Experimental Evidence of Orbital Ferrimagnetism by X-Ray Magnetic Circular Dichroism

We have experimentally investigated the magnetic properties of $CoMnO_3$(0001) epitaxial films known to exhibit orbital ferrimagnetism. The films were grown by the reactive rf magnetron sputtering method. The magnetic anisotropy of $CoMnO_3$(0001) thin films was found to have a large negative value of K_u=–15.6±0.8 Merg/cm^3 at 300 K. X-ray magnetic circular dichroism revealed that the spin momenta of Mn and Co ions are oriented opposite to each other and a large orbital momentum comparable to the spin angular momentum emerges only on Co, indicating the orbital ferrimagnetic nature of $CoMnO_3$.

$CoMnO_3$ has an ilmenite structure ($R\bar{3}$)[1–3] in which Co^{2+} and Mn^{4+} layers are alternately stacked along the *c*-axis [3]. Since Co^{2+} (d^7HS) and Mn^{4+} (d^3) possess S = 3/2, and are antiferromagnetically coupled through the superexchange interaction, the spin angular momenta of Co and Mn cancel each other out. However, the orbital angular momentum of Co^{2+} in the crystal field remains [3]. Since the experimentally estimated value of the magnetic moment is 0.72 μ_B/f.u. (M_s = 140 emu/cm^3 at 2 K), i.e., the same order of magnitude expected of the orbital angular momentum of Co^{2+} in an octahedral crystal field, this compound is termed an "orbital ferrimagnet."

Because of the remaining orbital angular momentum, $CoMnO_3$ possesses significantly large magnetic anisotropy. Cloud and Jesson succeeded in roughly estimating the magnetic anisotropy of $K_{u1}+2K_{u2}$~–14 ± 2 Merg/cm^3. They found that the *c*-axis is a magnetically hard axis, indicating that $CoMnO_3$ has a negative magnetic anisotropy constant [4]. Both the large orbital momentum and the large negative uniaxial anisotropy were partially explained within the framework of the single ion model of Co^{2+}, originally adopted for $CoFe_2O_4$ by Slonczewski [5]. Moreover, $CoMnO_3$ has a Néel temperature of 391 K [1]. Because of the unique origin of magnetism with a high transition temperature, $CoMnO_3$ is an attractive compound from the viewpoint of both fundamental magnetism and applications including novel devices for spintronics based on the orbital angular momentum. However, previous experimental works regarding $CoMnO_3$ were mainly carried out on miniature single crystals, powder, or ceramic samples, not on sufficiently large single crystals or epitaxial films suitable for investigating the details of various properties [1–3, 6, 7]. Therefore, the magnetic and other physical properties of $CoMnO_3$ are not yet well understood.

$CoMnO_3$ thin films were grown by the reactive rf magnetron sputtering technique. X-ray magnetic circular dichroism (XMCD) experiments were also performed at BL-16A to evaluate the element-specific spin angular and orbital angular momenta [8–10]. XMCD measurements for the film were performed at room temperature. The photon energy range was for the Mn and Co $L_{2,3}$ edges. The XAS were determined by the total electron yield (TEY) method. A set of XAS (μ_+ and μ_-) was measured using circularly polarized light with opposite helicity in a magnetic field of 50 kOe, applied parallel to the photon direction. The film plane was inclined at 30° to both the incident photons and the magnetic field. The magnetic anisotropy constant (K_u) was measured by a magnetotorque meter.

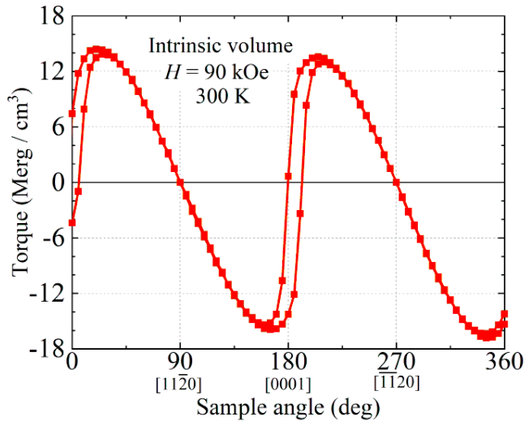

Figure 1: Magnetotorque curve of a $CoMnO_3$ thin film with a film intrinsic thickness of 69 nm and a nominal thickness of 90 nm at 90 kOe. The intrinsic thickness was evaluated by subtracting the magnetic dead layer from a nominal thickness.

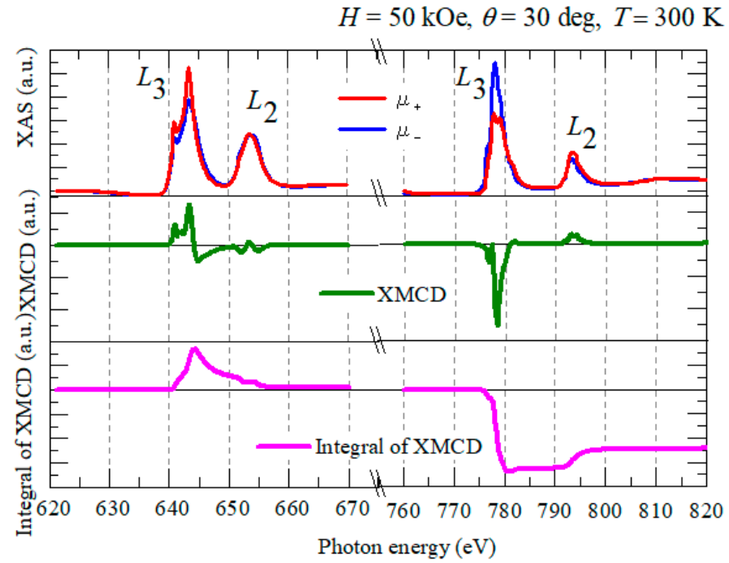

Figure 2: Mn and Co circularly polarized XAS (μ_+ and μ_-), XMCD spectra ($\mu_+ - \mu_-$), and the integral of XMCD spectra.

The magnetotorque curve of the $CoMnO_3$ thin film at 90 kOe is shown in Fig. 1. The measurements were performed at 300 K. The torque curve indicates that the magnetic easy axis lies in plane. Even at 90 kOe, the torque curve exhibits a sawtooth-wave-like shape with clear rotational hysteresis, indicating that the anisotropy field is significantly greater than 90 kOe. The uniaxial magnetic anisotropy energy can be written as $E = K_u^{\mathrm{eff}} \sin^2\theta$, where K_u^{eff} and θ are the effective uniaxial magnetic anisotropy constant and the angle between magnetization ***M*** and the normal to the film, respectively. The observed K_u^{eff} comprises both magnetic anisotropy contributions from magnetocrystalline anisotropy K_u and shape anisotropy. K_u^{eff} is determined from peak to peak of the torque curve. Note that shape anisotropy is negligibly smaller than the value of K_u. Therefore, the intrinsic $K_u \approx K_u^{\mathrm{eff}}$ of the $CoMnO_3$ thin film is found to be –15.6 ± 0.8 Merg/cm^3 at 300 K, which is almost the same as the previously reported value of K_u = –14 ± 2 Merg/cm^3 at room temperature [4].

Figure 2 shows Mn 2*p* XAS and XMCD spectra as well as those of Co 2*p* observed for the $CoMnO_3$ film. From XMCD sum rules of the XMCD spectra shown in Fig. 2, the m_{orb} / m_{spin} ratios are determined to be 0.02 ± 0.00 for Mn and 0.33 ± 0.03 for Co. It is obvious that the larger contribution of the orbital angular moment comparable to the spin angular moment was found only for Co. In addition, by comparing the shapes of the XMCD spectra for Co and Mn, we can conclude that the magnetic moments of Mn and Co are oriented in opposite directions. These results strongly indicate the orbital ferrimagnetic nature of $CoMnO_3$.

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BEAMLINES

BL-12C and BL-16A

H. Koizumi[1], S. Sharmin[1], K. Amemiya[2, 3], M. S-Sakamaki[2, 3, 4], J. Inoue[1] and H. Yanagihara[1, 5] ([1]Univ. of Tsukuba, [2]KEK-IMSS-PF, [3]SOKENDAI, [4]Gunma Univ., [5]TREMS)