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Incident angle dependence of MCD at the Dy M_5-edge of perpendicular magnetic Dy_xCo_{100-x} films

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Abstract

X-ray absorption and magnetic circular dichroism spectra (XAS–MCD) of the Dy $M_{4,5}$-edges of Dy_xCo_{100-x} ($x = 15$–33) thin films are measured. The advantage of element and orbital selectivity in the XAS–MCD measurements enables us to directly observe the magnetic moments of specified elements. The MCD intensity at the Dy M_5-edge is maximum when the incident X-ray is normal to the surface and the incident angle dependence of the MCD intensity shows a sine curve. That gives us information about the angular distribution of the Dy magnetic moments with respect to the total magnetization direction and the opening angle of the cone. Applying the sum rule, the expectation value of orbital magnetic moment was estimated. The half opening angle of the random distribution of the moments were estimated and it was found that the angle has its minimum value around $x = 20$.

Keywords: Magneto-optical recording; Thin film; Perpendicular magnetism; X-ray absorption spectra; Magnetic circular dichroism; Orbital moment; Magnetic anisotropy; Full-multiplet calculation; Dy_xCo_{100-x}

1. Introduction

Magnetic amorphous films consisting of rare-earth and transition-metal (α-RE–TM) alloys [1–3] have attracted much interest because its characteristic magnetic properties are useful, making them leading candidates as materials for magneto-optic recording, a high-bit-density, reading–write storage technology. For example, the RE–TM film with a specific composition, the so-called compensation composition of RE and TM elements, exhibits strong perpendicular anisotropy and large Kerr rotations. By making the best use of the advantages of α-RE–TM, magneto-optical (MO) recording based on the RE–TM amorphous films with perpendicular magnetic anisotropy is currently established as a storage technology.

The magnetic properties, especially magnetic anisotropy energy and magnetization, of the RE–TM amorphous films have been studied by many researchers [4,5]. Generally, in RE-based magnetic materials, the origin of the magnetic anisotropy is said to be caused by the large orbital moment of the well-localized 4f electrons of an RE ion. Moreover, the magnetic moments of TM and heavy RE elements in the RE–TM alloy film are anti-parallel and the moment of individual heavy RE (except gadolinium) is oriented at random within a cone around the easy axis of magnetization [3]. The net magnetic moments are composed of RE and TM sub-network moments and the RE–TM film is, therefore, a

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ferri-magnet. The conical distribution of magnetic moments for the RE element is likely to play a significant role in the perpendicular magnetism and there still exists much research interest in the RE–TM films in order to elucidate the origin of the perpendicular anisotropy. It is important to study the relationship between the RE composition and microscopic magnetic properties such as expectation orbital moment L_z of the elements.

Magnetic circular dichroism (MCD) in the core-level absorption spectrum (XAS) in soft X-ray regions, namely, the differences between the absorption spectra with the parallel and anti-parallel helicity of the incoming circularly polarized X-ray respect to magnetization of sample, has been used for spectroscopic study of ferro- and ferri-magnetic materials [6]. Analyzing the XAS and MCD at the $M_{4,5}$ (3d to 4f transitions)-edges of the RE element, the 4f electronic and spin state of RE is revealed. In addition, XAS–MCD enables us to estimate the orbital and spin magnetic moments, projected to the incident light direction, applying the sum rule. To reveal the relationship between the perpendicular magnetic anisotropy and the electronic state of an α-RE–TM film, the value of orbital magnetic moments of the RE ion when the metal composition ratio of the RE–TM film or the kind of RE and TM elements in the film is changed should be investigated.

α-Tb–Fe–Co alloy has been intensively studied with the aim of improving storage density. We have reported the microscopic magnetic properties of α-Tb–Fe–Co thin film elucidated by means of XAS and MCD [7,8]. α-Dy–Co is one in the family of α-RE–TMs, however, there are only a few studies of this alloy. For improvement of storage density, a candidate for new recoding media is desired. α-Dy–Co can be a good candidate for this new material, for such as a switching layer, memory layer, and writing layer. As above-mentioned, analysis of XAS–MCD signals based on the sum rules provides us the orbital magnetic moment as well as the spin magnetic moment [9,10]. In this paper, the incident angle dependence of MCD intensity is investigated. The expectation values of magnetic orbital moment are estimated from MCD–XAS measurement. Comparing these with the full-multiplet calculation result, the half opening angle of the conical distribution of orbital moment for various dysprosium concentrations (x) was calculated.

2. Experimental methods

Dy_xCo_{100-x} ($x = 15$–33) thin amorphous films with perpendicular magnetization were deposited on a Cr underlayer (20 nm)/Si(1 0 0) single crystal substrate using a dc-magnetron sputtering system with a power of 300 W and deposition rate of 0.5 nm/s. The thickness of the formed amorphous films was 25 nm. The base pressure was less than 8×10^{-6} Pa and the Ar sputtering pressure in all sample preparations was 0.5 Pa. A 5 nm thick SiN layer was deposited on the top of the Dy_xCo_{100-x} films as a protective layer against oxidation. In this composition of $x = 15$–33, the Dy_xCo_{100-x} thin film has perpendicular magnetism. Saturation magnetization (M_s) and perpendicular coercivity (H_c) measured perpendicular to the film surface are shown as functions of dysprosium composition in Fig. 1. H_c is maximum and M_s is minimum value at about $x = 20$–25, respectively.

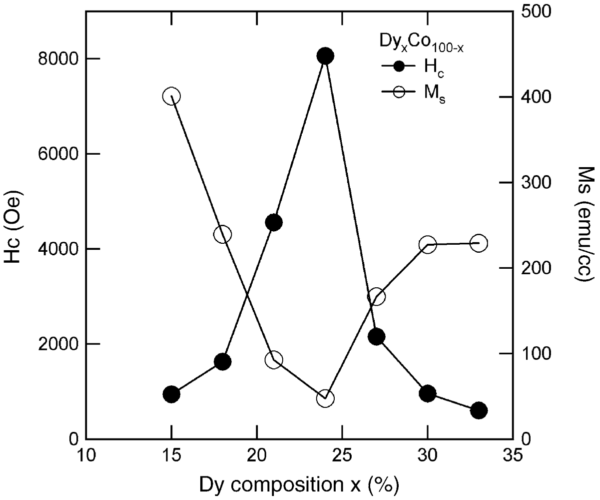

Fig. 1. M_s and H_c of Dy_xCo_{100-x} amorphous films measured perpendicular to the film surface as a function of Dy composition.

The measurements at the Dy $M_{4,5}$-edges of the Dy_xCo_{100-x} films were carried out using synchrotron radiation at the soft X-ray beamline BL23SU at SPring-8, Japan, where an APPLE-2-type undulator was adopted as a light source and a varied space plane grating type monochromator with a 1000 lines/mm grating was used. The energy-resolution $E/\Delta E$ was greater than 7000 during the measurements.

The current (I_1) was normalized to the incident photon intensity (monitor current, I_0) and normalized absorption intensity ($I = I_1/I_0$) was used for comparison. Hereafter, XAS intensities obtained by using photons of parallel and anti-parallel polarizations in the magnetization direction are denoted by I_+ and I_-, respectively. The MCD intensity was taken to be as the difference between the two absorption spectra, expressed as $I_{MCD} = (I_- - I_+)$. During MCD measurements, the undulator and monochromator were controlled synchronously, i.e., the photon helicity was switched at each photon energy point [11]. This method has a great advantage in that it avoids energy disparity between the two differently circularly polarized X-rays caused by mechanical errors of the monochromator. The energy value of the incident photon was calibrated referring to the energy of Dy $M_{4,5}$-edges of Dy_2O_3 [12]. We defined the incident angle of an X-ray as the angle between the sample surface and the incident X-ray. We carried out the measurements at 300 K, which is much lower than Curie temperature of the Dy_xCo_{100-x} amorphous films. The films were magnetized perpendicular to the film surface, and the incident direction of photons was normal to the surface. The remanent magnetization state was used for the MCD measurement.

3. Results

The MCD near the $M_{4,5}$-edges for Dy gives us information on the angular distribution of magnetic moments for the Dy 4f electron with respect to the direction of the easy axis of magnetization, and the maximum opening angle of the conical distribution. Fig. 2 shows MCD spectra of the Dy $M_{4,5}$-edges of $\mathrm{Dy}_x\mathrm{Co}_{100-x}$ of various x compositions, where the M_5 and M_4 white lines due to core-level 3d spin–orbit splitting are shown. The MCD peak direction is reversed over the range $x = 24$–21. Thus, it is deduced that dysprosium has its major magnetic moment at $x > 24$ and cobalt has major magnetic moment at $x < 21$. In spite of the peak direction, the lineshape of the spectra are very similar for all compositions, and thus the Dy 4f electronic states are not significantly changed by the Dy concentration.

Fig. 3 shows the incident angle dependence of MCD intensity of Dy M_5-peak of $\mathrm{Dy}_x\mathrm{Co}_{100-x}$ ($x = 15$ (triangle), 25 (square) and 33 (circle)), which normalized as $(I_- - I_+)/\mathrm{MAX}\,(I_- + I_+)$ for each concentration. The experimental geometry is shown in the figure. The angle dependence fits well with a sine curve (solid curve). This shows that the magnetic moment estimated from XAS–MCD measurements can be taken to be the average of the projections of the magnetic moment, distributed over a cone, to a plane normal to the film. The details of the estimation of opening angle of the cone will be reported elsewhere [8].

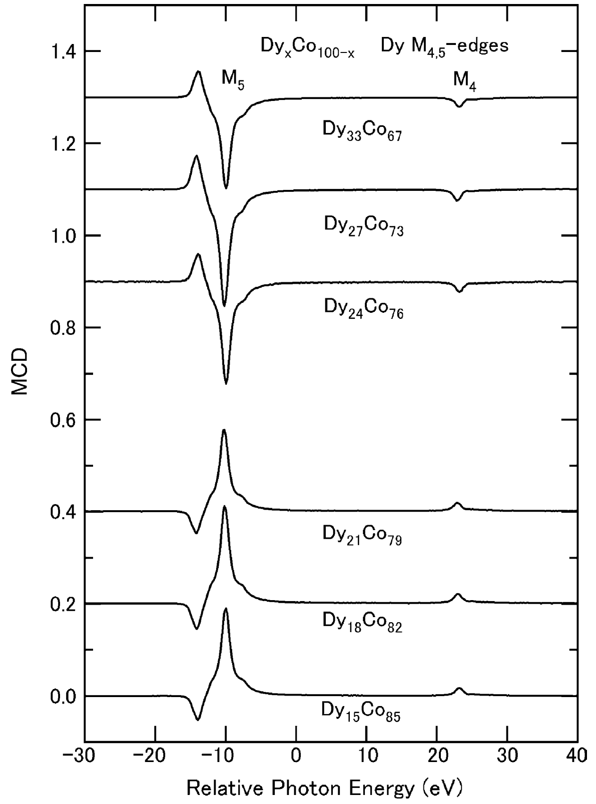

Fig. 2. The X-ray magnetic circular dichroism near the Dy $M_{4,5}$-edges of $\mathrm{Dy}_x\mathrm{Co}_{100-x}$ films ($x = 15$–33) films, which were taken to be the differences between I_- and I_+.

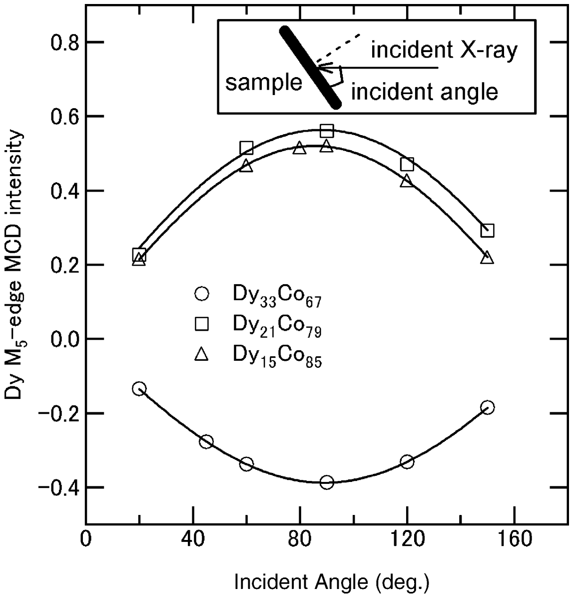

Fig. 3. The incident angle dependence of MCD intensity of the Dy M_5-edge of $\mathrm{Dy}_x\mathrm{Co}_{100-x}$ ($x = 15$, 21 and 33).

Thole and Carra [9,10] derived the sum rule, which allows us to estimate the expectation value of the orbital angular momentum $\langle L_z\rangle$ from XAS and MCD spectra. According to sum rules for the 3d to 4f transitions, the expectation value of $\langle L_z\rangle$ is expressed as:

$$\langle L_z\rangle = h_\mathrm{f}\frac{\int \mathrm{d}\omega(I_- - I_+)}{\int \mathrm{d}\omega(I_- + I_+)} \tag{1}$$

where ω is the photon energy of incident beam and h_f is the number of 4f holes in the RE ion. The integrals in the right hand side of (1) are taken for the whole region of the M_4- and M_5-edges. Generally, the sum rule of $\langle L_z\rangle$ is exactly applied to the core-level absorption region. Furthermore, using the sum rules, the $\langle L_z\rangle$ values for orbital moment of Dy 4f were estimated. The orbital magnetic moment (m_orb) relates to $\langle L_z\rangle$ by the formula: $m_\mathrm{orb} = -\langle L_z\rangle\cdot\mu_\mathrm{B}/\hbar$. Hereafter $\langle L_z\rangle$ is simply expressed in units of μ_B. The number of holes in the Dy 4f orbital was taken to be $h_\mathrm{f} = 5$ for this calculation. The absolute values of the expectation values of L_z for the films are around 0.8–$1.7\mu_\mathrm{B}$ (see open circles in Fig. 5), depending on the dysprosium concentration. The $\langle L_z\rangle$ values obtained in measurements are smaller than the expected values derived from the theoretical calculation for metals [13,14].

To provide a description of the spectral feature, line shapes of the MCD spectra were compared with theoretical values obtained by means of the ionic model [13–15]. Here, a theoretical model based on the full-multiplet calculation for a

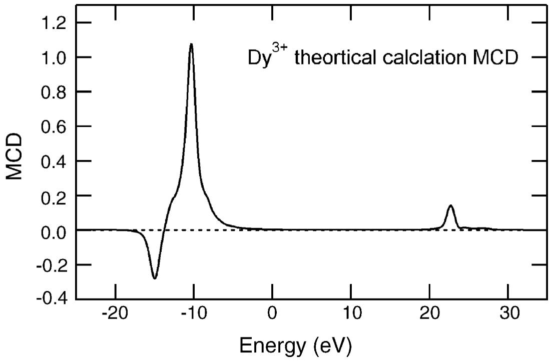

Fig. 4. The soft X-ray magnetic circular dichroism spectra of Dy^{3+} ion, which is deduced from the full-multiplet model calculation.

Dy^{3+} ion was used, which is known to a useful tool to analyze the experimental results of MCD spectra. Fig. 4 shows the calculated MCD spectra of the Dy $M_{4,5}$-edges for Dy^{3+} in a relative photon energy scale. The characteristic features of MCD results are fairly well reproduced the spectra of $M_{4,5}$-edges of modeled Dy^{3+} ions in the film reflecting the ionic features. The spectra of the $M_{4,5}$-edges of Dy^{3+} show characteristics which is attributed to the electronic structure of Dy $4f^9$ in the ground state of ${}^6H_{15/2}$.

On the other hand, the $\langle L_z \rangle$ value is calculated to be $5.1\mu_B$ from the theoretical spectra. The magnetic moment of Dy^{3+} is perfectly orientated in the direction perpendicular to the film surface in the atomic model, while the moments in the film are oriented at random within a cone. Because of the cone structure in the RE–TM amorphous film alloys, the maximum value of the Dy 4f orbital moment is smaller than the theoretical value ($5.1\mu_B$) expected from the ionic model, taking into account of the direction of the moment. When the Dy moments distribute randomly along the easy axis of magnetization, the opening angle of the cone is deduced from the projected moment of Dy 4f electron perpendicular to the film surface. The estimated cone half angles (solid circles) are shown in Fig. 5 as a function of dysprosium composition. The minimum value is around $x = 20$. Comparing Fig. 1 with Fig. 5, the concentration dependence of the opening angle and M_s (and H_c) are qualitatively similar, and thus it is expected that there is a relationship between microscopic magnetic behavior and macroscopic magnetic properties, e.g., the cone half opening angle of Dy 4f moment controls M_s and H_c.

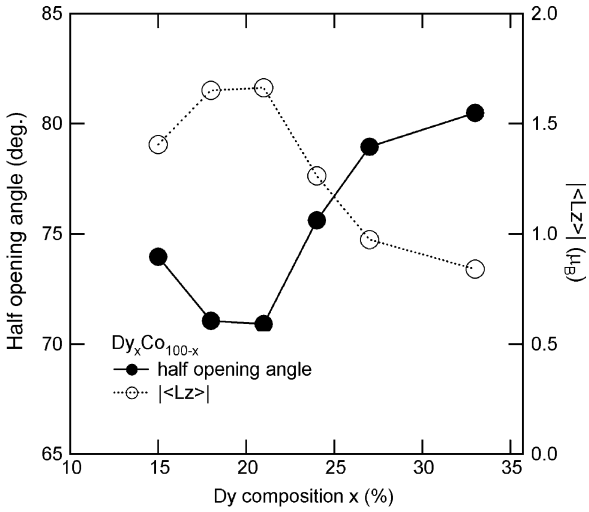

Fig. 5. The absolute value of the expectation value of L_z (open circle) and half opening angle (solid circle) of Dy 4f orbital moment as function of dysprosium concentration.

4. Conclusions

In this paper, the expectation values of orbital moment $\langle L_z \rangle$ of Dy 4f electron of Dy_xCo_{100-x} ($x = 15$–33) are estimated using soft X-ray absorption and circular dichroism spectra near the $M_{4,5}$-edges. The incident angle dependence of the MCD intensity of the M_5-peak was fitted by a sine curve, i.e., the magnetic moment estimated from XAS–MCD measurements can be taken to be as the average of projections of the magnetic moments, distributed over a cone perpendicular to sample surface. The $\langle L_z \rangle$ opening angle of Dy 4f electron was estimated, and its minimum value was around $x = 20$. In the future, the role of cobalt element and use of a normalized temperature to Curie temperature for each dysprosium composition should be investigated.

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