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Grain-size dependence of coercive force in sputtered and annealed iron films

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Magnetic properties such as coercive force, initial susceptibility, etc., are structure sensitive parameters. Especially, the coercive force depends on the existence of crystal defects such as dislocations and grain boundaries. In this study, the grain-size dependence of coercive force was investigated in an iron film, 150 nm in thickness, deposited by sputtering method on a (001) KCl substrate at room temperature. The grain size of the film was changed from 15 to 120 nm by annealing in a vacuum. The coercive force increased with the increase of grain size and was proportional to the square of grain size. The magnetic domain had a ripple structure when the grain size was smaller than 50 nm. A grain size larger than 50 nm led to an irregular domain wall configuration and the formation of domain walls in the grain.

1. Introduction

The magnetic properties of materials are strongly dependent on the existence of dislocations, grain boundaries and so-called 'structure-sensitive properties' [1, 2]. Of many magnetic parameters having structure sensitivity, coercive force, Hc, is one of the most useful parameters and can be applied in non-destructive testing [3]. The interaction of the dislocation introduced by plastic deformation with coercive force has been well documented by Kronmuller in Ref. [3]. Since grain boundaries act as impediments to domain wall motion, coercive force increases with decreasing grain size [4]. On the other hand, when grain size is smaller than 100 nm, Hc decreases rapidly with decreasing grain size. When the grain size is comparable to the effective domain wall width, δ, the magnetization may not follow the randomly oriented easy axis of each individual grain, and a common alignment of the magnetization in correlated grains may occur. The magnetocrystalline anisotropy constant may be averaged over several grains with the consequence that Hc decreases with decreasing grain size. This model is known as the random anisotropy model (RAM), which was proposed by Herzer [5]. The effective anisotropy $\langle K \rangle$ relevant to the magnetization process results from the mean fluctuation amplitude of the anisotropy energy within the volume of the ferromagnetic exchange length L_{ex}, and is given by $\langle K \rangle = K_1/\sqrt{N}$, where K_1 and N are the intrinsic magnetocrystalline anisotropy constant and the number of magnetically correlated grains. N is related to the grain size D and L_{ex} by $N = (L_{ex}/D)^3$ in three-dimensional structures. L_{ex} is related to $\langle K \rangle$ by $L_{ex} = (A/\langle K \rangle)^{1/2}$, where A is the exchange stiffness constant. As a result $\langle K \rangle$ for $D < L_{ex}$ is given by $\langle K \rangle \approx K_1^4 \cdot D^6/A^3$. Since the coercive force is closely correlated with the effective anisotropy $\langle K \rangle$ by $H_C = p\langle K \rangle/J_S$, where p and Js are the dimensionless pre-factor (1-2) and the saturation magnetization, respectively, the coercive force, H_c, is proportional to D^6 for bulk materials. The D^6 dependence of the coercive force has been experimentally detected in iron base soft magnetic materials, though the nanocrystalline sample consisting of two phases had been created by crystallization from the amorphous phase [6]. Experimental results of nanocrystalline nickel prepared with a gas-deposition method also show the D^6 dependence of coercive force [7, 8].

The coercive force is related to the motion of the magnetic domain wall. Since Lorentz microscopy is a useful tool for observing the magnetic domain structure, observation of the domain structure was performed mainly on iron base materials to clarify the relation between the microstructure and the domain structure [9, 10]. However, when samples of the iron base materials consist of two phases, i.e., ferromagnetic nanocrystalline alloys in an amorphous matrix, observation yields little information on the direct interaction between grain boundaries and the domain wall configuration. The aim of the present experiment was to measure the grain size dependence of the coercive field and to observe the magnetic domain structure in post-annealed iron film initially grown on (001) KCl at 300 K by the sputtering method. The samples for measurements of magnetization and grain size, and for observation of magnetic domain structure were prepared by cutting a large piece of as-sputtered Fe film on (001) KCl.

2. Experimental procedure

A polycrystalline iron film, 150 nm thick, was prepared by sputtering 99.9% pure iron onto the (001) plane of an air-cleaved KCl substrate. An Fe target in the form of a disc with a diameter of 50 mm and a thickness of 5 mm was used for deposition. Sputtering onto the

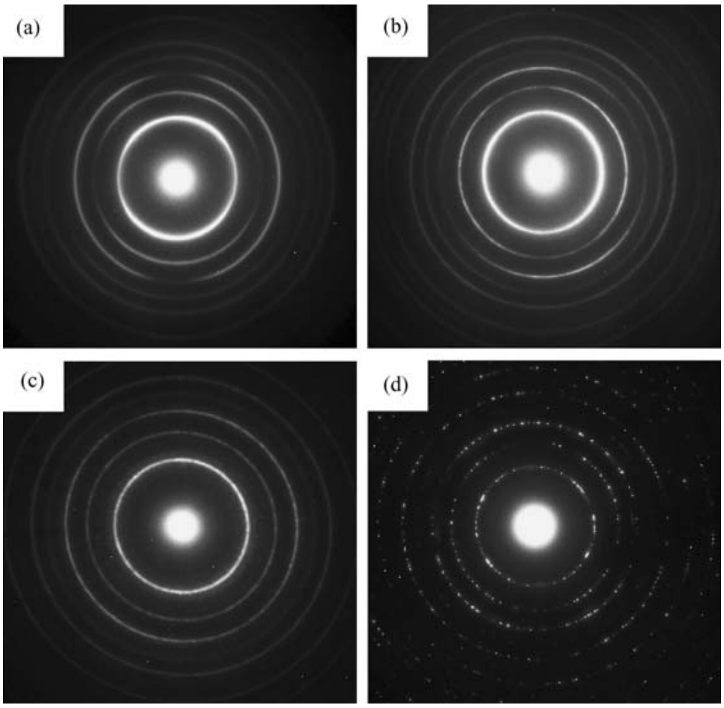

Figure 1 Electron diffraction patterns of as-deposited and post-annealed Fe films on (001) KCl substrates: (a) As-sputtered, (b) annealed at 420 K, (c) 620 K, and (d) 870 K.

substrate was carried out in argon at a rate of 0.4 nm/s at a substrate temperature of 300 K. The as-sputtered iron film on KCl was annealed at temperatures from 420 to 970 K in a vacuum using an infrared image furnace. Magnetization measurements were performed in a vibrating sample magnetometer (VSM) at a temperature of 293 K. Film with a KCl substrate was measured with the applied magnetic field parallel to the sample. After the KCl had been dissolved in water, the iron film was mounted on copper grids. Lorentz transmission electron microscopy was employed for examining as-sputtered and post-annealed samples. Electron microscopy was performed with a Philips Tecnai 30.

3. Experimental results and discussion

Fig. 1 shows electron diffraction patterns of as-deposited and post-annealed Fe film on a (001) KCl substrate. Even though deposition and annealing was conducted on (001) KCl, the diffraction patterns of all samples examined in the present experiment indicated a ring pattern, which can be indexed as bcc iron. The ring pattern showed a weak preferred orientation in an as-deposited sample (Fig. 1a). The preferred orientation seen in the as-deposited sample seemed to decrease after annealing. When the annealing temperature increased, reflection spots were identified as the result of grain growth, though they still formed the ring pattern. This indicates that the substrate orientation of (001) KCl did not affect the orientation of grains during grain growth by annealing.

Fig. 2 shows bright field images of annealed iron samples. The grain size increases with increasing annealing temperature. The image at the annealing temperature of 670 K, showing superimposition of grains with other grains, suggests that grains grew on the other grains. This means that 3–6 grains were included in a

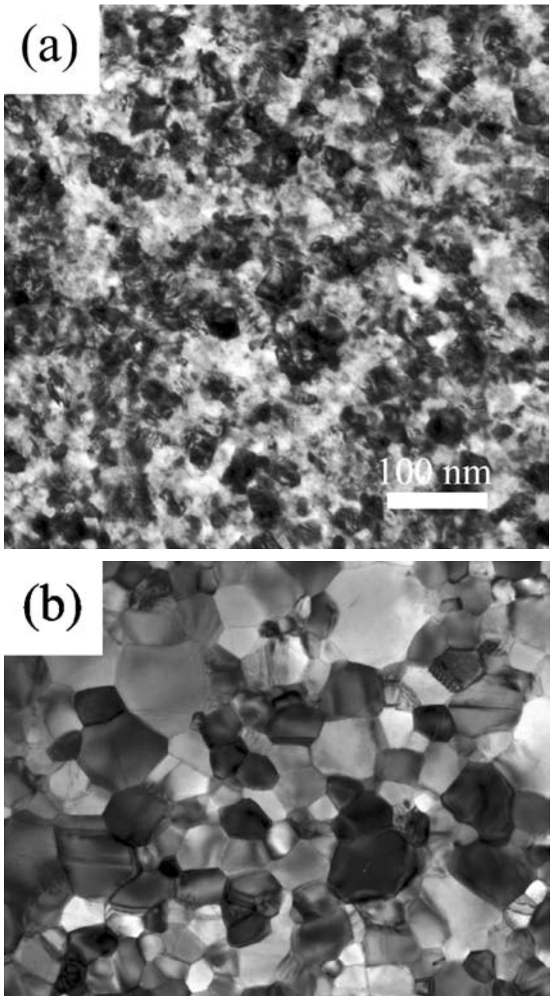

Figure 2 Bright field images of annealed iron samples: (a) Annealed at 670 K and (b) at 870 K.

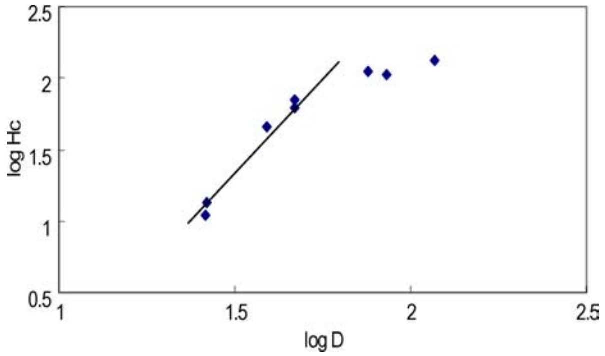

Figure 3 Average grain size dependence of coercive force of annealed Fe films.

150 nm-thick film in the direction of growth at annealing temperatures below 670 K, though the number of grains in the direction of growth depends on the grain size. On the other hand, a bright field image of the samples annealed above 770 K revealed a column of a single grain without any superimposition of grains in the direction of growth. The average grain size of as-sputtered Fe film was about 15 nm. The annealing resulted in grain growth, and according to the annealing temperature the average grain size varied from 25 to 120 nm in diameter. It should be pointed out that the distribution of grain size in a film annealed at 770 K was not homogeneous, but dispersed widely as shown in Fig. 2.

The coercive force of the post-annealed Fe film obtained from the magnetic field dependence of magnetization is shown as a function of average grain size in Fig. 3. It ranged from 10 to 130 Oe. The present experiment shows that the coercive force is proportional to $D^{2.5}$. From our data, the ranges of grain size and coercive force that could be covered were too small to explicitly verify their dependence. If we assume a two-dimensional structure, a D^2-variation of $\langle K \rangle$ is obtained from the random anisotropy model as the result of $N = (L_{ex}/D)^2$, though assumption of a two-dimensional structure was not completely satisfied in the films having several grains in the direction of growth in the case of the grain size smaller than 44 nm. For a grain size larger than 40–50 nm, the D^2-variation of the coercive force was not satisfied and the coercive force was almost constant for grain size larger than 70 nm in the present experiment. Two points are suggested by the grain size dependence of the coercive force shown in Fig. 3. One is that the grain size around 40–50 nm may be the critical size necessary to change the mechanism controlling the coercivity. We can calculate the effective domain wall width, δ, from the equation $\delta = \pi L_{ex}$ and obtain an effective wall width of 54 nm. This is a reasonable value for Fe [8]. Since the grain size is the same value as the effective domain wall width, the coercive force decreases with increasing grain size as a function of D^{-1} variation. The second point is that the small change of the coercive force with grain size larger than 70 nm may be due to the wide distribution of grain size. Since the coercive force is determined by the magnetic field necessary to move the domain walls from the strongest pinning site, the grain effectively pinning the domain may exist in the widely distributed grains, i.e., the grains with 50 nm in diameter exist in an annealed film even when the average grain size is larger than 70 nm. It should be noted that RAM is a simple model which only considers the exchange interaction and the magneto-crystalline anisotropy. Though the D^3-variation of the coercive force has been proposed for the case of two-phase nanocrystalline iron base materials, it is applicable only when the ferromagnetic grains are separated by the amorphous non-magnetic materials [11].

Fig. 4 shows low magnification images of the magnetic domain structure of the annealed Fe film at 420 K (a) and 770 K (b) using Lorentz microscopy. When

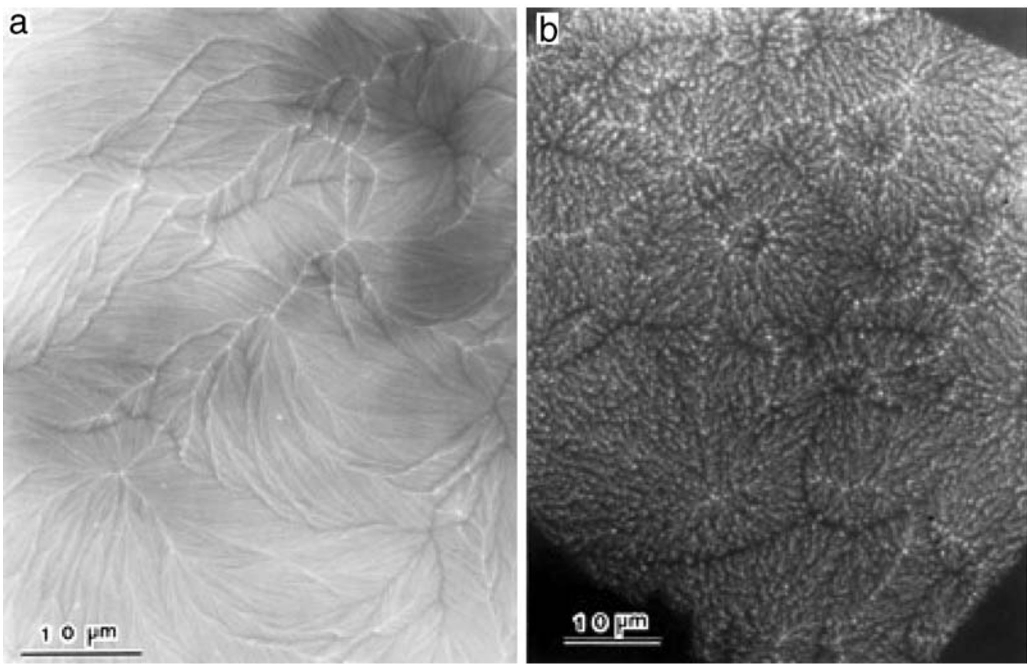

Figure 4 Low magnification images of magnetic domain structure of Fe films annealed at 420 K (a) and 770 K (b) using Lorentz microscopy.

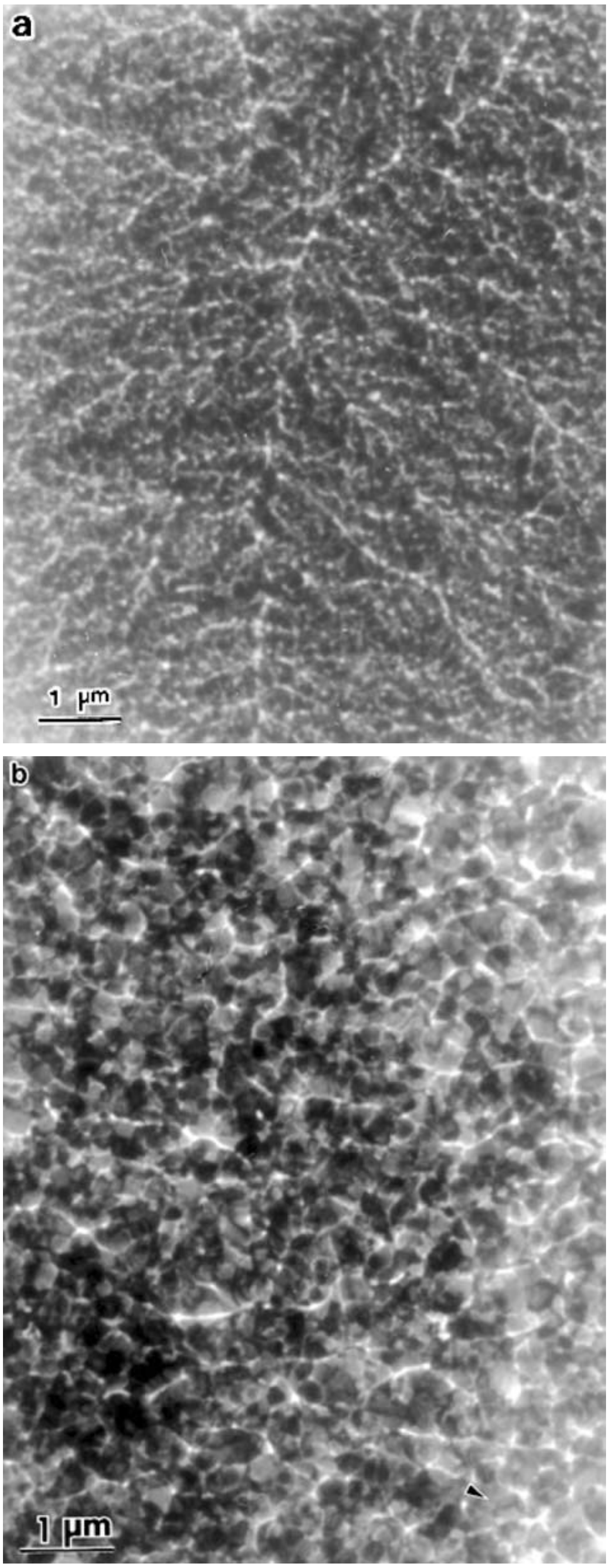

Figure 5 Enlarged image of the magnetic domain wall observed in annealed Fe films; (a) 420 K and (b) 870 K.

an Fe film showed an average grain size of 26 nm as a result of annealing at 420 K, the magnetic domain structure was the ripple structure [12]. The ripple structure which appeared in small grained deposited film is caused by small periodic variations in the direction of magnetization associated with a corresponding variation of the anisotropy [13]. This type of ripple structure was observed in the annealed Fe film below 620 K. On the other hand, macroscopically, the domain structure of annealed film at 770 K seems to be same as that of 420 K annealed film, but the shape of the ripples was seen as a dotted line rather than smooth lines observed in the annealed film at 420 K. An enlarged image of the magnetic domain wall observed in annealed Fe film is shown in Fig. 5. When a Fe film was annealed at 720 K, the ripple structure was retained, though the ripples were of irregular shape with wider spacing between them in comparison with those observed in an annealed film at 420 K. Fig. 5b shows the domain structure of an Fe film having an average grain size of 120 nm. The sample was divided into small domains about 1 μm in size and did manifest the ripple structure. The domain walls were both in grains and grain boundaries. Some grains had the domain structure in the grain as indicated by an arrow in Fig. 5b, because the domain structure of this sample having a grain size of 120 nm is determined by the magnetocrystalline anisotropy.

4. Conclusion

The grain size dependence of coercive force and magnetic domain structure of annealed Fe film with a thickness of 150 nm was investigated.

1. The coercive field was proportional to $D^{2.5}$ when the grain size was smaller than 50 nm. This can be explained based on the random anisotropy model for a two-dimensional structure.
2. The domain had a ripple structure at grain sizes smaller than 50 nm. The ripple structure gradually become irregular and then the domain wall was within the grain when the grain size was larger than 50 nm.

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