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# Evolution of structure and magnetic properties of nanocrystalline Fe$X$N thin films via Ta and Al addition*

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Nanocrystalline FeAlN and FeTaN films are prepared by direct growth and crystallization of their as-deposited amorphous films, respectively. The two films both show soft magnetism of nanocrystalline, but their uniaxial anisotropy is observed to be different from each other. Measurements of microstructure reveal that Ta addition leads to higher N-solubility in these films, and results in larger lattice dilation and more compressive stress. The uniaxial anisotropy is the consequence of the anisotropic distribution of interstitial N atoms in an $\alpha$-Fe lattice. Al is easy to react with nitrogen, therefore the $\alpha$-Fe is purer in the FeAlN film than in the FeTaN film and the stress is tensile in the FeAlN film. The difference in anisotropy may be attributed to the different microstructures in both films.

**Keywords:** Fe–$X$–N thin film, soft magnetic properties, uniaxial anisotropy, nanocrystalline
**PACC:** 7500, 7570A, 7550S

## 1. Introduction

In order to achieve ultrahigh density magnetic recording, media coercivity has to be larger than 240,000 A/m. For this reason, an inductive head pole material must exhibit high saturation magnetization that is accompanied with a difficulty in achieving combination of good soft magnetic properties. Soft magnetic Fe$X$N ($X$=Ta, Al, Ti, etc) films are very promising candidates for the next generation of inductive head because of their high moments ($4\pi M_{\rm s} = 1.8 - 2.0$T).[1,2]

It is well-known that the basic requirement for good soft magnetic properties is low or vanishing magnetic anisotropy.[3] The relevant anisotropy contributions increase in the following order in magnitude: magneto-crystalline anisotropy, magneto-elastic anisotropy, magnetic-field-induced anisotropy. Thus, the prerequisite for soft magnetic behaviour is that the effective magneto-crystalline anisotropy is low. Superior properties additionally require a low magneto-elastic anisotropy. Once these two contributions are sufficiently suppressed, the magnetic properties are mainly determined by induced anisotropy.

The soft magnetism in Fe$X$N films originates from the incorporation of N atoms which are instrumental in refining $\alpha$-Fe grain structure.[4] When a grain size is usually smaller than the ferromagnetic exchange coupling length (magnetic correlation length), $L_{\rm ex}(= 3.5 \times 10^{-9}\text{m})$,[5,6] the magnetization will not follow the preferred orientation of the axis of the individual grains, but will be forced to align parallel by exchange interaction. As a consequence, the effective anisotropy is an average over several grains, and, thus, is considerably reduced in magnitude. However, the incorporation of N strongly depends on the nature of element $X$. Meanwhile, the addition of N also gives the film a uniaxial anisotropy. It is known that the uniaxial anisotropy is "set" in the design of head in order to control the magnetic domain structure and improve the frequency response of the head.[7] In this work, the structure and magnetic properties of FeAlN and FeTaN films are investigated. It is found that both films show the difference in uniaxial anisotropy under suitable conditions, and then the mechanism is

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discussed.

## 2. Experimental procedure

Two different methods can be used to prepare nanocrystalline soft magnetic Fe$X$N films. The high-temperature technique is widely used to anneal amorphous as-deposited Fe$X$N alloys. The soft magnetic properties are enhanced by promoting a fine nanostructure from the amorphous structure. Another method is the direct growth of as-deposited soft magnetic nanostructure Fe$X$N. The soft magnetic FeTaN films were prepared by the former method because it is easier to prepare as-deposited amorphous FeTaN films. The latter method was used to grow nanocrystalline FeAlN films.

All samples were prepared using a magnetron sputtering system, in which composite targets were used. Small chips of Ta or Al were placed on the surface of pure iron target. The concentration of Ta or Al in the films was adjusted by varying the number of the chips. In this study, FeTaN films with high Ta (4at%) content were deposited on glass substrates cooled by water, which is helpful to form amorphous films. Nanocrystalline FeAlN films with low Al content (about 2.5at%) were deposited on the heated substrates. The concentration of N in the films was moderated by adjusting the nitrogen partial pressure, $P(\mathrm{N_2})$, where $P(\mathrm{N_2})$ is the ratio of nitrogen gas pressure to total mixed gas(Ar $+\mathrm{N_2}$) pressure in sputtering. Table 1 lists the deposition condition.

**Table 1.** Sputtering condition of FeAlN and FeTaN films.

| | FeAlN | FeTaN |
|---|---|---|
| background pressure/Pa | $6.7 \times 10^{-5}$ | $6.7 \times 10^{-5}$ |
| Sputtering Pressure/Pa | $2.7 \times 10^{-1}$ | $2.7 \times 10^{-1}$ |
| Nitrogen Pressure/% | 0–7 | 0–10 |
| Sputtering paved density/(w/m$^3$) | $2.3 \times 10^6$ | $7.3 \times 10^6$ |
| Substrate temperature/°C | 20–220 | 20 |
| Deposition rate/($10^{-11}$m/s) | 3.3–5.0 | 3.3–5.0 |

The magnetic properties, such as saturation magnetization and coercivity ($B_\mathrm{s}$ and $H_\mathrm{c}$), were measured using a highly sensitive vibrating sample magnetometer (VSM). Structure analyses of the films were carried out by x-ray diffraction (XRD), transmission electron microscope (TEM) and conversion electron Mossbauer spectra (CEMS).

## 3. Results and discussion

Figure 1 gives the x-ray diffraction patterns of as-deposited FeTaN films and post-450°C-annealed ones, which were prepared at different nitrogen partial pressures $P(\mathrm{N_2})$. The comparison between magnetic properties in as-deposited and annealed states is shown in

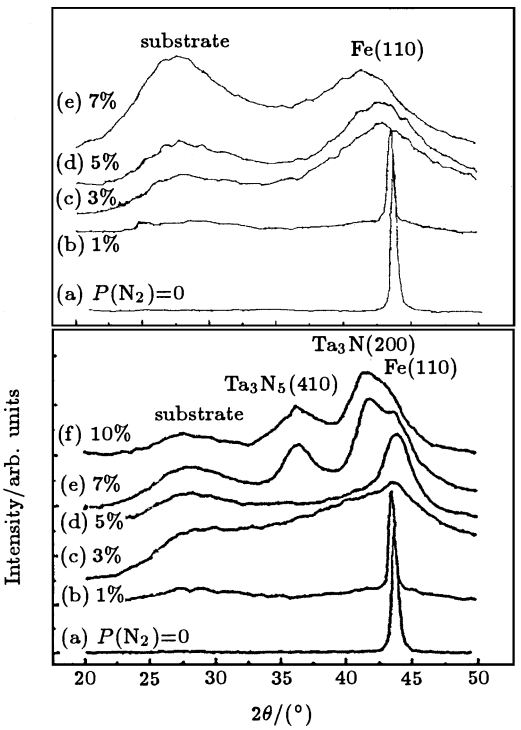



**Fig.1.** X-ray diffraction patterns of FeTaN films at different nitrogen partial pressures $P(\mathrm{N_2})$ Upper: as-deposited films; lower: post-annealed films.

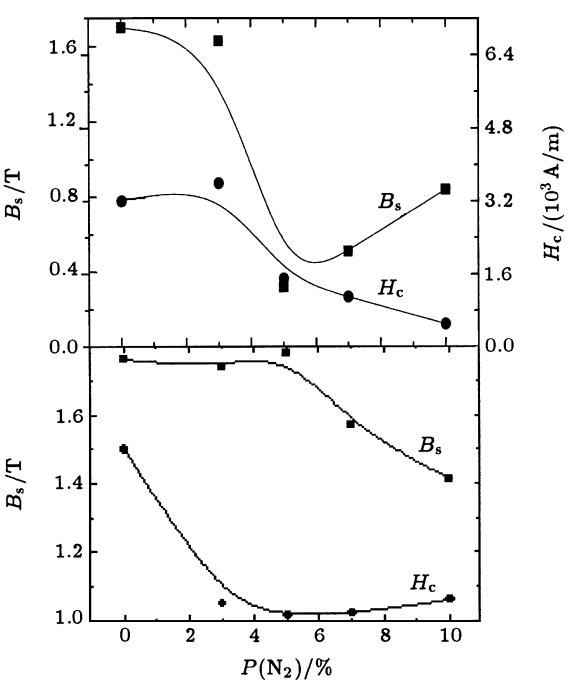



**Fig.2.** Magnetic properties of FeTaN films at different pressures $P(\mathrm{N_2})$. Upper: as-deposited films; lower: post-annealed films.

Fig.2. The film prepared at $P(N_2) = 5\%$ exhibits excellent soft magnetic properties. The film consists of nanocrystalline $\alpha$-Fe, which is crystallized from its as-deposited amorphous film.

FeAlN film prepared at $P(N_2)$=3% exhibits best soft magnetic properties.[8,9] Figures 3 and 4 show the XRD patterns and magnetic properties of the films deposited at different substrate temperatures, $T_s$, respectively. The film deposited at $T_s$=150°C has the minimum coercivity and exhibits good soft magnetic behaviour. It is composed of mainly nanocrystalline $\alpha$-Fe and a small amount of $\gamma$-$Fe_4N$.

**Table 2.** Typical structural and magnetic properties of both films.

| | FeAlN | FeTaN |
|---|---|---|
| Phase Structure | $\alpha$-Fe and small amount of $\gamma$-$Fe_4N$ | $\alpha$-Fe and residual amorphous matrix |
| Grain Size / $D10^{-10}$ m | <10 | <10 |
| Lattice dilation $\delta d(110)/d(110)$ | –0.15% | 2.69% |
| Saturation Magnetization $B_s$/T | 1.58 | 1.8 |
| Coercivity $H_c$/(A/m) | 80–160 | <80 |

Table 2 summarizes the magnetic properties and structural features of both films. The lattice dilation of $\alpha$-Fe is described by $\delta d(110)/d(110)$, where $d(110)$ is the interplane spacing of (110) plane.

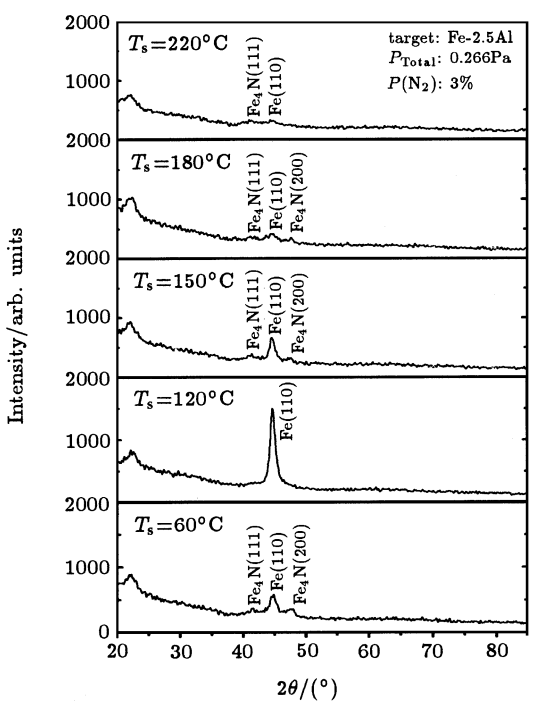



**Fig.3.** XRD patterns of FeAlN films deposited at various temperatures, $T_s$.

We notice that the $\alpha$-Fe grains have preferential orientation (110) to the film plane in both films. The grain size is smaller than the ferromagnetic exchange coupling length, $L_{ex}$(=$3.5\times10^{-9}$m), and both films show a good soft magnetic behaviour. The $\alpha$-Fe grain size in the FeTaN film is smaller than that in the FeAlN film, and the FeTaN film exhibits excellent magnetic properties, because the more the grain number is in the ferromagnetic correlation volume( $V=L_{ex}{}^3$), the stronger the exchange coupling between $\alpha$-Fe grains is and the smaller the local magneto-crystalline anisotropy is.

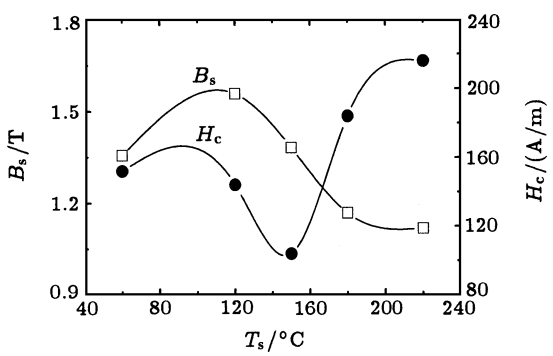



**Fig.4.** $B_s$ and $H_c$ of FeAlN films deposited at various temperatures, $T_s$.

It is interesting that both films show the difference in uniaxial anisotropy. Both films were deposited in an SPF-312 magnetron sputtering system. The leakage flux of rare-earth permanent magnet fixed under the sputtering target produces a magnetic field as high as 16,000 A/m in the substrate, actually. The films would be affected by the field in the deposition process. Therefore it is possible that uniaxial anisotropy may be induced by the external field, and the preferred orientation of the axis of uniaxial anisotropy in both films should be the same as and parallel to the field direction. Actually, the preferentially oriented axis of the FeTaN film is parallel to the direction of the magnetic field, but for the FeAlN film, the preferentially oriented axis of uniaxial anisotropy is perpendicular to the field. The effective magneto-crystalline anisotropy is effectively reduced by the ferromagnetic exchange

coupling between nanocrystalline $\alpha$-Fe grains in both films. Therefore it is reasonable to suppose that the difference in uniaxial anisotropy between both films may be related to magneto-elastic anisotropy or induced anisotropy.

The magneto-elastic anisotropy is proportional to stress. Usually, thin film stress consists of two main components: a thermal component resulting from the difference between thermal expansion coefficients of the film and the substrate, and an intrinsic component resulting from the structure of the film, for example the lattice dilation. It is believed that 75% of the film stress remains intrinsic.[2] We notice from Table 2 that the lattice dilation in the FeTaN film accounts for 2.69%. It suggests that the increase of lattice dilation of $\alpha$-Fe is a direct consequence of N addition to $\alpha$-Fe lattice. Some N atoms enter into interstitial sites between (110) planes to cause $d(110)$ lattice expansion. Therefore, the stress in the FeTaN film is compressive. For the FeAlN film, the lattice dilation $\delta d(100)/d(110)$ is negative and smaller than that of the FeTaN film. It means that no N atoms enter into $\alpha$-Fe lattice as interstitial atoms and the stress in the FeAlN film is tensile. This structure characteristic of the FeAlN film is proven by Mossbauer measurement. Figure 5 shows the conversion electron Mossbauer spectra (CEMS). This gives more detailed information about the microstructure in both films. By comparing the widths and intensities of lines in the spectra, we can draw the following conclusions:

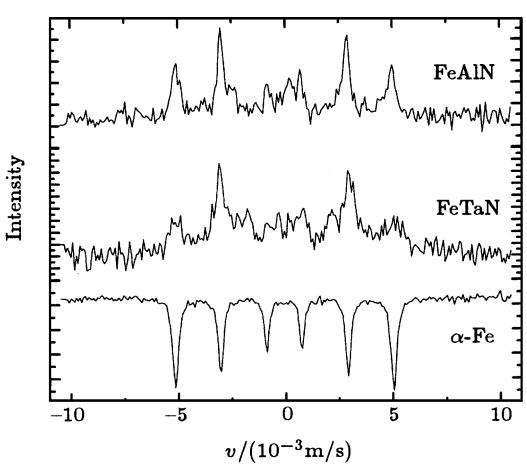



**Fig.5.** Conversion electron Mossbauer spectra of FeAlN and FeTaN films (upper: FeAlN films; lower: FeTaN films) and $\gamma$-ray transmission Mossbauer spectra of bulk Fe.

1. The spectrum of the FeAlN film has sharper and narrower lines than the FeTaN film spectrum, and the lines look more similar to those of the pure Fe foil in width than those in FeTaN spectrum. Therefore, the $\alpha$-Fe phase in the FeAlN film is purer than that in the FeTaN film.

2. If we "delete" pure Fe foil spectra from FeAlN or FeTaN film spectra, then we can see at "0" velocity some lines which belong to nonmagnetic phase. In the FeTaN film spectrum, the nonmagnetic phase is very small or equal to 0. It means there are some nonmagnetic phases in the FeAlN film spectrum.

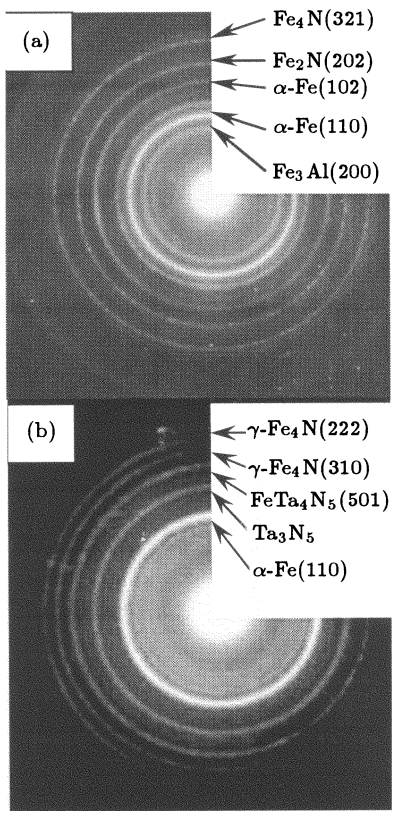



**Fig.6.** Selected area diffraction of TEM of FeAlN films and FeTaN films. (a) FeAlN films; (b) FeTaN films.

The structural features mentioned above are also confirmed by selected area diffraction (SAD) of TEM of both films in Fig.6. The difference in structures and magnetic properties can be interpreted by the chemical affinity of Ta and Al with N. The interaction coefficient $e_{\mathrm{N}}^{\mathrm{Al}} = 0.025$.[10] Al is easy to react with N to form nitrogen–aluminium compound, which is nonmagnetic and degrades the saturation magnetization, $B_{\mathrm{s}}$, and meanwhile, results in relatively low N-solubility in the FeAlN film. For the FeTaN film, the high interaction coefficient ($e_{\mathrm{Ta}}^{\mathrm{N}} = -0.035$) leads to lower tendency to form Ta–N compound and larger lattice dilation due to higher N-solubility. We know well that some N atoms fill in interstitial sites in the (110) plane to cause the anisotropy to increase,[11] and induced anisotropy

could have its origin in anisotropic ordering of defects (vacancies), pair ordering of atoms and anisotropy ordering of interstitial atoms. Classic studies of interstitial N in bulk Fe indicate that uniaxial anisotropy is the consequence of the anisotropic distribution in $\alpha$-Fe lattice.[12] Therefore, the uniaxial anisotropy in the FeTaN film shows induced anisotropy property and is the consequence of anisotropic ordering of interstitial atoms. Van De Riet and his colleagues pointed out that the binding axis between the Ta and N atoms determined the direction of induced anisotropy.[13] For the FeAlN film, there is no induced anisotropy resulting from anisotropic ordering of interstitial N because the $\alpha$-Fe phase in the FeAlN film is purer than that in the FeTaN film. The difference in uniaxial anisotropy may be due to the different stress states in both films.

## 4. conclusion

The soft magnetism of the Fe$X$N film comes from the incorporation of interstitial N atoms, which results in lattice expansion of $\alpha$-Fe and refines the grain size. The N atom incorporation strongly depends on the nature (i.e. chemical affinity) of added element $X$. Al is easy to react with N and form nitrogen-aluminium compound, which is nonmagnetic. Ta has low tendency to form metal–nitrogen and leads to high N-solubility resulting in large lattice dilation, compared with Al. Therefore, the FeTaN film has higher saturation magnetization. The uniaxial anisotropy is the consequence of the anisotropy distribution of N atoms in the $\alpha$-Fe lattice. For the FeAlN film, the $\alpha$-Fe phase is pure and there is no induced anisotropy resulting from anisotropic ordering of interstitial N atoms. In addition, the intrinsic stress of $\alpha$-Fe lattice in the FeAlN film is tensile and opposite to that in the FeTaN film. These differences in microstructures can account for the difference in uniaxial anisotropy.

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