(※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 2017/1/4
課題番号 Project No. 2017A0094 実験課題名 Title of experiment Search for magnetic excitations in a Zn-Nd-Zn single ion magnet 実験責任者名 Name of principal investigator Maiko Kofu 所属 Affiliation J-PARC Center, JAEA	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 2017/6/12 – 2017/6/14 2017/6/30 – 2017/7/1

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.

Nd complex ($C_{40}D_{39}NdN_8O_{11}S_2Zn_2$)

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 single-molecule magnet (SMM) is a metal complex that behaves as an individual nanomagnet. Each molecule, containing several metal centers with unpaired electrons, possesses a giant resultant spin. Given that the giant spin exhibits easy-axis anisotropy ($D < 0$), the magnetization reversal between the ground states with $S_z = \pm S$ is hindered by the potential barrier of DS_z^2. The barrier yields a slow relaxation of the magnetization reversal that is characteristic of SMMs. Early SMM researches have focused on the complexes containing multiple transition metal atoms. During the past decade, lanthanide SMMs have received much attention as promising materials with high blocking temperature, i.e. stable SMMs [1]. Owing to a large contribution of angular momentum, lanthanide complexes can become SMMs containing only one or two magnetic ions. However, the relaxation behaviors of lanthanide SMMs are rather complicated and several types of mechanisms are discussed, for example Raman, direct and quantum tunneling processes. Clarifying the mechanism of magnetic relaxation is a key issue in lanthanide SMMs and assists formulation of designing strategies for engineering long-living SMMs.

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

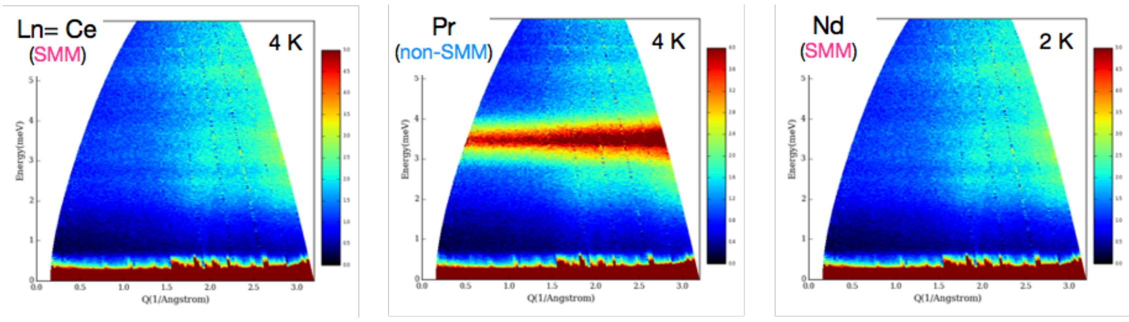

Fig. 1: $S(Q,\omega)$ maps of Ce, Pr, and Nd complexes taken on AMATERAS with E_i of 7.7 meV at base temperature.

We have first studied a Tb–Cu dinuclear SMM by means of inelastic (INS) and quasielastic neutron scattering (QENS) [2,3]. We are now investigating trinuclear Zn–Ln–Zn complexes (Ln = Ce, Pr, Nd). The system has only one magnetic center in a molecule. Interestingly, the complexes with Kramers ion (Ce : J=5/2 and Nd : J=9/2) exhibit the SMM behavior while that with non-Kramers ion (Pr : J=4) does not [4]. Firstly, the INS measurements were made on AMATERAS for the Ce and Pr complexes (2014B0064). In the present measurement, the Nd complex was examined. Figure 1 shows the observed $S(Q,\omega)$ maps for the three complexes. A magnetic excitation is observed at 3–4 meV in the Pr complex but undetected in the Ce and Nd ones. It indicates that the existence or non-existence of the low-energy excitation is closely related to whether the ac susceptibility exhibits the non-SMM or SMM behavior. To search for magnetic excitations in the Nd complex, an additional INS measurement in the high-energy region (E > 15meV) is planned.

We have also performed QENS measurements for the Nd complex. Figure 2 shows Arrhenius plots of relaxation times obtained, together with the data taken on DNA (Proposal No. 2017A0068) and from ac magnetic susceptibility. It is evident that the relaxation times do not follow Arrhenius law. So far, the Orbach process through tunneling between pairs of degenerate magnetic sublevels has been considered as a major mechanism for SMMs. However, (i) the absence of magnetic excitation at 3 meV corresponding to the potential barrier of 35 K estimated from ac magnetic susceptibility and (ii) the non-Arrhenius behavior of relaxation time suggest that the magnetic relaxation does not take place through the standard Orbach process in the Zn–Ln–Zn SMMs. Recently, other mechanisms of spin-phonon relaxation including Raman process and an anharmonic phonon effect are much discussed [5,6]. We believe that our study can provide insight into the mechanism of magnetization reversal in rare-earth based SMMs.

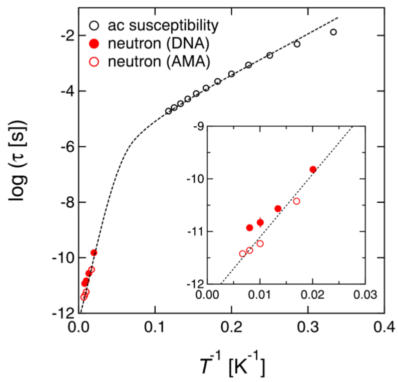

Fig. 2: Arrhenius plots of relaxation times obtained from QENS and ac susceptibility measurements in the Nd complex.

[1] R. Sessoli et al., Coordin. Chem. Rev., 253, 2328 (2013). [2] M. Kofu et al., Phys. Rev. B, 88, 064405 (2013). [3] M. Kofu et al., Chem. Phys., 427, 147-152 (2013). [4] C. Takehara et al., Dalton Trans., 44, 18276 (2015). [5] Y. Rechkemmer et al., Nat Commun, 7, 10467 (2016). [6] A Lunghi et al., Nat Commun, 8, 14620 (2017).