Theory of magnetic excitation for coupled spin dimer and spin chain system Cu$_2$Fe$_2$Ge$_4$O$_{13}$

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Abstract. Cu$_2$Fe$_2$Ge$_4$O$_{13}$ is a quantum magnet consisting of Cu spin dimers and Fe spin chains. It shows an antiferromagnetic phase transition at low temperatures due to the magnetic moment of Fe. Since the two parts are weakly coupled, magnetic excitations are well separated into high-energy Cu dimer part and low-energy Fe chain part. We study the magnetic excitations in the ordered phase of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ based on a theory used for coupled spin dimer systems such as TlCuCl$_3$. Analyzing the dynamical spin correlation functions, we find several high-energy modes with longitudinal spin fluctuations peculiar to the coupled spin dimer and spin chain system. This is consistent with the recent experimental result of inelastic neutron scattering. A possibility to detect such longitudinal modes is discussed by means of magnetic Raman scattering.

1. Introduction
Cu$_2$Fe$_2$Ge$_4$O$_{13}$ is a unique quantum magnet which is built from blocks of CuGeO$_3$ and Fe oxide chains [1]. The crystal structure of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ indicates that it consists of Cu spin dimer part and Fe spin chain part (see Fig. 1). The former is formed with $S = 1/2$ spin of Cu$^{2+}$, while the latter is formed with $S = 5/2$ spin of Fe$^{3+}$. Since the two parts are coupled, Cu$_2$Fe$_2$Ge$_4$O$_{13}$ shows an antiferromagnetic (AF) long-rage ordering at 39 K [2], where Fe ions mainly provide the ordered moment and Cu dimers work as a bridge between the Fe ions. Below the Néel temperature, inelastic neutron scattering measurement observed low-energy excitation mode below 10 meV with substantial dispersion relation [2]. In addition to this, they also found a high-energy excitation mode around 24 meV with weak dispersion relation [3, 4]. The former is well described by a spin wave excitation of the ordered Fe$^{3+}$ moment with a small anisotropy gap of about 1 meV. The latter is an excitation from the spin singlet to triplet of the Cu dimer. Exchange interaction parameters, they are estimated from the neutron scattering experiments, are listed in Table 1 [4]. We use this set of parameters in this paper.

2. Formulation
Since the Fe chain and Cu dimer parts are weakly coupled, the excitation modes are well separated in energy and we can treat them independently. In reality, however, Fe ions are coupled with Cu dimers. Since the Cu dimer is likely to reduce the ordered moment, we can
Figure 1. Crystal structure of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ [1, 2, 3, 4]. Blue and red circles indicate Cu$^{2+}$ and Fe$^{3+}$, respectively. The Fe chain runs in the $b$ direction. This layer structure is stacked along the $c$ axis, where the Fe ions are coupled between adjacent layers by $J_c$. A and B represent two sublattices in a spin cluster unit in the ordered phase, where the spin cluster is composed of two Fe ions and one Cu dimer.

Table 1. AF exchange interaction parameters estimated by inelastic neutron scattering measurements [4].

| Parameter       | Value   |
|-----------------|---------|
| $J_{Cu}$ (meV)  | 2.4(2)×10 |
| $J_b$ (meV)     | 1.60(2)  |
| $J_c$ (meV)     | 0.12(1)  |
| $J_{Cu-Fe}$ (meV)| 2.4(2)   |

expect quantum effects of the dimer in magnetic excitations. The purpose of this paper is to search for a novel unrecognized excitation mode peculiar to the coupled spin dimer and spin chain system.

To carry out this, we need a theory of magnetic excitations that can describe both the low-energy and high-energy modes. Then we consider a spin cluster consisting of two Fe ions and one Cu dimer as indicated by A or B in Fig. 1. We first solve a mean field problem at zero temperature by diagonalizing a mean field Hamiltonian for the spin cluster. We obtain then a spin multiplet with 144 (=$2\times2\times6\times6$) states which are mixtures of the Fe$^{3+}$ spins and Cu dimer. Wavefunctions of low-energy states are dominated by the singlet state of the dimer, while the triplet component of the dimer dominates high-energy states. The mean field groundstate is accompanied by a substantial magnetic moment at the Fe sites of the spin cluster. A finite magnetic moment is also induced at the Cu sites due to the finite coupling between Fe$^{3+}$ and Cu$^{2+}$. Since the spin multiplet is coupled with each other via $J_b$ and $J_c$ shown in Fig. 1, we can regard Cu$_2$Fe$_2$Ge$_4$O$_{13}$ as a system of coupled spin multiplets. For such coupled spin multiplets system, we can apply a generalized Holstein-Primakoff theory which has been introduced to describe excitation modes of higher rank multipoles of localized 4$f$ electron systems [5, 6, 7]. This theory is equivalent to the bond operator formulation introduced for coupled spin pair (or coupled spin dimer) systems [8, 9, 10, 11] and is applied also to anisotropic spin dimer systems [12]. The advantage of the generalized Holstein-Primakoff theory is that the bosonic Hamiltonian for excited states can be obtained easily compared to the bond operator formulation. Therefore, it is suitable for the complex coupled spin multiplets system Cu$_2$Fe$_2$Ge$_4$O$_{13}$. Details of the formulation will be published elsewhere.

3. Dynamical spin correlation function

We show dynamical correlation functions along the particular directions in reciprocal space in Fig. 2. As observed by the inelastic neutron scattering experiment, there are several low-energy branches below 10 meV [see Fig. 2(a)]. The dispersion relation is consistent with experiment
Figure 2. Dynamical spin correlation functions along the \((h,k,l)\) directions in reciprocal space in arbitrary unit. Both transverse and longitudinal intensities are shown. (a) Transverse intensity of Fe component. (b) Transverse intensity of Cu component. (c) Longitudinal intensity of Fe component. (d) Longitudinal intensity of Cu component.

Figures 2(a) and 2(b) are transverse intensities for Fe and Cu components, respectively. Since the low-energy modes have strong intensities for Fe, they are understood as spin wave excitations of the ordered moment of Fe\(^{3+}\). As seen in Fig. 2(b), these modes have very weak intensities also in Cu component due to the coupling between Fe\(^{3+}\) and Cu\(^{2+}\). There is another mode around 25 meV with very weak dispersion. There are both transverse and longitudinal spin fluctuations in the singlet-triplet excitations of the Cu dimer. In the ordered phase, they split into two. The mode at 25 meV corresponds to the transverse singlet-triplet excitations of the dimer.

Figures 2(c) and 2(d) are longitudinal intensities for Fe and Cu components, respectively. The low-energy modes below 10 meV have no longitudinal intensity. In contrast to this, there are high-energy modes with finite longitudinal intensities. For Fe, there are two modes around 18 meV and 33 meV [see Fig. 2(c)]. Since the ordered moment of Fe\(^{3+}\) is shrunk by the coupled dimers, the longitudinal spin fluctuation mode has a finite intensity [11, 13, 14]. Thus, these are understood as longitudinal excitation modes of the ordered moment that are peculiar to the coupled spin dimer and spin chain system. With the increase of the coupling constant between Fe\(^{3+}\) and Cu\(^{2+}\), we note that the longitudinal intensity becomes strong, since the ordered moment shrinks more strongly. For Cu, there are two modes around 27 meV and 34 meV [see Fig. 2(d)]. The mode at 27 meV corresponds to the longitudinal excitation mode of the Cu dimer, which is raised slightly compared to the transverse mode at 25 meV shown in Fig. 2(b).
4. Raman intensity

We next discuss Raman scattering in Cu$_2$Fe$_2$Ge$_4$O$_{13}$. In coupled spin dimer systems such as TlCuCl$_3$, Kuroe et al. found that the highest-lying excitation mode is Raman active in the field-induced ordered phase [15]. It is reported that the Raman-active mode is accompanied by longitudinal spin fluctuations in spin dimer systems [16]. In the same way, the longitudinal excitation modes can be detected by Raman scattering in Cu$_2$Fe$_2$Ge$_4$O$_{13}$. Figure 3 shows the field dependence of the Raman intensity. We can see three Raman-active modes at $H = 0$ T. They correspond to the longitudinal excitation modes at 18 meV, 27 meV and 33 meV [see Fig. 2(c) and 2(d)]. Under a magnetic field, the mode at 27 meV split into two. The energy difference between them is more than 1 meV at 16 T, which can be detected by high-field Raman scattering measurements. It is important to detect the energy shift to distinguish the magnetic Raman scattering from phonon scattering, where the latter does not change with field.

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