Journal of Korean Magnetics Society, Volume 5, Number 5, October 1995

ANNEALING BEHAVIOR OF FeN THIN FILMS

S. Park, Y.Choi and S. Jo
Dept. of Electronic Engineering, Soong Sil University, Seoul, 156-743, Korea

Abstract-FeN thin films were deposited on glass by RF diode reactive sputtering. The films were annealed in the air and in vacuum. The film annealed in the air showed sharp decrease of saturation magnetization and change of easy axis direction to hard axis direction and vice versa after 300 ℃ anneal. The coercivity decreased down to 0.5 Oe after 400 ℃ anneal. After 450 ℃ anneal, the film showed ε-$Fe_{2\text{-}3}N$ phase. The films annealed in vacuum showed coercivity increase after 300 ℃ anneal for the film deposited with initial substrate temperature of 35 ℃ and after 400 ℃ anneal for the film deposited with initial substrate temperatue of 170 ℃. These films showed $Fe_{16}N_2$ X-ray peaks after 450 ℃ anneal.

I. INTRODUCTION

The magnetic properties of FeN thin films can be controlled by the composition and micro-structure of the films[1,2]. They have good corrosion resistance and abrasion resistance. Furthermore, FeN films have high saturation magnetization (about 20 KG), low coercitivity (below 0.5 Oe), and high operating frequency[3]. Therefore, they are expected to be used as thin film head materials for high density recording.

Up to now, results have been reported for the films deposited by RF diode sputtering[4,5], facing target sputtering[6], plasma evaporation[7] and ion beam deopsition[8].

In this paper, annealing behavior of FeN thin films, which are deposited by RF diode reactive sputtering method, are reported. Annealing temperature and initial substrate temperature are varied. Annealing in the air and in vacuum is performed and the results are compared.

II. EXPERMENTAL PROCEDURE

The films were deposited onto corning glass 7059 substrates using Perkin-Elmer 2400 8L RF-diode sputtering system (8″ targets). The three layer FeN film annealed in the air was deposited with the base pressure of 4.0×10^{7} Torr and RF power of 1050 W. The flow ratio of N_2 and Ar was 7 : 10, and gas pressure was 3 mT. The SiO_2 layers deposited between the FeN layers were deposited with the conditions of 30 Å of thickness, 100 W of power and 10 mT of Ar pressure. The total thickness of the three layer film was about 12,500 Å. Two single FeN layer were annealed in vacuum. 800 W (2.47 W/㎠) input power, 3 mT gas pressure and N_2 to Ar gas flow ratio of 6.6 : 100 were sputtering conditions for the films. Thickness of the films were 6,000 Å. 45 Oe of DC magnetic field was applied by holding $SmCo_5$ magnets around the substrate table to induce the magnetic anisotropy. Substrate heating was provided by quartz lamp located above the substrate table and temperature characteristics were obtained using a K-type thermocouple.

The annealing temperature was increased from 100 ℃ to 450 ℃ by 50 ℃ and annealing time was 1 hour each. The films were left in the oven to cool to room temperature. When annealed, magnetic field was not applied, and the vacuum was maintained below 10^{5} Torr.

The easy and hard axis coercivities (Hc,e and Hc,h) and the saturation magnetization ($4\pi Ms$) were measured by a vibrating sample magnetometer (VSM). The film thickness was determined with a Tencor alpha-step profilometer. The film microstructure was analyzed by X-Ray diffractometer (XRD) after each anneal. The incident X-ray was Cu-Kα(wavelength 1.54Å) radiation and the incident angle was increased by 0.1° .

Ⅲ. EXPERIMENTAL RESULTS AND DISCUSION

Fig. 1 shows the change of coercivity and saturation magnetization of a 3 layer FeN thin film with annealing temperature. The annealing was done in the air. Up to 400 ℃, the coercivity initially increased somewhat and then decreased. However after 450 ℃ anneal, the coercivity increased sharply. When annealing temperature was increased from 250 ℃ to 300 ℃, the easy axis changed to the hard axis, and vice versa. The reason for this is not yet clear.

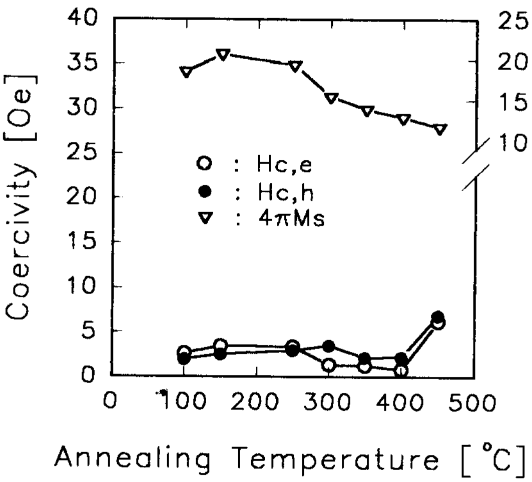
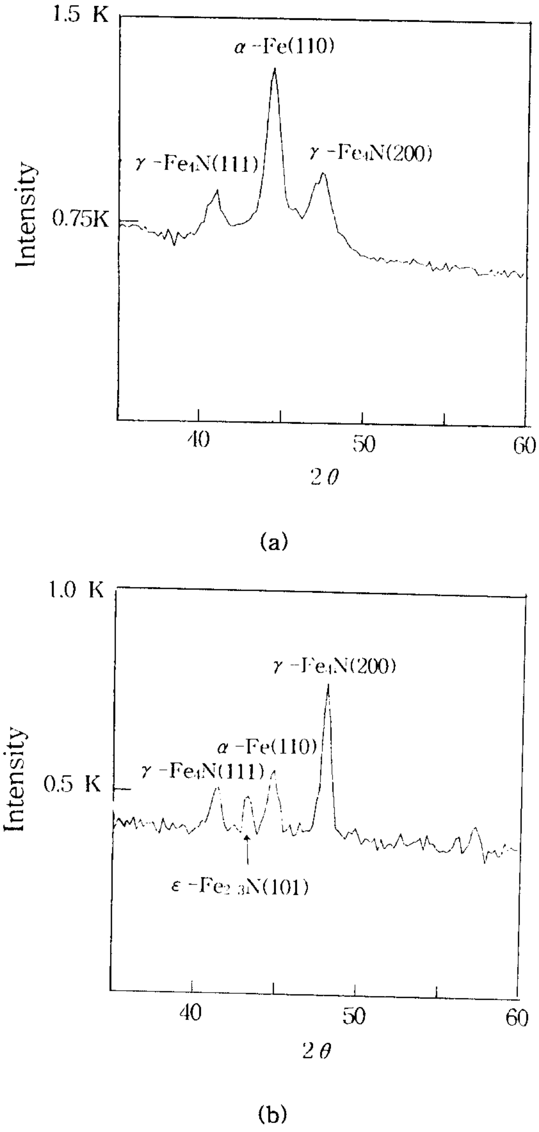


Fig. 1 Variations of Hc and 4πMs with annealing temperature. Annealed in the air.
Film deposition conditions : Input power 1050 W, Gas pressure 3mT, Flow ratio 7 : 100 (N_2 : Ar), 3 FeN layers, Total thickness 12,500 Å.

The saturation magnerization initially increased to 20.7 KG, then decreased rapidly as the annealing temperature became higher than 300 ℃.

Fig. 2 shows X-ray diffraction peaks of as deposited film (a) and the film after 450 ℃ of annealing (b). Comparing Fig. 2 (a) and Fig. 2 (b), intensity of α-Fe (110) phase decreased, γ-Fe_4N (200) phase increased and ε-$Fe_{2\text{-}3}N$ (101) phase was created after the annealing. According to FeN phase diagram[9], ε-$Fe_{2\text{-}3}N$ phase can be created if the N atomic content is higher than 20 % at 450 ℃. Thus, it is believed that ε-$Fe_{2\text{-}3}N$ phase is created during the annealing due to the incoporation of N_2 in the air to the film.



(a)

(b)

Fig. 2 XRD patterns (a) As-deposited (b) annealed at 450 ℃ in the air.

Fig. 3 and Fig. 4 show the coercivity and saturation magnetization variation with annealing temperature of single layer films which were deposited with initial substrate temperature of 35 ℃ and 170 ℃, respectively.

These films were annealed in vacuum. When the initial substrate temperature was 35 ℃ (Fig. 3), coercivity did vary much until 350 ℃, then increased sharply after 450 ℃ anneal. On the other hand, the film deposited with substrate temperature of 170 ℃ (Fig. 4) showed gradual increase of coercivity after 300 ℃ of anneal. The saturation magnetization did not vary up to 300 ℃ anneal, then decreased slowly down to 17 KG after 400 ℃ anneal. However, after 450 ℃ anneal, the saturation magentization increased about 1 KG, which coincided with the appearance of $Fe_{16}N_2$ phase (Fig. 6).

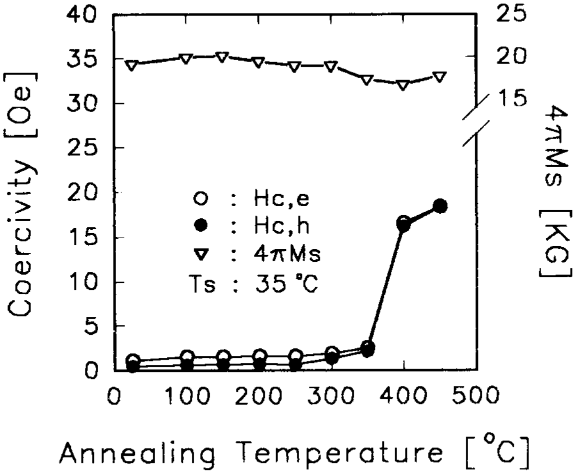


Fig. 3 Variations of Hc and 4πMs with annealing temperature Film deposition conditions : Input power 800 W, Gas pressure 3mT, Thickness 6,000 Å, Flow ratio 6.6 : 100 (N_2 : Ar), Single layer, Substrate temperature (Ts) 35 ℃.

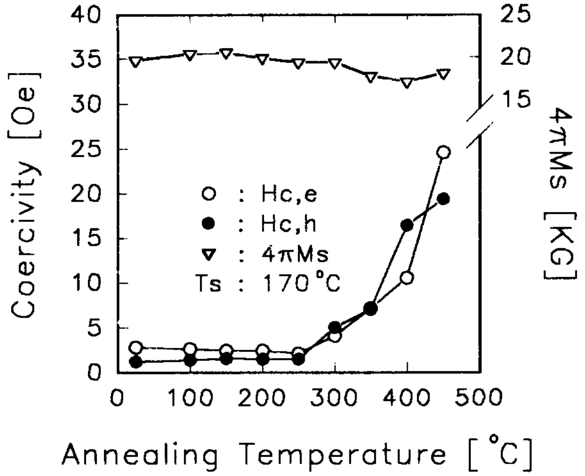
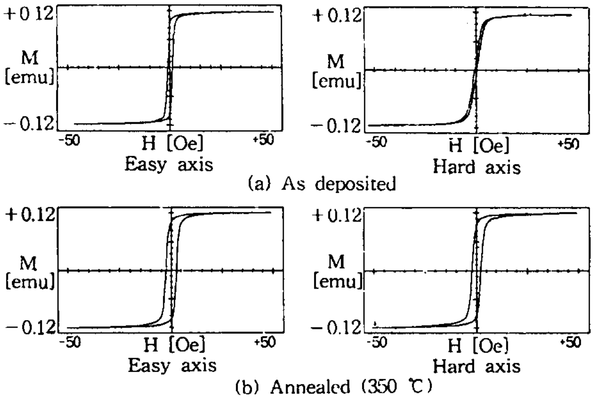


Fig. 4 Variations of Hc and 4πMs with annealing temperature. Film deposition conditions : Input power 800 W, Gas pressure 3mT, Thickness 6,000 Å, Flow ratio 6.6 : 100 (N_2 : Ar), Single layer, Substrate temperature (Ts) 170 ℃.

Fig. 5 shows hysteresis loops of the film deposited with initial substrate temperature of 35 ℃. Fig. 5 (a) shows the hysteresis loops in the easy and hard axis directions before the annealing. Fig. 5 (b) shows the hysteresis loops in the supposedly easy and hard axis directions after 350 ℃ annealing. Before the annealing, the hysteresis loop show typical easy and hard axis loops. However, after the annealing, the hysteresis loops indicates that the film actually became isotropic due to the increase of thermal energy which overcame the original induced anisotropy energy. The hysteresis loops of the film deposited with the initial substrate temperature of 170 ℃ exihibited more or less isotropic loops even before the annealing.



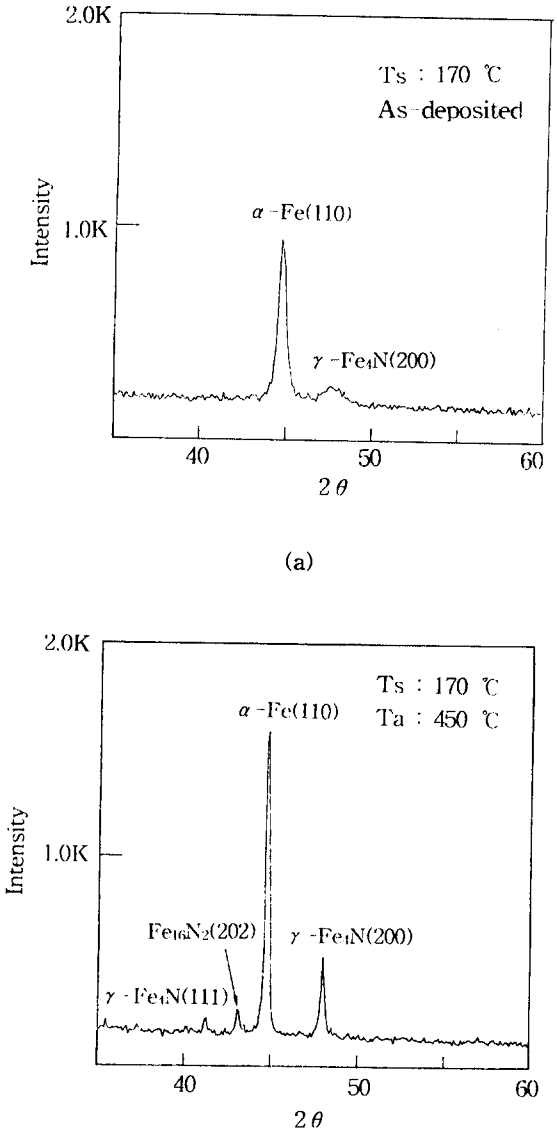
(a) As deposited

(b) Annealed (350 ℃)

Fig. 5 Hysterisis loops of the film deposited with substrate temperature of 35 ℃ (a) As-deposited (Hc,e : 1.12 Oe, Hc,h : 0.51 Oe) and (b) after annealing at 350 ℃ (Hc,e : 2.56 Oe, Hc,h : 2.20 Oe).

Fig. 6 shows X-ray diffraction peaks of the film deposited with substrate temperature of 170 ℃. Fig. 6 (a) shows X-ray peaks of the as-deposited film and Fig. 6 (b) shows that of 450 ℃ annealed film. Before annealing α-Fe and Fe_4N phases were shown but after 400 ℃ anneal, Fe_4N (111) phase started to show and the coercivity increased sharply. After 450 ℃ anneal $Fe_{16}N_2$ (202) phase appeared.

Also the intensity of the peaks of the α-Fe phase increased and the width decreased after the anneal. Therefore the grain size of the film increased.



(a)

(b)

Fig. 6 XRD patterns (Ts : 170 ℃) (a) As-deposited (b) annealed at 450 ℃.

X-ray diffraction patterns of the film deposited with initial substrate temperature of 35 ℃ showed similar peaks, but lower intensity of Fe$_{16}$N$_2$ phase.

Fig. 7 shows the change of grain sizes with the annealing temperature. As the annealing temperature increased, the grain size remained the same up to 250 ℃, then increased. The grain size was larger for the film deposited with lower initial substrate temperature (Ts = 35 ℃), which incidently exihibited lower coercivities.

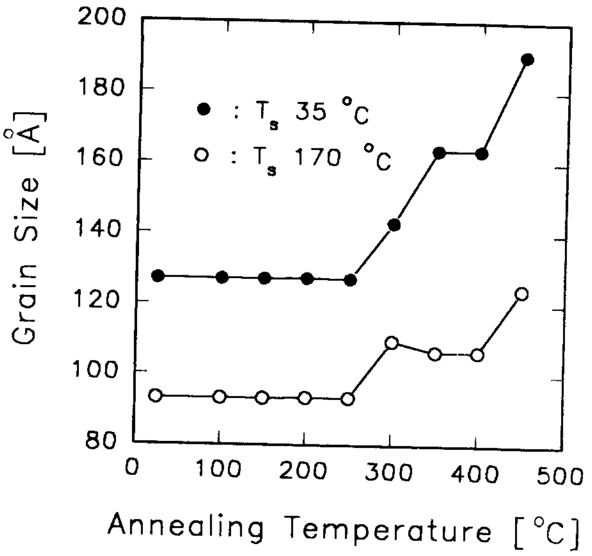


Fig. 7 Variations of grain size with annealing temperature.

IV. CONCLUSION

FeN films were depoisted with RF diode sputtering method and their annealing behavior was investigated. The film annealed at 450 ℃ for 1 hour in the air showed ϵ-Fe$_{2-3}$N phase along with α-Fe and γ-Fe$_4$N phases which were present before the annealing. However, films annealed in vacuum did not show ϵ-Fe$_{2-3}$N phase, and rather showed Fe$_{16}$N$_2$ phase after 450 ℃ of anneal.

Comparision of annealing results of the films deposited near room temperature (35 ℃), and at elevated temperature (170 ℃) indicated better temperature stability for the film deposited near room temperature. This suggests that the RF diode sputtering increases the substrate temperature during the deposion, and there is no need for additional substrate heating[10].

ACKNOWLDEGEMENT

This research was supported by Soong Sil University Research Fund, 1994.

REFERENCES

[1] T. K. Kim and M. Takahashi, Appl. Phys. Lett. 20. 492 (1972).

[2] N. Terarda, Y. Hosh, M. Naoe and S. Yamanaka, IEEE Trans. Magn., MAG-20, 1451 (1984).

[3] S. Wang. M. H. Kryder and D. J. Roger, IEEE Trans. Magn., MAG-28, 2418 (1992).

[4] C. Chang, J. M. Siversten, IEEE Trans. Magn., MAG-23, 3636 (1987).

[5] S. Wang and M. H. Kryder, J. Appl. Phys., 67 (9), 5134 (1990).

[6] A. Morisaka, M. Naoe, J. Appl. Phys., 63 (8), 3230 (1988).

[7] T. Tanaka, E. Fujita, J. Magn. Soc. Japan 14, 256 (1990).

[8] D. S. Rickerby, J. Vac. Sic. Technol., A4 (6), 2809 (1986).

[9] T. B. Massalski, Binary Alloy Phase Diagrams, Second Edition, Vol. 2, APDIC, p.1728.

[10] S. Jo, S. Park, Y. Choi. Presented at the '95 INTERMAG Conference, paper CP-09.