Spin wave dispersion softening in the ferromagnetic Kondo lattice model for manganites

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Spin dynamics is calculated in the ferromagnetic (FM) state of the generalized Kondo lattice model taking into account strong on-site correlations between $e_g$ electrons and antiferromagnetic (AFM) exchange among $t_{2g}$ spins. Our study suggests that competing FM double-exchange and AFM super-exchange interaction lead to a rather nontrivial spin-wave spectrum. While spin excitations have a conventional $Dq^2$ spectrum in the long-wavelength limit, there is a strong deviation from the spin-wave spectrum of the isotropic Heisenberg model close to the zone boundary. The relevance of our results to the experimental data is discussed.

The revival in the study of manganites has lead to experimental re-examination of their different properties and interplay between nontrivial magnetism and highly anisotropic charge transport. One of the puzzling features is the non-universality of the magnetic and transport properties of FM manganites at doping $x \sim 0.3$. According to the conventional theory of the Double Exchange (DE), the spin dynamics of the FM state that evolves at temperatures below the Curie temperature $T_C$ is expected to be of nearest-neighbor Heisenberg type. This picture seems to be indeed reasonably accurate for manganites with high value of $T_C$. Recently, however, several experimental results have shown a strong deviation of the spin-wave dispersion (SWD) from the typical Heisenberg behavior. The unexpected softening of the SWD at the zone boundary has been observed in several manganites with low but very different $T_C$: Pr$_{0.63}$Sr$_{0.37}$MnO$_3$ ($T_C=301$ K), La$_{0.7}$Ca$_{0.3}$MnO$_3$ ($T_C=242$ K), and Nd$_{0.7}$Sr$_{0.3}$MnO$_3$ ($T_C=198$ K). These observations are very important as they indicate that some aspects of spin dynamics in manganites have not been entirely understood yet. During last few years this problem was discussed by several theoreticians. Khalilullin and Kilian in Ref.\textsuperscript{2} proposed a theory of anomalous softening in FM manganites based on the modulation of magnetic exchange bonds by orbital degree of freedom of double-degenerate $e_g$ electrons. They found out that charge and coupled orbital-lattice fluctuations can be considered as the main origin of the softening phenomena. Soloviev and Terakura in Ref.\textsuperscript{2} have argued that the softening of spin wave at the zone boundary and the increase of the spin-wave stiffness constant with doping have purely magnetic origin.

The DE mechanism as an explanation of ferromagnetic order in doped manganites\textsuperscript{2} was proposed shortly after the first experiments in these compounds\textsuperscript{1}. In this model it is assumed that the hopping of $e_g$ electrons between neighboring sites is easier if the local spin on the sites are parallel, so an effective ferromagnetic coupling between the local spins is induced by the conduction electrons lowering their kinetic energy. For quite a long time it was believed that DE model gives full understanding of the FM metallic state in manganites. Later it was shown both from theory and experiment that, in fact, it provides only a qualitative explanations and it is necessary to extend DE model by taking into account the orbital degree of freedom of double-degenerate $e_g$ electrons, phonons, AFM superexchange interaction between localized spins, etc. Particularly, for the Curie temperature the value estimated by pure DE was much larger than the experimentally observed one. Taking into account AFM superexchange interaction among localized spins of $t_{2g}$ electrons gives possibility to diminish the value of $T_C$ in the framework of DE model\textsuperscript{2}. Recently, a Monte Carlo study of the Ferromagnetic Kondo Lattice Model (FKLM) for doped manganites (see, Ref.\textsuperscript{3}) confirmed the scaling of the $T_C$ suppression with a bandwidth narrowing induced by the antiferromagnetic frustration. In our recent paper\textsuperscript{4} we have also shown that AFM superexchange interaction plays very important role in determining the phase diagram in manganites.

The aim of our work is to show that the competition between AFM superexchange interaction among localized spins and double-exchange interaction among itinerant electrons is one of the mechanisms responsible for the spin wave softening phenomena.

In the present paper we continue to study the spin dynamics within the generalized one-orbital FKLM\textsuperscript{4}. The effective Hamiltonian of FKLM model\textsuperscript{4} can be written as

\[
H = \sum_{ij} (t_{ij} - \mu \delta_{ij}) c^\dagger(i)c(j) + U \sum_i n_{\uparrow}(i)n_{\downarrow}(i) - J_H \sum_i S(i)s(i) + 1/2 \sum_{ij} J_{ij} s(i)s(j)
\]

where $c(i)$ and $c^\dagger(i)$ are annihilation and creation operators for electrons in the spinor notation at site $R(i)$:

\[
c(i) = \begin{pmatrix} c_{\uparrow}(i) \\ c_{\downarrow}(i) \end{pmatrix}
\]
μ is the chemical potential; \( n_\sigma(i) = c_\sigma^\dagger(i)c_\sigma(i) \) is the density operator of electrons with spin \( \sigma \); the spin operator is given by \( s_\sigma(i) = \frac{1}{2}c_\sigma^\dagger(i)s_\sigma c_\sigma(i) \), \( s_\sigma(k = 1, 2, 3) \) are the Pauli matrices; \( S(i) \) is the localized Mn core spin of \( t_{ij} \) electrons with \( S = 3/2 \); \( t_{ij} \) is the nearest-neighbor hopping parameter; \( J_H \) is the FM Hund coupling interaction among the localized and delocalized spin subsystems, \( J_{ij} \) is the nearest-neighbor antiferromagnetic superexchange interaction among localized spins.

The magnetic properties of the above given Hamiltonian has a dual character: there exists a purely Heisenberg-like contribution from the exchange interaction among localized spins and the itinerant contribution driven by electronic excitations. The itinerant contribution leads to the effective FM exchange interaction between localized spins due to the DE mechanism. To treat appropriately this dual character of the magnetism one should describe both contributions on the equal level of approximation as well as retain SU(2) spin symmetry of the Hamiltonian. To this end, we employ the Composite operator method (COM) approach by introducing a new set of operators which well describe collective excitations in the system and are constructed from the original electron operators. These composite operators can have fermionic or bosonic character. They are created by interactions among the electrons and the localized spins, and, therefore, their properties will be determined by the dynamics and boundary conditions and must be computed self-consistently.

First we start by discussing the electronic excitations of the system, responsible for the itinerant contribution to spin dynamics. We introduce the 4-component fermionic basis:

\[
\psi(i) = \begin{pmatrix} \xi_\uparrow(i) \\ \eta_\uparrow(i) \\ \xi_\downarrow(i) \\ \eta_\downarrow(i) \end{pmatrix},
\]

where \( \xi_\sigma(i) \) and \( \eta_\sigma(i) \) are Hubbard composite excitations:

\[
\begin{align*}
\xi_\sigma(i) &= c_\sigma(i)(1 - n_\sigma(i)) \\
\eta_\sigma(i) &= c_\sigma(i)n_\sigma(i)
\end{align*}
\]

The \( \eta \) excitation describes an electron restricted to move on sites already occupied with an electron of opposite spin whereas \( \xi \) demands that there be no prior occupancy on the site.

Let us consider the equation of motion for the field \( j(i) \):

\[
i\frac{\partial}{\partial t} j(i) = [\psi(i), H],
\]

where \( j(i) \) is the current operator. By considering the Hamiltonian given by \( (\ref{eq:hamiltonian}) \), we obtain the following expression for the current operator:

\[
j(i) = \begin{pmatrix} -\mu \xi_\uparrow(i) - 6t \xi_\uparrow(i) - 6t \pi_\uparrow(i) - J_H/2 [\xi_\uparrow(i)S^-\downarrow(i) + \xi_\downarrow(i)S^+\uparrow(i)] \\
-(\mu - U) \eta_\uparrow(i) + 6t \pi_\uparrow(i) - J_H/2 [\eta_\uparrow(i)S^-\downarrow(i) + \eta_\downarrow(i)S^+\uparrow(i)] \\
-\mu \xi_\downarrow(i) - 6t \xi_\downarrow(i) - 6t \pi_\downarrow(i) - J_H/2 [\xi_\downarrow(i)S^-\uparrow(i) - \xi_\uparrow(i)S^+\downarrow(i)] \\
-(\mu - U) \eta_\downarrow(i) + 6t \pi_\downarrow(i) - J_H/2 [\eta_\downarrow(i)S^-\uparrow(i) - \eta_\uparrow(i)S^+\downarrow(i)] \end{pmatrix},
\]

where \( \pi(i) = \frac{1}{2} \sigma^\mu n_\mu(i)c^\sigma(i) + c(i)c^\sigma(i) \) is a higher order composite field; \( n_\mu(i) = c^\dagger(i)\sigma_\mu c(i) \) - the number \( (\mu = 0) \) and spin \( (\mu = 1, 2, 3) \) density operator with the notation \( \sigma_\mu = (1, \sigma) \), \( \sigma^\mu = (-1, \sigma) \), \( \sigma \) being the Pauli matrices; and the notation \( c^\sigma(i) = \sum_j \alpha(i,j)c(j) \) stands to indicate the field \( c \) on the first neighbor sites with \( F.T.\alpha(i,j) = \frac{1}{4}(\cos k_x + \cos k_y + \cos k_z) \) (abbreviation \( F.T. \) stands for Fourier transform).

Formally, we can write the \( n \)th element of the current as

\[
\begin{equation}
\begin{split}
\epsilon_{nm}(i,j) = \sum_j \sum_m \epsilon_{nm}(i,j)\psi_m(j) + \delta j_n(i) \end{split}
\end{equation}
\]

By projecting out the correction part \( \delta j_n(i) \), we linearize the Heisenberg equation of motion as

\[
i\frac{\partial}{\partial t} \psi_n(i) = \sum_j \epsilon_{nm}(i,j)\psi_m(j),
\]

where the energy matrix \( \epsilon(i, j) \) is calculated by means of the equation

\[
\epsilon(i, j) = \sum_i m(i, l)I^{-1}(l, j).
\]
where \( n \equiv < n > \) is the particle density and \( < s_z > = 1/2 < n_3 > \). The \( m \) matrix is hermitian and the elements different from zero are given by

\[
\begin{align*}
m_{11} &= -\mu I_{11} - 6t \Delta_1 - 6t \alpha(k)(I_{11} - I_{22} + p_t) - J_H(a + b_2), \\
m_{12} &= 6t \Delta_1 + 6t \alpha(k)(I_{22} + p_t), \\
m_{22} &= -(\mu - U)I_{22} - 6t \Delta_1 - 6t \alpha(k)p_1 + J_H b_2, \\
m_{33} &= -\mu I_{33} - 6t \Delta_1 - 6t \alpha(k)(I_{33} - I_{44} + p_1) - J_H(-a + b_1), \\
m_{34} &= 6t \Delta_1 + 6t \alpha(k)(I_{44} + p_1), \\
m_{44} &= -(\mu - U)I_{44} - 6t \Delta_1 - 6t \alpha(k)p_1 + J_H b_1,
\end{align*}
\]

(10)

where \( \sigma = \uparrow (1) \) or \( \sigma = \downarrow (2) \). Matrices \( m(k) \) and \( I \) involve correlation functions which we defined as

\[
\begin{align*}
\Delta_\sigma &= \langle \xi_{\sigma}^+(i)\xi_{\sigma}^-(i) \rangle - \langle n_{\sigma}^+ (i)n_{\sigma}^- (i) \rangle, \\
p_{\sigma} &= \frac{1}{2}[\langle n_{\sigma}^+ (i)n_{\sigma}^- (i) \rangle + 2(1-\sigma)n_{\sigma}^0 (i)n_3 (i)] - \langle c_{\sigma i}^+ c_{\sigma j}^- + c_{\sigma j}^+ c_{\sigma i}^-(i) \rangle^a, \\
a &= 1/2 < S_z >, \\
b_\sigma &= 1/2(< s^- (i)S^+(i) > - (1-\sigma)n_\sigma(i)S^z(i)).
\end{align*}
\]

The parameters \( \Delta_\sigma \) and \( p_{\sigma} \) are static inter-site correlation functions which describe, respectively, a constant shift of the bands and a bandwidth renormalization in itinerant subsystem.

The retarded Green function \( (GF) \) can be written as

\[
S^F(i, j) = \langle R(\psi(i), \psi^+(j)) =
\int \frac{d\Omega}{(2\pi)^4} \int d^3k \int \omega \exp^{ik(r_i - r_j) - i\omega(t_i - t_j)} S^F(k, \omega), \tag{12}
\]

where the \( k \)-integration is over the Brillouin zone (BZ) and \( \Omega \) is the inverse volume of the BZ. By means of the linearized Heisenberg equation (7) the Green’s function has the following expression:

\[
S^F(k, \omega) = \frac{1}{\omega - m(k)I^{-1}(k)} I(k).
\]

or, in the spectral form:

\[
S^F(k, \omega) = \sum_{i=1}^{4} \frac{\sigma^i(k)}{\omega - E_i(k) + i\delta}, \tag{13}
\]

where the energy spectra \( E_i(k) \) and the spectral functions \( \sigma^i(k) \) can be easily evaluated. The energy spectra \( E_i(k) \) are the eigenvalues of the matrix \( \omega(k) \). Since they depend on a set of external parameters such as electron density \( n \), temperature \( T \), Coulomb interaction \( U \), Hund coupling \( J_H \) and AFM Heisenberg exchange interaction \( J \), and a set of internal parameters \( (\mu, m, \Delta_\sigma, p_\sigma, a, b_1, b_2) \), they must be calculated in a self-consistent way. Internal parameters are expressed as expectation values of composite fields. If these composite fields belong to the fermionic or bosonic basis they can be expressed in terms of the corresponding GF. However, it may happen that some of the parameters are expressed as expectation values of higher-order composite fields that do not belong to the basic set and we need to evaluate by imposing some symmetry requirements, as it will be discussed later.

As we have already mentioned the spin dynamics of the present model is governed by both localized and itinerant spin subsystems. To treat these two contributions at the same level of the approximation we consider a bosonic sector by defining a basis of composite fields as

\[
B(i) = \begin{pmatrix}
s^+(i) \\
S^+(i)
\end{pmatrix}, \tag{14}
\]

The procedure then is similar to the fermionic sector; the propagator for the boson field \( B(i) \) will contain another set of parameters which must be self-consistently determined. After some algebra the following expressions of the bosonic normalization matrix

\[
\tilde{I}(k) = F.T. \langle [B(i), B^\dagger(j)]_{E.T.} >
\]

and the frequency matrix which is given by the commutator of the current and the bosonic fields

\[
\tilde{m}(k) = F.T. \langle [i\partial_{\tau_i} B(i), B^\dagger(j)]_{E.T.} >
\]

can be easily derived:

\[
\begin{align*}
\tilde{I}_{11} &= 2 < s_z >, \\
\tilde{I}_{12} &= \tilde{I}_{21} = 0, \\
\tilde{I}_{22} &= 2 < S_z >; \\
\tilde{m}_{11} &= J_H d + 6tw(1 - \alpha(k)), \\
\tilde{m}_{12} &= \tilde{m}_{21} = -J_H d, \\
\tilde{m}_{22} &= J_H d - 6f(1 - \alpha(k)),
\end{align*}
\]

(15)\(\tag{16}\)

with coefficients defined as

\[
\begin{align*}
w &= \langle c^i(\tau)\sigma^i(\tau) >, \\
d &= 2 < s_z(i)S_z(i) > + < s^+(i)S^-(i) >, \\
f &= 2 < S_\sigma^0(i)S_z(i) > + < (S^+)\sigma S^-(i) >.
\end{align*}
\]

It is important to emphasize that \( w \) is the kinetic energy and, hence, the effective FM exchange scales with the kinetic energy.

The retarded Green’s function for the boson field \( B(i) \) can be expressed as \( S^B(i, j) = \langle R[B(i)B^\dagger(j)] \rangle \). In the polar approximation the bosonic GF can be written as

\[
S^B(k, \omega) = \sum_{i=1}^{2} \frac{\tilde{\sigma}^i(k)}{\omega - \tilde{E}_i(k) + i\delta}, \tag{18}
\]

where \( \tilde{\sigma}^i(k) \) and \( \tilde{E}_i(k) \) are bosonic spectral functions and the energy spectra, respectively. \( \tilde{E}_i(k) \) are two branches
of spin excitations: acoustic and optical modes. In the long-wavelength limit the acoustic mode is \( E_1(k) \sim Dk^2 \), and the spin stiffness scales as \( D \sim (ztw - J) \) which clearly shows the competitions of the effective, induced by the electron hopping, FM exchange \( J_{FM} \) and AFM superexchange \( J \).

There are new internal parameters appearing in the bosonic sector. From the analysis given above, the bosonic and fermionic GF depend on 12 internal parameters:

\[
< s_z >, \quad < S_z >, \quad < s^+(i)S^-(i) >, \quad \mu, \quad \Delta, \quad p_r, \quad w, \quad f, \quad < n_\sigma(i)S_z(i) >.
\]  

(19)

Now we need to construct a closed self-consistent scheme for fixing these parameters. The static correlation functions (CF) \( C_{lm}(i,j) \) can be computed from the corresponding retarded GF by the fluctuation-disipation theorem:

\[
C^{F(B)}(i,j) = \begin{pmatrix}
\langle \psi(i)\psi^+(j) \rangle \\
\langle B(i)B^+(j) \rangle
\end{pmatrix} = \frac{\Omega}{(2\pi)^4} \int d^3k \int d\omega e^{i(k_\parallel r_\perp t_\perp - i\omega t_\parallel t_\perp)} C(k,\omega),
\]

where

\[
C^{F,B}(k,\omega) = \begin{pmatrix}
1 + \tanh(\omega/2T) & \text{Im}S^F(k,\omega) \\
1 + \coth(\omega/2T) & \text{Im}S^B(k,\omega)
\end{pmatrix}.
\]

(21)

For convenience, we will use the following notation for the static CF: \( C_{lm} = C_{lm}(i,i) \), \( C_{lm}^\alpha = C_{lm}(i,i^\alpha) \).

It should be pointed out \([13]\) that one can use Eq. (21) for calculation of correlation functions \( C(k,\omega) \) in terms of the commutator bosonic GF only for ergodic systems when there is no \( \delta(\omega) \) term in the spectral density. Otherwise one has to use the anticommutator or causal GF for bosonic operators to take into account nonergodic terms. In our case of FM ordered system with \( < S_z \neq 0 > \) and \( < s_z \neq 0 > \) the bosonic GF \([13]\) describes the one-particle, spin-wave-like excitations which are ergodic for all \( q \neq 0 \).

The first self-consistent equation is given by the definition of the magnetization per site:

\[
m = \frac{1}{2}(n_+ - n_-) = \frac{1}{2}[C_{11}^F - C_{22}^F].
\]

(22)

The boundary condition which fixes the particle number as an external parameter gives us the equation for fixing the chemical potential \( \mu \):

\[
n = 2 - C_{11}^F - C_{22}^F - C_{33}^F - C_{44}^F.
\]

(23)

The other three parameters are expressed through the matrix elements of the CF \([11]\) as follows:

\[
\Delta_\uparrow = C_{11}^{\sigma \alpha} - C_{22}^{\sigma \alpha} \\
\Delta_\downarrow = C_{33}^{\sigma \alpha} - C_{44}^{\sigma \alpha} \\
< s^+(i)S^-(i) > = C_{12}^B.
\]

(24)

The kinetic energy \( w \) can also be expressed through the linear combination of GF matrix elements:

\[
w = -\left( \sum_{n,m=1}^2 + \sum_{n,m=3}^4 \right) C_{nm}^{F}. \]  

(25)

The other parameters are not strictly bound to the dynamics and they must be evaluated by other means. This aspect is a general property of the Green’s function formalism \([18]\). The equations of motion are not sufficient to completely determine the Green’s functions which refer to a specific choice of the Hilbert space. The information concerning the representation must be supplied. As discussed in Refs. \([17]\) and \([18]\), this information is supplied by determining the parameters not bounded by the dynamics in such a way that the Hilbert space has the right properties to conserve the relations among matrix elements imposed by symmetry conditions. In particular, the main attention should be paid to the fact that the chosen representation does not violate the symmetry required by Pauli principle. Let us note that by Pauli principle we mean all relations among considered operators dictated by their algebra. The Pauli principle for the electron propagator requires that

\[
C_{12}^F = 0, \quad C_{34}^F = 0, \quad C_{11}^F = C_{33}^F.
\]

(26)

There is one more self-consistent equation which connects matrix elements of fermionic and boson GF:

\[
C_{11}^B = C_{44}^F.
\]

(27)

To close our self-consistent scheme, we have to do some approximation and we use the following decoupling scheme:

\[
< n_\sigma(i)S_z(i) > \gg < n_\sigma(i) > < S_z(i) >.
\]

(28)

Having solved the system of the self-consistent equations we can study various properties of the FKLM. In this paper we will look at the spin wave spectrum. The study of other properties will be published elsewhere.

In Fig. 1 we plot the dispersion of magnetic excitations along various direction in the Brillouin zone, and we compare it with nearest-neighbor Heisenberg form with the spin stiffness \( D = \lim_{q \to 0} E(q)/q^2 \), where \( E(q) \) is the acoustic mode of the bosonic spectrum calculated in the mean field approximation (see, formula (24) in Ref. \([13]\). We note that in this approach the acoustic spin mode already occurs in the mean-field approximation as the result of the full rotational symmetry of the spin system.
At small momenta the spectrum exhibits conventional Heisenberg behavior. With increasing momentum the spectrum shows the softening which has the maximum value close to the \((\pi,\pi,\pi)\) AFM vector. However, in experiment the maximum softening was observed in \((\pi,0,0)\) and \((\pi,\pi,0)\) directions. In order to reproduce the experimental data it is thus important to consider orbital degree of freedom as it was considered by Khaliullin and Kilian (see Ref. \[5\]). How strong the orbital fluctuations modulate exchange bonds between Mn ions depends on their characteristic time scale. If the typical frequency of orbital fluctuations is higher than the one of spin fluctuations, the magnon spectrum is renormalized only due to AFM fluctuations and then the orbital state affects the spin dynamics only by restoring the cubic symmetry of exchange bonds. If orbitals fluctuate slower then spins then the anisotropy imposed upon the magnetic exchange bonds by the orbital degree of freedom forms spin dynamics. The presence of Jahn-Teller distortions quenches the dynamics of orbitals and can lead to the long range orbitally ordered state which corresponds to the minimal frequency of orbital fluctuations. Then both orbital fluctuations and the competition between the double-exchange interaction and the AFM superexchange interaction are responsible for the strong renormalization of the magnetic excitation spectrum.

In \(\text{Pr}_0.63\text{Sr}_{0.37}\text{MnO}_3\) there is a tendency to the planar \((x^2-y^2)\) ferrotype orbital ordering. That means that orbital fluctuations mostly renormalize exchange bonds in two dimensions. Let us note that magnons in \((\pi,\pi,\pi)\) direction are sensible to all three spatial directions of the exchange bonds and thus, their dispersion remains unaffected by orbital fluctuations and softening of the spectrum in this direction is purely due to the AFM spin fluctuations. In our approach we attribute softening phenomena to the effect of the suppression of the FM ordering only by the AFM superexchange interaction and thus on the direction \((\pi,\pi,\pi)\) we estimate ordering correctly. In other direction we have to add influence of orbital degree of freedom to fit experimental results.

In summary, we have shown, that the spin wave spectrum obtained by the COM analysis of FKLM shows the softening close to the magnetic zone boundary. In our approach, we have accounted for both the intersite AFM exchange among localized \(t_{2g}\)-electron spins and the strong intra-atomic Hund coupling among the \(t_{2g}\) and \(e_g\) electrons and proved that the competition between these two interactions can lead to the strong renormalization of the magnon spectrum and suppression of the FM ordering.

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