MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2016B0129 実験課題名 Title of experiment μSR study on the single crystal of S = 1/2 kagome lattice antiferromagnet $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ 実験責任者名 Name of principal investigator Hiroyuki Yoshida 所属 Affiliation Hokkaido University	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/（BL No.） μSR spectrometer at D1 channel 実施日 Date of Experiment 2017, 2/16-19

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Single crystals of Ca-Kapellasite $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

Our study focused on the magnetic properties of S = 1/2 Kagome lattice antiferromagnet Ca-Kapellasite $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$. In the previous bulk magnetic measurements, we revealed that the Ca-Kapellasite possessed competing magnetic interactions J_1 = K (AF), J_d = K (AF), and J_2 = K (F). The magnetic susceptibility and the heat capacity measurements indicated that an unusual magnetic transition at T^* = 7.2 K where the χ_{ab} showed the cusp and the heat capacity exhibited the tiny peak anomaly. Remarkably, the temperature-linear (T-linear) term 5.9 mJ/CumolK^2 was observed in the heat capacity, in spite of the compound is an insulator. This shows that the unconventional magnetic excitation underlies the ground state of the Ca-Kapellasite, and suggesting the realization of the fluctuating ground state.

We carried out the μSR experiments on single crystals of Ca-Kapellasite using the μSR spectrometer at D1 channel in MLF. Hundreds of single crystals were aligned on the Ag sample holder as shown in Fig. 1. In this geometry, the incident muon spin direction is parallel to the c-axis of the crystal. In order to examine the slow spin fluctuation, we used the dilution refrigerator and the measurements were conducted at various temperatures, T = 0.082, 0.15, 0.3, 0.5, 0.7, 1, 2, 3, 4, 6.75, 11.5, 15 K. The zero-field (ZF)-μSR were taken at each temperature

and the longitudinal-field(LF)-μSR were taken at T = 0.082, 0.7, 2 K up to magnetic field of 2000 Oe. Figure 2a shows the ZF-μSR spectra at several temperatures. We did not observe a clear oscillation of the spectrum even at the lowest temperature which is smaller than the transition temperature T^* by two orders of magnitude. The characteristic feature of the spectral shape is the duality of the rapid and slow relaxation components. In order to obtain the relaxation rate from these spectra, we fitted these data by assuming two exponential ingredients and constant background contribution. Figure 3 summarized the temperature dependence of relaxation rates λ_1 and λ_2. The λ_1 and λ_2 become almost temperature independent below around 2 K which suggests that a dynamical spin component may exist down to the lowest temperature (0.082 K). The field dependence of LF-μSR spectra is shown in Fig. 2b. The spectrum was shifted to increase the asymmetry by applying the magnetic fields indicating the internal field of Ca-Kapellasite below T^* was decoupled by the longitudinal magnetic fields. The field dependence of the total asymmetry (A_1+A_2) is plotted in Fig. 4. There is an inflection point around 350 Oe which is consistent with a roughly estimated internal field from the value of λ_1 (~ 30 μs^{-1}) at the lowest temperature. Furthermore the slow relaxation component, which is related to spin fluctuation, seems to exist even well above the longitudinal field of 350 Oe. This implies that the ground state of Ca-Kapellasite contains a fluctuating spin component such as spin-liquid states. This spin fluctuation may also relate to the unusual magnetic excitation observed in the heat capacity. Although the magnetic state of Ca-Kapellasite has not been completely understood so far, the ground state of Ca-Kapelasite is not a conventional one, but should be an exotic state owing to the strong frustration and the quantum fluctuation on the Kagome lattice.

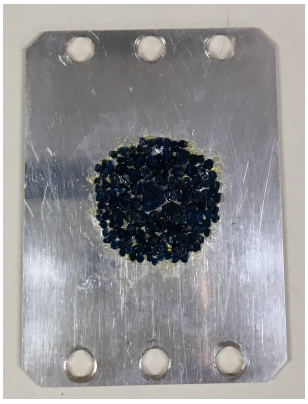

Fig. 1 Single crystals of Ca-Kapellasite

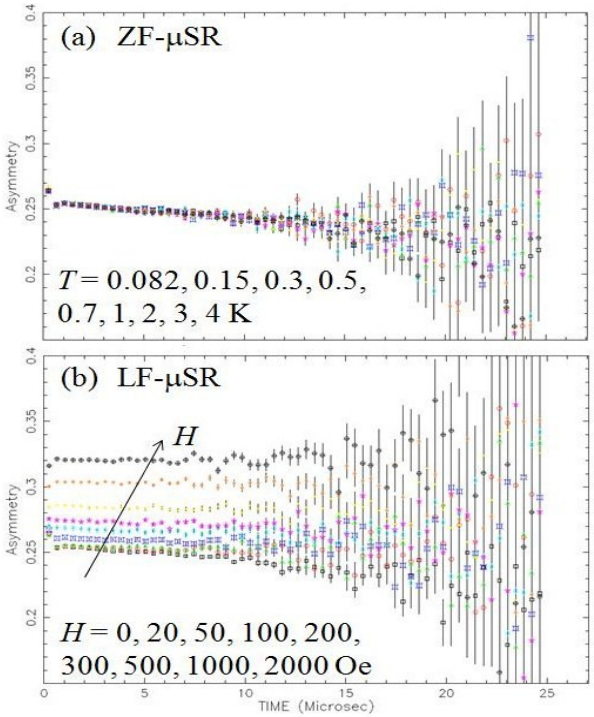

Fig. 2 (a) ZF-μSR and (b) LF-μSR spectra.

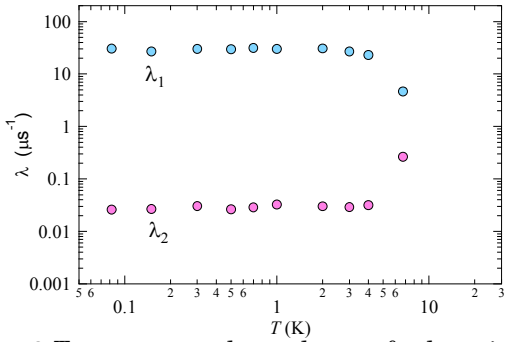

Fig. 3 Temperature dependence of relaxation rates.

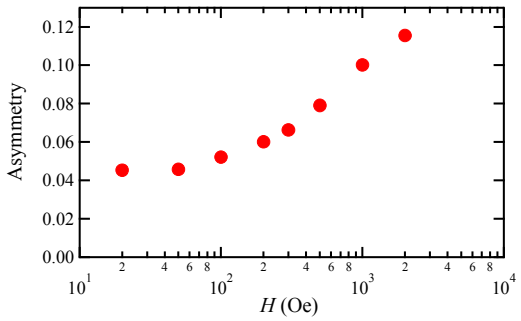

Fig. 4 Field dependence of asymmetry.