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 2014/5/12 承認者 Approver Ryoichi Kajimoto 提出日 2014/5/12
課題番号 Project No. 2014A0082 実験課題名 Title of experiment Inelastic neutron scattering study of the magnetic excitations 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	装置責任者 Name of Instrument scientist Ryoichi Kajimoto 装置名 Name of Instrument/（BL No.） BL01 実施日 Date of Experiment 2014/4/28 11:00 – 2014/4/30 11:00 (2 days)

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.
11 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 investigated $La_5Mo_4O_{16}$, a 4*d* electron material with localized magnetic moment, using a combination of elastic and inelastic neutron scattering techniques at iMATERIA and 4SEASONS.

In $La_5Mo_4O_{16}$, there are perovskite layers where Mo ions form a quasi-square lattice. Mo ions in the perovskite layers have S = 1/2 and S = 1 spins, and the layers are magnetically separated along the *c*-axis. Bulk measurements reported that 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 [1]. Speculating

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

from the similar compound $La_5Mo_{2.75}V_{1.25}O_{16}$[2], collinear antiferromagnetic and ferrimagnetic structures are expected in $La_5Mo_4O_{16}$.

Inelastic neutron scattering measurements were performed at BL01 4SEASONS. Approximately 11 g of powder $La_5Mo_4O_{16}$ was packed into an aluminum foil. Diameter and height of cylinders were 15 and 28 mm, respectively. The aluminum foil was then set to a closed-cycle refrigerator. Beam slits were 50(h) × 40(w) - 35(h) × 25(w) mm. We selected the setup with incident energies of E_i = 10.5, 15.3, 24.4, 45.2, and 110.6 meV with the Fermi chopper frequency f_{Fermi} = 250 Hz. The measurements were performed at T = 4, 100, and 220 K.

By cutting time-of-flight data along a Q-direction near the $\hbar\omega$ = 0 region, the diffraction pattern can be obtained. We compared the diffraction patterns at T = 4 and 220 K. Magnetic reflections are observed at Q = 0.874, 1.175, and 1.488 $Å^{-1}$ at 4 K. These magnetic reflections are indexed as (0,0,1.5), (1,1,0.5), and (1,1,1.5), respectively. Therefore, the magnetic propagation vector is $\mathbf{k}$ = (0,0,0.5), and the corresponding magnetic structure is shown in Fig. 1. For detailed analysis on the magnetic structure of $La_5Mo_4O_{16}$, we performed the diffraction measurement at iMATERIA (2014A0258).

A $S(Q, \hbar\omega)$ map of $La_5Mo_4O_{16}$ at 4 K with E_i = 45 meV is shown in Fig. 2. Band like excitations are observed around $\hbar\omega$ = 10 and 20 meV. Specurating from the bulk measurement and the diffraction result, we cosnsider the following Hamiltonian for $La_5Mo_4O_{16}$:

$$H = J\sum[\delta(S^xT^x + S^yT^y) + S^zT^z] + D\sum(S^z)^2$$

where J, δ, and D are exchange constant, Ising parameter, and single-ion anisotropy. **S** and **T** represent the spin operators of S = 1/2 and 1 spins. Using the best parameters so far, the calculated $S(Q, \hbar\omega)$ map at 4 K is shown in Fig. 3. The calculation succeeded in reproducing the band like excitations around $\hbar\omega$ = 10 and 20 meV. However, the statistics of the data is not enough to unambiguously determine the parameters and discuss the LS coupling in $La_5Mo_4O_{16}$. We are planning a futher inelastic scattering experiment.

[1] K. Kobayashi and T. Katsufuji, Phys. Rev. B **83**, (2011) 100411(R).

[2] F. Ramezanipour *et al.*, J. Sol. Sta. Chem. **184**, (2011) 3366.

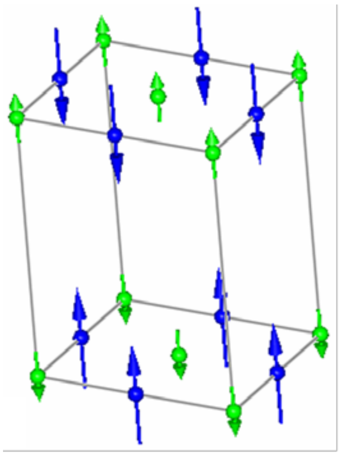

Fig. 1

Expected magnetic structure of $La_5Mo_4O_{16}$.

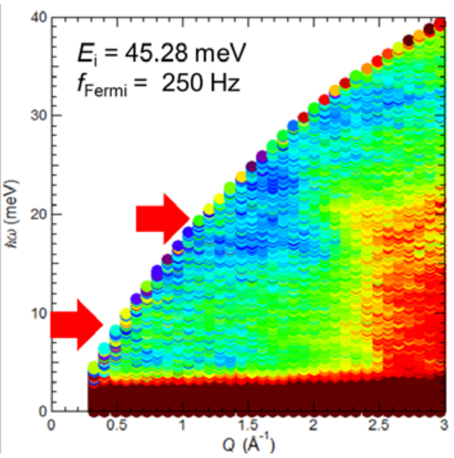

Fig. 2

Observed $S(Q, \hbar\omega)$ at T = 4K and E_i = 45.2 meV in $La_5Mo_4O_{16}$.

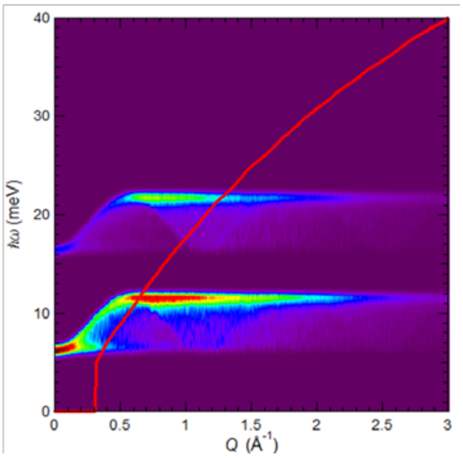

Fig. 3

Calculated $S(Q, \hbar\omega)$. of $La_5Mo_4O_{16}$ at T = 4 K.