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Study of nanocrystalline Fe–Al–N soft magnetic thin films*

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Fe–Al–N films were fabricated by reactive sputtering using a radio-frequency magnetron sputtering system. The effects of Al and N content and annealing temperature on microstructure and magnetic properties were investigated. The Fe–Al–N films, which have good soft magnetic properties, consist of nanocrystalline α-Fe grains and a small amount of other phases in the boundaries of α-Fe grains. The average α-Fe grain size is about 10–15nm. A slight amount of Fe–N and Al–N compounds precipitate in the boundaries of α-Fe grains and suppress their growth. Annealing improves the soft magnetic properties slightly by releasing the residual stress and reducing defects.

Keywords: soft magnetic properties, nanocrystalline, lattice deformation
PACC: 7530E, 7150B, 7550S

1. Introduction

The magnetic storage system represents the fastest developing high technology field owing to the requirement of the larger capability of information storage systems. Research focuses on achieving higher recording density. For longitudinal recording mode, the research has shown[1] that in order to achieve ultra-high density magnetic recording, the magnetic particles in the medium should be as small as possible, but this would bump up the supermagnetism limit. The magnetic poles would fluctuate even at room temperature and the recorded data would be lost gradually. So the lower limit of particle size is to avoid the supermagnetism. This means $K_u V \gg K_B T$, that is, the crystal anisotropy constant K must be high and the medium has high coercivity. For example, to realize more than 15.5Gbit/cm^2 density, the coercivity of the medium must be as high as 560kA/m.

In order to write data effectively in a high-coercivity medium, the field near the top pole of the write head should be three or four times as large as the coercivity of the medium. When coercivity is about 560kA/m, the write field is at least 2T. So, the study on write head materials mainly focuses on high B_s materials.[2] Write head materials must also have soft magnetic properties such as low coercivity H_c and high-frequency permeability. FeN films have B_s as high as 2.2T and also show good soft magnetic properties.[3] However, their thermal stability retards their practical applications. The addition of a third element (Ta, Hf, Nb, Zr, Al, etc) can effectively improve the thermal stability.

Soft magnetic properties of Fe–M–N films (where M represents Ta, Hf, Nb, Zr, Al, etc) originate from the incorporation of interstitial N atoms, which are instrumental in expanding the crystalline lattice and refining the grain size.[4] When the grain size is smaller than the ferromagnetic exchange length,[5,6] the local magnetic anisotropy can be substantially reduced.[7] Additionally, N atom incorporation and the lattice constant strongly depend on the nature of the added element, in terms of chemical affinity.[8] Therefore, each of the added elements also affects the soft magnetic behaviour in an individual way . In this paper the influence of Al and N concentration, sputtering conditions and annealing temperature on microstructure and magnetic properties of Fe–Al–N films are investigated.

2. Experimental procedure

A composite target was used to fabricate the Fe–Al–N films. The target was composed of a pure iron plate covered with small Al chips. The Al content was controlled by adjusting the total area of Al chips. The concentration of N was controlled by the nitrogen partial pressure in the argon–nitrogen sputtering gas mixture. The films were deposited on glass substrates

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by radio-frequency magnetron sputtering. The sputtering parameters used were: base vacuum pressure 9×10^{-5}Pa, Ar pressure 0.3Pa, nitrogen partial pressure 0–0.03Pa, and target power density 2.3W/cm^2. The thickness of the films was approximately 500nm. The heat treatment was carried out in a vacuum furnace with a pressure less than 6×10^{-4}Pa at a temperature up to 400°C.

The Fe and Al content in the films were determined by energy diffraction x-ray (EDX) analysis. The saturation magnetization B_s and coercivity H_c were measured using a high sensitivity vibrating sample magnetometer (VSM) and a sensitive fluxmeter, respectively. The film structure analysis, such as phase identification, crystalline grain size (Scherrer method) and interplanar spacing (d), were carried out by x-ray diffraction (XRD) and were verified by transition electron microscope (TEM).

3. Results and discussion

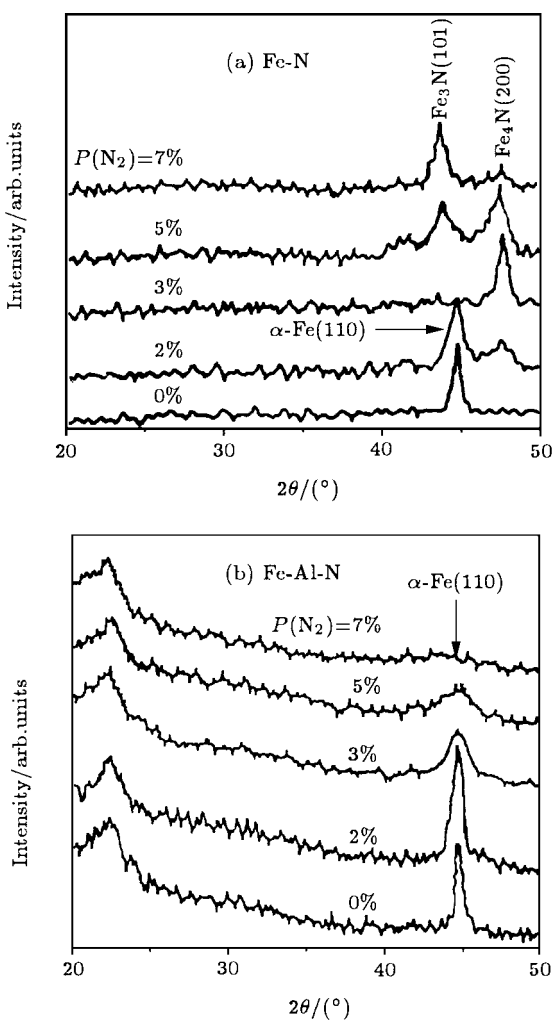


Fig.1. XRD patterns of as-sputtered films: (a) Fe–N and (b) Fe–Al–N.

Figures 1(a) and (b) show the XRD patterns of Fe–N and Fe–Al–N as-deposited films, respectively, with nitrogen partial pressure $P(N_2)$ as a parameter. The main peak in the spectra of the Fe–Al–N films is the (110) peak of α-Fe, which indicates that the films have (110) preferential orientation parallel to the film plane. It is also noted from Fig.1(b) that the intensity of the (110) peak decreases, but the width increases with increasing $P(N_2)$. When $P(N_2)\geq5\%$, the films become amorphous. For the Fe–N films, the γ-Fe_4N phase appears when $P(N_2)=2\%$. With increasing $P(N_2)$, not only γ-Fe_4N, but also other Fe–N compounds such as Fe_3N phase are produced. Therefore, the addition of Al effectively inhibits the formation of Fe–N compounds.

The dependence of magnetic properties of Fe–Al–N films on Al content is given in Fig.2. The films were sputtered at $P(N_2)=3\%$ and annealed at 350°C. Since Al is non-magnetic, the saturation magnetization B_s decreases monotonically with increasing Al content. We find that in a nitrogen-free film, the film consists of polycrystalline α-Fe, the lattice constant of polycrystalline α-Fe is about 0.2864nm and smaller than that of pure α-Fe (a=0.2866nm). This means that the added Al atoms replace Fe atoms in the α-Fe lattice and a substituted solid solution α-Fe(Al) is formed because the Al atom size is smaller than the Fe atom. Meanwhile, we know that soft magnetism in Fe–M–N films originates from the incorporation of interstitial N atoms, which is instrumental in expanding the crystalline lattice and refining the grain structure, but the N incorporation strongly depends on the M–N affinity. For Fe–Al–N films, the N incorporation is low because the interaction coefficient (e_N^{Al}=0.0025) is relatively low,[9] and Al leads to small lattice dilation due to low N solubility in the α-Fe lattice and a higher tendency to form metal nitrides. Therefore, it is reasonable to suppose that some of the Al atoms replace Fe atoms in α-Fe and form a substitutional solid solution, while others combine with Fe and N to form non-magnetic compounds. The coercivity, H_c, first decreases, approaching its lowest value at Al content 2.5%, and then increases as the Al content is increased. It is known that Fe–N films with (110) α-Fe orientation exhibit good soft magnetic properties at a critical deformation of the (110) lattice spacing, $\Delta d(110)/d(110)$ of about 0.28%.[10] Especially, along the critical line of $\Delta d(110)/d(110)=0.28\%$, the value of μ_i shows a fairly high value of about 2000 at a grain size less than 15nm. Figure 3 shows the ef-

fect of Al content on grain size D and deformation $\Delta d(110)/d(110)$. D decreases but $\Delta d(110)/d(110)$ increases as the Al content is increased. The value of $\Delta d(110)/d(110)$ approaches 0.28% for an Al content of about 2%–3% and D is less than 15nm. Therefore, the good soft magnetic properties of the Fe–2.5Al–N films may be attributed to the suitable size of the α-Fe grains and to the optimum value of $\Delta d(110)/d(110)$.

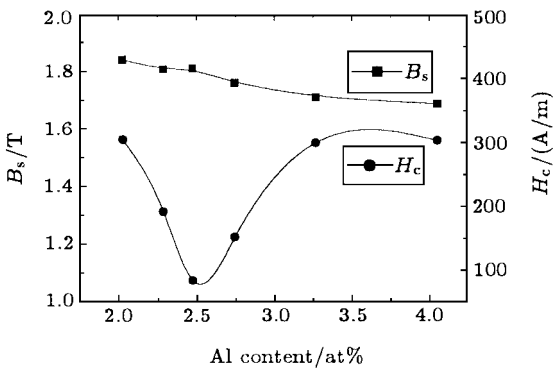


Fig.2. Dependence of B_s and H_c on Al content.

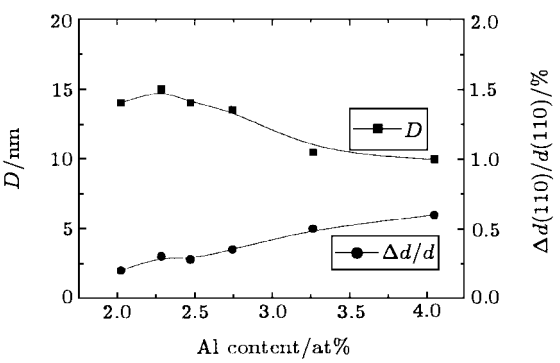


Fig.3. Effect of Al content on D and $\Delta d/d$.

Figure 4 shows the dependence of the magnetic properties of Fe–2.5Al–N films on nitrogen partial pressure $P(N_2)$. With increasing $P(N_2)$, B_s first decreases slowly, and then decreases rapidly when $P(N_2)$ >3%. H_c decreases drastically approaching its minimum value, and then increases slowly with $P(N_2)$. The change of magnetic properties is related to the microstructure of the film. It is clear from Fig.1(b) that the intensity of the (110) peak decreases and the peak broadens with increasing $P(N_2)$. When $P(N_2)$ $\geq$3%, the film consists of nanocrystalline α-Fe. When $P(N_2)$ >5%, the sputtered films are amorphous.[11] It is well known that, for Fe–N based soft magnetic materials, the refining of α-Fe grains originates from the incorporation of interstitial N atoms. We can suppose that, with increasing $P(N_2)$, more and more N atoms enter into α-Fe lattice sites as interstitial atoms and refine the α-Fe grains, until the amorphous structure is formed. When $P(N_2)$=3%, the film shows the best soft magnetic properties. This could be due to the increment of ferromagnetic exchange coupling among α-Fe grains, because the grain size is smaller than the ferromagnetic exchange coupling length and the local magnetic anisotropy is substantially lowered.

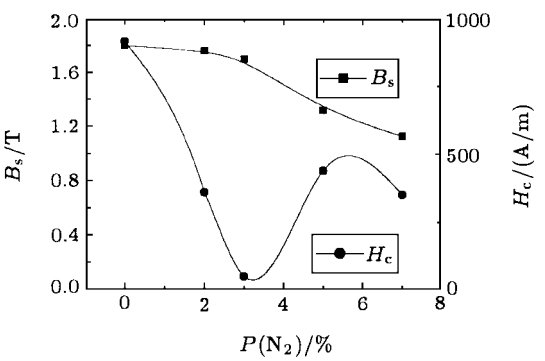


Fig.4. Dependence of B_s and H_c on $P(N_2)$ in Fe–2.5Al–N films.

Figure 5 shows TEM images and electron diffraction pattern of a Fe–Al–N film prepared at $P(N_2)$=3%. The dark-field and bright-field images are taken from the α-Fe (110) diffraction line. These images indicate that the average α-Fe grain diameter is approximately 15nm. We can also see from the electron diffraction patterns in Fig.5 that there are other phases such as Fe–N and Fe–Al compounds in the film. These compounds are extremely fine (roughly 1–3nm) and dispersed uniformly throughout the film. We can suppose that, as $P(N_2)$ increases, more Fe–N and Fe-Al compounds are formed. Because the saturation magnetization of the Fe–N and Fe–Al compounds are smaller than that of the α-Fe, the B_s of the films would decrease. When more Fe–N and Fe–Al compounds precipitate in the boundaries of α-Fe grains and lower the ferromagnetic exchange coupling, the increment of H_c is natural.

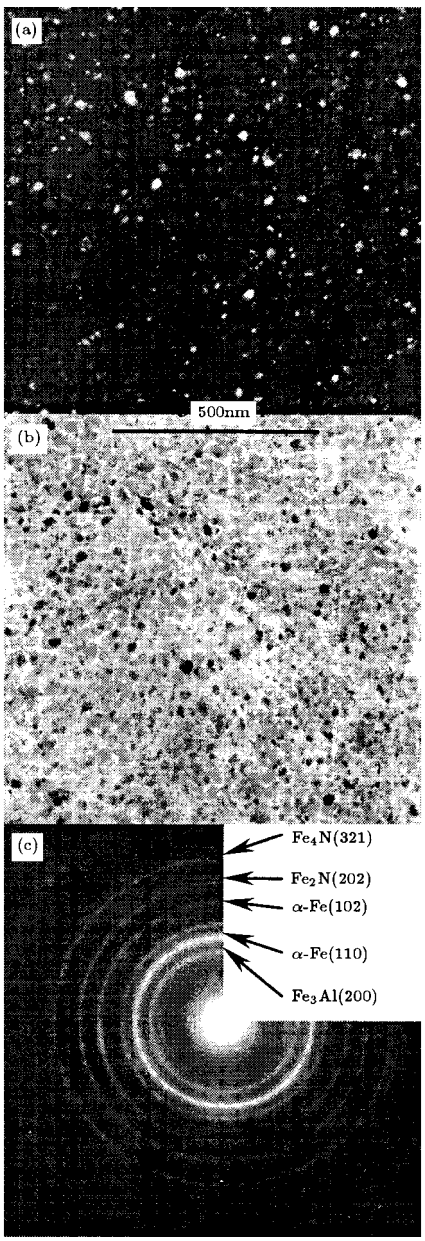


Fig.5. TEM images and electron diffraction pattern of Fe–Al–N films prepared at $P(N_2)$=3%.

The effect of the annealing temperature T_a on the magnetic properties of the Fe–2.5Al–N film is shown in Fig.6. The film was deposited at $P(N_2)$=3%. B_s remains constant at different annealing temperatures. When $T_a \leq 350°C$, the annealing slightly improves H_c, but above 350°C, H_c increases rapidly. This behaviour suggests that the annealing does not change the structure of the film. Annealing at low temperatures decreases the stress in the films and improves crystallization. The grain size increases as a result of annealing at high temperature, as verified by XRD patterns.

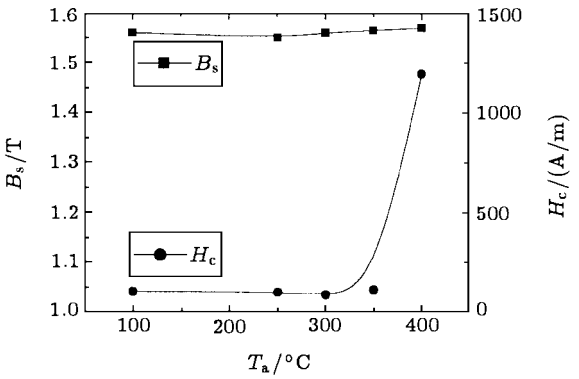


Fig.6. Effect of annealing temperature T on B_s and H_c of the Fe–2.5Al–N film.

Figure 7 shows the XRD patterns of Fe–2.5Al–N films, as-deposited and after annealing. These films were sputtered at $P(N_2)$=3%. The as-deposited film mainly consists of (110) textured α-Fe phase with bcc lattice. When $T_a \leq 350°C$, the patterns show no obvious difference. When T_a=450°C, the intensity of the (110) peak increases and its width narrows. We can also see from Fig.7 that the shape of the (110) peak is not Gaussian. Therefore, it is reasonable to assume that there are traces of Fe–N or Fe–Al compounds in the films, which precipitated in the boundaries of α-Fe grains[12] and suppressed their growth.

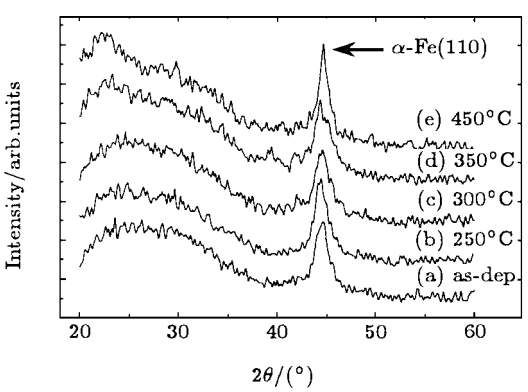


Fig.7. XRD patterns of Fe–2.5Al–N films at different annealing temperatures.

4. Summary

The nanocrystalline Fe–Al–N films were deposited by reactive sputtering. It is found that the addition of Al effectively inhibits the formation of Fe–N compounds. The structure and magnetic properties of Fe–Al–N films are sensitive to the amount of Al and N present in the films. Besides the bcc α-Fe, there are a few other phases such as Fe–N and Fe–Al compounds in the films. These compounds precipitate in the boundaries of α-Fe grains and suppress

their growth. Annealing at T_a <350°C improves the soft magnetic properties of Fe–Al–N films by releasing the residual stress and removing crystal defects. The good soft magnetic properties originate from the suitable grain size and lattice deformation of α-Fe.

References

[1] Grady K O, Whit R L and Grundy P J 1998 *J. Magn. Magn. Mater.* **177-181** 886

[2] Romankiw L T 2000 *J. Magn. Soc. Jpn.* **24** 1

[3] Takanashi M, Shoji H, Shimatsu T, Komaba H and Wakiyama T 1990 *IEEE. Trans. Magn.* **26** 1503

[4] Russak M A, Jahnes C V, Klokholm E, Lee J W, Re M E and Webb B C 1992 *J. Magn. Magn. Mater.* **185** 104

[5] Herzer G 1989 *IEEE. Trans. Magn.* **25** 3327

[6] Hoffman H 1978 *Thin Solid Films* **58** 223

[7] Takahashi M and Shimatsu T 1995 *IEEE. Trans. Magn.* **31** 3973

[8] Viala B, Minor M K and Barnard J A 1996 *J. Appl. Phys.* **80**(8) 3941

[9] Penkle R D and Elloitt F 1960 *AIME Trans.* **218** 1088

[10] Takahashi M, Shimatsu T and Shoj H 1992 *Proc. ICF* p 1483

[11] Lin D M, Wang H S, Wu Y C and Lin M L 1999 *Acta Phys. Sin.* (Overseas Edition) **9** 654

[12] Wang B Y, Suwabo S, Tsunida F and Laughlin D 1995 *IEEE. Trans. Magn.* **31** 3967