Magnetic Ordering Stabilized by Quadrupolar Ordering

in the Spin-Orbit Coupled Insulator Ba₂MgReO₆

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Multipole ordering is one of the most fundamental orders exhibited by d-electrons, in which spins and orbitals are entangled [1,2]. However, experimental results that can be compared with theory are limited due to the lack of model materials and difficulties in experimental observation. Currently, Ba₂MgReO₆ is the only material for which multipole ordering in the *d*-electron system has been established [3,4]. Clarifying the details of the anomalous noncollinear magnetic structure, which is closely related to quadrupolar ordering, will validate the theory and provide insight into the interaction between pseudospins in spinorbit coupled states. The moment size of pseudospins will also allow quantitative evaluation of the spin-orbit coupled electronic states. Comparison of theoretical and experimental results will lead to a microscopic understanding of the quantum phases produced by spin-orbitally entangled *d*-electrons.

This study aims to quantitatively determine the magnetic structure stabilized by quadrupolar ordering in Ba2MgReO6. In particular, we performed single crystal neutron diffraction measurements to reveal the moment size and orientation of the pseudospins. We have performed neutron diffraction (ND) experiments on a single crystal of Ba2MgReO6 with the size of $3 \times 3 \times 2.5$ mm³ in a 3 K closed-cycle refrigerator mounted on а four-circle diffractometer. The measurement was conducted with an incident neutron beam with wavelength $\lambda = 1.246$ Å at FONDER, JRR-3.

As shown in Fig. 1, fundamental reflections such as 4 4 0 reflection are clearly observed at 3 K. The obtained structural parameters and lattice constants are consistent with the previously reported values for Ba₂MgReO₆ [4]. Then, we attempted to observe magnetic reflections at 3 K. According to the previous resonant X-ray scattering experiments, the propagation vector of the magnetic order below $T_{\rm N} = 18$ K is reported to be $\mathbf{k} = [0\ 0\ 1]$ [4]. We have performed measurements at various expected magnetic reflection positions. However, no magnetic reflection was observed. It was found that the moment size of pseudospin was too small to detect under the experimental conditions. (smaller than our expectation of 0.4 $\mu_{\rm B}$) The following measurements will be performed on a single crystal with increased crystallinity and detwinned by applying pressure or a magnetic field in order to overcome the challenges.

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[3] D. Hirai *et al.* J. Phys. Soc. Jpn. **88**, 064712 (2019).

[4] D. Hirai *et al.*, Phys. Rev. Research, **55**, 10701 (2020).

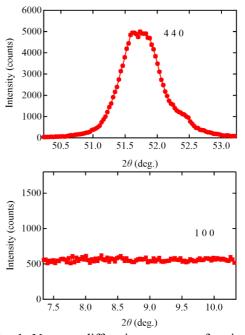


Fig. 1. Neutron diffraction patterns of a single crystal of Ba_2MgReO_6 around 4 4 0 and 1 0 0 reflection positions at 3 K.