Quadrupolar Orders in the Spin–Orbit-Coupled 5d Mott Insulator Ba₂MgReO₆

The crystal structure of the double perovskite Ba₂MgReO₆, having spin–orbit-entangled 5d electrons, was investigated by X-ray diffraction measurements using high-quality single crystals. The high-intensity and high-resolution synchrotron X-ray source at BL-8A and AR-NE1A enabled us to detect extremely small structural changes through the quadrupole order transition at \( T_q = 33 \) K. We observed a slight elongation and a rhomboid distortion of the ReO₆ octahedron, which reveal that the quadrupole order is composed of antiferroic \( Q_{u,v} \) and ferroic \( Q_{u,v} \) quadrupole moments. These findings demonstrate a unique symmetry-breaking of the multipolar degree of freedom in 5d electron systems.

Recently, 5d transition metal (TM) compounds have been attracting attention because of the combined effect of electron correlations and spin–orbit interactions (SOIs). For example, strong SOIs in Sr₃IrO₆ effectively enhance electron correlations, and a spin–orbit-entangled Mott insulating state is realized [1]. The 5d electrons with strong spin–orbit entanglement may experience various symmetry-breaking transitions, called multipolar orders [2]. Indeed, multipolar orders are predicted to be realized in 5d electron systems has not yet been experimentally observed for the superlattice reflections, we conclude that the actual quadrupolar order is a linear combination of the \( Q_{u,v} \) and \( Q_{u,v} \) orders. Our diffraction study establishes the existence of quadrupolar order for the correlated spin–orbit-entangled d electrons in the DP Ba₂MgReO₆. This compound provides an opportunity to experimentally investigate the symmetry-breaking of the multipolar degree of freedom in 5d electron systems.

REFERENCES

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Figure 1: a Double perovskite structure of Ba₂MgReO₆. b The peak splitting of the (0 0 24) reflection at \( T_c \). c Temperature dependence of the intensity of a superlattice reflection (4 1 0) of the tetragonal cell.

Figure 2: a The quadrupolar moments \( Q_{u,v} \) and \( Q_{u,v} \), which couple with the \( c \) and \( e \) displacement modes represented by the green and blue arrows, respectively. b Observed quadrupolar order patterns in Ba₂MgReO₆.