10 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.

10-1 Magnetic interactions between nanocrystalline grains in Nd-Fe-B hot-deformed magnets using scanning transmission X-ray microscopy

The mechanism of high coercivity in Nd-Fe-B magnets is important for achieving Dy-free high coercivity permanent magnets. A recent development is an Nd-Fe-B nanocrystalline hot-deformed magnet with infiltration of Nd-Cu eutectic alloys, which enhances coercivity [1]. In Nd-Fe-B nanocrystalline magnets, grain boundaries play an essential role in decreasing exchange interactions, and it is suggested that magnetic dipolar interaction becomes important. The significance of magnetic interactions between grains in the magnetization reversal process is not well understood, since quantitative measurement of the magnetic interactions in magnetic materials is rather difficult.

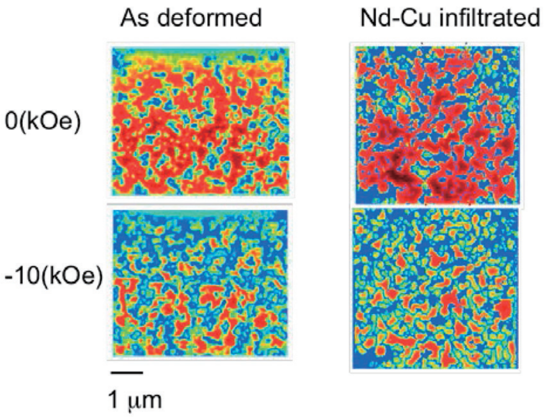

Fig 1: Dipolar energy density maps of nanocrystalline Nd-Fe-B magnets with and without Nd-Cu infiltration.

In this study, we have developed a quantitative method for visualizing the dipolar energy and exchange energy distributions in Nd-Fe-B nanocrystalline magnets with and without the Nd-Cu infiltration process using scanning transmission X-ray microscopy (STXM) [2-4] and large-scale micromagnetic simulation. We have also performed small-angle neutron scattering (SANS) measurements for bulk Nd-Fe-B nanocrystalline magnets to analyze the magnetic interactions between grains [5-7].

Figure 1 shows the dipolar energy distribution of Nd-Fe-B nanocrystalline magnet without and with the Nd-Cu infiltration process taken after application of a magnetic field of 0 and –10 kOe. In Nd-Cu infiltrated magnets the change in dipolar energy is smaller than in deformed magnets.

10-2 Magnetic structures and rare-earth site preferences in permanent magnets probed by neutron powder diffraction

Magnetic structure of $Sm_2Fe_{17}N_3$

The rare earth intermetallic compound $Sm_2Fe_{17}N_3$ exhibits notable magnetic properties such as high Curie temperature and high coercivity which are very suitable for permanent magnets. Although the microscopic magnetic structure is basic information for magnetic materials, to our knowledge there is no report on the magnetic structure of $Sm_2Fe_{17}N_3$. This is because the neutron absorption cross section of samarium is so large that researchers hesitate to conduct neutron diffraction experiments of Sm compounds. We have carried out powder neutron diffraction measurement of $Sm_2Fe_{17}N_3$ with a straightforward solution to the problem of long measurement time. Synchrotron X-ray diffraction measurements with single crystal have also been done to obtain initial crystal structure parameters for magnetic structure analysis and we have succeeded in analyzing the magnetic structure of $Sm_2Fe_{17}N_3$ at room temperature. Among four Fe sites in the unit cell, while one Fe site which is the nearest neighbor of nitrogen shows smaller magnetic moment than normal iron, two Fe sites show enhancement in their magnetic moments. This phenomenon can be understood as 'cobaltization' of Fe by the adjacent nitrogen through hybridization.

Site preferences in $(Nd, Dy)_2Fe_{14}B$

Under a high-temperature environment, the coercivity of Nd-Fe-B permanent magnet needs to be reinforced by Dy-substitution due to the low Curie temperature of $Nd_2Fe_{14}B$. From the view point of the national elements strategy, however, Dy usage should be minimized because of its scarcity. Despite the large amount of Dy used for (Nd, Dy)-Fe-B magnets, a quantitative analysis of the mechanism of coercivity enhancement by Dy-substitution has not been presented so far, especially from the microscopic point of view. For the case of the grain boundary distribution process, Dy is substituted in only a few layers from grain boundaries and this makes it difficult to obtain quantitative data with microscopic techniques. In such a case, numerical simulations will play an important part. Here, we present the site preference of Nd and Dy ions in $(Nd, Dy)_2Fe_{14}B$ determined from powder diffraction using neutrons and synchrotron X-rays and show that the combination of thermodynamical calculation and *ab-initio* calculation reproduces the site preference well. This result provides important parameters for numerical simulations for elucidating the mechanism of coercivity enhancement in Dy-substituted Nd-Fe-B magnets.

10-3 Magnetization reversal in hot-deformed Nd-Fe-B magnets as seen by magnetic small-angle neutron scattering

Magnetic small-angle neutron scattering (SANS) is a powerful experimental technique that can probe microstructures and magnetic domain structures in the order of several nanometers to micrometers of bulk ferromagnetic materials simultaneously. Hot-deformed Nd-Fe-B magnet is known to consist of $Nd_2Fe_{14}B$ grains of several 100 nm and the infiltration of RE-Cu (RE = Pr or Nd) eutectic alloy enhances their coercive force.

In order to reveal the mechanism of the enhancement of the coercive force by the RE-Cu infiltration, we have carried out a magnetic SANS study on as-deformed and RE-Cu-infiltrated Nd-Fe-B magnets. The SANS experiment was performed at the V4 beamline at the BER-II research reactor in Helmholtz-Zentrum Berlin, Germany. Small-angle neutron scattering intensities were obtained during the magnetization reversal process from the fully magnetized initial state by using a two-dimensional position-sensitive detector. Scattering intensities along the direction perpendicular to the nominal *c*-axis of $Nd_2Fe_{14}B$ grains were maximized at the coercive force in all samples. External magnetic field dependence of the magnetic scattering intensity showed suppression of the intensity variation in the RE-Cu-infiltrated samples compared to the as-deformed sample [8, 9]. It is indicated that the $Nd_2Fe_{14}B$ grains are magnetically isolated in the RE-Cu-infiltrated samples that result in the higher coercive force than that of the as-deformed one.

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