Three-dimensional visualization of magnetic domain structure with strong uniaxial anisotropy via scanning hard X-ray microtomography

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An X-ray tomographic technique was developed to investigate the internal magnetic domain structure in a micrometer-sized ferromagnetic sample. The technique is based on a scanning hard X-ray nanoprobe using X-ray magnetic circular dichroism (XMCD). From transmission XMCD images at the Gd L2 edge as a function of the sample rotation angle, the three-dimensional (3D) distribution of a single component of the magnetic vector in a GdFeCo microdisc was reconstructed with a spatial resolution of 360 nm, using a modified algebraic reconstruction algorithm. The method is applicable to practical magnetic materials and can be extended to 3D visualization of the magnetic domain formation process under external magnetic fields. © 2018 The Japan Society of Applied Physics

Magnetic domain structures1) are known to reflect the fundamental magnetic properties of materials, such as magnetostatic interaction, exchange energy, and magnetic anisotropy. Nucleation of magnetic domains and pinning of domain-wall propagation govern the magnetization-reversal processes and determine the macroscopic coercivity. Therefore, observation of the magnetic domain structure is important for understanding the magnetic characteristics of systems, including practical magnetic materials. Since the first confirmation of domain structure by Bitter,2) researchers have developed a variety of domain observation techniques, including magneto-optical Kerr effect (MOKE) microscopy,1) magnetic force microscopy,3) Lorentz transmission electron microscopy,4) scanning probe magnetic microscopy,5) photoelectron emission microscopy (PEEM),6) and transmission soft X-ray microscopy in the scanning and transmission soft X-ray microscopy.12) Donnelly et al. recently reported the 3D observation of vector domains of GdCo alloys via X-ray ptychography.13) However, there have been few studies, and it is important to develop different approaches of 3D magnetic microscopy for extending the capabilities and applicability of this emerging technique.

In this study, we demonstrate magnetic tomography via scanning hard X-ray microscopy based on X-ray magnetic circular dichroism (XMCD).14) We present the feasibility of the technique for studying a microfabricated structure of the soft ferromagnet GdFeCo, overcoming the limitations of neutron, electron, and soft-X-ray probes.10–12) The technique adopts a flexible setup enabling future studies under external fields and in combination with X-ray fluorescence microtomography,15) which is a potential advantage over hard X-ray ptychographic techniques.13)

A magnetic film of SiN (60 nm)/Gd22.00Fe68.25Co9.75 (5,000 nm)/SiN (5 nm) was grown on a SiN membrane substrate with a thickness of 1 μm via magnetron sputtering and then fabricated into a disc shape via optical lithography and Ar ion milling. The designed diameter and the thickness were 10 and 5 μm, respectively. Our MOKE measurement revealed the perpendicular magnetization of the unpatterned film. Maze-like domain structures with stripe widths of 2–3 μm were observed at the remnant magnetization state. From an XMCD measurement at the Gd L3 edge, the XMCD contrast of ±5% with respect to the polarization-averaged X-ray absorption coefficient was obtained. Element-specific magnetization measurement in the unpatterned film revealed a
characteristic hysteresis loop with perpendicular magnetization and the formation of multiple domain structures. The coercivity of the unpatterned sample was estimated to be ~50 Oe.

Magnetic X-ray computed tomographic imaging measurement was performed using the scanning hard X-ray nanoprobe at BL39XU of the SPring-8 synchrotron radiation facility.\(^{16}\) Figure 1 shows the experimental setup. The X-ray energy was tuned at the L3 resonance of Gd (7.247 keV), at which the maximum XMCD contrast was obtained, using a Si 111 double-crystal monochromator. A 0.45-mm-thick diamond X-ray phase retarder was used to generate circularly polarized X-ray beams of switchable photon helicity. The photon helicity was switched at 37 Hz, and the lock-in detection technique\(^{17}\) was used to improve the signal-to-noise ratio of the dichroic signal. The time constant of the low-pass filter was 30 ms. The output of voltages of the lock-in amplifier were converted into transistor–transistor logic pulses at the corresponding frequency using a voltage–frequency converter and then counted at a 100-ms duration synchronously with the position encoder output pulses of the X stage. These data-acquisition conditions ensured that the spatial resolution for the X-direction was approximately 100 nm, assuming the scan velocity, sampling rate, and time constant of the lock-in amplifier. The mechanical spatial resolution for the Z-direction was 100 nm, as determined by the scan step of the translation stage. The mechanical resolutions in the X- and Z-directions were smaller than the focused X-ray beam size (130 × 140 nm\(^2\) in FWHM), and the practical resolution was determined by the focused X-ray beam size. The projected images of XMCD, \(\Delta \mu(X, Z, \theta)\) and XAS \(\mu(X, Z, \theta)\) were collected at angles of ~70 to +70° with a step of 5°. The blind regions (−90° < θ < −70°, 70° < θ < 90°) were due to the window size of the membrane substrate. The XAS and XMCD projections were acquired simultaneously, and the acquisition time was 30 min for each projection angle.

We show that conventional reconstruction algorithms can be applied for reconstructing the 3D magnetic domain structure in the case where the sample has strong uniaxial anisotropy and a domain structure with uniaxial magnetization is formed. The bottom of Fig. 2 shows a one-pixel slice of the distribution of the sample magnetization in the X–Y plane, which is perpendicular to the Z-axis for rotation. The X–Y coordinate system is fixed to the X-ray beam and the experimental system, whereas the x–y coordinate system is assumed to be fixed at the sample and rotates about the Z-axis by an angle of θ. In this geometry, the XMCD amplitudes \(\Delta \mu_i\) from a local part of the sample are proportional to the magnetization of the local volume \(-m(x, y) = (m_x, m_y, m_z)\) projected to the direction of the incident X-ray beam; i.e., \(\Delta \mu_i \propto m_i(x, y) \sin \theta + m_z(x, y) \cos \theta\). As shown in the top of Fig. 2, the XMCD projection is given by an integral of \(\Delta \mu_i\) along the X-ray path, which is parallel to the Y-direction:

\[
\Delta \mu(X, \theta) = \int \Delta \mu_i(x, y) \, dY = \int [m_i(x, y) \sin \theta + m_z(x, y) \cos \theta] \, dY.
\]

We assume that the sample has strong magnetic uniaxial anisotropy so that only the magnetization component is parallel to the y-direction and the other components are zero; i.e., \(m(x, y) = (0, m_y, 0)\). In this case, XMCD projection is given by

\[
\Delta \mu(X, \theta) = \int m_i(x, y) \cos \theta \, dY.
\]

This formula is similar to the Radon transform\(^{18}\) but includes an additional factor of \(\cos \theta\). If a proper correction for this...
Principle of the tomographic reconstruction of the magnetization distribution from the projected image of XMCD.

Figure 2. Principle of the tomographic reconstruction of the magnetization distribution from the projected image of XMCD.

Figure 3. Selected images of the 2D projections of a GdFeCo disc at different rotation angles obtained using (a) polarization-averaged X-ray absorption and (b) XMCD signals.

 фактор is made, a conventional reconstruction algorithm can be directly applied for reconstruction of the 3D magnetic domains. We preprocessed the recorded projected XMCD images, as follows:

\[
g (X, \theta) = \frac{\Delta \mu (X, \theta)}{\cos \theta}
\]

By dividing the original projection image by a factor of \( \cos \theta \), one may obtain a corrected projection of \( g (X, \theta) \), which can be regarded as the Radon transform of the distribution of the uniaxial magnetization, \( m_1 (x, y) \). The standard algebraic reconstruction technique\(^{19} \) was applied to 29 projected images taken in the angular range of \(-70^\circ \) to \(70^\circ \) with \(~50\) iterations. No extrapolation or complement procedure was applied for lacking angles of \(-90^\circ < \theta < -70^\circ \) and \(70^\circ < \theta < 90^\circ \).

Figure 4 shows selected images of the (a) XAS projection and the (b) corresponding XMCD projection of the GdFeCo disc recorded at different angles. The origin of \( \theta \) was defined as the angle at which the normal of the sample substrate was parallel to the X-ray beam direction. As shown in the XAS image taken at \( \theta = 0^\circ \), the sample shape differs from the designed circular shape. The observed diameter of approximately \( 7 \mu m \) was smaller than the designed value of \( 10 \mu m \), probably because of over-etching in the Ar ion-milling process. Nevertheless, the outline shapes of the disc agreed well between the XAS and XMCD projected images. The XMCD images demonstrate the clear magnetic contrasts of magnetic domains with the typical width of \(~1 \mu m \). The magnetic contrast is the highest in the image taken at \( 0^\circ \) and decreases at larger angles. This result provides evidence in support of the perpendicular magnetic domains.

The XAS reconstruction revealed the trapezoidal shape of the GdFeCo disc, which had diameter of \( 6.7 \mu m \) and a thickness of \( 2.5 \mu m \). The sample was mostly homogeneous in composition. In Fig. 4(b), a cutaway view of the XMCD reconstruction result demonstrates the 3D distribution of the magnetization inside the GdFeCo disc. The color scales correspond to the direction and the amplitude of magnetization perpendicular to the film. Five striped magnetic domains were observed: three positive and two negative.

The sliced images of the \( x-Z \) plane at different \( y \) positions, which are shown in Figs. 4(c)–4(f), reveal that the magnetic domain structures and the boundaries are similar in the planes perpendicular to the film (easy magnetization direction). The cross sections in the \( y-Z \) [Fig. 4(g)] and \( x-y \) planes [Fig. 4(h)] clearly indicate the straight domain boundaries along the easy axis. This is a direct observation that the perpendicular magnetic domains are formed through the...
entire volume of the disc. In Figs. 4(i) and 4(j), the spatial resolutions of the 3D XMCD reconstructed image are estimated from the 10–90% widths of the observed domain boundaries under the assumption that the real boundary widths are significantly smaller than the experimental resolution. The resolution in the x- and Z-directions are given by $w_x = 311 \text{ nm}$ and $w_Z = 360 \text{ nm}$ for the same domain boundary. The obtained spatial resolutions are almost three times larger than the X-ray beam size. For the x-direction, this is because of the limited number of projection angles, which may be insufficient for obtaining reconstruction results with the optimum spatial resolutions. Thermal drifts in the sample position during the measurement might degrade the spatial resolutions in both the x- and Z-directions, although positional corrections to the reconstruction algorithm have been made.

As described above, we successfully visualized the magnetization directions and amplitudes of internal magnetic domain structures with a spatial resolution of a few hundred nanometers for a bulk sample a few micrometers thick, which has not yet been simultaneously achieved using the neutron, electron, or soft X-ray probes.10-12 However, the spatial resolution is approximately three times worse than that of magnetic vector tomography using the hard X-ray phytographic technique.13 Adopting multilayer mirror optics enables focused X-ray beams on the order of sub-10 nm,25 and the spatial resolutions in our scanning setup can be further improved.

The maximum size of observable samples is approximately 10 µm in diameter, which is restricted by strong X-ray absorption for a thick sample. If one adopts the criterion that the transmittance of the sample should be $I/I_0 < e^{-2}$, the measurable sample sizes is estimated to be 14 µm for Nd$_2$Fe$_{14}$B at the Nd L$_2$ edge (6.24 keV) and 7 µm for Co$_{50}$Pt$_{50}$ at the Pt L$_3$ edge (11.6 keV). Preparation of samples having such micrometer sizes is feasible using the focused ion beam technology currently available.

Our technique is applicable to the 3D observation of internal magnetic domain structures in several kinds of soft and hard magnetic materials. In particular, study of the Nd$_2$Fe$_{14}$B sintered permanent magnet would provide us with insight regarding the origin of the highly coercive field, as the nucleation and evolution of the magnetic domain structure should govern the magnetization-reversal mechanism of this material.23,24 The evolution of 3D magnetic domain structures has been intensively studied via numerical simulations23 but has not been elucidated experimentally thus far. To this end, tomographic magnetic imaging measurements involving external magnetic fields that rotate with the sample to keep the domain structure unaffected must be performed. Our scanning hard X-ray tomography setup with the long mirror-sample distance is suitable for introducing a specially designed magnet and allows 3D observation of the nucleation and evolution of magnetic domains under a variable magnetic field. Moreover, the microstructure of the sintered magnet is comprised of a Nd$_2$Fe$_{14}$B main phase and other several phases with different chemical compositions.23,24 Nd-rich phases surrounding the sintered magnetic material is comprised of a Nd$_2$Fe$_{14}$B main phase and other several phases with different chemical compositions.23,24 Our scanning X-ray setup can easily be modified for X-ray fluorescence microtomography,10 and used to study the correlation between the magnetic domains and the elemental distribution of the sintered magnet via 3D imaging.

To summarize, we developed a tomographic imaging technique to investigate the internal magnetic domain structure of micrometer-sized ferromagnetic samples based on scanning hard XMCD microscopy. The technique was applied to a GdFeCo disc that exhibited perpendicular magnetic anisotropy, and the interior uniaxial magnetic domain structures were successfully revealed three-dimensionally with a spatial resolution of 360 nm. This hard X-ray magnetic tomography method is applicable to various soft and hard magnetic materials, including strong permanent magnets, which is of great importance for practical applications.

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