Visualization of magnetic domain formation in neodymium magnet via scanning hard X-ray nanoprobe

Many kinds of magnets support modern life. Among them, neodymium magnet ($Nd_2Fe_{14}B$; abbreviated to Nd-Fe-B) is one of the most useful because it is the strongest permanent magnet currently available. This material has been applied to various products including high-efficiency electric motors for low-emission hybrid/electric vehicles, energy-saving air conditioners, and electric power generators. This powerful magnet is regarded as vital for achieving a green society. To further improve the energy-saving performance of high-power motors, an alternative permanent magnet that can maintain superior performances even at high temperatures is still desired. In other words, it is necessary to develop a new magnetic material that has an increased maximum energy product, $(BH)_{max}$, or an enhanced coercive field. Understanding the mechanism of the magnetization reversal process of permanent magnets will be a key to achieving this because the coercivity value is related to the reversal mechanism. A Nd-Fe-B magnet is produced by sintering fine grains with diameters of a few micrometers. As a result, the material has an inhomogeneous microstructure, which is likely the origin of the high coercivity of Nd-Fe-B sintered magnet. To clarify the relation between the microstructure and the magnetic properties, microscopic observation of the chemical and magnetic states is essential.

A scanning hard X-ray nanoprobe is one of the most promising techniques for the microscopic observation of sintered magnets. The technique at SPring-8 **BL39XU** [1,2] enables magnetic and chemical imaging with a spatial resolution of 100 nm under high magnetic fields of up to 22 kOe. Additionally, this X-ray nanoprobe allows the observation of uneven surfaces, such as the fractured surface of the bulk of a sintered magnet. This feature is a major advantage over other magnetic microscopy techniques such as Kerr microscopy, Lorentz transmission electron microscopy, and scanning transmission soft X-ray microscopy, which are only applicable to optically flat or polished surfaces or very thin specimens.

In this study, we used the scanning hard X-ray nanoprobe to visualize the formation process of magnetic domains at the fractured surface of a Nd-Fe-B sintered magnet during the demagnetization cycle [2]. In addition to the magnetic imaging, we performed X-ray fluorescence (XRF) microscopy measurement of the sample to discuss the correlation between the elemental distribution and the magnetic properties. Figure 1(a) shows a scanning electron microscopy (SEM) image of the fractured surface of a $Nd_{14.0}Fe_{79.7}Cu_{0.1}B_{6.2}$ magnet. Approximately 20 grains with diameters of ~5 μm can be seen at the fractured surface, where the microstructures are almost conserved as the bulk state. Figures 1(b–d) show XRF images of the same area as the SEM image in Fig. 1(a). The scan step was 250 nm for all images. The SEM and XRF images were obtained with no magnetic field. In the XRF images, elemental distributions of (a) Fe, (b) Nd, and (c) Cu are reproduced. The $Nd_2Fe_{14}B$ main-phases with a high Fe concentration and Nd-rich phases adjacent to the $Nd_2Fe_{14}B$ main-phase grains are clearly resolved. Cu is distributed non-uniformly at the boundaries of the $Nd_2Fe_{14}B$ main-phase grains. Areas of high Cu concentration are found adjacent to the Nd-rich regions at the triple junctions. By local X-ray absorption spectroscopy (XAS) analysis at the Cu *K* edge, the chemical state of the segregated Cu was determined to be an alloy with Nd [2].

Figures 2(a–g) show scanning XMCD images of magnetic domain structures whose magnetization directions are parallel to the crystallographic *c*-axis (perpendicular to the surface). A nanofocused and circularly polarized X-ray beam was scanned over a 15×15 μm² area corresponding to the middle parts of

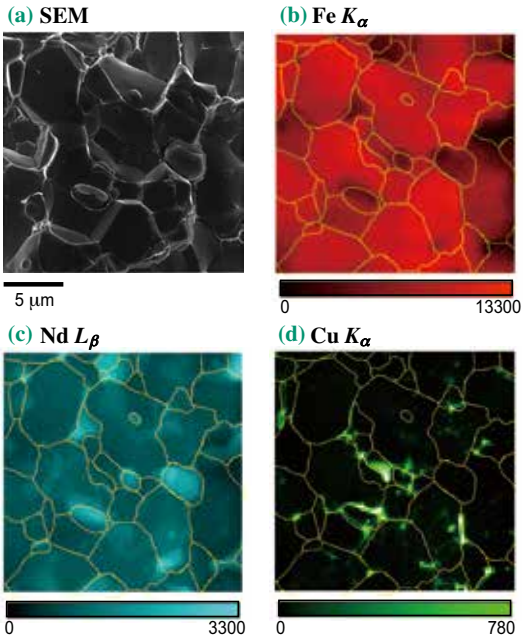

Fig. 1. **(a)** Scanning electron microscopy (SEM) image and scanning X-ray fluorescence images of **(b)** Fe, **(c)** Nd, and **(d)** Cu obtained at the fractured surface of a $Nd_{14.0}Fe_{79.7}Cu_{0.1}B_{6.2}$ sintered magnet. Horizontal color scales indicate the X-ray fluorescence intensity counted per 0.1 s.

the XRF images shown in Figs. 1(b–d), and the XMCD signals at the Nd L_2 edge (6.725 keV) were recorded by the fluorescence mode using a four-element silicon drift detector. The scan step was 300 nm and the acquisition time was 0.8 s per pixel (0.4 s each for left- and right-circular polarization). The resulting magnetic images clearly demonstrate the creation of the initial magnetic domains followed by the expansion of domain structures in a grain-to-grain manner with increasing demagnetization field.

The evolution of magnetic domains in the magnetization reversal process was found to proceed in the following steps: i) nucleation of initial multiple domains in particular grains; ii) expansion of reversed magnetic domains inside the grains by domain wall motion; and iii) propagation of reversal domains to the neighboring grains across the grain boundaries by intergranular exchange coupling. Furthermore, we confirmed that the formation of the magnetic domain structures is reproducible. In another demagnetization cycle, the domain evolution process was similar to the previous one. The initial reversed domains were created in the same grains with almost identical patterns. The subsequent domain extension process occurred in a similar manner. This result suggests the presence of particular nucleation sites that trigger the reversal of magnetization in the Nd-Fe-B sintered magnet.

By comparing the magnetic domain images in Figs. 2(a–g) and the XRF image of Cu in Fig. 1(d) and by performing statistical analyses, the magnetization reversal in $Nd_2Fe_{14}B$ grains adjacent to the segregated Cu tends to occur earlier than that of the other grains apart from the Cu grains. This suggests that the microstructures in the region around the Nd-rich phase with segregated Cu play a key role in determining the magnetic nucleation sites. Please refer to Ref. 2 for a detailed discussion.

In summary, magnetic imaging by a scanning hard X-ray nanoprobe is a novel technique for studying the correlation between the chemical microstructure and the magnetic domain structures of permanent magnets and their evolution processes. The element specificity provided by this technique will be useful for studying other varieties of magnet that have microstructures with characteristic elemental distributions. Another scanning soft X-ray nanoprobe has recently been developed at SPring-8 **BL25SU** [3,4], which will provide opportunities for magnetic microscopy with higher magnetic image contrast for 3*d* transition metals, such as Fe, with a superior spatial resolution, as well as novel sample environments with strong magnetic fields as high as 80 kOe.

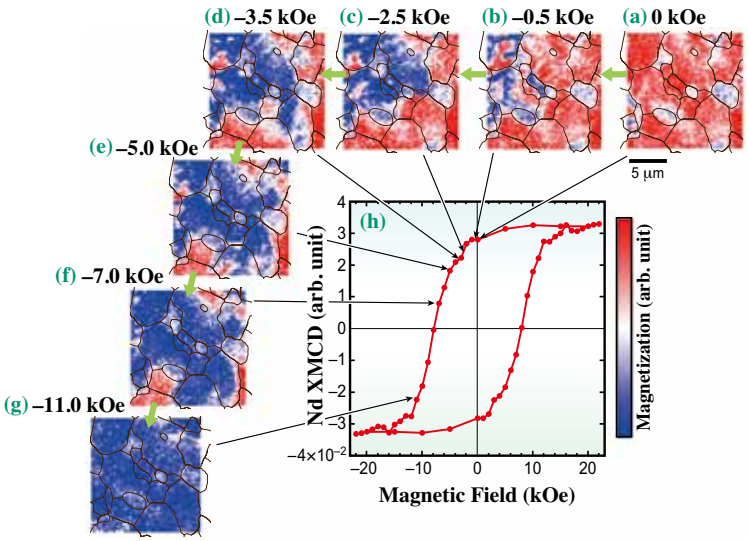

Fig. 2. **(a)–(g)** Magnetic domain structures at the fractured surface of a $Nd_{14.0}Fe_{79.7}Cu_{0.1}B_{6.2}$ sintered magnet as a function of external magnetic field. The magnetic images were obtained in a demagnetization cycle at various field points indicated on the magnetic hysteresis loop in **(h)**.

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References

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