

## Magnetic structure of Spin $S=1/2$ linear trimer system $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$

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Low dimensional quantum spin systems attract much attention. In particular, the frustrated quantum spin systems due to geometrical arrangement or competing interactions are expected to exhibit various interesting properties.  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$  has a  $\text{Cu}_3\text{O}_8$  trimer formed of edge-sharing three  $\text{CuO}_4$  square planes [1]. For  $\text{Cu}^{2+}$  spins ( $S=1/2$ ) within the  $\text{Cu}_3\text{O}_8$  trimer, the second-neighbor exchange interaction  $J_2$  is antiferromagnetic ( $J_2 > 0$ ), and the nearest-neighbor exchange interaction  $J_1$  is weak ferromagnetic or antiferromagnetic (AF). Such competing interactions between  $J_1$  and  $J_2$  can lead to novel quantum magnetic phenomena.

The T-dependence of magnetic susceptibility of  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$  in the T-region  $70 \text{ K} < T < 650 \text{ K}$  can be explained by the isolated  $S = 1/2$  Heisenberg trimer model, and it is obtained  $J_2/k_B = 340 \pm 20 \text{ K}$  (AF), and  $J_1/k_B = 30 \pm 20 \text{ K}$ . [2] On the other hand, T-dependence of of  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$  in the T-region  $4 \text{ K} < T < 70 \text{ K}$  can be explained by the  $S = 1/2$  uniform Heisenberg chain model called as Bonner-Fisher model [3], and it is obtained  $J_3/k_B = 18 \pm 1 \text{ K}$  where  $J_3$  is an inter-trimer exchange interaction.[2] The ground state of the isolated  $S = 1/2$  trimer is found that two spins of the edge in the  $\text{Cu}_3\text{O}_8$  trimers form a nonmagnetic singlet state by strong AF interaction  $J_2$ . The center spin of the  $\text{Cu}_3\text{O}_8$  trimer only survives in low T-region  $T < 70 \text{ K}$ . The behavior of the specific heat in the T-region  $4 \text{ K} < T < 70 \text{ K}$  can also be explained by the  $S = 1/2$  uniform Heisenberg chain model. As results of measurements of magnetic susceptibility, specific heat, and dielectric constant, the center spin of the  $\text{Cu}_3\text{O}_8$  trimer of  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$  exhibits an antiferromagnetic transition at  $T_N = 2 \text{ K}$  accompanied with a ferroelectricity (called multiferroic phenomenon). From the detailed magneti-

zation measurements, we found the existence of  $dM/dH$  anomaly at  $H_c = 0.37 \text{ T}$  at  $T = 1.9 \text{ K}$  ( $< T_N$ ). It is indicating that the magnetic structure changes at  $H_c$ . The determination of the magnetic structure at  $H = 0$  and  $H = 1 \text{ T}$  ( $> H_c$ ) give us important information to understand the magnetic behavior of the  $S = 1/2$  linear trimer system.

We investigated the magnetic structure of  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$  below  $T_N$  through powder neutron diffraction experiments using cold neutron powder diffractometer DMC at PSI. We used amount of 15 g of powder for  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$ . The superconducting magnet and the dilution refrigerator were used to reach down to 0.1 K and up to 1 T.

We obtained powder neutron diffraction patterns at  $T = 0.1 \text{ K}$ , 3K, and 12 K, respectively, in  $H = 0$  as well as that of  $T = 0.1 \text{ K}$  applied the magnetic field  $H = 1 \text{ T}$ . The figure (a) show the diffraction patterns at  $T=0.1 \text{ K}$  and 12 K, respectively. We can clearly observe the super-lattice magnetic Bragg peaks below  $T_N$  assigned by the arrows. The figure (b) show the intensity profile of difference between at  $T = 0.1 \text{ K}$  and 12 K,  $I(0.1\text{K}) - I(12\text{K})$ . We also found the possible magnetic Bragg peaks assigned by the short arrows. By applied the magnetic field  $H = 1 \text{ T}$ , no difference found the magnetic Bragg intensities at  $T = 0.1 \text{ K}$  in the error bar. Then, changing the magnetic structure at  $H_c$  seems to be small. Recently, the aligned powder of  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$  can be obtained in the magnetic field  $H = 9 \text{ T}$ . For determination of the magnetic structure of  $\text{Na}_2\text{Cu}_3\text{Ge}_4\text{O}_{12}$ , the results of the various measurements of the aligned powder give us important information. We are analyzing the neutron diffraction data and various measurements data.

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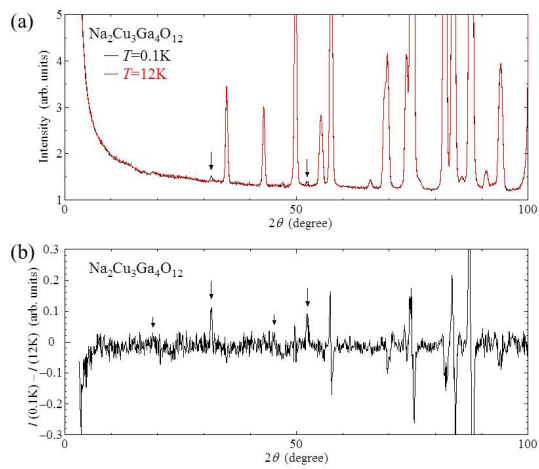


Fig. 1. (a). Neutron powder diffraction patterns measured at 0.1 K and 12 K. The arrows indicate the magnetic Bragg peaks. (b). Intensity profile of difference between at  $T = 0.1\text{ K}$  and 12 K.