Electromagnon excitation and longitudinal mode studied on the basis of symmetric spin-dependent electric polarization

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Abstract. Since a symmetric spin-dependent electric polarization makes quantum spin systems couple to an electric field, it is one of a possible origin of electromagnon excitations in noncollinear (spiral) magnetically ordered states. In collinear ordered states, however, it is believed that the electromagnon excitation is difficult to realize. In the present study, we focus on the symmetric spin-dependent electric polarization and study the electromagnon excitation in both collinear and noncollinear states. In one-magnon excitation process, it is found that the symmetric spin-dependent polarization couples to a longitudinal fluctuation of the ordered moment and that the electromagnon measurements probe the longitudinal (Higgs) mode selectively. In noncollinear states, we report that both Higgs and Nambu-Goldstone (transverse) modes are observable.

1. Introduction
Electromagnon is a magnetic excitation caused by an electric-field component of an electromagnetic wave in one-magnon process [1, 2]. To excite the electromagnon, it is necessary that spins couple to the electric field. The electric polarization can be described by spin operators, such as in symmetric $S_i \cdot S_j$ or antisymmetric $S_i \times S_j$ forms [3, 4, 5, 6, 7]. The former and the latter can be derived from magnetostriction and spin current mechanisms, respectively. The latter has been studied to account for multiferroic properties in noncollinear magnets $RMnO_3$ [6]. Electromagnon excitations were studied on the basis of the antisymmetric polarization [8, 9]. As in the antisymmetric case, the symmetric spin-dependent polarization can excite an electromagnon in noncollinear states [10, 11, 12, 13, 14, 15, 16], where a transverse magnon mode (Nambu-Goldstone mode) is excited. In contrast, it has been believed to be difficult to excite the electromagnon in collinear magnets. However, it is reported that the symmetric spin-dependent polarization couples to a longitudinal fluctuation and excites the longitudinal mode selectively in collinear states [17]. Since the longitudinal mode is formed as a consequence of the amplitude fluctuation of the order parameter (ordered moment in the present case), such mode in condensed matter physics is termed “Higgs mode” in analogy with the Higgs boson in high-energy physics [18]. In this sense, the electromagnon excitation probes the Higgs mode selectively in collinear magnets.

The Higgs mode is hard to detect in conventional magnetic compounds, since the ordered magnetic moment is large and stiff. However, the Higgs mode can survive in the vicinity of a quantum critical point, since the size of the moment is shrunk by a quantum effect and it does
not cost so high energy to modulate the moment size. Spin dimer system TiCuCl₃ is one of a typical compound showing the Higgs mode, where inelastic neutron scattering measurements revealed how the massive Higgs mode and the massless Nambu-Goldstone (N-G) mode evolve with pressure through the quantum critical point [19, 20, 21, 22]. In this paper, we report how the Higgs mode is detectable by the electromagnon measurements on the basis of the symmetric spin-dependent electric polarization.

This paper is organized as follows. In Sect. 2, we introduce a model Hamiltonian and give formulation of the extended spin-wave theory. The spin-dependent electric polarization is also discussed. The obtained results are presented in Sect. 3. The last section gives summary and discussion.

2. Formulation

2.1. Model Hamiltonian

As a simple system showing a quantum phase transition, we focus on an $S = 1$ spin system with a large easy-plane ($D > 0$) single-ion anisotropy $[D(S^z)^2]$.

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_i D(S^z_i)^2$$

(1)

Here, $\mathbf{S}_i$ is the spin operator at the $i$-th site. $J_{ij}$ is the coupling constant of the intersite exchange interaction. As shown in Fig. 1(a), we assume a tetragonal system with antiferromagnetic (AF) nearest ($J_{ab}$ and $J_1$) and a next nearest neighbor ($J_2$) interactions. Since $J_1$ and $J_2$ are competing, collinear and noncollinear states are stabilized for $J_2/J_1 < 1/4$ and $J_2/J_1 > 1/4$, respectively [15]. The local energy level scheme is shown in Fig. 1(b), where twofold degenerate ($S^z = \pm 1$) excited states are separated by $D$ from the singlet ($S^z = 0$) groundstate. For weak intersite interactions, a nonmagnetic (disordered) state is stabilized even at low temperatures. In the presence of the intersite interactions, the douplet $S^z = \pm 1$ excited states can move around and the excitation becomes dispersive. The excitations are twofold degenerate with a finite excitation gap in the nonmagnetic phase (see Fig. 2). When we increase the strength of the intersite interactions, the magnon band width becomes wider and the lowest mode goes soft at the quantum critical point, showing a linear dispersion relation around the AF wavevector $Q$. In the ordered phase, a finite magnetic moment lifts the degeneracy and the excitation modes split into a massless N-G and a massive Higgs modes, as shown in Fig. 2 [25, 26].

2.2. Extended spin-wave theory

In low-dimensional cases, the low-energy physics of the $S = 1$ system can be mapped to a nonlinear sigma model [23]. In the present study, we consider a three-dimensional case with a large $D$ term such as in NiCl₂⁺ ($S = 1$) system NiCl₂-4SC(NH₂)₂ [24]. We apply the extended spin-wave theory to describe magnetic excitations in both nonmagnetic and magnetic phases. First, we solve a mean-field (classical) solution on the basis of a mean-field Hamiltonian within the local three ($S^z_i = 1, 0, -1$) states. The AF wavevector is given by $Q = (\pi, \pi, Q_c)$. Here, $Q_c$ characterizes the spiral structure along the $c$-axis. It is determined as $Q_c = \cos^{-1}(-(J_1/4J_2)$ for $J_2/J_1 > 1/4$ and as $Q_c = \pi$ for $J_2/J_1 < 1/4$. The quantum critical point is located at $J_{eff}/D = 1$, where $J_{eff} = 8J_{ab} - 4J_1 \cos Q_c - 4J_2 \cos 2Q_c$ represents an effective intersite exchange interaction.

Next, we introduce three ($m = 0, 1, 2$) bosons which create and annihilate the local energy eigenstates (mean-field solutions). The bosons are subjected to the local constraint $\sum_{m=0,1,2} b_{i,m}^\dagger b_{i,m} = 1$. Here, $b_{i,0}$ is for the mean-field ground state, while $b_{i,1}$ and $b_{i,2}$ are for the excited states. The spin operators and the Hamiltonian are described by the bosons. Bose-Einstein condensation of the $b_{i,0}$ boson gives the mean-field (classical) value. Using the local constraint, we replace the $b_{i,0}$ boson as $b_{i,0} \rightarrow (1 - \sum_{m=1,2} b_{i,m}^\dagger b_{i,m})^{1/2}$. Here, the square root
is expanded assuming dilute excited bosons. The Hamiltonian is then described only by the $b_{i,1}$ and $b_{i,2}$ bosons. We retain up to the quadratic order of the bosons. The Hamiltonian is diagonalized by a Bogoliubov transformation and we obtain the magnon modes. Details of the formulation is given in Ref. 17.

2.3. Symmetric spin-dependent electric polarization and electromagnon excitation

The symmetric spin-dependent electric polarization operator can be expressed as [3, 4, 5, 7].

$$P = \sum_{\langle ij \rangle} \Pi_{ij} (S_i \cdot S_j)$$  \hspace{1cm} (2)

Here, $\Pi_{ij}$ is the local polarization which is determined by the crystal symmetry. As a possible example, we consider a tetragonal system shown in Fig. 3. The polarization is along the c-axis (z direction) and is characterized by a modulation wavevector $Q_p = (0, 0, \pi/2)$. The spin operator in Eq. (2) can be separated into a static ($M_j$) and a fluctuating ($\delta S_i$) parts as $S_i = M_i + \delta S_i$. Substituting this into Eq. (2), we obtain the following local polarization in one-magnon process:

$$P^z_i = \Pi^z \mathbf{H}^\omega_{i,\text{eff}} \cdot \delta \mathbf{S}_i, \quad \mathbf{H}^\omega_{i,\text{eff}} = \sum_j (\Pi^z_{ij}/\Pi^z) M_j.$$  \hspace{1cm} (3)

Since the alternating electric field is coupled to the spins by $\mathbf{H}'_p = -\sum_i \mathbf{P}_i \cdot \mathbf{E}^\omega_i$, $\mathbf{H}^\omega_{i,\text{eff}}$ can be understood as an alternating effective magnetic field acting on the local spin $S_i$. It comes from the static component ($M_j$) from the nearest-neighbor sites and gives rise to the electromagnon excitation [13, 14, 15]. In Figs. 4(a) and 4(b), we show how the electromagnon is excited in collinear and noncollinear states, respectively.

In collinear states, the effective field $\mathbf{H}^\omega_{i,\text{eff}}$ is parallel to the local moment. This indicates that the electric field excites the Higgs mode selectively [see Fig. 4(a)]. In conventional AF magnets, however, there is no Higgs mode and this one-magnon process has been discarded. In our study, we emphasize that the Higgs mode is observable as the electromagnon excitation even in the collinear case when the magnetic moment is shrunk by quantum effects.

In noncollinear states, $\mathbf{H}^\omega_{i,\text{eff}}$ has both the parallel and perpendicular components, as shown in Fig. 4(b). Correspondingly, the electromagnon intensity can be separated into two parts.
Figure 3. Schematic of position-dependent electric polarization (red thick arrow). It is characterized by $Q_p = (0, 0, \pi/2)$. The blue circle and red cross represent the magnetic ion site and the inversion center, respectively.

Figure 4. Magnetic moments viewed from the $c$-axis. The moment is aligned in the $ab$-plane under the easy-plane anisotropy. (a) For collinear states. The alternating effective magnetic field $H_{r,\text{eff}}^{i}$ is parallel to $M_i$ and it couples to the longitudinal fluctuation $S_i$. (b) For noncollinear states. $H_{r,\text{eff}}^{i}$ has both parallel and perpendicular components relative to $M_i$.

proportional to $\sin^2 Q_c$ and $\cos^2 Q_c$ [17]. The former and the later are from the transverse and longitudinal fluctuations of the local moment, respectively. The two fluctuations couple to each other by the $J_1$ and $J_2$ interactions along the $c$-axis. As the result, the two excitation modes are formed as a mixture of the two local fluctuations [17]. In the noncollinear case, we note that both the N-G and Higgs modes are observable as electromagnon excitations.

3. Result

In Fig. 5, we show the magnon dispersion relation and dynamical spin correlation function for $J_{ab} = J_1 = 3.5 J_2 (Q_c = 0.84\pi)$. In the nonmagnetic phase, the twofold excitation modes have a finite gap [see Fig. 5(a)]. The red open circle represents the mode excited by a magnetic-field component of light. The excitation gap closes at $Q$ on the quantum critical point, as shown in Fig. 5(b). In the ordered phase, the modes split into a N-G and a Higgs modes [see Fig. 5(c)]. The red closed squares represent the modes excited by an electric-field component. In a deep ordered phase region, energies of the N-G and the Higgs modes well separate [see Fig. 5(d)].

In Fig. 5(e), we show the $J_{\text{eff}}/D$ dependence of intensities of the light absorption caused by the magnetic- and electric-field components. The former and the latter intensities are obtained from the dynamical correlation functions of spin and electric polarization, respectively. We can see that there are magnetically excitable two modes ($E_{0T}$ and $E_{0L}$) [open circles in Figs. 5(a)-5(d)]. The electromagnon intensity appears only in the ordered phase, since it is proportional to the squared moment ($M^2$). The lower ($E_{Q_c,T}$) and the higher ($E_{Q_c,L}$) modes [closed squares in Figs. 5(c)-5(d)] are the N-G and the Higgs modes, respectively. As discussed in Sec. 2.3 (see Fig. 4), both modes are observable in noncollinear states. In Fig. 5(f), we show the $J_{\text{eff}}/D$ dependence of the electromagnon intensities for $E_{Q_c,T}$ and $E_{Q_c,L}$. Contributions from the transverse and longitudinal fluctuations of the local moment are shown separately. They are proportional to $\sin^2 Q_c$ and $\cos^2 Q_c$, respectively. In the vicinity of the quantum critical point, the two fluctuations are coupled strongly to form the excitation modes. In a deep ordered phase region (large $J_{\text{eff}}/D$), however, we can see that the N-G and the Higgs modes are formed purely from the transverse and longitudinal fluctuations of the local moment, respectively.

At the end, let us discuss collinear states. In this case, only the Higgs mode is excitable as
Figure 5. Dynamical spin correlation function (in arbitrary unit). Solid lines are magnon modes. Folded branches are also shown. (a) $J_{\text{eff}}/D = 0.9$. (b) $J_{\text{eff}}/D = 1$. (c) $J_{\text{eff}}/D = 1.1$. (d) $J_{\text{eff}}/D = 3$. Since the alternating magnetic ($\mathbf{H}_\omega$) and electric ($\mathbf{E}_\omega$) fields couple to spins by $\mathbf{H}_m' = -g\mu_B \sum_i \mathbf{S}_i \cdot \mathbf{H}_\omega$ and by $\mathbf{H}_p' = -\sum_i \mathbf{P}_i \cdot \mathbf{E}_\omega$, respectively, the magnetically and electrically (electromagnon) excitable modes are located at $(0,0,0)$ ($E_{0T}$ and $E_{0L}$) and at $Q_p = (0,0,\pi/2)$ ($E_{Q_pT}$ and $E_{Q_pL}$) as pointed by the open circles and filled squares, respectively. Here, $Q_p$ is the wavevector characterizing the spatial dependence of the polarization. (e) $J_{\text{eff}}/D$ dependence of intensities for light absorption. The dashed line represents the excitation energy of the soft mode at $Q$. (f) Electromagnon intensities for $E_{Q_pT}$ and $E_{Q_pL}$. Contributions from the $\cos^2 Q_c$ and the $\sin^2 Q_c$ terms are shown separately.

the electromagnon [see Fig. 4(a)]. For magnetically excitable modes, both the N-G and the Higgs modes are observable.

4. Summary and Discussion
The electric-field component of light can couple to quantum spin systems through the symmetric spin-dependent electric polarization ($P \propto \mathbf{S}_i \cdot \mathbf{S}_j$). In one-magnon process, the electric-field acts on a local moment. In collinear magnets, the electric field shakes the moment parallel to it, owing to the collinear nature in $\mathbf{S}_i \cdot \mathbf{S}_j$. As the result, the Higgs (longitudinal) mode is excited selectively. In noncollinear case, the electric field shakes the moment in both parallel and perpendicular directions and both the Higgs and Nambu-Goldstone (transverse) modes are excited. This is summarized in Fig. 4. In the present study, we focused on the $S = 1$ systems. However, the obtained result also holds in other systems when the ordered moment is shrunk by quantum effects, such as spin dimer systems [21, 22] or $S \geq 1$ systems with the easy-plane single-ion anisotropy as $\text{Ba}_2\text{CoGe}_2\text{O}_7$ ($S = 3/2$ system) [27, 28, 29]. Similarly to
the light absorption, we note that one-magnon Raman scattering is another probe for the Higgs mode. Indeed, the electrically excited one-magnon mode in the field- and pressure-induced ordered phase of $\text{TlCuCl}_3$ has been reported [30, 31, 32]. In conclusion, we investigated the electromagnon excitation and found that it is connected with the Higgs mode. This reveals rich physics in light absorption measurements in magnetic materials.

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