MLF Experimental Report

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</td><td>提出日 Date of Report
2014/5/19</td></tr>
<tr><td>課題番号 Project No.
2014A0258
実験課題名 Title of experiment
Study of the magnetic structure in the localized-4d system $La_5Mo_4O_{16}$
実験責任者名 Name of principal investigator
Ryoichi Kajimoto
所属 Affiliation
J-PARC Center</td><td>装置責任者 Name of responsible person
Toru Ishigaki
装置名 Name of Instrument/(BL No.)
BL20
実施日 Date of Experiment
2014/5/9 13:00 – 2014/5/10 13:00
(1 day)</td></tr>
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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.

3.7 g powder $La_5Mo_4O_{16}$.

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.

Generally, with an atomic number increasing, a spin-orbit (LS) coupling becomes significant for physical properties. Due to the strong LS coupling, 5*d* electron systems, for instance, show unconventional phenomena such as a topological insulator in $SrIrO_3$. On the other hand, in 3*d* electron systems, it is well known that the LS coupling can be treated perturbatively as a single-ion anisotropy. However, how the LS coupling works in the 4*d* electron systems still remains as a question. In addition, localized magnetism of the 4*d* electrons was only discovered in few compounds, and thus is not investigated much. Therefore, we are investigating $La_5Mo_4O_{16}$, a 4*d* electron material with localized magnetic moment. The most intriguing phenomenon we found in $La_5Mo_4O_{16}$ so far is the long-time decay of the magnetization as mentioned below, and to clarify its origin is the purpose of this work. Here, we performed the powder neutron scattering experiment on $La_5Mo_4O_{16}$ to study the magnetic structures without an external magnetic field.

From bulk measurements, a phase diagram of $La_5Mo_4O_{16}$ is reported [1]. At zero magnetic field there is a phase transition from a paramagnetic to an antiferromagnetic phase at T_{AF} = 190 K, and then another one from the antiferromagnetic to a ferrimagnetic phase at T_F = 70 K. Since it is difficult to make $La_5Mo_4O_{16}$ system, the

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

magnetic structure has not been investigated by neutron scattering. Instead, the magnetic structure of V-doped $La_5Mo_{2.75}V_{1.25}O_{16}$ was reported; a collinear ferrimagnetic structure develops in *ab*-plane and it stacks antiferromagnetically along *c*-axis [2]. Speculating from $La_5Mo_{2.75}V_{1.25}O_{16}$, we expect that similar magnetic structures in the antiferromagnetic and ferrimagnetic phases of $La_5Mo_4O_{16}$, and to confirm them by means of neutron diffraction technique is purpose of this experiment.

Neutron diffraction measurements were performed at BL20 iMATERIA using a dual frame. Approximately 3.7 g of $La_5Mo_4O_{16}$ was packed into a vanadium can. The can was sealed by an indium wire. Diameter and height of the can were 6 and 65 mm, respectively. The sample can was filled with ^{4}He gas, which was then set to a closed-cycle refrigerator. Measurements were performed at T = 4, 40, 100, and 220 K.

Figure 1 shows the neutron powder diffraction patterns of $La_5Mo_4O_{16}$ measured at (a) T = 4, (b) 100, and (c) 220 K. Almost all Bragg peaks in the paramagnetic phase at T = 220 K are indexed by the reported structure of $La_5Mo_4O_{16}$ [3]. There are several magnetic Bragg peaks observed at T = 4 and 100 K in the long range magnetic ordered state. Contrary to the bulk measurement results, however, the diffraction patterns at T = 4 and 100 K are almost same; as mentioned above the bulk measurements reported the phase transition from the antiferromagnetic to ferrimagnetic phase at 70 K. Both at 4 and 100 K, magnetic Bragg peaks were observed at Q = 1.135, 1.175, 1.4, and 1.49 Å^{-1}, which are indexed as (-1 1 0.5), (1 1 0.5), (-1 1 1.5), and (1 1 1.5), respectively; we assume the $C/2m$ space group in this report. Since symmetry of $La_5Mo_4O_{16}$ is monoclinic, magnetic propagation vector is **k** = (0, 0, 0.5). Thus, the model spin structure expected from the bulk measurements below 70 K, the ferrimagnetic structure with **k** = (0, 0, 0), is not correct. Our powder diffraction measurements revealed that the magnetic structure of $La_5Mo_4O_{16}$ below T_N ~ 190 K is an antiferromagnetic structure, which is very similar to that of $La_5Mo_{2.75}V_{1.25}O_{16}$ [2]: a collinear ferrimagnetic structure in *ab*-plane with antiferromagnetic stacking along *c*-axis. We are now doing Rietveld refinements of both the crystal and magnetic structures to determine the magnetic structure in $La_5Mo_4O_{16}$.

[1] K. Kobayashi and T. Katsufuji, Phys. Rev. B **83**, (2011) 100411(R).
[2] F. Ramezanipour *et al*., J. Sol. Stat. Chem. **184**, (2011) 3366.
[3] M. Ledesert *et al*., J. Sol. Stat. Chem. **105**, (1993) 143.

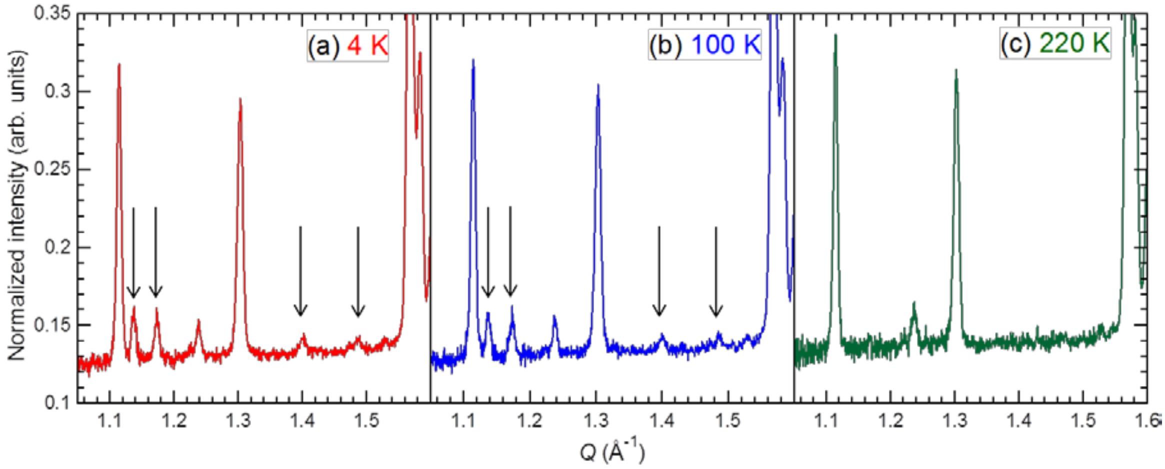

Fig. 1. Neutron diffraction patterns of $La_5Mo_4O_{16}$ at (a) T = 4, (b) 100, and (c) 220 K using iMATERIA. Magnetic reflections are indicated by solid arrows.