MLF Experimental Report

課題番号 Project No. 2014B0141	提出日 Date of Report
実験課題名 Title of experiment Inelastic neutron scattering measurements on $La_5Mo_4O_{16}$ using the small angle detector at HRC	装置責任者 Name of responsible person S. Itoh, T. Yokoo, S. Ibuka
実験責任者名 Name of principal investigator Kazuki Iida	装置名 Name of Instrument/(BL No.) HRC (BL 12)
所属 Affiliation CROSS	実施日 Date of Experiment 2015/3/23 – 2015/3/27

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.

Powder $La_5Mo_4O_{16}$, 50 g

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 a strong LS coupling, 5*d* electron systems, for instance, are expected to show unconventional phenomena such as a topological insulator and a spin liquid state induced by the Kitaev interaction. 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 or anisotropic exchange interactions. However, how the LS coupling works in the 4*d* electron systems still remains as an open question. In addition, localized magnetism of the 4*d* electrons was only discovered in few compounds, and thus is not investigated much. We are now investigating $La_5Mo_4O_{16}$, a 4*d* electron material with localized magnetic moments, using a combination of elastic and inelastic neutron scattering techniques at BL20 (iMATERIA), BL01 (4SEASONS), and BL12 (HRC) in J-PARC.

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 [1]. Speculating from the similar compound $La_5Mo_{2.75}V_{1.25}O_{16}$[2], collinear antiferromagnetic structure

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

is expected in $La_5Mo_4O_{16}$, which was confirmed by our neutron diffraction experiment at BL20 iMATERIA as shown in Fig. 1. We have already performed inelastic neutron scattering measurements at BL01 4SEASONS. However, since the magnetic structure of $La_5Mo_4O_{16}$ is a basically ferrimagnetic, most of the spectral weights of magnetic excitations are expected to locate at a low Q region. Our data at BL01 was not enough to determine the spin Hamiltonian in $La_5Mo_4O_{16}$ without an ambiguity. Therefore, we planned to measure further inelastic neutron scattering at BL12 HRC using the small angle detectors to observe the low Q magnetic excitations.

Inelastic neutron scattering measurements were performed at BL12 HRC using a small angle detector. Approximately 45 g powder $La_5Mo_4O_{16}$ was packed into an aluminum foil [32 × 32 ×20 (depth) mm^3]. The aluminum foil with He gas was put in an aluminum sample can, which was then set to a closed-cycle refrigerator (a bottom loading type). A 30 × 30 mm^2 Cd beam mask and a 0.3° collimator were installed. We selected the setup of incident energies of E_i = 32.06 and 102.4 meV with a Fermi chopper frequency f_{Fermi} = 400 Hz. The measurements were performed at T = 4 and 220 K.

A $S(Q, \hbar\omega)$ map of $La_5Mo_4O_{16}$ at 4 K with E_i = 102.4 meV is shown in Fig. 2. Band like excitations are observed around $\hbar\omega$ = 10 and 26 meV. Specurating from our bulk measurement and neutron diffraction results, we cosnsider the following model Hamiltonian for $La_5Mo_4O_{16}$:

$$H = J_1\sum[\delta_1(S^xT^x + S^yT^y) + S^zT^z] + J_2\sum[\delta_2(S^xS^x + S^yS^y) + S^zS^z] + J_2\sum[\delta_2(T^xT^x + T^yT^y) + T^zT^z] + D\sum(T^z)^2$$

where J_n, δ_n, and D are n-th nearest neighbor 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 and instrumental energy resolution, 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 26 meV. Optimum parameters so far are J_1 = 7.5 meV, δ_1 = 0.8, J_2 = 1.2 meV, δ_2 = 0.8, and D = -0.8 meV. This result suggests that $La_5Mo_4O_{16}$ has a strong easy axis anisotropy, which is consistent with the single crystal magnetization results.

[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.

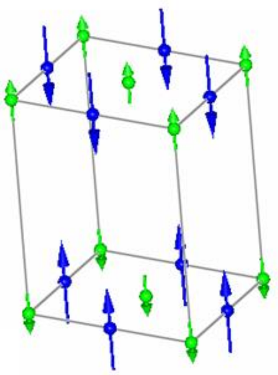

Fig. 1
Obtained magnetic structure of $La_5Mo_4O_{16}$ from BL20.

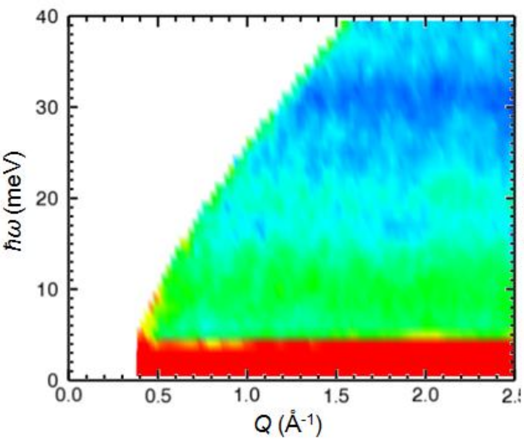

Fig. 2
Observed $S(Q, \hbar\omega)$ map at T = 4K and E_i = 102.4 meV in $La_5Mo_4O_{16}$.

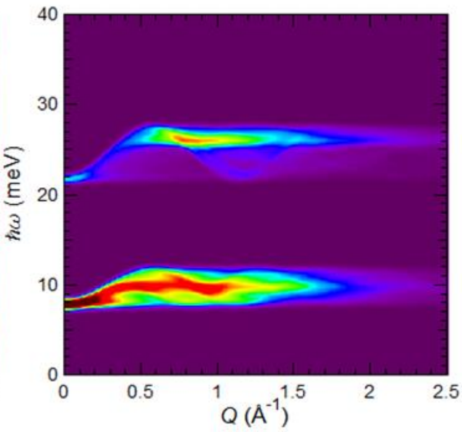

Fig. 3
Calculated $S(Q, \hbar\omega)$ map of $La_5Mo_4O_{16}$ at T = 4 K.