Investigation on magnetic devil's staircase in $La_5Mo_4O_{16}$

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Study of competing or frustrated interactions in materials is one of the important issues of the material science, because they often induce interesting magnetic properties. Such competing and/or frustrated interactions sometimes induce a long-period ordered structure whose periodicity shows discrete changes as a function of external field (so-called devil's staircase). This long-period structure is intimately related with the novel phenomena characterizing the materials as seen in recent examples of the stripe ordering in layered Ni perovskite oxides, charge ordering in the spin-ladder compound NaV_2O_5, the multiferroics in RMnO$_3$, and the giant magnetoresistance in $SrCo_6O_{11}$. Recently, we found another material, $La_5Mo_4O_{16}$ that shows the magnetic devil's staircase as a function of external magnetic field (B). In this material, several magnetization steps (e.g. 1/7 plateau) were observed in the magnetization curve under $B \parallel c$ at 50 K.

$La_5Mo_4O_{16}$ is a layered perovskite compound with monoclinic symmetry, and there are three inequivalent Mo sites (Mo1, Mo2, and Mo3) as shown in Fig. 1(a). Corner-sharing MoO_6 octahedra consisting of Mo1 and Mo2 form a quasi-square checkerboard lattice. Meanwhile, two Mo3 form a Mo_2O_{10} pillar, which is located between the perovskite layers and connects Mo2 octahedra. The valences of the Mo1, Mo2, and Mo3 sites are 5+, 4+, and 4+, respectively, and the Mo1 and Mo2 sites have spins $S = 1/2$ ($4d^1$) and $S = 1$ ($4d^2$). On the other hand, the molecular orbital in the edge-sharing bioctahedral Mo_2O_{10} is in the low spin state. Since Mo_2O_{10} pillars are nonmagnetic and the distance between the layers is large, the interlayer interaction is expected to be very weak. Under zero magnetic field, $La_5Mo_4O_{16}$ undergoes the magnetic phase transition below $T_N = 190$ K, where the in-plane collinear ferrimagnetic structure stacks antiferromagnetically along the c axis [K. Iida *et al.*, J. Phys. Soc. Jpn. **86**, 064803 (2017).]. The antiferromagnetic (AFM) interlayer coupling is fragile against the magnetic field, and $La_5Mo_4O_{16}$ undergoes a ferrimagnetic state with ferromagnetic (FM) interlayer coupling above $B \sim 0.5$ T. Therefore, the devil's staircase phenomenon in the magnetization curve of $La_5Mo_4O_{16}$ can be interpreted as a mixture of the FM and AMF coupled layers and discrete changes of their ratio. However, the actual way of stacking of the layers has not been confirmed due to lack of microscopic measurements. To clarify the period of the long-period ordering gives the fundamental information to understand the devil's staircase phenomenon and important hint of the characters of the competing interactions underlying the phenomenon.

We performed neutron diffraction measurements using a single crystal of $La_5Mo_4O_{16}$ with mass of $\sim$ 40 mg [Fig. 1(b)] under the external magnetic field ($B \parallel c$) at the single crystal diffractometer CORELLI in SNS to determine the wave vectors characterizing the magnetization steps. We also measured external magnetic field dependences of diffraction patterns in $La_5Mo_4O_{16}$.

Figures 1(c) and 1(d) show the external magnetic field dependences of diffraction patterns of $\left(-1 -1 -\frac{1}{2}\right)$ (AF) and $(-1 -1 -1)$ (FM) with field ramping up to 2 T at 50 K. When the field is increased, the AF magnetic peak disappears above 1.2 T [Fig. 1(c)], while the intensity of the FM peak increased above 1.2 T [Fig. 1(d)]. Interestingly, with field ramping down to 0 T, as shown in Figs. 1(e) and 1(f), field dependences of both $\left(-1 -1 -\frac{1}{2}\right)$ and

$(-1-1-1)$ were different from those of field ramping up. On the other hand, no incommensurate magnetic peak was observed (not shown). These results indicate that $La_5Mo_4O_{16}$ shows the hysteresis as expected for the ferrimagnetic nature, consistent with the bulk magnetic measurements. On the other hand, the 1/7 plateau cannot be explained by the long-priod magnetic structure with the magnetic propagation vetor of $\left(0, 0, \frac{1}{7}\right)$. Instead, the magnetization steps originate in the balance of AF and ferrimagnetic structures (or the domain effect due to the monoclinic structue), which could be explained by a function of temperature and external magnetic field.

The complete data set of neutron diffractions obtained by CORELLI under the external magnetic field will provide us great insights to construct the microscopic model to unserstand the exotic magnetisms in $La_5Mo_4O_{16}$.

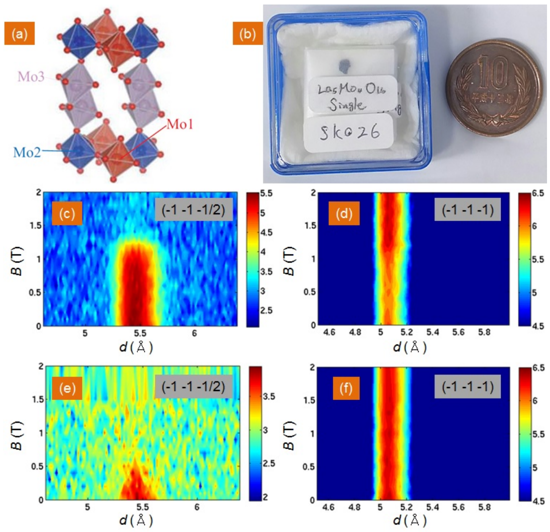

Fig. 1. (a) Crystal structure of $La_5Mo_4O_{16}$. (b) Picture of single crystalline $La_5Mo_4O_{16}$. (c) – (f) External magnetic field dependences of Bragg peaks. (c) and (d) are field ramping up while (e) and (f) are field ramping down.