Microstructural study on multilayer \([\text{FeTaN}/\text{TaN}]_5\) films

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Abstract

The microstructure of \([\text{FeTaN}/\text{TaN}]_5\) multilayer films has been investigated by transmission electron microscopy (TEM) and high-resolution electron microscopy (HREM) in cross section and plan view. Each layer shows a small surface roughening less than 1 nm. The FeTaN layers are composed of \(b.c.c.\) Fe with Ta incorporated substitutionally and N interstitially, denoted as \(\text{Fe(Ta,N)}\); while the TaN layers mainly consist of \(f.c.c.\) TaN phase. A \(\{110\}\) texture of \(\text{Fe(Ta,N)}\) has been formed in the FeTaN layers. The columnar grain structure is a typical feature in FeTaN layers.

Keywords: Microstructure; Thin films; Magnetic materials; Transmission electron microscopy (TEM); Sputtering

1. Introduction

The density of stored information in magnetic recording systems has been increasing gradually with a corresponding increase in the coercivity of the media, thus a fast improvement in recording heads is required. The ideal recording head material requires optimal properties such as high saturation magnetization, low coercivity, suitable thermal stability and small magnetostriction. Though permalloy (Ni–Fe) film has been widely used as magnetic recording head material in the industry, its relatively low saturation flux density limits the choice of media and therefore the recording density \([1,2]\). The Fe–N film has very high flux density, but it has the disadvantages of high magnetostriction and poor thermal stability \([3]\). Third elements, such as Ta, Al, Hf, were then added to form ternary Fe–X–N (X=Ta, Al, ...) films. Because of the exceptional combination of high saturation magnetization and excellent soft magnetic properties, Fe–X–N films as a potential head material for high-density magnetic recording have received considerable attention \([4–10]\). Recently, improved magnetic properties over single layer films have been found in multilayers, such as \(\text{FeAlN/SiO}_2(\text{Al}_2\text{O}_3)\) \([11]\), \(\text{FeTaN}/\text{TaN}\) \([12, 13]\). They have lower coercivity in multilayers with nonmagnetic interlayers \([11]\) or enhanced high-frequency permeability with insulating interlayers due to a reduction of eddy currents \([14]\) as compared to single layer films of the same materials. Researchers...
have made great efforts on films preparation as well as optimization of the high saturation soft magnetic properties and encouraging progress have been obtained. However, reports on the related microstructure, especially the detailed microstructure at atomic scale, were very limited. Since the microstructure may strongly influence magnetic properties of these materials, it is necessary to study the detailed microstructure of the multilayer films. In the present work, the microstructure of [FeTaN/TaN]5 multilayer films prepared by reactive radio frequency (RF) magnetron sputtering deposition technique were characterized using high-resolution transmission electron microscopy and associated techniques.

2. Experimental procedure

The [FeTaN/TaN]5 multilayers were synthesized on (100) oxidized Si substrate using reactive RF magnetron sputtering on a Denton Vacuum Discovery-18 Deposition System [15]. A 3-in. Fe target with Ta chips that cover about 2.0–3.0% of the efficient Fe target surface area was used for FeTaN layers. A mixture of pure Ar/N2 was induced in the deposition chamber and the total pressure of processing ambient gas was kept at 2.0 mTorr. The percentage of N2 partial pressure $P_N$ in the Ar/N2 ambient gasses was varied for different layers. For FeTaN layers, $P_N$ was set at 3.0% and the RF power was kept at 100 W. For TaN layers, $P_N$ was set at 20.0% and the RF power was kept at 70 W.

The cross-section and plan-view specimens suitable for TEM and high-resolution electron microscopy (HREM) observations were prepared by standard techniques. For the cross-sectional specimen, it was cut into $3 \times 0.5$ mm strips along the [001] direction of Si by a diamond low speed saw. Two pieces of Si substrate carrying (FeTaN/TaN)$_5$ multilayer films were then glued together with the film surfaces facing each other using M-bond 610 adhesive and clamped till the epoxy was cured. The cured specimens were carefully grinded mechani-

![Fig. 1. (a) Low magnification cross-sectional image of [FeTaN/TaN]$_5$ films grown on (100) Si substrate. Higher magnification cross-sectional (b) bright-field and (c) dark-field images of the multilayers. (d) The corresponding selected area electron diffraction pattern. The four strong spots inside the (110)$_{Fe(Ta,N)}$ ring were {111} diffraction of Si substrate.](image-url)
cally to a thickness of about 70 μm, followed by dimpling in a Gatan dimpler. The ion-beam milling was done by Ar\(^+\) bombardment. Moreover, to protect the interface and to reduce the effect of the different milling rates of Si and FeTaN/TaN system, shields were used during thinning (see, e.g. Ref. [16] for a schematic). The plan-view specimen was made parallel to the (100) Si plane and thinned from the substrate side.

The specimens were examined in a JEM 2010 high-resolution transmission electron microscope (TEM) with a point to point resolution of 0.19 nm. Chemical composition of the films was analyzed using a HF 2000 cold field-emission-gun TEM equipped with an Oxford Link energy dispersive spectroscopy system (EDS). The microscopes were operated at 200 kV.

3. Results and discussion

The low magnification cross-sectional image of [FeTaN/TaN]\(_5\) multilayers is shown in Fig. 1a. The clear contrast difference between the FeTaN and TaN layers results from the scattering and absorption effects for different atoms. The average thickness of FeTaN and TaN layers is 30 and 5 nm, respectively. In order to reveal more clearly the growth characteristic of the film, high magnification cross-sectional bright-field and dark-field images are given in Fig. 1b,c, respectively. A columnar grain structure in the FeTaN layers was observed with an average diameter of 12 nm. Moreover, the bright contrast between the multilayers and the substrate, as indicated by an arrow in Fig. 1b, represents the oxide layer of Si.

Fig. 1d is the corresponding selected area electron diffraction (SAED) pattern taken from the cross-sectional FeTaN/TaN multilayers and the Si substrate. Only b.c.c. Fe and f.c.c. TaN phases were identified in the multilayers. Due to the relatively low N\(_2\) partial pressure used during the preparation of the FeTaN layers, no iron nitride has been formed. The average lattice parameter of b.c.c. Fe was determined to be 0.2889 nm by the internal calibration with the Si substrate, which is 0.79% larger than that of bulk b.c.c. Fe with \(a=0.28664\) nm. It is known that the

![Fig. 2. HREM images of the FeTaN/TaN multilayer films in cross section, showing f.c.c. TaN grains in TaN layers and Fe(Ta,N) grains in FeTaN layers with (a) [001] and (b) [111] directions of Fe(Ta,N) grains parallel to the electron beam.](image-url)
atomic radius of Ta is larger than Fe and it would substitute into the Fe lattice in sputtered FeTaN thin films. Moreover, N would occupy the octahedral interstitial sites of b.c.c. Fe in FeTaN films. As a result, both the interstitial N and substitutional Ta act to expand the lattice of b.c.c. Fe. The solid solution of b.c.c. Fe with Ta and N will be referred to as Fe(Ta,N) hereafter.

Previous researches showed that metastable f.c.c. TaN phase with NaCl type can be obtained only at high temperature, e.g. 1700 °C, in a bulk TaN system prepared by heating Ta in a high pressure of N2 [17], but the f.c.c. TaN phase can be obtained easily in sputtered thin films under suitable preparation conditions, such as relatively high N2 partial pressure. During sputtering deposition, it is high energetic ions in the plasma generated by the RF that supply the energy required for forming the f.c.c. TaN phase [18].

It is worth to note that the distribution of diffraction spots in the {110} ring of Fe(Ta,N) is not even. The ring has strong intensities parallel to the film normal direction [100] and 60° away from the normal, as indicated by arrows, revealing the presence of ⟨110⟩ texture in the FeTaN layers.

Fig. 2a is a typical cross-sectional HREM image of the multilayers. Each layer shows a small surface roughening less than 1 nm. Two grains of Fe(Ta,N) with their [001] direction parallel to the electron beam are observed in the FeTaN layer. Both of them exhibit obvious columnar growth along ⟨110⟩ direction. The tilting angle of the two grains is about 15° as indicated by two ⟨110⟩ planes. TaN grains with f.c.c. structure were also clearly observed in the TaN layers with ⟨110⟩ axis parallel to the incident electron beam. Fig. 2b gives another cross-sectional HREM image of the multilayers, showing a Fe(Ta,N) grain with its ⟨111⟩ direction parallel to the electron beam. Again, its ⟨110⟩ direction is nearly parallel to the film normal.

According to the results of cross-sectional HREM and SAED given above, it is thus clear that Ta and N formed a solid solution with b.c.c. Fe in the FeTaN layers while the TaN layers consisted of f.c.c. TaN phase.

Fig. 3a shows a typical plan-view dark-field image of the multilayer films. The corresponding EDP is shown in Fig. 3b. Except Fe(Ta,N) and f.c.c. TaN, there is no indication of other nitrides in the EDP, which is consistent with the results of cross-sectional observations.

The coercivity values of the multilayers may be found in our previous work (Fig. 2a of Ref. [13]), and a decreased coercivity (less than 2.0 Oe) was achieved in our multilayer films [13]. It is known that grain size is an important parameter of films. Microstructure with fine crystals has been confirmed as an effective means to obtain films with improved soft magnetic properties [1,3,19]. The plan-view dark-field image of the films (Fig. 3a) reveals that the grain size is small, about 25 nm. As shown by plan-view HREM image (Fig. 4), however, the factual grain size is even smaller, because the apparent grains in the dark-field image are grain clusters,
in fact. A cluster consists of several smaller grains, which have similar orientations or diffraction conditions, and therefore display similar contrast. The HREM images of such clusters are shown in Fig. 4, where the close-packed planes of \((110)_{\text{Fe(Ta,N)}}\) are indicated. The dashed lines represent the grain boundaries. The three Fe(Ta,N) grains in Fig. 4a have very similar orientation, and contact with each other with low-angle grain boundaries. The three grains in Fig. 4b form large-angle grain boundaries. The similar cluster structure was also observed in polycrystalline Fe–N films [20].

In order to image the TaN layer, very thin specimen is required, since the top TaN layer is only 5 nm in thickness. However, the thickness of the thinnest part of specimens thinned by ion-beam milling is usually larger than 5 nm. Therefore, even at the edge part of the specimen, the observed plan-view specimen certainly contains both TaN and

Fig. 4. HREM images of two clusters, consisting of Fe(Ta,N) grains. (a) Three Fe(Ta,N) grains contact with each other with low-angle grain boundaries. (b) Three Fe(Ta,N) grains form large-angle grain boundaries.

Fig. 5. Plan view HREM image of a thinner area, showing small grains of Fe(Ta,N) and f.c.c. TaN.
FeTaN layers. Fig. 5 gives a plan-view HREM image of a thinner area, showing small grains of Fe(Ta,N) and f.c.c. TaN.

The composition of the FeTaN layers was also analyzed by nanometer-beam EDS with a spatial resolution of 2–3 nm. The typical result is given in Fig. 6. Quantitative analysis showed that the content of Ta in the FeTaN film is about 7 wt.%. Quantitative N content was not pursued because the N peak cannot be resolved from that of C resulted from the contamination during EDS analysis.

4. Conclusions

A microstructural characterization of sputtered [FeTaN/TaN]$_5$ multilayer films was performed by TEM and HREM. Each layer shows a small surface roughening less than 1 nm. A columnar grain structure in the FeTaN layers was clearly observed. Ta and N formed a solid solution with b.c.c. Fe in the FeTaN layers while the TaN layers consisted of f.c.c. TaN phase. Grain clusters consisting of small grains are present in the plan-view observations containing both FeTaN and TaN layers.

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References