The Effect of Annealing on the Microstructure and Magnetic Properties of Fe-Ta-Cr-N Thin Films

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Abstract — **The effect of T_{anneal} on Fe-Ta-Cr-N thin films has been studied. Crystallization of Fe was found to precede TaN. Results showed that T_{anneal} controls not only the grain size but also the amount of Ta and N which is in solution within the Fe matrix and the magnetic properties was determined largely by these two factors. In addition, TaN was found to nucleate both homogeneously and heterogeneously, according to the K-S OR, with respect to the Fe grain.**

I. INTRODUCTION

It has been demonstrated that nanocrystalline Fe thin films adorned with non-magnetic intermetallic carbides or nitrides at the grain boundaries possess excellent soft magnetic properties and are suitable for high density metal-in-gap(MIG) recording head applications[1]. The as-deposited film is amorphous and nanocrystallinity is obtained through post-deposition annealing, wherein Fe grain growth is restricted by the fine network of intermetallic particles. The family of Fe-X-N(X=Ta,Nb,Hf,Zr) films has been studied extensively [2-4]. The saturation flux density (B_s), coercivity (H_c), permeability (μ) and magnetostriction (λ_s) have been shown to be very sensitive to the annealing temperature (T_{anneal}) and chemical composition, which together control the microstructure. However, the understanding of the nucleation and growth of such systems is still incomplete. This work examined the microstructure of Fe-Ta-Cr-N films at different T_{anneal} in order to better address the correlation between structure and properties in these systems.

II. EXPERIMENTAL PROCEDURES

$Fe_{73.5}Ta_{10.0}N_{14.0}Cr_{2.5}$ films, 2μm thick, were deposited onto $CaTiO_3$ substrates by reactive RF magnetron sputtering. The total gas pressure was 0.3 Pa with a N_2/Ar partial pressure ratio of approximately 0.1. The films were annealed in N_2 for 30min at temperatures between 400 and 650°C. The magnetic properties were measured with a VSM. λ_s was obtained by the cantilever beam method. The specimens annealed at 400°C, 560°C and 650°C, hereafter designated as films A, B and C, were examined with both conventional (Philips 420T) and high resolution (JEOL 4000EX) transmission electron microscopy (HRTEM).

III. RESULTS AND DISCUSSIONS

The magnetic properties of the three specimens are listed in Table 1. B_s is enhanced with increasing T_{anneal}, whereas μ initially increases with T_{anneal} but declines rapidly as the temperature is raised beyond 560°C. H_c is below 1 Oe for all specimens except the one annealed at 650°C. The relationship between λ_s vs T_{anneal} is plotted in Figure 1. λ_s switches from positive to negative as the temperature is raised and a zero crossing is registered at 520°C.

TABLE 1.
The magnetic properties of Fe-Ta-Cr-N films annealed at 400°C, 560°C and 650°C.

Film	A	B	C
T_{anneal}	400°C	560°C	650°C
B_s	1.32T	1.56T	1.56T
H_c	0.6Oe	0.5Oe	3.5Oe
μ	3000	4500	250

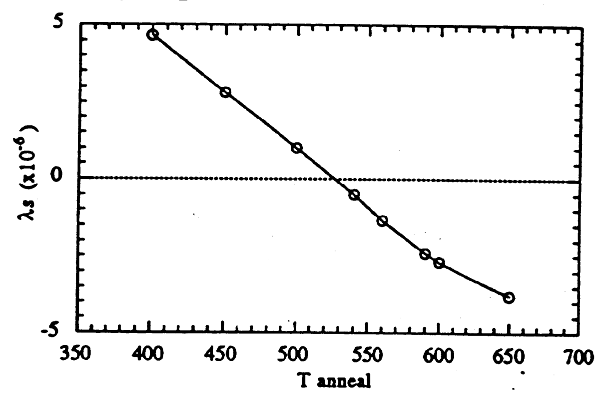

Figure 1. Plot of λ_s vs T_{anneal}.

In the x-ray spectra of films A and B shown in Figure 2, TaN peaks are indistinguishable from the background and the Fe peaks are very weak. From the position of the Fe (110) peak in film A's spectrum, the interplanar spacing was calculated to be around 2.08Å. In film C, distinctive peaks from Fe and TaN can be observed in the diffraction spectrum reflecting an improvement in crystallinity within the film. The Fe (110) interplanar spacing is now 2.03Å, which is same as pure bcc Fe.

The corresponding selected area diffraction (SAD) patterns for the three specimens are shown in Figure 3. At 400°C, the SAD pattern already shows bcc Fe rings. However, one can also find a thick amorphous ring (arrow Figure 3a) around the Fe (110) ring which indicates a mixture of amorphous and crystalline phases co-existing in the film. In film B, the amorphous ring is no longer present but a faint TaN (111) ring can be observed inside the Fe (110) ring. Thus,

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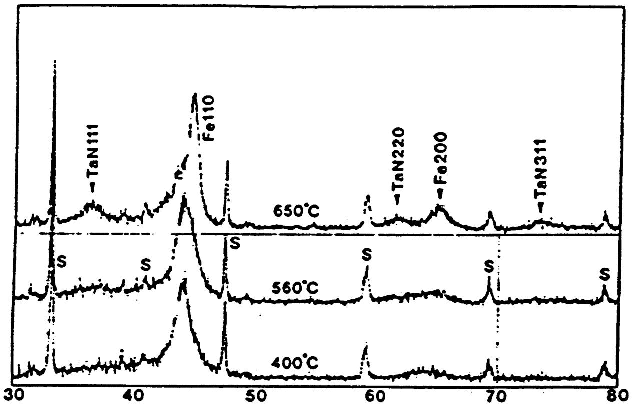

Figure 2. X-ray diffraction spectra of Fe-Ta-Cr-N films annealed at 400°C, 560°C and 650°C. (S:substrate)

crystallization is completed and TaN has begun to form. At 650°C, the TaN (111) (arrow Figure 3c) ring has increased in both intensity and sharpness, revealing an increase in the volume fraction and the size of TaN. The bright field (BF) images of these three films are shown in Figure 4. The

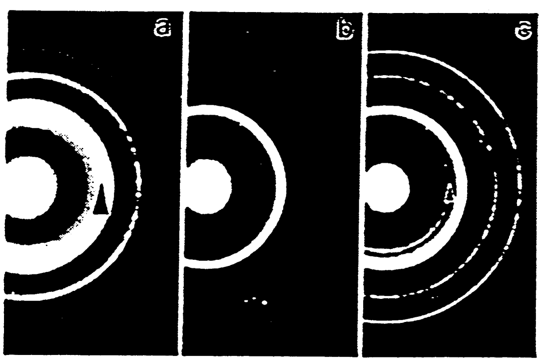

Figure 3. SAD patterns of Fe-Ta-Cr-N films annealed at (a) 400°C, (b) 560°C and (c) 650°C.

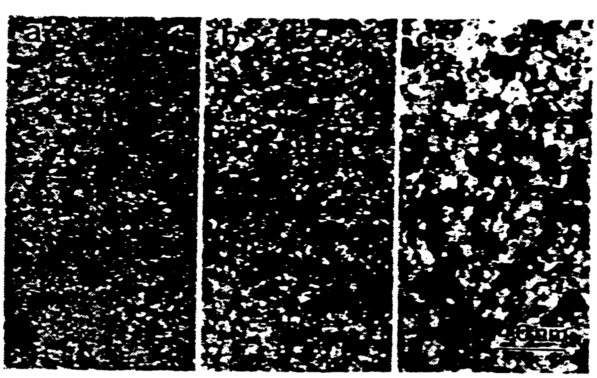

Figure 4. BF images Fe-Ta-Cr-N films annealed at (a) 400°C, (b) 560°C and (c) 650°C.

grains in film A have weak diffraction contrast and the estimated grain size is around 4nm. In film B, the grain size has increased because of higher T_{anneal} and it now ranges from 4nm to 16nm. In these two films, the grain boundaries are not well defined. At 650°C, the grains have a well developed morphology and their size averages to 22nm. The uniformity is probably a result of recrystallization of the smaller Fe grains and restriction of Fe grain growth by TaN particles. The behavior of H_c with respect to T_{anneal} can thus be explained by the variation in grain size as fine grains are necessary for low coercivity in soft magnetic films. In addition, μ is also very sensitive to the grain size since high permeability is favored by small grains. The well defined grain structure in film C may have caused in a reduction in intergranular exchange coupling between Fe grains and resulted in the degradation of soft magnetic properties.

A HRTEM image of film A is shown in Figure 5. Fe nanocrystals are present along with some amorphous contrast. The Fe (110) interplanar spacing was measured to be 2.09Å, higher than the equilibrium value of 2.03Å. Furthermore, no TaN grains can be observed through out the specimen. This, along with the x-ray spectra, suggests that Ta, and also N, is in solution within the Fe matrix thus expanding the Fe lattice. The incorporation of non-magnetic

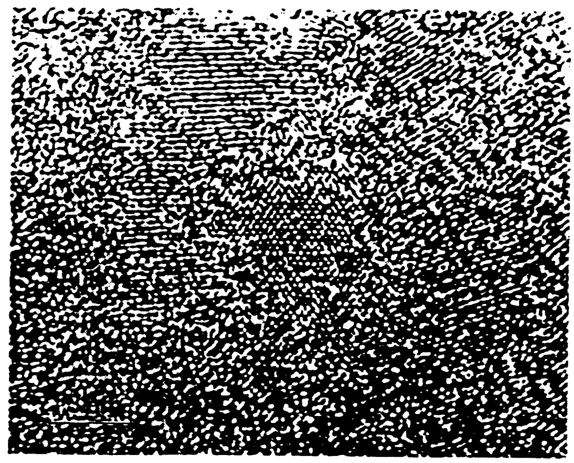

Figure 5. HRTEM image of the film Fe-Ta-Cr-N annealed at 400°C.

elements resulted in a lower B_S. In film B (Figure 6), small TaN particles, measuring 1.2nm to 3.5nm, are observed throughout the film. One such TaN nanocrystal can be found in region B, situated next to a Fe particle in the lower left hand corner of Figure 6a. In the upper right hand corner, local expansion within the Fe grain lattice can be seen in region P. Two of the lattice spacings within this expanded regime were measured to be 2.32Å and 2.10Å (Figure 6b), which is larger than bcc Fe {2.03Å for (110) and $(1\bar{1}0)$} but smaller than the equilibrium spacing for fcc TaN {2.50Å for (111) and 2.19Å for (200)}. This may have been caused by a local partitioning of Ta and N from the Fe matrix as the diffusivity becomes favorable at higher T_{anneal}, which along with the larger undercooling, leads to homogeneous nucleation of TaN within the Fe lattice. Since the lattice mismatch between Fe and TaN is relatively large, a compressive distortion of TaN lattice is induced. This large strain field will restrict the growth of such homogeneously nucleated TaN particles and seriously deform the lattice surrounding this nuclei as observed in Figure 6b. The orientation relationship (OR) between the two lattices is that of Kurdjimov-Sachs[5] (K-S): $(1\bar{1}0)_{TaN}//(\bar{1}11)_{Fe}$, $[111]_{TaN}//[110]_{Fe}$ (Figure 7). The TaN (111) and Fe (011) is at a 60° angle with respect to each other. This is the same angle measured between TaN and Fe in region B which indicates that TaN also nucleated heterogeneously on Fe according to the K-S OR. Such OR is known to minimize the interfacial energy and can explain the selection of fcc TaN over the equilibrium hcp TaN.

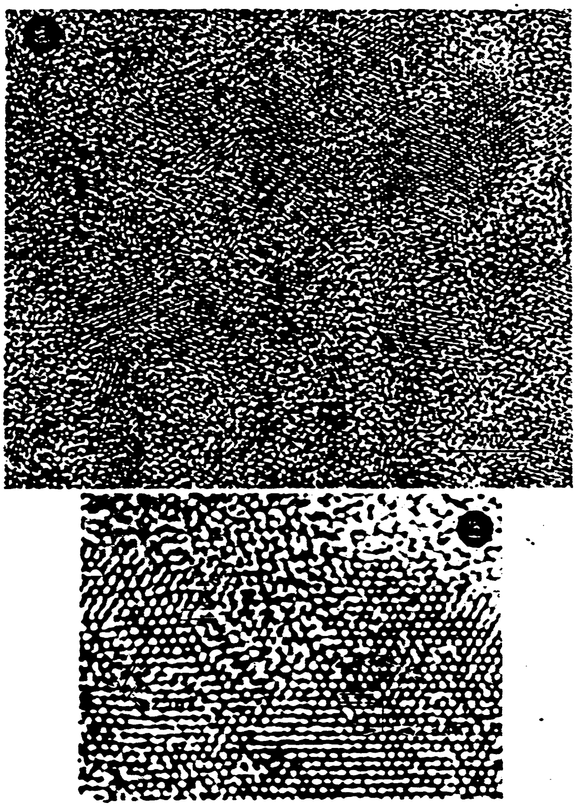

Figure 6. (a) HRTEM of the film Fe-Ta-Cr-N annealed at 560°C. (b) a magnified images of area P in (a).

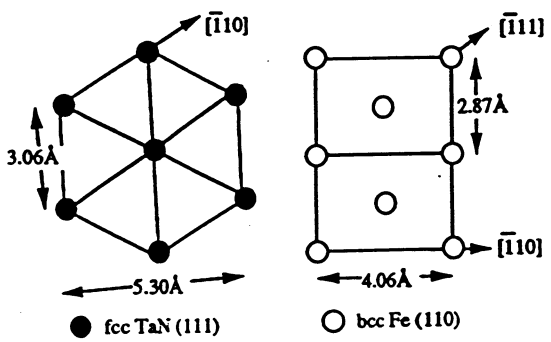

Figure 7. Schematic of the OR between fcc TaN and bcc Fe.

In film C, a network of TaN particles can be found throughout the film. They are mostly located at triple grain junction as shown in Figure 8. The Fe grains have well defined boundaries and the Fe (110) interplanar spacing now resumes the equilibrium value of 2.03Å. This suggests the complete partition of Ta and N from the Fe matrix which brought higher B_s. Furthermore, it also shows that the λ_s is very sensitive to the composition of the Fe matrix. As T_{anneal} is raised, the amount of non-Fe element incorporated within the Fe matrix after annealing is progressively less. At 520°C, the optimum chemical composition was probably achieved and resulted in zero magnetostriction.

Figure 8. HRTEM of film Fe-Ta-Cr-N annealed at 650°C.

IV. Conclusions

We have conducted microstructural investigations on Fe-Ta-Cr-N films. Fe particles formed before TaN during the annealing process. Finer grain size leads to lower H_c and higher μ. On the other hand, B_s and λ_s are very sensitive to the chemical composition (Ta,N) within the Fe matrix which changes with T_{anneal}. TaN has been observed to nucleate both homogeneously within the Fe matrix and heterogeneously at the Fe grain boundaries. It was found that fcc TaN assumes the K-S OR with respect to bcc Fe thus making its formation more favorable with respect to the equilibrium hcp phase.

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