6 Elements Strategy Initiative Center for Magnetic Materials (ESICMM)

– in situ analysis using neutrons and X-rays –

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6-1 Introduction

The microstructure and magnetic microstructure are the dominant factors of coercivity in permanent magnets. We carried out quantum beam (neutron and synchrotron radiation) experiments to clarify the microstructure and magnetic microstructure of sintered and hot-deformed rare-earth permanent magnets and rare-earth free magnets.

6-2 Precise structural and magnetic structure analysis of magnetic materials using neutrons and synchrotron radiation

The most essential and fundamental aspects of the material characterization of magnetic materials, which are the main phases of permanent magnets, are the crystal and magnetic structures, and it is essential to know such information for the research and development of magnetic materials. X-ray diffraction is mainly used for a precise crystal structure analysis. However, the magnetic structure is closely related to the magnetic properties of the main phase. The magnetic structure information within the range from low temperature to the Curie temperature is essential to understand the role of the main phase of a magnetic material. To evaluate the magnetic structure, it is impossible to determine the magnetic structure because the conventional X-ray diffraction method is insensitive to magnetism. Therefore, it is necessary to use neutrons as probes for magnetism. By using neutron diffraction, it is possible to clarify the magnetic structure and its temperature variation and obtain information such as the quantitative values of the magnetic moment at each site of the crystal that constitutes the magnet. In addition, the site occupancy of rare earth permanent magnets, which indicates the elements occupied at each site of the crystal structure, is also significant for the magnetic properties, and neutron diffraction is essential to quantitatively evaluate the site occupancy. It is difficult to determine the magnetic structure of magnetic materials using neutron diffraction because a complicated data analysis is necessary.

Under such a background, we succeeded in developing a method to quantitatively determine the magnetic structure and site occupancy of magnetic materials using neutron diffraction.

The crystal and magnetic structures of 1-12 compound magnetic materials (Sm,Zr)Fe12-xMx (M=Zr,Ti,Co) were determined using neutron diffraction. An example of an *in situ* neutron diffraction pattern is shown in Fig. 1, where the magnetic structure was precisely determined at each temperature through *in situ* neutron diffraction.

In an analysis of the neutron diffraction shown in the figure, there are many parameters for the magnetic structure. It is difficult to determine the magnetic structure from only the experimental

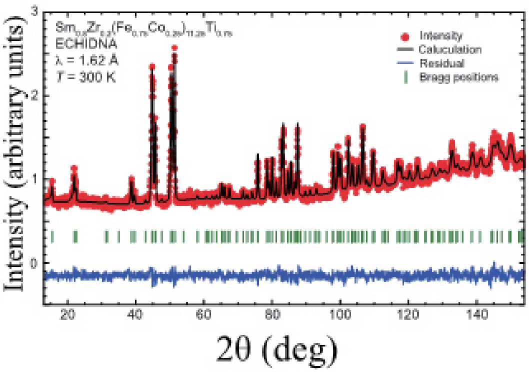


Fig. 1: Neutron diffraction pattern of 1-12 REPM

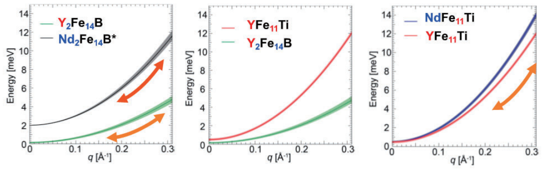
data, however. To solve this problem, we developed a new data analysis method combining neutron diffraction and first-principle calculations, and clarified the temperature dependence of the elemental occupancies and magnetic moments at each site for rare earth and transition metal sites. Our idea for the newly proposed data analysis method is to use data from first-principle calculations of the electronic structure. The results of such calculations are fed back to the Rietveld analysis of the neutron diffraction, and the analysis is repeated until both the inputs and outputs agree. The analysis converges after two or three rounds, revealing new insights into the site-selectivity of Zr and its contribution to the magnetization of each sub-lattice. This method enables us to analyze the origin of magnetism in 1-12 compound magnetic materials, that is, the optimal compound composition. It is difficult to reduce the amount of Ti addition beyond a specific lower limit because of the tradeoff between the structural stability of SmFe12 and the loss of the magnetic properties.

This result enables material researchers who are unskilled in neutron diffraction to obtain the magnetic structure and site occupancy with high accuracy.

6-3 Evaluation of the fundamental physical properties of magnetic materials

It is essential to obtain the fundamental physical parameters such as the crystal magnetic anisotropy and exchange stiffness constants for magnetic materials. We developed a new method to determine such constants of polycrystalline samples using neutron and synchrotron radiation experiments and optical measurements. The spin-wave dispersions of rare-earth and transition metals in the 2-14-1 and 1-12 magnetic materials were determined and are shown in Figure 2. These results provide new insight into the effects of the interactions of rare-earth ions and crystal structures on the magnetocrystalline anisotropy and exchange stiffness constants.

J_{TT} : Fe-Fe interaction



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	J_{TT} [meV Å^2]	n_{RT} [meV]
$Y_2Fe_{14}B$	48(3)	0.15(6)
$Nd_2Fe_{14}B$	100	2
$YFe_{11}Ti$	121(1)	0.6(1)
$NdFe_{11}Ti$	140(2)	0.5(1)

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2-14-1 type
Large J_{TT} change with RE

1-12 type
Small J_{TT} change with RE

From the viewpoint of J_{TT}
1-12 type > 2-14-1 type

To do : Spin wave measurement
in Sm compounds

Fig. 2: Spin-wave dispersions of 1-12 and 2-14-1 REPMs

6-4 Short- and long-range magnetic ordering in magnetic materials

To elucidate the short- and long-range magnetic ordering in magnetic materials, we also developed a method to reveal the magnetic fluctuations near the Curie temperature through small-angle neutron scattering. A combination of neutron and synchrotron radiation experiments, micromagnetic simulations, and informatics have been used to clarify the short- and long-range magnetic order during the magnetization process and the magnetic interactions between grains. Figure 3 shows the simulation results of the micro-magnetics. As this figure illustrates, we succeeded in visualizing the anisotropic and dipole fields inside the hot-deformed magnet. We also developed a methodology for predicting a magnetization reversal behavior by combining micromagnetic simulations and information science.

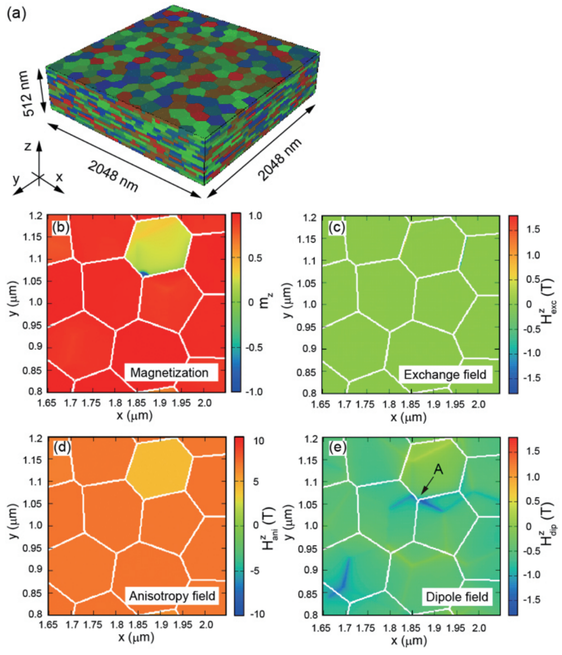

Fig. 3: Visualization of magnetization, exchange field, anisotropy field, and dipole field in hot-deformed magnet