Electromagnon in one-dimensional frustrated chain

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Abstract. Recently, a novel one-magnon excitation induced by electric components of light, so called electromagnon, has been paid attention. Theoretically, a mechanism to induce a one-magnon excitation by electric component of light has been proposed. Such a magnetic excitation can be possible in spins with noncollinear structures due to the couplings between symmetric spin-dependent electric polarizations and electric fields. We adopt the mechanism to one-dimensional frustrated Heisenberg systems, where cycloidal spin states are the ground state in highly frustrated parameter region, and discuss the possible electromagnon processes.

1. Introduction

Multiferroics, where ferromagnetism and ferroelectricity coexist, have attracted both experimental and theoretical interests [1]. One of the hot topics in multiferroics is an electromagnon, i.e. electric component of light induced spin excitations. In multiferroics RMnO₃, such electric absorptions by magnons have been observed in an optical spectroscopy at terahertz frequencies [2–12]. Recent theory indicates that parts of the absorption can be explained well by considering the one-magnon excitation induced due to the couplings between electric fields \( \vec{E}_\omega \) and symmetric spin-dependent polarization \( P_S \propto \vec{S}_i \cdot \vec{S}_j \) [9, 13]. So far, it was believed that couplings between \( \vec{E}_\omega \) and \( P_S \) can induce simultaneous two-magnon excitations in Heisenberg systems and can not induce one-magnon excitation as well known in Néel ordered state [14]. However, in non-collinear structures, even one-magnon excitation can be induced by oscillating electric field \( \vec{E}_\omega \). This becomes possible because the ground state itself is a symmetry broken state, which makes a matrix element between ground state and one-magnon excitations non-zero.

In this paper, we review the fact that one-magnon absorption is possible in non-collinear structure. In Sec. 3, as a typical example, we show that such an electromagnon can be induced in one-dimensional frustrated Heisenberg system. Summary is given in Sec. 4.

2. Electromagnon in spins with noncollinear structures

The spin Hamiltonian under the external electric field \( \vec{E} \) can be written as \( \mathcal{H} = \mathcal{H}_0 - \vec{E} \cdot \vec{P}_S \), where \( \mathcal{H}_0 \) is a Heisenberg Hamiltonian and \( \vec{P}_S \) is a symmetric spin-dependent electric dipole [14, 15]:

\[
\vec{P}_S = \sum_{<i,j>} \vec{\Pi}_{ij} (\vec{S}_i \cdot \vec{S}_j).
\]

(1)

Note that if the center of the bond has an inversion symmetry, \( \vec{\Pi}_{ij} \) vanishes.
For spins with noncollinear structures, one-magnon can be induced by the oscillating electric fields through the couplings $\vec{E} \cdot \vec{P}_S$. It can be understood by considering one bond connecting noncollinear spins on site 1 and 2 (Fig. 1) with $\vec{P}_S = \vec{\Pi}_{12} (\vec{S}_1 \cdot \vec{S}_2)$. When we apply the oscillating electric field $\vec{E}\omega$, Hamiltonian of the field can be represented as

$$-\vec{E}\omega \cdot \vec{P}_S = -\frac{1}{2} (\vec{H}_1^{\omega} \cdot \vec{S}_1 + \vec{H}_2^{\omega} \cdot \vec{S}_2)$$

(2)

by using effective fields $\vec{H}_1^{\omega}$ and $\vec{H}_2^{\omega}$, which coupled to $\vec{S}_1$ and $\vec{S}_2$ respectively. Here $\vec{H}_1^{\omega} = (\vec{E}\omega \cdot \vec{\Pi}_{12}) \vec{S}_2$ and $\vec{H}_2^{\omega} = (\vec{E}\omega \cdot \vec{\Pi}_{12}) \vec{S}_1$. When $\vec{S}_1$ and $\vec{S}_2$ are noncollinear, $\vec{H}_1^{\omega}$ and $\vec{H}_2^{\omega}$ have a transverse component with respect to each spin, whose magnitude is approximated as $H^{\omega}_\perp \sim (\vec{E}\omega \cdot \vec{\Pi}_{12}) S \sin \theta$. Here $\theta$ is the relative angle between $\vec{S}_1$ and $\vec{S}_2$ as shown in Fig. 1. In this way, the effects of electric component of light can be described as an effective magnetic field, which induces a one-magnon resonance.

Figure 1. One bond model with a symmetric spin dependent polarization $\vec{\Pi}_{12} (\vec{S}_1 \cdot \vec{S}_2)$. By applying the electric fields $\vec{E}\omega$, spins feel effective transverse magnetic fields $H^{\omega}_\perp \sim (\vec{E}\omega \cdot \vec{\Pi}_{12}) S \sin \theta$ as shown by red arrows. $\theta$ is an angle between $\vec{S}_1$ and $\vec{S}_2$.

3. Electromagnon in zig-zag chain model

Let us consider a classical $J_1$-$J_2$ chain model:

$$\vec{H}_0 = \sum_{n.n.} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{n.n.n.} J_2 \vec{S}_i \cdot \vec{S}_j. \quad (3)$$

For antiferromagnetic next nearest neighbor interaction $J_2$ ($J_2 > 0$), cycloidal state is the ground state in the condition $J_2/|J_1| > 0.25$. Low lying magnetic excitations at low-temperatures can be described well by the linear spin wave theory [16]. After rotations of local axis and Holstein-Primakoff approximations $\alpha_k = c_k a_k + s_k a_k^\dagger$, where $c_k^2 - s_k^2 = 1$, the spin Hamiltonian (3) for the cycloidal state, whose rotation angle is $\theta$, is written by the spin wave annihilation (creation) operator $\alpha_k^\dagger$ ($\alpha_k$) as

$$H_0 = S \sum_k \hbar \omega_k \alpha_k^\dagger \alpha_k + \text{const.} \quad (4)$$

The spin wave frequencies are given by

$$\hbar \omega_k = 2 \sqrt{A_k^2 - B_k^2}.$$ 

(5)

where

$$A_k = -J_1 \cos \theta + J_1 \cos^2 \frac{\theta}{2} \cos k_z - J_2 \cos (2\theta) + J_2 \cos^2 \theta \cos (2k_z),$$

(6)

$$B_k = J_1 \sin^2 \frac{\theta}{2} \cos k_z + J_2 \sin^2 \theta \cos (2k_z).$$

(7)
To discuss the electromagnon in zig-zag chain model, we assume three types of polarization structures as shown in Fig. 2 (a)-(c):

$$P_{S_z}^{(a)} = \sum_i (-1)^i \Pi(\vec{S}_i \cdot \vec{S}_{i+1}),$$

$$P_{S_z}^{(b)} = \sum_i \left[ \cos \left( \frac{\pi}{2} r_i \right) \Pi(\vec{S}_i \cdot \vec{S}_{i+1}) \right],$$

$$P_{S_z}^{(c)} = \sum_i \sqrt{2} \cos \left( \frac{\pi}{2} r_i + \frac{\pi}{4} \right) \Pi(\vec{S}_i \cdot \vec{S}_{i+1}).$$

To produce polarization between nearest neighbor spins, inversion symmetry can not exist at the center of the bonds. We assume that $C_2$ rotation symmetry exists along the chain, then, the polarizations are aligned along chains. In the structure (a), inversion center exists at all spin sites, which produces the staggered polarizations. When dimerization occurs and inversion center exists at one of the bonds, polarization structure is described in Fig. 2 (b). If there are two inequivalent spin sites and there is no inversion symmetry at one of them, the structure in Fig. 2 (c) is expected. In the electric field along $z$ direction $E_z$, these spin dependent polarizations are also represented by the spin wave operators respectively:

$$E_z P_{S_z}^{(a)} \sim \sqrt{2 S^3 N} E_z \Pi \sin \theta (c_{\pi} - s_{\pi}) \left( \alpha^\dagger_{\pi} + \alpha_{\pi} \right) + \cdots,$$

$$E_z P_{S_z}^{(b)} \sim \frac{\sqrt{S^3 N}}{2} E_z \Pi \sin \theta \left( c_{\pi/2} - s_{\pi/2} \right) \left\{ e^{-i\pi/4} \left( \alpha^\dagger_{\pi/2} + \alpha_{-\pi/2} \right) + e^{i\pi/4} \left( \alpha^\dagger_{-\pi/2} + \alpha_{\pi/2} \right) \right\} + \cdots,$$

$$E_z P_{S_z}^{(c)} \sim -i \sqrt{\frac{S^3 N}{2}} E_z \Pi \sin \theta \left( c_{\pi/2} - s_{\pi/2} \right) \left( \alpha^\dagger_{\pi/2} - \alpha_{\pi/2} - \alpha^\dagger_{-\pi/2} + \alpha_{-\pi/2} \right) + \cdots.$$  

In this way, these polarization can induce one-magnon excitation by oscillating electric fields. For the configuration (a), zone-edge magnon is induced, whereas magnon at $k_z = \pi/2$ is excited in the configurations (b) and (c). As obvious from Eqs. (11)-(13), such an excitation can be possible only for cycloidal spin state, and it is prohibited for ferromagnetic ($\theta = 0$) and antiferromagnetic ($\theta = \pi$) ordered states. If the staggered antiferroelectric spin dependent polarization is realized as in Fig. 2 (a), a magnetic resonance due to electric field occurs at the resonance frequency $\omega_{\pi}$:

$$\hbar \omega_{\pi} = |J_1| \left( 1 - \frac{J_1}{4J_2} \right).$$

For polarization structures in Figs. 2 (b) and (c), the resonance frequency $\omega_{\pi/2}$ is written as

$$\hbar \omega_{\pi/2} = |J_1|,$$

which does not depend on the interaction ratio $J_2/|J_1|$, i.e. the cycloidal angle $\theta$. For $J_2/J_1 = \pm 0.3$, $\pm 0.5$, and $\pm 1.0$ ($J_2 > 0$), the dispersion of one-magnon excitation and the electromagnon energies are shown in Fig. 3.
Figure 3. Magnon dispersions in one-dimensional $J_1$-$J_2$ Heisenberg model for $J_2/J_1 = \pm 0.3, \pm 0.5$, and $\pm 1.0 (J_2 > 0)$. Electromagnon energies induced by the $E_z \vec{P}^{(a)}_{S_z}$ term $\omega_\pi$ are represented by squares. Electromagnon energies due to $E_z \vec{P}^{(b)}_{S_z}$ and $E_z \vec{P}^{(c)}_{S_z}$ terms $\omega_{\pi/2}$ are shown by a circle, which does not depend on the interaction ratio $J_2/|J_1|$.

4. Conclusion

In this paper, we investigated the electromagnon absorption in the one-dimensional zig-zag chain system. Once the unit cell is larger, several magnitudes of spin dependent polarization can exist. Such a situation can make it possible to induce one-magnon resonance at several frequencies. For example, when polarization is described as a sum of $P_A$ and $P_B$ ($P_C$), the resonances at both $\omega_\pi$ and $\omega_{\pi/2}$ are expected. Such a one-magnon resonance is likely observable in frustrated spin systems quite generally, as already observed in $RMnO_3$.

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