^3 band which due to a strong Hund's rule coupling form a core $3/2-$spin at each Mn site and $1-x$ electrons that go into a quasi two-fold degenerate e_g band. For $x = 0$ the Manganese atoms are $(+3)$ and the compound has a nominal filling equal to 1. Since the bands are originated from partially filled d-orbitals, on general grounds one expects that correlation effects should play an important role in the low energy behavior, which is indeed the case in many other transition metal oxides (TMO) with the perovskite structure [1].

At $x = 0$ the parent compound, $LaMnO_3$, is an insulator with a layer antiferromagnetic structure. The spins are ferromagnetically ordered within a layer and the antiferromagnetic order is in the direction perpendicular to the layers. (*i.e.*, stacks of alternating ferromagnetic layers.) Upon doping one finds a transition to a ferromagnetically ordered state. The basic physics of this class of CMR compounds seems to be qualitatively captured by the "double exchange" mechanism proposed by Zener almost fifty years ago [2]. Anderson and Hasegawa provided an explicit realization of this mechanism [3], which was subsequently taken over by de Gennes [4] to demonstrate that, within a mean field treatment, a 3-dimensional lattice at small doping can also show "canted" antiferromagnetic spin order. Kubo and Ohata [5] worked out the mean field theory of the ferromagnetic Kondo lattice model which was considered a minimal model for the manganese oxides, and obtained a large negative magnetoresistance which, however, was not in good agreement with experiments. More recently, Furukawa [6] investigated the same model using a recently developed technique, the dynamical mean field theory [7], to show that it does account for many experimental observations, most notably the dependence of the magnetization with the resistivity. While the studies on the ferromagnetic Kondo lattice model demonstrate that the double exchange mechanism is certainly a main ingredient, additional interactions must be added to obtain a more correct picture [8]. Among those, the dynamical Jahn-Teller effect [9] was studied in detail [10,11] and there is experimental evidence of strong polaronic effects in many compounds [12]. In spite of this recent progress, our understanding of the CMR compounds is still incomplete. In particular at low temperature and within the ferromagnetic (FM) phase, where there are several experimental observations that remain unaccounted for: i) the enhancement of the density of states near the Fermi energy with decreasing T observed in photoemission [13,14], ii) the unusual redistribution of optical spectral weight as function of the temperature which occurs in the range of the eV [15], iii) the strong enhancement of the A coefficient of the T^2 term in the resistivity as function of the composition in $La_{1-x}Sr_xMnO_3$ [16], and iv) the suppression of the resistivity with applied pressure [17].

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These features are not exclusive of the CMR manganese compounds, on the contrary, they have been observed in other (early) transition metal oxides with perovskite structure. For instance, in V_2O_3, a redistribution of spectral weight in the optical response [18] and a small enhancement of the density of states near the Fermi energy in the photoemission [19] is observed as the temperature is lowered within the paramagnetic metallic phase. The $La_xSr_{1-x}TiO_3$ system, on the other hand, displays a notable increase of the T^2 coefficient of the resistivity as a function of x [20]. Both compounds also show suppression of the resistivity with applied pressure [21,22]. These experiments on early TMO have recently received a qualitative interpretation within a dynamical mean field theory of the Hubbard model that becomes exact in the limit of large dimensions [23–25,7]. The key feature is to realize that the mean field solution of the Hubbard model predicts a quasiparticle resonance at the Fermi level when the system is close to a metal insulator transition (MIT). Thus, the proximity to a MIT provides with a dynamically generated small energy scale which allows for the qualitative interpretation of the unusual behavior observed in the experimental compounds.

In regard of these similarities between some early TMO and the manganese perovskites, one is motivated to consider the question whether a similar underlying mechanism may be responsible for the low energy behavior. However, an apparent reason that would mean to immediately discard this idea is that the observed phenomena in the CMR compounds occur as they evolve into a ferromagnetic (FM) state with a magnetic moment that saturates close to the classical value. Naively one may think that this is incompatible with the presence of quasiparticles with an enhanced renormalized mass. Thus, the goal of this paper is to introduce a model which contains some realistic features which where not previously considered, namely local Coulomb repulsion and band degeneracy [26], and demonstrate that in the parameter regime relevant for the CMR manganates the system remains close to a MIT with a dynamically generated small energy scale as it goes in to the fully polarized FM state at low temperatures.

The full Hamiltonian for the CMR compounds has the following form [10,7]:

$$H = \sum_{\langle ij\rangle,a,b,\sigma} t_{ij}^{ab}\ c_{ia\sigma}^{\dagger} c_{jb\sigma} - J_H \sum_{i,a,\sigma} \mathbf{S}_{ci}\cdot\mathbf{s}_{di} + \quad H_{J-T} + \sum_{i,a,b,\sigma,\sigma'} U_{ab\sigma\sigma'} n_{ia\sigma} n_{ib\sigma'}(1-\delta_{ab}\delta_{\sigma\sigma'}) \qquad (1)$$

where $\sigma = \pm 1/2$, $a,b = 1,2$ are the orbital indexes of the e_g bands, and the local spins $3/2$ are described by $\mathbf{S}_c$. The first two terms define the ferromagnetic Kondo lattice model and give a realization of the DEM while the third is a Jahn-Teller term that brings in polaronic effects. The role of the dynamical correlations due to the local Coulomb repulsion described by the fourth term still remains largely unaccounted and is a main concern in this work. Recent resonant photoemission experiments indicate that the manganese parent compound is in an intermediate state between a charge transfer and Mott-Hubbard insulator with $U \sim 3.5$ eV [13]. It is important to point out that this energy is similar to the value of the ferromagnetic Kondo coupling, which casts some doubts on the appropriateness of the commonly used simple ferromagnetic Kondo lattice. Another important aspect which we shall consider explicitly here is the orbital degeneracy, and, as it turns out, we shall see that these degrees of freedom will play a crucial role within the proposed scenario.

In order to better focus on the role of local repulsion and orbital degeneracy we shall simplify the Hamiltonian (1). Firstly, as we are concerned with the electronic state in the FM phase we neglect H_{J-T}. This term is most relevant around and above T_c, but moving deep into the FM metallic phase its strength rapidly decreases [27,28,10]. Also, well into the FM phase, we can assume the local spins $3/2$ to be uniform and static. Therefore, by means of the ferromagnetic coupling J_H, their main effect on the conduction electrons is to produce an effective local magnetic field h_{loc} as they become polarized under the action of an implicit external field. Once more, we emphasize that the driving force of the FM state is in the double exchange mechanism. It is important to note that the effective local field, being of electronic origin, may be very strong and is estimated to be about twice the bandwidth [6].

Thus our effective model Hamiltonian becomes,

$$H_{eff} = \sum_{\langle ij\rangle,a,b} t_{ij}^{ab}\ c_{ia\sigma}^{\dagger} c_{jb\sigma} + \sum_{i,a,b,\sigma,\sigma'} U_{ab\sigma\sigma'} n_{ia\sigma} n_{ib\sigma'}(1-\delta_{ab}\delta_{\sigma\sigma'}) - h_{loc}\mathbf{m}_i\cdot\mathbf{s}_i \qquad (2)$$

where $\mathbf{m}_i^2 = 1$.

It is important to state that our simplified model is not an alternative to the double exchange mechanism. We are rather assuming that double exchange is indeed responsible for the basic physics such as the phase diagram and some of the CMR behavior, and we are trying to build up the ingredients which may provide for a qualitative understanding of the experimental observations mentioned before. As it turns out, we shall see that the orbital degrees of freedom will be playing a crucial role within the proposed scenario.

We shall solve this model within the dynamical mean field theory which becomes exact in the limit of large$-d$, with d being the number of spatial dimensions. This limit is equivalent to the limit of large lattice connectivity. As usual, in order to get a non-trivial problem allowing the itinerant and local terms in the Hamiltonian to compete, the hopping is renormalized $t \to t/\sqrt{d}$ [29]. We shall assume a semi-circular density of states $\rho^0(\epsilon) = 1/(\pi D)\sqrt{1-(\epsilon/D)^2}$, with the half bandwidth $D = 1$ and $D = 2\sqrt{2}t$. This ρ^0 is realized in a Bethe lattice, and our choice is motivated by both, simplicity and the realistic finite bandwidth that it provides.

In order to numerically solve the model we shall make some further non crucial simplifications. Firstly, we take $U_{ab\sigma\sigma'} = U$. Secondly, we set $t_{ij}^{ab} = -t\ \forall a,b$, as on general grounds, one may expect that the off-diagonal

hopping amplitudes have a similar magnitude as compared to the diagonal ones. This choice actually forces the system to remain orbitally disordered even at zero doping; while if we choose $t^{aa} \neq t^{ab}$, we would have an orbitally ordered state (analogous to an AFM) at zero doping with a small ordering temperature $T_N^{orb} \approx 4[(t^{aa})^2 - (t^{ab})^2]/U$ [23]. In any case, one should keep in mind that in the low doping limit our effective model Hamiltonian is no longer applicable for the CMR compounds since the Jahn-Teller splitting becomes relevant. It is important to emphasize that our choice of a single value for the different hopping amplitudes is made just for simplicity. The main role of the off-diagonal hopping is to contribute to the stability of the para-orbital state that we shall encounter latter on. On the other hand, one should also realize that there is a second, and possibly more important, feature that assures the stability of the disordered orbital state, namely, the rather large doping $x \gtrsim 0.175$, which is experimentally necessary for the CMR state to occur. Doping an ordered Mott insulator favours disordered states, which is a familiar fact from the physics of the high T_c superconductors. In the usual one band Hubbard model one finds a spin paramagnetic state upon doping the antiferromagnetic state at $x = 0$, while in our case, since the orbital degrees of freedom of our Hamiltonian are playing a role analogous to the spin, it is an orbitally disordered state that results upon sufficient doping. Thus, it is this second and important effect what actually renders our simplifying choice of a single valued hopping $t_{ij}^{ab} = t$ non-crucial, as any non-extremely asymmetric choice for the hopping amplitudes will leave our results unmodified.

The Hamiltonian can now be mapped onto its associated impurity problem (a degenerate Anderson impurity in a magnetic field), which is supplemented with a self-consistency condition that enforces the translational invariance [30–32]. The self-consistency condition reads,

$$[\mathcal{G}^0_{\sigma a}(z)]^{-1} = z + \mu - \sigma h_{loc} - t^2[G_{\sigma a}(z) + G_{\sigma b}(z)] \quad (3)$$

where $\mathcal{G}^0$ and G denote local Green functions, and μ the chemical potential. It should be now clear from equation (3) how the role of the off-diagonal hopping that we discussed above is to provide *frustration* in orbit space, thus favoring the stability of a para-orbital (orbital disordered) state. From the equation we see that the electronic mean field at each site is the *average* of both orbital reaction fields $t^2 G_{\sigma a}$ and $t^2 G_{\sigma b}$. This situation is in close analogy with the spin-frustrated two sublattice model introduced in reference [23].

We numerically solve the model using a quantum Monte-Carlo technique [32–34]. To demonstrate that the model remains near a MIT as it goes into the FM phase we shall compute the mass renormalization m^*/m. In the limit of large dimensions the self-energy $\Sigma = {\mathcal{G}^0}^{-1} - G^{-1}$ is local, thus, $m^*/m = 1 - \partial\Sigma/\partial\omega$. Since our results are obtained at low but finite temperature we shall estimate this value using $m^*/m \approx 1 - \Sigma(\omega_1)/\omega_1$, where $\omega_1 = \pi T$ is the first Matsubara frequency. Another quantity that we shall obtain is $\langle n \rangle$ *versus* μ with n the particle number. The slope of this curve is proportional to the compressibility, therefore the (Mott) insulating states will be indicated by plateaux.

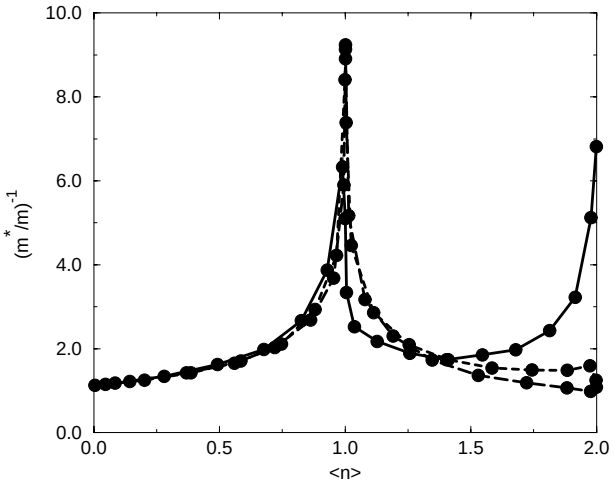

Fig. 1. Renormalized mass as a function of the particle occupation for $U = 3$, $T = 1/8$ and $h_{loc} = 0,\ 0.5,\ 1$ (full, dashed, long dashed).

The physics of the two band degenerate Hubbard model has been recently considered within the dynamical mean field approach [32,35]. One of the main results was that, within the paramagnetic state, the phase diagram shows lines of Mott insulating states at integer fillings for values of the interaction $U > U_c(n)$ and low enough temperatures. As these lines are approached as a function of filling, a divergency in the renormalized mass is observed which signals a correlated metallic state with an effective Fermi energy which vanishes as $(m^*/m)^{-1}$. These features are reminiscent of the solution of the single band Hubbard model which is known in quite detail [36,7]. The key observation that we shall demonstrate in this paper is that the two band model with $n \leq 1$ and moderate U is near a MIT line even as it goes into a saturated FM state. Thus, irrespective of the magnetization the system always remains in a correlated metallic state with a reduced effective Fermi energy $\sim \frac{m}{m^*} D$. The underlaying reason is simple, as the electrons become fully polarized one may map the two band model into a *single band* Hubbard model where the usual role of the spin indices is played by the orbital ones. In other words, under a strong magnetic field the operators carrying a, say, $\downarrow$ spin disappear from the effective Hamiltonian (2).

In Figure 1 we show m^*/m as a function of the number of particles and different magnetic fields [37]. The most striking feature is how, for any field, the mass renormalization maintains its divergent behavior when $\langle n \rangle \to 1$ from below. As we argued above this occurs because the system crossover from two- to one-band behavior. In particular note that for the highest h_{loc} the m^*/m plot shows symmetry around $\langle n \rangle = 1$ as is the case in a single band model. Our results predict that the compounds with $x \gtrsim 0.175$ should have an $m^*/m \lesssim 3$ which is consistent with the enhancement observed in experiments on $La_{1-x}Sr_xMnO_3$ [38].

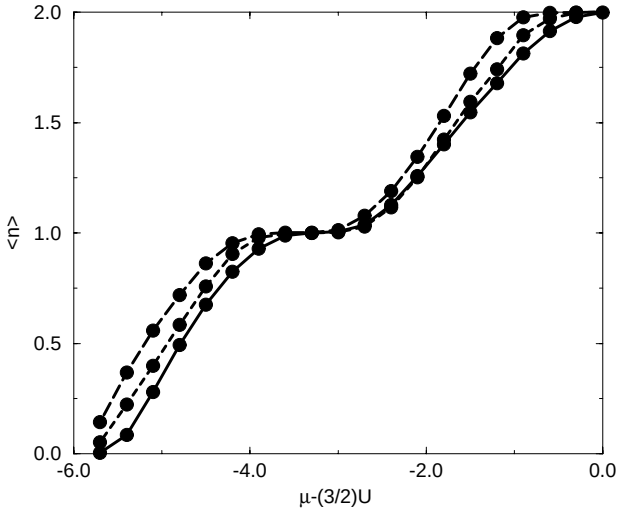

Fig. 2. Particle occupation as a function of the chemical potential for $U = 3$, $T = 1/8$ and $h_{loc} = 0,\ 0.5,\ 1$ (full, dashed, long dashed).

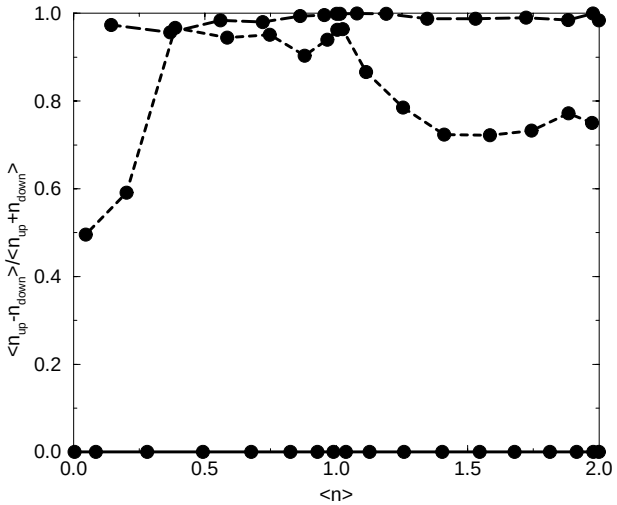

Fig. 3. Relative magnetic moment of the conduction electrons as a function of the particle number for $U = 3$, $T = 1/8$ and $h_{loc} = 0,\ 0.5,\ 1$ (full, dashed, long dashed).

In Figure 2 we show the number of particles $\langle n \rangle$ as a function of μ for different local magnetic fields. We observe that plateaux are always present for fillings $\langle n \rangle = 1$ and 2 even in the case of a strong h_{loc}. Since the slope of the curves is proportional to the compressibility, the system becomes an insulator at those fillings. In the case of $\langle n \rangle = 1$ we argued before that under a strong local magnetic field the Hamiltonian maps onto a half-filled para-orbital single band model, therefore, the insulating state corresponds to a Mott-Hubbard insulator for all h_{loc}. However, the character of the insulating state at $\langle n \rangle = 2$ strongly depends on h_{loc}. For $h_{loc} = 0$ the state is a Mott insulator since the bands are both half-filled. On the other hand, in the polarized state the insulator should be better thought of as a band insulator since the bands can accommodate only one electron each.

The previous discussion relayed heavily on the assumption that for $h_{loc} = 1$ the system is fully polarized. Thus, in Figure 3 we plot the relative magnetic moment $\langle n_\uparrow - n_\downarrow \rangle / \langle n_\uparrow + n_\downarrow \rangle$ *versus* $\langle n \rangle$ at different magnetic fields in order to check the validity of the assumption. We observe that at zero field there is no magnetic moment as expected while for the largest field the magnetic moment is close to unity which indicates that all the electrons are almost fully polarized.

It is interesting to comment on the behavior of the magnetization in the intermediate case [39], which illuminates aspects of the competition between coherence and magnetization. At small fillings, the correlation effects due to the on-site repulsion are not important and the magnetic moment relative to the particle number is rather small. As the particle occupation increases the correlation effects become more important (the effective mass increases) and the magnetic moment grows rapidly due to the enhanced susceptibility of the correlated metal. This almost saturated state persist upto filling one and is surprising to observe that this dramatic change in the magnetization has almost no noticeable effect on either m^*/m nor the compressibility (Figs. 1 and 2). The orbital degrees of freedom are now playing a crucial role in order to maintain the correlated metallic state. As we fill the system further, the associated impurity model goes into a mixed valence state and the enhanced charge fluctuations have the effect of lowering the magnetic susceptibility. Finally, approaching $\langle n \rangle = 2$ the repulsive interaction renders the electrons almost localized in a Mott state and the polarization grows again due to the large susceptibility of the almost free moments.

Before closing we would like to make a remark on whether the double exchange mechanism alone can account for the large observed values of the dc-resistivity [40]. The presently proposed scenario basically deals with the state of the conduction electrons moving in a static magnetic background provided by the core spins, thus, is quite independent of the double exchange mechanism (which is of course responsible for much of the scattering and the stability of the FM state!). Nevertheless, we note that since the correlated metallic state occurs rather close to a MIT, this may imply that an additional (and possibly important) source of scattering may play a role in the observed dc-resistivity. This scattering would be associated to the orbital degrees of freedom in a mechanism which could be though of as an "orbital Kondo" effect in analogy to the usual Kondo scattering that involves the spin degrees of freedom [41]. This observation follows from the fact that within the dynamical mean field theory, one thinks of each lattice site as immersed in a conduction medium by virtue of the close analogy of the dynamical mean field equations and those of the single impurity problem [7]. In the case of our particular effective model for the ferromagnetic state of the CMR compounds, we showed that the associated local problem, when the states are fully spin polarized, becomes analogous to a single impurity Anderson model, where the role of the usual spin is being precisely played by the orbital degree of freedom.

To conclude we have introduced a model that contains realistic features of the perovskite manganese oxides, namely, band degeneracy and strong electronic correlations. We demonstrated that for parameters which are appropriate for the CMR compounds the system remains in a correlated metallic state and rather close to a MIT as it goes into the saturated FM phase at low temperatures. The low energy physics can then be identified with that of a single band Hubbard model in the proximity of a MIT. We argued that, in analogy to other perovskite TMO, this may allow for the qualitative interpretation of a variety experiments which suggest the existence of a small energy scale. We identify this energy scale with the renormalized Fermi energy of the coherent quasiparticle peak that characterizes the proximity to the MIT in the dynamical mean field theory of the Hubbard model.

Finally, is interesting to observe that in our proposed scenario the orbital degrees of freedom are playing a crucial role. This seems to be emerging as a generic feature of correlated electron systems which contain quasi-degenerate bands as was recently demonstrated in the neutron scattering experiments on the classical transition metal oxide V_2O_3 [42] and also in $YTiO_3$ [43]. The application of similar techniques on the CMR compounds may serve as an experimental test for the validity of the scenario proposed in this work.

Useful discussions with M. Altarelli and P. G. Radaelli are gratefully acknowledged.

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