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Growth Rate Effects in Soft CoFe Films

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We report on growth rate effects in sputter-deposited CoFe films prepared using high target utilization sputtering technology (HiTUS). We find that the grain structure of these polycrystalline films is closely related to the growth rate. By changing the growth rate, samples were prepared with different grain structure, which in turn had the effect of changing the magnetic properties of the films. We demonstrate control of the coercivity, which varied by a factor of more than ten. This was achieved via grain size control in CoFe films of thickness 20 nm. Furthermore, by employing a two-step sputtering process, in which two extreme growth rates are used sequentially, we were able to tune the saturation magnetization.

***Index Terms*—Control of M_s and H_c in CoFe films, grain size effects, high moment soft CoFe films, high target utilization sputtering technology (HiTUS).**

I. Introduction

CoFe thin films are widely used in sensing devices such as read heads, magnetic random access memory cells, or as the write element in a magnetic recording head. However, depending upon the specific application, the required magnetic properties of CoFe films (e.g., coercivity H_c and saturation magnetization $4\pi M_s$) can vary. Controlling both H_c and $4\pi M_s$ of CoFe is difficult and usually involves the use of additives or seed layers [1], which are inconvenient in terms of cost and final device capabilities. In this paper, we demonstrate the preparation of CoFe thin films with control of H_c and $4\pi M_s$ over a broad range of values. A change in H_c by a factor of ten and a maximum change in $4\pi M_s$ by a factor of 3.5 have been achieved by using high target utilization sputtering technology (HiTUS), which we have described previously [2], [3]. The change in both H_c and $4\pi M_s$ has been achieved by tuning the growth rate, which we have shown has the effect of modifying the grain size in the sputtered films [3]. We prepared CoFe samples from sputtering targets with two different compositions ($Co_{35}Fe_{65}$ and $Co_{60}Fe_{40}$).

II. Experimental Results

CoFe films of thickness 20 nm were sputtered directly onto unheated Si (100) substrates using HiTUS. This is a novel design in which the plasma is generated by means of an RF antenna, in a side arm, remote from the sputtering chamber. Because the plasma generation is independent of the sputtering cathode, a high-density plasma can be achieved in addition to the capability of sputtering at very low Ar process pressures ($\sim 10^{-5}$ mbar). Changing the RF power, process pressure, or target dc bias voltage gives close control of the growth rate and the grain size [4]. In all of our depositions the base pressure was 2.8×10^{-7} mbar, Ar process pressure 2.75×10^{-3} mbar, RF power was kept constant at 1.75 kW, and the dc bias voltage was used as a control parameter to change the growth rate.

TABLE I
Sample Characteristics

Sample	Target	Bias (-V)	Growth (Å/s)	<D> (nm)	H_c (Oe)	$4\pi M_s$ (kG)
A1	$Co_{35}Fe_{65}$	120	0.1	10	8	10.3
B1	$Co_{35}Fe_{65}$	1000	0.8	150	130	23.3
A2	$Co_{60}Fe_{40}$	120	0.1	10	15	5.4
B2	$Co_{60}Fe_{40}$	1000	0.8	150	130	19.4

Mean grain sizes are estimated from TEM images. $4\pi M_s$ and H_c were obtained from hysteresis loops after calibration to absolute magnetization.

Samples were sputtered in a dc field of $\sim$500 Oe from two different targets with nominal composition $Co_{35}Fe_{65}$ and $Co_{60}Fe_{40}$. The first target composition corresponds to the highest magnetization per unit volume achievable in CoFe alloys, while the second target generates a lower magnetization and has been chosen for comparison studies of our technique. Samples A1 and B1 were sputtered from the $Co_{35}Fe_{65}$ target, and samples A2 and B2 were sputtered from the $Co_{60}Fe_{40}$ target under the deposition conditions indicated in Table I.

The grain structure was investigated by TEM imaging of carbon support grids coated simultaneously with the Si substrates. We observed a close relationship between the sputtering rate and the final grain size structure of the films. It appears that a low sputtering rate promotes the formation of grains as small as 10 nm (see Fig. 1(a)-Sample A2), while a fast sputtering rate results in films with mean grain size of 150 nm (see Fig. 1(b)-Sample B2).

The crystal structure was identified as bcc. It is apparent that the diffraction pattern rings of sample A2 [Fig. 1(a)] are broadened, which corresponds with a fine grain structure. The diffraction patterns of sample B2 [Fig. 1(b)] are much sharper and better defined, which is indicative of a larger grained polycrystalline film. X-ray diffraction (XRD) analysis of these samples (not shown here) confirmed the bcc structure with a preferred (110) orientation when the samples are grown on Si (100).

The magnetic properties of the samples were determined from room temperature hysteresis loop measurements using a vibrating sample magnetometer (VSM) and are shown in Figs. 2 and 3. Measurements of absolute magnetization were

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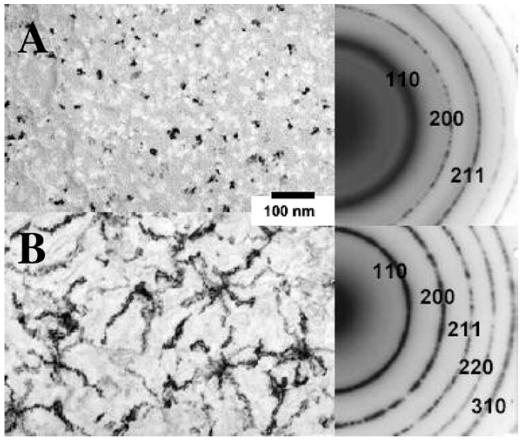

Fig. 1. TEM bright field images and corresponding diffraction pattern rings for samples A2 [Fig. 1(a)] and B2 [Fig. 1(b)].

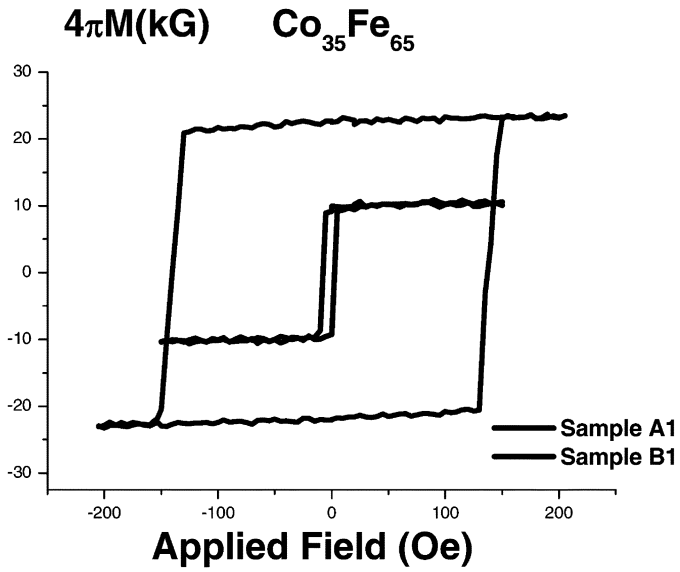

Fig. 2. Magnetization as a function of applied field.

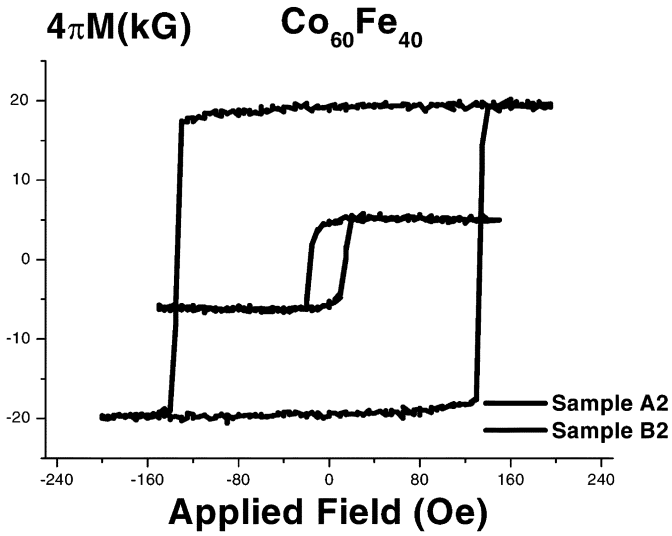

Fig. 3. Magnetization as a function of applied field.

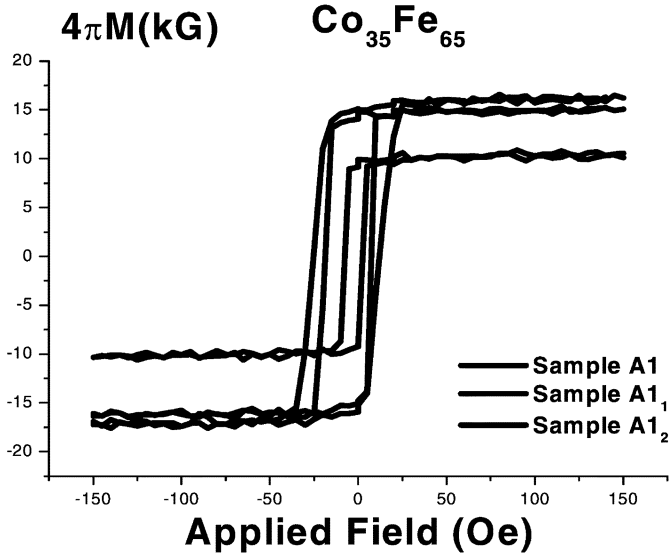

Fig. 4. Magnetization as a function of applied field.

achieved by growing the samples on precut substrates of dimensions 5×5 mm. Film thickness was monitored using an Inficon quartz crystal thickness rate monitor with a 0.1-Å resolution. Magnetic measurements were made using a PAR 4500 VSM calibrated with a Pd standard. It is clear that samples A1 and A2 with small grain size are magnetically soft and in-plane anisotropic, while samples B1 and B2 with large grains are magnetically harder with coercivities exceeding 120 Oe. These samples are in-plane isotropic [1]. The observed change in the coercivity with grain size is in agreement with the random anisotropy model [5], [6]. However, the saturation magnetization of the two sets of samples is also modified by a factor ranging from 2.2 to 3.6. The TEM images and XRD patterns indicate that samples A1 and A2 contain small grains with seemingly noncrystalline regions between them [see Fig. 1(a)]. The origin of the low $4\pi Ms$ values is not clear but appears to be related to poor crystallinity of the samples with small grains (A1, A2). It is known that amorphous iron can suffer a large reduction in $4\pi Ms$ [7], which would account for the lower $4\pi Ms$ observed in the small grain samples (A1, A2). An alternative explanation is that the amorphous material between the grains is more susceptible to oxidation [8]. However, oxide layers would be expected to increase Hc, which does not occur. These data imply that an increase in the saturation magnetization could be achieved by increasing the crystalline fraction of the films.

In order to increase the $4\pi M_s$ of the soft samples A1 and A2, a two-step sputtering process has been employed. Four additional samples have been prepared (two for each target composition) in the following way: 2-nm CoFe was sputtered at 0.8 Å/s followed by 18 nm at 0.1 Å/s for samples $A1_1$ and $A2_1$. Then, samples $A2_1$ and $A2_2$ were sputtered in a similar way, except that the first step layer was 4 nm followed by a 16-nm layer at the lower sputtering rate. The results shown in Figs. 4 and 5 indicate a clear increase in $4\pi M_s$, while the coercivity remains low. Sample $A1_2$ shows a maximum 240% increase in $4\pi M_s$ (Fig. 4) and sample $A2_2$ shows a maximum 150% increase. It should be noted that the samples remained magnetically soft. Since the amount of crystalline material controls the $4\pi M_s$ [7] and the size of the crystallites (grains) controls the H_c [5], [6],

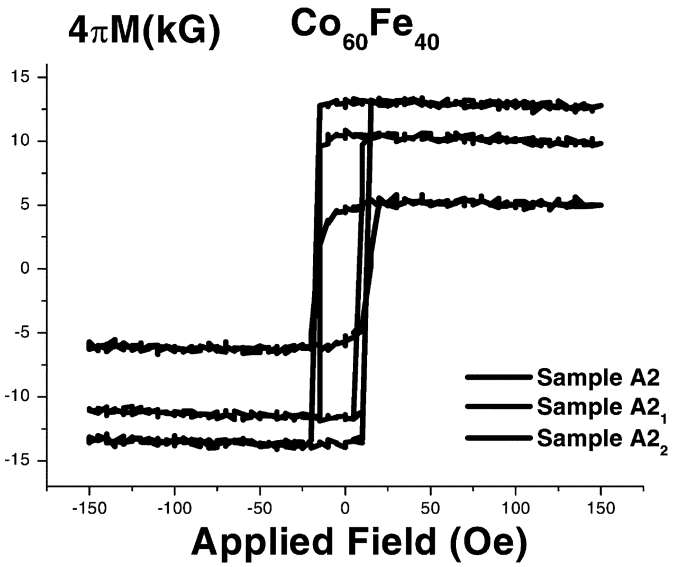

Fig. 5. Magnetization as a function of applied field.

this result implies that the mean grain size is unchanged but the packing fraction is increased in the two-step deposition samples. We believe that this is due to an increase in the number of nucleation sites, which is promoted by the two-step sputtering process. The film growth starts by a nucleation process, followed by grain growth and coalescence [9]. Each stable nucleus develops into a grain. For very thin films, where coalescence does not occur, the number of nuclei determines the number of grains. In the two-step sputtering process, the first step is the growth of an ultrathin layer at a fast growth rate during which a large number of nuclei are formed. However, grain growth is not initiated, as this step is very short. By continuing with the second step at low sputtering rate, grain growth is continued, but the final grain structure will be determined by the sputtering rate according to the relationship "fast growth rate $\rightarrow$ large grains" and "slow growth rate $\rightarrow$ small grains"[3]. Since the second growth step is at a slow rate, the grain structure of the two-step samples will resemble that of samples A1 and A2 except that the amount of "amorphous" phase is reduced and a larger number of small grains are formed. In turn, $4\pi M_s$ will be enhanced by the two-step sputtering procedure, while the coercivity remains low (Fig. 4 and 5). For the two-step samples in which the first layer is thicker than 4 nm, the $4\pi M_s$ approaches the value of the bulk material but the coercivity also increases.

This interpretation is supported by TEM images and diffraction patterns showing the grain structure of the samples sputtered using the two-step method. Bright field TEM images indicate a clear increase in the number of small grains formed in the two-step process (Figs. 1(a) and 6). Also, the diffraction pattern rings for sample $A2_2$ (Fig. 6) are sharper, which indicates better crystallinity in this sample in comparison with sample A2 [Fig. 1(a)].

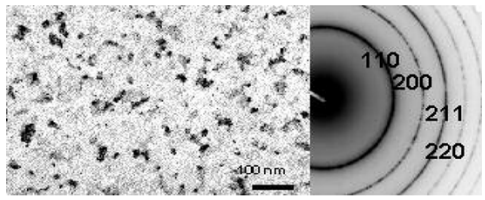

Fig. 6. TEM bright field image and corresponding diffraction pattern rings for sample $A2_2$.

This two-step method is very important because it could be used to prepare various polycrystalline thin films with different grain sizes and packing fractions. By changing these two parameters, the structural and magnetic properties of the films can be altered substantially, depending upon the specific application. Of course, one of the main questions is how close the two-step sputtering process can come to producing films with $4\pi Ms = 24$ kG while maintaining low coercivity. We intend to undertake FMR measurements to verify the values of $4\pi Ms$ and to check for multiple phases in the samples.

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REFERENCES

[1] H. S. Jung, W. D. Doyle, and S. Matsunuma, "Influence of underlayers on the soft properties of high magnetization FeCo films," *J. Appl. Phys.*, vol. 93, pp. 6462–6464, 2003.
[2] M. J. Thwaites, "High density plasmas," U.S. Patent 6 463 873, Oct. 2002.
[3] M. Vopsaroiu, G. V. Fernandez, M. J. Thwaites, J. Anguita, P. J. Grundy, and K. O'Grady, "Deposition of polycrystalline thin films with controlled grain size," *J. Phys. D, Appl. Phys.*, vol. 38, pp. 490–496, 2005.
[4] M. Vopsaroiu, M. J. Thwaites, S. Rand, P. J. Grundy, and K. O'Grady, "Novel sputtering technology for grain size control," *IEEE Trans. Magn.*, vol. 40, no. 4, p. 2443, Jul. 2004.
[5] R. Alben, J. J. Becker, and M. C. Chi, "Random anisotropy in amorphous ferromagnets," *J. Appl. Phys.*, vol. 49, pp. 1653–1658, 1978.
[6] M. Vopsaroiu, M. Georgieva, P. J. Grundy, G. V. Fernandez, S. Manzoor, M. J. Thwaites, and K. O'Grady, "Preparation of high moment CoFe films with controlled grain size and coercivity," *J. Appl. Phys.*, vol. 92, May 2005.
[7] P. J. Grundy, "The structure and magnetic properties of amorphous magnetic thin films," *J. Magn. Magn. Mater.*, vol. 21, pp. 1–23, 1980.
[8] W. Yu, J. A. Bain, Y. Peng, and D. E. Laughlin, "Magnetization reduction due to oxygen contamination of bias sputtered $Fe_{65}Co_{35}$ thin films," *IEEE Trans. Magn.*, vol. 38, no. 5, p. 3030, Sep. 2002.
[9] C. V. Thompson, "Grain growth in polycrystalline thin films," in *Proc. Mat. Res. Soc. Symp.*, vol. 343, 1994, pp. 3–12.

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