MLF Experimental Report	提出日 Date of Report 2018. 3. 13.
課題番号 Project No. 2017A0027 実験課題名 Title of experiment Dynamical structure factors on S=1/2 triangular antiferromagnetic system with bond randomness $Cs2CuCl_{4-x}Br_x$ 実験責任者名 Name of principal investigator Toshio Ono 所属 Affiliation Osaka Prefecture University	装置責任者 Name of responsible person Dr. Kenji Nakajima 装置名 Name of Instrument/(BL No.) BL 14 実施日 Date of Experiment 2017. 11. 10 – 2017. 11. 13.

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.

Cs_2CuCl_4 and Cs_2CuBr_4 are isomorphic and are spatially anisotropic S=1/2 triangular antiferromagnets. We are investigating the effects of the bond randomness on the triangular antiferromagnet using the mixture system $Cs_2CuCl_{4-x}Br_x$. On the present experiment, we have performed the inelastic neutron scattering for the sample with x=3.4 which has antiferromagnetically ordered ground state. Three single crystals those were grown by slow evaporation method were co-aligned so as to make the bc-plane is parallel to the scattering plane. The total mass of the crystals used was 14g and a mosaic of the crystal is approximately ~1˚.

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.

Three co-aligned single crystals were cooled down to 0.3K using ^{3}He refrigerator. In order to correct the four-dimensional scattering intensity $I(\boldsymbol{Q},\omega)$, crystals were rotated 150˚ in the scattering plane with acquiring multiple datasets. Measurements were also performed at T = 10K to check the temperature dependence of $I(\boldsymbol{Q},\omega)$. Incident neutron energies were chosen as 3.0, 5.6 and 13.7 meV using the multiple E_i chopper system [1].
Figure 1 shows the static structure foctor as a function of h and k. The intensity is energy integrated for $\hbar\omega$= [-0.1, 0.1] meV. Measurements were carried out at 10 K (Fig. 1 (a)) and 0.3 K (Fig. 1 (b)). Antiferromagnetic phase transition was observed at T = 0.6 K on the magnetic measurement for the present sample. So that the reflections observed only at T = 0.3 K indicated by the black triangles should correspond to the magnetic Bragg peaks. In addition to the nuclear Bragg reflections expected for end member, such as h + k = even, weak reflection indicated by white triangle was observed at Q = (0, 1, 0). The violation of the extinction rule might be caused by random substitution of halogen ions. The position of

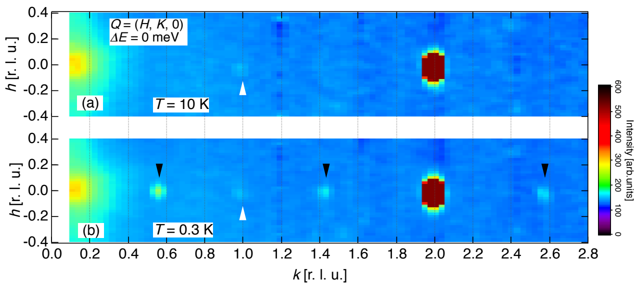

Figure 1 Static structure factors in [hk0] plane measured at T = 10K (a) and 0.3K (b).

observed magnetic Bragg reflection was $Q = (0.\ q, 0)$ with $q = 0.56(1)$. Within a classical treatment, the relationship between q and the ratio of exchange interactions $\alpha = J_2 / J_1$ are described as $\pi q = \arccos(-\alpha/2)$ on the present system. Where J_1 and J_2 are the exchange interactions shown in the inset of Fig. 2. By applying the relation to the present result, α is obtained as $\alpha = 0.37$. Weihong *et al.* studied the quantum renormalization effects on the ground state and the excitation properties of anisotropic triangular antiferromagnet using the series expansion method [2]. They obtained the relationship between q and α as shown by a solid line in Fig. 2. When q is smaller than 2/3, α estimated from their theory is larger than that of classical model. Applying this relationship to the present experiment, we obtain $\alpha \sim 0.64$ for the sample with $x = 3.4$.

Figure 3 shows a contour map of the excitation spectrum along (0 k 0) direction. Spin wave modes which is similar to those of end members were clearly observed. The dashed lines are the spin-wave dispersion relations of anisotropic triangular antiferromagnetic model with $\alpha = 0.37$ [3]. By adjusting the amplitude of the model to the experimental result, the exchange energy J_1 can be estimated as $J_1 = 2.0 \pm 0.1$ meV. We should be noticed that the estimated J_1 using the classical model is enlarged by quantum renormalization effect. For the sample with $x = 0$, Cs_2CuCl_4, J_1 was found to be larger than the real magnitude of the interaction about 1.6 times [4].

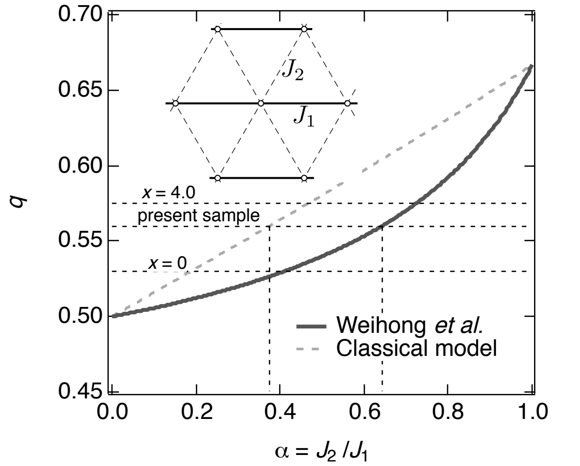

Figure 2 Static structure factors in [hk0] plane measured at T = 10K (a) and 0.3K (b).

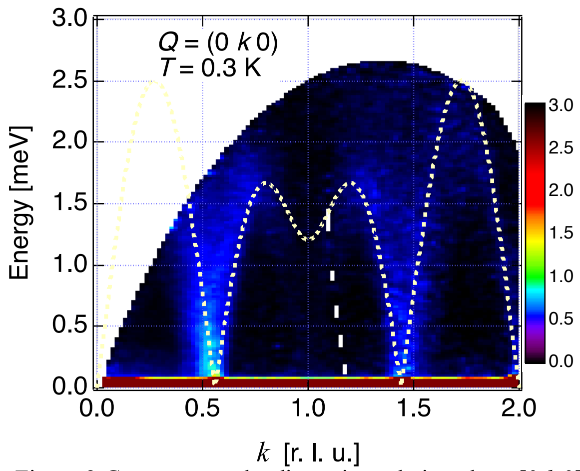

Figure 3 Contour map the dispersion relation along [0 k 0] direction. Dashed line indicates spin-wave dispersion relation of anisotropic triangular antiferromagnet.

Figure 4 is the scattering intensity as a function of energy around $Q = (0, k, 0)$ with $k \sim 0.76$ measured at $T = 0.3$K. Excitation spectrum of present experiment which is indicated by red circles shows rather broad peak around 1.65 meV. Blue line is the result of the sample with $x = 4.0$, Cs_2CuBr_4, measured with energy resolution $\Delta E \sim 1.7$ meV which is almost equivalent to the present experiment. The line width of the spin-wave excitation of the present experiment is approximately 2 times larger than that of $x = 4.0$. This result qualitatively reproduces the randomness effect on the line shape which is predicted for triangular antiferromagnet by Shimokawa *et al.* [5].

As described above, the ground state and the dynamical structure factor of the present sample is identical to those of end member except for the magnitude of exchange interactions. This fact indicates that the halogen ion substitution of the present system does not significantly alter the fundamental properties as the anisotropic triangular antiferromagnetic system. By performing comparative study using the sample with more strong randomness is essential to verify the random singlet state which is theoretically expected.

[1] K. Nakajima *et al.*: J. Phys. Soc. Jpn. **80** SB028 (2011).
[2] Z. Weihong *et al.*: Phys. Rev. B **59** 14367 (1999).
[3] R. Coldea *et al.*: Phys. Rev. B **68** 134424 (2003).
[4] R. Coldea *et al.*: Phys. Rev. Lett. **88** 137203 (2002).
[5] T. Shimokawa *et al.*: Phys. Rev. B **92** 134407 (2015).

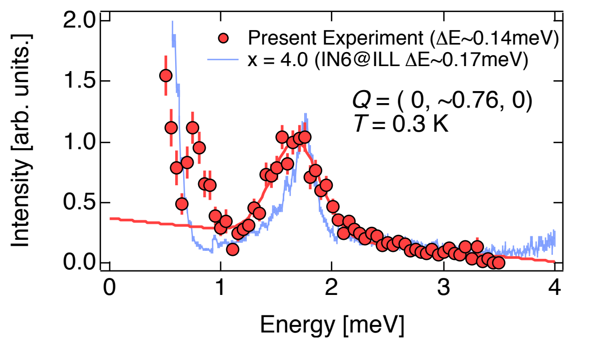

Figure 4 Measured scattering intensity at $Q = (0, \sim 0.76, 0)$ as a function of energy (red circles). Blue line denotes the result for the sample with $x = 4.0$.