49th MMM Conference, Nov. 2004, Jacksonville (USA)

paper code: ED-02

Preparation of high moment CoFe films with controlled grain size and coercivity

M. Vopsaroiu[1], M. Georgieva[2], P.J. Grundy[2], G. Vallejo Fernandez[1], S. Manzoor[1], M.J. Thwaites[3], K. O'Grady[1]

[1] Department of Physics, The University of York, York, YO10 5DD, UK

[2] Department of Physics, University of Salford, M5 4WT, UK

[3] Plasma Quest Ltd., Hook, RG27 9UT, UK

In this paper a new preparation method for high moment CoFe thin films with soft magnetic properties is reported. A full control of coercivity in a series of 20 nm thick CoFe films has been achieved without using seed layers, additives or thermal annealing. The films were sputtered directly onto Si substrates and the coercivity was varied by changing the mean grain size in the sputtered films. The mean grains size was in turn controlled via the sputtering rate. A reduction in the coercivity has been observed from 120 Oe for samples with mean grain size larger than 17 nm down to 12 Oe for a sample with a mean grain size of 7.2 nm. The results are in good agreement with the "random anisotropy model" relating the coercivity to the mean grain size in polycrystalline ferromagnetic films.

I. INTRODUCTION

High moment CoFe alloy films are essential for applications such as soft underlayers in perpendicular media [1] and as the core material of write elements in modern recording heads [2,3]. However, CoFe thin films do not naturally exhibit soft magnetic properties and usually seed layers or additives are used to reduce the coercivity. Thompson et al. [4] found that CoFe films grown on Au/MgO seed layers exhibited soft magnetic properties with a coercivity of 16 Oe. Similar results were reported by Platt et al. [5] for CoFe films grown on CoO with H_c as low as 12 Oe. This reduction in H_c was related to a reduction in the grain size from typically 20-35 nm to 5-15 nm. Recently, the use of several seed layers to produce soft CoFe films was reported by Jung et al. [6]. They showed that a Cu underlayer as thin as 2.5 nm could reduce the coercivity of 50 nm CoFe films to 12 Oe from 120 Oe in samples without the seed layer. They extended the studies to other seed layers and similar results were found for Ru, NiFe and Ta/NiFe underlayers. Again, the reduction of the coercivity was related to a reduction in the grain size of the CoFe films.

In the present study we report the preparation of soft 20 nm CoFe films without the use of seed layers or additives. A reduction in coercivity from 120 Oe down to 12 Oe has been obtained in agreement with the previously published work [6]. This has been achieved by directly controlling the grain size through the plasma sputtering process. Our objective is to relate the growth process to the magnetic properties rather than to produce soft underlayers themselves.

II. EXPERIMENT

CoFe thin films were prepared using a special plasma sputtering technology (HiTUS) [7], which has the capability to control the grain size in sputtered polycrystalline thin films [8]. This is due the design of the system that allows close control of the sputtering rates via the deposition parameters [7]. Our previous work [8] has shown that three parameters affect the grain size in films produced using the HiTUS system. These are the sputter gas pressure, the RF power and the DC bias. We have shown that each of these affects the growth rate and that it is the growth rate that controls the grain size. Due to the ability of the HiTUS system to vary the growth rate over a wide range of values close control of grain size is possible.

A set of seven CoFe samples was sputtered directly onto Si substrates without using seed layers. A magnetic field of 100 Oe was applied during deposition to induce a uniaxial magnetic anisotropy. All films were sputtered after pumping to a base pressure of 5 x 10^{-7} mbar with an Ar process pressure of 2.7 x 10^{-3} mbar. The RF power used to generate the plasma in the HiTUS system was also constant at 1.75 kW. Prior to each deposition, target and substrate plasma cleaning were applied to eliminate contamination or oxide layers. Substrate heating was not employed and during sputtering the substrate temperature lay in the range of 60 - 100^0 C.

It is well known that grain size in sputtered polycrystalline films is related to the growth rate [9]. Hence, in order to change the grain size, the sputtering rate was varied via the DC bias voltage (Table 1). The thickness of the films was kept constant at 20 nm while the sputtering rate ranged between 0.1 to 0.8 Å/s (Table 1). However, these rates are characteristic of our sputtering plant which is a research laboratory unit utilizing 5 cm diameter target with a target to substrate separation of 25 cm. Using larger targets (10 cm), sputtering rates as high as 35 Å/s can be achieved.

Table 1. growth conditions and sample specifications

Sample	Bias Voltage (- V)	Growth rate (Å/s)	Mean grain size D (nm)	Coercivity H_c (Oe)
A1	120	0.1	7.20	12
A2	200	0.2	14.1	20
A3	300	0.3	17.1	120
A4	400	0.4	20.0	126
A5	600	0.55	21.2	117
A6	800	0.7	24.1	108
A7	1000	0.8	26.3	121

Grain size measurements were performed using TEM imaging of grids that were attached to each substrate. TEM images were acquired for each sample in bright field mode at 120 kV and x150k magnification. The mean grain size was obtained by measuring and counting over 500 particles for each sample using a Zeiss particle size analyzer. Typical TEM images and size distributions for samples A1 and A7 are shown in figures 1 and 2. The crystal structure was determined using X ray diffraction and was found to be BCC with a dominant (110) component. Magnetic hysteresis properties were measured using a VSM.

III. RESULTS AND DISCUSSIONS

Figures 1 and 2 show examples of TEM images and the corresponding distributions. These data are for the samples with the smallest (A1) and the largest (A7) mean grain size respectively. The data has been fitted to log-normal distributions and the mean grain size determined from the fit (Table 1). A change of more than a factor 3 in the mean grain size is observed as a function of the sputtering rate. Mean grain diameters as small as 7.2 nm have been achieved at a sputtering rate of 0.1 Å/s. At a sputtering rate of 0.8 Å/s a mean grain size of 26.3 nm resulted. Figure 3 shows the change in the mean grain size as a function of the sputtering rate for all samples.

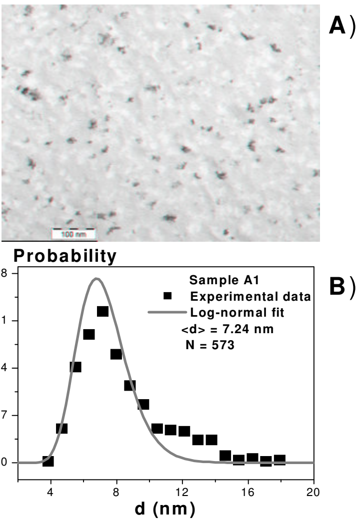

Figure 1. A) TEM image of sample A1. B) Typical distribution and log-normal fit

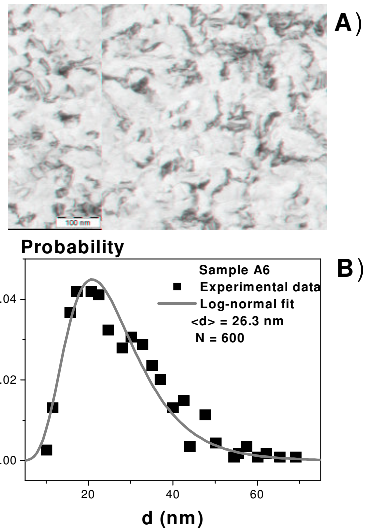

Figure 2. A) TEM image of sample A7. B) Typical distribution and log-normal fit

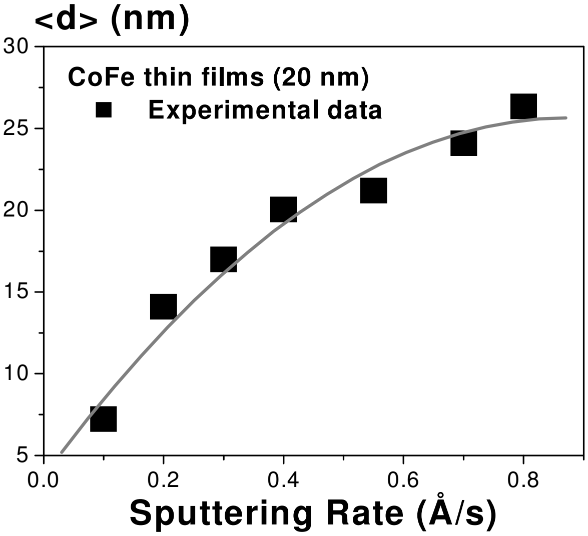

Figure 3. Mean grain size as a function of the sputtering rate

Figure 4 shows the hysteresis loop of each sample measured in the easy axis direction. A clear distinction between hard and easy axis behavior was only observed for the samples with the smallest coercivities, A1 and A2. Although the loops have all very similar squareness with sharp magnetization reversal, there is a clear difference between the coercivity of the samples having the smallest mean grain size (A1 and A2) and those with larger mean grain size (A3 to A7).

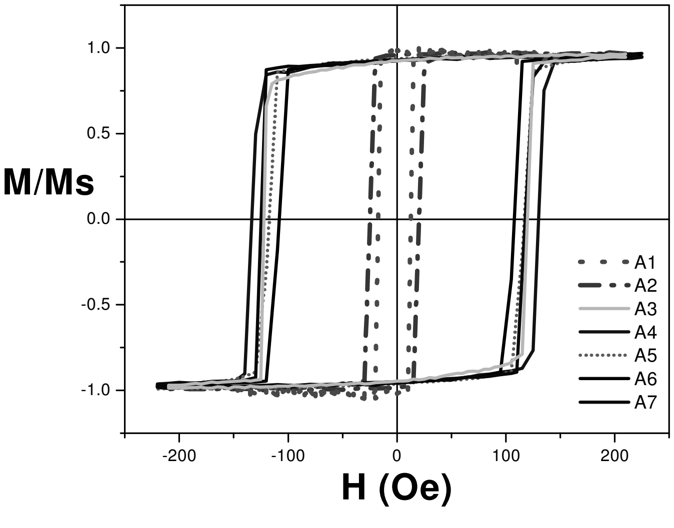

Figure 4. Room temperature Hysteresis loops of CoFe samples A1 – A7

Figure 5 shows the coercivity as a function of the mean grain size. Samples with mean grain diameter below 15 nm show soft magnetic properties with coercivities less than 25 Oe and a minimum of 12 Oe. As the mean grain size increases there is a sharp increase in the coercivity up to around 120 Oe for grain sizes larger than 15 nm. The coercivity remains almost constant for grain sizes in the range 15-26 nm. The coercivity in polycrystalline ferromagnetic materials is well described by the random anisotropy model [10, 11].

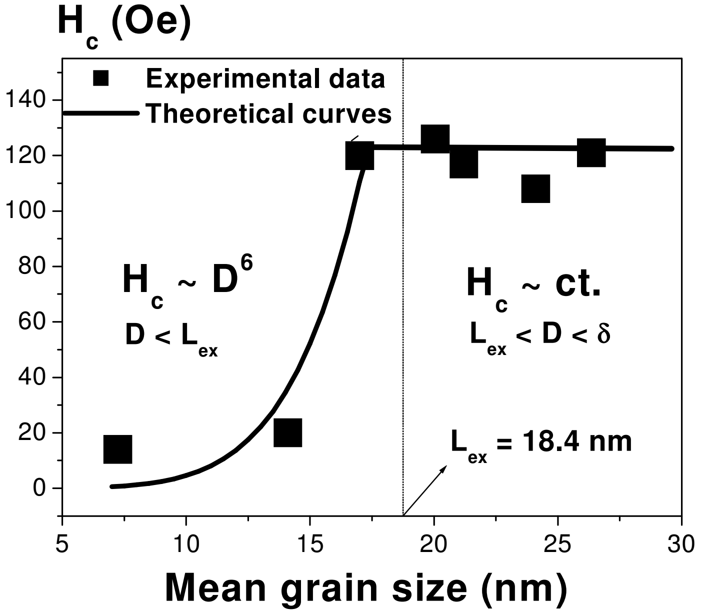

Figure 5. Coercivity variation versus mean grain size.

According to this model, the coercivity variation with the grain size is described by:

$$H_c = \begin{cases} \rho \times \dfrac{K_u{}^4}{A^3 M_s} \times D^6 \text{.............} D < L_{ex} & \text{1.a)} \\ \rho \times \dfrac{K_u}{M_s} \text{........................} D \cong L_{ex} & \text{1.b)} \\ \rho \times \dfrac{\sqrt{AK_u}}{M_s} \times \dfrac{1}{D} \text{.............} D > \delta & \text{1.c)} \end{cases} \qquad (1)$$

Where L_{ex} is the exchange length, δ is the domain wall width, K_u is the magnetocrystalline anisotropy constant, M_s is the saturation magnetization, A is the exchange constant and ρ is a dimensionless factor related to the crystal structure. For $D < L_{ex}$ the coercivity increases sharply as $\sim D^6$ (eq. (1.a)) and is independent of D for $D \cong L_{ex}$ (eq. (1.b)). This is consistent with our experimental data. If $D > \delta$, the coercivity decreases with the grain size as $\sim D^{-1}$.

In order to demonstrate the agreement between the theory and our experimental data we compared the experimental mean grain size values with the calculated exchange length and domain wall widths of our samples. The exchange length indicates the minimum distance over which the magnetization may change direction without involving exchange energy. Below this, the exchange energy overcomes the magnetic anisotropy resulting in an effective anisotropy averaged over a number of grains and therefore reduced in magnitude. The exchange length L_{ex} is given by:

$$L_{ex} = \sqrt{A / K_u} \tag{2}$$

where A is the exchange constant which for our samples was calculated, using the Curie temperature approximation, as $A = 0.911 \times 10^{-8}$ erg/cm. Using $K_u = 2.7 \times 10^5$ erg/cm^3 we obtained $L_{ex} = 18.4$ nm. This value is in excellent agreement with the mean grain size below which a significant reduction in coercivity is observed or above which the coercivity is relatively constant as shown in figure 5. The plateau region in figure 5 can be explained by comparing D with the domain wall width given for a BCC structure by:

$$\delta = \frac{\pi}{\sqrt{2}} L_{ex} \tag{3}$$

Using equation (3), the domain wall width for our samples is $\delta = 41$ nm, which is larger than the largest mean grain size. Hence, in our experimental data only the first two coercivity regimes (eq. 1.a and 1.b) are observed and the plateau region would normally extend up to about 41 nm.

In conclusion we have demonstrated a method of controlling the coercivity in CoFe sputtered thin films based on control of the grain size. Grain size can be controlled via the sputtering rate. Further studies are in progress on thicker films for applications in recording media.

AKNOWLEDGMENTS

The authors would like to acknowledge the financial support to this project by the UK Department of Trade and Industry, LINK ISD Programme. We also acknowledge the EU Research Training Network "NEXBIAS" and Gov. of Pakistan scholarship that funded Dr. S. Manzoor under the PDFP 2003 grant no. PD-II/Ph-UK040532.

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