Spin Wave Theory of Double Exchange Ferromagnets

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We construct the 1/S spin-wave expansion for double exchange ferromagnets at T=0. It is assumed that the value of Hund’s rule coupling, $J_H$, is sufficiently large, resulting in a fully saturated, ferromagnetic half-metallic ground state. We evaluate corrections to the magnon dispersion law, and we also find that, in contrast to earlier statements in the literature, magnon-electron scattering does give rise to spin wave damping. We analyse the momentum dependence of these quantities and discuss the experimental implications for colossal magnetoresistance compounds.

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The phenomenon of colossal magnetoresistance (CMR), with its potential technological applications, has motivated an extensive experimental and theoretical research effort directed at the understanding of properties of doped manganese oxides [1]. In particular, considerable attention has been paid to magnetic properties of these compounds. An ubiquitous feature shared by the numerous theoretical models of the CMR compounds is the presence of a strong ferromagnetic Hund’s rule exchange coupling, $J_H$, between the spins of itinerant $e_g$ electrons and the core spins of Mn ions. The kinetic energy of itinerant electrons is then minimized when the ionic spins are parallel to each other; this gives rise to the conduction-electron mediated double exchange ferromagnetism [2]. The physics of double exchange is thus completely different from that of both Heisenberg exchange and RKKY interaction (the latter corresponding to the case of small $J_H$).

Since the value of core spin is relatively large, $S = 3/2$, studying the effects of double exchange interaction in the large-S limit should provide at least a qualitative description of the low-temperature properties of the manganites. At the same time, the quantum nature of the core spins does affect the behaviour of the system in a profound way, as indicated by the variational results. At finite $S$, the continuum of Stoner-like single-particle excitations with finite energies is present even at $J_H \rightarrow \infty$ [3]; also, the spin wave spectra obtained variationally show appreciable quantum-spin corrections [4,5]. These conclusions are fully supported by the exact diagonalization studies of one-dimensional double exchange systems [6,7].

It is therefore somewhat surprising that, apart from the leading-order calculations [8–11] (which do not account for any quantum corrections), the perturbative spin wave theory of double exchange magnets remains underdeveloped. The objective of the present paper is to partially fill this gap. Our approach (unlike that of Ref. [3]) remains valid in the experimentally relevant case of large carrier densities. We will show that the subleading terms in the 1/S expansion, which originate from magnon-electron scattering, provide corrections to the magnon dispersion law, and also give rise to magnon damping. We will also see how the momentum dependence of these quantities is affected by the Fermi surface geometry. The experimental data will be discussed briefly.

We start with the usual double exchange Hamiltonian,

$$
\mathcal{H} = -\frac{t}{2} \sum_{\langle i,j \rangle, \alpha} (c_{i\alpha}^\dagger c_{j\alpha} + c_{j\alpha}^\dagger c_{i\alpha}) - \frac{J_H}{2S} \sum_{i,\alpha,\beta} \vec{S}_i \sigma^{\alpha\beta} c_{i\alpha}^\dagger c_{i\beta}.
$$

(1)

Here $c_{j\alpha}$ are the electron annihilation operators, $\vec{S}_i$ are the operators of the core (localized) spins located at the sites of a square (or simple cubic) lattice, and the vector $\sigma^{\alpha\beta}$ is composed of Pauli matrices. We assume, for simplicity, that only one conduction-electron orbital (with two possible values of spin projection, $\alpha = \uparrow, \downarrow$) is available per each site $i$. Throughout the paper we use units in which hopping $t$, $\hbar$, and the lattice spacing are all equal to unity, and we consider the $T=0$ case.

There is presently no reason to doubt that for $S \geq 3/2$ the ground state of the Hamiltonian (1) is ferromagnetic, at least for an infinite system in two or three dimensions and with a finite number of electrons per site, $x < 1$ (this is corroborated by the variational calculations [8–11,3]; see also Ref. [4] for the 1D case, and Ref. [6] for finite-size systems). To leading order in $1/S$, the electron spectrum in the ferromagnetic state is given by

$$
\epsilon_{k}^{\uparrow} = \epsilon_{k}^{\downarrow} = \epsilon_k + J_H/2,
$$

for the tight-binding model of Eq. (1), $\epsilon_k = -\sum_{\alpha=1}^{d} \cos k_\alpha d$, and $d$ is the dimensionality of the lattice [12]. We will only consider the half-metallic case [6] when the chemical potential lies below the bot-
ton of the upper band, $\epsilon_F < \frac{1}{4}J_H - d + \mathcal{O}(1/S)$, so that only spin-up electrons are present in the ground state.

The magnon operators $a_i$ are introduced by means of the Holstein–Prakashoff transformation (including the subleading terms) of the operators $\hat{S}_i$, and the canonical transformation $[\hat{H}] \rightarrow \hat{H}' = \exp(-U) \hat{H} \exp(U)$ with

$$U = \frac{J_H}{\sqrt{2}N} \sum_{\vec{k},\vec{p}} \left( c_{\vec{k}}^\dagger c_{\vec{k} + \vec{p}} a_{\vec{p}}^\dagger - \text{h.c.} \right)$$

(where $N$ is the number of lattice sites and the summation is over the first Brillouin zone) is then applied to the Hamiltonian, thereby accomplishing the transition to the “true magnons”. The resultant Hamiltonian can be written as an expansion in powers of $S^{-1/2}$:

$$\hat{H}' = \sum_{\vec{k},\alpha} e_{\vec{k}} c_{\vec{k}}^\dagger c_{\vec{k}}^\alpha + \hat{H}_2' + \hat{H}_3' + \hat{H}_4' + \ldots \ .$$

Here, the term $\hat{H}_1' \propto S^{-1/2}$ has been eliminated by the canonical transformation, and

$$\hat{H}_2' = \frac{J_H}{4NS} \sum_{1,2,3,4} \left( \left\{ c_{1\vec{k}}^\dagger c_{1\vec{q} + \vec{p}} c_{2\vec{q}} c_{2\vec{p}} + \epsilon_1 - \epsilon_{1\vec{k} + \vec{p}} c_{1\vec{k}}^\dagger c_{2\vec{p}} a_{1\vec{k}}^\dagger a_{1\vec{p}} \right\} + \epsilon_{2\vec{k} + \vec{q}} - \epsilon_{2\vec{k}} c_{2\vec{k}}^\dagger c_{1\vec{q}} c_{1\vec{k}}^\alpha a_{4\vec{k}} a_{4\vec{q}} 

- \epsilon_{2\vec{k}} c_{2\vec{q}} c_{1\vec{k}}^\dagger c_{1\vec{q}}^\alpha a_{4\vec{k}} a_{4\vec{q}} + \text{h.c.} \right) - \frac{J_H}{2NS} \sum_{\vec{k},\vec{q}} c_{\vec{k}}^\dagger c_{\vec{q}} \ - \epsilon_{2\vec{k}} c_{2\vec{q}} c_{1\vec{k}}^\dagger c_{1\vec{q}}^\alpha a_{4\vec{k}} a_{4\vec{q}}$$

Here, the subscripts $1, 2, 3, 4$ stand for $\vec{p}_1, \vec{p}_2, \ldots$, and $\epsilon_{1\vec{k}}$ means that the quasi-momentum conservation law is enforced. The spin wave energy $\omega_{\vec{q}}$ is equal to the on-shell value of the real part of the magnon self energy (cf. Ref. [12]). The leading-order term in self energy, which originates from $\hat{H}_2'$, is real and coincides with the earlier results [11,13,14,15], which in the $J_H \rightarrow \infty$ limit reduce to

$$\omega_{\vec{q}}^{(0)} = -(2S)^{-1} \int n_{\vec{q}}(\epsilon_{\vec{q}} - \epsilon_{\vec{q} + \vec{p}}) d^d q / (2\pi)^d ,$$

where $n_{\vec{q}}$ is the Fermi distribution function for the spin-up electrons. Using the explicit form of $\epsilon_{\vec{q}}$, one obtains a Heisenberg-like expression, $\omega_{\vec{q}}^{(0)} = |E|/(d + \epsilon_{\vec{q}})/2dS$. Here, $E = \int \epsilon_{\vec{q}} n_{\vec{q}} d^d q / (2\pi)^d$ is the total energy of electrons calculated with respect to the center of the lower band.

Evaluation of the higher-order terms in Eq. (3) involves repeated commutations of the operator $\hat{U}$ (see Eq. (4)) with the original Hamiltonian, Eq. (1). In order to calculate the leading correction to the energy of a single magnon, we will need to collect only the terms which contain neither $c_{\vec{k}1}$ nor $c_{\vec{k}2}$ and are quadratic in magnon operators. Such terms do not occur in $\hat{H}_3'$, and we find

$$\hat{H}_4' \equiv \frac{J_H}{32S^2 N^2} \left\{ - \sum_{1+4} c_{1\vec{k}}^\dagger c_{1\vec{q} + \vec{p}} a_{1\vec{k}}^\dagger a_{1\vec{q}} - \sum_{\vec{q}} \left[ \frac{c_{1\vec{k}}^\dagger c_{1\vec{q}}^\dagger c_{2\vec{p}} a_{1\vec{k}}^\dagger a_{1\vec{q}} a_{1\vec{p}}}{\epsilon_{1\vec{k}} - \epsilon_{1\vec{q}} - \epsilon_{2\vec{p}} + \epsilon_{2\vec{q} + \vec{p}} + \epsilon_{1\vec{k} + \vec{p}}} \right] \right\}$$

for

$$\omega_{\vec{q}}^{(1)} = \frac{1}{16S^2} \int \left( 3\epsilon_{\vec{q}} - \epsilon_{\vec{q} + \vec{p}} \right) n_{\vec{q}} \frac{d^d q}{(2\pi)^d} + \text{Re}\Sigma_{\vec{q}}^{\pi}(0, \vec{p}) ,$$

$$\Sigma_{\vec{q}}^{\pi}(\omega, \vec{p}) = \frac{1}{16S^2} \int \left( \epsilon_{\vec{q}} + \epsilon_{\vec{q} + \vec{p}} - 2\epsilon_{\vec{p} + \vec{q}} \right) (1 - n_{\vec{q}}) n_{\vec{q}} \ d^d q d^d k / (2\pi)^d .$$

For the $x \ll 1$ case, this agrees with Ref. [12].

For a 2D system with band filling values $x = 0.3$ and $x = 0.4$, the quantity $\omega_{\vec{q}}^{(1)} S^2$ is plotted in Fig. 1a (solid and dotted lines, respectively). The dashed lines represent the corresponding Heisenberg-like fits, $\omega_{\vec{q}}^{(1)} S^2 = 2D^2(2 + \epsilon_{\vec{q}})S^2$, where $\epsilon_{\vec{q}}$ is the first quantum correction to spin stiffness. The deviation of $\omega_{\vec{q}}^{(1)}$ from $\omega_{\vec{q}}^{(1)} S^2$ is not large, but it increases dramatically for smaller $x$, in agreement with the numerical and variational results.

The doping dependence of the spin stiffness, $D = D_0 + D^{(1)}$, for $S = 3/2$ is shown in Fig. 1b (solid line). The dashed line represents the leading-order term, $D^{(1)} = |E|/8S$. We see that for $S = 3/2$ the value of $D^{(1)}$ is not small numerically [10], implying that quantum corrections to the spin wave spectrum cannot be omitted in any quantitative treatment [10]. Note that $D(x)$ is not symmetric with respect to quarter filling, $x = 0.5$, reflecting the loss of particle-hole symmetry at finite $S$.

Spin wave damping is given by the on-shell value of the imaginary part of magnon self energy, and the first-order perturbation theory terms obviously do not contribute to it. It is also easy to see that to second order in $1/S$, $\text{Im}\Sigma_{\vec{q}}^{\pi}(\omega_{\vec{q}}^{\pi}, \vec{p}) = \text{Im}\Sigma_{\vec{q}}^{\pi}(0, \vec{p}) = 0$. This is because the energies of all the intermediate states occurring in the second-order perturbation theory terms are higher than that of a single-magnon state (which to leading order equals the ground state energy). Therefore, the integration contour in the energy space terminates at the pole
(cf. Eq. 6 with $\omega = 0$), so that the latter does not give rise to any imaginary contribution. In the third order in $1/S$, a multitude of new diagrams appear, combining all possible vertices from $H_2^p$, $H_3^p$, and $H_4^p$. By the above reasoning, their leading-order imaginary parts vanish on shell (20), with one exception. This exception is the second diagram in Fig. 2, which corresponds to the term with the second-order pole in the standard third-order perturbation theory formula (21). This is nothing but a self-energy correction to $\Sigma_2(\omega, \vec{p})$ (Eq. (8)), and the net third-order contribution to the magnon relaxation rate is thus given by the diagram on the r. h. s. in Fig. 2 (22):

$$\Gamma(\vec{p}) = \frac{\pi}{48\pi^2} \int \frac{d^d\vec{q}}{(2\pi)^d} J(\vec{p}, \vec{q}) \delta(\omega(\vec{p})) \delta(\omega(\vec{q})), \quad (9)$$

$$J(\vec{p}, \vec{q}) = \int (\epsilon_{\vec{q}} - \epsilon_{\vec{p}+\vec{q}})^2 (1 - n_{\vec{q}+\vec{r}}) n_{\vec{q}} \times$$

$$\times \delta(\omega(\vec{p}) - \omega(\vec{q})) \frac{d^d\vec{q}}{(2\pi)^d} \delta(\omega(\vec{q})), \quad (10)$$

Note that $J(\vec{p}, \vec{q})$ is of the order of $1/S$.

Thus in a double exchange system, the spin wave linewidth remains finite even in a $J_H \to \infty$, $T = 0$ case. While such a possibility was discussed earlier (1) in connection with recent experiments (23), it is at variance with the prevalent view (24, 25). Magnon damping in a double exchange ferromagnet is due to magnon-electron scattering: a magnon can scatter into a lower-momentum state, while exciting an electron from under the Fermi level (see Fig. 2). The momentum transfer into the electronic subsystem allows us to speculate that an electrical current may arise in this process.

The value $\Gamma(\vec{p})$ for different values of bandfilling $x$ in the 2D case is plotted in Fig. 2 c. It shows strong momentum dependence, reaching (for $S = 3/2$) up to 10% of the corresponding magnon energy. We note a difference from both a Heisenberg ferromagnet (with $\Gamma(\vec{p}) \equiv 0$ at $T = 0$) and an RKKY ($s-f$ exchange) ferromagnet, where the leading term in $\Gamma(\vec{p})$ is threshold-like (26).

We note that Eqs. (10) (as well as Eqs. (9, 11)) remain unchanged if the first term in the Hamiltonian (8) is written as $\sum_{k, \alpha} \epsilon_k c_k^\dagger c_{k+q}^\alpha$, with an arbitrary $\epsilon_k$. It is thus possible to use Eqs. (10) to evaluate $\Gamma(\vec{p})$ for an arbitrary single-band electron dispersion. The overall profile of $\Gamma(\vec{p})$ (as well as that of $\omega(\vec{p})$) is sensitive to both the carrier concentration value $x$ and the details of electron bandstructure throughout the Brillouin zone. There are, however, two generic features in the behaviour of $\Gamma(\vec{p})$:

1. **Long-wavelength limit.** When $p$ is small in comparison to the Fermi momentum $k_F$, $\Gamma(\vec{p})$ is proportional to $p^2$ in the 2D case and to $p^6$ in three dimensions (23). Here, the factor $p^2$ originates from the matrix element $(\epsilon_\vec{q} - \epsilon_{\vec{p}+\vec{q}})^2$ in Eq. (11), and is multiplied by a $p^4$ corresponding to the space volume available to the virtual magnon with momentum $\vec{p} - \vec{r}$ and energy $\omega_{\vec{p} - \vec{r}} < \omega(\vec{p}) \approx D(0)p^2$ (see Fig. 2). The remaining factor of $p^4$ comes from electron kinematics. In the one dimensional case, $\Gamma(\vec{p}) \equiv 0$ everywhere at $p < k_F$.

2. **The anomaly at $p = k_F$.** The Fermi surface geometry manifests itself through a weak singularity of $\Gamma(\vec{p})$ at $p = k_F$ (27). Indeed, the quantity $J(\vec{p}, \vec{r})$ (see Eq. (10)) has a singularity at $r = 2k_F$, and vanishes at $\omega_{\vec{p} - \vec{r}} = \omega_q(0)$. The anomaly in $\Gamma(\vec{p})$ is due to the tangency between these two surfaces (in $\vec{r}$-space), which occurs at $p = k_F$. At this point, the second (in 2D) or third (in 3D) derivative of $\Gamma(p)$ suffers either a jump or a logarithmic divergence, depending on the local geometry of these two tangent surfaces. These singularities are too weak to be visible in Fig. 2 c, except for $x = .49$, when owing to the flatness of the Fermi surface the singularity acquires a nearly-1D (jump in the first derivative) character.

The anomaly in $\Gamma(\vec{p})$ is of course accompanied by a singularity in the third-order (in $1/S$) term in $\omega_F$. It is contained in the real part of the same diagram, and for an isotropic dispersion law amounts to a jump in the second derivative (2D) or to a logarithmic divergence in either the first (1D) or third (3D) derivative of $\omega_F$.

In view of the anticipated Fermi surface geometry of the CMR compounds (28), we mention the case when the Fermi surface is nearly-cubic (nearly square). As the magnon isoenergetic surfaces are unlikely to have the same shape (29), this case is not reducible to 1D. In fact, when $\vec{p}$ is perpendicular to a flat part of the Fermi surface, the singular term in $\Gamma(\vec{p})$ is proportional to $p - k_F)^3/\theta(p - k_F)$ in 2D and to $(p - k_F)^3/\theta(p - k_F)$ in 3D (27). The singularity in $\omega_F$ is given by $-\ln(p - k_F)^3/\theta(p - k_F)$ in 2D and by $(p - k_F)^2\ln|p - k_F|$ in 3D.

Some comments are in order concerning the experimental situation. Low temperature spin dynamics, including spin wave damping and the deviation of magnon spectrum from the Heisenberg-like form, is studied intensively in both 3D perovskite (23, 26, 31) and quasi-2D layered (22) CMR manganites. In both cases, unexpectedly large low-$T$ values of $\Gamma(p)$ at large $p$ were found, which is consistent with our results (in some compounds (31), $\Gamma(p)$ may be due in part to electron-phonon coupling). However, the lack of detailed knowledge of electron bandstructure restricts one’s options in comparing theoretical and experimental results quantitatively.

The low-momentum measurements of $\Gamma(\vec{p})$ in the perovskite manganites (24) yield the $p^4(\ln(T/\omega_F))^2$ dependence, attributable to the magnon-magnon scattering (33). We suggest that these measurements, both in 2D and 3D compounds, should be performed at lower temperatures in order to reveal the $p^5$ or $p^6$ contribution of magnon-electron scattering (which is likely to have a numerically small prefactor (23)). One should also try to identify the $p = k_F$ anomalies in $\Gamma$ and $\omega$ (the irreversibility seen in the data of Ref. (23)) at $p \sim \pi/2$ may be a possible candidate). We also suggest that a systematic study of doping dependence of spin stiffness should be
In conclusion, we have calculated the spin wave linewidth and correction to the spin wave energy for a double exchange half-metallic ferromagnet. The proposed new measurements would verify to what extent the double exchange model accounts for the low-temperature spin dynamics of the CMR compounds.

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Note added in proof: Our Eq. (4) agrees with Ref. [3], reporting a 1/S expansion for the model with a strong ferromagnetic exchange, $J_f$, between the core spins. However, the second-order calculation in itself is not sufficient to evaluate the magnon damping at $J_f = 0$, given by Eq. (8) above.

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FIG. 1. (a) The leading 1/S-corrections to the spin wave energy in a 2D system with carrier concentration $x = 0.3$ (solid line) and $x = 0.4$ (dotted line) as functions of momentum. The dashed lines represent opposite Heisenberg-like fits, $\omega^{(1)} = S^2$. (b) Doping dependence of spin stiffness $D$ in a 2D system with $S = 3/2$. The dashed line corresponds to the classical value, $D^{(0)}$. (c) Momentum dependence of spin wave damping, $\Gamma(p)$, for a 2D system with $x = 0.49$, $x = 0.4$, and $x = 0.3$ (solid, dotted and dashed lines, respectively). For $x = 0.49$, note the $p = k_F$ anomaly, visible at $\bar{\rho} \approx \pi/2, \pi/2$ (see the inset), and the smallness of $\Gamma(2k_F)$.

FIG. 2. Magnon self energy diagrams. Dashed and solid lines are the unperturbed Green’s functions of magnons, $(\omega + i\theta)^{-1}$, and of the spin-up electrons, respectively. The bold dashed line is the exact magnon Green’s function, which to required accuracy is given by $(\omega - \omega^{(0)} + i\theta)^{-1}$. The boxes represent the interaction, i.e. the $c_\uparrow^\dagger c_\uparrow a^\dagger a$-terms in Eq. (4).
Fig. 1
Fig. 2