

# Chapter 7

## Magnetic Thin Films for Perpendicular Magnetic Recording Systems

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### 7.1 Introduction

In the advanced information society of today, information storage technology, which helps to store a mass of electronic data and offers high-speed random access to the data, is indispensable. Against this background, hard disk drives (HDD), which are magnetic recording devices, have gained in importance because of their advantages in capacity, speed, reliability, and production cost. These days, the uses of HDD extend not only to personal computers and network servers but also to consumer electronics products such as personal video recorders, portable music players, car navigation systems, video games, video cameras, and personal digital assistances.

IBM introduced the first HDD of the IBM 350 Disk File in 1956, which was the storage unit of the IBM RAMAC 305 computer [1]. It was composed of fifty 61 cm aluminum disks coated with iron oxide paint, and provided an areal density of 0.002 Mbit/inch<sup>2</sup>; in the last half century, the density has been increasing to beat the band.

Such an increase in the density was achieved through epochal developments in the magnetic thin film materials used in magnetic recording media and heads. After a magnetic thin film had been made by using electroless deposition in place of coating, a rapid rise in the density was achieved. Product PATTY@ by Nippon Telegraph and Telephone (NTT), which used electroless Co-Ni-P film reached a density of 9 Mbit/inch<sup>2</sup> [2]. NEC Corp. had continuously developed higher level of 24.5 Mbit/inch<sup>2</sup>. In a disk-manufacturing process, an electroless Ni-P film with amorphous structure was also used for the underlayer plated on the aluminum alloy substrate. A higher system of perpendicular magnetic recording media was being developed by electroless Co-Ni-Re-P and Co-Ni-P alloy films [3, 4].

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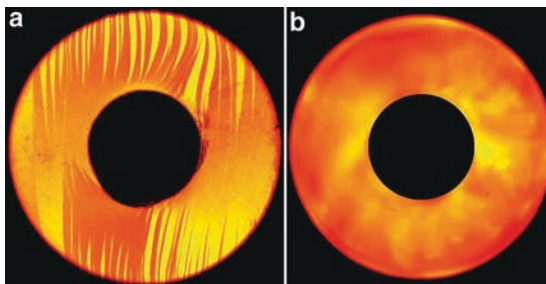
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In this section, our recent study on a challenge of new materials for the next generation of magnetic recording approaching an ultra high areal recording density (beyond 1 Tbit/inch<sup>2</sup>) is overviewed.

## 7.2 Soft Magnetic Underlayer by Electroless-Deposition for Perpendicular Magnetic Recording

A double-layered perpendicular magnetic recording medium [5–7], which is composed of a hard magnetic recording layer with perpendicular magnetic anisotropy and a soft magnetic underlayer (SUL), is a promising system for realizing an ultra high areal recording density. Therefore, we should pay attention not only to the recording layer but also the SUL in order to develop high performance perpendicular magnetic recording media. Concerning the SULs, a large value of  $B_s \delta$ , where  $\delta$  is the thickness of the SUL, is required for making the most of the writing ability of the magnetic recording head, and various sputter-deposited SULs with the thickness of more than a 100 nm have been reported [8–11]. However, we face the following critical issues in developing the SULs for practical use. First, mass productivity is a rather low when such a thick SUL is sputter-deposited. Second, peculiar layered structures are needed to suppress the “spike noise” caused by magnetic domain boundaries in the SUL [12–14].

We have investigated the possibility of the application of Co–Ni–Fe-based alloy thin films electroless-deposited on a disk substrate to the SUL, as a trial to solve the above-mentioned issues. We succeeded in electroless-depositing the SULs of a Co–Ni–Fe–P or Co–Ni–Fe–B alloy without marked macroscopic magnetic domain boundaries [15–17]. Figures 7.1a and b show magnetic domain images observed through the *Kerr effect* for the Co–Ni–Fe–P SULs, which were prepared without and with the applied magnetic field of ca. 800 Oe during electroless-deposition, respectively.

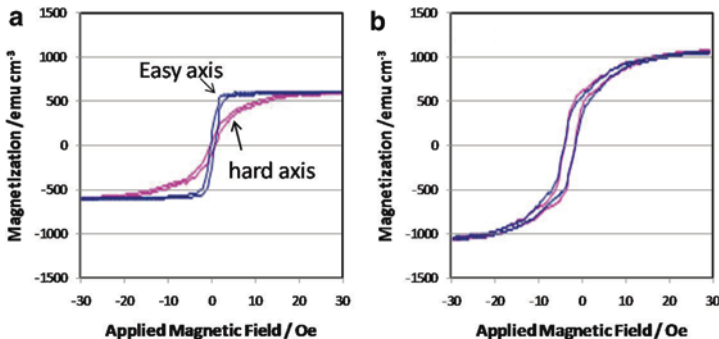


**Fig. 7.1** Magnetic domain images were observed through the Kerr effect of Co–Ni–Fe–P soft magnetic underlayer on a disk substrate electroless-deposited (a) without and (b) with an external applied magnetic field. The thickness of each sample is 1  $\mu\text{m}$  [15]. © 2004 IEEE

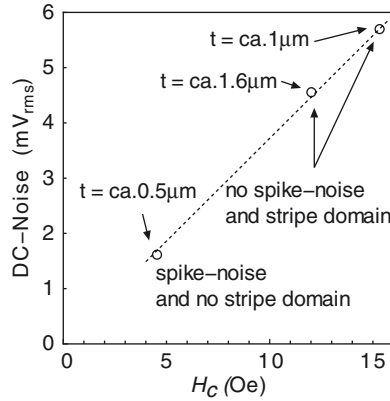
The magnetic field was applied in the direction parallel to the surface of disk substrate while the disk substrate was rotated. The Co–Ni–Fe–P SUL deposited without the magnetic field tends to show clear and complicated magnetic domain boundaries, whereas, the SUL deposited with the magnetic field has no marked domain boundaries. The magnetic domain boundaries of the former are readily moved by an imposition of an external magnetic field, and the domain boundaries cause an problem of spike noise. On the other hand, no domain boundaries are induced in the latter even when the external magnetic field was imposed.

Figures 7.2a and b show in-plane  $M$ – $H$  hysteresis loops of the Co–Ni–Fe–P SULs in Figs. 7.1a and b, respectively. The SUL deposited without the magnetic field had a good soft magnetic property, and it exhibited magnetic anisotropy with the easy axis in the film plane. On the other hand, we obtained an interesting result that the Co–Ni–Fe–P SUL deposited with the magnetic field exhibited almost isotropic magnetic property in the film plane, but slightly perpendicular magnetic anisotropy, which is likely to be induced by fine stripe magnetic domains. The stripe magnetic domains of ca.  $1\ \mu\text{m}$  in width were actually observed by precise magnetic force microscopy in this kind of SUL [18]. We consider that the complicated fine stripe domains make macroscopic magnetic domain boundaries inconspicuous. This phenomenon is similar in the case of the so-called anomalous magnetic thin films, e.g., permalloy thin films with a large Ni composition more than 85 wt.% [19]. From read and write experiments, it was confirmed that the double-layered perpendicular magnetic recording medium with the SUL shown in Fig. 7.1b generated *no* spike noise, whereas, that with the SUL shown in Fig. 7.1a showed, explicitly, the spike noise.

However, in a magnitude of root-mean-square noise amplitude, i.e. DC noise, the SUL without the generation of spike noise takes poor level. Figure 7.3 shows a variation in DC noise voltage of the Co–Ni–Fe–B SUL with in-plane coercivity. DC noise was increased with increasing the value of coercivity [18]. Since the coercivity



**Fig. 7.2**  $M$ – $H$  hysteresis loops of the samples shown in Fig. 7.1(a) Co–Ni–Fe–P soft magnetic underlayer electroless-deposited, (a) without and (b) with an external applied magnetic field [15]. © 2004 IEEE



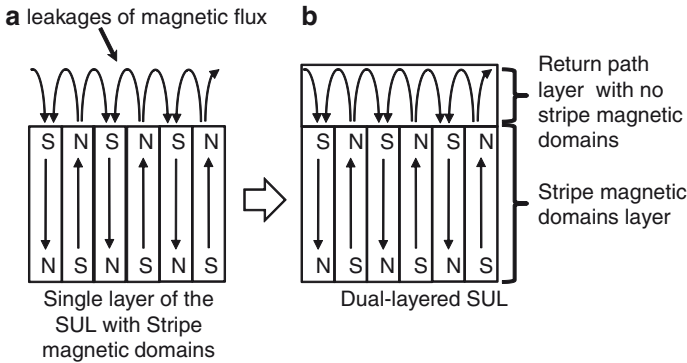
**Fig. 7.3** DC noise voltage of Co–Ni–Fe–B films as a function of in-plane coercivity [18]. © 2005 IEEE

of the SUL without the generation of spike noises inevitably takes a large value by the perpendicular magnetic leakage of striped magnetic domains, then DC noise of the SUL without spike noise cannot be reduced more than that of SUL with the generation of spike noise.

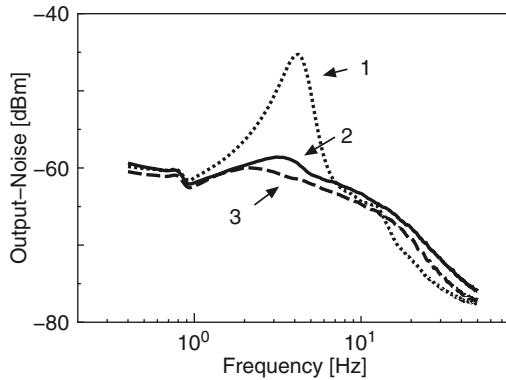
In order to solve this problem, we proposed that the dual-layered SUL consists of two layers having different magnetic domains [20]. The schematic of the double-layered SUL is shown in Fig. 7.4. The lower layer is the film with the stripe domain layer, and the upper layer is the film with the both of small coercivities and no stripe domain. The upper layer works as a return-path layer, i.e., the perpendicular magnetic leakage generated from the lower layer is refluxed into the SUL through the upper layer. The optimized thickness of the upper layer is the height of the magnetic leakage on the surface, and Favieres reported that the height becomes equal to the width of the striped magnetic domains [21]. Figure 7.5 shows the noise spectra for these SULs, which were measured by using a spin-stand tester with a merged GMR head. The double-layered SUL has allowed the noise to be reduced to the same level as in the SUL without the striped magnetic domain. Thus, we have successfully developed a double-layered medium with the SUL fabricated by electroless-deposition, which generates no spike noise. This study has just opened up a new way to manufacture SULs with high mass productivity.

### 7.3 Control of Characteristics of Magnetic Recording Media by Metal Clusters Formed by Electrochemical Process

In a double-layered perpendicular magnetic medium, an intermediate layer between a recording layer and an SUL plays an important role as a seed to control the crystal growth of the recording layer and to reduce the magnetic domain size of the recording layer. Moreover, the intermediate layer should be as thin as possible to minimize the



**Fig. 7.4** Schematic of the dual-layered SUL



**Fig. 7.5** Noise spectra for electrolessly Co-Ni-Fe SULs. 1: SUL without the generation of spike noise (stripped magnetic domain emerged film), 2: Dual layered SUL, 3. SUL with the generation of striped magnetic domain (low  $H_c$  film) [20]

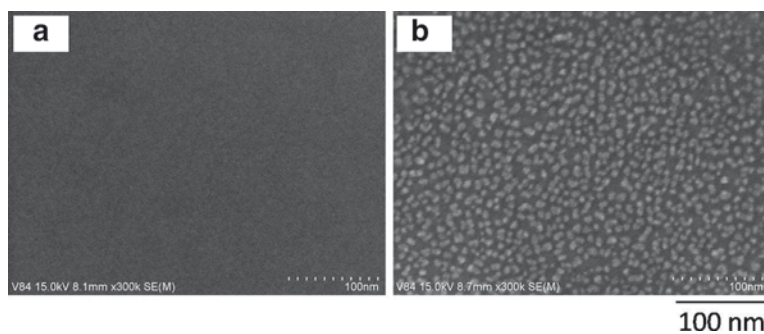
magnetic spacing loss between the recording layer and the SUL for high recording efficiency. The intermediate layers meeting these demands have been developed by sputtering for various perpendicular magnetic recording media.

For instance, in the case of the medium with Co/Pd multilayered perpendicular magnetization film [22, 23], Pd-based intermediate layers such as Pd/ITO [24], Pt-B/Pd/MgO [25], Pd-Si-N [26], and Pd/Si [27–29] were reported to successfully improve magnetic properties of the Co/Pd multilayered films. In these intermediate layers, fine Pd grains provide the suitable nucleation site for the magnetically isolated Co/Pd crystal grains, leading to weak intergranular exchange coupling and good read and write characteristics. On the other hand, we have focused on an electrochemical substitution deposition technique as a novel method to prepare the metal seed for the double-layered media. Electrochemical process is useful to modify a

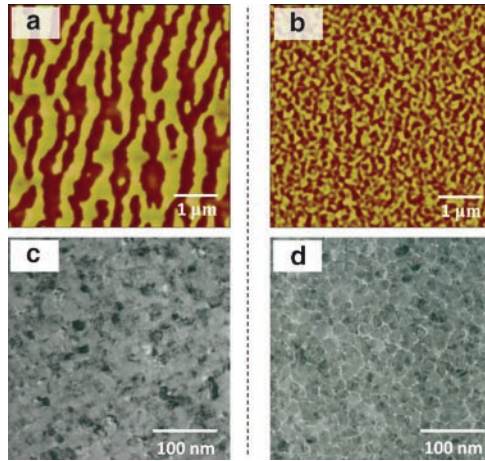
film surface and to form uniformly nanoscopic structure on the surfaces. Since the electrochemical substitution reaction takes place only at the interface between the surface and the electrolyte solution, a deposited metal is expected to form a very thin film or island-like clusters rather than a thick continuous film, which could be applicable to the seed for the double-layered media. In this section, the Pd formation on a Co-based Co–Zr–Nb SUL as the seed for the sputter-deposited Co/Pd perpendicular magnetization film using the substitution reaction caused by the difference in redox potential between Pd and Co is mainly described [30, 31].

Figures 7.6a and b show high resolution SEM images of the Co–Zr–Nb SUL surface without any treatment and that treated with PdCl<sub>2</sub> solution, respectively. An island-like structure with the mean diameter of ca. 9 nm was formed on the SUL with a high density, where the mean distance between adjacent clusters was ca. 18 nm. From X-ray photospectroscopy and diffractometry, the island-like structure was identified as the Pd metal with fcc structure [30], which is hereafter called “Pd cluster seeds”.

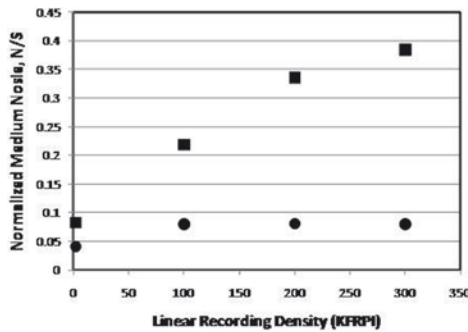
Figures 7.7a and b show MFM images at an ac-demagnetized states of the Co/Pd multilayered films with a sputter-deposited continuous Pd seedlayer (film I) and with the Pd cluster seeds (film II). The maze-like domain pattern with large magnetic domains was observed in the former, whereas, smaller magnetic domains with a diameter of ca. 100 nm were observed in the latter [31]. These results indicated that the Pd cluster seeds formed by the electrochemical process were quite effective to suppress the intergranular exchange coupling, leading to the reduction of magnetic domain size. To elucidate the effect of the Pd cluster seeds, TEM observations were carried out. Figures 7.7c and d show the plan-view TEM bright field images of films I and II. Well-defined grain boundaries were observed in film II, providing the Co/Pd grains with the mean diameter of 15.8 nm, while the boundaries were hardly seen in film I [31]. From this result, it is found that the electrochemically formed Pd cluster seeds worked as nucleation sites for the Co/Pd multilayered film, and enhanced the columnar grain growth of the Co/Pd multilayered film with wide grain boundaries. Apparently, in the Co/Pd multilayered film, the grain boundaries



**Fig. 7.6** SEM images of: (a) Co–Zr–Nb soft magnetic underlayer surface without any treatment and (b) that treated with PdCl<sub>2</sub> solution [31]



**Fig. 7.7** MFM images at the ac-demagnetized states of the Co/Pd multilayered films: (a) with the sputter-deposited 10 nm-thick Pd seedlayer (film I) and (b) with the Pd cluster seeds (film II). Plan-view TEM bright field images of (c) film I and (d) film II [31]



**Fig. 7.8** Dependence of normalized medium noise on the linear recording density for the recording media consisting of film I (squares) and film II (circles) [31]

of voids with low material density contributed to the reduction of intergranular exchange coupling in the lateral direction, resulting in a fine magnetic domain structure as shown in Fig. 7.7b.

Figure 7.8 shows the dependence of normalized medium noise ( $N/S_0$ ) on the linear recording density for the recording media consisting of films I and II, where the medium noise ( $N$ ) was normalized to the reproduced voltage at the low recording density of 2 kFRPI ( $S_0$ ). Medium II with the Pd cluster seeds exhibited lower medium noise than medium I with the sputter-deposited Pd seed layer. This result suggested that the transition noise caused by the fluctuation of the magnetization transition region between recorded bits was suppressed by employing the Pd cluster

seeds, which bring about weak intergranular exchange coupling and fine magnetic domains [31].

Our study clarified that the size and nucleation density of the Pd cluster seeds were controlled by applying a pretreatment with a  $\text{SnCl}_2$  solution, which mostly produced the finer Pd cluster seeds with a higher density and further decreased the intergranular exchange coupling, magnetic domain size, and medium noise. Moreover, the Pd cluster seeds formed on the electroless-deposited Co–Ni–Fe based SUL, mentioned in the previous section, were also effective for improving the magnetic properties of Co/Pd multilayered films [32]. This is a promising way to improve not only the magnetic properties but also the mass productivity of double-layered perpendicular magnetic recording media. Through these studies, we have demonstrated that the combination of wet and dry processes is useful in fabricating perpendicular magnetic recording media.

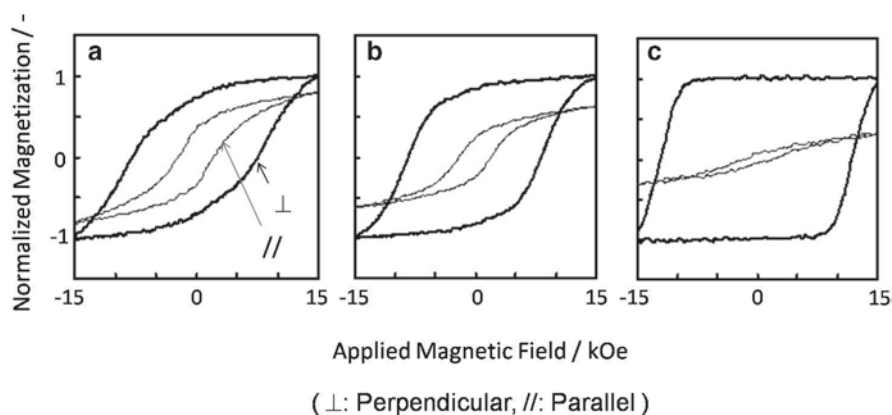
## 7.4 Development of Permanent Magnet Thin Films with Very High Perpendicular Magnetic Anisotropy

From the viewpoint of the materials of recording layer, thin films exhibiting high perpendicular magnetic anisotropy are indispensable for increasing areal recording density against superparamagnetism. Various attempts have been initiated to achieve the goal of ultra high areal density beyond 1 Tbit/inch<sup>2</sup>, and, in particular, a thin film with high perpendicular magnetic anisotropy with an anisotropy constant,  $K_u$ , greater than  $1.2 \times 10^7$  erg/cm<sup>3</sup> is required to maintain the stability of magnetization in exceedingly small crystal grains less than 5 nm in diameter against thermal agitation [33, 34]. The  $\text{SmCo}_5$  alloy, a representative permanent magnet material, is a promising candidate for the materials because of its extremely high uniaxial magnetocrystalline anisotropy, whose  $K_u$  value is  $1.1 \times 10^8$  erg/cm<sup>3</sup> or greater in the form of bulk alloy [34]. However, an  $\text{SmCo}_5$  thin film exhibiting distinct perpendicular magnetic anisotropy had not been prepared, although the film with in-plane anisotropy is relatively easy to prepare, for instance, by using a Cr underlayer [35–39]. Recently, we succeeded in developing sputter-deposited  $\text{SmCo}_5$  thin films with very high perpendicular magnetic anisotropy [40–45]; it will be stated in detail in this section.

The introduction of appropriate underlayer is a key to controlling the magnetic properties. First, various materials were examined as an underlayer of Sm–Co thin films in order to impart perpendicular magnetic anisotropy. As a result, it was found that the Sm–Co thin films deposited on a glass substrate coated with a (111)-oriented fcc-Cu underlayer with a large thickness of 100 nm or more exhibited distinct perpendicular magnetic anisotropy. The Sm–Co thin films with the Cu underlayer contained the hexagonal  $\text{SmCo}_5$  phase with (001) orientation, revealed by an X-ray diffractometry. Thus, the Cu underlayer was found to be useful for affording perpendicular magnetic anisotropy to  $\text{SmCo}_5$  thin films [40]. This was the first time when the  $\text{SmCo}_5$  thin films exhibiting perpendicular magnetic anisotropy were developed. Takei et al. almost simultaneously reported a similar result [46].



Figure 7.9a shows  $M$ - $H$  hysteresis loops for a typical sample with the Cu underlayer (sample A). The substrate temperature was set at an elevated temperature of  $345^\circ\text{C}$  for the deposition of all layers. The values of  $H_c$  and squareness ratio (SQR) measured in the perpendicular direction are listed in Table 7.1 together with the values of  $K_u$  and  $\Delta\theta_{50}$  of the  $\text{SmCo}_5(002)$  reflection in an X-ray diffractometry, defined as the values of full width at half maximum obtained from the rocking curves. Both  $H_c$  and SQR in the perpendicular direction were greater than those in the film plane, indicating that perpendicular magnetic anisotropy was clearly generated. The  $K_u$  value of  $1.7 \times 10^7 \text{ erg/cm}^3$  is greater by a factor of 10 than a conventional material for recording layer, such as a Co-Cr-Pt-based alloy. Figure 7.9b shows  $M$ - $H$  hysteresis loops for sample B, which has the same layer configuration with sample A deposited under a different condition; namely, the substrate temperature was set at a room temperature of  $20^\circ\text{C}$  for the deposition of the underlayer, followed by the deposition of the Sm-Co layer at  $325^\circ$ . The values of  $H_c$ , SQR, and  $K_u$  were greater than those of sample A. The enhancement of perpendicular magnetic anisotropy was revealed to be attributable to the suppression of the surface roughness of Cu underlayer [42, 43]. The surface roughness was attempted to decrease further by introducing



**Fig. 7.9**  $M$ - $H$  hysteresis loops of  $\text{SmCo}_5$  thin films with perpendicular magnetic anisotropy. (a) Sample A, (b) sample B, and (c) sample C correspond to those in Table 7.1. Thick and thin lines represent the loops measured in the directions perpendicular and parallel to the film plane, respectively [45]

**Table 7.1** Magnetic properties and values of full width at half maximum of  $\text{SmCo}_5(002)$  reflection ( $\Delta\theta_{50}$ ) for samples A, B and C shown in Fig. 7.9

Sample	Underlayer	$H_c$ (kOe)	SQR	$K_u$ ( $\times 10^7$ erg/cm $^3$ )	$\Delta\theta_{50}$ SmCo $_5(002)$
A	Cu	7.7	0.72	1.7	Undetectable
B	Cu	8.2	0.85	2.2	10.1
C	Cu/Ti	12.0	1	4.0	3.1

a Ti layer prior to depositing the Cu underlayer. The Ti layer was expected to make smoother the Cu layer deposited upon it, because the melting point and the surface energy of Ti are higher than those of Cu. Indeed, the surface roughness of underlayer for sample C became smaller than that for sample B [42]. The  $\Delta\theta_{50}$  values for samples B and C were  $10.1^\circ$  and  $3.1^\circ$ , respectively. This indicates that the higher degree of preferred orientation of the *c*-axis (easy axis) of hexagonal  $\text{SmCo}_5$  was achieved with samples having a smoother underlayer surface. The  $M$ - $H$  hysteresis loop for sample C is shown in Fig. 7.9c. The magnetic properties of sample C were significantly improved; the values of  $H_c$  and SQR became 12.0 kOe and unity, respectively. The  $K_u$  value reached an extremely high value of  $4.0 \times 10^7 \text{ erg/cm}^3$ . We estimate the minimal stable grain diameter of sample C to be 3.7 nm from the  $K_u$  value, which is a value highly advantageous for an ultra high-density magnetic recording medium.

In summary, we developed, by using a conventional sputtering process, the  $\text{SmCo}_5$  thin film with very high perpendicular magnetic anisotropy. This result contributes to the progress in applications of the  $\text{SmCo}_5$  thin films to various magnetic devices, especially to magnetic recording media with ultra high areal recording density.

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