The anomalous metallic ferromagnetic state of Sr doped manganites.

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We deduce a model relevant for the anomalous metallic state of Sr doped manganites at low temperatures within the ferromagnetic phase. It provides a natural explanation to several anomalous features observed experimentally, such as the vanishing Drude contribution in optical conductivity, the pseudo-gap in the density of states, and the unusual dispersion observed in photoemission.

The fascinating physics of the perovskite manganite systems is continuously attracting the interest of experimentalists and theorists alike. Initially, most of the attraction focused on the colossal magnetoresistance (CMR) effect, due to its potential technological applications. Nevertheless, these systems brought us other remarkable and perhaps more puzzling surprises as, for instance, the strange nature of the metallic state at low temperatures. Optical conductivity experiments in Sr doped manganites La\(_x\)Sr\(_{1-x}\)MnO\(_3\) by Tokura et al. \(^1\), have demonstrated the presence of an extremely small Drude part with a strong mid-infrared contribution dominating the low frequency response. The dc-resistivity, on the other hand, shows conventional behaviour, but its coefficient \(A\) has a strong doping dependence, a feature typical of strongly correlated systems \(^2\). In contrast, however, the specific heat experiments revealed a rather small effective mass enhancement \(^3\). Recent X-ray photoemission studies on n-layer Sr doped systems (n=2 and \(\infty\)) by Dessau et al. \(^4\) showed additional puzzling aspects. For instance, in the n=\(\infty\) compound at \(x = 0.4\) there is a strong suppression of spectral weight near the Fermi edge, which correlates with the appearance of a pseudogap in the n=2 bi-layer system. Remarkably, this bi-layer system still shows a strong CMR effect with a rather good metallic ferromagnetic (FM) state at low \(T\).

Providing theoretical insight to understand this puzzling combination is a challenging problem of strong current interest and the goal of the present work. One possible approach would be to assume that the anomalous behaviour is due to the combined interaction of the many degrees of freedom which are \textit{a priori} present in the perovskite manganites, such as, spin, orbital and local lattice distortion \(^5\). Here we take a different stance and by some reasonable simplifying assumptions, we are lead to a minimal model Hamiltonian that can be reliably solved. It will provide valuable physical insight into the nature of the FM metallic state.

A rather general Hamiltonian for the low energy physics of the CMR manganite systems is, \(^6\)

\[
H = \sum_{<ij>\alpha\sigma} t_{ij}^{\alpha\sigma} c_{i\alpha\sigma}^\dagger c_{j\alpha\sigma} + h.c. - J_H \sum_i \vec{S}_i \cdot \vec{s}_i + H_{JT} + \frac{U}{2} \sum_{i\alpha\sigma\sigma'} n_{i\alpha\sigma} n_{i\alpha\sigma'} (1 - \delta_{\alpha\sigma,\alpha'\sigma'}) \tag{1}
\]

where \(a, b = 1, 2\) are the orbital indexes of the \(e_g\) bands, \(\sigma\) is the spin, \(t_{ij}^{\alpha\sigma}\) correspond to the amplitudes for nearest neighbours hopping, and \(\vec{S}\) denotes a local spin \(3/2\) due to the three electron in the \(t_{2g}\) orbitals The local spin density \(\vec{s}\) of the \((1-x)\) conduction electrons in the \(e_g\) bands is coupled to the latter through the Hund’s rule. The first two terms define a ferromagnetic Kondo lattice model and give a realization of the double exchange mechanism (DEM) \(^7\), the third includes Jahn-Teller polaronic effects \(^1\). These terms are widely considered to be responsible for much of the physics associated to the CMR effect that occurs at intermediate \(T\) of the order of the ferromagnetic transition \(T_c\). However, it was recently emphasized that the model also predicts phase separation \(^8\) in various regions of parameter space, which may play an important role for the complete understanding of the CMR. The last term accounts for local correlations due to a Hubbard-like interaction. While this interaction is recognized as an essential ingredient in every other transition metal oxide, it has only received very limited attention in manganites so far \(^9,10\). As we shall see, it will play a most important role in the anomalous FM phase at low \(T\).

The exact solution of the full model \(^1\) is clearly not possible, so we shall make a few simplifying assumptions which are well justified in the FM phase at low enough temperatures. In passing we recall that the stability of the FM state is due to the DEM, that favors the gain in kinetic energy when the system is doped with \(x\) holes per site \(^11,18\). In a first approximation, we can neglect the Jahn-Teller polaronic term \(H_{JT}\) since the dynamical local distortions of the lattice, which have a notable effect in insulating phases and close to \(T_c\), are found to disappear when one enters the metallic FM state \(^11,18\). On the other hand, we can also safely assume the local spins \(3/2\) to be uniform and static and the magnetization of the \((1-x)\) conduction electrons to be perfectly aligned to them. This assumption is justified by the experimental observation that at low enough temperatures (below \(\approx 200K\) at \(x \approx 0.2\)) the local magnetization is found to saturate at the classical value \(^2\). In conse-
quence, in Fig. 1 we can further drop all terms containing operators labeled with, say, the down spin index \( \downarrow \). At this point, we may take the limit of large lattice connectivity (or \( d \to \infty \)) and use the dynamical mean field theory (DMFT) \([21,7]\) as a controlled approximation to solve our model. The lattice Hamiltonian is mapped onto an impurity problem with a self-consistency condition, and the resulting local action reads,

\[
S_{\text{loc}} = \sum_{nab} d_n^\dagger (i\omega_n) [G_0^{-1}(i\omega_n)]_{ab} d_n (-i\omega_n) + \beta \frac{U}{2} \sum_{\substack{n \neq b}} n_a(i\omega_n) n_b(-i\omega_n)
\]

where \([G_0^{-1}(i\omega_n)]_{ab}\) obeys the self-consistency condition \([G_0^{-1}(i\omega_n)]_{ab} = (i\omega_n + \mu) \delta_{ab} + \Sigma_{\gamma'} t^\gamma_{\gamma'} [G(i\omega_n)]_{\gamma'\gamma'} t^{\gamma'}_{\gamma'ab}\) with \( G \) the local Green’s function \([22]\). For realistic \( \epsilon_g \)-bands the off diagonal hopping elements of the matrix \( t^\gamma_{\gamma'} \) are of the same order of magnitude as the diagonal ones, thus, for simplicity, we may set \( t^\gamma_{\gamma'} = -t \). With a rotation of the impurity operators \( \{d_1, d_2\} \to \{\gamma = (d_1 + d_2)/\sqrt{2}, \gamma' = (d_1 - d_2)/\sqrt{2}\} \), the matrix \( G_0^{-1}(i\omega_n) \) becomes diagonal and the self-consistency condition now reads,

\[
G_0^{-1}(i\omega_n) = \begin{bmatrix} i\omega_n + \mu - (2t)^2 G(i\omega_n) & 0 \\ 0 & i\omega_n + \mu \end{bmatrix}
\]

This equation tells us that in the new basis the \( \gamma \)-electrons are mobile with hopping amplitude \( 2t \), while the \( \gamma' \)-electron is localized. As the interaction term in \( [4] \) is rotationally invariant, the self-consistent system of equations \( [3] \) and \( [4] \) becomes formally identical to that of a Falicov-Kimball model (FKM) \([23,24]\). In this analogy, the orbital degrees of freedom labeled by \( \gamma \) and \( \gamma' \) correspond to the light and heavy spin-less electrons of the FKM.

Before discussing the physics of the model in the present context, we would like to show that after the various manipulations and simplifying assumptions made we have not lost touch with reality, and on the contrary, we did gain precious insight. To this end, we use density functional theory and obtain the realistic bandstructure of ferromagnetic LaSrMnO\(_3\) within the local density approximation. The bandstructure for the majority spin electrons is plotted in Fig. 2 along one of the main directions of the cubic structure. The results shown correspond to the two Mn bands with \( e_g \) symmetry averaged over the Brillouin zone. It can be clearly observed in the figure that the \( d_{z^2} \) band is dispersive and crosses the Fermi energy, while the \( d_{x^2-y^2} \) band near the Fermi energy is essentially flat. The results of this realistic calculation give strong support to the simplified model that we derived before. We have therefore gained valuable physical insight as it will turn out that the anomalous metallic behavior is due to the strong scattering of fast electrons moving in the \( d_{z^2} \) channel colliding with the very slow electrons in the \( d_{x^2-y^2} \) channel. Indeed, this is the basic mechanism that leads to non-Fermi liquid behavior in the DMFT solution of the FKM \([23,26]\). This effect occurs near half filling (in the so called “pinning” region) and the incoherent metallic behavior can be thought of arising from the superposition of the upper and lower Hubbard bands that are split by the interaction strength \( U \).

![FIG. 1. LDA bands for majority spin Mn orbitals of FM La\(_{1-x}\)A\(_x\)MnO\(_3\) with x=0.175. The results are for bands with \( d_{z^2} \) (a) and \( d_{x^2-y^2} \) (b) character along the \( K_x \) direction. The intensity corresponds to the average over \( K_x \) and \( K_y \) in the Brillouin zone. Doping is similited in the Virtual Crystal Approximation.](image)

We shall now show that many anomalous properties of the FM metallic state observed in \( Sr \)-doped manganites are qualitatively captured by our model, using realistic values for the parameters. For the particular case of the FKM it is well known \([25,26,18]\) that the qualitative behavior of the spectral functions does not depend on the choice of the non-interacting density of states (DOS). Therefore, we shall use a Lorentzian DOS \([25,24]\) that allow for fast and simple calculations. In our units \( 2t=1eV \), that gives a bandwidth \( W(\text{FWHM})=2eV \), as suggested by bandstructure calculations. The value of the local Coulomb repulsion is set to \( U=3eV \), which is consistent with the experimental estimates of either the charge transfer or the Mott gap in Ref. \([27]\).

In Fig. 3 we show the results for the optical conductivity \([28]\) at various temperatures and small doping \([1,4]\). We clearly observe the lack of a Drude part which is due to the non-Fermi liquid character of the metallic state. This Drude contribution is absent even at \( T = 0 \) and we can understand this result as due to the presence of localized electrons that break the translational invariance. More physically, the incoherent metallic state is due to the strong scattering of itinerant electrons at the posi-
tions occupied by the localized ones. We thus find that the optical spectra is dominated by the mid-infrared contribution which has a $T$ dependence very similar to the experimental results of Okimoto et al. [1]. Note that the lower values of $T$ were chosen to be of the same order of magnitude as in the experiments.

\[ \sigma(\omega) \]

**FIG. 2.** Optical conductivity $\sigma(\omega)$ calculated for various temperatures $T/W=0.005, 0.015, 0.05, 0.15$ (solid, dashed, long dashed, dot-dashed) and total number of particles $n=0.93$.

In Fig. 2 we plot the results for the optical response as we doped the system away from half filling. We observe that the Drude part contribution rapidly emerges as the model recovers its translational invariance and the metallic state looses its non-Fermi liquid character. As we show in the inset of the figure, the reason for this behaviour is that when we reduce the chemical potential $\mu$ (decreasing the total number of particles) the occupation of the localized $\gamma'$-electrons rapidly decreases and, at the same time, the occupation of the itinerant $\gamma$-electrons increases. Thus we have the simultaneous effects of a reduction of the scattering and an increase in the number of carriers with doping. We would like to point out, that this behavior is in good qualitative agreement with recent experimental results of Tokura et al. [28].

\[ \rho(\omega) \]

**FIG. 4.** The top figure is the density of states $\rho(\omega)$ obtained for $T=0.005$ and $n=0.93$. The bottom figure shows the $\epsilon$-resolved density of states $\rho(\omega)$ calculated for various values of $\epsilon$ across the Brillouin zone. Note that the vertical axis correspond to the frequency $\omega$ and the the curves are plotted along the horizontal axis to better display the dispersion of the quasiparticle feature. The spectra $\rho(\omega)$ were shifted (in the horizontal direction) by their corresponding $\epsilon$ value. The thick dotted line is an aid to the eye to follow the dispersion of the quasiparticle feature as a function of $\epsilon$ across the Brillouin zone. The thin dashed line shows the itinerant $\gamma$-electron band dispersion of the non interacting FK model.

We now turn to the calculation of the density of states (DOS) which allows for the interpretation of the remarkable X-ray photoemission results [3]. The photoemission experiments were performed on $n-$layer systems at $x = 0.4$. The $n=2$ is a bi-layer system and the $n=\infty$ corresponds to the usual 3-dimensional compound. In the first case the samples can be cleaved and angle resolved photoemission results were obtained. They revealed the unexpected feature of a well defined quasiparticle resonance that shows usual band dispersion behavior away from the Fermi surface, but the dispersion flattens out as it approached the (expected location of the) Fermi surface, and remains flat without crossing $\omega=0$. After angle integration, this behavior leads to the presence
of a so called pseudogap at the Fermi energy. In the $n=\infty$ compound, angle resolved experiments are not possible and the angle integrated photoemission also shows a strong suppression of the spectral weight at $E_F$. In the upper part of Fig.4 we show the model prediction for the DOS that clearly show a suppression of spectral weight near $\omega=0$. This is due to the splitting of the upper and lower Hubbard bands by an energy of order $U$. To gain further insight on the nature of the Hubbard bands and to make comparison of the predictions of the model to the angle resolved data, we compute the analog to a momentum resolved DOS within DMFT. In this theory [21] the Green’s function implicitly depends on momenta through the single particle energy $\epsilon \equiv \epsilon_\mathbf{k}$ as, $G(\epsilon, \omega) = 1/(\omega - \epsilon - \Sigma(\omega))$ where $\Sigma(\omega)$ is the local self-energy. Therefore, the connection to the usual $\vec{k}$-resolved DOS is made by noting that momenta close to the zone center ($\Gamma$-point) correspond to values of $\epsilon \sim -W$; momenta near $E_F$, to values of $\epsilon \sim 0$; and momenta close to the zone boundary, to $\epsilon \sim W$. In the lower part of Fig.4 we plot the results for the imaginary part of $G(\epsilon, \omega)$ for various values of $\epsilon$ across the Brillouin zone. These results provide insights on the nature of the dispersion of the interacting model and can be qualitatively compared to the angle resolved data of Ref. [3]. Using the same realistic values for the model parameters, we clearly see in the figure a distinct quasiparticle feature that disperses across the lower Hubbard band in the occupied part of the spectra ($\omega < 0$), and another one that has similar behaviour in the upper Hubbard band for the unoccupied part ($\omega > 0$). The first one, which is to be contrasted to photoemission experiments, shows a rather usual dispersive pattern for $\epsilon \sim -W$ (near the zone center), but it then flattens out as $\epsilon \to 0$, and remains flat up to higher values of $\epsilon$ (near the zone boundary). It is important to note that as the quasiparticle peak fails to cross the Fermi energy, it also becomes broader and has a rapid loss of weight, indicating its progressive incoherent nature as it approaches the low frequencies $\omega \sim 0$. This behaviour is in remarkable qualitative agreement with the angle resolved data in the bi-layer system [4].

To conclude, we have derived and studied a model relevant for the anomalous FM metallic state of Sr doped manganites at low $T$. Our results provide a consistent description of several unusual observations in the experimental spectroscopy. The analogy of our model with an orbital Falicov-Kimball model, also explains the experimental finding that at upon doping to high values of $x$, the incoherent metal crosses-over to a regular Fermi liquid. Even more interestingly, at low values of doping, the analogy with the FKM suggest also an explanation for the driving force in the recent observation by Endoh et al. [31] of charge-ordering in the FM insulating phase as due to the well known charge density wave instability [24,29] of the model in bipartite lattices near half filling.

We stress that our present work remains at a qualitative level, and a detailed calculation of the phase boundaries [30] and the eventual modifications that one may have to introduce to make more precise comparison with experiments are left for future work.

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