MLF Experimental Report

課題番号 Project No. 2017A0013 実験課題名 Title of experiment Spin dynamics in the quantum spin liquid state of $CaCu_3(OH)_6Cl_2$ $0.6H_2O$ 実験責任者名 Name of principal investigator Kazuki Iida 所属 Affiliation CROSS	提出日 Date of Report 装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/（BL No.） AMATERAS (BL14) 実施日 Date of Experiment 2017/6/8 – 2017/6/12

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.

Co-aligned single-crystal $CaCu_3(OD)_6Cl_2$ $0.6D_2O$.

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.

Ground state of the spin-1/2 perfect kagome lattice is believed to be a quantum spin liquid (QSL). It is of great interest whether or not intrinsic spin gap exists in the QSL state [S. Yan *et al*., Science **332**, 1173 (2011)], which can provide us strong constrain to construct the QSL theory. So far, all reported spin-1/2 perfect kagome-lattice compounds have impurity (*i.e.*, atomic site randomness between Cu^{2+} and nonmagnetic ions), which prevents us to investigate the existence of spin gap because impurity gives rise to low-energy scattering [T.-H. Han *et al*., PRB **94**, 201111 (2016).]. To discuss the spin gap problem in the spin-1/2 kagome lattice, we performed single-crystal inelastic neutron scattering measurements on newly discovered spin-1/2 kagome compound $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$ using AMATERAS.

We prepared co-aligned deuterated single crystals [about 400 pieces, see Fig. 1(a)], which were well aligned confirmed by the diffraction pattern: we can clearly see the six-fold symmetry as shown in Fig. 1(b). The mosaic of the co-aligned sample was 4.3° (FWHM). The sample holder was attached to the ^3He cryostat. Measurements were performed at 0.3 and 40 K. Set of E_is of 12.5, 5.2, and 2.9 meV were selected. In this report, we only show the data with E_i = 12.5 meV.

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

Figure 2(a) shows the constant energy map in $CaCu_3(OD)_6Cl_2{\cdot}0.6D_2O$ at 0.3 K. Interestingly, this constant energy map can be reproduced quantitatively assuming the singlet-to-triplet excitations on the kagome lattice (not shown). We also show the dispersion relation along *H* and *HH* as shown in Figs. 2(b) and 2(c). The broad continuum excitation was observed. These two dispersion relations are the direct evidence of the fractionalized excitation, or the spinon continuum, in the spin-1/2 kagome lattice. Thus, our observation elucidated that the fractionalized excitation really exists in the spin-1/2 kagome lattice in the absence of chemical disorder.

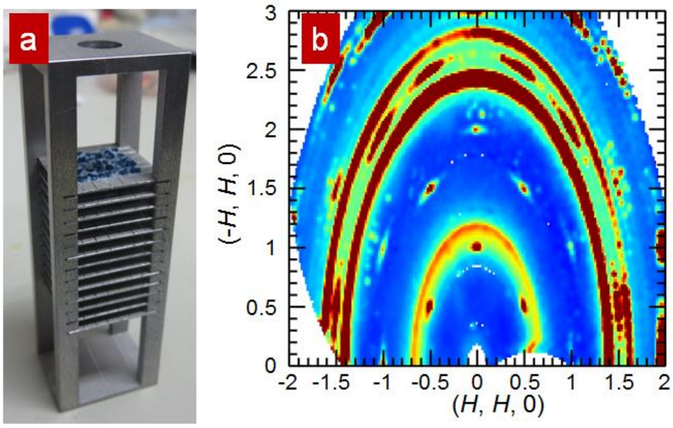

Fig. 1. (a) Co-aligned $CaCu_3(OD)_6Cl_2$ $0.6D_2O$. (b) Diffraction pattern at 0.3 K. *L* and $\hbar\omega$ were integrated in [-0.7, 0.7] and [-0.1, 0.1] meV, respectively.

Furthermore, we have succeeded in observing the gapped magnetic excitation using lower E_i (not shown). Therefore, our single-crystal inelastic neutron scattering measurements show the gapped fractionalized excitation in the QSL state in the spin-1/2 perfect kagome $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$.

Finally, we would like to thank our local contacts Dr. Kenji Nakajima and Dr. Seiko Ohira-Kawamura for their nice support on the experiment. We also appreciate the staff of the MLF SE team.

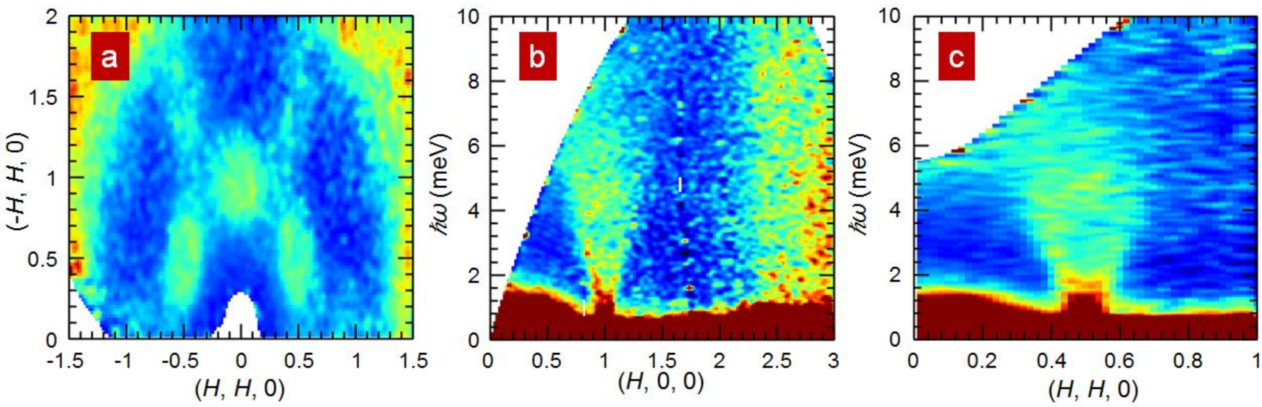

Fig. 2. (a) Constant energy map in $CaCu_3(OD)_6Cl_2$ $0.6D_2O$ at 0.3 K. *L* and $\hbar\omega$ were integrated in [-0.7, 0.7] and [2.5, 5.5] meV. (b) Dispersion along (b) *H* and (c) *HH*. *L* was integrated in [-0.7, 0.7]. (b) *HH* was integrated in [-0.1, 0.1], and (c) *H* was integrated in [0.4, 0.6], respectively.