Non–Heisenberg Spin Dynamics of Double-Exchange Ferromagnets with Coulomb Repulsion

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With a variational three–body calculation we study the role of the interplay between the on-site Coulomb, Hund’s rule, and superexchange interactions on the spinwave excitation spectrum of itinerant ferromagnets. We show that correlations between a Fermi sea electron–hole pair and a magnon result in a very pronounced zone boundary softening and strong deviations from the Heisenberg spinwave dispersion. We show that this spin dynamics depends sensitively on the Coulomb and exchange interactions and discusses its possible relevance to recent experiments in the manganites.

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The interaction between itinerant carrier spins and localized magnetic moments leads to ferromagnetic order in a wide variety of systems [1]. Examples include the manganese oxides (manganites) $\text{R}_1\text{A}_x\text{MnO}_3$ ($\text{R}=\text{La, Pr, Nd, Sm, · · ·}$ and $\text{A}=\text{Ca, Ba, Sr, Pb, · · ·}$) and the III–Mn–V ferromagnetic semiconductors [2]. Such systems are of great current interest due to their novel potential applications. For example, the manganites display colossal magnetoresistance [2], while ferromagnetic semiconductors raise the possibility of multifunctional quantum devices that combine information processing and storage on a single chip with low power consumption [4]. In such materials, the magnetic and transport properties are intimately related and can be controlled by varying the itinerant carrier concentration and dimensionality.

In the manganites, $n=1-x$ itinerant electrons per Mn atom partially fill a $d$–band with $e_g$ symmetry. Their concentration, $n$, is controlled by the hole doping, $x$. The $d$–band kinetic energy $K$ is determined by the hopping energy between the neighboring lattice sites, $t \sim 0.2–0.5$eV. The itinerant electron spins interact strongly with localized spin–$S$ magnetic moments (Hund’s rule coupling $H_{\text{exch}}$ with strength $J \sim 2$eV$\times t$). $S=3/2$ comes from the three electrons in the tightly bound $t_{2g}$ orbitals. This ferromagnetic interaction competes with the direct antiferromagnetic interactions ($H_{\text{AF}}$) between neighboring local spins, $J_{\text{AF}} \sim 0.01t$. The largest energy scale in the manganites is given by the on-site Coulomb repulsion between the itinerant electrons, $U \sim 3.5–8$eV ($H_U$). This Coulomb interaction is generally difficult to treat and its effects have received less attention. Here we focus on the role of $U$ on the spin dynamics in the concentration range $0.5 \leq n \leq 0.8$ where metallic behavior is observed in both 3D and quasi–2D (layered) manganites.

The ferromagnetic order in the manganites can be interpreted to first approximation by invoking the double exchange mechanism and the $J \rightarrow \infty$ limit of the minimal Hamiltonian $K + H_{\text{exch}}$ [2, 3]. An itinerant carrier is allowed to hop on a lattice site only if its spin is parallel to the local spin on that site. The kinetic energy is thus reduced when all spins are parallel. This favors the ferromagnetic state $|F\rangle$, which describes local spins with $S_z = S$ on all lattice sites and a Fermi sea of spin–$\uparrow$ electrons. The above spins are often treated as classical, justified for $S \rightarrow \infty$ [2]. In this limit, the system can be described by a nearest neighbor Heisenberg model with ferromagnetic interaction. Quantum effects are often treated perturbatively in $1/S$ [6, 7]. To $O(1/S)$, one thus obtains noninteracting Random Phase Approximation (RPA) magnons, whose dispersion in the strong coupling limit coincides with that of the nearest neighbor Heisenberg ferromagnet $\tilde{H}_0$. Such a dispersion was observed experimentally for concentrations $n > 0.7$ [8].

However, strong deviations from the short range Heisenberg model spinwave dispersion were observed for $n \leq 0.7$ in both 3D [9, 10, 11] and quasi–2D manganites [12]. Most striking is the strong spinwave softening close to the zone boundary [9, 10, 11]. This indicates a new spin dynamics in the metallic ferromagnetic phase whose physical origin is still unclear [11]. The proposed mechanisms involve orbital degrees of freedom, magnon–phonon interactions, disorder, bandstructure effects, and the Hubbard repulsion [2, 6, 10, 11, 13, 19]. The zone boundary softening can be fitted phenomenologically by adding long range interactions to the Heisenberg Hamiltonian $\tilde{H}_0$ [10, 11]. Ye et.al. [11] found that the above softening increases with $x=1-n$, while the dispersion for low momenta only changes weakly. They argued that none of the existing theories can explain these experimental trends [11].

In this paper we study the concentration dependence of the spinwave dispersion predicted by the model Hamiltonian $H = K + H_{\text{exch}} + H_U + H_{\text{AF}}$ [2, 6, 14] with a single $e_g$ orbital per lattice site. We treat exactly the long–range magnon–Fermi sea pair three–body correlations induced by the interplay between $H_U$ and $H_{\text{exch}}$ with a variational wavefunction. We show that such correlations lead to strong deviations from the RPA and Heisenberg spinwave dispersions. These deviations, as well as the stability of the ferromagnetic order, depend
sensitively on \( H_U \). Our approach interpolates between
the strong/weak coupling and \( n = 0 \)/\( n = 1 \) limits with the
same formalism and can therefore address the intermediate interactions and \( n \) relevant to the manganites. At the same
time, it recovers the \( 1/S \) expansion \( [6] \) and exact numerical results \( [15, 16] \) as special cases. We find
that magnon–Fermi sea pair correlations due to \( U \) result
in a pronounced zone boundary spinwave softening that
increases with \( x \) (similar to the experiment \( [11] \)) in a way
that depends on \( U \) and \( J \). Our variational calculation sets
a lower bound on the magnitude of this softening.

**Method**—We use the variational wavefunction \(|Q\rangle = M_1 Q \langle F | \), where the operator \( M_1 Q \) conserves the total
momentum \( Q \) and lowers the \( z \)–component of the total
spin by 1. This spin reversal can be achieved either by lowering
the localized spin \( z \)–component, via the collective spin
operator \( S_\perp \) \([14]\), or by coherently promoting an electron
from the spin–up to the spin–down band; it may also be accompanied
by the scattering of Fermi sea pairs. Neglecting multipair excitations, the most general \( M_1 Q \) is \([14]\)

\[
M_1 Q = S_Q + \sum_\nu X_\nu Q^\dagger c^\nu_\perp c_\perp^\dagger + \sum_{\alpha \mu} c^\alpha_\mu c^\dagger_\mu \times \\
[\Psi_{\alpha \mu} S_{Q+\mu-\alpha} + \frac{1}{2} \sum_\nu \Phi_{\alpha \mu \nu} c^\dagger_\nu Q_{\perp-\alpha-\nu+\nu} c^\dagger_\nu] \tag{1}
\]

where \( c^\alpha_\mu \) creates a spin–\( \sigma \), momentum–\( \mathbf{k} \) electron. \( \nu, \mu \)
(\( \alpha \)) label states inside (outside) the Fermi sea. The first
two terms create a magnon of momentum \( Q \). The last
two terms describe magnon scattering, \( Q \rightarrow Q + \mu - \alpha \),
accompanied by electron scattering across the Fermi surface,
\( \mu \rightarrow \alpha \) (Fermi sea pair shakeup). By setting
\( \Psi = \Phi = 0 \) we recover the RPA results \([14]\). However,
here the variational parameters \( X_\nu Q, \Psi_{\alpha \mu} \) and \( \Phi_{\alpha \mu \nu} \) are
not restricted in any way; unlike in Ref. \([17]\), we do not
assume any particular form or momentum dependence.

By solving the full variational equations numerically for
fairly large \( N \times N \) lattices (\( N \sim 20 - 30 \)), we put up an upper
bound on the spinwave excitation energies \( \omega_Q \) (with
respect to \( |F\rangle \)) that converges with \( N \) and thus reflects
the thermodynamic limit. We can therefore conclude that \( (i) \)
the exact dispersion is at least as soft as our results, \( (ii) \)
\( \omega_Q < 0 \) means that \( |F\rangle \) is not the ground state.

The wavefunction Eq.\( (1) \) offers several advantages. It
gives exact results in the two concentration limits \( n \rightarrow 0 \)
(one electron) and \( n = 1 \) (half–filling). In the special
cases \( H_U = H_{AF} = 0 \) and \( H_{TF} = H_{AF} = 0 \) it agrees
very well with exact results \( [14, 10] \). Our results also become
exact in the atomic limit \( t \rightarrow 0 \) \([14, 13]\) and should therefore
handle local correlations well. While the latter
dominate in the strong coupling limit, long range correlations become important as \( J/t \) and \( U/t \) decrease \([13]\).
The experiment \([10, 11]\) points out the importance of
long–range interactions. Eq.\( (1) \) treats exactly all correlations
between a single Fermi sea pair and a magnon. The
only restriction of Eq.\( (1) \) is that it neglects contributions from
two or more Fermi sea pairs, which are however
suppressed for large \( S \) \([14]\) and in 1D \([15, 18]\).

**Results**—Fig.\( 1 \) shows the calculated three–body
spinwave dispersion for \( U = 25t \) (Fig.\( 1 \)a) and \( U = 10t \)
(Fig.\( 1 \)b)). It compares this to the RPA \( (\Psi = \Phi = 0 \) \)
and the results of Ref. \([17]\), which we recover by expanding
the RPA to \( O(1/S) \) and \( O(1/(JS+NU)) \). Fig.\( 1 \)a also
compares to the Heisenberg dispersion obtained by taking
the limit \( J \rightarrow \infty, U = 0 \) of the RPA (rather than by
fitting). The latter deviates strongly from our intermediate
coupling results. While the RPA agrees well with Ref. \([6]\),
the Fermi sea pair–magnon correlations lead to a
very strong softening (deviations \( \sim 100\% \) from the RPA).

The on–site Coulomb repulsion \( U \) increases the spinwave
energies and therefore the stability of the ferromagnetic state \( |F\rangle \). Fig.\( 1 \)c demonstrates this hardening
along \( \Gamma - X \) \((0,0) \rightarrow (\pi,0) \) as \( U \) increases in steps
\( \Delta U = 5t \). While initially the energies increase strongly
with \( U \), their relative change decreases with increasing
\( U \). Nevertheless, full convergence to the \( U \rightarrow \infty \) result
(dashed curve in Fig.\( 1 \)c) only occurs for very large \( U \).

Ref. \([14]\) treated the effects of strong \( U \) by mapping the
problem to a Hamiltonian with \( U = 0 \) \([14]\) and renormalized
hopping \( t(n) \). The magnon excitations were then described
within the RPA. Due to the increase in the effective
\( J/t(n) \), \( U \) resulted in higher spinwave energies. Here
we show that carrier–magnon correlations beyond the
RPA, induced by \( U \), lead to a pronounced zone boundary
softening as compared to the RPA. This can be seen in
Fig.\( 1 \)d, which shows the percentage deviation from the RPA
at the \( X \)–point as function of \( U \) (maximum is \( 100\% \)). While the deviations from the RPA decrease with increasing \( U \), they remain quite large for the typical \( U \).

We now focus on the dependence of the \( X \)–point en-
energy on \( n \). Refs.\[8, 10, 11\] found that the deviation, at this zone boundary, of the nearest–neighbor Heisenberg model dispersion that fits the experiment at small \( Q \) increases with \( x=1-n \). The experimental dispersion along all directions in the Brillouin zone was fitted by a Heisenberg model with both 4th–nearest–neighbor \((J_4)\) and next–nearest–neighbor \((J_1)\) exchange couplings; 2nd– and 3rd–nearest–neighbor interactions were negligible.\[11\]. The ratio \( J_4/J_1 \propto x \) becomes strong for \( n \leq 0.7 \) \[11\].

Our numerical results can also be fitted very well to the \( J_1-J_4 \) Heisenberg model. Fig. \ref{fig:fig2} shows the behavior of \( J_1(n)/J_1(n) \) (and thus the spinwave softening) for different \( J \). The crucial role of the pair–magnon correlations is clear by comparing to the RPA. The RPA gives small \( J_1/J_1 \) (in the strong coupling limit it coincides with the nearest–neighbor Heisenberg dispersion). However, the pair–magnon correlations greatly enhance \( J_4/J_1 \) (and the softening), typically by a factor 3–4 or higher in Fig. \ref{fig:fig2} \( J_4/J_1 \) increases rapidly with \( x=1-n \) until it reaches its maximum. For large \( J/t \), \( J_4/J_1 \) increases more slowly with \( x \). This increase is sharp for smaller \( J \), as the ferromagnetic state becomes less stable (compare Figs. \ref{fig:fig2} \( a \) and \ref{fig:fig2} \( d \)). On the other hand, \( J_4/J_1 \) is small for \( n > 0.7 \).

Next we turn to the spinwave dispersion for small \( Q \). Its behavior is characterized by the stiffness \( D(n) \), obtained by fitting the small–\( Q \) dispersion to the form \( DQ^2 \). Fig.\ref{fig:fig3} \( a \) compares our results to Ref.\[8, \] Eq.\[1\], and the RPA. The pair–magnon correlations decrease \( D(n) \) by as much as \(~100\%\) as compared to Ref.\[8\] and by as much as \(~50\%\) from the RPA. Fig.\ref{fig:fig3} \( a \) demonstrates a plateau as function of \( n \), where \( D(n) \) remains fairly constant within a wide range of \( n \) relevant to the manganites. The pair–magnon correlations decrease the dependence of \( D \) on \( n \) for such concentrations (compare the three curves in Fig.\ref{fig:fig3} \( a \)). As shown in Fig.\ref{fig:fig3} \( b \), \( U \) increases the stiffness. Overall, Figs. \ref{fig:fig2} and \ref{fig:fig3} are consistent with the main experimental trends.\[11\]. However, in Ref.\[11\] \( D(n) \) was found to be fairly constant over a wider range of \( n \). Fig.\ref{fig:fig2} and Fig.\ref{fig:fig3} show that the pair–magnon correlations suppress the dependence of \( D \) on \( n \) while enhancing \( J_4 \). We speculate that the differences from the experiment may be due to the bandstructure effects neglected here.

We now turn to the origin of the zone boundary softening and show that it is dominated by strong correlations due to \( U \). We set \( J_{AF} = 0 \). Similar to Ref.\[14\], the spinwave dispersion \( \omega_Q \) is determined by the amplitude \( X_Q \), Eq.\[1\], describing the coherent spin\(↑→\)spin\(↓ \) electron excitation (\( \propto \sum_{\mu} X_{\mu} \)) and by the amplitude \( \Psi \) describing magnon–pair scattering. The dominant new effect here comes from the renormalization of \( X_{\mu} \) by the scattering, due to \( U \), of a spin\(↑→\)spin\(↓ \) excitation with a Fermi sea pair. The corresponding interaction process is described by the amplitude \( \Phi \) in Eq.\[1\] and is shown schematically in Fig. \ref{fig:fig4} \( a \). The Fermi sea pair (\( \mu, \alpha \)) is created by interacting with the spin\(↓ \) electron via \( U \). Such scattering gives a contribution \( \propto U \sum_{\mu} \Phi_{\mu\mu} \) to \( X_{\mu} \). In Fig.\ref{fig:fig3} \( c \) we plot this correlation contribution, both for \( Q \) close to the \( X \)–point and for small \( Q \), as function of momentum \( \mu \) for \( n = 0.6 \) where the softening is pronounced. We consider momenta \( \mu \parallel Q \) (\( \mu_x \), contribution \( \Phi_{\parallel} \)) and momenta \( \mu \perp Q \) (\( \mu_y \), contribution \( \Phi_{\perp} \)). As can be seen in Fig.\ref{fig:fig3} \( c \), the largest correlation contribution comes for \( \mu \parallel Q \) close to the Fermi surface (which for the concentrations of interest is close to the zone boundary) and for \( Q \) close to the zone boundary. In Fig.\ref{fig:fig3} \( d \) we compare the spinwave energy from the full calculation with the results obtained by neglecting \( \Phi_{\perp} \) and/or \( \Phi_{\parallel} \). It is clear that the strong softening of the spinwave dispersion as compared to the
RPA comes from $\Phi^\parallel$, i.e. from the renormalization of $X_\mu$ by the scattering of a spin $\uparrow \rightarrow \downarrow$ excitation with a Fermi sea pair for momenta $\mu$ along $\Gamma$–$X$.

With decreasing $J/t$, the magnon energy for intermediate $n$ turns negative at the $X$–point while the magnon stiffness is still positive. This variational result allows us to conclude instability of the ferromagnetic state. On the other hand, for small $n$, the spinwave energy first turns negative at the $(\pi, \pi)$ point (antiferromagnetic correlations). Finally, for larger $n$, the spinwave energy turns negative at small momenta first, $D < 0$. By identifying the minimum values of $J$, $J_c(n)$, where $\omega_Q \geq 0$ for all momenta, we can definitely conclude, due to the variational nature of our calculation, that the ground state is not ferromagnetic for $J < J_c$. On the other hand, for $J > J_c$, the stability of $|F\rangle$ is not guaranteed.

$J_c(n)$ is shown in Fig.4(b). By comparing to the RPA, it is clear that the pair–magnon correlations lead to a very pronounced upward shift of the ferromagnetic phase boundary. While for large $n$ the correlation effects diminish, and the RPA becomes exact at $n=1$, for $n < 0.7$ the deviations from the RPA exceed 100%. As $n$ decreases further, the RPA fails completely and we can conclude that it grossly overestimates the stability of the ferromagnetism. Even though additional effects (e.g. phase separation [2,6] and charge ordering [20]) will further increase $J_c(n)$ for some $n$, our variational calculation allows us to conclude that Fermi sea pair–magnon correlations are strong in the manganites and should be treated beyond the mean field theory of Refs. [6,20].

We conclude that non–perturbative long range electron–hole pair–magnon correlations play a very important role in the spin dynamics of the manganites. Most important is the strong softening of the spinwave dispersion and the decrease in the stability of the ferromagnetic state. These correlation effects depend sensitively on the onsite Coulomb repulsion and on its interplay with the magnetic exchange and superexchange interactions. We propose that the scattering of magnons by charge excitations plays an important role in interpreting recent experiments [11]. Our work can be extended to other itinerant ferromagnetic systems (e.g. III(Mn)V semiconductors) that are far from the strong coupling limit. The correlations discussed here should also play an important role in the ultrafast magnetization dynamics measured by pump–probe optical spectroscopy [21].

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