Magnetic neutron diffraction study on alkali-metal superoxide CsO_2

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In the alkali-metal superoxide CsO_2, the doubly degenerate π^* orbital of the O_2^- molecule accommodates three electrons and is in the spin-1/2 state. The crystal structure at room temperature is tetragonal (I4/mmm), and structural phase transitions occur at 150 K and 70 K. Both of the two phases at low temperatures appear to be orthorhombic in X-ray diffraction, but their crystal structures have not been clarified. The magnetic susceptibility shows a broad maxima at low temperatures and fits well with the Bonner-Fisher model of 1D Heisenberg antiferromagnetism [1,2]. Antiferromagneic order occurs below the Néel temperature of T_N = 9.6 K. A density functional theory (DFT) calculations showed that the crystal symmetry lowering is due to the tilting of O_2^- molecules, and the π^* orbital ordering due to the cooperative Jahn-Teller effect is predicted to give rise to the one dimensional magnetism [1]. However, these mechanisms have not been clarified experimentally, since there is no information on the crystal structure and magnetic structure at low temperatures. In this study, we aim to clarify the magnetic structure at the lowest temperature by neutron diffraction, and to elucidate the mechanism of the peculiar magnetism in CsO_2.

CsO_2 powder samples were synthesized at Okayama Univ. and neutron diffraction measurements were performed at JRR-3 T1-1. We found that the 1/2 0 1 diffraction peak appeared near T_N and the intensity increased upon cooling, confirming that it was a magnetic reflection. In addition, the 1/2 0 0 reflection also appeared at low temperatures. On the other hand, the 001 reflection was not observed. From these results, the most plausible magnetic structure is one in which the magnetic moments are arranged antiferromagnetically in the *a*-axis and ferromagnetically in the *b*-axis, as shown in Fig. 1. The direction of the easy axis has not yet been determined. This magnetic structure is close to the prediction by Riyadi *et al* [1]. The magnitude of the ordered moment was found to be strongly suppressed to about 0.2 μ_B.

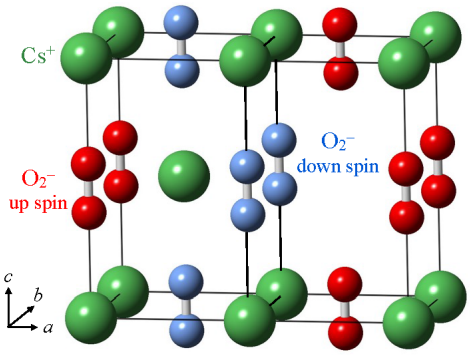

Fig. 1. Schematic illustration of magnetic structure of CsO_2 expected from the present study. The magnetic moments of the red-colored and the blue-colored O_2^- molecules order antiparallel to each other.

[1] S. Riyadi *et al*., Phys. Rev. Lett. **108**, 217206 (2012).
[2] M. Miyajima *et al*., J. Phys. Soc. Jpn. **87**, 063704 (2018).