Zn-substitution effects on distorted tetrahedral spin-chain system $Cu_3Mo_2O_9$†

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The Zn-substitution effects on $Cu_3Mo_2O_9$ were studied. This compound has a quasi one-dimensional distorted tetrahedral spin system made of S = 1/2 Cu^{2+} ions.[1)] The multiferroic properties below the Néel temperature T_N = 8 K has been reported based on macroscopic measurements.[2)] The substituted Zn ions cut the magnetic chain directly and reduce the magnetic order. We have reported a novel magnetic ground state based on some macroscopic measurements for the heavily (5.0%) Zn-substituted sample.[3)] To obtain a microscopic viewpoint of the Zn-substitution effects on $Cu_3Mo_2O_9$, we measured muon spin rotation/relaxation spectra in $(Cu,Zn)_3Mo_2O_9$ with ARGUS spectrometer at Port 2. We prepared single crystals of lightly (0.5%) and heavily (5.0%) Zn substituted $Cu_3Mo_2O_9$ through continuous solid-state crystalization.[4)] The sliced single crystals are placed in the Variox cryostat with the ^{3}He sorption refrigerator. We measured the backward-forward asymmetry spectrum $A_{BF}(t)$ defined as

$$A_{BF}(t) = [A_B(t) - \alpha A_F(t)]/ [A_B(t) + \alpha A_F(t)] \quad , \qquad (1)$$

where $A_B(t)$ and $A_F(t)$ are the signal from the backward and the forward counters, respectively. A parameter $\alpha \sim 1$ is introduced to correct the small misalignment of the system. The signals from the muons stopping at the Ag foil on the crystals are removed using the comparison of $A_{BF}(t)$ under the transverse magnetic field of 20 G at temperatures below and above T_N. We found that approximately 75% of the implanted muons are stopped at the crystal.

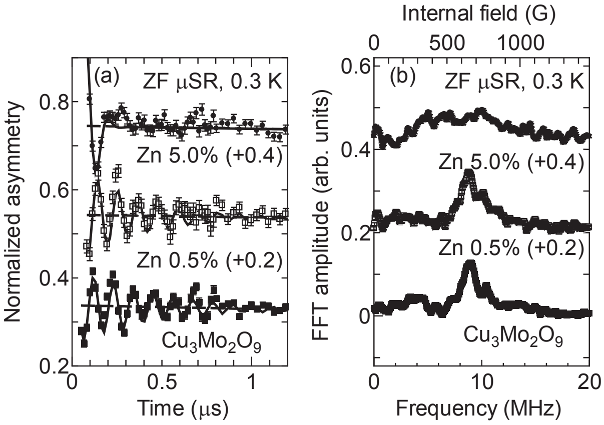

Fig. 1. Normalized asymmetry spectra at 0.3 K in $(Cu,Zn)_3Mo_2O_9$ in (a) and their fast Fourier transformation in (b). The upper scale in (b) denotes the internal field working on the muon stopping site(s).

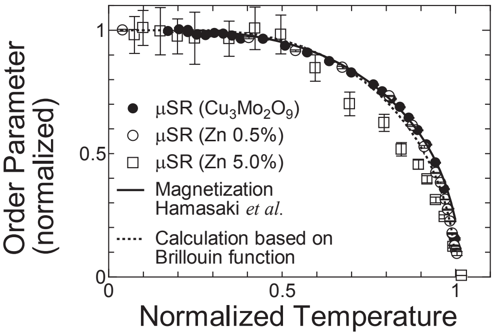

Fig. 2. Normalized internal fields in $(Cu,Zn)_3Mo_2O_9$, the magnetization taken from ref. 1, and the saturation magnetization calculated based on the Brillouin function as functions of the temperature normalized by T_N.

Figures 1(a) and 1(b) show the μSR time spectra and their fast Fourier transformations, respectively. In pure $Cu_3Mo_2O_9$ and the Zn-0.5% sample, the signals are very similar, indicating the same magnetic ground states. The oscillation frequencies of the μSR time spectra in Fig. 1(a) correspond to the dominating components in the frequency-domain spectra of Fig. 1(b) due to the muon precession around the internal field of 650 G. The beat on the oscillating spectrum at approximately 0.7 μs in Fig. 1(a) and the weak peak at 750 G in Fig. 1(b) indicate the two kinds of internal magnetic fields. In the Zn-5.0% sample, the rapidly decaying oscillation in the time-domain spectrum and the widely distributed frequency-domain spectrum were observed as shown in Figs. 1(a) and 1(b), respectively. We conclude that the magnetic ground state of the Zn-5.0% sample is different from the ones in pure $Cu_3Mo_2O_9$ and the Zn-0.5% sample.

In Fig. 2, we show the normalized amplitudes of the dominating internal field in $Cu_3Mo_2O_9$ and the Zn-0.5% sample and that of the averaged internal field in the Zn-5.0% sample as functions of temperature normalized by T_N. These normalized amplitudes have similar temperature dependences with the normalized magnetization because of the weak ferromagnetic component of the spin moment in pure $Cu_3Mo_2O_9$[1)] as well as the temperature variation of the saturation magnetization in a ferromagnet calculated based on the Brillouin function. These facts indicate that the order parameter of this multiferroic phase transition is the sublattice magnetization.

References

1) T. Hamasaki *et al.*: Phys. Rev. B 77, 134419 (2008).
2) H. Kuroe *et al.*: J. Phys. Soc. Jpn. 80, 083705 (2011).
3) H. Kuroe *et al.*: J. Kor. Phys. Soc. 63, 542 (2013).
4) K. Oka *et al.*: J. Cryst. Growth 334, 108 (2011).

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† Condensed from the article in JPS Conf. Proc. **2**, 010206 (2014)

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