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LETTER TO THE EDITOR

Magnetic properties of the double-exchange model

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Abstract. We study the *ferromagnetic* (FM) Kondo lattice model in the strong-coupling limit (the double-exchange (DE) model). The DE mechanism proposed by Zener to explain ferromagnetism is found to have unexpected properties when there is more than one itinerant electron. We find that, in general, the many-body ground state of the DE model is *not* globally FM ordered (except for in special filled-shell cases). Furthermore, the low-energy excitations of this model are distinct from those in usual Heisenberg ferromagnets, which will result in unusual dynamic magnetic properties.

The double-exchange (DE) model [1–3] has attracted much recent attention [4–12] because of its anticipated relevance to the Mn oxide perovskite $La_{1-x}A_xMnO_3$ (A = Ca, Sr, Ba, Pb) materials exhibiting colossal magnetoresistance (CMR) [13–17]. To explain the ferromagnetism in these Mn oxides, Zener [1] introduced a DE mechanism, in which local $S = 3/2$ spins of the three Mn t_{2g} d electrons become ferromagnetically coupled due to coherent hopping of e_g electrons. Due to the coupling between the spin, lattice and orbital degrees of freedom, the CMR materials have unusual dynamical magnetic properties compared to the ferromagnetic (FM) Heisenberg model [18, 19]. Very recently, a surprising experimental observation via inelastic neutron scattering showed that the spin-wave dispersion in $La_{0.7}Pb_{0.3}MnO_3$ can be fitted throughout the Brillouin zone [20] by a FM Heisenberg model with nearest-neighbour exchange coupling. For a theoretical understanding of these unusual magnetic properties in the real materials, it is important to first understand the intrinsic magnetic properties of the DE model. Here, we show that these properties are not those of a simple FM.

The DE mechanism can be derived from the following (FM) Kondo lattice model Hamiltonian [2, 4]:

$$H = -t \sum_{\langle i,j\rangle\alpha} c^{\dagger}_{i\alpha} c_{j\alpha} - J_H \sum_{i\alpha\beta} \boldsymbol{S}_i \cdot \boldsymbol{\sigma}_{\alpha\beta} c^{\dagger}_{i\alpha} c_{i\beta} \tag{1}$$

where $\boldsymbol{\sigma}$ is the Pauli matrix and $\boldsymbol{S}_i$ is the local spin $S = 3/2$ of three Mn t_{2g} d electrons. The operators $c_{i\alpha}$ ($c^{\dagger}_{i\alpha}$) annihilate (create) a mobile e_g electron with spin α at site i. The DE mechanism requires the Hund's rule coupling $J_H \gg t/S$.

The DE model was studied by Anderson and Hasegawa for a single electron on two sites [2]. The model (1) was also studied in a spin-wave approximation [4, 21], mean-field approximations [4, 10, 11], the dimension $D = \infty$ and $S = \infty$ limit [6], and a coherent-state path integral approach in a semiclassical approximation [7]. In these approximations, the magnetic properties of the DE model are described by an effective-FM Heisenberg model. However, the DE model describes a partially *itinerant* magnetic system, which may behave

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differently. In this letter, we present finite-size exact-diagonalization and analytical results for the magnetic properties of the DE model. In 1D, we show that the ground state (GS) of the model (for periodic boundary conditions (PBC) and $t > 0$) is a spin singlet ($S^t = 0$) for even numbers of electrons, and maximally polarized ($S^t = S^t_{max} \equiv NS + n/2$) for odd numbers of electrons, where N is the number of sites, and n is the number of electrons. This property was also discovered numerically for short spin-1/2 chains by Kubo previously [22]. For even n, there are states in each spin sector $S < S^t_{max}$ which are lower in energy than that of the FM GS [23]. In 2D and 3D, we find numerically on up to 8^2 and 4^3 lattice sizes that the GS is not maximally polarized for general fillings. The low-energy excitations of the model are also different from those of the Heisenberg FM for fillings with *both* a FM GS and a non-FM GS, yielding distinctive spin susceptibilities.

Let us first discuss the GS properties of the model (1). The Hamiltonian (1) conserves total momentum $\boldsymbol{K}$, total spin S^t, and S^t_z. For the FM states [24], $S^t = S^t_{max}$. It can be easily seen that the wavefunctions of FM eigenstates are simply the Slater determinants of spinless fermions $|\boldsymbol{k}_1, \boldsymbol{k}_2, \ldots, \boldsymbol{k}_n\rangle \otimes |\mathrm{FM}\rangle$, with energy $\sum_i (-J_H S + \epsilon_{\boldsymbol{k}_i})$, where $\epsilon_{\boldsymbol{k}}$ is the non-interacting electron dispersion. $|\mathrm{FM}\rangle$ denotes the FM state for local spins. We will denote the lowest FM eigenenergy by E_0. For our numerical calculations, we set $t = 1$, $J_H = 40$ for the GS and $J_H = \infty$ for spin excitations and susceptibility. In the manganese perovskites $S = 3/2$; however, we will take $S = 1/2$ for most of our numerical calculations. Sample calculations were repeated for smaller systems at $S = 3/2$, with qualitatively similar results.

For one electron ($n = 1$), it can be shown analytically that in the ground state $S^t = S^t_{max}$ for $J_H > 0$, which is consistent with the DE mechanism. At half-filling $n = N$, however, there is an induced AFM coupling $\sim t^2/J_H$ between spins due to the Pauli principle; thus the GS at half-filling is AFM [25]. What happens for general fillings? In 1D it is straightforward to show that non-FM ground states exist for periodic systems with n even in the limit where the local spins are classical ($S \to \infty$). Consider a 1D ring where the local spins uniformly circle the north pole at an angle θ. The magnitude of the effective hopping t_{eff} is reduced as θ increases, but an additional magnetic flux (a Berry phase) is simultaneously added. It is easy to verify that for large J_H the change in energy is $\Delta E = -c_1 t^2 \sin^2(\theta)/J_H$, where c_1 is a positive constant for an even number of electrons. For finite J_H, the Berry phase reduces the total energy more than t_{eff} increases it, and the spins prefer to lie on the equator. For finite S, one can show [26] that the leading $1/S$ correction is $\Delta E = -c_1 t^2 \sin^2(\theta)/J_H - c_2|t| \sin^2(\theta)/S$, where c_1 and c_2 are positive constants for an even number of electrons. Quantum fluctuations also favour a single twist state with spins lying in the equator.

In 1D, the GS for any even number of electrons $n = 2m$ (m integer) is non-FM for any N and S (for $t > 0$) [27]. This can be proven by constructing the following wavefunction with $S^t = S^t_{max} - 1$ [28]:

$$|\Psi\rangle = S^-_t(q_0)|\mathrm{vac}\rangle - C\, S^-_t(0)\, c^\dagger_{-mq_0\uparrow} c_{(1-m)q_0\uparrow}|\mathrm{vac}\rangle \tag{2}$$

with $|\mathrm{vac}\rangle = c^\dagger_{(1-m)q_0\uparrow} \cdots c^\dagger_{mq_0\uparrow}|0\rangle|\mathrm{FM}\rangle$ and $q_0 = \pm 2\pi/N$. Here we have used $S^-_t(q) = \sum_l \mathrm{e}^{-\mathrm{i}ql}(S^-_l + c^\dagger_{l\downarrow} c_{l\uparrow})$ and $C = 1/(2NS + 2m)$. It can be shown that the variational energy $E_v < E_0$, so there is at least one eigenstate with $S^t = S^t_{max} - 1$ and energy less than E_0. The difference between even and odd numbers of electrons arises as follows: in the FM states, the electrons fill the non-interacting states up to the Fermi level; for an odd number of electrons, the $\pm k_f$-states at the Fermi level are all filled; for an even number of electrons, only one of the $\pm k_f$-states is filled. Due to this degeneracy, the wavefunctions (2) have an

energy lower than E_0. In 1D systems, we calculated the GS and some low-energy excited states using Lanczos methods for sizes up to $N = 16, n = 2$, and $N = 10, n = 6$. We find that the GS always has $S^t = 0$ and $K = \pi$ for even numbers of electrons, and $S^t = S^t_{max}$ and $K = 0$ for odd numbers of electrons. These exact-diagonalization results in 1D are consistent with a previous calculation described in reference [22].

The trial wavefunction in 1D can be extended to general D for *two* electrons. We denote the zone centre $\boldsymbol{k}_0 = 0$ with (lowest Bloch) energy ϵ_0, and the first excited states with energy ϵ_1 by $\boldsymbol{Q}_i$, with $i = 1, \ldots, M$ $(M \geqslant 2)$. Then we can write a wavefunction with $S^t = S^t_{max} - 1$ as follows:

$$|\Psi\rangle = \sum_{i=1}^{M} S_t^-(\boldsymbol{Q}_i)\, c^\dagger_{0\uparrow} c^\dagger_{-\boldsymbol{Q}_i\uparrow} |0\rangle|\mathrm{FM}\rangle. \tag{3}$$

The variational energy is $E_v = E_0 - (\epsilon_1 - \epsilon_0)(M-1)/(2SN + M + 1)$. If the number of sites $N > 2$, the energy difference $E_0 - E_v$ is finite. We speculate that the GS of model (1) is non-FM in higher dimensions whenever the single-particle states at the Fermi level of the FM GS are not completely filled. This has been confirmed in our finite-size exact-diagonalization calculations. From the single-electron dispersion of the non-interacting systems, we see that the filled-shell cases are 1, 7 for the $\sqrt{8} \times \sqrt{8}$ lattice and 1, 5, 9 for the $\sqrt{10} \times \sqrt{10}$ lattice. For these fillings, we find that the GS is FM. The more numerous non-filled-shell GSs have

$$\begin{aligned}
&S^t = 0\,(2), 1/2\,(3), 0\,(4), 1/2\,(5), 0\,(6), 0\,(8) && \text{for } N = 8\\
&S^t = 0\,(2), 5/2\,(3), 4\,(4), 7\,(6) && \text{for } N = 10\\
&S^t = 0\,(2) && \text{for } N = 16.
\end{aligned}$$

(The numbers in parentheses are the numbers of electrons.) In all of these cases the ground states are non-FM. One different feature in a 2D (e.g. $\sqrt{10} \times \sqrt{10}$) lattice is that the ground states are not necessarily at the smallest S^t for non-filled-shell cases. But for all of the cases that we have checked, there is at least one state in each $S^t < S^t_{max}$ sector which has an energy lower than E_0. For larger clusters, e.g. 6^2 $(n = 2, 3, 4)$, 8^2 $(n = 2, 3, 4)$, 3^3 $(n = 2, 3)$ and 4^3 $(n = 2, 3)$, we have confirmed [26] that the GS in the $S^t_z = S_{max} - 1$ sector is lower than that in the $S^t_z = S_{max}$ sector. We also calculated the 1D and 2D $S = 3/2$ cases up to $N = 8$ and $n = 4$. The results are similar to those for $S = 1/2$.

In the above calculations, we have used PBC. Anti-periodic boundary conditions do not change the qualitative results except that the filled-shell and open-shell cases will be changed. For open boundary conditions (OBC), the FM states are more robust: the GS in 1D has $S^t = S_{max}$ [22]. (See reference [29] for a more detailed study of the 1D system using OBC.) We find, however, that the GS can still be non-FM in $D > 1$ finite clusters ($2 \times 3, 3 \times 4$, etc) even with OBC. For the remainder of this report we will use PBC.

What is the magnetic structure of the non-FM GS? At $n = N$, the GS is AFM on a bipartite lattice, as described above. At $n = 2$, the GS also has $S^t = 0$, but the spins have *local* FM ordering, which is evident in the correlation functions (see table 1 for a 4×4 2D lattice). For other fillings with non-FM ground states $S^t < S^t_{max}$, and the magnetic structures are most likely to be locally but not globally FM ordered (see figure 1). For non-OBC 1D systems, the local spin structures are *spiral* with a pitch equal to the length of the chain [22]. This can be seen in the correlation function $\langle(\boldsymbol{S}_1 \times \boldsymbol{S}_2) \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_{i+1})\rangle$, which is only weakly dependent on i and has magnitude $\sim(0.25 \sin(2\pi/N))^2$ [26]. Preliminary numerical results suggest that the 2D and 3D non-FM GS is also non-collinear for non-filled-shell cases. The details of the spin structures for 1D and higher D are not the major concern here and will be

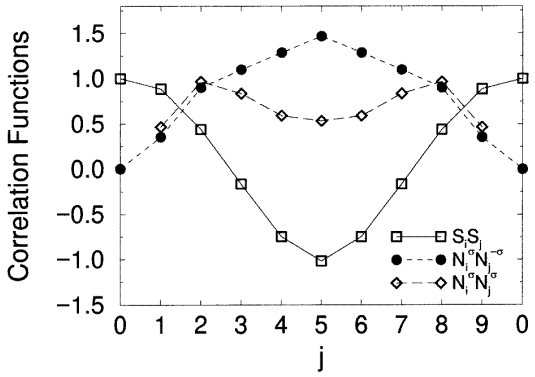

Figure 1. The correlation functions $\langle n_1^\sigma n_{j+1}^{-\sigma}\rangle$, $\langle n_1^\sigma n_{j+1}^{\sigma}\rangle$, $\langle \boldsymbol{S}_1 \cdot \boldsymbol{S}_{j+1}\rangle$ for the 1D $N = 10$, $n = 4$, $S = 1/2$ system. The curves are rescaled to fit on the same graph, so only the relative magnitudes are representative.

Table 1. The GS static correlation functions for 2D, $N = 16$ (4×4), $n = 2$, $S = 1/2$. $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j\rangle$ is the correlation function of the total spin (local + itinerant). The index for site (i_x, i_y) is labelled as $i = i_x + 4(i_y - 1)$.

(i, j)	$\langle n_i^\sigma n_j^{-\sigma}\rangle$	$\langle n_i^\sigma n_j^{\sigma}\rangle$	$\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j\rangle$
(1, 2)	0.003 264	0.001 489	0.1655
(1, 3)	0.005 771	0.002 301	−0.0131
(1, 6)	0.005 771	0.002 301	−0.0131
(1, 7)	0.008 117	0.002 820	−0.2333
(1, 11)	0.010 515	0.003 084	−0.5559

discussed elsewhere. What we can conclude from these correlation functions (see figure 1 and table 1) is that the spin-up electrons are repelled from spin-down electrons, and the local spins with greatest separation are anti-parallel; thus there is local (i.e. FM domain) but not global FM ordering.

Equally importantly, for *both* FM and non-FM GSs, the excitation spectra of model (1) are also unusual. For the open-shell fillings, the low-energy excitation spectrum is complicated [26] and we cannot interpret the low-lying $\Delta S = 1$ states as quasi-spin-wave (SW) states (see figure 2(c)). For the filled-shell cases, there are quasi-SW excitations where the dispersion softens at large K (or high energy) (see figure 2(b)) due to a 'spin-polaron' effect: as K increases, the electrons increasingly avoid the flipped spin, so the effective spin–spin coupling J becomes smaller. In figure 3(a) we show explicitly the SW dispersion softening for different filled-shell cases. One can show using, e.g., a Bethe *ansatz* that the $n = 1$ excitations shown in figure 3(a) cannot have an energy greater than $2t[1-\cos(\pi/N)]$. In figures 2 and 3 the effective spin–spin coupling J is extracted from the low-energy SW excitations [26]; the dispersion of the SW excitations is rescaled to coincide at $K = 0, 2\pi/N$. In figure 3(b), we also show the spectral weight $C|\langle\Psi_K|S_t^-(K)|\Psi_0\rangle|^2$, where $|\Psi_0\rangle$ is the FM GS wavefunction and $|\Psi_K\rangle$ is the SW wavefunction with momentum K. This spectral weight should be proportional to the neutron scattering cross-section at the SW frequency. From figure 3(b) we can see that the spectral weight decreases at large K due to the spin-polaronic effect noted above. The multi-magnon states in the

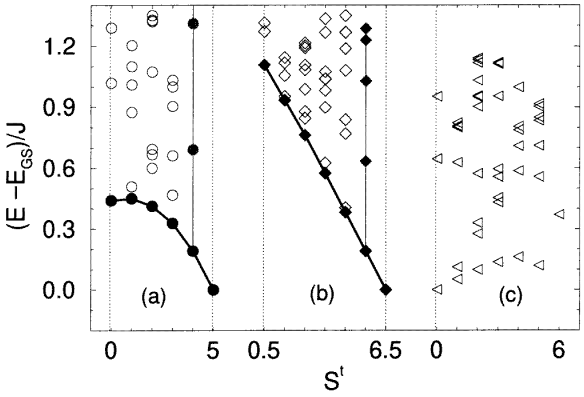

Figure 2. Low-energy excitation spectra for 1D $N = 10$ systems. (a) The Heisenberg system; (b) the filled shell, $n = 3$; (c) (rescaled by $\times 2$) the unfilled shell, $n = 2$. Note that the high-energy (high-momentum) spin-wave states (grey symbols along the vertical line) in (b) show softening as compared to those in (a) (see figure 3). The lowest multi-magnon states (single-magnon and FM states are also included) are the black symbols on the thick lines in (a) and (b).

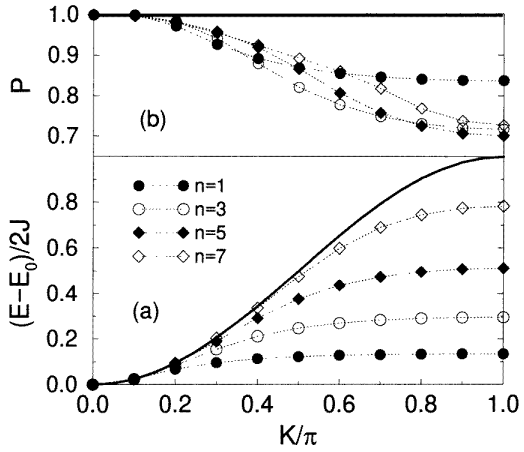

Figure 3. The dispersion and spectral weight of SW states at $N = 20, n = 1, 3, 5, 7$. The lower four curves are SW dispersions. The solid line is the cosine curve, i.e. SW dispersion in the Heisenberg system. Parameters: $S = 1/2$, $J_H = \infty$. The upper curves are their spectral weights, $P = C|\langle \Psi_K | S_t^-(K) | \Psi_0 \rangle|^2$.

filled-shell DE model also behave differently from those in the FM Heisenberg model (see figure 2(a)). In the Heisenberg model, the lowest curve $E(S^t)$ is not linear, due to interactions between magnons, as shown in figure 2(a). In the DE model, the lowest-energy multi-magnon states have momentum mq_0 and $\Delta E^m_{\pm mq_0} = m\,\Delta E^1_{\pm q_0}$ with high accuracy, where ΔE^m_K is the energy difference between the m-magnon state with momentum K and the ground state. This linear behaviour persists for different system sizes and different filled-shell cases. It is surprising that this linear behaviour is accurate even for m-magnon states with large momentum mq_0, if one notes that there is substantial energy softening in

the large-momentum SW states. However, the 'sum rule' $\Delta E^m_{q_1+\cdots+q_m} = \sum_{i=1}^m \Delta E^1_{q_i}$ for multi-magnon energies is not satisfied, if different-momentum (e.g. $q_i \neq q_j$) SW states or higher-momentum (e.g. $|q_i| \geqslant 2q_0$ for at least one i) SW states are involved. So this linear behaviour in equation (2) is not due to the absence of magnon–magnon interaction in the DE model.

The energy shifts in the SW states and low-energy multi-magnon states discussed in the previous paragraph may affect the low-temperature spin susceptibilities. At high temperatures, the spin excitations are coupled to the charge excitations, and the spin susceptibility will also be changed. We have calculated the spin susceptibility for 1D, $N = 7, 8, 9, 10, n = 2, 3$, and 3D, $N = 2^3$, $n = 1, \ldots, 8$ lattices [26]. We have also calculated the properties of the DE model with a high-temperature series expansion for both the q-coordinated Bethe lattice and the simple cubic lattice [30]. The results in the DE model cannot be rescaled into those of the Heisenberg model.

In 1D we have shown that the GS for even numbers of electrons is a spin singlet, while for odd numbers the GS is maximally polarized. In 2D and 3D, we find numerically that the GS is non-FM for open-shell fillings for finite clusters up to 8^2 and 4^3. From the results in the 1D classical spin limit, the energy differences between the non-FM GS and the lowest FM state scale as $1/N^3$. Because of the filling dependence, it is difficult to extrapolate the GS properties to the thermodynamic limit. Although the non-FM states are not maximally polarized, locally there still exist FM correlations. It is also conceivable that this model may have *global* FM ordering in the thermodynamic limit. However, such a recovery of global FM ordering from the non-collinear spin states is non-trivial. This distinguishes the DE system from the Heisenberg system. We also find that even for the filled-shell cases, the spectrum of low-energy spin excitations is different from that in the Heisenberg FM, which can show up in the finite-temperature spin susceptibility. The softening of large-momentum SW states and the decrease of the spectral weight should be observable via neutron scattering. The anomaly in the low-energy multi-magnon spin excitations may also be observable in non-linear multi-magnon neutron experiments. The recently observed spin-wave dispersion in $La_{0.7}Pb_{0.3}MnO_3$ measured via neutron scattering [20] can be well described by the FM Heisenberg model [20, 21]. However, we note that the model Hamiltonian (1) does not necessarily fully describe the magnetic properties of CMR materials: additional spin–lattice and spin–orbit coupling probably play important roles [7, 9]. The recovery of Heisenberg ferromagnetism may be due to the inter-orbital electron–electron interactions [26]. There is also experimental evidence for slow 'spin' dynamics [18, 19] which cannot be explained by the simple FM Heisenberg model.

In this work, we have shown that the DE model has unusual magnetic properties. The GS of the model is not maximally polarized for open-shell cases. In the DE model (1), there are O(N) $S^t < S^t_{max}$ states with energy lower than the FM GS energy E_0, except for in special filled-shell cases. This was shown in 1D for any system size N and spin S. Our exact-diagonalization calculations also confirmed this for higher-D clusters up to 8^2 and 4^3. The spin correlations show that the non-FM low-energy states have local FM-domain-like structures (spiral ordering in 1D). Our calculation also shows that even for filled-shell cases the spectra of low-energy multi-magnon states and high-momentum SW states are different from those in the usual Heisenberg model.

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Note added in proof. After this paper was submitted, we received a preprint from T A Kaplan and S D Mahanti. They also calculated the SW dispersion of the DE model for a small number of electrons or holes at $S = 3/2$. They conclude that the deviation of the SW dispersion from that of Heisenberg model will vanish at a *hole* concentration $x \sim 0.1$–0.5. We have calculated the SW dispersion for arbitrary S and larger systems, which gives results consistent with theirs. However, even for fillings where the SW dispersion agrees well with that in the Heisenberg model, the wavefunctions constructed using the SW operator are not a good approximation to the eigenstates. These results will be presented elsewhere.

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