Spin and orbital magnetization loops obtained using magnetic Compton scattering

The magnetic moment of a material is the sum of the spin (μ_S) and orbital moments (μ_L). Both are fundamental quantities for understanding the macroscopic magnetic properties of materials. However, there are very few experimental techniques that can be used to disentangle the spin and orbital contributions from the total magnetic moment. Most conventional magnetometer-based techniques, e.g., vibrating sample magnetometry (VSM) and superconducting quantum interference device magnetometry (SQUID), can only measure the total magnetic moment of the samples. X-ray magnetic circular dichroism (XMCD) is one of the useful tools that provide the ratio μ_L/μ_S within the framework called the sum rule procedure.

In this report, an X-ray magnetic Compton scattering (MCS) technique is described. MCS is one of the useful techniques for disentangling the spin and orbital contributions. Since MCS probes only the spin magnetization, it can provide the absolute value of the spin component of magnetization. Combined with conventional techniques, the orbital component of magnetization and its curve as a function of magnetic field can be uniquely deduced. Recent improvements of the MCS technique [1] can successfully separate the magnetic part from total Compton scattering as a function of magnetic field, enabling independent measurements in the positive and negative sides of spin magnetization curves. This development is useful for magnetic materials with exchange spring coupling, in which the magnetization curves show asymmetry with respect to the origin.

When the incident X-rays are circularly polarized, the cross section for Compton scattering is given as [2,3]

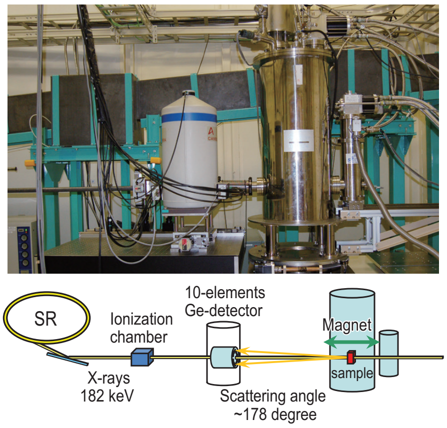

Fig. 1. Photograph and schematic drawing of MCS spectrometer.

$$\frac{d\sigma}{d\Omega} = \left(\frac{d\sigma}{d\Omega}\right)_{charge} + \left(\frac{d\sigma}{d\Omega}\right)_{spin}$$
$$= C_{charge}N + C_{mag}P_c\mathbf{S}\cdot(\boldsymbol{k}\cos\theta + \boldsymbol{k}')\mu_s$$

where P_c is the degree of circular polarization of X-rays, $\mathbf{S}$ is the spin direction, $\boldsymbol{k}$ ($\boldsymbol{k}'$) is the wavevector of incident (scattered) X-rays, and θ is the scattering angle. C_{charge} and C_{mag} are constants. The first term, which contains the total number of electrons, N, corresponds to the charge Compton scattering. The second term is the magnetic term that contains the spin moment, μ_S.

The scattered X-ray intensity is proportional to the scattering cross section. To determine the absolute spin moment in a sample, we measure the intensities of Compton scattered X-rays from the sample under sampling condition (I) and non-magnetized condition (I_{non}). The magnetic effect R is defined by the following equation.

$$R = \frac{I - I_{non}}{I_{non}} = \frac{I_{spin}}{I_{charge}}$$
$$= \frac{C_{mag}P_c\mathbf{S}\cdot(\boldsymbol{k}\cos\theta + \boldsymbol{k}')}{C_{charge}} \cdot \frac{\mu_s}{N} = A\left(\frac{\mu_s}{N}\right)$$

Once the coefficient A is given, the spin moment can be determined from the experimentally determined magnetic effect R, since the total number of electrons, N, is known. The coefficient A is usually determined by measuring a well-characterized material, such as Fe metal, the spin moment of which is already known.

The demonstrations [1] were carried out at the High Energy Inelastic Scattering beamline **BL08W**. Elliptically polarized X-rays emitted by the elliptical multipole wiggler are monochromatized to 182.6 keV with a degree of circular polarization of ~0.55. The spectrometer consists of a 3T-superconducting magnet, a sample cryocooler and a 10-element Ge detector. Compton scattered X-rays from a sample are detected by the detector at a scattering angle of 178 degrees.

Figure 2(a) shows the MCS intensity loop (filled circles) and VSM data (solid line) of $GdAl_2$ at 5 K. The total data acquisition time for MCS is ~14 h. The total count, which includes both the charge and magnetic contributions, is ~2×10^7 at each data point. The orbital moment of ferromagnetic $GdAl_2$ is almost quenched so that VSM can measure the response of the spin magnetic moment to the applied magnetic field. Thus, $GdAl_2$ is a good magnet for a feasibility test and the

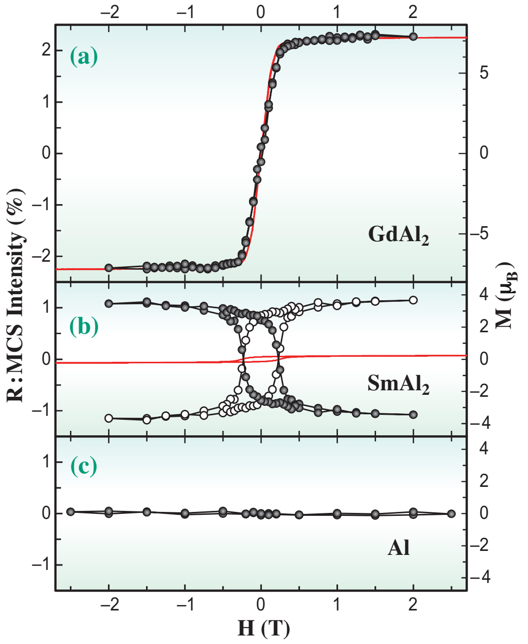

Fig. 2. MCS intensity loop (filled circles) and magnetization loops measured by VSM (solid line) of **(a)** GdAl$_2$, **(b)** SmAl$_2$ and **(c)** Al. The MCS intensity is corresponds to spin magnetization. The orbital magnetization loop (open circles in (b)) was obtained by subtracting the spin magnetization from the total magnetization data by VSM.

calibration of the MCS spectrometer. The left axis shows the magnetic effect *R*, which is the ratio of MCS intensity to charge Compton scattering intensity and the right axis shows the magnetization measured by VSM. The MCS spectrometer was calibrated by fitting the MCS intensity curve to the VSM data. Hence, the spin magnetization can be evaluated in the unit of Bohr magneton in later experiments using the spectrometer. The MCS intensity loop is in excellent agreement with the VSM data, demonstrating that the MCS loop captures the response of spin magnetization to the applied magnetic field.

Figure 2(b) shows the MCS intensity loop at 10 K (filled circles) as well as the VSM loop at 5 K (solid line) for SmAl$_2$. The total count is 2.6×10^7 at each point and the total data acquisition time is ~8 h. The MCS intensity is calibrated by GdAl$_2$ data, and the corresponding spin magnetization is given as μ_B in the right axis. In contrast to GdAl$_2$, the MCS intensity loop differs in its magnitude and sign from the VSM loop because of a large orbital magnetization in SmAl$_2$. The VSM data show a total magnetization of 0.2 μ_B at 2 T, while the MCS intensity loop reveals a spin magnetization of 3.5 μ_B. This leads to the orbital magnetization of 3.7 μ_B at 2 T. The spin and orbital magnetizations obtained here are in good agreement with the results of a previous work [4]. The orbital magnetization loop (open circles), obtained from the difference between the VSM and MCS intensity loops, shows an almost reverse hysteresis feature compared with the spin magnetization loop. This result confirms that the spin and orbital magnetization loops cancel each other out to make a small total magnetization loop in SmAl$_2$.

The sensitivity of the MCS spectrometer is estimated from the MCS intensity loop of Al. The intensity is almost constant because of its non-magnetic property (Fig. 2(c)). The standard deviation in the magnetic effects, *R*, is ~0.025%, which is equivalent to ~0.025 μ_B for Fe and ~0.07 μ_B for Gd as the minimum limit of spin magnetization measurement.

The temperature-dependent magnetization is also measured. Figure 3 shows the MCS intensity of SmAl$_2$ as a function of temperature at a magnetic field of 2.5 T. The MCS intensity starts to increase at around 100 K and then rapidly increases as the temperature decreases. The temperature dependences show almost the same curves with decreasing and increasing temperature.

Finally, the decomposition of total magnetization into spin and orbital contributions serves as a probe of atomic-level magnetic anisotropy and coupling, and plays an essential role in understanding the magnetization reversal process. This MCS technique is a new means for studying ferro- and ferrimagnetisms.

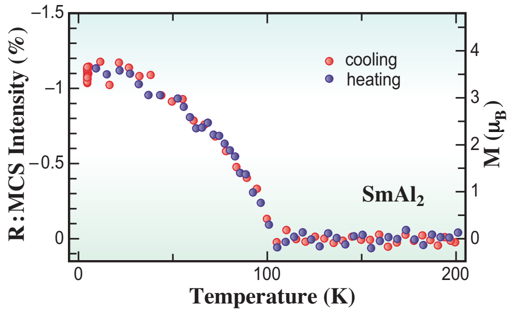

Fig. 3. Temperature dependence of MCS intensity of SmAl$_2$.

Masayoshi Itou

SPring-8/JASRI

E-mail: mito@spring8.or.jp

References

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