Tetra-magnetism: all-in/all-out spin arrangement on the pyrochlore lattice

Pyrochlore oxide is a class of minerals in various substance groups of the form $A_2B_2O_7$ ($A_2B_2O_6O'$). The cubic pyrochlore structure possesses high symmetry in the space group $Fd\bar{3}m$. The structure comprises two interpenetrating networks composed of A-O' and B-O. The pyrochlore lattice is a three-dimensional network of tetrahedra sharing a vertex obtained by connecting only A or B atoms (see the inset of Fig. 1). Pyrochlore oxides have been studied extensively to search for exotic magnetic phenomena. When spins reside on the vertices of tetrahedra and interact antiferromagnetically with each other, a conventional magnetic long-range order like a Néel order tends to be suppressed owing to geometrical frustration. It is theoretically predicted that a quantum mechanically liquid state with a large fluctuation is realized even at zero temperature.

The pyrochlore oxide $Cd_2Os_2O_7$ discovered in 1974 shows a metal-insulator (MI) transition with an antiferromagnetic transition at 225 K [1]. A Slater transition, designated as a mechanism of an MI transition produced by the doubling of the unit cell due to the antiferromagnetic order, has been indicated. However, no magnetic peaks for identifying the magnetic structure in the insulating phase have been observed yet by means of neutron scattering. Thus, to reveal the magnetic structure, the resonant X-ray magnetic scattering method (RXMS) was employed, instead of the neutron scattering method, which suffers from the Cd absorption problem. X-ray magnetic scattering is a method of selectively enhancing the signal of a substance by using a resonant effect at the absorption edge of the element.

The measured sample was a high-quality single crystal of $Cd_2Os_2O_7$ with a 0.2×0.2 mm^2 surface of the (001) plane grown in a temperature gradient furnace. RXMS experiments were carried out at the absorption edge of Os at beamlines **BL19LXU** and **BL02B1**. Figure 1 shows the X-ray absorption spectra near the Os-L_{III} edge and the intensity of the 006 reflection at 10 K [2]. The peak-maximum position of 006 reflection is in good agreement with the Os-L_{III} edge, as depicted by the dotted line, implying the enhancement of the 006 reflection intensity by the resonance effect. The 006 reflection observed indicates the violation of the $Fd\bar{3}m$ extinction rule. Moreover, neither additional reflection breaking of the face-centered lattice nor long-period structures was observed.

Figure 2(a) shows the temperature dependence of the intensity of the 006 forbidden reflection, which increases gradually below 227 K, indicating a continuous second-order transition. Anomalies in forbidden reflection, magnetic susceptibility, and resistivity are observed at nearly the same temperature. Moreover, in the 006 forbidden reflection the polarization dependence emerges as shown in Fig. 2(b); the intensity was observed in only the π (incident beam) - σ' (scattered) channel. From these results, we therefore conclude that a commensurate magnetic structure appears at the propagation vector $\boldsymbol{q} = 0$ at low temperatures in this compound.

The $\boldsymbol{q} = 0$ magnetic structure should be described in terms of an Os tetrahedron because the magnetic primitive cell includes one Os tetrahedron. Here, the possible spin arrangements of the Os site can be classified by representation analysis; that is, the spin arrangements of the 12-basis functions are given. The basis functions represent one and eight kinds of spin arrangement with cubic and tetragonal magnetic symmetries, respectively. All of these magnetic structures are noncollinear antiferromagnetic spin arrangement with a zero net moment on the Os tetrahedron. The other three kinds of spin arrangement represent ferromagnetic spin arrangements. Generally, in a transition metal oxide, the lattice and magnetic structure are strongly coupled through exchange striction and spin-orbit (SO) interaction, resulting in a low-symmetry spin alignment that almost always induces crystallographic distortion. Thus, the magnetic symmetry is expected to be associated with the lattice symmetry in $Cd_2Os_2O_7$. Here, the preservation of the cubic spatial symmetry was confirmed by Raman scattering measurement, which is the most powerful technique for detecting small lattice distortions [2]. Therefore,

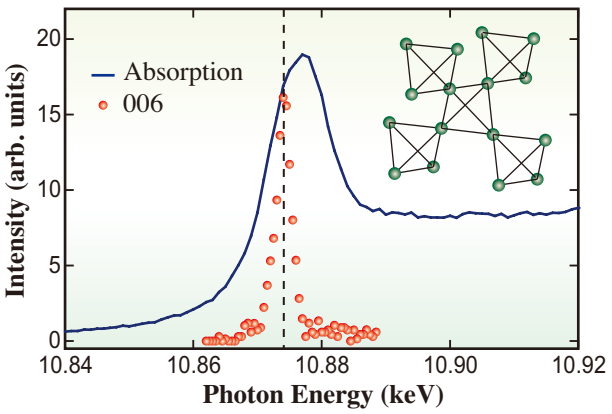

Fig. 1. Spectra of the intensity of the 006 forbidden reflection at 10 K and X-ray absorption near the Os-L_{III} edge at room temperature. The inset shows the pyrochlore lattice.

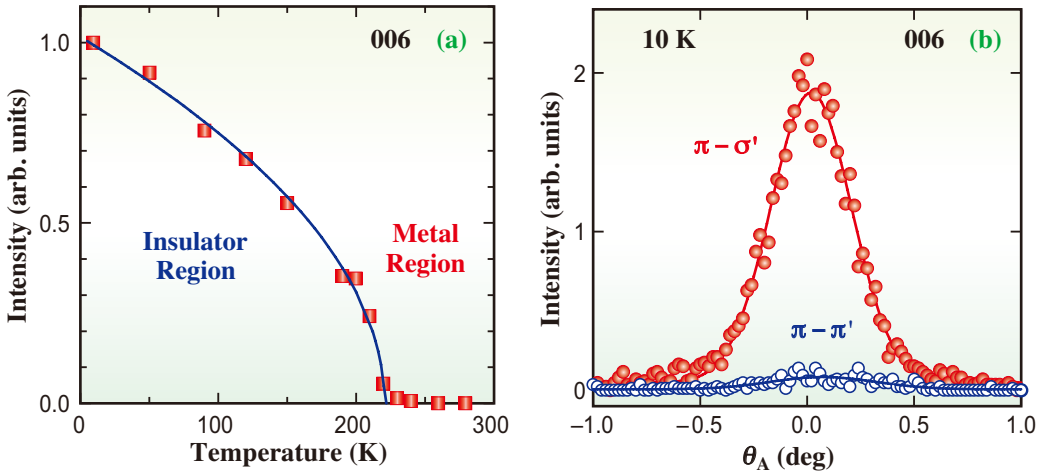

Fig. 2. **(a)** Temperature dependence of the integrated intensity of the 006 forbidden reflection. **(b)** Polarization dependence of the 006 magnetic reflection at the Os-L_{III} edge at 10 K.

only the spin arrangement with cubic magnetic symmetry is a unique solution to the magnetic ordered structure. The arrangement denotes the all-in/all-out antiferromagnetic spin arrangement with a zero net moment on the Os tetrahedron, in which all the spins point to (all-in) or away (all-out) from the center of the tetrahedron, as illustrated in Fig. 3. This spin arrangement is identical to the results of the polarization dependence. The all-in/all-out magnetic structure can be regarded as a negative/positive magnetic octupole: the term "tetra-magnetism" would be designated by the pair of negative/positive magnetic octupoles, which is different from the collinear-type antiferromagnetism and helimagnetism. The tetra-magnetism compound is expected to exhibit interesting physical properties such as an anomalous magnetization and a magnetostriction effect [3].

The present spin arrangement with $\boldsymbol{q}$ = 0 indicates that these transition is not due to Slater-type transitions. Consequently, the band-gap formation of $Cd_2Os_2O_7$ is thought to arise from an alternative mechanism that is presumably related to the all-in/all-out spin arrangement: a tetrahedral magnetic structure on the pyrochlore lattice without any spatial symmetry breaking. The emergence of a tetrahedral magnetic order triggers such cooperative band shifts so as to form a small gap through a strong SO coupling. Actually, it was proposed that the SO coupling plays a significant role in band-gap formation in the band structure calculation [4].

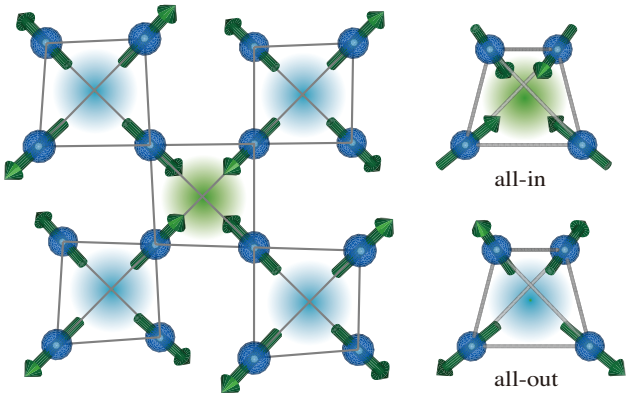

Fig. 3. The all-in/all-out magnetic structure is composed of Os tetrahedral "all-in" and "all-out" units.

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