Development of hard X-ray magnetic circular dichroism microscope and its application to NdFeB magnet

A Nd-Fe-B magnet (neodymium magnet) [1] is the strongest permanent magnet ever made and has been used for a wide range of applications in modern products, such as motors in cordless tools, hard disk drives, magnetic fasteners and synchrotron undulators. These magnets are produced by sintering and annealing processes and thus exhibit multiphase microstructures. One of the major drawbacks of neodymium magnets is their low curie temperature. In order to overcome this drawback, some of the Nd is replaced with Dy. It is considered that Dy atoms in the borders of micrograins play an important role in improving the Curie temperature. To understand this mechanism, observation of the microstructure and revealing the relationship between the microstructure and micromagnetics are indispensable. In this study, the magnetic microstructures and element distributions in a Nd-Fe-B magnet were observed by a hard X-ray magnetic circular dichroism (HXMCD) microscope with Kirkpatrick-Baez (K-B) mirrors at beamline **BL16XU**, and it was found that the local magnetic moment and the concentration of Nd are strongly correlated [2].

Figure 1 shows the composition of our microscope system at beamline BL16XU [3]. Monochromatized linearly polarized X-rays are converted to either left circularly polarized (LCP) or right circularly polarized (RCP) X-rays by passing them through a diamond X-ray phase retarder (XPR). Then, the X-rays enter a pair of K-B mirror systems to form a focused beam. At the time of our experiments, the size of the focused beam on the sample was estimated to be 1.92 (horizontal) × 1.83 (vertical) μm^2. A sample of the neodymium magnet was placed in a 2 T electromagnet. Figure 2 shows how the (H)XMCD spectrum was measured and an example of the Nd L_{II} HXMCD spectrum of the neodymium magnet. The concept of XMCD spectroscopy was first proposed by Schütz *et al.* in 1987 [4]. One can understand the mechanism of X-ray absorption as the transition of electrons from an occupied inner state to an empty outer state. For a magnetic material, the outer shell has a spin moment, which is given by the imbalance of spin-up and spin-down electrons or equivalently (except for the sign) by the imbalance of spin-up and spin-down holes. If we can make spin-dependent X-ray absorption, it will be possible to detect the difference in the number of empty states with up spin and down spin. This spin-dependent X-ray absorption is realized by the use of RCP absorption and LCP absorption, which transfer their angular momentum to the excited photoelectrons. As a result of absorption measurements by RCP and LCP, the spectra show a slight difference, and the difference is the so-called (H) XMCD spectrum.

For our mapping experiments, the photon energy was fixed to 6.721 keV, which is the standard characteristic of Nd L_{II} XMCD and Nd L_{II} XAFS (see Fig. 2). For the XMCD mapping, the signal of Nd L_{II} XMCD was derived from the fluorescence of Nd $L\beta$. Figure 3(a) shows the fluorescence Nd-$L\alpha$ intensity map for a 44 × 20 μm^2 rectangular area, which reflects the Nd concentration. It should be noted that the fluorescence of Nd $L\beta$ contains the XMCD signal because its origin is the relaxation from the spin polarized N5 shell, while Nd $L\alpha$ contains little of the XMCD signal because its origin is the relaxation from the unpolarized (filled) M shells. Several islands with areas of several μm^2 with relatively high Nd concentrations can be observed in Fig. 3(a). Figure 3(b) shows the mapping of the magnetic moment per Nd atom derived from HXMCD signals. With our experimental configuration, only the magnetic moment parallel to the horizontal direction can be detected. It is possible to find similar regions by comparing Figs. 3(a) and 3(b). In area (A) in Fig. 3, the HXMCD signal is strong and the Nd concentration is low. In contrast, area (B) exhibits a weaker HXMCD signal and a higher Nd concentration than those in the surrounding areas. We conclude that area (A) is a crystalline $Nd_2Fe_{14}B$ alloy while area (B) consists of paramagnetic Nd-rich precipitates, as previously reported. This strongly indicates that Nd-rich three-grain junction precipitates are paramagnetic in the neodymium materials.

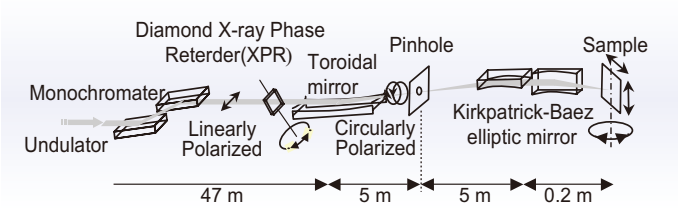

Fig. 1. Schematic drawing of our magnetic/elemental microscope system at BL16XU.

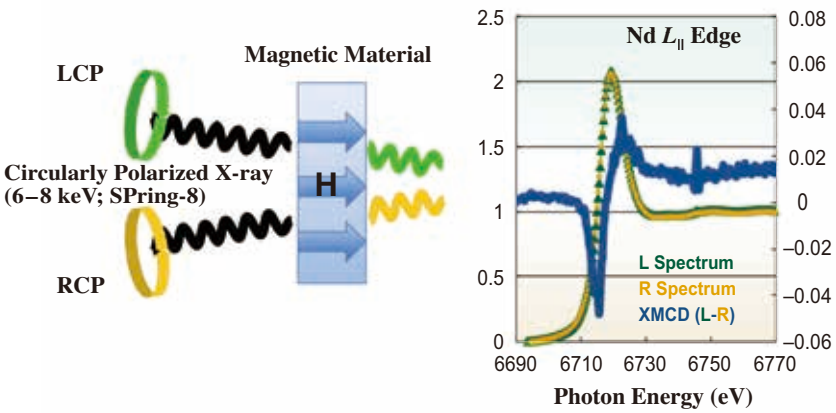

Fig. 2. (Left) Conceptual drawing of (H)XMCD measurement. The arrows in the magnetic material indicate the direction of magnetization. The X-ray absorption probability depends on the relation (parallel/antiparallel) between the X-ray helicity (LCP/RCP) and magnetized direction. (Right) Example of Nd L_{II} HXMCD spectrum derived from two spin-dependent spectra.

In this work, we performed simultaneous 2D mapping of fluorescence X-ray and HXMCD signals by scanning a hard X-ray microbeam and demonstrated the feasibility of studying magnetic materials with our microscope system. Our results clearly indicate the correlation between the Nd concentration and the magnetization of Nd-rich three-grain precipitates. The results also show that microspectrometry with hard X-rays is a powerful tool for revealing the correlation between the magnetic microstructure and the mapping of element buried under the surface oxide layer. Our technique shows strong potential for *in situ* observation of the magnetization reversal of bulk rare-earth magnets under a high magnetic field.

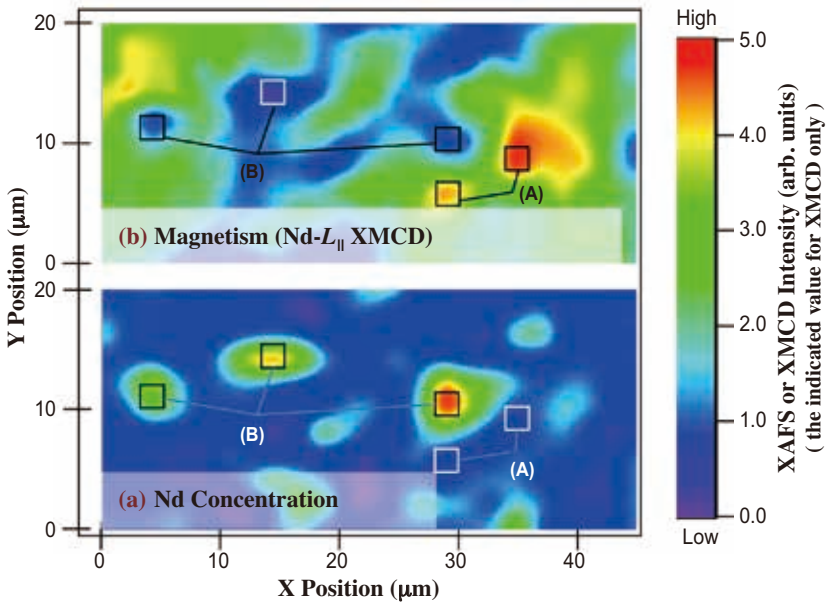

Fig. 3. Experimental results obtained using our magnetic/elemental microscope at BL16XU. **(a)** Elemental map for Nd concentration obtained by measuring Nd-*Lα* fluorescence and **(b)** HXMCD signal map of Nd-*Lα* XMCD. Area (A) is a crystalline $Nd_2Fe_{14}B$ alloy and area (B) consists of Nd-rich paramagnetic precipitates.

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