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	June, 15 2017
課題番号 Project No.	2016B0023
実験課題名 Title of experiment	Exploring magnetic excitations in fully frustrated dimerized quantum magnet
実験責任者名 Name of principal investigator	Hidekazu Tanaka
所属 Affiliation	Tokyo Institute of technology
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	February 3-6 and 10-13, 2017

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.

$Ba_2CuSi_2O_6Cl_2$ and $Ba_2NiSi_2O_6Cl_2$

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.

A new series of two-dimensional (2D) dimerized antiferromagnets $Ba_2MSi_2O_6Cl_2$ (M = Co [1], Cu [2], Ni [3]), abbreviated hereafter as B*M*SOC, attracts recent attention owing to their intriguing quantum phenomena. The 2D magnetic network is illustrated in Fig. 1 where J and J_{ij} represent intradimer and interdimer exchange interactions, respectively. The ground state of B*M*SOC is a nonmagnetic singlet with a finite gap to the excited state. When magnetic field exceeding the critical field H_c is applied, magnetic excitations, magnons, are mapped onto the boson lattice. Magnons move to the neighboring site with amplitude proportional to $J_{11}+J_{22}-J_{12}-J_{21}$. When $J_{11}+J_{22}=J_{12}+J_{21}$, the interdimer interactions are fully frustrated and hence the hopping of magnons becomes completely suppressed.

It was reported that BCoSOC, which is described by an effective spin S = 1/2 XXZ model, exhibits a sharp stepwise magnetization plateau at one-half of the saturation value M_s [1]. Recent inelastic neutron scattering measurements have shown that the magnetic excitations are completely dispersionless. These results provide evidence that the frustration of interdimer interactions is almost perfect and hence, above H_c, magnons form a periodic array regarded as a Wigner crystal. Similar conclusion might be drawn for the S =1 analog BNiSOC where the magnetization process is stepwise with plateaus at $(1/4)M_s$ and $(1/2)M_s$ [3]. On the other hand, BCuSOC undergoes an XY antiferromagnetic ordering at H_c above which the magnetization continuously increases with increasing field [2], characteristic of the weak frustration, i.e. $J_{11}+J_{22}\neq J_{12}+J_{21}$.

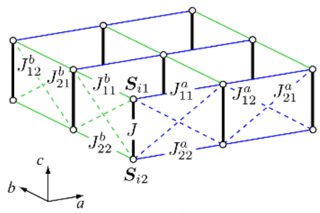

Fig. 1: 2D magnetic network in $Ba_2MSi_2O_6Cl_2$. J and J_{ij} represent intradimer and interdimer exchange interactions, respectively.

In this project, we measured magnetic excitations of BCuSOC and BNiSOC single crystals using a cold-neutron diskchopper spectrometer AMATERAS (BL14) installed at J-PARC, Japan. For both the measurements, the wave vector k_i of incident neutron was first set to be parallel to the c* axis. The scattering

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

were collected by rotating the sample around the *a*- (or *b*-) axis to clarify excitation spectra in a wider momentum-energy space. The intensities along the *c** direction are integrated assuming that the interlayer exchange interactions are negligibly small. The measurements were carried out mainly at the base temperature of 0.3 K and 4 K for BCuSOC and BNiSOC, respectively, which are sufficiently smaller than their gap energies.

Figure 2(a) shows the excitation spectra of BCuSOC for (*h*, 0) at 0.3 K with the incident neutron energy E_i = 5.9 meV. In contrast to the case of BCoSOC, a dispersive excitation with the width of ~1 meV is observed. The lowest excitation energy of 1.7 meV is comparable to the gap energy of 20.4 K [2]. The dispersive behavior is also found for (0, *k*) while the excitation spectrum along the *c** direction is almost flat, indicative of the good two-dimensionality. Together with the reported magnetization process with magnetization slope above H_c, we can conclude that the frustration of interdimer interactions is weak in BCuSOC. As the dispersion curve exhibits minima at Γ point, $J_{11}+J_{22} < J_{12}+J_{21}$ is probably realized. Interestingly, the scattering intensities become significantly decreased at 2.6 meV, which is close to J = 28.1 K [2]. The gap-like feature with the splitting width of 0.2 meV is found at 2.6 meV, independently of Q and of E_i. This result could be explained by the presence of unequal interdimer interactions, which can produce a band gap at the energy corresponding to the J value. Since this scenario is not applicable for the crystal structure of *Cmca* determined at room temperature [2], further experiments such as low temperature X-ray measurements are necessary.

Figure 2(b) shows the excitation spectra of BNiSOC for (*h*, 0) at 4 K with E_i = 9.9 meV. Almost dispersionless four excitations are observed. Although the reason why the excitation spectra constitute two similar excitations both at ~5 meV and ~7 meV have not been identified yet, these two energies probably relate to excitation gaps from the singlet ground state to the first and second excited states, respectively. In fact, the former and latter gaps are deduced to be 5.6 meV and 9.6 meV, respectively [3]. The present result provides evidence that interdimer interactions are strongly frustrated in BNiSOC, consistent with the reported stepwise magnetization process [3]. It is of interest that the two excitations at ~ 5 meV and the other two excitations at ~ 7 meV are intense when both *h* and *k* are integers and half-integers, respectively. It is noted that the similar lattice periodicity of the scattering intensities was found in the excitation spectra of BCoSOC where the two excitations with Q-dependent scattering intensities can reasonably be explained by the presence of unpaired spin dimers [4].

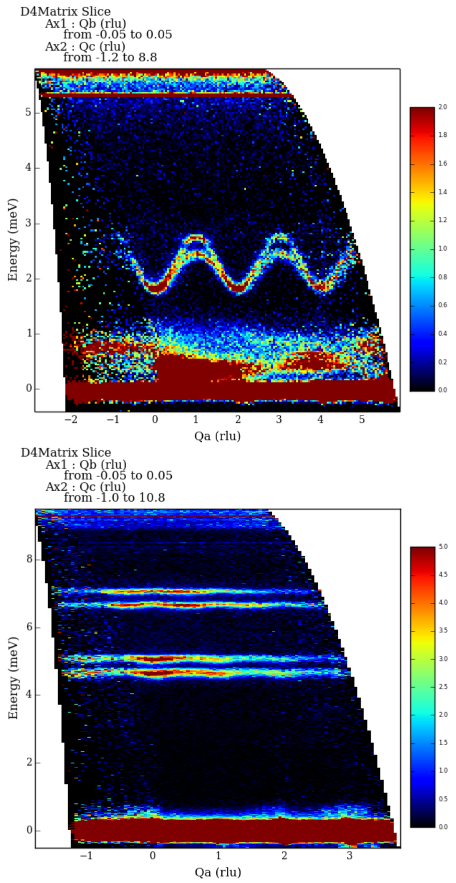

Fig. 2: Energy-momentum maps of the neutron scattering intensity for Q_a = (*h*, 0) in (a) $Ba_2CuSi_2O_6Cl_2$ at T = 0.3 K with E_i = 5.9 meV and (b) $Ba_2NiSi_2O_6Cl_2$ at T = 4 K with E_i = 9.9 meV. The scattering data were collected by rotating the samples around the *a*- (or *b*-) axis. The intensities along the *c** direction are integrated.

References

[1] H. Tanaka, N. Kurita, M. Okada, E. Kunihiro, Y. Shirata, K. Fujii, H. Uekusa, A. Matsuo, K. Kindo, and H. Nojiri, J. Phys. Soc. Jpn. **83**, 103701 (2014).

[2] M. Okada, H. Tanaka, N. Kurita, K. Johmoto, H. Uekusa, A. Miyake, M. Tokunaga, S. Nishimoto, M. Nakamura, M. Jaime, G. Radtke, and A. Saúl, Phys. Rev. B **94**, 094421 (2016).

[3] M. Koike, Master's thesis, Tokyo Institute of Technology (2017).

[4] N. Kurita *et al.*, in preparation.