

Hollow-cone Foucault imaging method

Ken Harada1,2*, Atsushi Kawaguchi2, Atsuhiro Kotani2, Yukihiro Fujibayashi2, Keiko Shimada1, and Shigeo Mori2

1CEMS, RIKEN (The Institute of Physical and Chemical Research), Hatoyama, Saitama 350-0395, Japan
2Department of Materials Science, Osaka Prefecture University, Sakai, Osaka 599-8531, Japan

E-mail: kharada@riken.jp

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A hollow-cone Foucault (HCF) imaging method using Lorentz microscopy was developed. Hollow-cone illumination was realized by using deflectors above the specimen and an inclined electron beam circulating with respect to the optical axis. The advantage of the HCF method, having the bright and dark-field modes, is that it can simultaneously visualize both magnetic domains and magnetic domain walls under the in-focus condition. Furthermore, schlieren images, obtained under the specific inclination angle of the illumination beam by adjusting the angle between the bright-field and dark-field modes, can qualitatively visualize the electromagnetic fields in spaces around the specimen.

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In conventional transmission electron microscopy (TEM), observation of biological materials and organic materials with high contrast is difficult because these materials are composed of light elements, resulting in weak scattering for electron beams. Electromagnetic structures in metals or semiconductors, for example, magnetization distributions and dielectric polarization distributions, are also difficult to observe for the same reason. To observe these structures, visualization of phase distributions by electron beams or waves is effective and practical. For this purpose the following observation methods have been developed: electron holography,1–3 Lorentz microscopy,4–8 phase microscopy,9–11 focus-modulation microscopy,12–14 diffractive microscopy,15–17 and ptychography.18–20 These methods, however, require additional equipment and/or techniques to conventional TEM and are, therefore, not suitable for wide and practical applications. Among these methods, Lorentz microscopy is the most effective and widely utilized. There are two methods in Lorentz microscopy: the Fresnel imaging method, in which focus conditions of the specimen images are varied to visualize domain walls, and the Foucault imaging method, in which some of the deflected electron beams are selected for imaging under the in-focus condition to realize high-contrast magnetic or dielectric domain observations. In the Fresnel method, high spatial resolution cannot be obtained because of defocused images. On the other hand, in the Foucault method asymmetric images can be formed because an angle-limiting aperture—an objective aperture in ordinary optics—is placed at the asymmetric position with respect to the optical axis, resulting in azimuthally dependent image contrasts. Furthermore, in Lorentz microscopy electromagnetic fields are not observed because no contrasts are obtained.

To overcome these difficulties in obtaining information of weak scattering objects, the hollow-cone Foucault (HCF) imaging method was developed, where an incident electron beam on the specimen was tilted with respect to the optical axis with an inclination angle and was circulated in all azimuths around the optical axis.21,22 Both magnetic domains and domain walls were simultaneously visualized with sufficient contrasts under the in-focus condition. Furthermore, the schlieren mode allows observation of electromagnetic fields around specimens by appropriately adjusting the inclination angle at the position between the bright-field and dark-field modes.23

Figure 1 shows a schematic diagram of the optical system for HCF imaging. Instead of a conventional objective lens, an objective mini-lens is used to focus the crossover on the selected area (SA) aperture plane. The SA aperture works as an angle-limiting tool just like the conventional objective aperture. The beam deflectors above the specimen incline and circulate the illumination beam in all azimuthal directions around the optical axis.

Fig. 1. (Color online) Schematic diagram of the optical system for the HCF imaging. Instead of a conventional objective lens, an objective mini-lens is used to focus the crossover on the selected area (SA) aperture plane. The SA aperture works as an angle-limiting tool just like the conventional objective aperture. The beam deflectors above the specimen incline and circulate the illumination beam in all azimuthal directions around the optical axis.
specimen having a wide area of about 85 μm in diameter with the inclination angles in the X and Y directions controlled by using the beam deflector system placed above the specimen. The circulating electron beam is illuminated in all azimuthal directions around the optical axis. We note that the special condition of the magnetic field-free space above the specimen due to switching off the objective lens leads to realization of small-angle-tilted hollow-cone beams with an inclination angle as small as $10^{-3}$ rad.

Figure 2(a) shows composite trajectories of the zeroth-order diffraction spots on the SA aperture plane for five inclination angles, 3.3, 4.1, 4.9, 5.7, and $6.5 \times 10^{-3}$ rad. (b) a relation between the input amplitude value and the inclination angle for 0.8 Hz observation. Linear and precise control of the illumination beam are realized.

Figure 2(a) shows composite trajectories of the zeroth-order diffraction spots on the SA aperture plane for five inclination angles. Each trajectory ring was recorded by the hollow-cone beam with about eight turns in 10 s (corresponding to 0.8 Hz in the azimuthal rotation). The diameter of the trajectory rings corresponds to the inclination angle of the hollow-cone beam. Figure 2(b) shows the relation between the input amplitude value and the inclination angle, indicating that a linear and precise angle control was realized in the hollow-cone inclination. The experiment was performed using a 200 kV thermal emission TEM (JEM-2100F, JEOL Ltd) having the optionally attached hollow-cone illumination system. The HCF images were recorded with a 14-bit charge-coupled device camera (2k × 2k pixels, Ultrascan camera, Gatan Inc.). Each image was recorded through about 12 turns in the illumination azimuthal rotation in 7.5 s, corresponding to 1.6 Hz in the azimuth rotation.

To test the validity and efficiency of the developed HCF imaging method, we applied it to observe magnetic domains and domain walls in an Fe₀.₈₈Ga₀.₁₂ alloy (in at%) which has large magnetostriction at room temperature. The thin film of 250 nm thickness with [001] orientation for TEM observation was prepared using a focused ion beam instrument (NB-5000, Hitachi High-Technologies Corp.).

To show the good performance of the HCF imaging method, magnetization structures were observed for comparison using conventional Lorentz microscopy, Fresnel and Foucault methods, under the same experimental conditions as those used in the HCF method.

Figure 3 shows the Fresnel observation results: (a) under-focused image (defocus value: $\Delta f = 336 \mu m$), (b) in-focused image ($\Delta f = 0 \mu m$), and (c) over-focused image ($\Delta f = -336 \mu m$). In the inset in Fig. 3(a) a Bragg diffraction pattern of an Fe₀.₈₈Ga₀.₁₂ thin film is shown. The contrast of the Bragg diffraction pattern is reversed for a better view.

The direction of the thin film was adjusted to the [001] orientation. In the Fresnel images in Figs. 3(a) and 3(c), magnetic domains with 180° and 90° domain walls are observed side by side. Two dark and curved lines in the central part of the specimen film are bend contour lines indicating that the specimen is a single crystal [see, Fig. 3(b)].

Figure 4 shows Foucault observation results: (a) Foucault image corresponding to spot (I) shown in the inset in Figs. 4(a), 4(b) Foucault image corresponding to spot (II), and (c) Foucault image corresponding to spot (III). To observe the Foucault images, an SA aperture of 20 μm in diameter was used as an angle-limiting tool corresponding to $2.6 \times 10^{-3}$ rad. The inset in Fig. 4(a) shows a small-angle electron diffraction (SmAED) pattern of the central region of an Fe₀.₈₈Ga₀.₁₂ thin film (5 μm in diameter) recorded with a camera length of 42 m, indicating three magnetization domains with 180° and about 90° domains. The contrast of the SmAED pattern is also reversed for a better view. Streaks among the spots indicate that the domain walls were of Bloch type. The Foucault images in Figs. 4(a)–4(c) clearly show magnetic domains.

The Fresnel images and Bragg diffraction pattern in Fig. 3 and the Foucault images and SmAED pattern in Fig. 4 are all the observation results that conventional Lorentz microscopy can produce.

Figure 5 shows the HCF images for different inclination angle conditions. The SA aperture with opening size 100 μm in diameter corresponding to $1.30 \times 10^{-3}$ rad was utilized as the angle-limiting tool. Figure 5(a) shows a hollow-cone image for 0 rad inclination angle; this image corresponds to the bright-field micrograph in Fig. 5(b). Figures 5(b) and 5(c) are bright-field...
Fig. 4. Conventional Lorentz microscopy observations in the Foucault mode: (a) Foucault image from the imaging spot (I) in the SmAED pattern shown in the inset, (b) Foucault image from spot (II), (c) Foucault image from spot (III). Each Foucault image was selected by the SA aperture of the opening size 20 μm in diameter corresponding to 2.6 × 10⁻⁴ rad. Inset in (a) shows an SmAED pattern recorded with the camera length of 42 m in reversed contrast.

Fig. 5. Hollow-cone Foucault images: (a) electron micrograph for 0 rad inclination angle corresponding to the conventional bright-field image, (b) bright-field HCF image for 1.6 × 10⁻⁴ rad inclination angle, (c) bright-field HCF image for 2.8 × 10⁻⁴ rad, (d) schlieren image for 3.6 × 10⁻² rad inclination angle, (e) dark-field HCF image for 4.4 × 10⁻⁴ rad inclination angle, (f) dark-field HCF image for 5.0 × 10⁻⁴ rad, and (g) conventional dark-field image for 7.2 × 10⁻⁴ rad taken by the whole azimuthal angle around the optical axis.

HCF images for 1.6 × 10⁻⁴ rad and 2.8 × 10⁻⁴ rad inclination angles, respectively, where domain walls are observed as white lines and several domains have slight dark contrast. The contrast of domains and domain walls disappears in Fig. 5(d) for 3.6 × 10⁻⁴ rad inclination angle. This corresponds to a schlieren image, to be discussed later. Figures 5(e) and 5(f) are dark-field HCF images for 4.4 × 10⁻⁴ rad and 5.0 × 10⁻⁴ rad inclination angles, respectively. In these figures, domain walls are observed with black lines and several domains also have slight bright contrast. The contrast in the dark-field HCF images in Figs. 5(e) and 5(f) are reversed with respect to the bright-field HCF images in Figs. 5(b) and 5(c), respectively. Figure 5(g) shows a hollow-cone image for 7.2 × 10⁻⁴ rad, a dark-field micrograph taken by the whole azimuthal angle around the optical axis. The contrast of the magnetization disappeared because deflected beams by the magnetization distribution inside the Fe₉₀Ga₁₀₁₂ material were completely filtered out by the SA aperture. We note that in Fig. 5 both magnetic domains and magnetic domain walls are simultaneously observed under the in-focus condition: Figs. 5(b) and 5(c) (bright-field HCF) and Figs. 5(e) and 5(f) (dark-field HCF). On the other hand, in Fig. 3 (Fresnel method) only magnetic domain walls were observed and in Fig. 4 (Foucault method) only magnetic domains were observed.

A special condition for schlieren imaging corresponds to the specific inclination angle of the illumination beam changed from the bright-field mode to dark-field mode. The schlieren imaging method is known as a high-speed imaging method applicable to low refractive index media, such as air, and is also known as old-fashioned phase-contrast optical microscopy. It is one of the asymmetric imaging methods using only a half of the diffraction pattern controlled by an angle-limiting aperture. Figure 5(d) corresponds to the image by the schlieren optical system. Figure 6 shows its wider view results: Fig. 6(a) shows imaging of the inside of specimen and Fig. 6(b) shows imaging in the outside space around the specimen. The brightness fluctuation around the specimen qualitatively shows that magnetic field leaked out and spread out from the specimen over a wide range and to a far distance. This is the reason that in electron holography reference waves must be placed far away from specimens for electromagnetic observation.

In conclusion, we developed a HCF imaging method with which simultaneously observation of magnetic domains and magnetic domain walls was realized under the in-focus condition. By adjusting inclination angles, observations in the bright-field mode and in the dark-field mode became possible. When the mode changed from the bright-field mode to the dark-field mode, we obtained schlieren images with which electromagnetic fields in the space around the specimen were qualitatively visualized.

The developed HCF imaging method has the advantages of both the Fresnel and Foucault imaging methods, therefore the HCF method may be called the third Lorentz microscopy. We hope the HCF method will be widely used for analyzing electromagnetic properties in materials in the future.
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