

Development of Spin SEM Technology for Observation of Magnetic Domains

Teruo Kohashi, Dr. Sc.

Kumi Motai, Dr. Sc.

OVERVIEW: Electron microscope technology developed by Hitachi is used for testing and analysis of a wide range of semiconductors and other hardware devices. Spin SEMs are able to observe the magnetic structure of a device at the nanometer scale and are already used for applications such as evaluating the shape of storage bits on magnetic media. Hitachi has succeeded in implementing two new observation techniques in order to deal with the increasingly diverse range of device functions and evaluation criteria, namely observation of structural changes in magnetic domains at high temperatures (up to 500°C) and observation of samples with remanent magnetization resulting in magnetic stray fields. It is anticipated that these observation techniques will contribute to improvements in the performance of magnetic devices, including playing a role in the development of powerful new magnetic materials.

INTRODUCTION

BECAUSE high-resolution observation of magnetic domains allows device characteristics to be analyzed at the micro level, it is an effective technique for the evaluation of magnetic devices as they continue to improve in performance. Electron spin is a physical quantity, and the spin-polarized scanning electron microscope (spin SEM) is an instrument for the observation of magnetic domains that works by detecting and imaging the spin of electrons inside ferromagnetic samples (see Fig. 1).

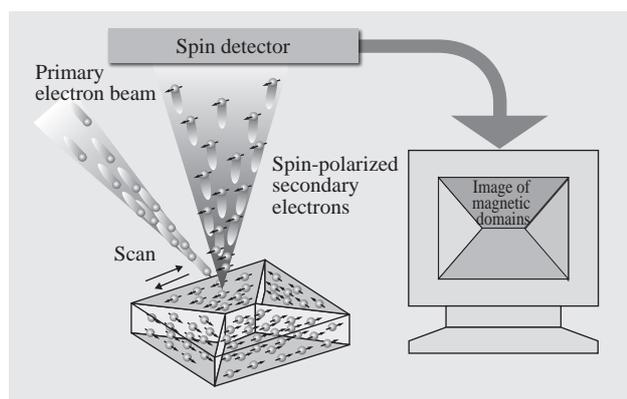


Fig. 1—Diagram of Magnetic Domain Observation Using Spin SEM.

By exposing a sample to a primary electron beam and detecting the spin polarization of the emitted secondary electrons, the magnetic orientation at that point on the sample can be determined. This primary electron beam can then be scanned over the sample to build up an image of its magnetic domains.

A common cause of magnetism is when the electron spins inside a sample of magnetic material become aligned (polarized). It is known that, if an electron beam is applied to a sample and it causes electrons from inside the sample to be emitted, those electrons will retain this spin polarization. Accordingly, if a tightly focused primary electron beam is applied to the sample and a detector is used to measure the spin polarization of the emitted secondary electrons, the magnetic orientation at the point from which the secondary electrons were emitted can be determined. By progressively scanning the primary electron beam across the surface of the sample, an image of its magnetic domains can be obtained. The key feature of this instrument is the spin detector for measuring the electron spin polarization.

It works by Mott scattering whereby spin-polarized electrons incident on a thin metal foil result in an asymmetric pattern of scattering directions⁽¹⁾. This principle can be exploited to obtain a quantitative, three-dimensional map of a material's magnetic orientations, with characteristics that include an excellent ability to handle any sample shapes and the ability to get an information about magnetization separately from physical shape. Hitachi led the world in developing spin SEMs⁽¹⁾, and since then has made continued improvements to sensitivity and resolution^{(2),(3)}, utilizing the instrument to measure the characteristics of a wide range of different magnetic devices.

In the past, spin SEM measurement has not been well suited to observation of permanent magnets and other materials with strong remanent magnetization, and examples of such use have been rare. This is because the magnetic stray field from such samples affects both the incident primary electron beam and the emitted secondary electrons that constitute the signal, degrading performance characteristics such as the resolution and signal-to-noise (S/N) ratio of the image. Similarly, experiments that involve raising a sample to high temperature can have a strong influence on things like vacuum integrity and electron lensing. Consequently, reports of such use in the literature are all but non-existent. In recent years, however, performance improvements and a greater diversity in the nature of magnetic devices have made these types of observations more important. They are including observations of changes in magnetic domains caused by temperature and external magnetic fields, and also observations of the structure of magnetic domains in materials with remanent magnetization, such as permanent magnets. Accordingly, we have developed new mechanisms that allow a spin SEM to be used for observations of samples that have been raised to 500°C, and samples with a magnetic field of 80 kA/m at the material surface.

These mechanisms are described here, together with example applications.

SAMPLE STAGE INCORPORATING HEATING AND MAGNETIC STRAY FIELD SHIELDING MECHANISMS

This section describes the design of a sample holder that incorporates the mechanisms referred to above. Sample heating is performed using a pyrolytic boron nitride (PBN) ceramic heater. Fig. 2 shows how the different parts are positioned in the sample holder. Fig. 2 (a) shows a plan view and (b) shows a lateral cross section. Although not described here, a mechanism for applying a magnetic field to the sample was developed in parallel. As this involves locating a magnet directly under the sample to provide the magnetic field, the placement of the heater needed to leave a gap of approximately 10 mm for this purpose. The heat from the heater is conveyed to the sample via a thermally conductive copper sheet. As overheating the under-sample magnet would degrade its magnetism, a thermally insulating plate (stainless steel) is positioned between it and the heater. This design succeeded in minimizing the locations that are raised to a high temperature and limited any negative effects of sample heating, such as loss of vacuum or

impairment of the electron lensing apparatus located just above the sample.

For observations of material with remanent magnetization, a magnetic shield is required that can block the magnetic stray field and reduce its influence on the primary and secondary electron beams. This mechanism was designed and developed using a three-dimensional simulation of the electron path. The magnetic shield was made from 0.5-mm permalloy sheet cut to size so that it could be placed like a shroud over the top of the sample holder, and cut with a slit 5 mm long and 1 mm wide. The shield was then positioned so that the slit aligned with the region of sample surface to be observed. The probe electron beam passes through the slit to the sample surface,

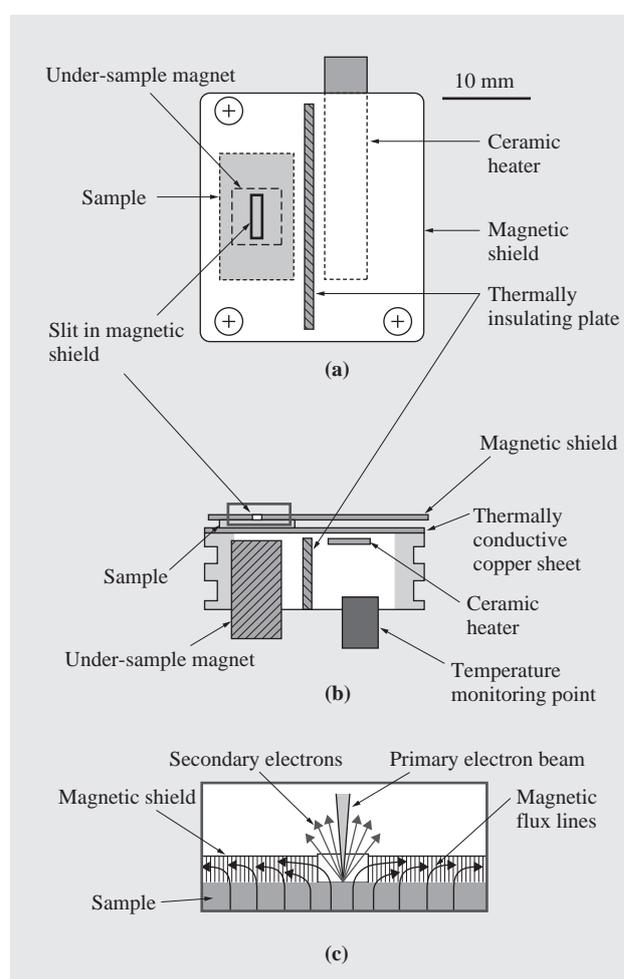


Fig. 2—Plan View (a), Lateral Cross Section (b), and Detail (c) of Sample Holder.

Diagrams (a) and (b) show the relative positions of the sample, heater, under-sample magnet, and magnetic shield in the mechanism for sample heating and observing samples with remanent magnetization. Diagram (c) is an enlargement of the gray box in diagram (b) and shows how the magnetic shield blocks the magnetic stray field from the sample.

and the secondary electrons are also captured by the detector through the slit. As the magnetic stray field from the sample is blocked by the magnetic shield, as shown in Fig. 2 (c), its influence on the primary electron beam and secondary electrons is significantly reduced. The mechanism was fabricated and then tested on material with a magnetic stray field to demonstrate that the shielding functioned adequately.

SAMPLE HEATING TESTS

Using the sample heating mechanism described above, high-temperature observations were made of the magnetic domains on the surface of a cobalt (Co) singlecrystal (0001). Fig. 3 shows the images captured at 100°C intervals from 100°C up to 500°C.

While the sample has a magnetic easy axis (axis at which the material is easily magnetized) perpendicular to its surface [the (0001) axis], it is also known that its structure is characterized by closure magnetic domains on its surface due to magnetostatic energy. As the observations show only a small component perpendicular to the surface, the images in Fig. 3 were produced by color-coding the result of calculating the magnetic orientations that lay on the sample surface.

The 100°C image shows magnetic domains with a size of 2 to 3 μm , reflecting the symmetry of the crystal structure. While any change in domain shape as the temperature increases remained comparatively small up to 200°C, the small domains had all disappeared by 300°C as the structure changed to one with large magnetic domains of 10 μm or more. As the temperature rose further from 400°C to 500°C, the formation of small new domains with sizes of 2 to 3 μm was observed to occur inside the large domains.

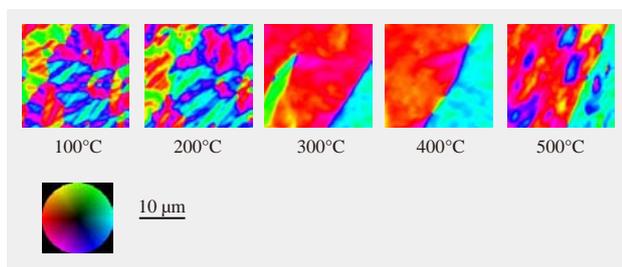


Fig. 3—Changes in Magnetic Domains with Rising Temperature in Co (0001).

The images were color coded to indicate the magnetization vector (as indicated by the color circle on the bottom left). Major changes in magnetic domain structure were evident between 200°C and 300°C, and between 400°C and 500°C. These are believed to correspond to changes in the magnetic anisotropy and in the crystal structure that occurred between these temperatures.

It is known that the magnetic easy axis in the (0001) orientation that exists in the Co singlecrystal at room temperature changes to a (0001) in-plane orientation at temperatures above the phase transition that occurs around 200°C. At temperatures below the phase transition, therefore, the magnetization inside the sample is oriented in the (0001) axis, whereas magnetostatic energy causes the structure on the sample surface to be characterized by small closure magnetic domains. At temperatures above the phase transition, on the other hand, the (0001) in-plane orientation (that is, an orientation along the surface) becomes an easy axis and therefore large stable domains oriented along in-surface axes form both in the interior and on the surface of the sample. In this experiment, it is assumed that this phase transition was the cause of the major change in magnetic domains that occurred between 200°C and 300°C. Similarly, as Co singlecrystal changes from an hcp structure to an fcc structure at around 450°C, it is assumed that this was the reason for the second change in domain structure. Using the spin SEM in this way to observe the magnetic domains of a Co (0001) surface allowed direct observations to be made of the magnetic domain changes associated with phase transitions reported in the literature, and therefore it demonstrated that the newly developed sample heating mechanism was working correctly.

The intention is to use this technology for the observation of magnetic devices, the high-temperature magnetic properties of which are a subject of interest. In the case of the powerful neodymium-iron-boron (NdFeB) magnets used in the electric motors that drive hybrid cars, for example, there is a need to assess changes in magnetic domains at high temperatures in order to make further improvements in the magnets' high-temperature coercivity (resistance to becoming demagnetized).

EXAMPLE OBSERVATIONS OF REMANENT MAGNETIZATION

This section describes observations of a permanent magnet (a material with remanent magnetization)⁽⁴⁾. The sample was a NdFeB magnet with fine-grained anisotropic crystals and the experiment, described below, studied the relationship between changes in the magnetic domains and crystal grain shape. First, spin SEM images of the magnetic domains and surface shape were obtained for the magnet in a thermally demagnetized state (in which it had zero magnetization). The sample was then removed and

placed in a separate apparatus where it was exposed to a magnetic field of 80 kA/m. After the magnetic field was turned off, leaving the material in a state of remanent magnetization, the sample was then returned to the spin SEM and further observations were made. This procedure was then repeated with the applied magnetic field increased by 80 kA/m each time until the magnetization became identical in the field of view. The changes in the remanent magnetization of the magnetic domains at each stage were observed.

Images (a) to (k) in Fig. 4 show some example results. The images are oriented so that the magnetic easy axes of the crystal grains point upwards. In the thermally demagnetized state shown in image (a), the bands of light and dark represent the magnetic domains. The light regions indicate an upward orientation, the dark regions a downward orientation, and non-magnetic regions (corresponding to impurities, etc.) appear gray. The areas of the light and dark regions in this view are roughly equal.

Image (b) shows the shape for the same area as image (a). A crack can be seen running up the left side of the image and an impurity with a size slightly less than $1\ \mu\text{m}$ is visible on the right. The granular structures with sizes in the 0.3 to $0.5\text{-}\mu\text{m}$ range observable elsewhere in the image as thin lines contrasted against the background correspond to the individual crystals that make up the magnet. As the contrast is inadequate in some places, not all the crystals can be resolved.

To investigate the relationship between the shape image and magnetic domain walls, image (c) was generated by overlaying the magnetic domain walls obtained from image (a) on image (b). This shows some places where the magnetic domain walls coincide with the crystal grain boundaries as well as some places where they do not. As the magnetic domain walls tend to run in straight lines in order to minimize their area, they sometimes transect crystal grains. Grains where this occurs have multiple magnetic domains, which means they have two different directions of magnetization within the same grain. As the spin SEM can be used in this way to study shape and magnetization in parallel, it provides a way to analyze the relationship between crystal shapes and magnetic domain walls.

Images (d) to (k) in Fig. 4 show the magnetic domains for the sample in a state of remanent magnetization resulting from the successive exposure of magnetic fields oriented in the upward direction. As exposure of the magnetic fields required the sample to be removed from the microscope, the crack and

impurities visible in the shape image (b) were used as landmarks to align the viewpoints for each observation. The position of a particular crystal grain at the bottom left of the shape image (b) is enclosed by a white border and indicated by an arrow. The same crystal grain is also shown in the magnetic domain images to indicate the relationship between the viewpoints in each image.

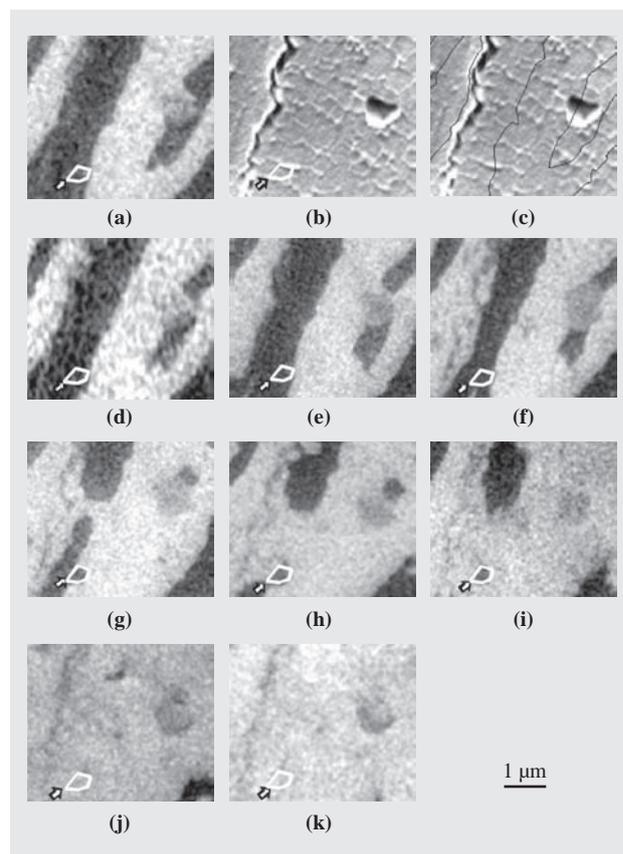


Fig. 4—Spin SEM Images of Sample at Varying Degrees of Magnetization.

The images show the magnetic domains and shape respectively at varying degrees of magnetization, from a thermally demagnetized state up to remanent magnetization after exposure of a field of 640 kA/m. Images (a) and (b) show the magnetic domains and shape images respectively when the sample was in a thermally demagnetized state, and image (c) shows the magnetic domain walls overlaid on image (b). Images (d) to (k) show the magnetic domains for the sample in a state of remanent magnetization after exposure of increasingly strong magnetic fields, the respective field strengths being: 80 kA/m (d), 160 kA/m (e), 240 kA/m (f), 320 kA/m (g), 400 kA/m (h), 480 kA/m (i), 560 kA/m (j), and 640 kA/m (k). In the magnetic domain images, the light regions indicate an upward magnetic orientation and the dark regions a downward magnetic orientation. Although the viewpoint shifts slightly in the images that show the sample in a state of remanent magnetization, the position of a particular crystal grain is enclosed by a white border and highlighted by an arrow. Reprinted with permission from the Journal of The Magnetics Society of Japan⁽⁴⁾.

Looking at the shapes of the magnetic domains, no major changes are evident between the thermally demagnetized sample in image (a) and the 240-kA/m image (f), except for a gradual thinning of the dark regions and an expansion of the light regions. In the 320-kA/m remanent magnetization image (g), the large dark magnetic domain visible in the earlier images has shrunk and split in two, and this shrinkage becomes rapidly more pronounced in images (h) through (j) (400 to 560 kA/m) until it is seen to disappear.

Through this process, the shapes of the magnetic domain walls can be observed changing from smooth and linear in the thermally demagnetized image (a) to more complex and undulating lines in the images for 320 kA/m and higher. At around 0.3 to 0.5 μm , the size of these undulations is similar to that of the crystal grains, suggesting that the magnetic orientation is being determined at the level of single grains. For example, the crystal grain highlighted at the bottom-left of the magnetic domain images remains part of the large dark domain from the thermally demagnetized image (a) up to the 240-kA/m remanent magnetization image (f). In the 320-kA/m image (g), this crystal grain still retains its magnetic orientation but the magnetization of part of the surrounding area has flipped polarity, leaving this crystal grain protruding from the area of dark magnetic domain and causing the magnetic domain wall to take on an undulating shape. In the 400-kA/m image (h), the flipping of polarity in the surrounding area is more advanced, and by the 480-kA/m image (i) the crystal grain has become its own isolated magnetic domain. Finally, in the 560-kA/m image (j), the magnetic polarity of this crystal grain also flips.

These results show how the retention and flipping of magnetic polarity occur at the level of crystal grains and are different to the multi-domain structure suggested by the sample when it was in a thermally demagnetized state. Also flipping of the magnetization polarity occurs at the level of individual crystal grains at different magnetic fields. The use of data such as these to provide pointers on how to improve the coercivity of the magnet will be a topic for future work.

During this experiment, the remanent magnetization of the sample was progressively increased until it finally reached more than 0.2 T. While there were concerns about the magnetic stray field at the surface of the sample, the measures described earlier meant that magnetic domain images could be obtained without magnetic stray fields influencing the results.

The research described in this article was conducted jointly with the NEOMAX Company, Hitachi Metals, Ltd.

CONCLUSIONS

Nearly 30 years after the spin SEM was first developed, advances in this field continue to be made. This article has described an observation technique for investigating changes in magnetic domains. Meanwhile, further planned developments include work on achieving higher resolutions and on satisfying user demands such as the ability to make observations of the grain boundary regions in NdFeB magnets. Hitachi aims to continue contributing to improvements in magnetic device performance through the use of microscopic analysis, while also making enhancements to detector sensitivity and other aspects of microscope technology.

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ABOUT THE AUTHORS



Teruo Kohashi, Dr. Sc.

Joined Hitachi, Ltd. in 1991, and now works at the Advanced Measurement and Analysis Center, Central Research Laboratory. He is currently engaged in work on spin-polarized electron optics and its application to the analysis of materials and devices. Dr. Kohashi is a member of The Magnetics Society of Japan, The Japan Institute of Metals, and The Japanese Society of Microscopy.



Kumi Motai, Dr. Sc.

Joined Hitachi, Ltd. in 1994, and now works at the Advanced Measurement and Analysis Center, Central Research Laboratory. She is currently engaged in work on surface analysis of materials and devices using auger electron spectroscopy and spin-polarized scanning electron microscopy. Dr. Motai is a member of The Surface Science Society of Japan, The Physical Society of Japan, and The Japan Society of Applied Physics.