Soft Magnetic Properties of Nanocrystalline Amorphous HITPERM Films and Multilayers

M.-Q. Huang, Y.-N. Hsu, M. E. McHenry, and D. E. Laughlin

***Abstract*—Amorphous precursors to HITPERM (Fe, Co)–Zr–B–Cu (a-HITPERM) films and a-HITPERM/SiO_2 multilayer films have been deposited on glass or Si substrates by a rf sputtering system with a target of composition $(Fe_{0.7}Co_{0.3})_{88}Zr_7B_4Cu_1$. It was found that the a-HITPERM single layer film with a thickness of 100 nm possessed good soft magnetic properties with a saturation magnetization of $4\pi M_s \geq 14$ kG and a coercivity of $H_c \sim 0.9$ Oe at room temperature. Hc increases from 0.9 Oe to 25 Oe when the film thickness increases from 100 to 150 nm. To obtain excellent soft magnetic properties at larger thicknesses, a-HITPERM/SiO_2 multilayer films have been synthesized. In these, H_c drops from 25 Oe (single layer) to 0.25 Oe (multilayers) with the same total a-HITPERM thickness. The intervening SiO_2 layers play an important role in reducing the coercivity. Experimental results show the optimum thickness for SiO_2 was 2–4 nm. M–H loops for the multilayers films exhibit pronounced two step magnetization reversal processes for temperatures between 5 and 25 K. This behavior can be attributed to magneto-static coupling between a-HITPERM layers and sequential switching of the layers. The coupling exhibits itself in a blocked phenomenon with wide (stepped) hysteresis curves for $T < 25$ K and smooth narrow hysteresis curves for $T > 25$ K. The effects of magneto-static coupling on the magnetic properties of a-HITPERM/SiO_2 films will be discussed.**

***Index Terms*—High moment material, HITPERM, magnetic static coupling, nanocrystalline, soft magnetic thin film, thermally activated switching.**

I. Introduction

A NEW class of nano crystalline alloys, HITPTERM (Fe, Co)–M–B–Cu (M = Zr, Hf, Nb and etc.) magnets have been developed in our previous work [1]. It was found that HITPERM alloys exhibited excellent soft magnetic properties at elevated temperatures. In the present work, we describe thin films of the amorphous precursor to HITPERM (a-HITPERM) and a-HITPERM/SiO_2 multilayer films. These are suggested for high temperature recording head application. a-HITPERM single or a-HITPERM/SiO_2 multilayer films were fabricated and characterized in the temperature range of 5 and 300 K. The soft magnetic properties of nanocrystalline a-HITPERM films are reported here.

Manuscript received October 13, 2000.

This research was supported by the Data Storage Systems Center at CM, by the Air Force Office of Scientific Research, Air Force Material command, USAF, under Grant F49620-96-1-0454.

The authors are with the Department of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, PA 15213 USA (e-mail: mh8f@andrew.cmu.edu).

Publisher Item Identifier S 0018-9464(01)06564-5.

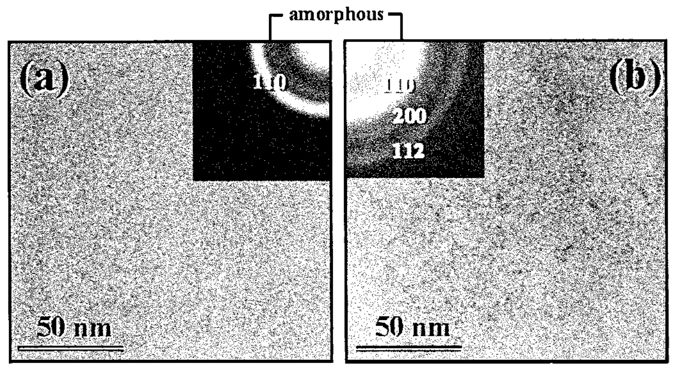

Fig. 1. TEM plane-view bright field images and diffraction patterns. (a) 50 nm. (b) 130 nm a-HITPERM single layer film.

II. Experiments

a-HITPERM single layer films or a-HITPERM/SiO_2 multilayer films were deposited on glass or Si (100) substrates using a Leybold–Heraeus Z400 rf-sputtering system with an alloy target of composition $(Fe_{0.7}Co_{0.3})_{88}Zr_7B_4Cu_1$. The best sputtering conditions used for developing good soft magnetic properties were Ar pressure of 10 mTorr, sputtering power density of 2.3 W/cm^2, and substrate temperature of $\sim$24°C. Magnetic properties (coercivity H_c, anisotropy field H_k, and hysteresis loop squareness B_r/B_s) at room temperature were measured by an B–H hysteresis loop tracer in fields up to 100 Oe with a frequency of 2 Hz. A superconducting quantum interference device (SQUID) magnetometer was also used to calibrate the saturation magnetization $4\pi M_s$, and investigate the magnetic properties at 5–300 K. A Philips EM 420T transmission electron microscopy (TEM) was employed to study the film's microstructure.

III. Results and Discussions

TEM diffraction patterns for as-deposited single layer films with thicknesses of 50 nm or 130 nm have been shown in Fig. 1. Both films show amorphous rings, indicating that the amorphous phase exists in these two films. However, the intensity of the diffraction patterns resulting from the bcc structured grains is enhanced for the thick a-HITPERM film. This implies that the film becomes more crystalline with increasing thickness. The diffuse bcc diffraction rings indicate that the bcc structure grains are very small.

The corresponding magnetic properties of films are as follows.

0018–9464/01$10.00 © 2001 IEEE

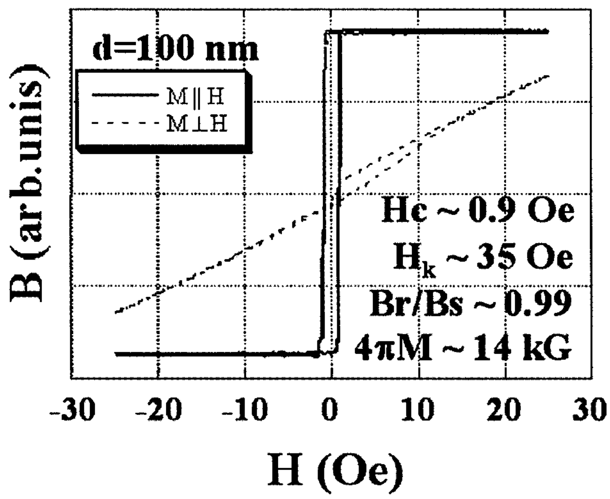

Fig. 2. B–H loop of a-HITPERM single layer film with $d = 100$ nm.

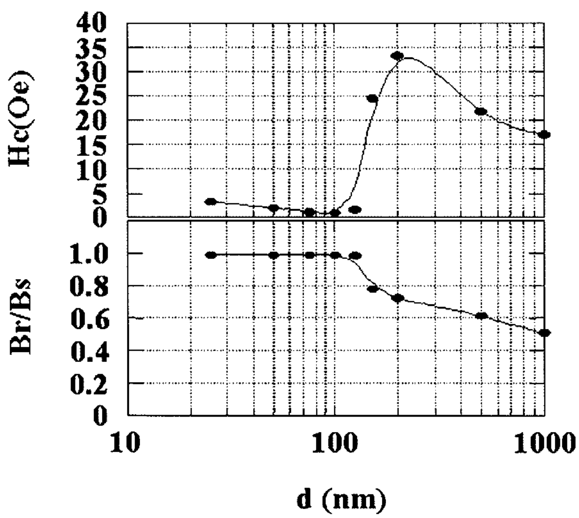

Fig. 3. a-HITPERM thickness (d) dependence of (a) Hc and (b) Br/Bs for a-HITPERM single layer films.

A. a-HITPERM Single Layer Films

A typical hysteresis loop for the a-HITPERM single layer film with a thickness of $d = 100$ nm is shown in Fig. 2. It shows good soft magnetic properties, $Hc \sim 0.9$ Oe, $Br/Bs \sim 0.99$, and 4 $\pi M_s > 14$ kG. We have observed that this film exhibits an in-plane uniaxial anisotropy with an anisotropy field (H_k) of $\sim$35 Oe at room temperature. This may result from forming aligned ferromagnetic atom-pairs due to their strong magnetic exchange coupling during deposition process. The soft magnetic properties are degraded when the film thickness further increases. Hc increases from 0.9 to 25 Oe and Br/Bs decreases from 0.99 to 0.78 when d is increased from 100 to 150 nm.

As shown in Fig. 3, Hc, Br/Bs, and anisotropy behavior vary significantly with the film thickness (d). The films with $d < 125$ nm exhibit an in-plane uniaxial anisotropy, low Hc (<4.5 Oe), and high Br/Bs (0.98~0.99). It implies that there exists a strong exchange coupling among nano-grains or particles, which are of small size and small separation. As d increases to 150–200 nm, the coupling may be reduced by the increase of separation. In this case, magnetization reversal

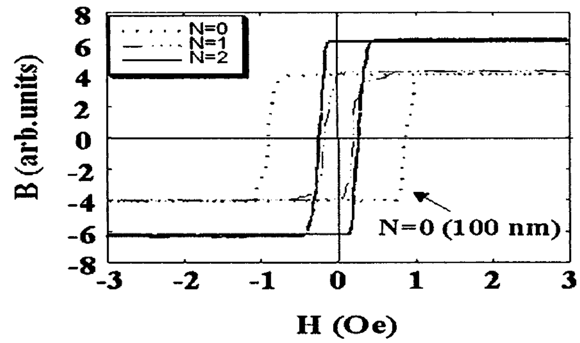

Fig. 4. B–H loops of a-HITPERM/SiO_2 multilayer films with one ($N = 1$) or two ($N = 2$) SiO_2 layers.

process may be partially dominated by a rotation of the magnetization vector. Consequently, Hc increases up to 25~32 Oe and Br/Bs decreases to 0.76~0.78. When d further increases, the multidomain particles may form, resulting in decrease of both Hc and Br/Bs. Chen *et al.* [2] also observed the similar results in sputtered Co films.

B. a-HITPERM/SiO_2 Multilayer Films

In order to maintain excellent soft magnetic properties at larger thicknesses, a-HITPERM/SiO_2 multilayer films have been synthesized. For the multilayer films with only one SiO_2 interlayer ($N = 1$), they were deposited in a layer structure of a-HITPERM(d)/SiO_2(t)/a-HITPERM(d)/substrate. For the multilayer films with two SiO_2 interlayers ($N = 2$), they were deposited in a layer structure of a-HITPERM(d)/SiO_2(t)/a-HITPERM(d)/SiO_2(t)/a-HITPERM(d)/substrate.

The typical hysteresis loops for both type films with $d =$ 50 nm and $t = 2$ nm are plotted in Fig. 4. It was found that with the same total thickness (D) of a-HITPERM, Hc for multilayer films are much smaller than that of single layer films. In the case of $N = 1$, with $D = 100$ nm, the Hc drops from 0.9 Oe (single layer) to 0.2 Oe (multilayer). In the case of $N = 2$, with $D = 150$ nm, the Hc drops from 25 Oe (single layer) to 0.25 Oe (multilayer). Similar phenomena have been also observed by Naoe *et al.* in a nanocrystalline Fe–Cu–Nb–Si–B/Al multilayer film [3]. The intervening SiO_2 layers play an important role in reducing the coercivity Hc. This behavior can be attributed breaking of the exchange coupling between the layers. However, at low temperatures, interlayer magneto-static coupling induced by interface roughness in magnetic/nonmagnetic multilayer films leads to a larger coercivity. The concept of so-called orange-peel magnetostatic coupling was first proposed by Neel [4] and further developed more recently by several other scientists [5], [6].

We have also observed that Hc of the multilayer films ($N =$ 1) strongly depends on the thickness of interlayer SiO_2(t) as well as on the thickness of a-HITPERM layer (d). As shown in Fig. 5, for the multilayer films ($N = 1$) with different d (50 nm or 100 nm), Hc vary significantly with the thickness of interlayer SiO_2(t). To obtain the lowest Hc, the optimum thickness t is 2–4 nm for the multilayer film with $d = 50$ nm, and $t > 4$ nm for the multilayer film with $d = 100$ nm. It seems that t (and t/d) should be thick enough to well form magneto-static coupling. Otherwise, it results in higher Hc when $t < t$ (optimum).

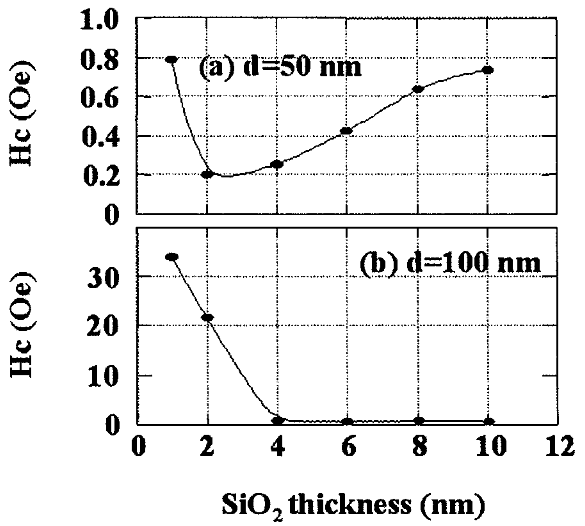

Fig. 5. Thickness of interlayer $SiO_2(t)$ dependence of Hc for a-HTIPERM multilayer films. (a) $d = 50$ nm. (b) $d = 100$ nm.

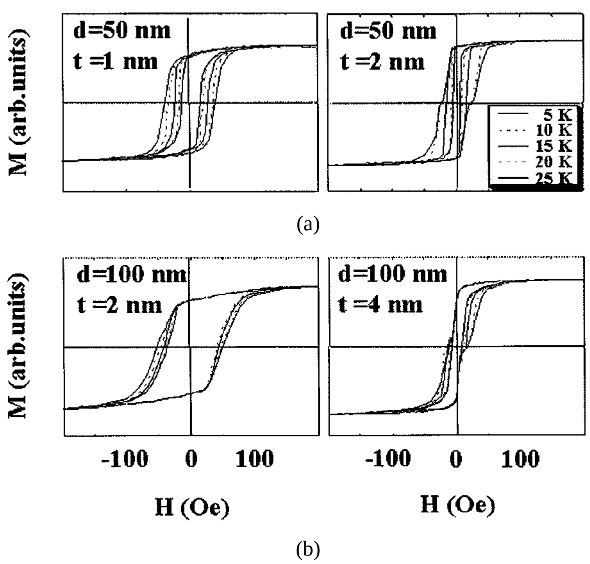

Fig. 6. M–H loops of the multilayer films ($N = 1$). (a) $d = 50$ nm. (b) $d = 100$ nm at 5–25 K.

On the other hand, t (and t/d) should be thin enough to retain strong coupling. Otherwise, it causes reduce in the coupling and increase in Hc when $t > t$ (optimum). In our case, it appears that the optimum value for t/d is ~0.04.

The magnetic properties of the above $N = 1$ type multilayer films with $d = 50$ or 100 nm have been investigated at cryogenic temperatures (5–25 K). As shown in Fig. 6, the M–H loops show a smooth hysteresis curves with a larger Hc_1 at 5–25 K when $t < t$ (optimum). As expected, Hc_1 is decreased when $t = t$ (optimum) and the $M = H$ loops show a smooth hysteresis curves at temperatures between 15 and 25 K. Below 15–25 K, the M–H loops exhibit a pronounced two step magnetization reversal process with a lower value of Hc_1 and a higher

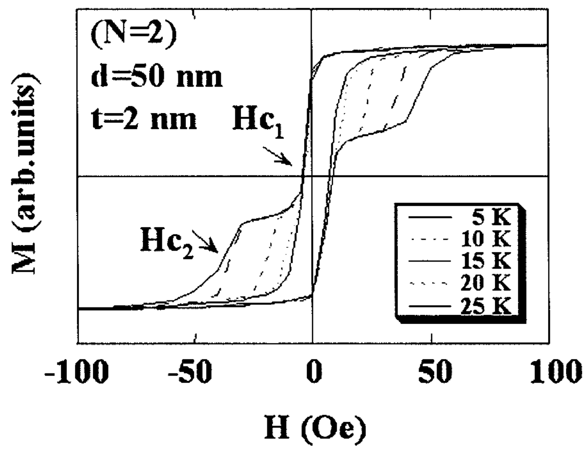

Fig. 7. M–H loop of multilayer film ($N = 2$) at 5–25 K.

value of Hc_2, which decreases monotonically with increasing temperature. This behavior has been also observed in $N = 2$ type multilayer film, which shows two step hysteresis curves with $Hc_1 \sim 5$ Oe and $Hc_2 \sim$ 12–38 Oe.

In comparison with the $N = 1$ films, the steps of $N = 2$ film are much sharper than that of $N = 1$ type multilayer films (Fig. 7). We believe that this behavior can be attributed to magneto-static coupling between a-HITPERM layers and sequential switching of layers. The coupling exhibits itself in a blocked phenomenon with wide (stepped) hysteresis curves for $T \leq 25$ K, and smooth narrow hysteresis curves for $T \geq 25$ K. It suggests that higher values of Hc_2 are caused by a weak orange-peel interlayer exchange coupling due to an enhancement of topological coupling at cryogenic temperature. A Similar phenomenon has been observed in Spin Valve type multilayers at room temperature [7]. The temperature dependence of H_{c1} and H_{c2} could be related to the thermal activation and the variation of the magneto-crystalline energy at cryogenic temperature.

REFERENCES

[1] M. A. Willard, M.-Q. Huang, D. E. Laughlin, M. E. McHenry, J. O. Cross, V. G. Harris, and C. Franchetti, "Magnetic properties of HIT-PERM $(Fe,Co)_{88}Zr_7B_4Cu_1$ magnets," *J. Appl. Phys.*, vol. 85, p. 4421, 1999.

[2] T. Chen and G. B. Chairan, "High coercivity and high hysteresis loop squareness of sputtered Co–Re thin film," *J. Appl. Phys.*, vol. 50, p. 4285, 1979.

[3] M. Naoe, H. Matsumiya, T. Ichihara, and S. Nakagawa, "Preparation of soft magnetic films of nanocrystalline Fe–Cu–Nb–Si–B alloy by facing targets sputtering," *J. Appl. Phys.*, vol. 83, p. 6673, 1998.

[4] L. Neel, "On a new mode of coupling between the magnetizations of two thin ferromagnetic layers," *Comptes. Rendus*, vol. 255, p. 1676, 1962.

[5] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, "Giant magnetoresistance in soft ferromagnetic multilayers," *Phys. Rev.*, vol. B43, p. 1297, 1991.

[6] J. Zhamg and R. M. White, "Topological coupling in spin valve type multilayers," *IEEE Trans. Mag.*, vol. 32, p. 4630, 1996.

[7] ——, "Topological coupling in magnetic multilayer films," *Appl. Phys.*, vol. 79, p. 5113, 1996.