RAPID PUBLICATION

Soft Magnetic Properties of Nonequilibrium bcc Fe-Hf Alloys Produced by rf Sputtering

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Microstructure and magnetic properties of Fe-rich Fe-Hf alloy films produced by rf sputtering have been investigated as a function of Hf content. The films in the concentration range 1–6 at%Hf have a nonequilibrium bcc phase with fine grains of 5 to 20 nm and exhibit high saturation magnetization and soft magnetic properties. In particular, the bcc Fe_{95.6}Hf_{4.4} film annealed at 673 K for 3.6 ks exhibits a saturation magnetization of 1.9 T and an effective permeability of 1800 at 1 MHz.

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I. Introduction

A high-density magnetic recording media has required magnetic heads with high saturation magnetization⁽¹⁾. Since a high saturation magnetization up to 2 T at room temperature can be obtained only for Fe metal and Fe-rich alloys, the Fe-based films with either a single phase or a multilayered structure have intensively been investigated^{(2)–(11)}. In general, soft magnetic properties for polycrystalline materials are strongly dependent on grain size and crystallographic orientation. It has been reported in bcc Fe metal and Fe-rich alloy films that soft magnetic properties are improved by the refinement of grain size and the control of preferred orientation^{(12)–(14)}. Furthermore, the morphology of the sputter-deposited films is also known to influence soft magnetic properties, i.e., the coercivity of the films decreases when the formation of a columnar structure is suppressed^{(15)–(17)}. Recently, the present authors have found that nonequilibrium bcc Fe-TM (TM=Zr, Hf, Nb or Ta) alloy films produced by vapor quenching are good candidates for high-density recording head materials⁽¹⁸⁾. This paper is intended to clarify further detailed dependences of concentration and annealing temperature on soft magnetic properties and microstructure of Fe-Hf alloy films produced by rf sputtering.

II. Experimental Procedures

Fe-Hf alloy targets were prepared from 99.9 mass%

Fe and 95.0 mass% Hf by arc-melting in an argon atmosphere. The commercial grade Hf used in the present study contained 3 mass% of Zr. The alloy films were deposited on an indirectly water-cooled glass or a polyimide substrate by a conventional rf sputtering method. The films deposited on the polyimide substrate were used only for the saturation magnetization measurements. The back-pressure was about 1×10^{-4} Pa and the Ar gas pressure during rf sputter-deposition (RFS) was 0.8 Pa. The input power was 80 W. The composition was determined mainly by inductively coupled plasma spectroscopy and partly by electron probe microanalysis, indicating good agreement within ± 0.2 at%.

X-ray diffraction patterns were obtained using Cu $K\alpha$ radiation with a graphite monochromator. The 2θ angle was calibrated with Si powder. Microstructure of the films was observed by transmission electron microscopy (TEM) and scanning electron microscopy (SEM). The saturation magnetization at 290 K was measured using a vibrating sample magnetometer in an applied field below 0.8 MA/m (10 kOe). The coercivity at 290 K was measured using a dc hysteresis loop tracer at a maximum magnetic field of 8 kA/m (100 Oe). The relative permeability was measured at 1 MHz using an HP4191 impedance analyzer. The magnetostriction at 290 K was measured in a field up to 8 kA/m (100 Oe) by an optical cantilever method.

III. Results and Discussion

Figure 1 shows the X-ray diffraction patterns of the RFS Fe_{100-x}Hf_x ($x=0$ to 8.5 at%) alloy films. Only bcc peaks are observed in the composition range below 6 at% Hf and a further increase in the Hf content results in an appearance of asymmetric peaks due to the precipitation

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Fig. 1 X-ray diffraction patterns of sputter-deposited $\text{Fe}_{100-x}\text{Hf}_x$ ($x=0, 2.5, 4.4$ and 8.5 at%) films.

of the other phase. Figure 2(a) shows the lattice parameter of the RFS $\text{Fe}_{100-x}\text{Hf}_x$ alloy films. The lattice parameter increases monotonously with increasing Hf content because of a larger atomic radius of Hf. The concentration dependence of the half-width of the bcc (110) peak, $2\theta_{50}$ for the RFS $\text{Fe}_{100-x}\text{Hf}_x$ alloy films is shown in Fig. 2(b). With an increase of Hf content, the $2\theta_{50}$ slightly increases probably because of the decrease of the grain size.

Figure 3 shows the bright-field electron micrographs of the RFS bcc Fe and Fe–Hf films. The grain size of the RFS $\text{Fe}_{95.6}\text{Hf}_{4.4}$ film is smaller than that of the RFS Fe film, which is consistent with the results of the X-ray diffractometry. It is confirmed by SEM that the RFS bcc Fe and Fe–Hf films have a fine equiaxed microstructure which is known^{(15)–(17)} to be favorable for the appearance of good soft magnetic properties.

Figure 4 shows the concentration dependence of the saturation magnetization, σ_s , and coercivity, H_c , for the $\text{Fe}_{100-x}\text{Hf}_x$ films. Although the σ_s decreases monotonously with increasing Hf content, the H_c decrease drastically from 1800 to 200 A/m at 1 at%Hf and remains constant in the range up to 7 at%Hf. With a further increase of Hf content, the H_c increases because of the coexistence of bcc and the other phase.

The annealing temperature dependence of the reduced coercivity represented by the ratio of H_c (annealed)/ H_c (as deposited) is shown in Fig. 5. The addition of about 1 to 5 at%Hf brings about a further decrease in H_c by

Fig. 2 Lattice parameter (a) and half-width of the (110) peaks (b) of the bcc $\text{Fe}_{100-x}\text{Hf}_x$ films produced by sputter deposition.

Fig. 3 Bright-field electron micrographs of sputter-deposited Fe (a) and $\text{Fe}_{95.6}\text{Hf}_{4.4}$ (b) films.

Fig. 4 Changes of the magnetization (σ_s) and coercivity (H_c) at 290 K as a function of Hf content for sputter-deposited $\text{Fe}_{100-x}\text{Hf}_x$ films.

Fig. 5 Changes in the reduced coercivity as a function of annealing temperature for sputter-deposited $\text{Fe}_{100-x}\text{Hf}_x$ films.

annealing at temperatures ranging from 523 to 673 K, though the H_c increases rapidly at annealing temperatures above 723 K due to the precipitation of the other phase. The decrease in H_c by annealing is presumably due

Fig. 6 Changes in the coercivity (H_c) and the effective permeability (μ) at 1 MHz along the hard-axis for sputter-deposited $\text{Fe}_{100-x}\text{Hf}_x$ films as a function of Hf content. The open and closed marks represent the specimens annealed at 573 and 673 K, respectively.

Fig. 7 Change in the magnetostriction at $8 \times 10^4 \text{ A/m}$ as a function of Hf content for sputter-deposited Fe-Hf films annealed at 673 K for 3.6 ks.

to the release of the residual stress in the films.

The coercivity and relative permeability at 1 MHz for the RFS $\text{Fe}_{100-x}\text{Hf}_x$ films annealed for 1 h at an optimum temperature are shown in Fig. 6. The annealing temperature was 573 K for the 1.3 and 2.8 at%Hf samples and 673 K for the 0, 4.4, 5.6 and 8.5 at%Hf samples. The rather high permeability of more than 1000 at 1 MHz is

realized for the films in the bcc single phase region.

Figure 7 shows the concentration dependence of the magnetostriction under an applied field of 8×10^4 A/m for the RFS $\text{Fe}_{100-x}\text{Hf}_x$ films annealed at 673 K. The curve becomes zero at about 4 at%Hf, and hence the maximization of the permeability at 1 MHz for the 4.4 at%Hf films is presumed to originate from the nearly zero magnetostriction. Thus, the bcc $\text{Fe}_{96}\text{Hf}_4$ film produced by rf sputtering seems to be appropriate for high-density recording head applications.

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