Magnon Broadening Effect by Magnon-Phonon Interaction in Colossal Magnetoresistance Manganites

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In order to study the magnetic excitation behaviors in colossal magnetoresistance manganites, a magnon-phonon interacting system is investigated. Sudden broadening of magnon linewidth is obtained when a magnon branch crosses over an optical phonon branch. Onset of the broadening is approximately determined by the magnon density of states. Anomalous magnon damping at the brillouine zone boundary observed in low Curie temperature manganites is explained.

KEYWORDS: Colossal magnetoresistance manganites, zone boundary magnon broadening, magnon-phonon interaction

Introduction: One of the main interests in colossal magnetoresistance (CMR) manganites is to investigate changes in the microscopic behaviors upon the band-width control. Compounds with larger ionic radius for A-site ions have relatively high Curie temperature \((T_c)\) and exhibit canonical half-metallic double exchange (DE) behaviors. This is considered to be due to large \(e_g\) electron hopping and thus wide bandwidth of the itinerant electrons which favors the ferromagnetic state to maximize the kinetic energy. On the other hand, decreasing the radius makes \(T_c\) lower and finally drives the system into charge-ordered insulator. Magnetoresistance phenomena becomes prominent in the intermediate region where ferromagnetism and charge ordering are competing. In order to study the effect of such competitions to various physical properties, it is important to make an investigation of these compounds in the intermediate region.

One example of such systematic studies is the measurement of magnon dispersion relations of these compounds in the ferromagnetic state. For high-\(T_c\) compounds, \(e.g., (La,\text{Sr})\text{MnO}_3\) and \((La,\text{Pb})\text{MnO}_3\) with hole-carrier concentration \(x \sim 0.3\), cosine-type dispersion relations are reported. This behavior is reproduced by the linear spin-wave approximation of the single-band DE model at \(S \to \infty\) and \(J_H \to \infty\), where \(S\) is the \(t_{2g}\) spin and \(J_H\) is the Hund’s coupling, respectively. For low-\(T_c\) compounds \((Pr,\text{Sr})\text{MnO}_3\), \((Nd,\text{Sr})\text{MnO}_3\) and \((La,\text{Ca})\text{MnO}_3\), however, magnon dispersion is reported to deviate from the cosine-band. Prominent softening of magnon at the zone boundary is observed.

Although zone boundary softening occurs at finite \(J_H\) and finite \(S\) in the single-band DE model, it is not sufficiently large compared to the data. Taking into account the realistic electronic structure, Solovyev et al. discuss that the spin wave softening is of purely magnetic origin. It is claimed to be essential to include the Mn \(t_{2g}\) and O 2p orbitals. On the other hand, Khalliliun et al. have calculated the spin wave dispersion in the presence of orbital fluctuations as well as phonons to explain the zone boundary softening. In such a case, softening occurs as a precursor of ferromagnetic orbital ordering accompanied by the \(A\)-type antiferromagnetism.

Another point of issue is broadening effect of the magnons. In wide band compounds \((La,\text{Sr})\text{MnO}_3\) and \((La,\text{Pb})\text{MnO}_3\) with high \(T_c\) \((x = 0.2 \sim 0.3)\), magnon dispersion is observed throughout the brillouine zone at low temperature limit. There is no magnon damping at high magnon-frequency region expected for itinerant weak ferromagnets. Within the DE model with large \(J_H\), the density of states for itinerant \(e_g\) electrons shows a large exchange splitting. The Stoner excitation is gapped and its continuum lies at \(\omega \sim 2J_H\), which does not interact with low energy magnons. Magnon damping due to thermally induced minority band scales as \(\Gamma \propto (1 - m^2) \cdot \omega_g\), where \(m = M_{sat}/M_{tot}\) is the total magnetization \(M_{tot}\) scaled by the saturation magnetization \(M_{sat}\) at the lowest temperature \(M_{sat}\). This scaling form is indeed observed in the wide bandwidth compound at small \(q\).

However, in narrow band compound \((Pr,\text{Sr})\text{MnO}_3\), sudden broadening of the magnon linewidth near the zone boundary is observed. More quantitative measurement has been done for another low-\(T_c\) compound, \((La,\text{Sr})\text{MnO}_3\) with less doping \(x = 0.15\). The inelastic neutron scattering data show narrow magnon linewidth at small \(q\) region and sudden increase of linewidth near the zone boundary. The experiment is performed at low...
temperature with saturated magnetization $m \sim 1$ where no Stoner contribution is expected. Indeed, in the small $q$ region magnon linewidth obeys the scaling law due to the magnon-magnon scattering, $\Gamma_{\text{mag}} \propto q^4 \ln^2(T/\omega_q)$. On the other hand, magnons at the zone boundary have far wider linewidth than the scaling form. They are heavily damped so that the linewidth are comparable to the magnon energy. Such magnon damping should be attributed to some mechanisms other than Heisenberg-type interactions. In the case of (Nd, Sr)MnO$_3$ at $x = 0.3$, zone boundary magnons are overdamped and below the sensitivity of the measurement.

Investigation of the mechanisms for such anomalous magnon damping is very important to understand the magnetic and electronic behaviors of CMR manganites in the low $T_c$ intermediate bandwidth region. This may cast doubt on adapting the half-metallic electronic structure for these low $T_c$ compounds if the damping mechanism is due to Stoner continuum absorption. Another possibility is that strong spin-orbital exchange interaction causes the damping of magnon coupled to the orbital liquid states.

Recent experiment by Dai et al. shows that the onset of the linewidth broadening coincides with the point where the magnon branch crosses the longitudinal optical phonon branch, for various directions in the Brillouine zone. Magnon linewidth below the crossing frequency is narrow. Therefore, they emphasized the role of magnon-phonon coupling as a mechanism of the anomalous magnon broadening. Another important aspect observed here is that phonon broadening is not reported in the entire the Brillouine zone, even though strong magnon-phonon coupling is speculated. In this letter, we focus on the role of the magnon-phonon interaction and calculate the linewidth broadening of magnons and phonons.

**Magnon-Phonon interaction:** In CMR manganites, cooperative Jahn-Teller effect which strongly couples lattice distortion to conduction electrons has been discussed. In such case, magnon-phonon coupling will be produced by the modulation of the DE interaction $J$ by lattice displacements in the form $J(x) = J(x_0) + (\partial J/\partial x) \cdot \delta x$, where $x_0$ and $\delta x = x - x_0$ are the atomic distance in the equilibrium and the atomic displacement, respectively. Such interaction of spin and lattice degrees of freedom which conserves the spin quantum number is described in a generic form

$$\mathcal{H}_{\text{int}} = \sum_{kq} \left\{ g(k,q)a_{k+q}^\dagger a_k b_q + h.c. \right\},$$

where $a$ ($b$) is the magnon (phonon) annihilation operator and $g$ is the magnon-phonon interaction vertex function. The vertices are illustrated in Fig. 1. We define the magnon dispersion by $\omega_k$ which crosses the phonon dispersion $\Omega_q$ at generic points of the Brillouine zone denoted by $k^\ast$.

![Fig. 1. Magnon-phonon vertices. Solid lines and dashed lines represent magnon and phonon propagators, respectively.](image)

**Magnon linewidth:** Linewidth of magnons is calculated from the self-energy diagram in Fig. 2(a). From the lowest order perturbation calculation at $T = 0$, magnon linewidth broadening $\Gamma_{\text{mag}}$ due to the vertex (a) is given by

$$\Gamma_{\text{mag}}(k) = \text{Im}\Pi_{\text{mag}}(k, \omega_k) \propto \sum_q \delta(\omega_k - \omega_{k-q} - \Omega_q),$$

where $\Pi_{\text{mag}}$ is the magnon self-energy.

![Fig. 2. Self-energy diagrams for (a) magnon and (b) phonon. Shaded areas represent vertex corrections.](image)

If we consider an Einstein phonon $\Omega_q = \Omega_0$, we have

$$\Gamma_{\text{mag}}(k) \propto D_{\text{mag}}(\omega_k - \Omega_0)$$

where $D_{\text{mag}}$ is the magnon density of states. In three dimension, $D_{\text{mag}}(\omega) \sim \sqrt{\omega}$ for $\omega \geq 0$ while $D_{\text{mag}} = 0$ for $\omega < 0$. The result is schematically illustrated in Fig. 3. Namely, for $\omega_k < \Omega_0$ the linewidth of the magnon due to the magnon-phonon interaction is small. As the magnon dispersion crosses the phonon branch, the linewidth suddenly becomes broad. In the case of finite dispersion of phonon frequency, the onset of the linewidth broadening at the magnon-phonon crossing point will be smeared out, as is indicated by the grey curve in Fig. 3. For two-dimensional magnons, the density of states is step-function like, and a more abrupt increase of magnon linewidth is expected.

The result is intuitively understood as follows. When magnon frequency is larger than the crossing frequency, it is possible that a magnon with momentum and frequency $(k, \omega_k)$ is scattered into a magnon $(k - q, \omega_{k-q})$

$$\frac{\partial J}{\partial x} \cdot \delta x,$$
and a phonon \((q, \Omega_q)\) obeying \(\omega_k = \omega_{k-q} + \Omega_q\) through the magnon-phonon interaction \(\Delta\). Thus magnon energy dissipates into the bosonic bath of optical phonons. Near the zone center \(k \ll k^*\), magnons with smaller frequencies do not suffer from such processes.

In the strong coupling region, a self-consistent treatment of phonons and magnons gives a qualitatively similar result. Finite lifetime of magnons and phonons creates the broadening of the delta-function in \((2)\) which again causes a smearing of the onset of the magnon broadening. At the onset, relevant contributions come from small frequency magnons \(\omega_k\) at \(k \sim 0\) and phonons at the crossing point \(q \sim k^*\). Therefore the broadening of the delta-function should not be large. An incoherent part of the propagators creates a weakly \(q\)-dependent background in magnon width.

**Phonon linewidth:** Phonon linewidth is calculated from the self-energy diagram in Fig. 2(b). Note that an magnon-phonon interaction which conserves spin quantum number creates a self-energy with only magnon-antimagnon diagrams. In this case, we have

\[
\Gamma_{ph}(q) \propto \sum_k \delta(\Omega_q - \omega_{k+q} + \omega_k) \cdot (n(\omega_k) - n(\omega_{k+q})), \tag{4}
\]

where \(n(\omega)\) is the Bose distribution function. At \(T \to 0\), we have \(n(\omega_k) \to 0\) so that \(\Gamma_{ph} \to 0\). Thus, the difference in the type of self-energy diagram (Fig. 2(a-b)) explains that phonon linewidth does not show broadening even if the magnon-phonon interaction is strong enough to exhibit anomalous magnon broadening. Only at finite temperature there is a finite contribution from the thermal spin fluctuation.

In the presence of spin-orbit \((L-S)\) coupling, spin quantum number is not conserved. In such a case, magnon-phonon hybridization terms e.g. \(\sum \lambda(k)(a_k^\dagger b_k + h.c.)\) appear. These terms create hybridization gap at the magnon-phonon crossing point. Since in high-\(T_c\) \((\text{La,Sr})\text{MnO}_3\) a smooth crossing of magnon and phonon branch is experimentally observed, such a hybridization term should be small, although the anomalous Hall effect in the ferromagnetic phase suggests it be non-zero.

Since the hybridization terms are bilinear in magnon and phonon operators, this interaction does not cause the magnon broadening effect. Hybridization causes the mixture of magnon and phonon wavefunctions in both excitation spectra. If hybridization is small, as expected, the qualitative behavior in the linewidth does not change. Broadening suddenly occurs at the "optical" branch above the crossing point which is mostly magnon-like but has non-zero phonon character as well. In Fig. 4 we schematically illustrate the dispersion relation of the magnon and phonon branches as well as the magnon linewidth in the presence and absence of magnon-phonon hybridization terms.

**Discussion:** A crucial test for the present model is to measure the temperature dependence of the phonon linewidth. From eq. \((4)\) we expect an increase of the linewidth as temperature is increased, through the magnon population \(n(\omega_k)\). At a fixed temperature, \(n(\omega)\) is maximum at \(k \to 0\) \((\omega_k \to 0)\) and eq. \((4)\) suggests that \(\Gamma_{ph}\) is maximum at \(\Omega_q \simeq \omega_q\). Namely, the
phonon linewidth should be maximum around the crossing point. At the Curie temperature $T_c$, magnon population should critically increase, so yet another anomalous broadening is expected in the phonon linewidth.

The magnon damping effect given here can be generalized to any system where magnons couple with other bosonic degrees of freedom through the interaction in the form $\Delta E = \frac{1}{2} \hbar \gamma^2 \mathbf{q} \cdot \mathbf{a}(\mathbf{q}) \mathbf{a}(\mathbf{q})^\dagger$. In manganites, another possible source of damping is the orbital wave excitations. Namely, accurate measurement of magnon dispersion and linewidth might become an indirect method to investigate the orbital dynamics. At the same time, for metallic manganites in the low-temperature ferromagnetic phase at around $x \sim 0.3$, no anomalous magnon broadening has been reported so far in the low energy region $\lesssim 10\text{meV}$. This might indicate that it is not likely to assume low-energy orbital fluctuations. The mechanism for magnon broadening discussed here does not require an intrinsic modification to the picture of half-metallic double exchange ferromagnet as the low temperature state of manganites in the low energy region.

By decreasing A-site radius in manganites, increase of the magnon damping at the zone boundary is prominent, and at the same time spin stiffness constant which is a measure for electron kinetic energy only makes a small change. Similarly, decrease of $T_c$ by A-site ionic radius control has been known to be unexpectedly large compared to the nominal bandwidth change. Therefore, it is not likely to simply assume an increase of magnon-phonon coupling through the decrease of electronic bandwidth while keeping the lattice couplings constant. A possible origin of a sudden increase in the coupling constant might be due to the fact that the metallic state is in a close vicinity to the charge ordered insulating state, through fluctuations toward the phase with larger lattice distortions. Note that such increase is speculated to occur both at low temperature and at $T \sim T_c$, although it has been considered that dynamical lattice distortions are suppressed in the low temperature ferromagnetic phase. Further study of such dynamical fluctuations may give more clear understandings in the intermediate compounds at low temperature, including a possible phase separation phenomena toward ferromagnetic metal and charge ordered insulator.

To summarize, the effects of the magnon-phonon interaction to the broadening of magnons are studied at the presence of magnon-phonon dispersion crossing. When the interaction conserves spin quantum number, the magnon linewidth becomes broad only when magnon frequency is larger than the magnon-phonon crossing frequency. Anomalous broadening at the zone boundary, which is observed in the narrow bandwidth CMR manganites, will be reproduced if we simply assume that the magnon-phonon coupling is strong enough. Phonon linewidth does not show broadening at low temperature.

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