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。）

CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2015/4/13 承認者 Approver Kaoru Shibata 提出日 Date of Report 2015/4/13
課題番号 Project No. 2014B0180 実験課題名 Title of experiment Low energy magnetic excitations in a single molecule magnet Mn_6 実験責任者名 Name of principal investigator Kazuki Iida 所属 Affiliation CROSS	装置責任者 Name of Instrument scientist Kaoru Shibata 装置名 Name of Instrument/(BL No.) DNA (BL02) 実施日 Date of Experiment 2015/4/8 – 2015/4/11

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.

$[n\text{-}BuNH_3]_{12}[Mn_6Cl_6(SbW_9O_{33})_2]\cdot 6H_2O$ or Mn_6 in short

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.

Isolated magnetic clusters can exhibit quantum phenomena such as quantized magnetic states and quantum tunneling between them. When the interactions between the clusters become strong, a magnetic long range order with collective spin wave excitations can appear at low temperatures. If the clusters are arranged in a frustrating lattice with a triangular motif, however, the magnetic order can be suppressed, leading to exotic states. For instance, $LiZn_2Mo_3O_8$ [1] in which Mo_3O_{13} clusters with $S = 1/2$ quantum spins form a triangular superlattice has a ground state of a collective disordered valence bond solid with a gapless excitation spectrum. In general, for a coupled molecular system, its ground state is determined by the interplay between the effective spin value of the magnetic molecules (S) and the nature of the “inter”-cluster and “intra”-cluster couplings. It is important to investigate different regions of the phase diagram to search for possible exotic states, and thus, we are investigating Mn_6 [2], which are coupled clusters in a frustrating triangular lattice as shown in Fig. 1.

To study detailed magnetic correlations in Mn_6, we have performed time-of-flight measurements on

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

powder Mn_6 at DCS, NIST. Figure 2 shows $\hbar\omega$ dependences of the scattering intensities, $S(Q = 0.25\ \text{Å}^{-1}, \hbar\omega)$, at 1.5, 3.0, and 6.0 K. Gapless low energy magnetic fluctuations are observed at 6.0 K, while strong peak centered at $\hbar\omega$ = 0.26 meV and small peaks around 0.54 meV are observed at 1.5 and 3.0 K. Interestingly, our exact diagonalization study shows that both $\hbar\omega$ and Q dependences can be well explained by a spin Hamiltonian describing two neighboring Mn_6 clusters that have ferromagnetic "intra"-cluster and antiferromagnetic "inter"-cluster couplings, and an easy-plane anisotropy. All the results so far suggest that Mn_6 is the system where the magnetic clusters are arranged in a frustrating lattice with a triangular motif and the ground state is most likely short-range order. Although our previous study revealed the existence of antiferromagnetic short-range order of Mn_6 clusters, our calculation expects that there are additional magnetic excitations.

To confirm the validity of our model which can explain the exotic magnetism in the Mn_6 system, we performed inelastic neutron scattering measurements at BL02 DNA, MLF, J-PARC. We have prepared deuterated 4 g powder sample, which was put into a Cu can and then attached to the dilution refrigerator. Si(111) reflections and 3x3 slits were used so that the energy resolution was 3 μeV at the elastic channel. By combining the five different phases, we obtained the fine-energy-resolution intensity maps in the range of $-0.4< \hbar\omega<0.8$ meV and $0.1<Q<1.8\ \text{Å}^{-1}$. Here, we show the energy spectra from Mn_6 at 0.05 and 1.5 K using DNA. Q was integrated in $0.2 < Q < 1.0\ \text{Å}^{-1}$. As observed at DCS, the sharp peak was observed at $\hbar\omega$ = 0.265 meV both in 0.05 and 1.5 K. Surprisingly, additional peaks evolves at 1.5 K at 0.225 and 0.245 meV, which are very close the 0.265 meV peak. We are now considering how we can explain this "new" result using our model. We believe that the result at DNA supports our model in which Mn_6 is the system where the magnetic clusters are arranged in a frustrating lattice with a triangular motif and the ground state is most likely short-range order.

Finally we would like to thank the ISSP, CROSS, and JAEA staff for their great help and effort with preparing and conducting our experiment.

[1] J. P. Sheckelton *et al*., Nat. Mater. **11**, 493 (2012).
[2] T. Yamase *et al*., Inorg. Chem. **45**, 7698 (2006).

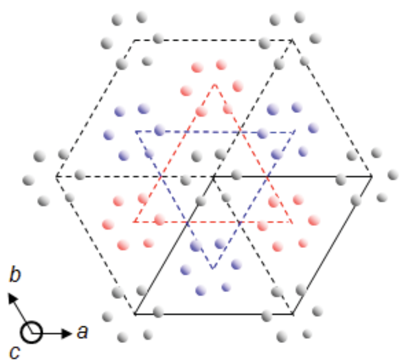

Fig. 1
Crystal structure of Mn_6.

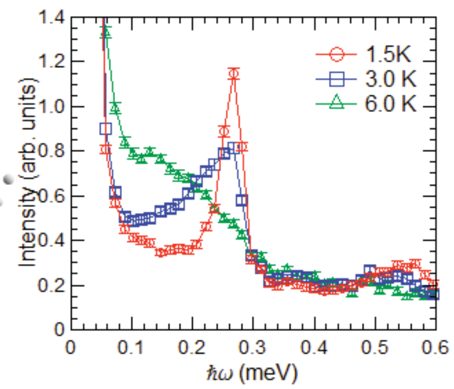

Fig. 2
$\hbar\omega$ dependences of $S(0.25\ \text{Å}^{-1}, \hbar\omega)$ at 1.5, 3.0, and 6.0 K using DCS.

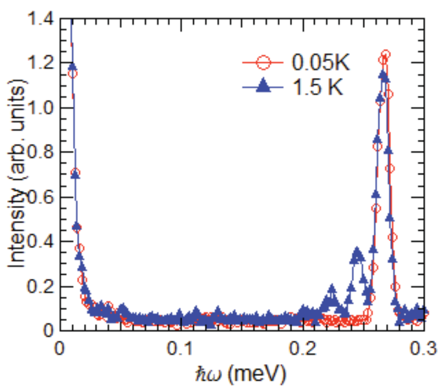

Fig. 3
Energy spectra at 0.05 and 1.5 K using DNA.