Double exchange magnets: Spin-dynamics in the paramagnetic phase

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The electronic structure of perovskite manganese oxides is investigated in terms of a Kondo lattice model with ferromagnetic Hund coupling and antiferromagnetic exchange between $t_{2g}$-spins using a finite temperature diagonalization technique. Results for the dynamic structure factor are consistent with recent neutron scattering experiments for the bilayer manganite La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$. The susceptibility shows Curie-Weiss behaviour and is used to derive a phase diagram. In the paramagnetic phase carriers are characterized as ferromagnetic polarons in an antiferromagnetic spin liquid.

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Double exchange (DE) as an explanation of ferromagnetic order in doped manganites was proposed shortly after the first experimental work on these compounds. Open questions are connected with the proper understanding of the paramagnetic (PM) phase above the Curie temperature $T_C$ and the giant magnetoresistance at the phase transition. Millis \textit{et al} argued that DE is neither sufficient to explain the size of the resistivity above $T_C$ nor the activated form of $\rho(T)$ observed in many compounds, and suggested that Jahn-Teller polaron formation is essential. A different point of view is that these systems are governed by strong correlations while spin disorder scattering is crucial for the understanding of the resistivity. Further complications not considered here may arise from orbital degeneracy and charge disorder.

A detailed investigation of the spin-dynamics is therefore important for a deeper understanding of the manganites. Inelastic neutron scattering experiments in the PM-phase were performed recently by Perring \textit{et al} for the highly doped bilayer manganite system La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$. Particularly surprising was the observation of growing antiferromagnetic (AF) scattering at low energy parallel to increasing ferromagnetic (FM) critical scattering as the FM $T_C$ was approached from above. Our aim here is to shed light on these experiments by an investigation of the spin-dynamics within a standard model for these compounds.

The appearance of ferromagnetism was explained by Zener in terms of a strong Hund coupling $J_H$ of the $e_g$ conduction electrons to $S=3/2$ core spins $S_i^e$ formed by the energetically well separated $t_{2g}$-electrons. The $e_g$ electron in Mn$^{3+}$ couples to a $S=2$ high-spin configuration. A relevant model is the \textit{ferromagnetic} Kondo lattice model

$$H_{KLM} = -\sum_{\langle ij \rangle} t_{ij} d_{i\sigma} \dagger d_{j\sigma} - J_H \sum_{i>\sigma} S_i^e \cdot d_{i\sigma} \dagger \sigma d_{i\sigma} + J_{AF} \sum_{<ij>} S_i^e \cdot S_j^e + U \sum_i n_{i\uparrow} n_{i\downarrow},$$

The operators $d_{i\sigma}$ and $S_i^e$ refer to $e_g$-electrons and $t_{2g}$-spins, respectively, while $n_{i\sigma} = d_{i\sigma}^\dagger d_{i\sigma}$ denotes the $e_g$-density operator. A typical value for the Hund coupling is $J_H \sim 1$eV, while the nearest neighbor hopping $t_{ij} = t \sim 0.15eV$. In view of the large local repulsion $U \sim 8$ eV, we shall furtheron exclude double occupancy of $e_g$-orbitals, and define $J_{H}/J = 6$. The model explains ferromagnetism via the optimization of the kinetic energy of the $N_e = \sum n_{i\sigma}$ conduction electrons in the partially filled $e_g$ band when the spins are aligned at low temperature. At low doping manganite materials are usually AF insulators. Antiferromagnetic superexchange interaction $J_{AF}$ between the $t_{2g}$-electrons or spins is an obvious source of such correlations.

The interplay of the global AF exchange interaction with the local FM alignment induced by the motion of $e_g$-electrons via the DE mechanism is a subtle problem. A simple physical picture, which is consistent with our numerical study, is that of \textit{ferromagnetic} spin polarons, where a local FM surrounding of $t_{2g}$-spins moves with the holes through an otherwise AF background.

Our calculations are based on a finite temperature diagonalization approach developed by Jaklič and Prelošek, which was successfully used for the study of the $t$-$J$ model. Here due to the much larger Hilbert space only relatively small systems can be solved. Nevertheless due to the high density of low energy excitations we believe that our results reflect already the physics of large systems. We have found large similarity between 1D and 2D systems, in striking contrast to the physics of the $t$-$J$ model. We shall present here mainly 1D results, since the resolution in momentum space is larger. Our energy and temperature units are $t = 1$, while $S = 1$ is used instead of $S = 3/2$ $t_{2g}$-spins to save computer time.

A global measure for the evolution of magnetic correlations is given by the square of the total spin $S = \sum S_i$, which includes both core- and $e_g$-spins. Results for different $e_g$-concentration and $J_{AF}$ are shown in Fig. 1(a). The high-$T$ limit is determined by independent spins (i.e. for $T < J_H$; for even higher $T$ also the $e_g$-spins decouple). In the absence of $J_{AF}$ FM correlations develop as the temperature is lowered. For quite small $J_{AF}$ this be-
behaviour is already changed. For $J_{AF} = 0.1$ ($S^2$) decreases with decreasing temperature for a single hole. This suggests the evolution of AF correlations, while at higher doping FM correlations are established via the DE mechanism. Further insight is gained from the uniform susceptibility $\chi = \langle S^2 \rangle / N k_B T$. The corresponding $1/\chi(T)$-data, Fig. 1(b), show Curie-Weiss behavior, similar to experiments [14,15]. It is quite remarkable that for these small systems with $N = 6$ and 8 sites $1/\chi(T)$ follows a linear Curie-Weiss law to quite low temperatures, which allows us to determine a mean-field transition temperature $T_{C_{MF}}$. Deviations at lower temperatures are expected from non mean-field behavior of low-dimensional systems and finite size effects.

![Graph](image)

FIG. 1. Square of total spin (a) and inverse susceptibility (b) vs. temperature for hole concentrations $x = (1 - N_e/N) = 1/6, 3/6$ (1D) and 1/8 (2D). A comparison with the FM polaron approach is shown for a single hole in 1D (triangles) and 2D (squares) for $J_{AF} = 0$.

The mean-field phase diagram (Fig. 2) reflects the interplay between antiferromagnetism and carrier induced ferromagnetism at higher doping. $T_{C_{MF}}$ scales with coordination number and is consistent with the experimental $T_C$-scale. The value chosen for $J_{AF} (\sim 0.1)$ is typical for real systems [8]. For $J_{AF} = 0$ our data is well described by $T_{C_{MF}} \sim x(1 - x)$ [8], while it differs in that respect from high-temperature series expansion results [17].

A simple estimate of $\langle S^2 \rangle$ in terms of a ferromagnetic spin-polaron picture has been given by Varma [6] for the model without AF-interactions. The spin polaron is assumed to be determined by a number $P$ of ferromagnetically aligned spins around the hole which allows the particle to improve its kinetic energy. The gain in kinetic energy is counterbalanced by the loss of spin entropy due to the coupling of $P$ spins, which can no longer rotate independently. An estimate for the change of free energy is $\delta E_P = at/P^p + P k_B T \ln(2S_2 + 1)$. The kinetic energy exponent $\eta = 2/3, 1$ in 3, 2, and 1-dimensions, respectively. Minimization leads to $P(T) = (\eta t/t_0)^{1/(1+\eta)}$ with $B = \ln(2S_2 + 1)$. Under the same assumptions the square of the total spin is given in terms of $S_P = (S_1 + PS_2)$

$$\langle S^2 \rangle = N \left[ xS_P(S_P + 1) + (1 - x - Px)S_2(S_2 + 1) \right],$$

where in the case of few $e_g$-holes $x$ is the hole concentration. Here $S_1 = 1$ and $S_2 = 3/2$ (instead of 3/2 and 2).

The effective spin $S_{eff}$ for a single polaron may be defined as $S_{eff}(S_{eff} + 1) = \langle S^2 \rangle_T - \langle S^2 \rangle_\infty$. Near $T_{C_{MF}}$ we obtain $S_{eff} \sim 7$ for the two-dimensional case. This corresponds to an enhancement factor $\sim 14$ with respect to the spin $S = 1/2$ of the $e_g$ conduction electron (hole). This result is qualitatively consistent with EPR experiments [14] where large effective spins at temperatures slightly above $T_C$ were observed.

![Graph](image)

FIG. 2. Mean-field phase diagram of a 1D chain for $J_{AF} = 0.1$ (triangles) vs. $(1 - x) = N_e/N$. The boundaries between AF, FM and PM phases (solid lines) are guides to the eye. (Anti)periodic boundary conditions were used for odd (even) electron number [18]. The $J_{AF} = 0$ data (squares) is compared with the predicted behavior $T_{C_{MF}} \sim x(1 - x)$.

The study of correlation functions (CF) gives further support to the FM polaron picture even at high doping concentration and with frustrating AF interactions, although the simple formulation above is certainly not sufficient for these cases. The temperature dependence of the $\langle S_i \cdot S_j \rangle$ correlation functions for $J_{AF} = 0.1$ is shown in Fig. 3. We distinguish (a) a low- and (b) a
high-doping case where the system approaches the AF (FM) phase (Fig. 2) at low temperature, respectively. In the single hole case (a) the nearest-neighbor (n.n.) CF becomes AF as T is lowered, while further neighbor correlations remain small. The CF’s for \( x = 0.5 \) (b) become all FM at low T. Yet it is interesting to notice that for intermediate T the n.n. CF is antiferromagnetic. For \( T \gg t \) all CF’s vanish.

To investigate the structure of the spin-polaron we calculate the relative correlation function (RCF)

\[
C^{n+1}_n = \sum_i \langle n_i^n S_{i+n} \cdot S_{i+n+1} \rangle,
\]

which measures the spin-CF \( \langle S_i \cdot S_j \rangle \) in a distance \( n \) from the position of the moving hole. Here \( n_i^n = (1 - n_i) \) and \( l \) is a lattice vector. The results in Fig. 3 show the FM alignement of the spins at the position of the hole and on the neighboring site, i.e. \( C^1_0 \), at low T even in the single hole case where \( \langle S_0 \cdot S_1 \rangle \) becomes antiferromagnetic. The large drop at very small T suggests that FM-polarons are unstable in the low temperature AF-phase consistent with Fig. 1(a). The CF’s \( C^1_0 \) are weakly antiferromagnetic at higher temperatures similar to \( \langle S_i \cdot S_j \rangle \).

In the single hole case (a) all other \( (n \neq 0) \) RCF’s become gradually AF with decreasing temperature, i.e. indicating that the AF-phase dominates. The CF’s for \( x = 0.5 \) (b) are indicative for frustrated antiferromagnetism at temperatures above \( T \sim 0.5 \), while at lower temperature all CF’s become ferromagnetic. Hence on the basis of these equal-time CF’s there is no evidence for significant AF correlations at low T near \( T^{MF}_C \) for high doping. Nevertheless there are clear AF-correlations in the low-frequency response.

The complete information about the spin-dynamics is contained in the dynamic structure factor

\[
S_q(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \exp(-i\omega t) \langle S_q S_{-q}(t) \rangle,
\]

which can be directly measured by inelastic neutron scattering. A central result of this work is the evolution of \( S_q(\omega) \) with temperature (Fig. 4). As an example results for a 6-site chain with 3 \( e_g \)-holes are shown for \( J_{AF} = 0 \) (a) and 0.1 (b) at different momenta. At high T critical scattering extends over the whole Brillouin zone, while at low temperature it is confined to \( q \sim 0 \) in the absence of AF-interactions. In case (a) FM magnons emerge when lowering the temperature already well above \( T^{MF}_C \sim 0.1 \). Their energy scale is consistent with theoretical work on the magnon dispersion [4] and neutron scattering in the FM-phase [21] of La\(_{0.7}\)Pr\(_{0.3}\)MnO\(_3\). Antiferromagnetic frustration appears to be small in this system, since already LaMnO\(_3\) consists of FM planes with a well developed magnon spectrum even at \( T_C \) [21]. The interlayer coupling is AF and weak in this system.
$q = \pi$ are growing as well. We note that our data is broadened by $\delta = 0.05$. The natural linewidth of the $\omega \sim 0$ structures appears to be smaller than $T$.

The low frequency response $\delta S(q) = \int_{-\infty}^{\omega_c} d\omega S_q(\omega)$ integrated over a small energy window ($\omega_c = 0.05$), is frequently measured in neutron scattering experiments [8]. It is interesting to contrast the momentum dependence of $\delta S(q)$ from the behavior of the static structure factor $S(q) = \int_{-\infty}^{\infty} d\omega S_q(\omega)$, which measures equal-time correlations (Fig. 5). In the high-temperature limit $S(q)$ is constant, reflecting the absence of spin-correlations. At low temperature $S(q)$ develops a peak centered at $q = 0$ in the model without antiferromagnetic interactions due to carrier induced ferromagnetic correlations. With increasing $J_{AF}$ the data shows the evolution of short-range (nearest-neighbor) AF correlations.

![FIG. 5. Comparison of $S(q)$ and integrated low-frequency response $\delta S(q)$; parameters as in Fig. 4.](image)

The observation of pronounced AF-correlations in $\delta S(q)$, e.g. for $J_{AF} = 0.1$, in the presence of FM critical scattering is consistent with the experimental finding of local AF correlations by Perring et al [8]. These experiments show strong AF correlations in the PM-phase (at low frequency) and their sudden disappearance at $T_C$ when the low-$T$ FM-order is established. We stress that the observation of a well defined AF peak in the low-frequency spin-fluctuation spectrum $S_q(\omega \to 0)$ is actually very surprising since these experiments were performed for a highly doped ($x = 0.4$) double-layer system and at temperatures slightly above the transition into the FM-phase. We find considerable qualitative agreement between the present model and neutron scattering, although we do not find a significant peak structure in the momentum dependence at $q = \pi$, except for larger values for $J_{AF}$ where the FM scattering is reduced. The disappearance of the AF signal in the low-temperature FM phase [8] is consistent with our studies [22].

In summary we have shown, that the competition between AF and FM correlations detected by neutron scattering in the highly doped double layer compound La$_{1.2}$Sr$_{1.8}$MnO$_7$ is consistent with the physics of the frustrated Kondo lattice model. From our study of various correlation functions, we conclude that the temperature and doping dependence of the magnetic correlations in the paramagnetic phase can be qualitatively understood in terms of FM polarons moving in a short-range AF-slip liquid. Slow spin-dynamics as a consequence of frustration may lead to localization of magnetic polarons.

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