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Dark Field Observation of Fe_3O_4 - $\gamma\text{Fe}_2\text{O}_3$ Multiphase Films

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Abstract. We investigated the phase separation of cubic and tetragonal systems in Fe_3O_4 - $\gamma\text{Fe}_2\text{O}_3$ intermediate thin films. An Fe thin film of 50 nm was deposited on a SiO_2 substrate using an electron beam evaporator at room temperature. The film was oxidized to $\alpha\text{Fe}_2\text{O}_3$ at 400°C in air, then the film was reduced to Fe_3O_4 at 320°C in methanol vapor. The gradual oxidation of the Fe_3O_4 film to $\gamma\text{Fe}_2\text{O}_3$ film at 240°C was terminated when the coercive force of the film increased more than 1000 Oe at an intermediate state. The intermediate film was examined by dark-field TEM observation with tetragonal diffraction spots. The result revealed that crystallites of about 10 nm of tetragonal structure are distributed in a matrix of cubic phase.

1. Introduction

The phase transition from cubic to tetragonal systems in the Fe_3O_4 - $\gamma\text{Fe}_2\text{O}_3$ intermediate thin films is particularly interesting because it is probably connected with the anomalous increase in coercive force observed in thin film and powder samples. The gradual oxidation from Fe_3O_4 to $\gamma\text{Fe}_2\text{O}_3$ in low temperatures near 200°C bring about a sharp increase in coercive force [1].

The origin of the increase in coercive force was discussed first in connection with the phase separation by Kojima and Haneda [2]. Evidence for the pinning of magnetic domain walls was searched in the films using electron microscope technologies [3]. In this paper, we examine the intermediate thin film showing a high coercive force in dark field images which are composed of the tetragonal diffraction spots.

2. Experimental

Pure iron of 99.99% was deposited onto vitreous SiO_2 substrates at room temperature using an electron beam evaporator at a residual pressure of 4×10^{-4} Pa. The thickness of the films was 50 nm and the deposition rate was 1 nm s^{-1} . The film was oxidized to $\alpha\text{Fe}_2\text{O}_3$ at 400°C for 1h in air. Subsequently the film was reduced to Fe_3O_4 in methanol vapor at 320°C.

The X-ray diffraction analysis of the reduced film indicates that no $\alpha\text{Fe}_2\text{O}_3$ or Fe phase was contained in the film. No preferential orientation was observed in the diffraction peaks. The Fe_3O_4 film was gradually oxidized in air by heating at 240°C. The coercive force of the film was measured during the gradual oxidation. The coercive force of the intermediate film increased as the oxidation proceeded from 400 Oe to 1200 Oe in the first 20 minutes and decreased to 300 Oe after 180 minutes oxidation. The color of the film changed from dark brown to light amber during the oxidation. The sample film was removed from the substrate when the coercive force increased more than 1000 Oe. All the processes of sample preparation for the TEM

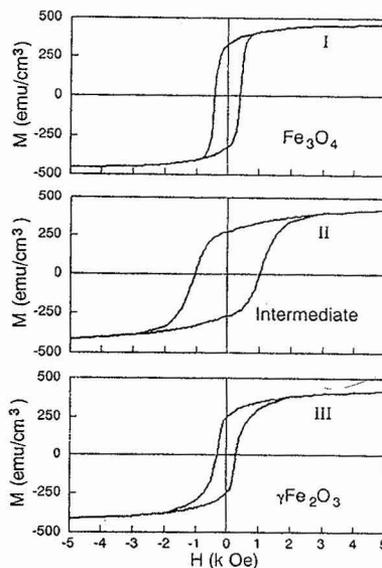


Fig.1 Hysteresis loops of Fe_3O_4 (I), intermediate film annealed for 20 min (II), and $\gamma\text{Fe}_2\text{O}_3$ (III).

observation were continued at room temperature in order to maintain the oxidation state. A high resolution electron microscope (JEM-2010HC) was used for the dark field observation.

3. Results and Discussion

Figure 1 shows the hysteresis loops of the Fe_3O_4 , intermediate and $\gamma\text{Fe}_2\text{O}_3$ films demonstrating the increase in coercive force. It is noted that the shape of loop is different between Fe_3O_4 and $\gamma\text{Fe}_2\text{O}_3$ films. Figure 2 shows the transmission electron diffraction patterns of the Fe_3O_4 (a) and $\gamma\text{Fe}_2\text{O}_3$ (b) films. The arrows "C, T" in Fig.2(b) indicate (220) and (111) diffractions of the cubic lattice. These diffractions are common to the cubic and tetragonal lattices. The arrow "T" indicates (213) and/or (116) diffractions which are peculiar to the tetragonal lattice of $\gamma\text{Fe}_2\text{O}_3$.

The dark field image of Fig. 3 is constructed by the diffraction spots on the ring "T". Figure 3 also shows a bright field image of the same area of the film. The arrows "T" in Fig. 3 indicate a crystallite of tetragonal structure which is surrounded with cubic matrix. The small size of the tetragonal crystallite suggests a pinning of magnetic domain walls which move through the intermediate film.

References

- [1] M.Namikawa, M.Satou, Y.Imaoka and S.Tochihara, *Denki Kagaku*, **31**, 37 (1963).
- [2] H.Kojima and K.Haneda, *IEEE Trans. Magn.*, **MAG-16**, 11(1980).
- [2] Y.Yamazaki, K.Okuda, N.Komachi, M.Sato and T.Namikawa, *Proc. 6th Int. Conf. on Ferrites*, 401(1992).

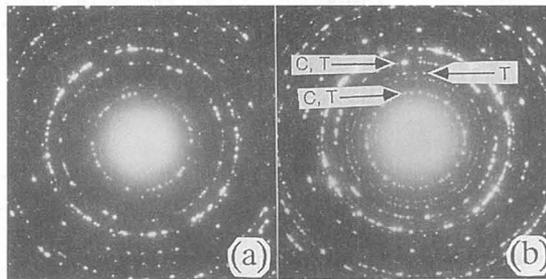


Fig. 2 THEED patterns of Fe_3O_4 (a) and $\gamma\text{Fe}_2\text{O}_3$ (b) films.

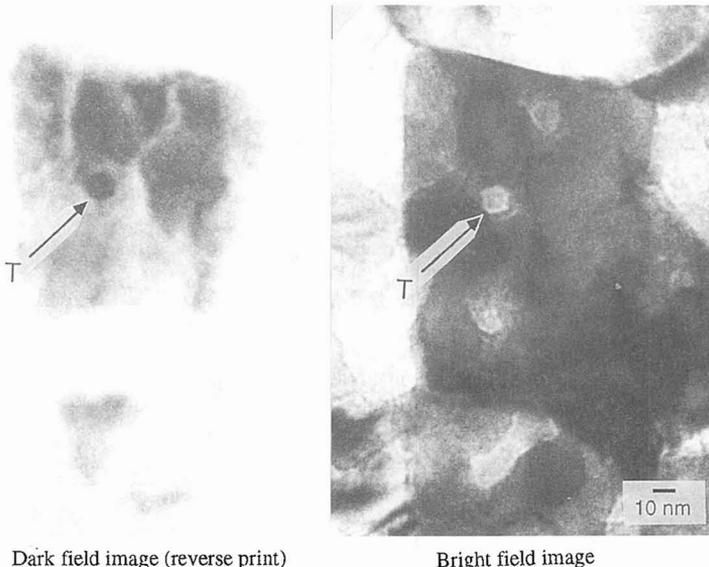


Fig. 3 Dark field and bright field images of an Fe_3O_4 - $\gamma\text{Fe}_2\text{O}_3$ intermediate film, annealed for 20 min, showing a high coercive force of 1010 Oe, HV=200kV.