X-Ray Detection of Magnetic Octupole Order Producing a Large Anomalous Hall Effect

We have observed X-ray magnetic circular dichroism (XMCD) arising from the magnetic octupole order in an antiferromagnetic metal, Mn_3Sn. This material has recently been attracting attention due to the large anomalous Hall and thermoelectric responses with a novel type of mechanism. Based on a comparison between experiment and theoretical calculation, we have revealed that the present XMCD signal arises from the magnetic octupole order, which has been difficult to detect so far. This finding suggests that XMCD could be useful for detecting higher-rank multipoles, and thus may expand the applicable targets of X-ray magnetic spectroscopy.

Recently, the anomalous Hall and thermoelectric (Nernst) effects in magnetic metals have attracted much attention as a potential factor for advanced spintronics, energy harvesting, and magnetic sensing. To date, these properties were mostly studied in ferromagnets, where the spin moments are uniformly aligned. In contrast, large anomalous Hall and Nernst effects are also found in an antiferromagnetic metal, Mn_3Sn [1]. Mn_3Sn has a triangular antiferromagnetic structure, and thus the total magnetization is vanishingly small (~0.002 μ_B/Mn) as depicted in Fig. 1(a). Therefore, the mechanism is expected to be different from that of ferromagnets. Theoretically, the origin is understood to be a novel type of order parameter called cluster magnetic octupole [2]. However, the magnetic octupole order has not been well established experimentally. To solve this problem, we have applied X-ray magnetic circular dichroism (XMCD) to Mn_3Sn [3]. Recent theoretical studies [4, 5] suggest that the XMCD signal can be detected in triangular antiferromagnets like Mn_3Sn through the magnetic dipole term called T_z term.

Figure 1(a) shows a schematic illustration of the experimental setup for XMCD measurement. Part of the experiment was performed at BL-16A. The angle between the X-ray beam and (0001) plane is defined as θ, and the magnetic field is parallel to the X-ray beam. Figure 1(b) shows a typical XMCD signal of Mn_3Sn for B = 0.1 T and 1 T. Clear positive and negative XMCD signals were observed at L_3 and L_2 edges, and this feature is indeed opposite to that of conventional ferromagnetic Mn compounds, where the XMCD signal is negative and positive for L_3 and L_2 edges, respectively. This anomalous sign of XMCD is one of the characteristics of the present XMCD. Also, the signal intensity and the shape of XMCD are mostly independent of the field strength. This feature is not consistent with the magnetization process, where the magnetization increases about four times between 0.1 to 1 T with a linear slope as shown in the inset. These features clearly indicate that the present XMCD is decoupled from the magnetization behavior, meaning that the origin of XMCD is different from that of the conventional XMCD, where the XMCD response is proportional to the static magnetization.

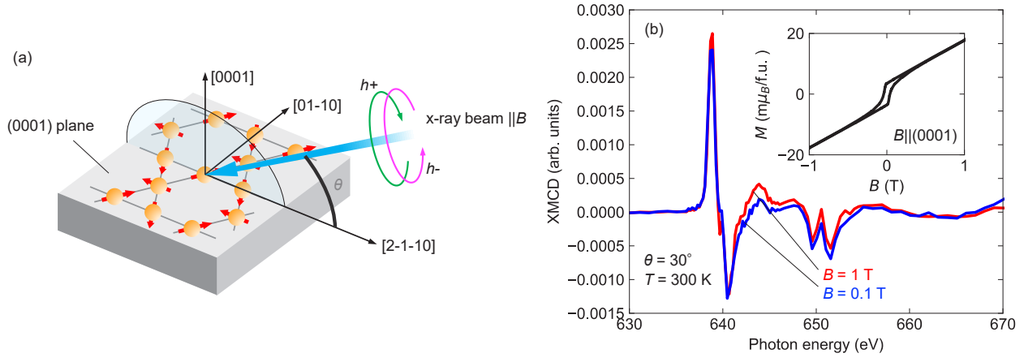

Figure 1: (a) Schematic of experimental setup of XMCD measurement. (b) Typical XMCD signal of Mn_3Sn for B = 0.1 and 1 T. (inset) Magnetization process of Mn_3Sn up to 1 T.

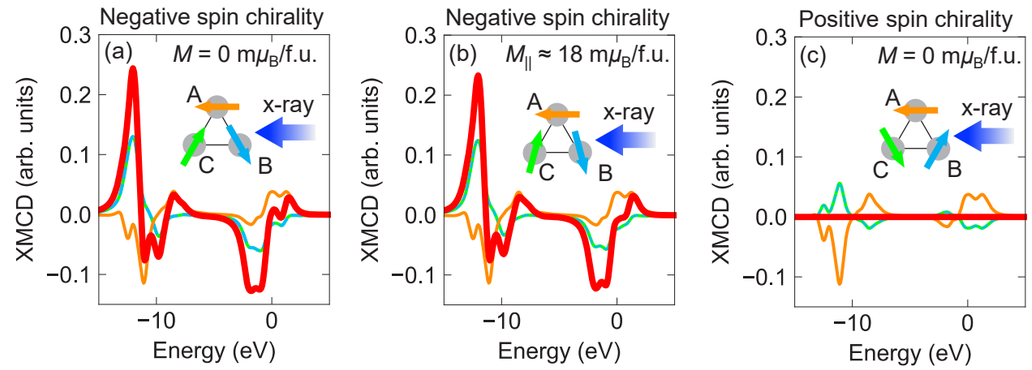

Figure 2: (a, b) Calculated XMCD signals for the case of negative spin chirality without in-plane magnetic field (a) and slight in-plane field corresponding to B ≈ 1 T (b). (c) Calculated XMCD for positive spin chirality.

To clarify the origin of the present XMCD, we performed theoretical model calculations of the XMCD spectrum [Fig. 2(a)–(c)]. Thick red lines indicate the total XMCD signal, which is the sum of each XMCD contribution from antiferromagnetic sublattices A, B, and C (indicated by thin orange, light blue, and light green lines, respectively). The corresponding magnetic structure is shown in the insets. In Fig. 2(a), a clear XMCD signal remains even though the sum of spin moment is zero, and the shape of XMCD is reasonably consistent with the experimental observation. Also, the calculated XMCD is mostly independent of the in-plane magnetic field as shown in Fig. 2(b). This means that the present XMCD does not originate from the field-induced moment but from the triangular magnetic structure itself, which is mostly preserved in such a weak magnetic field. On the other hand, in Fig. 2(c), individual XMCD contributions compensate each other, and thus the total XMCD is zero. The difference of the two magnetic structures in Fig. 2(a) and (c) is the sign of spin chirality. For the inset of Fig. 2(a), the spin moment rotates counterclockwise (–120°) when the position of the antiferromagnetic sublattice is changed clockwise as A to C. In this case, the spin chirality is negative. On the other hand, the spin chirality is positive for the magnetic structure of Fig. 2(c) since the spin direction rotates clockwise (+120°) for the same operation. Importantly, based on the group theory consideration, the TRS-broken cluster magnetic octupole only appears when the spin chirality is negative [2]. Thus, our calculation shows complete correspondence between the appearance of XMCD and cluster octupole order. Note that the magnetic structure of Fig. 2(a) is realized in Mn_3Sn, while that of Fig. 2(b) is a fictitious magnetic structure for comparison.

In this study, we have demonstrated that the unconventional XMCD in Mn_3Sn arises from the triangular antiferromagnetic structure characterized by the TRS-broken cluster magnetic octupole order. Our experimental observation shows the efficiency of XMCD for observing ferroic higher-rank multipole order. The present work may expand the applicable targets of X-ray magnetic spectroscopy.

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M. Kimata (Tohoku Univ.)