Microstructure and magnetic properties of FeCoN thin films

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Effects of nitrogen contents and substrate temperatures to the microstructure and magnetic properties of the FeCoN films have been investigated. According to the TEM and x-ray Scherrer's equation analyses, we found that the grain size of films with substrate temperature below 200 °C is roughly about 13 nm, however, it increases very fast for films with substrate temperature above 300 °C. N content in the films is saturated to 30 at. %, as N_2 flow ratio $N_2/(Ar+N_2)$ is higher than 5 vol. %. From the magnetization studies, we have found that the saturation magnetization $4\pi M_s$ of the optimum samples (with the substrate temperature near 200 °C) is 23.9 kG. The improvement of the magnetic properties is attributed to the combination of α-Fe with N to form the high magnetic moment FeN phases. [S0021-8979(98)35011-2]

I. INTRODUCTION

A soft magnetic film with high saturation magnetization $4\pi M_s$ is required for use in a high-density magnetic recording head. Recently, extensive efforts have been made to improve the soft magnetic properties of Fe-based films.[1–7] According to the Pauling–Slater curve, Fe–Co alloys have the highest magnetization of iron alloy. Takahashi *et al.*[8] prepared FeCo nitride film onto a polyimide substrate by ion-assisted normally vapor deposition method. They found that the film was columnar structure with large grains of about 500 Å and maximum $4\pi M_s$ occurred at the Co content of about 32 at. %. Their films have large perpendicular magnetic anisotropy and N content of the film is unknown. On the other hand, Liao[9] reported that electrodeposited $Fe_{90}Co_{10}$ films show a high $4\pi M_s$ value of 19 kG and good soft magnetic properties. Recently, Nakagawa *et al.*[10] examined the FeCoN and FeCoTaN films which prepared by facing targets sputtering with N_2+Kr mixture gas. They found that $4\pi M_s$ of the film was decreased with increasing N_2 partial pressure. Pure $Fe_{90}Co_{10}$ alloy film has an extremely large $4\pi M_s$ of about 24 kG. In this report, we investigate the effect of substrate temperature and N content on the magnetic properties and microstructures of sputtered FeCoN films.

II. EXPERIMENT

$(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films with $x=0-30$ were prepared on a precleaned glass substrate by reactive RF magnetron sputtering in N_2 and Ar mixed atmosphere. The flow ratio of nitrogen to argon and nitrogen, i.e., $N_2/(Ar+N_2)$, during sputtering was varied between 0 and 10 vol. %. The substrate temperature was varied between 25 and 400 °C. Composite target consisting of pure Fe disk overlaid with Co pieces was used in this experiment. The base pressure in the system with a turbo pump was 1×10^{-6} Torr, and after the high purity $Ar-N_2$ mixed gas was introduced, sputtering pressure was 1×10^{-3} Torr. Thickness of the films was about 200 nm.

Structure and grain size of the films were determined by using x-ray diffractometer (XRD). Thickness of the films was measured by a Sloan DEKTAK-3-0305ST α-step profilometer. Magnetic properties of the films were measured with vibrating sample magnetometer (VSM) at room temperature with maximum applied field of 12 kOe. Composition, N content, and homogeneity of the films were determined by depth profiling analysis of Auger electron spectroscopy (AES).

III. RESULTS AND DISCUSSION

Figure 1 shows the N contents in pure Fe, Co, and $Fe_{90}Co_{10}$ films as a function of N_2 flow ratio during sputtering, i.e., $(N_2/Ar+N_2)\times100\%$, with the substrate temperature T_s kept at 25 °C. It can be seen that N content in the films increases dramatically with increasing N_2 flow ratio when N_2 flow ratio is lower than 5 vol. %. This is because N atoms can easily occupy interstitial sites of crystal lattice during deposition as N_2 flow ratio is lower than 5 vol. %. N content in the film is saturated as N_2 flow ratio is higher than 5 vol. % for all films. This is due to that all interstitial sites of crystal lattices in the film are occupied. Saturated N content of Fe film is about 32 at. % which is equal to the N content of ζ-Fe_2N phase.[11] ζ-Fe_2N phase is nonmagnetic and has maximum equilibrium N content in Fe–N binary system.[12] The maximum N contents of $Fe_{90}Co_{10}$ and Co films are 30 and 23 at. %, respectively. Figure 2 presents a

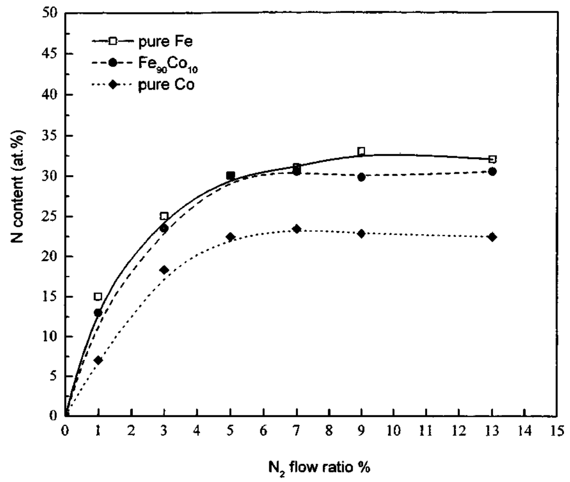

FIG. 1. Variation of N content with N_2 flow ratio of Fe, $Fe_{90}Co_{10}$ and Co films. The substrate temperature is kept at 25 °C.

typical depth profile of a FeCoN film analyzed by Auger electron spectroscopy (AES). We can see that the composition of this film is approximately $(Fe_{0.9}Co_{0.1})_{70}N_{30}$. It is calibrated by a standard of bulk $Fe_{50}Co_{50}$ alloy. Because the films were exposed to air prior to AES analysis, near the surface of the films, O and C were always observed as shown in Fig. 2.

Typical microstructure of the $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films is shown in Fig. 3. Figure 3(a) is a TEM photograph of the $(Fe_{0.9}Co_{0.1})_{77}N_{23}$ film with substrate temperature of 25 °C. We can see that the film has a nanocrystalline structure and its average grain size is about 13 nm. In Fig. 3(b), from the diffraction lines of the electron diffraction pattern of this film, we noticed that ϵ-$Fe_{2-3}N$, Co_2N, $\gamma'-Fe_4N$, and ζ-Fe_2N phases are included.

Figure 4 shows the saturation magnetization $4\pi M_s$ and coercivity H_c as a function of N content in $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films with its substrate temperature at 25 °C. It is clear that $4\pi M_s$, and both in-plane coercivity $H_{c\parallel}$ and perpendicular coercivity $H_{c\perp}$ of the films increase slowly from 22 to 22.8 kG, 12 to 18 Oe, and 22 to 30 Oe, respectively, for N less than roughly 13 at. %. However,

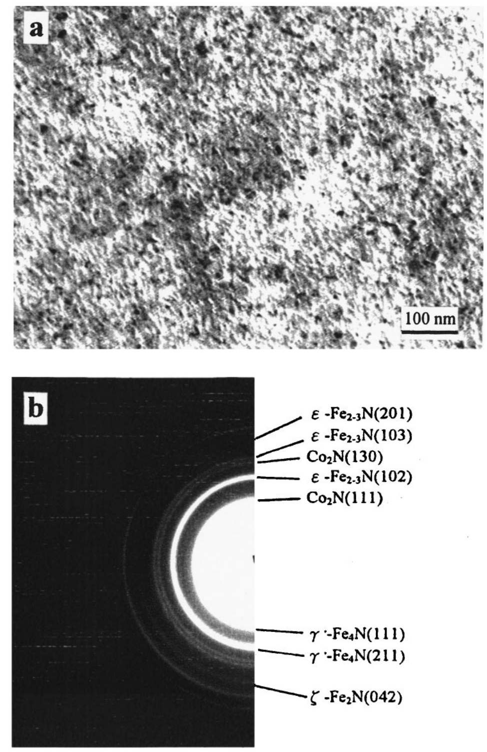

FIG. 3. TEM micrograph and electron diffraction pattern of $(Fe_{0.9}Co_{0.1})_{77}N_{23}$ film with $T_s = 25$ °C. (a) is the microstructure of planar section and (b) is the corresponding electron diffraction pattern.

$4\pi M_s$ decreases and both H_c's increase rapidly as N content is larger than 13 at. %. When N content is approached to 30 at. %, both H_c increase to about 150 Oe and $4\pi M_s$ decreases to about 2 kG due to the formation of ζ-Fe_2N phase in the film. The perpendicular coercivity $H_{c\perp}$ is slightly higher than that of the in-plane coercivity $H_{c\parallel}$. This indicates that the film is almost magnetic isotropy as substrate temperature is kept at 25 °C.

Figure 5 shows the relation between $4\pi M_s$, $H_{c\parallel}$, $H_{c\perp}$ and substrate temperature T_s of the $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films prepared with the flow ratio of N_2 at 1 vol. % during sputtering. According to the AES analysis, the N content of the

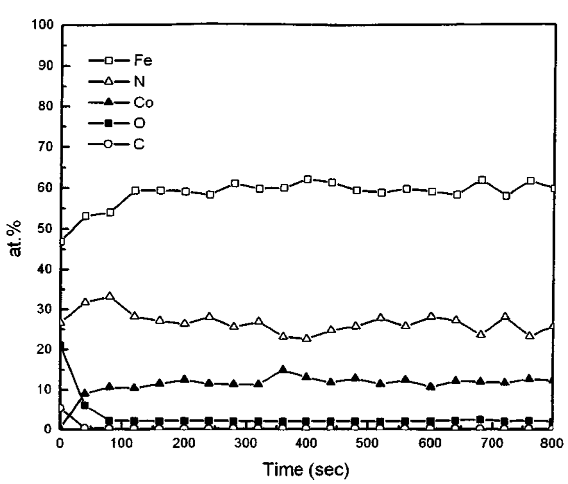

FIG. 2. Auger depth profile of a $(Fe_{0.9}Co_{0.1})_{70}N_{30}$ film.

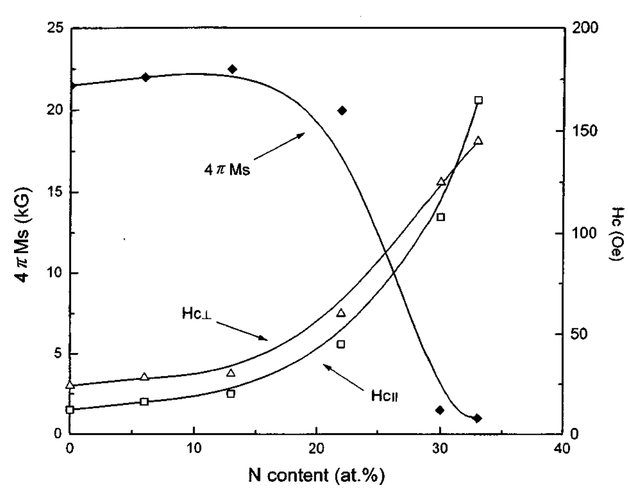

FIG. 4. Effect of N content on the $4\pi M_s$, $H_{c\parallel}$, and $H_{c\perp}$ of $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films. The substrate temperature is kept at 25 °C.

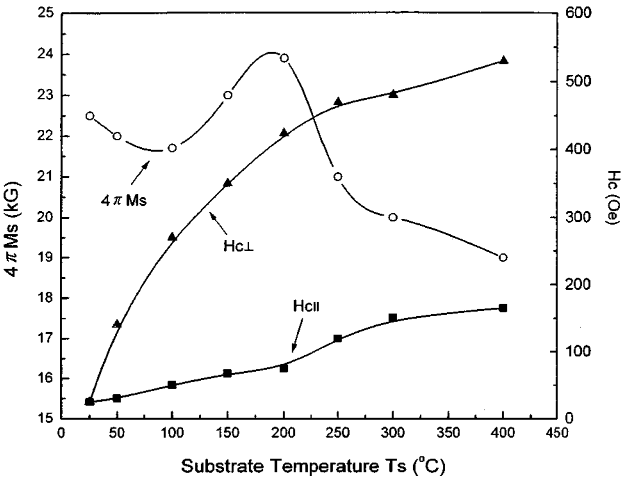

FIG. 5. The change of $4\pi M_s$, $H_{c\parallel}$, and $H_{c\perp}$ against T_s for $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films. N_2 flow ratio is 1 vol. % during sputtering.

film was found to decrease from 13 at. % for $T_s = 25$ °C to 10 at. % for $T_s = 100$ °C, 8 at. % for $T_s = 200$ °C, and 3 at. % for $T_s = 400$ °C. It can be seen that $H_{c\parallel}$ increases monotonously from 25 to 165 Oe when T_s increasing from 25 to 400 °C, but $H_{c\perp}$ of the film is increased rapidly from 25 to 530 Oe. It is obvious that magnetic easy axis is in the film plane, as T_s is higher than 25 °C. The increasing of coercivities with T_s is owing to that N content of the film is decreased with increasing T_s. From the x-ray diffraction patterns study of the films at various T_s, we found that the amount of high coercivity FeCo phase is increased with T_s. For example, Fig. 6 shows curves (a) to (c) with their substrate temperatures at 25, 100, and 200 °C, respectively. When T_s is higher than 300 °C, only the FeCo peak can be detected by x-ray diffractometer. According to the x-ray Scherrer's equation analysis from the diffraction peak of FeCo(100), we found that the grain size of the films with substrate temperature below 200 °C is roughly about 13 nm, however, it increases very fast for films with substrate temperature above 300 °C.

The $4\pi M_s$ in Fig. 5 shows a minimum value of 21.7 kG around 100 °C, and a maximum value of 24 kG at $T_s \cong 200$ °C. Comparing with the x-ray diffraction peaks of Figs. 6(a), 6(b), and 6(c), we can see that these films all contain high moment FeCo and $\alpha' - Fe_{6-10}N$ phases.[13] From this point of view, variation of the $4\pi M_s$ of film with T_s is due to the change of N content in the film, i.e., amounts of various FeN, FeCo, CoN, α-Fe phases in the film are changed.

IV. CONCLUSIONS

In conclusion, effects of nitrogen contents and substrate temperatures to the microstructure and magnetic properties of the $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films with $x = 0-30$ films have been investigated. TEM observation found that they have nanocrystalline structure. When N content of the film is lower than 13 at. %, it contains high moment $\alpha' - Fe_{6-10}N$, FeCo, and γ'-Fe_4N phases. The $(Fe_{0.9}Co_{0.1})_{92}N_8$ film which prepared at substrate temperature of about 200 °C and N_2 flow ratio of 1 vol. % has maximum $4\pi M_s$ of 23.9 kG. As N content of the film is higher than 20 at. %, $4\pi M_s$ of the film decreases rapidly due to the formation of nonmagnetic ζ-Fe_2N phase in the film.

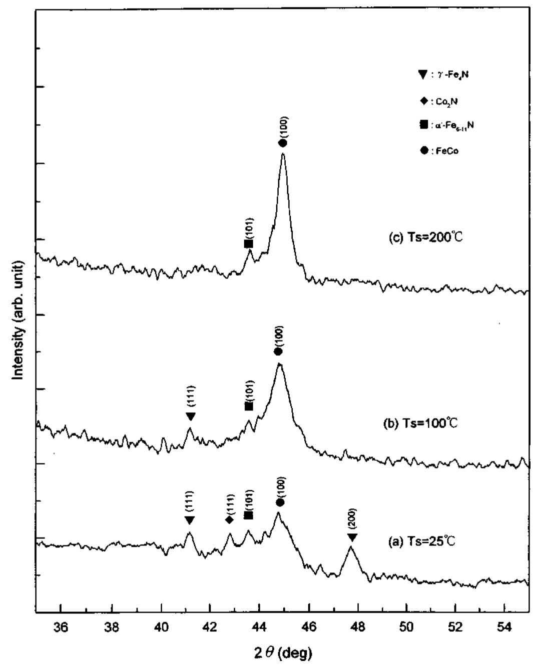

FIG. 6. X-ray diffraction patterns of various $(Fe_{0.9}Co_{0.1})_{100-x}N_x$ films. N_2 flow ratio is 1 vol. % during sputtering and substrate temperatures are (a) $T_s = 25$ °C, (b) $T_s = 100$ °C, and (c) $T_s = 200$ °C.

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