実験報告書様式（一般利用課題・成果公開利用）

（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。）

MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2015A0314 実験課題名 Title of experiment Investigation of the magnetic ground state in the edge-sharing spin tetrahedra system $K_2Cu_3O(SO_4)_3$ 実験責任者名 Name of principal investigator Masayoshi Fujihara 所属 Affiliation Tokyo University of Science	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D1 実施日 Date of Experiment 2016 2/20 – 22

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.
$K_2Cu_3O(SO_4)_3$ and $K_3Cu_3AlO_2(SO_4)_4$ To investigate the spin dynamics in $K_2Cu_3O(SO_4)_3$, we performed μSR measurement at 1D area of J-PARC MUSE. However, unfortunately this experiment ended in failure. Then, we performed the experiment on $K_3Cu_3AlO_2(SO_4)_4$.

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.

The diamond chain system is one of the low dimensional quantum spin systems, in which diamond shaped units of S = 1/2 compose chains. In spite of its simple structure, low dimensionality, quantum effect and geometrical frustration within a chain make this system very unique and exotic ground state has been studied theoretically [K. Okamoto *et al.,* J. Phys.: Condens. Matter 11 (1999) 10485-10498.]. Recently, we reported the interesting magnetic properties in the new highly one-dimensional S = 1/2 scalene diamond chain compound $K_3Cu_3AlO_2(SO_4)_4$ [M. Fujihala *et al.,* J. Phys. Soc. Jpn. 84 (2015) 073702]. $K_3Cu_3AlO_2(SO_4)_4$ contains the Cu^{2+} ions in a scalene diamond arrangement along the b-axis direction. Nonmagnetic potassium and aluminum ions are located in the inter-chain space, thus resulting in a long inter-chain distance. Therefore, magnetic long-range ordering is absent down to at least 0.5 K. As shown in Fig 2(a), the characteristic two broad peaks are observed in the temperature dependence of magnetic susceptibility around T_{HM} ~ 200 K and T_{LM} ~ 50 K. Similar χ(T) behavior has been observed in Azurite [H. Kikuchi et al., Phys. Rev. Lett. 94 (2005) 0227201(1-4) and many other works.], as two-stage development of short range correlation of dimers and monomers. In order to investigate spin dynamics in this unique spin liquid state, we conducted muon spin rotation and relaxation (μSR) measurement at D1 area of J-PARC MUSE. We performed zero field (ZF) and longitudinal field (LF) experiment on polycrystalline sample of $K_3Cu_3AlO_2(SO_4)_4$ at 5 ~ 300K. Figure 1 (a) is the asymmetry spectra of ZF-μSR measurement. We use following form for the fitting analysis $A(t) = A_0 \exp[-(\lambda t)^{\beta}]$, where λ is depolarization rate of muon spin, A_0 is the initial asymmetry, and β is a stretch parameter and β = 1.66.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

The blue and red circles in Fig.2 (a) shows the temperature dependence of λ and A_0 obtained from fitting the ZF-μSR data. And light colored arrows in Fig.2 (a) are guide to eyes. From these results, it is clear that long range ordering is absent. We will discuss the behavior of λ and A_0 considering the magnetic susceptibility in Fig.2 (b). In the temperature dependence of λ, there is characteristic increase from 200 K to 5 K. In this temperature range, since thermal fluctuation is reduced, we consider muon spins detect electron spin fluctuation. Short range correlation between monomers develops as the temperature goes down. Therefore, this increase in λ indicates fluctuation of monomer spins. Above 100 K, the behavior of A_0 looks flat compared to that of λ. More characteristic change is the reduction of A_0 below 100 K, which Fig.1 (a) and Fig.2 (a) exhibit. This is attributed to fast depolarization of muon spins observed as disappearance of asymmetry due to the limited time resolution (~ 0.1 μs). Considering $\chi(T)$ behavior, the correlation between monomers develops around 50 K. Thus we consider fluctuation of monomer spins contributes to fast depolarization of muon spins. Fig.1 (b) shows LF-μSR asymmetry spectra at 5 K fitted by the same form as that of ZF-μSR. It is clear that spectra at 10 G and 1000 G are almost the same, which shows small field 10 G decoupled muon spin relaxation. This result seems strange because results of ZF-μSR indicates presence of distributed internal field which realizes fast depolarization of muon spins, and such field cannot seems as weak as 10 G. Otherwise, different way of looking this result is that, this internal field could be very sensitive to external field. More discussion and investigation are still needed for fully understanding of these results. This experiment was the first challenge of μSR investigation of dimer-monomer spin liquid state ground state in the distorted diamond chain system. Combining results of previous experiments, we conclude that we were successful to observe the spin dynamics which changes with the development of short-range correlation of alternating dimer-monomer spin liquid state. We believe more detail of the spin dynamics in quantum spin liquid state in ideal one dimensional S = 1/2 antiferromagnet will be revealed in the next experiment on $K_3Cu_3AlO_2(SO_4)_4$.

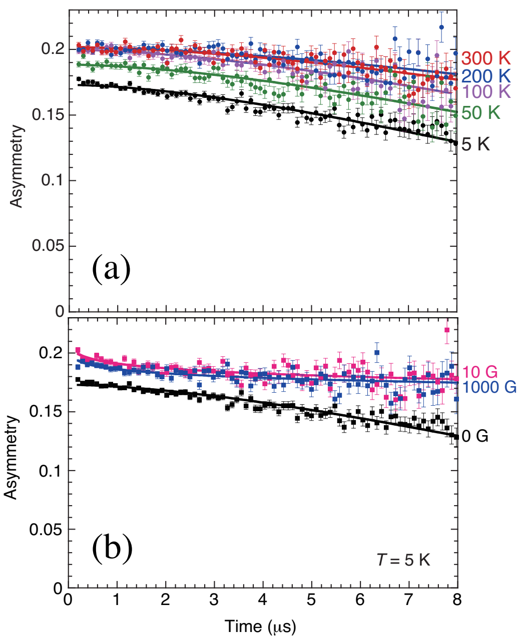

Fig. 1 (a) Zero-field muon spin asymmetry spectra and (b) longitudinal field muon spin asymmetry spectra of K3Cu3AlO2(SO4)4. Measured temperatures and fields are written in Figures.

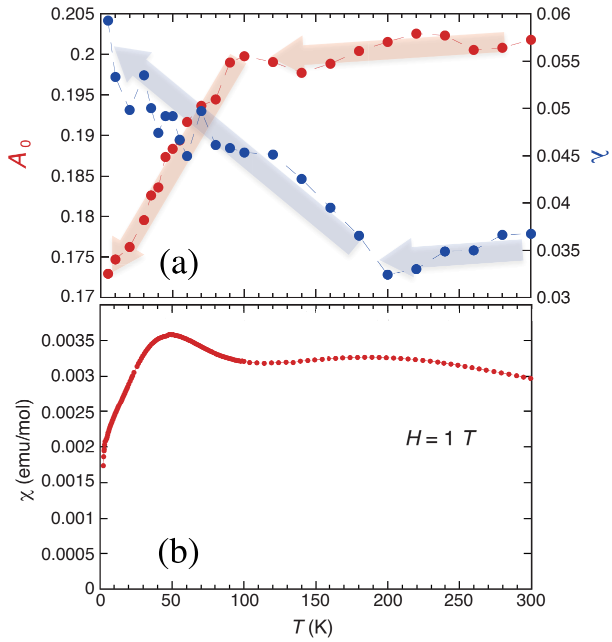

Fig. 2 (a) Temperature dependence of muon spin relaxation (blue circles) and initial asymmetry (red circles) of K3Cu3AlO2(SO4)4. (b) Temperature dependence of magnetic susceptibility K3Cu3AlO2(SO4)4.