Magnetic textures in a hexaferrite thin film and their response to magnetic fields revealed by phase microscopy

Atsuhiro Kotani1, Ken Harada2, Marek Malac3,4, Hiroshi Nakajima1, Kosuke Kurushima5, and Shigeo Mori1*

1Department of Materials Science, Osaka Prefecture University, Sakai, Osaka 599-8531, Japan
2Center for Emergent Matter Science, The Institute of Physical and Chemical Research (RIKEN), Hatoyama, Saitama 350-0395, Japan
3Nanotechnology Research Centre, National Research Council (NRC), Edmonton, Alberta T6G 2M9, Canada
4Department of Physics, University of Alberta, Edmonton, Alberta T6G 2E1, Canada
5Toray Research Center, Otsu, Shiga 520-8567, Japan

E-mail: mori@mtr.osakafu-u.ac.jp

Received February 20, 2019; revised April 15, 2019; accepted April 22, 2019; published online May 24, 2019

We investigated magnetic textures in a Sc-doped hexaferrite film by means of phase microscopy with a hole-free phase plate in a transmission electron microscope. In a zero magnetic field, the stripe-shaped magnetic domains coexist with magnetic bubbles. The magnetization in both magnetic domains was oriented perpendicular to the film and the domain walls have an in-plane magnetization. In the remnant state at 9.2 mT, a number of magnetic bubbles were formed with the formation of stripe-shaped magnetic domains, and the out-of-plane component in the stripe-shaped domains gradually appeared as the film thickness increased. As the film thickness increased further, the magnetic bubbles with clockwise or counterclockwise spin helicities formed a triangular lattice. These results in the remnant state suggest that the domain-wall energy in the magnetic bubble domains is lower in the thicker region. © 2019 The Japan Society of Applied Physics

1. Introduction

Phase microscopy (PM), especially with a hole-free phase plate (HFPP) in a transmission electron microscope (TEM), has been used to enhance phase contrasts of materials consisting of the light elements in biological fields.1,3 PM has a potential advantage in that the highly magnified images can be obtained in-focus, thus not suffering from Fresnel fringes caused by defocusing.4,5 Unlike in electron holography, PM observation does not require a reference wave. Therefore, PM has been utilized for imaging the magnetization distribution of magnetic textures.6 For example, skyrmions,7–10 vortex-like magnetic textures were observed using PM with an HFPP, and the semi-quantitative magnetic distribution was reproduced from the acquired PM image.11 In this paper, we report that PM with an HFPP can be applied to observe the nanoscale magnetic textures, such as stripe-shaped magnetic domains and magnetic bubbles,12–16 in a uniaxial ferromagnet Sc-substituted M-type hexaferrite, BaFe10.35Sc1.6Mg0.05O19 (BFSMO). It has been revealed by phase microscopy that the thickness dependence of the magnetization distribution in the magnetic textures in the remnant state was revealed in the PM observation.

2. Experimental methods

A single crystal of BFSMO specimens was synthesized via the floating zone method.19 Specimens for TEM observation were thinned using Ar ion milling. The observations using PM with an HFPP were performed in a 300 kV TEM (Hitachi HF-3300). We utilized a 13-nm-thick amorphous carbon film prepared by electron beam evaporation as an HFPP.13 After the installation of the HFPP to the microscope column, the HFPP was heated to approximately 200 °C and kept at that temperature during the experiment to prevent the HFPP film from contamination.21,22

We used the PM optics constructed in previous studies,11 as illustrated in Fig. 1. The HFPP was placed at the selected-area aperture plane and the condenser lens was adjusted to construct the crossover at the HFPP. The application of magnetic fields perpendicular to the thin film was achieved by exciting the objective lens. The deviation of the crossover from the selected aperture position caused by exciting the objective lens was compensated by using the condenser lens. Image focusing was achieved by adjusting the excitation of the intermediate lens.

The phase shift Δφ is expressed as follows:23

$$\Delta \phi(x, y) = C_E V_0 (x, y) I (x, y) - \frac{2 \pi e}{h} \int B(x, y) dS. \tag{1}$$

Here, $C_E$ and $V_0$ and $I$, are the interaction constant which is 0.00652 rad V$^{-1}$ nm$^{-1}$ for 300 keV electrons, mean inner potential and specimen thickness, respectively. $B$ is the magnetic flux of the specimen magnetization. The phase shift depends on both the mean inner potential and the specimen thickness. Assuming that the spatial change in the thickness can be negligible for the change in magnetization, the magnetization maps can be obtained from gradients of the phase distribution on the PM image acquired with an HFPP as follows:11

$$B = \frac{h}{2 \pi e} \Delta \phi(x, y) = \frac{h}{2 \pi e} \Delta I(x, y)$$

$$= \frac{h}{2 \pi e} \left( \frac{\partial I}{\partial x} \right). \tag{2}$$

Here, $B$, $\phi$ and $I$, are the magnetic flux of the specimen magnetization, phase shift due to the magnetic flux and...
intensity of the phase image. The absolute value of the in-plane magnetization can be obtained from the following equation:

\[ |B| = \frac{\hbar}{2\pi e} \left( \left( \frac{\partial I}{\partial x} \right)^2 + \left( \frac{\partial I}{\partial y} \right)^2 \right)^{1/2}. \]  

(3)

Note that the above equations are valid when an image recording device has a linear relationship between the number of detected electrons on the detector and the output intensity on the display, providing that the phase shift is proportional to the image contrast. Using these methods, we obtained magnetization maps from the PM images under magnetic fields applied externally.

3. Results and discussion

First, magnetic textures of BFSMO in zero magnetic field at RT were examined by PM observations. Figure 2(a) shows a PM image, showing the stripe-shaped magnetic domains and several magnetic bubbles. To extract the magnetization map of the BFSMO specimen in Fig. 2(b), differential images of the intensity in Fig. 2(a) were calculated according to Eq. (2). Figs. 2(c) and 2(d) show the gradient images of the x and y directions, respectively. The white and black indicate positive and negative differential values, respectively. The magnetization vector map in Fig. 2(b) was obtained from Figs. 2(c) and 2(d). The color map indicates the direction of magnetization, coded according to the color wheel, while the color saturation indicates the in-plane component of the magnetization intensity calculated using Eq. (3).

In Fig. 2(b) the in-plane magnetization is indicated by white arrows. This shows the magnetization in the domain walls between the stripe-shaped magnetic domains is oriented parallel to the in-plane direction. The in-plane component of the magnetization was not detected in the stripe-shaped magnetic domains and magnetic bubbles, which suggests the magnetization in those domains is mainly oriented parallel to the easy axis (perpendicular to the thin film). Furthermore, it appears that the magnetization in the circular domain walls of the magnetic bubbles rotates clockwise (CW) or counterclockwise (CCW) in the plane of the thin film. The magnetization distribution of the magnetic bubbles obtained with PM is similar to that obtained by phase reconstructions through an iterative calculation using a series of 32 defocused images.24)

The magnetic texture varied depending on the strength of the applied external magnetic field. Figs. 3(a) and 3(b) show the PM image and the magnetization map at 80 mT of the magnetic field applied, respectively. It can be seen in Fig. 3(a) that the width of the stripe-shaped magnetic domains is decreased and simultaneously the diameter of the magnetic bubbles is reduced as the strength of the magnetic field applied is increasing due to the Zeeman effect.

A Bloch line is formed, as indicated by the white arrowhead in Fig. 3(b). Note that a Bloch line is characteristic for the reversal of the domain-wall chirality, in which it has been accepted that the directions of the in-plane magnetization are reversed by gradual rather than abrupt rotation. The magnetization distribution obtained from Fig. 3(b) is schematically shown in Fig. 3(c). It has been recently recognized that the Bloch line plays an important role in the formation of magnetic bubbles.19) As understood by comparing Figs. 2(b) and 3(b), a Bloch line was formed from the stripe-shaped magnetic domains by applying an external magnetic field.

As the strength of the magnetic field increases to 120 mT, magnetic bubbles are formed from the stripe-shaped magnetic domains. Figure 3(d) shows the PM image of magnetic bubbles formed at 120 mT. Figure 3(e) is the magnetization map of magnetic bubbles indicated by the white dotted lines.

Fig. 1. (Color online) Schematic illustration of the optic for PM using an HFPP. Red arrows indicate a magnetic field applied by a weakly excited objective lens.

Fig. 2. (Color online) (a) PM image in a zero magnetic field at RT. (b) Magnetization map calculated from the differential images of (a) using the areas surrounded by the white dotted lines. Color and the white arrows indicate the magnetization direction as described by the color wheel. (c) and (d) show the differential images in (a) in the x and y direction, respectively.
in Fig. 3(d). The magnetic bubbles have CW or CCW rotation of the magnetization. From the magnetization maps, the diameter of magnetic bubbles inside the circular domain wall can be estimated to be approximately 230 nm at 0 mT, 150 nm at 80 mT and 100 nm at 120 mT, respectively. These results show that the diameters of magnetic bubbles decrease as the strength of the magnetic field increases.

After the external magnetic field up to 2 T was applied by exciting the objective lens, the lens was turned off to reduce the magnetic field quickly to 9.2 mT. Figure 4 shows that in the remnant state a number of magnetic bubbles were formed in the region far from the edge of the specimen and the magnetic stripe domain exists in the region near the edge. The specimen in this study was made by ion milling and has...
a wedge-like thickness profile with progressively lower thickness closer to the edge. The thickness values in the region indicated by I, II and III in Fig. 4 measured using electron energy-loss spectroscopy are 39, 96 and 163 nm, respectively. Therefore, it is shown that the stripe-shaped magnetic domains are formed in the thinner region and magnetic bubbles are formed in the thicker region.

Here, the magnetization distribution of stripe-shaped magnetic domains and magnetic bubbles are discussed. PM observations revealed changes in the magnetization distribution of the stripe-shaped magnetic domains and the transformation from the domains into magnetic bubbles with increasing the film thickness. Figure 5 shows the three magnetization maps of regions I, II and III in Fig. 4 calculated using Eq. (3). The white arrows indicate the magnetization direction. The magnetization map obtained from I shows that the magnetization in the stripe-shaped magnetic domain increases as the film thickness increases. In magnetization map II, the dark region where the magnetization is oriented perpendicular to the thin film in the stripe-shaped magnetic domain expands. These results show that the thickness dependence of the magnetization distributions of stripe-shaped magnetic domains can be clarified using PM with an HFPP.

For stripe-shaped magnetic domains, it has been reported that the thickness-driven reorientation of the magnetization from in-plane to perpendicular is caused because the demagnetization field whose contribution favors an in-plane preferential orientation for the magnetization decreases as the thickness increases. Critical thickness, \( t_C \), of the reorientation is given as follows:

\[
t_C \sim 27.2 A^{1/2} M_S^2/K_U^{3/2},
\]

where \( A \) is the exchange stiffness constant, \( M_S \) is the saturation magnetization and \( K_U \) is the uniaxial anisotropy constant. The parameters of the BFSMO specimen were reported to be \( A = 1.3 \times 10^{-6} \text{ erg cm}^{-1} \), \( M_S = 286 \text{ emu cm}^{-3} \) and \( K_U = 5.3 \times 10^6 \text{ erg cm}^{-3} \), resulting in \( t_C \sim 67 \text{ nm} \). The PM observations in Figs. 5(I) and 5(II) experimentally show the reorientation of the magnetization across \( t_C \).

As the film thickness increases to more than 100 nm, magnetic bubbles are formed. Fig. 5(III) shows a magnetization map of the region III in Fig. 4. The magnetization distribution of magnetic bubbles with the CW or CCW spin rotation in the magnetic domain wall can be seen clearly, and those magnetic bubbles are formed locally in a triangular lattice. As shown in the magnetization maps in Figs. 2 and 3, the black and white balls in Fig. 4 correspond to the magnetic bubbles with CW and CCW rotation of the magnetization, respectively. It can be seen in Fig. 4 that the same quantity of those magnetic bubbles exists and the magnetization helicities are oriented randomly. Therefore, it is suggested that the energies in the magnetic bubbles with the CW or CCW helicities are equivalent randomly. It appears that the formation of magnetic bubbles in the remnant state depends on the film thickness.

It was reported that the magnetic energy of the stripe-shaped magnetic domain is higher than that of the magnetic bubble by magnetic domain-wall energy.\(^{28}\) The wall energy \( \sigma_W \) of a Bloch domain wall in a uniaxial ferrimagnet is expressed as follows:\(^{29}\)

\[
\sigma_W = 4 \sqrt{AK}.
\]

The effective magnetic anisotropy \( K \) related to \( \sigma_W \) decrease as the thickness increases.\(^{30}\) Thus, judging from Eq. (4), the formation of magnetic bubbles in the remnant state is induced by lower \( \sigma_W \) in the thicker region, as shown in Fig. 5(III). The mechanism of thickness dependence in the formation of magnetic bubbles in the remnant state will be revealed in the future by the experiments with thickness-controlled films and theoretical simulations.

4. Conclusions

In conclusion, PM with an HFPP was utilized to reveal the magnetization distributions of magnetic textures, such as stripe-shaped magnetic domains and magnetic bubbles in BFSMO and their response to external magnetic fields applied perpendicular to the thin film. The stripe-shaped magnetic domains and a few magnetic bubbles coexisted in a zero magnetic field. PM observation revealed that the magnetic domain structures consist of the domain wall with the in-plane magnetization and the domain with the magnetization perpendicular to the film. In the remnant state after the external magnetic field was applied up to 2 T and was decreased, a number of magnetic bubbles with the stripe-shaped magnetic domains were observed. Changes in the magnetization distribution in the stripe-shaped magnetic
domains were revealed as the film thickness increased. In addition, it was revealed that magnetic bubbles with CW or CCW spin helicities were formed in a triangular lattice as the film thickness increased further. The PM with an HFPP will be a powerful tool to analyze the magnetic distribution of complex magnetic textures.

Acknowledgments

The authors gratefully acknowledge Kai Cui and Mark Salomons for technical support during the PM experiments and Jean Nassar for support with programming using the Python code. This work was partially supported by JSPS KAKENHI (Grant No. 18J12180) and the Graduate Program for System-inspired Leaders in Material Science (SiMS) of Osaka Prefecture University by the Ministry of Education, Culture, Sports, Science and Technology of Japan. This work was also supported by the National Research Council (NRC) in Canada. The experiments were made possible by outstanding support from Hitachi High Technologies by providing the Hitachi HF-3300 microscope at NRC-Nano.

ORCID iDs

Hiroshi Nakajima @ https://orcid.org/0000-0002-0366-5468

1) M. Malac, M. Beleggia, M. Kawasaki, P. Li, and R. F. Egerton, Ultramicroscopy 118, 77 (2012).
2) R. Danev, B. Bijsses, M. Khoshouei, J. M. Plitzko, and W. Baumeister, Proc. Natl Acad. Sci. 111, 15635 (2014).
3) Y. Fukuda, U. Laugks, V. Lučić, W. Baumeister, and R. Danev, J. Struct. Biol. 190, 143 (2015).
4) M. Malac, S. Hettler, M. Hayashida, M. Kawasaki, Y. Konyuba, Y. Okura, H. Iijima, I. Ishikawa, and M. Beleggia, Micron 100, 10 (2017).
5) M. Hayashida, K. Cui, A. M. Najarian, R. McCreery, N. Jehanathan, C. Pawlowicz, S. Motoki, M. Kawasaki, Y. Konyuba, and M. Malac, Micron 116, 54 (2019).
6) S. Pollard, M. Malac, M. Beleggia, M. Kawasaki, and Y. Zhu, Appl. Phys. Lett. 102, 192101 (2013).
7) S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Science 323, 915 (2009).
8) N. Nagaosa and Y. Tokura, Nat. Nanotechnol. 8, 899 (2013).
9) S. A. Montoya et al., Phys. Rev. B 95, 024415 (2017).
10) H. Nakajima, A. Kotani, M. Mochizuki, K. Harada, and S. Mori, Cit. Appl. Phys. Lett. 111, 192401 (2017).
11) A. Kotani, K. Harada, M. Malac, M. Salomons, M. Hayashida, and S. Mori, AIP Adv. 8, 055216 (2018).
12) S. R. Herd and P. Chaudhari, Phys. Status Solidi (a) 18, 603 (1973).
13) P. J. Grundy and S. R. Herd, Phys. Status Solidi (a) 20, 295 (1973).
14) A. Kotani, H. Nakajima, Y. Ishii, K. Harada, and S. Mori, AIP Adv. 6, 056403 (2016).
15) A. Kotani, H. Nakajima, K. Harada, Y. Ishii, and S. Mori, Phys. Rev. B 94, 024407 (2016).
16) H. Nakajima, A. Kotani, K. Harada, Y. Ishii, and S. Mori, Microscopy 65, 473 (2016).
17) X. Yu, M. Mostovoy, Y. Tokunaga, W. Zhang, K. Kimoto, Y. Matsu, Y. Kaneko, N. Nagaosa, and Y. Tokura, Proc. Natl Acad. Sci. 109, 8856 (2012).
18) X. Z. Yu, K. Shibata, W. Kohshibae, Y. Tokunaga, Y. Kaneko, T. Nagai, K. Kimoto, Y. Taguchi, N. Nagaosa, and Y. Tokura, Phys. Rev. B 93, 134417 (2016).
19) H. Nakajima, A. Kotani, K. Harada, Y. Ishii, and S. Mori, Phys. Rev. B 94, 224427 (2016).
20) Y. Tokunaga, Y. Kaneko, D. Okuyama, S. Ishiwata, T. Arima, S. Wakoimto, K. Kakurai, Y. Taguchi, and Y. Tokura, Phys. Rev. Lett. 105, 257201 (2010).
21) S. Hettler, M. Dries, P. Herrmann, M. Obermair, D. Gerthsen, and M. Malac, Micron 96, 38 (2017).
22) S. Hettler, E. Kano, M. Dries, D. Gerthsen, L. Pfaffmann, M. Bruns, M. Beleggia, and M. Malac, Ultramicroscopy 184, 252 (2018).
23) H. S. Park et al., Nat. Nanotechnol. 9, 337 (2014).
24) T. Tamura, Y. Nakane, H. Nakajima, S. Mori, K. Harada, and Y. Takai, Microscopy 67, 171 (2018).
25) N. Saito, H. Fujiwara, and Y. Sugita, J. Phys. Soc. Japan 19, 1116 (1964).
26) M. Hehn, S. Padovani, K. Ounadjela, and J. P. Bucher, Phys. Rev. B 54, 3428 (1996).
27) K. Kurushima, K. Tanaka, H. Nakajima, M. Mochizuki, and S. Mori, J. Appl. Phys. 125, 53902 (2019).
28) J. A. Cape and G. W. Lehman, J. Appl. Phys. 42, 5732 (1971).
29) A. Hubert and R. Schäfer, Magnetic Domains: The Analysis of Magnetic Microstructures Title (Springer, Berlin, 1998).
30) K. Chesnel et al., Phys. Rev. B 98, 224404 (2018).