Triplon-spinon hybridization in Cu$_3$Mo$_2$O$_9$ observed using inelastic neutron scattering

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Abstract. We have measured the inelastic neutron scattering in Cu$_3$Mo$_2$O$_9$ to study its dispersion curves of magnetic excitations. This system has the spin chains interacting with the isolated spin dimers. Therefore, the magnetic excitations of this system can be described as the hybridized modes between the magnetic excitations of the spin chain (spinon excitations) and those of the spin dimers (triplon ones). We estimate the amplitude of the exchange interaction in the spin chain (4.0 meV) and that of the spin dimers (5.8 meV). The hybridization parameter, i.e., the effective interaction between the spin chain and the spin dimer is estimated to be $E_{\text{int}} \sin (\pi k)$, where $E_{\text{int}} = 1.6$ meV. The modulation of the molecular field generated by the spin precession in the spinon excited state plays an essential role in the spinon-triplon hybridization.

1. Introduction
Magnetic excitations from the antiferromagnetic (AF) ground state have been extensively studied. In the classical viewpoint, the spin excitations are described as the spin precession around its equilibrium direction. To describe the dispersion curve of magnetic excitations in uniform one-dimensional (1D) $S = 1/2$ AF system, the fermionic excitations are introduced through the Jordan-Wigner transformation [1]. The fundamental magnetic excitations of this system is the one-spinon mode known as the des Cloizeaux and Pearson (dCP) mode [2], and the two-spinon continuum. Hereafter we call this kind of magnetic excitation spinon.

In some magnetic systems with AF interactions, the ground state is nonmagnetic. The three-dimensional spin-dimer network ACuCl$_3$ ($A = K$ and/or Tl) [3] and the alternating 1D $S = 1/2$ AF system [4], which includes the spin-Peierls phase of CuGeO$_3$ [5], are good examples of the system with a nonmagnetic ground state. The dispersion relation of these systems can be understood using the bond-operator representation with the Holstein-Primakoff transformations [6, 7, 8]. The fundamental magnetic excitation is the $S = 1$ singlet-triplet one. Hereafter we call this kind of magnetic excitation triplon.

Recently, a complex of different spin subsystems has begun to be realized. The interactions between the triplon excitation and the spinon one or the spin-wave one, such as Cu$_2$Fe$_2$Ge$_4$O$_{13}$ [10], have received much attention.
In this work, we report that Cu$_3$Mo$_2$O$_9$ is the first material, to our knowledge, in which the spinon–triplon hybridization, i.e., the energy-level anticrossing between the spinon and the triplon excitations, is clearly observed. This compound crystalizes in an orthorhombic crystal structure (space group Pnma, lattice constants $a = 7.667$ Å, $b = 6.862$ Å and $c = 14.608$ Å, number of chemical units in the unit cell $Z = 4$) at room temperature [11]. Three kinds of crystallographically different Cu sites exist (Cu1, Cu2 and Cu3), as portrayed in Fig. 1. All Cu ions are divalent and have spin-1/2 [12, 13]. Considering the four shorter bonds among Cu$^{2+}$ ions, the spin system can be regarded as the chain of the distorted tetrahedron, as depicted in Fig. 1. The Hamiltonian of a distorted tetrahedral chain ($\mathcal{H}_{dt}$) can be divided into the 1D spin system ($\mathcal{H}_c$) and the spin dimers ($\mathcal{H}_d$), which are connected through the intersubsystem interaction term ($\mathcal{H}_{cd}$):

$$\mathcal{H}_{dt} = \mathcal{H}_c + \mathcal{H}_d + \mathcal{H}_{cd},$$

$$\mathcal{H}_c = \sum_j [J_1 S_{1,j} \cdot S_{1,j+1} + D_j \cdot (S_{1,j} \times S_{1,j+1})],$$

$$\mathcal{H}_d = \sum_j J_3 S_{2,j} \cdot S_{3,j},$$

$$\mathcal{H}_{cd} = \sum_j [J_1 (S_{1,j} + S_{1,j+1}) \cdot S_{2,j} + J_2 (S_{1,j} + S_{1,j+1}) \cdot S_{3,j}],$$

where $S_{i,j}$ ($i = 1–3$) denotes the $S = 1/2$ spin at the $j$th Cu site and we introduced the exchange interactions $J_i$ ($i = 1–4$) as well as Dzyaloshinskii–Moriya (DM) interactions between $S_{1,j}$ and $S_{1,j+1}$ with the Dzyaloshinskii vector $D_j$. Below $T_N = 7.9$ K, the spin moments at the Cu1 site have the following two components: the AF component along the $b$ axis which shows the long-range order [12], and the weak-ferromagnetic (WF) component in the $ac$ plane, which is generated by the DM interactions. Another phase transition point has been reported at $T_c$ ($\sim 2.5$ K) [12], below which the long-range order of the WF component appears. When $T_c < T < T_N$, the long-range order of the WF component appears only under the magnetic field.

2. Experiments and results

Inelastic neutron scattering measurements with the single crystal were conducted on the thermal neutron triple-axis spectrometer TAS-2 installed at JRR-3 at the Japan Atomic Energy Agency. The single crystal ($\phi 3 \times 30$ mm$^3$), which was made using an infrared imaging furnace, was cooled to around 4 K (between $T_c$ and $T_N$). The final neutron energy and the horizontal collimator sequence were fixed, respectively, at 14.7 meV and “guide-80′-sample-80′-open”. All measurements with several wavevectors $q = h \alpha^* + k \beta^* + l \epsilon^* \equiv (h, k, l)$, where $\alpha^*$, $\beta^*$ and $\epsilon^*$ were the primitive reciprocal lattice vectors, were conducted at a zero magnetic field. As shown in Fig. 2(a), the spectra can be fitted by one or two Gaussian curves on the linear background, as denoted by the solid curves. These peaks weakened in intensity with increasing temperature. Therefore, these come from magnetic excitations. In the inelastic-neutron spectra with wavevectors $(0, 1, l)$ ($l = 0, 0.5$ and $1$), the intensity around 2 meV decreases with increasing $l$. We observed the
tail of the peak generated by the lower-energy magnetic excitations. Further inelastic-neutron scattering measurements using another spectrometer clarify the detailed magnetic dispersion curves below 2 meV and the interactions between the distorted tetrahedral chains, which will be published elsewhere [14]. Although several spectra are not shown in Fig. 2(a), we obtained the dispersion curves of magnetic excitations in Fig. 2(b).

3. Discussion

Figure 2(b) depicts two branches, which are considered to reflect the two magnetic subsystems. Without the effects of \( J_1 \) and \( J_2 \) superexchange interactions and the interchain interactions [12], the two branches can be reproduced by the dispersion curve of the pure 1D spinons \( \hbar \omega_{dCP} = pJ_4 |\sin \pi k| \) [2], where \( p = \pi/2 \) is the renormalization factor, and that of the isolated spin dimers \( \hbar \omega_t = J_3 \). As described in the following, we need to consider the effects of \( J_1 \) and \( J_2 \) superexchange interactions and the interchain interactions to discuss the observed dispersion curves. Indeed, the dispersion curves along \([0k0]\) or \([0k1]\) are dispersive against the flat \( \hbar \omega_t \), which indicates some interaction working on the spin dimers.

Because the lower branch of the dispersion curves along \([01l]\) were observed in the energy region smaller than 2 meV [14], the interaction between the distorted tetrahedral chains is thought to be weak. And then we can apply \( \hbar \omega_s(h,k,l) \approx \hbar \omega_{dCP}(h,k,l) \) for the dispersion curves of the spinon excitations above 2 meV. More general formula based on the Holstein-Primakoff transformation [solid curve in Fig. 2(b)] will be published elsewhere [14]. From the
momentum-independent triplon energy along [01l], we obtained that $\hbar \omega_{l}(q) = J_{3} = 5.8$ meV, as represented by the dashed lines in Fig. 2.

We consider the hybridization effect which originates from the intersubsystem interactions $\mathcal{H}_{cd}$. The effects of $J_{1}$ and $J_{2}$ cannot be separated. Therefore, we phenomenologically introduce the interaction energy $E_{\text{int}}(q)$. The energy-level anticrossing can be obtained using the standard secular equation technique:

$$
\hbar \omega_{\pm}(q) = \frac{\hbar \omega_{s}(q) + \hbar \omega_{t}(q)}{2} \pm \sqrt{\left[\hbar \omega_{s}(q) - \hbar \omega_{t}(q)\right]^{2} + \left|2E_{\text{int}}(q)\right|^{2}}.
$$

(5)

The deviation between $\hbar \omega_{+}(q)$ and $\hbar \omega_{-}(q)$ increases with increasing $k$ of $(0, k, 0)$ or $(0, k, 1)$ from $k = 1$. Around the top of the spinon dispersion curve, $\hbar \omega_{l}(h, 1.5, l)$, the effect of the hybridization is strong because $\hbar \omega_{l}(h, 1.5, l) \sim \hbar \omega_{l}(h, 1.5, l)$. Meanwhile, finite $E_{\text{int}}(h, 0, l)$ results in the incommensurate magnetic ground state, which has not been observed in our measurements. One can easily confirm it by searching the condition $\hbar \omega_{-}(q) = 0$ in eq. (5). Consequently, it is natural to consider that $E_{\text{int}}(q)$ has a form of $E_{\text{int}} \sin(\pi k)$, meaning that two magnetic subsystems with $k = n$, where $n$ is an integer, are decoupled. When $E_{\text{int}} = 1.6$ meV and $J_{4} = 4.0$ meV, the overall observed dispersion curves are reproduced by $\hbar \omega_{\pm}(q)$, as shown by the solid curves in Fig. 2. We notice here that the maximum amplitude of $E_{\text{int}}(q)$ is smaller than $J_{3}$ and $J_{4}$, which indicates that it is adequate to treat $\mathcal{H}_{cd}$ as a perturbation and to use eq. (5) in calculating $\hbar \omega_{\pm}(q)$ above 2 meV. In our results, the spinon-triplon hybridization induces the energy-level anticrossing between the dispersion curves of $\hbar \omega_{s}(q)$ [dot-dashed curves in Fig. 2(b)] and $\hbar \omega_{t}(q)$ [dashed lines in Fig. 2(b)].

As an conclusive remark, we measured the inelastic-neutron scattering in Cu$_{3}$Mo$_{2}$O$_{9}$ to observe the magnetic excitation dispersion curve and to estimate the magnetic parameters. We observed the strong hybridization effect between the spinon and triplon excitations.

Acknowledgments

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