Lorentz Microscopy of Magnetic Granular Films

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Lorentz microscopy is successfully applied to observe the magnetic structures of iron particles embedded in thin magnetic granular films: Fe-Mo and Fe-MgO films. Images of experimentally observed Fe particles, 25 and 60 nm in diameter, are compared with simulated ones. Single-magnetic-domain particles could be observed. Cylindrical Fe particles 2–8 nm in diameter and 10–20 nm thick in an Fe-MgO film showed clear black-and-white-pair contrast in a region about 30 nm in diameter, which seems to be the range of the area having magnetic correlation.

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Magnetic granular films, which consist of magnetic fine particles of nanometer size and an immiscible and nonmagnetic matrix, have been of great interest for new magnetic substances. They exhibit unique characteristics, such as giant magnetic resistance [1,2] and soft magnetic properties [3]. Their magnetic characteristics strongly depend on the fine structures, besides the combination of magnetic and nonmagnetic constituents, especially on their surface topology and the particle size. Although the relationship between these fine topological structures and the macromagnetic characteristics has been investigated systematically by many researchers [4], there has remained a desire to observe magnetic microstructures directly.

We have succeeded in observing the magnetic structure of fine particles in granular films by using Lorentz microscopy. There are two usual modes in which Lorentz microscopy is used: Fresnel (out-of-focus) mode and Foucault (displaced aperture) mode [5]. Foucault imaging has the advantage of taking images in focus, so higher resolution can be obtained. On the other hand, Fresnel imaging is suitable for observing a specimen whose magnetization is in random directions and for studying phenomena dynamically. Although the spatial resolution of Fresnel mode is poorer than that of Foucault mode, the resolution will be improved by using a highly coherent electron source, i.e., a field emission gun. In this study, we used Fresnel mode.

Two kinds of specimens were observed. One was an Fe-Mo granular film. An amorphous film of Fe-Mo was prepared by dc magnetron sputtering on an amorphous carbon film mounted on a Cu grid for electron microscopy at a temperature below 180 K. The sputtering power was 45–60 W and the duration was 3–5 min. The background pressure was 8×10^{-5} Pa and Ar pressure was 0.8 Pa. The as-grown Fe(80 at.%)-Mo(20 at.%) film had a weak saturation magnetization, as shown in Fig. 1(a). Films were annealed for 30 min at 770 K in an in-plane magnetic field in order to precipitate fine particles. From the phase diagram of binary alloys, we expected that the precipitation of Fe particles, including 2% or 3% of Mo, in $Fe_{60}Mo_{40}$ (ε phase) which has no magnetism. We confirmed the existence of precipitated Fe particles, which may have included a few percent of Mo, using an energy dispersive x-ray analyzer attached to a 200-kV transmission electron microscope (TEM). Figure 1(b), which

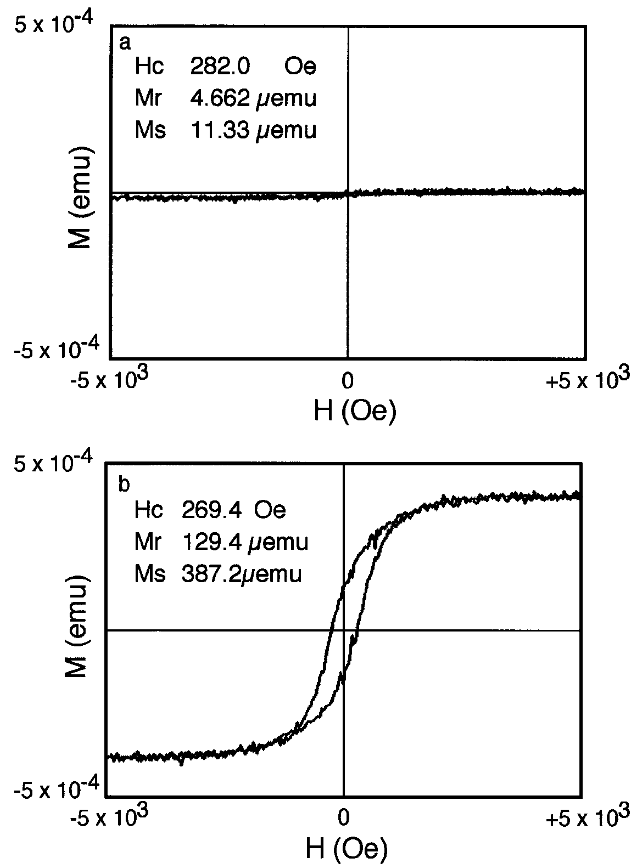

FIG. 1. Magnetization curves of Fe-Mo films: (a) as-grown and (b) annealed for 30 min at 770 K in an in-plane magnetic field.

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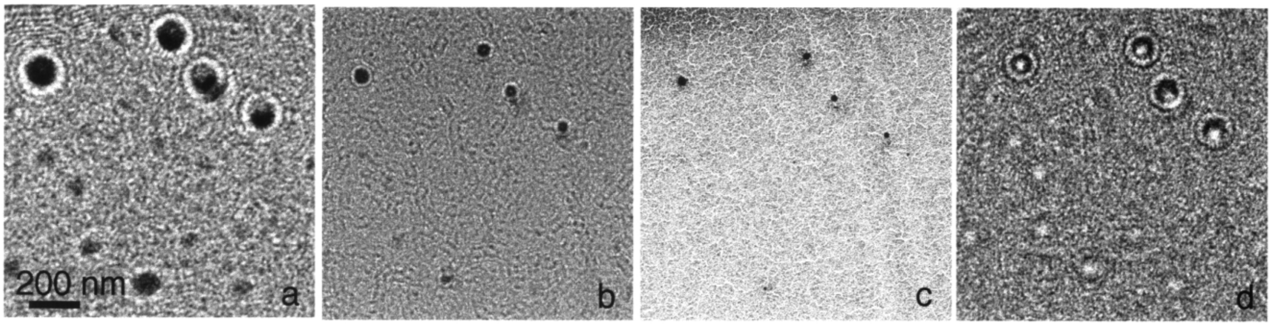

FIG. 2. Through-focus images of the annealed Fe-Mo film taken with *df* values of (a) −1.2 mm (underfocused), (b) −0.1 mm, (c) 0 mm (slightly underfocused), and (d) +1.2 mm (overfocused).

shows the magnetization of the annealed film, also indicates that ferromagnetic particles were precipitated.

The second kind of specimen was an Fe-MgO composite film, where iron particles were embedded in single-crystal magnesium oxide. The film was coevaporated on an NaCl substrate at 620 K using electron beam guns in a vacuum of 1×10^{-5} Pa. It has been reported [6] that an MgO film grows in [001] orientation and that Fe particles, which have a bcc structure, are epitaxially embedded as (001) [110]Fe‖(001) [100]MgO or $(01\bar{1})$ [100]Fe‖(001) [100]MgO. The observation of magnetic microstructures was achieved using a TEM equipped with a field emission gun and a low magnetic field objective lens [7]. The magnification was reduced to one-third of that provided by a standard objective lens, but the maximum value (×500 000) was enough for our Lorentz microscopy. The focusing step was determined by measuring the deviation of a (200) dark field image from a bright field image of MgO smoke, and it was estimated to be 42 times the displayed one.

Since Lorentz microscopy of ultrafine particles has never been reported before, we needed to confirm what contrast should be presented by single-magnetic-domain Fe particles. We simulated wave-optically the Lorentz image contrast using a model of a magnetic sphere, just as in electron holography [8]. In the Fresnel mode of Lorentz microscopy, the image of a thin magnetic film can be explained with only the deflection of electrons due to the Lorentz force, while the image of magnetic fine particles must be interpreted by considering an outline (or thickness) effect besides the magnetic effect. The Fresnel fringes due to their outlines show sometimes stronger contrast than the magnetic contrast. The simulation treated the magnetic induction inside the particle and

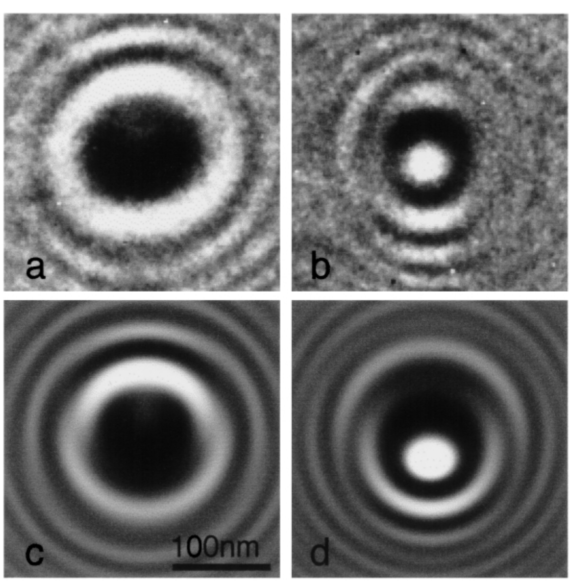

FIG. 3. Lorentz microscopy of an Fe particle 60 nm in diameter. Experimental images [(a), (b)] coincide with simulated ones [(c), (d)] at both foci $df = -1.6$ mm (underfocused) [(a), (c)] and +1.6 mm [(b), (d)].

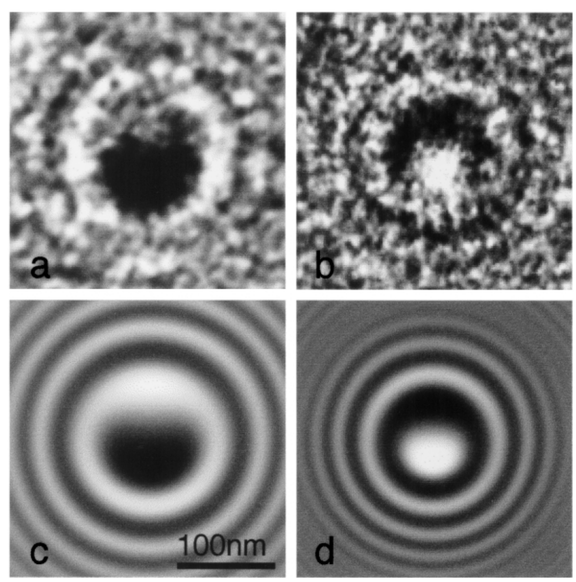

FIG. 4. Lorentz microscopy of an Fe particle 25 nm in diameter. Experimental images [(a), (b)] coincide with simulated ones [(c), (d)] at both foci $df = -1.2$ mm (underfocused) [(a), (c)] and +1.2 mm [(b), (d)].

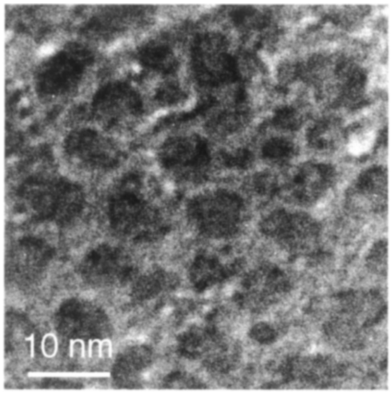

FIG. 5. TEM image of an Fe-MgO film taken in the slightly underfocused condition.

that leaking outside it, as well as an inner potential (i.e., an outline effect), but an absorption effect was ignored. Since actual particles are partly embedded in a film, in this study we took into consideration the effect of embedding by reducing the value of the inner potential of iron to $\frac{1}{2}-\frac{1}{10}$ of that for a bulk specimen. Details of the simulation will be reported elsewhere.

Figure 2 shows through-focus images of a 30 nm thick Fe-Mo film. Some of the precipitated Fe particles show the contrast of black-and-white pairs. It can clearly be seen that black-and-white pairs reverse their contrast between the underfocused image 2(a) and the overfocused image 2(d). The 0.1 mm underfocused image 2(b) and slightly underfocused image 2(c) do not show any magnetic contrast. These are typical characteristics of Lorentz microscopy. The contrast seen in the underfocused images is not simply the reverse of that in overfocused images. In Fig. 2(c), perfectly embedded precipitates, whose thickness may be less than their diameter, show weak contrast, which becomes stronger in Figs. 2(a) and 2(d). In the case of such small particles, however, the typical black-and-white pairs are not observed clearly.

Simulated images of an Fe particle in a vacuum with an inner potential reduced to 30% are shown in Figs. 3 and 4, for comparison with the experimental results. Lorentz images of a particle 60 nm in diameter show antisymmetric contrast inside strong Fresnel fringes in overfocused images [Figs. 3(a) and 3(c)], and in underfocused images the inside antisymmetry is weaker [Figs. 3(b) and 3(d)]. On the other hand, black-and-white pairs are very clear in both the overfocused and underfocused images of a particle 25 nm in diameter, as shown in Fig. 4.

Figure 5 shows a transmission microscrope image of an Fe-MgO film taken in a slightly underfocused condition. The particle diameters are judged to range from 2 to 8 nm. The thickness of the film was about 20 nm and that of the particles was estimated to range from 10 to 20 nm. A through-focus series of Lorentz micrographs is shown in Fig. 6. It is clear that black-and-white pairs, indicated by arrows, reverse their contrast between the underfocused and overfocused images. In the focused image the contrast disappears. This suggests that anisotropic regions of magnetization do exist somewhere. The areas 10–30 nm in diameter appear to be single-magnetic-domain particles, though the diameter of each particle is far less than that. The contrast simulation showed that such a high contrast black-and-white pair cannot be obtained from particles less than 10 nm in diameter. It seems that single-magnetic-domain iron particles in the 30 nm area all have magnetic correlation and the same direction of magnetization. The area where black-and-white contrasts align in one direction in the Lorentz micrograph seems to be where the magnetic induction flows linearly.

We have shown that the magnetization of fine particles in thin granular films can be observed by Fresnel-mode Lorentz microscopy. A simulation showed that the Lorentz contrast of a fairly large particle is difficult to be distinguished because of its strong Fresnel fringe contrast. Particles around 25 nm in size present clear black-and-white pair contrast. The Lorentz microscopy of an Fe-MgO granular film shows black-and-white pairs 20–30 nm in diameter, which seems to be the area where the magnetizations of particles 2–8 nm in diameter have correlation.

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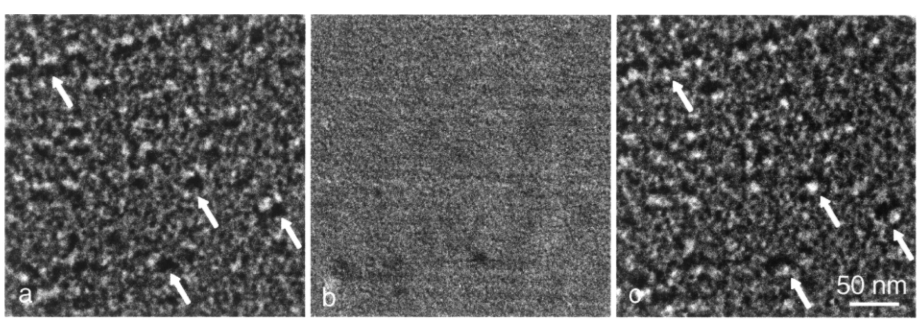

FIG. 6. Lorentz images of an Fe-MgO film taken at *df* values of (a) −0.5 mm (underfocused), (b) 0 mm (in focus), and (c) +0.5 mm.

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