

# Magnetic ordering in $\text{Cu}_6\text{O}_8\text{TbCl}$ probed during the $\mu\text{SR}$ measurement

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The copper-oxide family of  $\text{Cu}_6\text{O}_8\text{MCl}$  ( $M = \text{cation}$ ) has a caged structure ( $\text{Cu}_6\text{O}_8$  cage) in its crystal structure (crystal system: cubic, space group:  $Fm-3m$  (No. 225)). The form of the surface of the  $\text{Cu}_6\text{O}_8$  cage is similar to the  $\text{CuO}_2$  plane in high- $T_c$  cuprate superconductors. The  $\text{Cu}_6\text{O}_8$  cage forms a three-dimensional network, sharing their face (left panel of Fig. 1).<sup>1-3</sup> The  $\text{Cl}^-$  anions are located at the center of the  $\text{Cu}_6\text{O}_8$  cage, and the  $M$  cations, which have a valence of  $3+$  or  $4+$ , exist in the cuboid space located between  $\text{Cu}_6\text{O}_8$  cages. The average valence of  $\text{Cu}$  ions in the  $\text{Cu}_6\text{O}_8$  cage is  $2.33+$  for  $M^{3+}$ .

For the  $M = \text{Tb}^{3+}$  compound ( $\text{Cu}_6\text{O}_8\text{TbCl}$ ), the magnetic susceptibility data indicate a weak magnetic transition at approximately 40 K. Moreover, the electrical resistivity data exhibit a metal-insulator transition at approximately 40 K (right panel of Fig. 1). The effective Bohr magneton  $P_{\text{eff}}$  is estimated to be  $\sim 6.74$ , which is close to the theoretical value for  $\text{Tb}^{3+}$ , indicating that a localized magnetic moment exists at the  $\text{Tb}^{3+}$  site. However, whether the origin of the magnetic ordering state is long range (static) or not and the relationship between the magnetic state and other physical properties is not yet clear. In order to clarify these points,  $\mu\text{SR}$  measurement was performed at the RIKEN-RAL Muon Facility in U.K. using a polycrystalline sample of  $\text{Cu}_6\text{O}_8\text{TbCl}$ .

Figure 2 (a) shows the time dependence of asymmetry of the zero field (ZF)  $\mu\text{SR}$  spectra,  $A(t)$ , of  $\text{Cu}_6\text{O}_8\text{TbCl}$  measured at various temperatures. These spectra were observed using the double-pulse muon beam. The spectra show exponential-like depolarized behaviors, and the muon-spin depolarization becomes faster with decreasing temperature above 40 K. Significant loss of initial

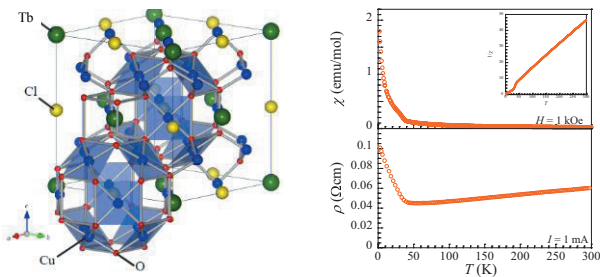


Fig. 1 (Left panel) Crystal structure of  $\text{Cu}_6\text{O}_8\text{MCl}$  (program VESTA was used.<sup>4</sup>) Solid line shows the unit cell. (Right panel) Temperature dependence of magnetic susceptibility and electrical resistivity of  $\text{Cu}_6\text{O}_8\text{TbCl}$ . Inset shows the inverted magnetic susceptibility as a function of temperature.

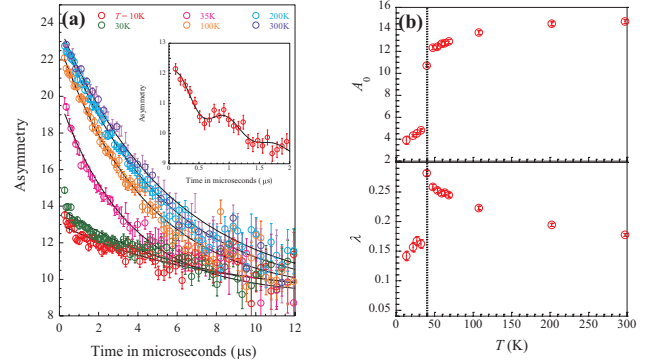


Fig. 2 (a) The time dependence of asymmetry of ZF  $\mu\text{SR}$  spectra of  $\text{Cu}_6\text{O}_8\text{TbCl}$  measured at various temperatures. Solid lines are fitting results using Eq. (1). (b) The temperature dependence of  $A_0$  and  $\lambda$  obtained by the fitting.

asymmetry (at  $t = 0$ ) is observed below 40 K. In order to observe the change of the spin dynamics in detail, all of the time spectra were analyzed using the following function:

$$A(t) = A_0 \exp(-\lambda t) + A_B, \quad (1)$$

where  $A_0$  is the initial asymmetry,  $\lambda$  is the depolarization rate of the muon spins, and  $A_B$  is the background. The parameters obtained from the best fit of Eq. 1 to the data in Fig. 2(a) (solid line) are shown in Fig. 2(b). From Fig. 2(b),  $A_0$  slightly decreases and  $\lambda$  increases above 40 K. Moreover,  $A_0$  and  $\lambda$  values rapidly change at approximately 40 K, indicating that the spin state drastically changes at 40 K. The inset of Fig. 2(a) shows an early time region of the ZF  $\mu\text{SR}$  spectra at 5 K (lowest temperature of this measurement) measured using the single-pulse muon beam. Muon-spin precession behavior was observed, indicating the existence of a long-range magnetic-ordered state. Consequently, the weak magnetic transition confirmed in the magnetic susceptibility data of  $\text{Cu}_6\text{O}_8\text{TbCl}$  is generated by the development of long-range magnetic order of  $\text{Tb}^{3+}$ . Moreover, it is suggested that the conducting carrier is trapped and that the ground state changes from metallic to semiconducting with the onset of long-range magnetic order.

## References

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