Recent progress in studies on crystalline phases and magnetic domain structures in high coercivity permanent magnets using synchrotron X-rays

T. Nakamura*

(JASRI, *ESICMM)

In the last three decades, synchrotron X-rays have been widely used in materials science. To date, they have become essential means of getting information about crystal structures and electronic and magnetic properties of materials. In the earlier studies, the structural, physical and chemical properties had been investigated mostly in pristine substance like the single crystals or the single phase compounds, whilst the recent applications of the synchrotron X-rays extend to practical materials which are generally polycrystalline and inhomogeneous. In addition, the *in situ* measurements are becoming more popular in order to get better understanding about what happens in materials during manufacturing processes or practical uses.

A study of permanent magnets using the synchrotron X-rays is such a recent subject since magnets are usually inhomogeneous with the microstructure. A Nd-Fe-B sintered magnet has been the strongest permanent magnet since its invention by Sagawa et al. in 1983 [1] and is widely used in many applications such as electric vehicles, wind power generators and voice coil motors, which are crucial for realizing a sustainable society. However, the coercivity mechanism in the Nd-Fe-B sintered magnets remains a debated issue in which the practical coercivity of approximately 1–2 T is rather moderate compared to the value expected from its magnetic anisotropy field of approximately 7.5 T. We have, therefore, carried out the synchrotron based characterizations for the analysis of structural and magnetic properties of the Nd-Fe-B sintered magnets by applying X-ray diffraction (XRD) and X-ray magnetic circular dichroism (XMCD) experiments, respectively. In this talk, I will present recent studies on structural and magnetic properties in high performance permanent magnets using the synchrotron X-rays at SPring-8. I will also illustrate the developments of measurement techniques which have were motivated by the necessity of the characterization of the permanent magnets.

Figure 1 shows the summary of the measurement techniques and their measured examples which have been obtained in studies of the Nd-Fe-B sintered magnets at SPring-8. In the experiment using soft XMCD, the fractured surface in Nd-Fe-B-Cu sintered magnet was investigated to evaluate the magnetic property of the grain boundary (GB) phases directly [2]. This work has clarified that the GB phase which is exposed in the fractured surface shows ferromagnetic at room temperature and have the lower Curie temperature than that of $Nd_2Fe_{14}B$ crystal by about 50 °C. Since this result was recorded using an unfocused soft X-ray beam with the irradiated area of about sub-mm^2 in the sample surface, the magnetic property were detected as the laterally averaged from a number of grains. In order to increase the spatial resolution in the soft XMCD measurement, we have developed a scanning soft XMCD microspectroscopy measurement technique with the apparatus equipped with an 8 T superconducting magnet at BL25SU of SPring-8 [3]. This observation technique using the focused soft X-ray beam with the beam size of about 100 nm makes us possible to visualize magnetic domains with the elemental distribution not only for the flat surface like the polished one but also for the irregular surface like the fractured one. The substantial difference in the magnetic domain reversal is observed for the polished and fracture surfaces [4]. The magnetic domain reversal behavior is also compared with the result of the FORK measurement, showing that the magnetic domain observation is valuable to give an interpretation of the FORK diagram [5]. Regarding the uses of the hard X-rays, we performed the scanning hard XMCD microspectroscopy experiment using the focused X-ray beam as small as 100 nm^2 at BL39XU of SPring-8 [6]. In the magnetic domain observation using the hard X-rays, the surface insensitive observation becomes possible though the deeper probing depth decrease the lateral spatial resolution. More recently, the observation technique for visualizing magnetic domains three dimensionally has been developed using the focused hard X-ray beam. This development directs the future uses of the magnetic domain observation techniques, where the soft and hard X-rays will probe the surface two dimensionally and the interior three dimensionally, respectively. In the structural analysis, the *in situ* XRD measurement at elevated temperature and the Rietveld analysis have also been applied to studies of the Nd-Fe-B-Cu sintered magnet in order to evaluate the variation of the constituent phases during the annealing process. As the result of the Rietveld analysis, it is implied that dhcp Nd phase in the Nd-Fe-B-Cu sintered magnet contains a certain amount of oxygen and shows the phase transition to the fcc structure when the internal stress is removed [7]. Here, I would like to emphasize that the structure and/or magnetic property of crystalline phases in materials with the microstructure are possibly different from their pristine materials, meaning that the new functional material would be found even in materials which is practically used at present. The synchrotron X-ray measurement technique with the nano-scale resolution will possibly becomes a tool to discover a new material which will make an innovation.

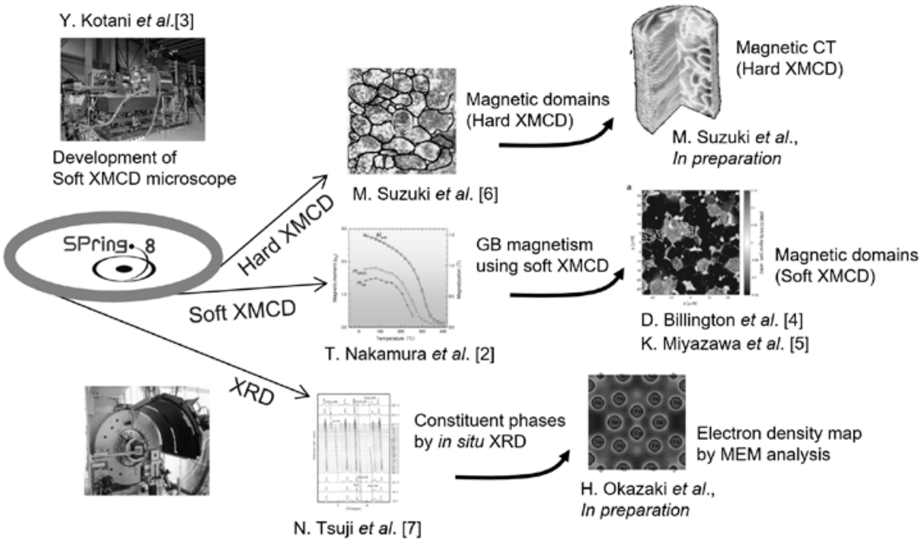

Fig.1 Measured examples in synchrotron X-ray experiments and developed measurements techniques at SPring-8 for studies of permanent magnets.

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