

## Microstructural Investigations of FeN and FeAlN Thin Films for Recording Head Applications

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**Abstract**--A series of FeN and FeAlN thin films were grown by reactive rf sputter deposition. Planar and cross-sectional specimens were investigated in the TEM. We observed that introduction of nitrogen is associated with improved magnetic properties and decreased grain size and that the introduction of Al is associated with improved thermal stability of both the magnetic properties and the microstructure. We also investigated the effect of lamination and the role of the interlayer. We found the best magnetic properties in annealed films with five 1000Å thick FeN layers and four 25Å thick SiO<sub>2</sub> interlayers. High resolution TEM studies of cross-sectional specimens revealed an amorphous structure in this interlayer, providing crystallographic decoupling between the FeN layers. It is proposed that this SiO<sub>2</sub> layer also serves to reduce magnetic exchange coupling between layers.

### I. INTRODUCTION

Iron nitride (FeN) thin films have been proposed for use in high density recording heads [1,2]. For this application a material should have a high saturation magnetisation ( $4\pi M_s > 20\text{kG}$ ), a low coercivity ( $H_c < 1\text{ Oe}$ ), a low saturation magnetostriction ( $\lambda_s < 1 \times 10^{-6}$ ), a high permeability ( $\mu > 4000$ ) and good thermal stability. In previous work [3], we employed macroscopic studies to investigate how the magnetic properties of FeN films depend on growth parameters. We then presented work on iron aluminium nitride (FeAlN) thin films and on lamination [4,5]. Using these modifications we obtained both improved magnetic properties and improved thermal stability. In this paper we present results of TEM investigations of the microstructure, on a series of single and multilayer FeN and FeAlN films with systematically adjusted growth parameters. We then discuss how these results relate to the magnetic properties.

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### II. EXPERIMENTAL DETAILS

FeN thin films were grown by rf sputter deposition from a pure iron target onto glass substrates. Prior to film deposition, nitrogen and argon gas were bled into the system so that reactive sputtering would occur. The amount of nitrogen in the system was controlled using the bleed pressure and flow rates. FeAlN thin films were produced in the same manner with up to 4% of the surface of the pure iron target covered with aluminium. Macroscopic structural characterisation was conducted using x-ray diffraction. TEM studies were performed on ion-milled planar and cross-sections in Philips 420T and JEOL 4000EX electron microscopes. Magnetic characterisation was conducted using an M-H loop tracer, and  $\lambda_s$  was obtained by measuring the anisotropy field under external stress. Auger spectroscopy was used to study the film composition.

We investigated the effect of: 1) varying N<sub>2</sub>/Ar flow rate ratio up to 8% during film growth and then annealing, 2) introducing up to 4% Al and then annealing, 3) laminating with 5 FeN layers and 3 different interlayers: SiO<sub>2</sub> (25Å), CoZrRe (80Å) and NiFe (130Å) and then annealing.

For all films the total FeN/FeAlN thickness was 500nm. All annealing was performed *in vacuo* for 2 hours at 300C.

### III. RESULTS

1) For Fe films, with no N<sub>2</sub> introduced,  $H_c$  was  $> 5\text{ Oe}$  and  $4\pi M_s$  was approximately 22kG. For FeN films these figures dropped with increasing N<sub>2</sub>/Ar flow rate ratio to 20kG and 1 Oe, respectively. From TEM micrographs of FeN planar sections (figure 1) we observed that the grain size dropped rapidly from an average of approximately 50nm, for films with no nitrogen introduced, to an average of approximately 10nm for films with N<sub>2</sub>/Ar flow rate ratios between 2 and 8%. For the pure Fe film selected area electron diffraction (SAD) revealed two phases. The majority phase was bcc  $\alpha$ -Fe with a lattice parameter of  $2.87 \pm 0.02\text{Å}$  and the secondary phase was magnetite (Fe<sub>3</sub>O<sub>4</sub>), probably due to a surface oxide layer. No strong crystallographic texturing was observed. With introduction of N<sub>2</sub>, while bcc  $\alpha$ -Fe was still the majority phase, we also observed the  $\gamma$ -Fe<sub>4</sub>N (Pm $\bar{3}$ m)

phase (figure 2). The amount of this phase was seen to increase with increasing  $N_2/Ar$  flow rate ratio, as did  $\lambda_s$ . Dark field imaging failed to reveal a particular microstructural feature with which the secondary phases could be associated.

Annealing produced a small decrease in both  $H_c$  and  $\lambda_s$  ( $H_c$  to 0.9 Oe and an  $\lambda_s$  to  $\sim 10^{-6}$ ), and an increase in grain size of up to a factor of two. SAD revealed no large change in the relative amounts of each phase.

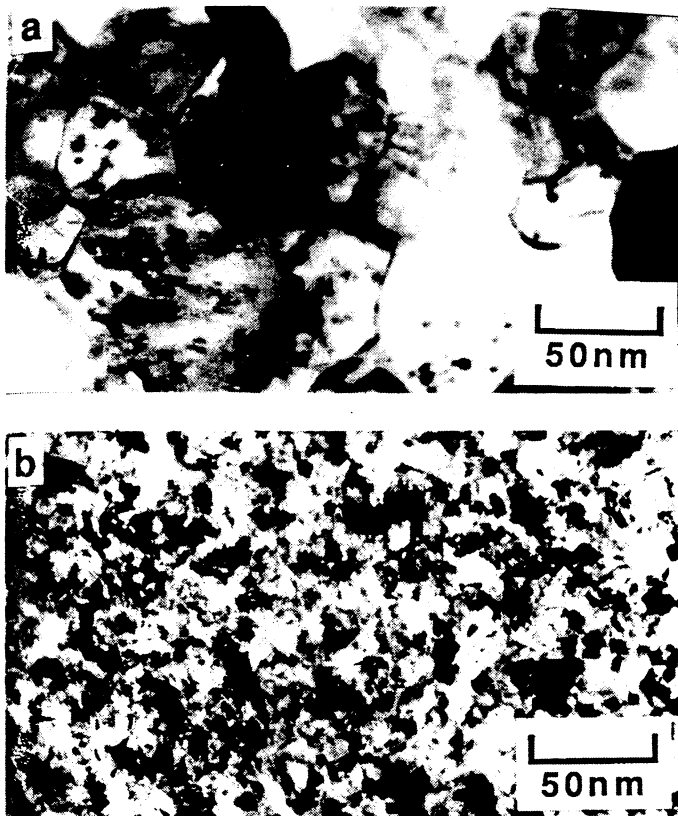


Figure 1 Bright field TEM micrograph of planar section of a) Fe thin film and b) FeN thin film grown with nitrogen/argon flow rate ratio of 4.5 %.

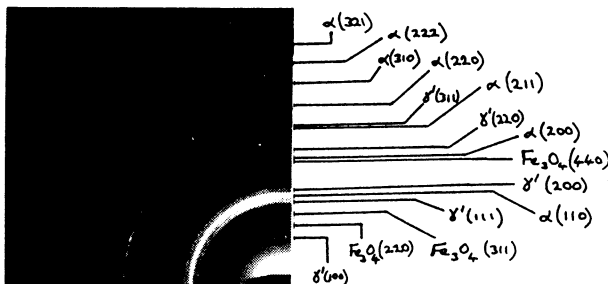


Figure 2 TEM selected area diffraction pattern showing the bcc  $\alpha$ -Fe,  $\gamma$ - $Fe_4N$  ( $Pm\bar{3}m$ ) and magnetite ( $Fe_3O_4$ ) phases.

2) For the FeAlN films the average grain size was between 10 and 15 nm for Al concentrations of up to 4%. Bcc  $\alpha$ -Fe was the majority phase and the amount of  $\gamma$ - $Fe_4N$  was seen to decrease with increasing Al concentration. The magnetic properties of FeAlN show a similar dependence on  $N_2/Ar$  flow rate ratio to the FeN films, but we see a lower  $H_c$  for near-zero  $\lambda_s$  than in FeN films ( $H_c = 0.5$  Oe for  $\lambda_s \sim 10^{-6}$ ). There were no distinct differences in microstructure apparent (including grain size) between samples before and after annealing (figure 3). This confirmed the proposition that improved stability of the magnetic properties in FeAlN as compared to FeN was associated with improved thermal stability of the microstructure.

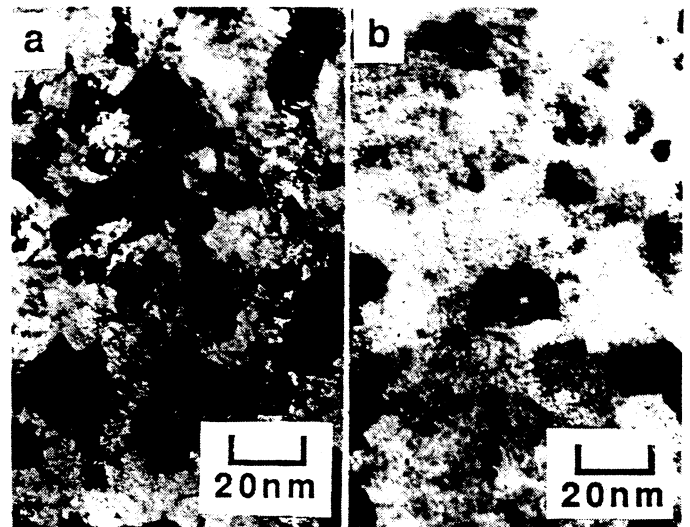


Figure 3 TEM micrographs showing planar specimens of FeAlN thin films a) before and b) after annealing.

3) High resolution TEM studies of cross-sectional FeN multilayer specimens revealed that both the  $SiO_2$  and the CoZrRe interlayers were amorphous while the NiFe interlayer was crystalline (figure 4). It is important to note that the  $SiO_2$  interlayer is the only one of these three which is non-ferromagnetic in nature [6]. We found that the average grain size was slightly smaller for the sample with  $SiO_2$  interlayers and that the grain size for all the samples increased with annealing. For all the films the interlayer was observed to seal the individual layers with no discontinuities for FeN grains to pass through. The growth of the FeN grains was arrested by the  $SiO_2$  interlayer whereas there appeared to be some correspondence of grain structure above and below the interlayer for the other two interlayer materials (figures 5 & 6). Many FeN grains appeared to extend through the entire thickness of the FeN layers for films with either the CoZrRe or NiFe interlayers. FeN grains in the film with  $SiO_2$  interlayers, however, were much reduced in extent. The

annealed film with the SiO<sub>2</sub> interlayers had the lowest H<sub>c</sub> for

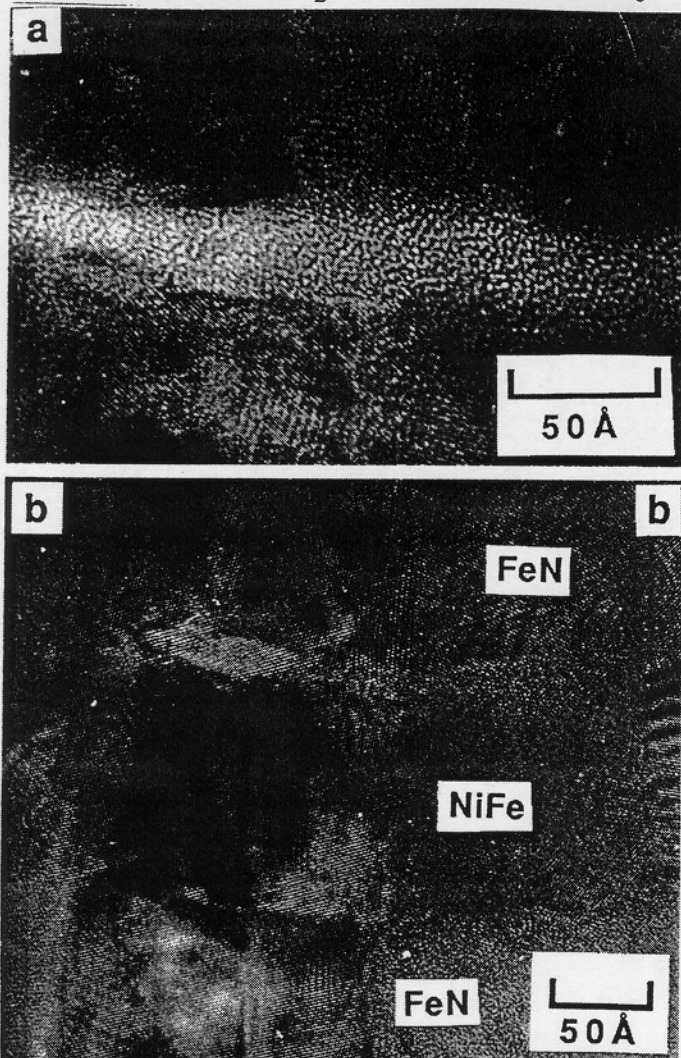


Figure 4 High resolution TEM micrographs showing a) amorphous SiO<sub>2</sub> interlayer decoupling individual FeN layers and b) crystalline NiFe layer with FeN grains above and below linked epitaxially through the interlayer.

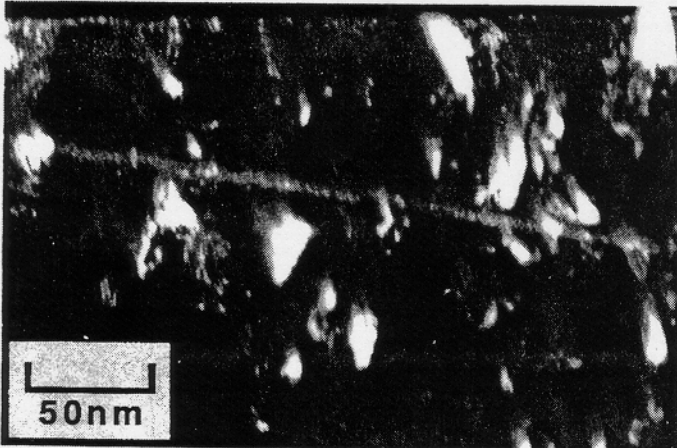


Figure 5 Dark field TEM micrograph of cross-sectional specimen of FeN multilayers with SiO<sub>2</sub> interlayers.

near zero  $\lambda_s$  ( $H_c = 0.4$  Oe for  $\lambda_s \sim 5 \times 10^{-7}$ ) and the annealed film with the CoZrRe interlayer had the highest H<sub>c</sub> (1 Oe).

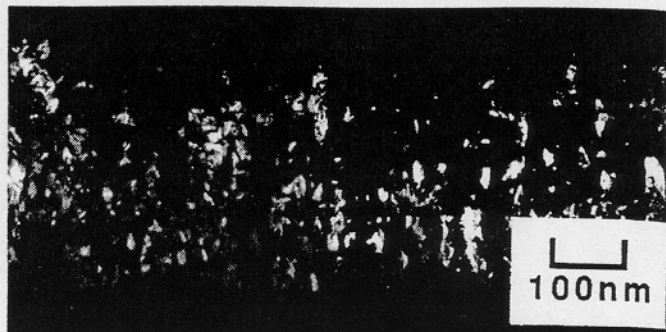


Figure 6 Dark field TEM micrograph of cross-sectional specimen of FeN multilayer with NiFe interlayers.

#### IV. CONCLUSIONS

We have shown that when N<sub>2</sub> is introduced in the sputtering process there is a marked reduction in both grain size and H<sub>c</sub>. As more N<sub>2</sub> is introduced there is an increase in the amount of  $\gamma$ -Fe<sub>4</sub>N phase [3]. Subsequent annealing at 300C is associated with a general increase in grain size and a slight reduction in H<sub>c</sub>. For FeAlN films we observed a decrease in the amount of  $\gamma$ -Fe<sub>4</sub>N phase with increased N<sub>2</sub> [4]. While H<sub>c</sub> decreases on annealing as for FeN, no change in microstructure has been observed. For the multilayer structure, the SiO<sub>2</sub> decouples the individual layers crystallographically and may also serve to reduce exchange coupling between layers [6].

Thus we have shown for FeN and FeAlN thin films that microstructural features, such as grain size, amount of certain phases and continuity of grains through an interlayer, may influence the magnetic properties. In this way we have gained more information on the nature of the mechanisms controlling the magnetic properties of FeN and FeAlN thin films.

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