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Non-Heisenberg spin dynamics of double-exchange ferromagnets with Coulomb repulsion

M. D. Kapetanakis and I. E. Perakis
Department of Physics, University of Crete, Crete, Greece
and Institute of Electronic Structure & Laser, Foundation for Research and Technology—Hellas, Heraklion, Crete, Greece

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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 spin-wave 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 spin-wave dispersion. We show that this spin dynamics depends sensitively on the Coulomb and exchange interactions and discuss 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[2] (manganites) $R_{1-x}A_xMnO_3$ (R=La,Pr,Nd,Sm,… and A=Ca,Ba,Sr,Pb,…) and the III-Mn-V ferromagnetic semiconductors.[3] 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_{exch} with strength $J\sim 2$ eV$>t$). $S=3/2$ comes from the three electrons in the tightly bound t_{2g} orbitals. This ferromagnetic interaction competes with the direct antiferromagnetic interactions (H_{AF}) between neighboring local spins, $J_{AF}\sim 0.01t$. 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 $0.5\leqslant n\leqslant 0.8$ where metallic behavior is observed in both three-dimensional (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 $K+H_{exch}$.[2,5] 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.[7] Such a dispersion was observed experimentally for concentrations $n>0.7$.[8]

However, strong deviations from the short-range Heisenberg model spin-wave dispersion were observed for $n\leqslant 0.7$ in both 3D (Refs. 9–11) and quasi-2D manganites.[12] Most striking is the strong spin-wave softening close to the zone boundary.[9–11] 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, band-structure 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.[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 spin-wave dispersion predicted by the model Hamiltonian $H=K+H_{exch}+H_U+H_{AF}$,[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_{exch} with a variational wave function. We show that such correlations lead to strong deviations from the RPA and Heisenberg spin-wave dispersions. These deviations, as well as the stability of the ferromagnetic order, depend sensitively on H_U. Our approach interpolates between the strong- and weak-coupling and $n=0$ and $n=1$ 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 spin-wave 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.

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Method. We use the variational wavefunction $|\mathbf{Q}\rangle = M_{\mathbf{Q}}^{\dagger}|F\rangle$, where the operator $M_{\mathbf{Q}}^{\dagger}$ conserves the total momentum $\mathbf{Q}$ 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 $\mathbf{S}_{\mathbf{q}}^{-}$,[14] or by coherently promoting an electron from the spin-$\uparrow$ to the spin-$\downarrow$ band; it may also be accompanied by the scattering of Fermi sea pairs. Neglecting multipair excitations, the most general $M_{\mathbf{Q}}^{\dagger}$ is[14]

$$M_{\mathbf{Q}}^{\dagger} = S_{\mathbf{Q}}^{-} + \sum_{\nu} X_{\nu}^{\mathbf{Q}} c_{\mathbf{Q}+\nu\downarrow}^{\dagger} c_{\nu\uparrow} + \sum_{\alpha\mu} c_{\alpha\uparrow}^{\dagger} c_{\mu\uparrow} \times \left(\Psi_{\alpha\mu}^{\mathbf{Q}} S_{\mathbf{Q}+\mu-\alpha}^{-} + \frac{1}{2} \sum_{\nu} \Phi_{\alpha\mu\nu}^{\mathbf{Q}} c_{\mathbf{Q}+\mu-\alpha+\nu\downarrow}^{\dagger} c_{\nu\uparrow} \right), \quad (1)$$

where $c_{\mathbf{k}\sigma}^{\dagger}$ creates a spin-σ, momentum-$\mathbf{k}$ electron. ν and μ (α) label states inside (outside) the Fermi sea. The first two terms create a magnon of momentum $\mathbf{Q}$. The last two terms describe magnon scattering, $\mathbf{Q} \rightarrow \mathbf{Q}+\mu-\alpha$, 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 $X_{\nu}^{\mathbf{Q}}$, $\Psi_{\alpha\mu}^{\mathbf{Q}}$, and $\Phi_{\alpha\mu\nu}^{\mathbf{Q}}$ are not restricted in any way; unlike in Ref. 17, we do not assume any particular form of momentum dependence. By solving the full variational equations numerically for fairly large $N \times N$ lattices ($N \sim 20-30$), we put an upper bound on the spin-wave excitation energies $\omega_{\mathbf{Q}}$ (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 and (ii) $\omega_{\mathbf{Q}} < 0$ means that $|F\rangle$ is *not* the ground state.

The wave function 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 $H_{exch} = H_{AF} = 0$ it agrees very well with exact results.[14,16] Our results also become exact in the atomic limit $t \rightarrow 0$,[14,18] and should therefore treat local correlations well. While the latter dominate in the strong-coupling limit, long-range correlations become important as J/t and U/t decrease.[18] The experiment[10,11] points out the importance of long-range interactions. Equation (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 (Ref. 14) and in 1D.[16,18]

Results. Figure 1 shows the calculated three-body spin-wave dispersion for $U=25t$ [Fig. 1(a)] and $10t$ [Fig. 1(b)]. It compares this to the RPA result ($\Psi=\Phi=0$) and the results of Ref. 6, which we recover by expanding the RPA to $O(1/S)$ and $O[1/(JS+nU)]$. Figure 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 spin-wave energies and therefore the stability of the ferromagnetic state

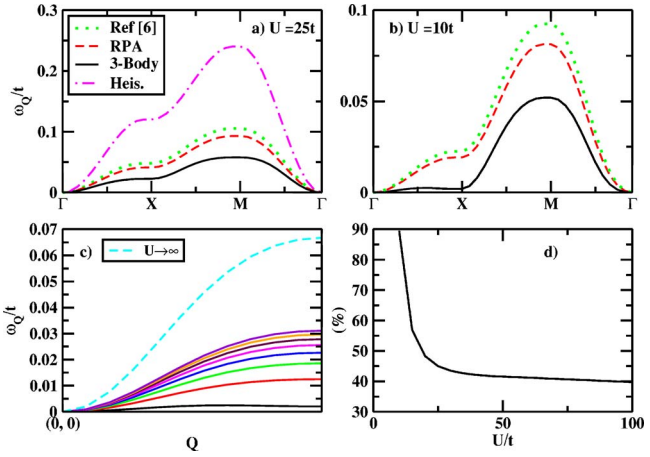

FIG. 1. (Color online) Spin-wave dispersion along different directions ($n=0.6$, $J=7t$, $J_{AF}=0.012t$). (a) $U=25t$. (b) $U=10t$. (c) Direction Γ-X: $U=10 \rightarrow 45t$ in increments of $5t$. (d) Deviation from the RPA: $1-\omega/\omega^{RPA}$ at X point.

$|F\rangle$. Figure 1(c) demonstrates this hardening along Γ-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.

Reference 19 treated the effects of strong U by mapping the problem to a Hamiltonian with $U=0$ (Ref. 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 spin-wave 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 a 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 energy on n. References 9–11 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* fourth-nearest-neighbor (J_4) and next-nearest-neighbor (J_1) exchange couplings; second- and third-nearest-neighbor interactions were negligible.[11] The ratio $J_4/J_1 \propto x$ becomes strong for $n \leqslant 0.7$.[11]

Our numerical results can also be fitted very well to the J_1-J_4 Heisenberg model. Figure 2 shows the behavior of $J_4(n)/J_1(n)$ (and thus the spin-wave softening) for different J. The crucial role of the pair-magnon correlations is made clear by comparing to the RPA. The RPA gives small J_4/J_1 (in the strong-coupling limit it coincides with the nearest-neighbor Heisenberg dispersion[14]). However, the pair-magnon correlations greatly enhance J_4/J_1 (and the softening), typically by a factor 3–4 or higher in Fig. 2. 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 in-

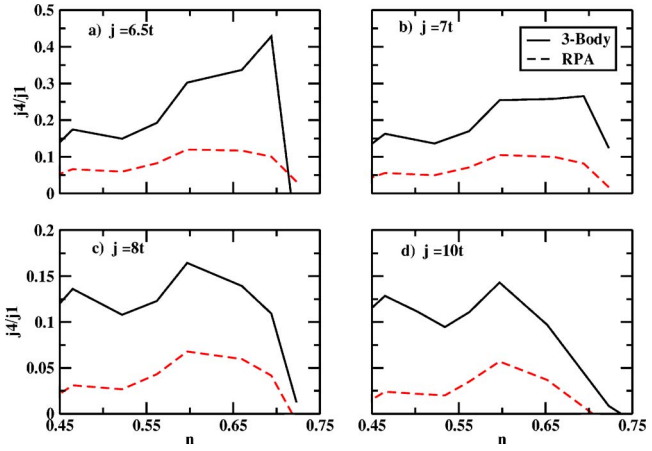

FIG. 2. (Color online) $J_4(n)/J_1(n)$ for $J_{AF}=0.012t$, $U=25t$ extracted by fitting our results to the first- plus fourth-nearest-neighbor Heisenberg model. The same behavior is exhibited by the spin-wave softening compared to the Heisenberg model.

crease is sharp for smaller J, as the ferromagnetic state becomes less stable [compare Figs. 2(a) and 2(d)]. On the other hand, J_4/J_1 is small for $n>0.7$.

Next we turn to the spin-wave 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. Figure 3(a) compares our results to Ref. 6, Eq. (1), and the RPA. The pair-magnon correlations decrease $D(n)$ by as much as $\sim$100% as compared to Ref. 6 and by as much as $\sim$50% from the RPA. Figure 3(a) demonstrates a plateau as a 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. 3(a)]. As shown in Fig. 3(b), U increases the stiffness. Overall, Figs. 2 and 3 are consistent with the main experimental trends.[10,11] However, in Ref. 11 $D(n)$ was found to be fairly constant over a wider range of n. Figures 2 and 3 show that the pair-magnon

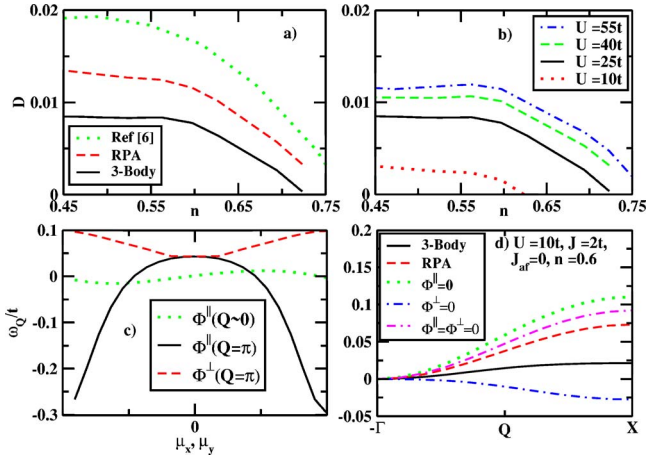

FIG. 3. (Color online) (a) Comparison of different approximations for $D(n)$ ($J=7t$, $U=25t$), (b) the effect of U on $D(n)$, (c) contribution of magnon-pair correlations for different momenta, and (d) origin of magnon softening. $J_{AF}=0.012t$.

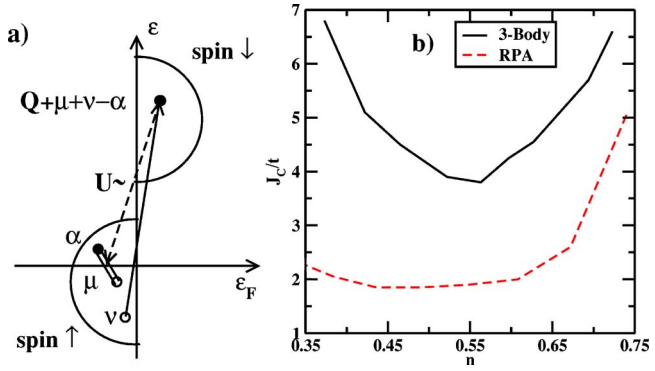

FIG. 4. (Color online) (a) Schematic describing the scattering of spin $\uparrow\rightarrow$spin $\downarrow$ electronic excitation with Fermi sea pair (μ,α) due to U. (b) $J_c(n)$ for $J_{AF}=0.012t$, $U=25t$. For $J<J_c$, the ferromagnetic state $|F\rangle$ is not the ground state.

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 band-structure 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 spin-wave dispersion $\omega_{\mathbf{Q}}$ is determined by the amplitude $X_\mu^{\mathbf{Q}}$, Eq. (1), describing the coherent spin $\uparrow\rightarrow$spin $\downarrow$ electron excitation ($\propto\Sigma_\mu X_\mu$) and by the amplitude Ψ, describing magnon-pair scattering. The dominant new effect here comes from the renormalization of X_μ by the scattering, due to U, of a spin $\uparrow\rightarrow$spin $\downarrow$ excitation with a Fermi sea pair. The corresponding interaction process is described by the amplitude Φ in Eq. (1) and is shown schematically in Fig. 4(a). The Fermi sea pair (μ,α) is created by interacting with the spin-$\downarrow$ electron via U. Such scattering gives a contribution proportional to $U\Sigma_{\alpha\nu}\Phi^{\mathbf{Q}}_{\alpha\nu\mu}$ to $X^{\mathbf{Q}}_\mu$. In Fig. 3(c) we plot this correlation contribution, both for $\mathbf{Q}$ close to the X point and for small $\mathbf{Q}$, as a function of momentum μ for $n=0.6$ where the softening is pronounced. We consider momenta $\mu\|\mathbf{Q}$ (μ_x, the contribution $\Phi^{\|}$) and momenta $\mu\perp\mathbf{Q}$ (μ_y, the contribution $\Phi^{\perp}$). As can be seen in Fig. 3(c), the largest correlation contribution comes for $\mu\|\mathbf{Q}$ close to the Fermi surface (which for the concentrations of interest is close to the zone boundary) and for $\mathbf{Q}$ close to the zone boundary. In Fig. 3(d) we compare the spin-wave energy from the full calculation with the results obtained by neglecting $\Phi^{\perp}$ and/or $\Phi^{\|}$. It is clear that the strong softening of the spin-wave dispersion as compared to the RPA comes from $\Phi^{\|}$, i.e., from the renormalization of X_μ by the scattering of a spin $\uparrow\rightarrow$spin $\downarrow$ excitation with a Fermi sea pair for momenta μ along Γ-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 that the ferromagnetic state is unstable. On the other hand, for small n, the spin-wave energy first turns negative at the (π,π) point (antiferromagnetic correlations). Finally, for larger n, the spin-wave energy turns negative at small momenta first, $D<0$. By identifying the minimum value of J, $J_c(n)$, where $\omega_{\mathbf{Q}}\geqslant 0$ 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 and 20.

We conclude that nonperturbative 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 spin-wave dispersion and the decrease in the stability of the ferromagnetic state. These correlation effects depend sensitively on the on-site 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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