Magnetic ground state of Cu_6O_8MCl (M = Y, Pb) with a caged structure

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Cu_6O_8MCl (M=cation) compound has a Cu_6O_8 cage which forms a three-dimensional Cu-O network by connecting their faces in its crystal structure [1]. The formal Cu valence in the Cu_6O_8 cage is +2.15 for M = Pb^{4+} and +2.33 for M = Y^{3+}, suggesting the existence of Cu^+ ($3d^{10}$), Cu^{2+} ($3d^9$) with S = 1/2 spin, and Cu^{3+} ($3d^8$) [2]. If there is partial existence of S = 1/2 spins on the Cu site in the Cu_6O_8 cage, the static magnetic ordered state is expected in the square-lattice and the dynamical spin fluctuation in the triangular-lattice i.e., the magnetic competition state is expected in the magnetic ground state of Cu_6O_8MCl. To elucidate the detailed physical properties of Cu_6O_8MCl, we focused on clarifying the magnetic ground states of Cu_6O_8PbCl, which is the semiconducting material, and compared the observed data with the based material of Cu_6O_8YCl, which is the metallic compound with paramagnetic behavior.

μSR experiments were performed at the RIKEN-RAL Muon facility at the Rutherford-Appleton Laboratory, UK. Fig. 1 shows the zero field (ZF) μSR spectra of Cu_6O_8MCl (M=Y, Pb) at various temperatures. With the decrease in the temperature, the initial asymmetry of Cu_6O_8PbCl rapidly decreased below 20 K (Fig. 1(b)). On the other hand, clear decreasing behavior of the initial asymmetry was not observed in the ZF-μSR spectra of Cu_6O_8YCl down to 0.3 K, indicating that there is no magnetic ordered state in this system (Fig. 1(a)). The ZF-μSR spectra in Fig. 1 were analyzed using the following function,

$$P(t) = A\exp(-\lambda t)G_{KT}(\Delta,t)+A_B \quad (1),$$

where A is the initial asymmetry at t = 0, λ is relaxation ratio of the muon spin, and A_B is the background signal. $G_{KT}(\Delta,t)$ is the static Kubo-Toyabe function with a half-width of Δ, describing the distribution of the nuclear-dipole field at the muon site [3]. Results of the best-fit of eq. 1 are indicated by the solid line in Fig. 1, and the observed adjusted parameters A, λ, and Δ of Cu_6O_8MCl (M=Y, Pb) as functions of temperature are shown in Fig. 2. A (a-relaxing) of Cu_6O_8YCl slightly decreases with the decrease in the temperature (Fig. 2), whereas λ and Δ of Cu_6O_8YCl are almost constant, being temperature independent. These facts indicate that there is no change of spin dynamic and long range magnetic ordered state in Cu_6O_8YCl, which is a metallic compound with paramagnetic behavior. For Cu_6O_8PbCl, the temperature dependence of a-relaxing, λ, and Δ change below 20 K, indicating the change in the magnetic spin state (Fig. 2). However, clear precession signal is not confirmed in the ZF-μSR spectra below 20 K. The Cu_6O_8 cage has a square-lattice and triangular-lattice on its surface, and the Cu sites in the Cu_6O_8 cage are occupied by various valences of Cu^+, Cu^{2+}, and Cu^{3+} [2]. These conditions encumber the formation of the completely static magnetic ordered state in Cu_6O_8PbCl. The observed behavior of ZF-μSR spectra and Fig. 2 data of Cu_6O_8PbCl indicate the growth of the short-range magnetic interaction between S = 1/2 spins below 20 K. Consequently, the magnetic ground state of Cu_6O_8PbCl does not have a static long range magnetic ordered state such as an antiferromagnetic state in high-T_c cuprate. There is possibility that the short range interaction of Cu_6O_8PbCl forms the spin glass state below 20 K like under-doping material in high-T_c cuprate. The magnetic ground state of Cu_6O_8MCl compound depends on the valence state of the M site ion.

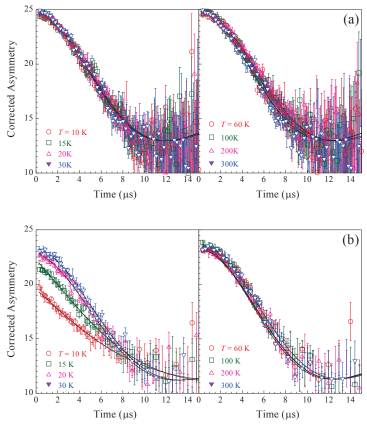

Fig. 1 ZF-μSR time spectra of Cu_6O_8MCl ((a) M = Y, (b) M = Pb) at various temperatures. Solid lines indicate the fitting results of eq. (1).

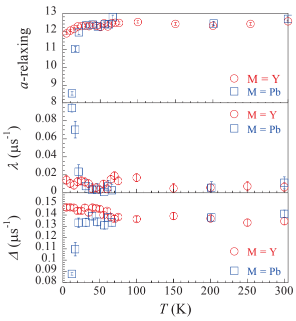

Fig. 2 Temperature dependence of the initial asymmetry A (a-relaxing), relaxation ratio λ, and Δ of Cu_6O_8MCl (M = Y, Pb) defined by the results of fitting for the Fig. 1 data.

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References

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