CROSS TOKAI Experimental Report J-PARC	提出日 Date of Report 2017/5/16 承認者 Approver Takashi Ohhara 提出日 Date of Report 2017/5/16
課題番号 Project No. 2017A0148 実験課題名 Title of experiment Neutron diffraction study of the devil's staircase in $La_5Mo_4O_{16}$ 実験責任者名 Name of principal investigator Ryoichi Kajimoto 所属 Affiliation J-PARC Center	装置責任者 Name of responsible person Takashi Ohhara 装置名 Name of Instrument/(BL No.) SENJU (BL18) 実施日 Date of Experiment 2017/4/9 – 2017/4/14

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.

Single crystal $La_5Mo_4O_{16}$

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.

$La_5Mo_4O_{16}$ consists of perovskite layers of MoO_6 octahedra, where 4*d* electrons at Mo sites in the perovskite layers have localized magnetic moments with strong Ising anisotropies, and Mo_2O_{10} pillars connect the perovskite layers. It is believed that Mo ions in the pillars are non-magnetic, and thus the inter-layer magnetic interaction between the perovskite layers are expected to be very weak. In fact, $La_5Mo_4O_{16}$ undergoes antiferromagnetic order with the magnetic propagation vector **k** = (0, 0, 0.5) below T_N = 200 K [Fig. 1(a)], and external magnetic field along the *c* axis (> 0.5 T) can induce the ferrimagnetic order with **k** = (0, 0, 0) [Fig. 1(b)]. Recently, a Waseda University group found so-called devil's staircase in $La_5Mo_4O_{16}$ under external magnetic field along the *c* axis as shown in Fig. 1(c). To investigate in detail the nature of the magnetic devil's staircase by means of the single crystal neutron diffraction technique is a purpose of this experiment.

A single crystal with 40 mg was grown by the floating zone technique. We aligned the crystal in the *ab* plane to apply the external magnetic field along the *c* axis using the "vertical 7T magnet". Most measurements were carried out at T = 50 K, and some zero-field measurements at 4 and 220 K. Magnetic field along the *c* axis was applied in the range of $-5 < H < 5$ T. Data was analyzed by the software suite "STARGazer".

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

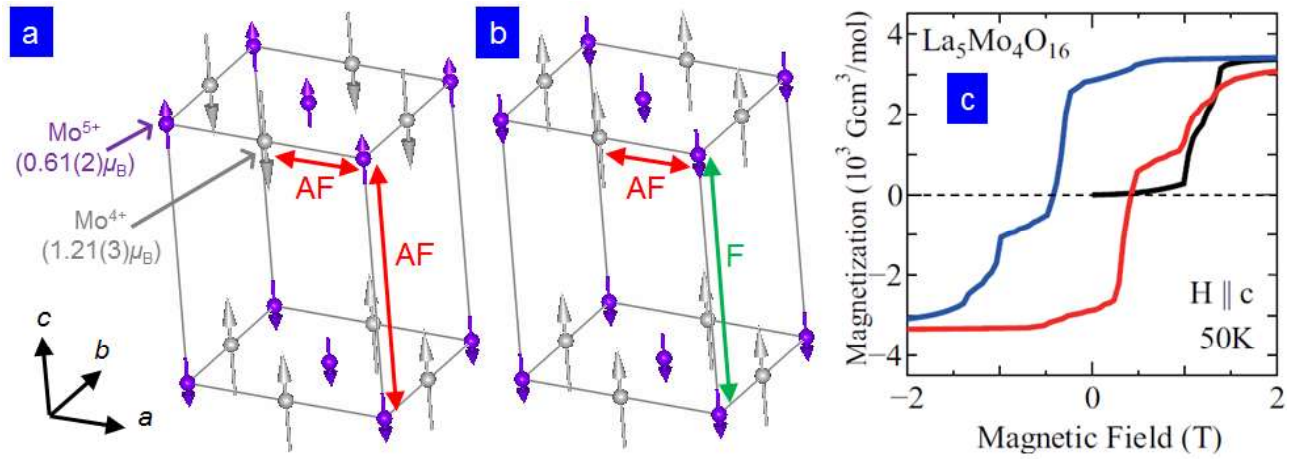

Fig. 1. (a) Antiferromagnetic structure and (b) ferrimagnetic structure of $La_5Mo_4O_{16}$. (c) Magnetization curve in $La_5Mo_4O_{16}$ under the external magnetic field along the *c* axis at 50 K.

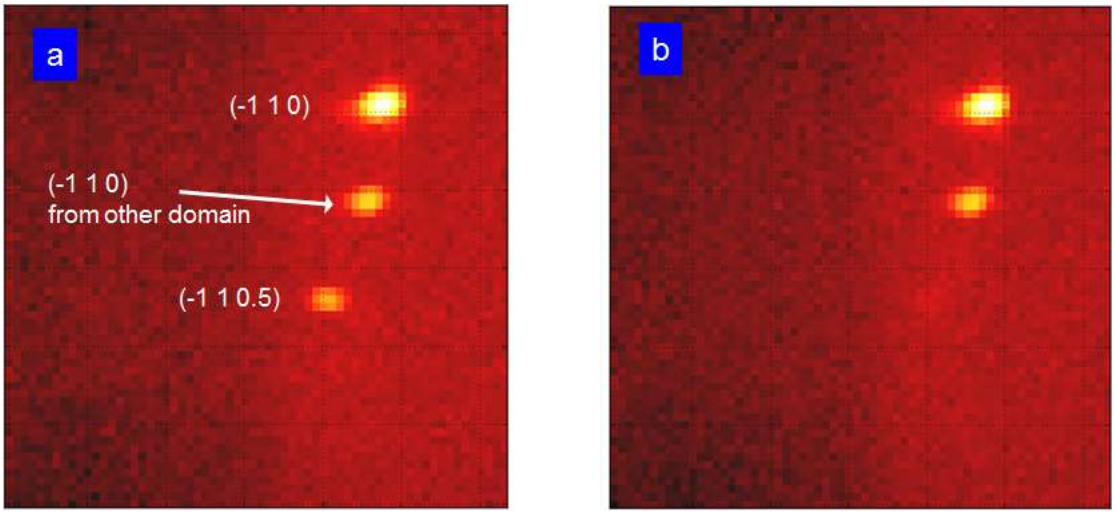

Fig. 2. Detector maps around (−1 1 *L*) at 50 K under the external magnetic fields of (a) 0 and (b) 5 T. The intensities are plotted in the log scale. Note that the Bragg peak from the other domain due to the monoclinic structure was also observed.

Figures 2(a) and 2(b) show neutron-intensity maps under the external magnetic fields of 0 and 5 T at 50 K using a single detector. In the absence of the external magnetic field, the magnetic Bragg peak was observed at the AF position of (−1 1 0.5). The AF Bragg peak disappeared at 5 T, indicating that the ferromagnetic structure is realized. On the other hand, no incommensurate magnetic peak was observed in any measured magnetic fields. Instead, the intensities at the AF and ferrimagnetic peaks are changed. These results indicate that the magnetization steps such as the 1/7 plateau cannot be explained by a long-period magnetic structure. Although further detailed analysis is required, the magnetic devil's staircase can be explained well by the balance of the occupancy of the AF and ferrimagnetic states, which are determined by the combination of temperature and external magnetic fields.

Finally, we would like to appreciate the local contacts, Dr. Akiko Nakao and Dr. Taketo Moyoshi, and the MLF sample environment team.