Neutron diffraction study on orbital order in $La_5Mo_4O_{16}$

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Orbital order is supposed to be ubiquitous among 3*d*/4*d* (and 4*f*/5*f*) electron systems, and experimental identification of orbital order in a realistic compound is extremely important because orbital order is known to induce abundant intriguing phenomena such as a giant magneto-resistive effect. In this proposal, we performed a single-crystal neutron diffraction study on orbital order in a 4*d*-electron layered perovskite $La_5Mo_4O_{16}$ by measuring the superlattice peaks.

$La_5Mo_4O_{16}$ consists of two octahedra of Mo^{4+} ($4d^2$, $S = 1$) and Mo^{5+} ($4d^1$, $S = 1/2$), and only $Mo^{4+}O_6$ octahedra are connected by nonmagnetic Mo_2O_{10} pillars along *c* [1]. Both Mo^{4+} and Mo^{5+} have orbital degree of freedom. $La_5Mo_4O_{16}$ undergoes in-plane collinear antiferromagnetic (AFM) order with AFM stacking along the *c* axis below $T_N = 190$ K [2]. Other transitions take place at $T_1 = 60$ K and $T_2 = 90$ K [3]. We call the phases at $T < T_1$ and $T_1 < T < T_2$ the low-*T* and middle-*T* phases, respectively. In our previous neutron scattering study at SENJU, MLF, J-PARC, we found superlattice peaks with $\delta \sim (0.5, 0.5, 0)$ and (0.6, 0.2, 0). On the other hand, the superlattice peaks are not detected by X-ray diffraction. These results indicate that the possible orbital order is induced by oxygen. However, the exact positions and the temperature dependence of the superlattice peaks were still unclear. Here, we measured the superlattice peaks, especially their temperature dependences, by means of a single-crystal neutron diffraction technique.

We used a $La_5Mo_4O_{16}$ single crystal with a mass of 40 mg whose scattering plane was (*HK*0). The neutron diffraction measurements were performed at 5G in JRR-3. Neutrons with $E_i = E_f = 34.05$ meV were used, and collimation was "open-80′-80′-open". We used a non-focusing PG monochromator and a focusing PG analyzer. The GM cryostat was used.

We searched the superlattice peaks at 3 and 75 K. We found the peak at $Q = (6.50(1), 3.50(1), 0)$ at 3 K, while the peak at $Q \sim (5.59(1), 5.21(1), 0)$ was found at 75 K. Therefore, the superlattice peaks with $\delta = (0.5, 0.5, 0)$ and (0.59, 0.21, 0) evolve in the low-*T* and middle-*T* phases. To see the temperature evolution of the superlattice peaks in detail, we performed θ-2θ scans for the (6.5, 3.5, 0) peak, and the integrated intensity as a function of temperature is shown in Fig. 1 (black symbols). The emergence of the peak below 60 K in the low-*T* phase can be seen. We also performed "sit-and-count" measurements at $Q = (5.6, 5.2, 0)$. The background subtracted intensity as a function of temperature is shown in Fig. 1 (red symbols). Interestingly, the peak can be seen only in the middle-*T* phase.

In summary, we have measured the temperature dependence of two superlattice peaks. We successfully identified superlattice peaks with $\delta = (0.5, 0.5, 0)$ and (0.59, 0.21, 0) in the low-*T* and middle-*T* phases, respectively. The results suggest that there are two different orbital orders in different phases of $La_5Mo_4O_{16}$.

References

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

[2] K. Iida *et al.*, J. Phys. Soc. Jpn. **86**, 064803 (2017).

[3] T. Katsufuji *et al.*, Phys. Rev. Research **3**, 013105 (2021).

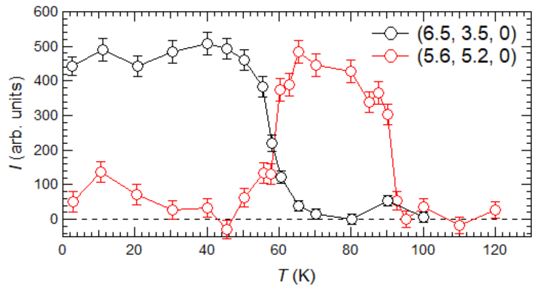

Fig. 1. Temperature dependence of the superlattice peaks at $Q = (6.5, 3.5, 0)$ (black) and (5.6, 5.2, 0) (red).