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 2016
課題番号 Project No. 2015A0161 実験課題名 Title of experiment Magnetic excitations in the highly frustrated dimer magnet $Ba_2CoSi_2O_6Cl_2$ 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 March 23 to 26, 2016

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.

$Ba_2CoSi_2O_6Cl_2$

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.

$Ba_2CoSi_2O_6Cl_2$ is magnetically described as an S=1/2 dimerized quantum magnet [1]. It is deduced from the crystal structure that the exchange network in the dimer layer parallel to the *ab* plane is closely related to that shown in Fig. 1(a), and the exchange interaction between layers is negligible. There are two kinds of interdimer exchange interactions, as illustrated by thin solid and dashed lines in Fig. 1(a). These two interdimer exchange interactions act to interfere each other's transfer of triplet. This situation is called "frustration" in the dimerized magnet. In the case of strong frustration, $J'_{=} \approx J'_{\times}$, the movement of triplet is much suppressed. In this situation, magnetic excitations become dispersionless. When such a strongly frustrated dimerized magnet is subjected to magnetic field, the ground state is determined by the Zeeman energy and the repulsive interaction between triplets owing to the longitudinal component of the interdimer exchange interaction. With increasing magnetic field, triplets are created on dimers at the critical field. However, the simultaneous occupation of triplets on neighboring dimers is unfavorable, because it causes a rapid increase in the interaction energy. Therefore, triplets form a periodic array regarded as a "crystallization" to avoid the repulsive force between triplets [2]. With a further increasing magnetic field, triplets occupy all the dimer sites, which leads to the saturation of magnetization. Consequently, magnetization varies stepwise. The magnetization curves in $Ba_2CoSi_2O_6Cl_2$ measured for two different field directions are sharply stepwise with a plateau at half of the saturation magnetization [1]. This magnetization process is typical of the fully frustrated case, $J'_{=} = J'_{\times}$.

In the present experiment, we measured magnetic excitations in $Ba_2CoSi_2O_6Cl_2$ using a cold-neutron diskchopper spectrometer AMATERAS (BL14) installed at J-PARC, Japan. The wave vector $\boldsymbol{k}_{\mathrm{i}}$ of incident neutron was first set to be parallel to the c^* axis. As shown in Fig. 1(b), completely dispersionless three excitations were observed at E=4-7 meV. The widths of these excitations are resolution-limited. Thus, all the excitation peaks are single peaks that are not the superposition of two or more excitation peaks. These excitations are considered to be related to S^z=±1 excitations, because the S^z=0 level is expected to be at E≈10 meV owing to

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

strong XY anisotropy. Unexpected observation is that the intensities of these excitations have different ***Q*** dependence, as shown in Figs. 1(c)-(e). The intensity of most intense middle excitation is independent of ***Q***, which is assigned as single singlet-triplet excitations to $S^z=\pm1$ states. On the other hand, the lower and higher excitations are intense when both *h* and *k* are integers and half-integers, respectively. Because the intensities of these two excitations have the lattice periodicity, and the energy deference between the higher and middle excitations and between the middle and lower excitations are order of the interdimer interaction [1], we can deduce that the interdimer interactions are responsible for these excitations. This verifies that interdimer interactions in $Ba_2CoSi_2O_6Cl_2$ are fully frustrated, i.e., $J'_{=} = J'_{\times}$. To the best of our knowledge, there is no report on such dispersionless excitations with ***Q***-dependent intensities. Although the origins of these excitations have not been clearly identified yet, it is certain that the excitations of neighboring two dimers are strongly correlated. One possible origin is follows: a single singlet-triplet transition to $S^z=0$ state occurs. Before the triplet excitation is relaxed to the singlet state, the neighboring dimer is excited to $S^z=\pm1$ states to form a bound state with the help of the interdimer interactions.

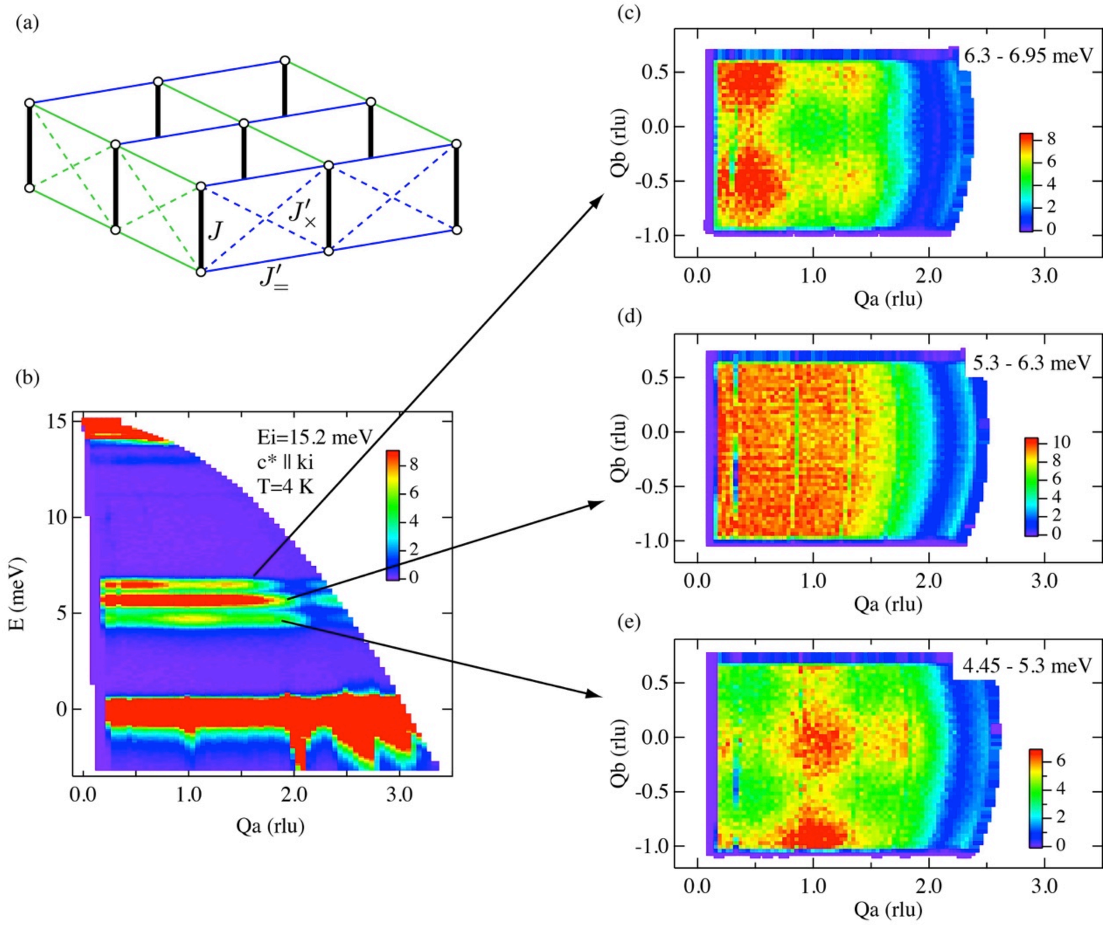

Fig. 1: (a) Illustration of exchange network in $Ba_2CoSi_2O_6Cl_2$. Thick bonds are strong intradimer exchange interactions *J*, while thin solid and dashed bonds, $J'_{=}$ and $J'_{\times}$ respectively, are weak interdimer exchange interactions. (b) Energy-momentum map of the scattering intensity along ***Q***=(*h*, 0, 0) in $Ba_2CoSi_2O_6Cl_2$. Intensity maps on ***Q***=(*h*, *k*, 0) for energies between (c) 6.3 and 6.95 meV, (d) 5.3 and 6.3 meV and (d) 4.45 and 5.3 meV.

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] T. M. Rice, Science **298**, 760 (2002).