Theory of spin waves in a ferromagnetic Kondo lattice model

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A random-phase approximation theory for spin waves in a ferromagnetic Kondo lattice model is presented. In the strong-coupling limit, this theory agrees with the existing theoretical results, in which the magnetic coupling is taken to be infinite *a priori*. It is shown explicitly that, in the strong-coupling limit, the spin-wave spectrum of the system is identical to that of a short-range Heisenberg model for the case of a single conduction band with short-range hopping integrals. In the intermediate coupling regime, on the other hand, more complicated effects, such as softening or disappearance of spin waves, are likely to be observed. [S0163-1829(98)04414-2]

This paper presents a random-phase approximation (RPA) theory of spin waves for the following ferromagnetic Kondo lattice model (FKLM) with the Hamiltonian,

$$H=-\sum_{\langle i,j\rangle,\mu}(t_{i,j}C^{\dagger}_{i,\mu}C_{j,\mu}+\text{H.c.})-J\sum_i c^{\dagger}_{i,\mu}(\tfrac{1}{2}\tau)_{\mu\nu}c_{i,\nu}\cdot\mathbf{S}_i\,, \tag{1}$$

where $c^{\dagger}/c$'s are the fermion creation/annihilation operators, i,j are site indices, μ, ν are spin indices, τ is the pauli matrix, and S_i's are the local quantum spins. The magnetic coupling constant J is positive. The $t_{i,j}$'s are the hopping integrals. Both the infinite J limit and the intermediate J regime are considered here.

It is generally believed that the FKLM model can account for the low-temperature magnetic properties of $La_{1-x}M_xMnO_3$ where M stands for a divalent ion and x is around 0.3. In this model the local $S=\frac{3}{2}$ quantum spins are formed by the localized t_{2g} electrons due to the strong Hund's rule coupling. The remaining $1-x$ (per Mn) e_g electrons have a finite hopping integral between Mn sites and couple ferromagnetically to the local spins. Because of the large Hund's rule coupling between the d electrons of Mn, J is estimated of the order of 1 eV. Most theoretical treatments of spin waves in FKLM so far have concentrated on the infinite J limit,[1,2] in which an effective single spin Hamiltonian (or action) is the starting point. In real systems the hopping integrals $|t|$ may be as large as 0.5 eV, which puts the infinite J approximation in question. Recent neutron-scattering experiments[3,4] have measured the spin-wave spectra in various manganites. While for high Curie temperature samples, Heisenberg ferromagnet behavior was observed,[3] in some lower Curie temperature samples, softening or disappearance of higher-energy spin waves were observed.[4] These considerations call for a more careful investigation of FKLM, particularly in the intermediate coupling regime, which is the purpose of this paper.

The rest of the paper is organized as follows. First, an RPA theory of spin waves for the FKLM is presented. The long-wavelength limit is then taken and an analytical expression for the spin-wave stiffness is obtained. In the strong-coupling limit, it is shown that the present theory agrees with the existing results. It is also explicitly shown that in this limit, for a single conduction band with short-range hopping integrals, the spin-wave spectrum is exactly that of a Heisenberg ferromagnet with short-range couplings. Then the intermediate coupling regime is discussed. A condition for the stability of ferromagnetic ground state assumed in the RPA treatment is presented. It is shown by numerical examples that in the intermediate coupling regime, both spin-wave softening and disappearance may happen. Finally, the effect of direct antiferromagnetic couplings between local spins due to superexchange is discussed.

The spin-wave spectrum is given by the poles of the following correlation function (for a reference on double time Green functions, see Ref. 5):

$$\begin{aligned}\chi^{+-}(\mathbf{q},\omega)&=\langle S^{+}(-\mathbf{q})|S^{-}(\mathbf{q})\rangle_{\omega}\\&\equiv\int dt(-i)\theta(t)\langle[S^{+}(-\mathbf{q},t),S^{-}(\mathbf{q},0)]_{-}\rangle\\&\quad\times\exp[i(\omega+i0^{+})t],\end{aligned} \tag{2}$$

where $\hbar=1$, $\theta(t)$ is the step function,

$$\langle\hat{O}(t)\rangle\equiv\frac{\text{Tr}[\exp(-H/T)\hat{O}(t)\rangle]}{\text{Tr}[\exp(-H/T)]}, \tag{3}$$

and

$$\mathbf{S}(\mathbf{q})\equiv\frac{1}{\sqrt{N}}\sum_i\mathbf{S}_i\exp(-i\mathbf{q}\cdot\mathbf{R}_i). \tag{4}$$

Applying the following equation of motion,

$$\begin{aligned}\omega\langle S^{+}(-\mathbf{q})|S^{-}(\mathbf{q})\rangle_{\omega}&=\langle[S^{+}(-\mathbf{q}),S^{-}(\mathbf{q})]_{-}\rangle\\&\quad-\langle[H,S^{+}(-\mathbf{q})]_{-}|S^{-}(\mathbf{q})\rangle_{\omega},\end{aligned} \tag{5}$$

we have

$$\begin{aligned}&\omega\langle S^{+}(-\mathbf{q})|S^{-}(\mathbf{q})\rangle_{\omega}\\&\quad=2\langle S^{z}\rangle-\frac{J}{\sqrt{N}}\sum_{\mathbf{q}'}\langle\hat{s}^{+}(-\mathbf{q}')S^{z}(\mathbf{q}'-\mathbf{q})|S^{-}(\mathbf{q})\rangle_{\omega}\end{aligned}$$

$$+\frac{J}{\sqrt{N}}\sum_{\mathbf{q}'}\langle\hat{s}^z(-\mathbf{q}')S^+(\mathbf{q}'-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega, \qquad (6)$$

where

$$\hat{s}(\mathbf{q})\equiv\frac{1}{\sqrt{N}}\sum_i \hat{s}_i\exp(-i\mathbf{q}\cdot\mathbf{R}_i)$$

$$=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}} c^\dagger_{\mathbf{k}-\mathbf{q},\mu}(\tfrac{1}{2}\tau)_{\mu,\nu}c_{\mathbf{k},\nu}. \qquad (7)$$

We then use the following RPA decoupling scheme

$$\langle\hat{s}^+(\mathbf{q}')S^z(\mathbf{q}'')|S^-(\mathbf{q})\rangle_\omega=\langle S^z(\mathbf{q}'')\rangle\langle\hat{s}^+(\mathbf{q}')|S^-(\mathbf{q})\rangle_\omega,$$

$$\langle\hat{s}^z(\mathbf{q}')S^+(\mathbf{q}'')|S^-(\mathbf{q})\rangle_\omega=\langle\hat{s}^z(\mathbf{q}')\rangle\langle S^+(\mathbf{q}'')|S^-(\mathbf{q})\rangle_\omega, \qquad (8)$$

to get

$$\omega\langle S^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega=2\langle S^z\rangle-J\langle S^z\rangle\langle\hat{s}^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega$$
$$+J\langle\hat{s}^z\rangle\langle S^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega. \qquad (9)$$

Similarly, we have

$$\omega\langle c^\dagger_{\mathbf{k}+\mathbf{q},\uparrow}c_{\mathbf{k},\downarrow}|S^-(\mathbf{q})\rangle_\omega=(\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}+\mathbf{q}})\langle c^\dagger_{\mathbf{k}+\mathbf{q},\uparrow}c_{\mathbf{k},\downarrow}|S^-(\mathbf{q})\rangle_\omega$$
$$+\frac{J(\langle\hat{n}_{\mathbf{k},\downarrow}\rangle-\langle\hat{n}_{\mathbf{k}+\mathbf{q},\uparrow}\rangle)}{2\sqrt{N}}$$
$$\times\langle S^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega+J\langle S^z\rangle$$
$$\times\langle c^\dagger_{\mathbf{k}+\mathbf{q},\uparrow}c_{\mathbf{k},\downarrow}|S^-(\mathbf{q})\rangle_\omega, \qquad (10)$$

from which we get

$$\langle\hat{s}^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega=\frac{J}{2N}\sum_{\mathbf{k}}\frac{\langle\hat{n}_{\mathbf{k},\downarrow}\rangle-\langle\hat{n}_{\mathbf{k}+\mathbf{q},\uparrow}\rangle}{\omega-J\langle S^z\rangle-(\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}+\mathbf{q}})}$$
$$\times\langle S^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega. \qquad (11)$$

Solving Eqs. (9) and (11), we obtain the main equation of this paper,

$$\left(\omega-J\langle\hat{s}^z\rangle+\frac{J^2\langle S^z\rangle}{2}\chi_0^{+-}(\mathbf{q},\omega)\right)\langle S^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega$$
$$=2\langle S^z\rangle, \qquad (12)$$

where

$$\chi_0^{+-}(\mathbf{q},\omega)\equiv\frac{1}{N}\sum_{\mathbf{k}}\frac{\langle\hat{n}_{\mathbf{k},\downarrow}\rangle-\langle\hat{n}_{\mathbf{k}+\mathbf{q},\uparrow}\rangle}{\omega-J\langle S^z\rangle-(\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}+\mathbf{q}})}. \qquad (13)$$

Thus the spin-wave dispersion is given by the solutions of the following equation:

$$-(\omega-J\langle\hat{s}^z\rangle)=\frac{J^2\langle S^z\rangle}{2}\chi_0^{+-}(\mathbf{q},\omega). \qquad (14)$$

In the long-wavelength limit, $|\mathbf{q}|\to 0$, expand χ_0 to the order of q^2, we have

$$\chi_0^{+-}(q\to 0)\approx\frac{2\langle\hat{s}^z\rangle}{J\langle S^z\rangle}\left(1+\frac{\omega}{J\langle S^z\rangle}\right)-\frac{1}{2J^2\langle S^z\rangle^2}$$
$$\times\left(\frac{1}{N}\sum_{\mathbf{k}}(\langle\hat{n}_{\mathbf{k}\uparrow}\rangle+\langle\hat{n}_{\mathbf{k}\downarrow}\rangle)(\mathbf{q}\cdot\nabla)^2\epsilon_{\mathbf{k}}\right)$$
$$+\frac{1}{2J^3\langle S^z\rangle^3}\left(\frac{1}{N}\sum_{\mathbf{k}}(\langle\hat{n}_{\mathbf{k}\uparrow}\rangle\right.$$
$$\left.-\langle\hat{n}_{\mathbf{k}\downarrow}\rangle)(\mathbf{q}\cdot\nabla\epsilon_{\mathbf{k}})^2\right). \qquad (15)$$

Thus the spin-wave stiffness is

$$D(\hat{\mathbf{q}})=S_{\rm tot}^{-1}\left(\frac{1}{4N}\sum_{\mathbf{k}}(\langle\hat{n}_{\mathbf{k}\uparrow}\rangle+\langle\hat{n}_{\mathbf{k}\downarrow}\rangle)(\hat{\mathbf{q}}\cdot\nabla)^2\epsilon_{\mathbf{k}}\right.$$
$$\left.-\frac{1}{4NJ\langle S^z\rangle}\sum_{\mathbf{k}}(\langle\hat{n}_{\mathbf{k}\uparrow}\rangle-\langle\hat{n}_{\mathbf{k}\downarrow}\rangle)(\hat{\mathbf{q}}\cdot\nabla\epsilon_{\mathbf{k}})^2\right), \qquad (16)$$

where $S_{\rm tot}=\langle S^z\rangle+\langle\hat{s}^z\rangle$.

In the strong-coupling limit, the majority-spin and minority-spin bands do not overlap with each other, and with the filling ratio no larger than 1, we have a half-metallic system, i.e., $\langle\hat{n}_{\mathbf{k}\downarrow}\rangle=0$. In the strong-coupling limit, we also have $\epsilon_{\mathbf{k}}/J\langle S^z\rangle\to 0$, and $\langle\hat{s}^z\rangle=1/2N\Sigma_{\mathbf{k}}\langle\hat{n}_{\mathbf{k}\uparrow}\rangle=(1-x)/2$, therefore from Eq. (16) we get

$$D(\hat{\mathbf{q}})_{J\to\infty}=\left(\langle S^z\rangle+\frac{1-x}{2}\right)^{-1}\left(\frac{1}{4N}\sum_{\mathbf{k}}\langle\hat{n}_{\mathbf{k}\uparrow}\rangle(\hat{\mathbf{q}}\cdot\nabla)^2\epsilon_{\mathbf{k}}\right), \qquad (17)$$

where x is the doping concentration. Equation (16) is in agreement with the results of Refs. 1 and 2

On the other hand, we can also take the infinite J limit to get an expression for the spin-wave spectrum over the whole Brillouin zone from Eq. (14). Note that in this limit, Eq. (13) gives

$$\chi_0^{+-}(J\to\infty)\approx\frac{1}{J\langle S^z\rangle}\frac{1}{N}\sum_{\mathbf{k}}\langle\hat{n}_{\mathbf{k},\uparrow}\rangle\left(1+\frac{\omega+\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}-\mathbf{q}}}{J\langle S^z\rangle}\right), \qquad (18)$$

thus we have for the spin-wave spectrum,

$$\omega(\mathbf{q})_{J\to\infty}=S_{\rm tot}^{-1}\frac{1}{2N}\sum_{\mathbf{k}}(\epsilon_{\mathbf{k}-\mathbf{q}}-\epsilon_{\mathbf{k}})\langle\hat{n}_{\mathbf{k}\uparrow}\rangle, \qquad (19)$$

which agrees with Ref. 1 that used a different approach. Taking the limit $q\to 0$, we again obtain Eq. (17).

For a single band tight-binding model with short-range hopping integrals, we have

$$\epsilon_{\mathbf{k}}=-2\sum_{\Delta}t_\Delta\cos(\mathbf{k}\cdot\Delta), \qquad (20)$$

where Δ's are the neighboring site indices and t_Δ's are the corresponding hopping integrals. Hence,

$$\omega(\mathbf{q})_{J\to\infty}=S_{\rm tot}^{-1}\frac{1}{N}\sum_{\mathbf{k}}\langle\hat{n}_{\mathbf{k}\uparrow}\rangle\sum_{\Delta}t_\Delta\cos(\mathbf{k}\cdot\Delta)$$
$$\times[1-\cos(\mathbf{q}\cdot\Delta)], \qquad (21)$$

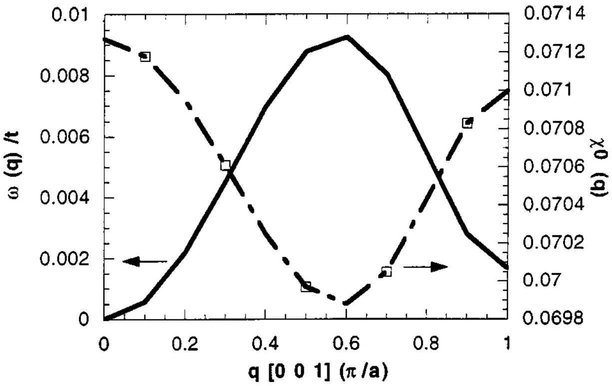

FIG. 1. Spin-wave dispersion (solid line) and bare susceptibility (dashed line) for a simple cubic FKLM with nearest-neighbor hopping, $E_f=-3.5t$, and $J=4.8t$.

where the sum of an odd function, $\Sigma_{\mathbf{k}}\langle \hat{n}_{\mathbf{k}\uparrow}\rangle \sin(\mathbf{k}\cdot\Delta)=0$, has been used. This is exactly the spin-wave spectrum of a Heisenberg ferromagnet with

$$J_{\Delta,0}=\frac{1}{2NS_{\text{tot}}}\sum_{\mathbf{k}}\langle \hat{n}_{\mathbf{k}\uparrow}\rangle t_\Delta \cos(\mathbf{k}\cdot\Delta).$$

This result is consistent with the observation that in higher Curie temperature samples, the spin-wave spectrum is that of a nearest-neighbor Heisenberg model.[3] The same conclusion was also reached by Furukawa[6] recently. We note in passing that at half filling ($\langle \hat{n}_{\mathbf{k},\uparrow}\rangle=1$, $\langle \hat{n}_{\mathbf{k},\downarrow}\rangle=0$), even in the limit of $J\rightarrow\infty$, the ferromagnetic ground state is not stable, which is evident from Eq. (16) since the first term in Eq. (16) vanishes.

We now turn our attention to the intermediate coupling regime. First we present a necessary condition for the stability of the assumed ferromagnetic ground state that is the basis for our RPA treatment. The stability of the ferromagnetic ground state requires that

$$\chi^{+-}(\mathbf{q}\neq 0,\omega=0)>0. \tag{22}$$

This in turn implies [cf. Eq. (11)]

$$\chi_0^{+-}(\mathbf{q}\neq 0,\omega=0)<\chi_0^{+-}(0,0)=\frac{2\langle \hat{s}^z\rangle}{J\langle S^z\rangle}. \tag{23}$$

In all our numerical results below, the stability condition has been checked and was found to hold.

We have shown that if the conduction electrons can be described by a single band tight-binding model, the corresponding FKLM in the strong-coupling limit has the same spin-wave spectrum as that of a Heisenberg model with short-range couplings. It is interesting to see if, in the intermediate coupling regime for the same single band model, a more complicated spin-wave spectrum than that of the simple Heisenberg model type may occur. We have found that, indeed, both spin-wave softening and the dispearance of spin waves into the Stonner continuum may occur in the intermediate coupling regime.

Figure 1 shows the spin-wave dispersion solved from Eq. (14) along [001] for a simple cubic model with nearest-neighbor hopping, $E_f=-3.5t$ ($n\approx 0.5$), and $J=4.8t$. The softening at the zone boundary is evident. We found that this softening is closely related to the peak around the [001] of χ_0^{+-} (also shown in Fig. 1). This is to be expected by inspecting Eq. (14). Since χ_0^{+-} is very much band-structure dependent, we have not tried to fit the neutron-scattering results of Hwang *et al.*[4]

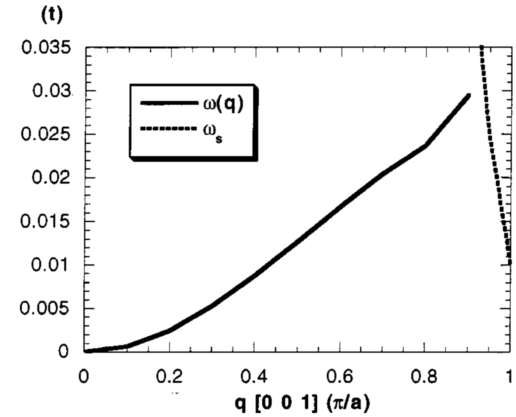

FIG. 2. Spin-wave dispersion (solid line) and Stoner threshold (dotted line) for a simple cubic FKLM with nearest-neighbor hopping, $E_f=-4.005t$, and $J=4.0t$.

For a different set of parameters ($E_f=-4.005t$ and $J=4t$), Fig. 2 shows how the spin waves merge into the Stonner continuum, which is a common feature in many metallic ferromagnets. In this case, a characteristic threshold energy ω_s is defined beyond which the imaginary part of χ_0^{+-} becomes finite.

Finally, we would like to comment on the role of possible superexchange (direct) antiferromagnetic coupling between the local spins. The Hamiltonian, in this case, is

$$H=-\sum_{\langle i,j\rangle,\mu}(t_{i,j}c_{i,\mu}^\dagger c_{j,\mu}+\text{H.c.})-J\sum_i c_{i,\mu}^\dagger(\tfrac{1}{2}\tau)_{\mu\nu}c_{i,\nu}\cdot\mathbf{S}_i+J'\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j, \tag{24}$$

where $J'>0$. It is straightforward to generalize Eq. (12) to

$$\left(\omega+2J'\lambda(\mathbf{q})-J\langle\hat{s}^z\rangle+\frac{J^2\langle S^z\rangle}{2}\chi_0^{+-}(\mathbf{q},\omega)\right)\times\langle S^+(-\mathbf{q})|S^-(\mathbf{q})\rangle_\omega=2\langle S^z\rangle, \tag{25}$$

where

$$\lambda(\mathbf{q})=\sum_\Delta [1-\cos(\mathbf{q}\cdot\Delta)]. \tag{26}$$

It is clear that in the strong coupling limit,

$$\omega(\mathbf{q})_{J\rightarrow\infty}=S_{\text{tot}}^{-1}\frac{1}{2N}\sum_{\mathbf{k}}(\epsilon_{\mathbf{k}-\mathbf{q}}-\epsilon_{\mathbf{k}})\langle\hat{n}_{\mathbf{k}\uparrow}\rangle-2J'\lambda(\mathbf{q}), \tag{27}$$

which can be viewed as the superposition of a ferromagnetic Heisenberg model spectrum and an antiferromagnetic Heisenberg model spectrum. That is to say that, as long as the ground state remains ferromagnetic, the spin-wave spectrum should still be that of a Heisenberg ferromagnet in the infinite J limit.

In conclusion, we have presented an RPA theory for spin waves in FKLM for both the strong-coupling limit and the intermediate coupling regime. In the strong-coupling limit, we show that if the conduction electrons can be described by a single band tight-binding model, a Heisenberg-like spin-wave spectrum is to be expected *throughout* the whole zone. Our results for the strong-coupling limit agree with existing results with $J\rightarrow\infty$ taken *a priori*.[1,2] We show that in the intermediate coupling regime, more complicated spin-wave spectra, such as softening or disappearance of spin waves can be observed. Both are closely related to the detailed structures of the bare susceptibility χ_0^{+-}. In our simple numerical model calculation, the softening is due to the additional peaks of $\chi_0(\mathbf{q},0)$ other than that at the Γ point. We also show that as long as the infinite J limit is taken, the inclusion of superexchange antiferromagnetic coupling between local spins does not change the nature of the spin-wave spectrum.

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