7 Elements Strategy Initiative Center for Magnetic Materials (ESICMM)

– in situ analysis using neutrons and X-rays –

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The goals of the Elements Strategy Initiative Center for Magnetic Materials (ESICMM) at the National Institute of Material Science (NIMS) are: (1) laboratory-scale synthesis of mass-producible high-performance permanent magnets without using critical rare-earth elements for the next generation and (2) framework-building and provision of basic science and technology for industrial R&D. To achieve these goals, ESICMM focuses on theoretical research and mining of new permanent magnet materials, and simultaneously pursues various processing technologies to improve the existing high-performance permanent magnet materials through cooperative activities in the three research fields of computer physics, structural and property characterization, and material processing. Another important mission of ESICMM is to train scientists who will contribute to the future development of magnetic functional materials.

In CMRC, the In-situ Analysis Using Neutrons and X-rays Project was started in July 2012 as an analysis group of ESICMM. The complementary use of neutrons at J-PARC/MLF and other facilities as well as synchrotron X-rays at the Photon Factory is very useful for characterizing magnetic materials from the atomic scale to micrometer scale.

7-1. Multiple magnetic scattering in small-angle neutron scattering (SANS) of Nd–Fe–B nanocrystalline magnet

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We have investigated the influence of multiple scattering on the magnetic SANS from a Nd–Fe–B nanocrystalline magnet. We performed sample-thickness and neutron-wavelength-dependent SANS measurements, and observed the scattering vector dependence of the multiple magnetic scattering. We revealed that significant multiple scattering exists in the magnetic scattering rather than the nuclear scattering of the Nd–Fe–B nanocrystalline magnet. It is considered that the mean free path of the neutrons for magnetic scattering is rather short in Nd–Fe–B magnets. We analyzed the SANS data by the phenomenological magnetic correlation model considering the magnetic microstructures and obtained the microstructural parameters.

SANS is recognized as a powerful experimental technique to characterize permanent magnet materials such as $Nd_2Fe_{14}B$ single crystal, Nd–Fe–B sintered magnets, hot-deformed nanocrystalline magnets, and nanocomposites. Because of the magnetic sensitivity and the high transparency of neutrons to matter, one can probe the magnetic properties of the materials to generate bulk averaged information. Moreover, the range of the scattering vector, q, matches the length scale of the microstructures and the magnetic domains in permanent magnet materials. These unique characteristics of SANS are complementary to experimental techniques such as transmission electron microscopy or X-ray microscopy that probe the local, rather than bulk, structure. For the development of high-performance permanent magnet materials, it is necessary to investigate magnetic structures and to reveal the origin of the coercivity. In the previous study, we

qualitatively discussed the magnetic SANS in the magnetization reversal process. However, it is essential to extract quantitative information such as particle-size distribution and magnetic correlation length during the magnetization reversal process from these SANS data.

Analysis of the small-angle X-ray or neutron scattering intensity considers, in general, the X-ray or neutrons to be singly-scattered, i.e. samples to be moderately thin. Such a single scattering approximation is invalid for thick samples in which the thickness exceeds the scattering mean free path (MFP). The scattering probability is proportional to the scattering cross-section of the material, and multiple scattering becomes noticeable for strongly scattering thick samples. Multiple scattering smears the small-angle scattering patterns, and blurs the "real", single scattering, signals. For quantitative analysis of SANS data, the sample has to be thin enough to preserve the single-scattering approximation regime. However, on the other hand, the sample must be thick enough to be regarded as a bulk representative.

In this study, we have investigated the influence of multiple scattering on the magnetic SANS from a Nd–Fe–B nanocrystalline magnet. Considering Molière's theory of multiple small-angle scattering, it is important to check the dependence of sample thickness on the mean free path of the neutrons. We clarify the multiple-scattering effect by either measuring samples of different thicknesses or using neutron beams with different wavelengths. SANS from a thermally demagnetized state of Nd–Fe–B nanocrystalline magnet were measured at room temperature, which includes both nuclear and magnetic scattering arising from the magnetic domain walls, and also measured above the Curie temperature, T_C, which only includes nuclear scattering. We reveal that significant multiple scattering exists in the magnetic scattering rather than the nuclear scattering. The phenomenological model fitting analysis was applied to the magnetic scattering intensities, and the microstructural parameters were obtained.

The effect of multiple scattering on nuclear scattering was measured at elevated temperature. One can observe only nuclear scattering signals when measured above T_C because $Nd_2Fe_{14}B$ becomes paramagnetic and magnetic interaction disappears. Figure 1(a) shows the nuclear scattering intensities $I_{nuc}(q)$ along the c-perpendicular direction for different t (t = 0.1 and 0.5 mm) and λ (λ = 0.5, 0.81 and 1.15 nm) observed at $T > T_C$. Nuclear scattering intensities seem to be identical for all sample thicknesses and neutron wavelengths investigated. This indicates that the effect of multiple scattering on nuclear scattering is negligible, at least in the observed q region in this experiment (0.02–0.4 nm^{-1}).

The magnetic scattering intensity $I_{mag}(q)$ was obtained by subtracting $I_{nuc}(q)$ from the scattering intensity obtained in the thermally demagnetized state $I(q)$: $I_{mag}(q) = I(q) - I_{nuc}(q)$. Figure 1(b) shows $I_{mag}(q)$ along the c-perpendicular direction for different sample thicknesses and neutron wavelengths. Magnetic scattering intensities show different q-dependences for different sample thicknesses and neutron wavelengths. Arrows in

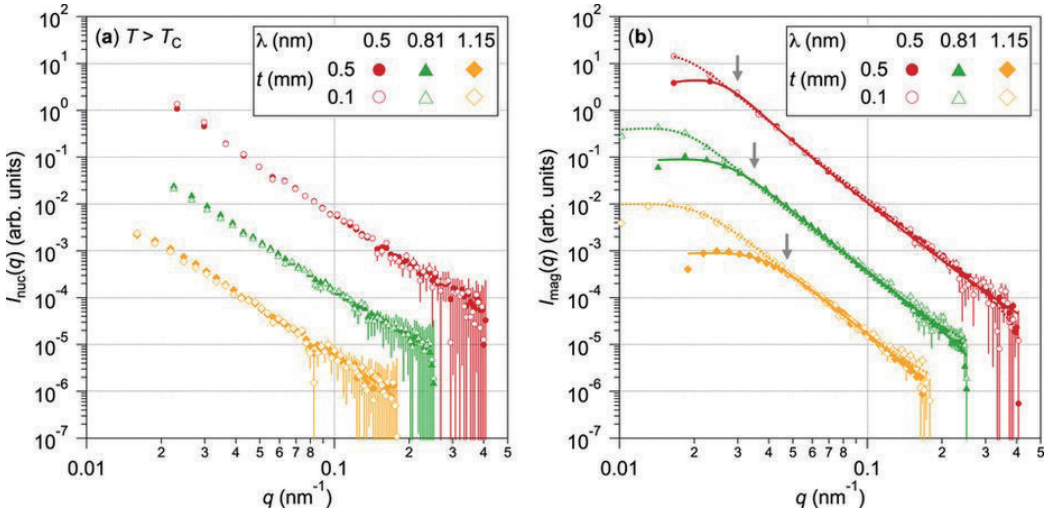

Fig. 1: Nuclear and magnetic scattering intensities for the Nd–Fe–B nanocrystalline magnet. (a) Nuclear scattering intensities $I_{nuc}(q)$ at $T > T_C$ and (b) magnetic scattering intensities $I_{mag}(q)$ for the Nd–Fe–B nanocrystalline magnet with different sample thickness t (t = 0.1 and 0.5 mm) and neutron wavelength λ (λ = 0.5, 0.81 and 1.15 nm).

Fig. 1(b) indicate the critical q points below which $I_{mag}(q)$ for t = 0.5 mm and t = 0.1 mm behave differently. Suppression of the intensity in the low q region is marked for the thicker sample measured with longer wavelength neutrons, which are one characteristic of the multiple-scattering effect. These characteristics are similar to multiple nuclear scattering in the literature, however, the shape of $I_{mag}(q)$ for the thin sample (t = 0.1 mm) is largely independent of the λ investigated.

The periodicity I and the correlation length ξ for different λ are plotted as a function of the sample thickness t in Fig. 2. For any neutron wavelength, d and ξ are smaller for the thicker sample than those for the thinner sample. Thus, d and ξ are underestimated if one uses SANS data with significant magnetic multiple scattering. Extrapolation of the linear regression of d and ξ to $t \rightarrow 0$ yields almost the same values for different λ. The extrapolation to t = 0 should, therefore, serve as a reasonable approximation for the true values of d and ξ in the single-scattering regime. The correlation length at t = 0, $\xi \sim 110$ nm, in the present case is interpreted as the radius of the $Nd_2Fe_{14}B$ grains, and the diameter $2\xi \sim 220$ nm is consistent with the diameter of $Nd_2Fe_{14}B$ grains of 160–300 nm which was determined by transmission electron microscopy. On the other hand, the magnetic periodicity at t = 0, d ~ 420 nm, is explained by magnetic domains formed by magnetically coupled grains, i.e. so-called interaction domains. These results indicate the applicability of the extrapolation to zero thickness to retrieve parameters for magnetic correlation function.

In conclusion, significant multiple-scattering effects have been observed in magnetic scattering rather than nuclear scattering, in a Nd–Fe–B nanocrystalline magnet. A phenomenological model fitting approach was applied to the magnetic scattering and the magnetic periodicity, d, and the correlation length, ξ, were obtained. The analysis yielded the anticipated values for the bulk magnetic domains in the thermally demagnetized state of Nd–Fe–B nanocrystalline magnet.

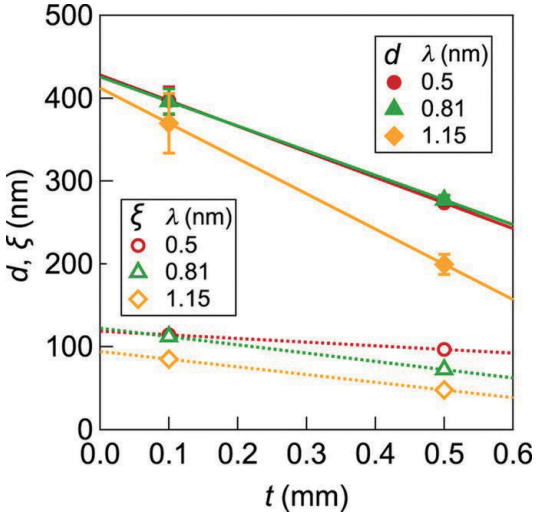

Fig. 2: Magnetic periodicity and correlation length versus sample thickness.