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Effects of Sputtering Conditions on Magnetic Properties of Co/Pd Multilayered Films

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Effects of substrate temperature and N_2 gas addition to Ar sputtering gas during deposition of Co/Pd multilayered films on their magnetic properties and crystalline microstructure were investigated. By depositing at the substrate temperature of 230 °C with N_2 partial pressure of 0.06 mTorr, the Co/Pd multilayered film with weak intergranular exchange coupling and high coercivity in the direction perpendicular to the film surface was successfully prepared. Transmission electron microscopy revealed that the Co/Pd crystal grains were physically isolated with vacant grain boundaries produced by depositing upon the heated substrate with N_2 additive gas, leading to the decrease in intergranular exchange coupling.

Key words: Co/Pd multilayered film, perpendicular magnetic recording medium, intergranular exchange coupling, substrate temperature, N_2 gas addition

1. INTRODUCTION

A Co/Pd multilayered film possesses high perpendicular magnetic anisotropy generated at the interface between Co and Pd sub-layers [1]. Therefore, it attracts much attention in respect of the application to the perpendicular magnetic recording medium with high thermal stability for hard disk drives [2]. However, it is a critical problem that the Co/Pd multilayered media exhibit high medium noise, which is brought about by large magnetic clusters resulting from strong intergranular exchange coupling in the lateral direction of the film surface.

It is well known that characteristics of sputter-deposited thin films are markedly affected by sputtering conditions, *e.g.*, sputtering power, pressure of process gas, substrate temperature, *etc.* In most studies, Co/Pd multilayered films have been sputter-deposited at room temperature under pure noble gas such as Ar, Kr, and Xe. On the other hand, the Co/Pt multulayered film prepared by electron beam evaporation showed weak intergranular exchange coupling when deposited upon heated substrate [3]. Furthermore, the addition of N_2 to Ar sputtering gas was reported to fine down the crystal grains of CoCr films [4].

Recently, we applied the fabrication processes, *i.e.*, the substrate heating during deposition and the addition of N_2 to Ar gas, to the sputter-deposited Co/Pd multilayered film in order to decrease the intergranular exchange coupling [5]. In this paper, the change in magnetic properties of Co/Pd multilayered films coming from a variation of the sputtering conditions was investigated and, in particular, the correlation between their magnetic properties and crystalline microstructure was focused on.

2. EXPERIMENTAL

The Co/Pd multilayered films were prepared by using multi-source dc magnetron sputtering system with a rotating substrate table (ANELVA, L-350S-C). The layer structure of the films was [Co (0.2 nm)/Pd (0.8 nm)]$_{20}$/Pd (5 nm)/substrate unless otherwise noted, where a 2.5 inch-diameter glass disk was used as the substrate. The sputtering conditions are listed in Table I. The substrate temperature (T_{sub}) during deposition of Co/Pd multilayer and seedlayers was set to 20 or 230 °C. The N_2 gas partial pressure (P_{N2}) was 0 or 0.06 mTorr. For read-write (R/W) experiments, the double-layered media composed of C (5 nm)/[Co (0.2 nm)/Pd (0.8 nm)]$_{20}$/Pd (5 nm)/C (5 nm)/$Co_{91}Zr_5Nb_4$ (300 nm)/substrate were used. The CoZrNb soft magnetic underlayer (SUL) had saturation magnetic flux density of 14 kG and coercivity of 10 Oe.

Magnetic properties of the films were measured with a vibrating sample magnetometer (VSM), a torque magnetometer, and a polar Kerr magnetometer. Magnetic anisotropy field (H_k) was calculated by using the following equation showing relationship between a magnetic anisotropy constant (K_u) and saturation magnetization (M_s),

Table I Sputtering conditions.

Base pressure [Torr]		< 3 x 10^{-7}
Ar pressure [mTorr]	Co/Pd	20 (+ N_2 0.06)
	Pd	100
	C	50
	CoZrNb	5
Sputtering power [W]	Co/Pd	42/68
	Pd	30
	C	300
	CoZrNb	100
Target diameter [inch]		4
Substrate temperature [°C]	Co/Pd, Pd	20, 230
	C, CoZrNb	20

Table II Magnetic properties and values of $D_{cluster}$ and D_{grain} for Co/Pd multilayered films.

Film	T_{Sub} [°C]	P_{N2} [mTorr]	M_s [emu/cm^3]	H_c [kOe]	α [-]	H_k [kOe]	$D_{cluster}$ [nm]	D_{grain} [nm]
A	20	0	320	5.3	3.6	23.4	169 ± 30	13.9 ± 2.5
B	230	0	340	4.7	3.6	20.0	167 ± 37	20.1 ± 3.6
C	20	0.06	240	4.2	2.6	21.2	143 ± 25	13.3 ± 2.2
D	230	0.06	310	5.2	2.0	17.7	135 ± 26	19.8 ± 3.5

$$H_k = 2K_u / M_s, \quad (1)$$

where the value of K_u was obtained by adding demagnetization energy, $2\pi M_s^2$, to the experimental value of magnetic anisotropy constant (K_{eff}). Crystalline microstructure was analyzed with a transmission electron microscope (TEM) with an acceleration voltage of 200 kV, and with an X-ray diffractometer (XRD) using Cu-$K\alpha$ radiated at an acceleration voltage of 50 kV and an emission current of 200 mA. The composition in the micro-region of the films was determined by energy dispersive X-ray spectroscopy (EDS). Magnetization state was observed with a magnetic force microscope (MFM). The *R/W* characteristics were evaluated with a merged MR head for recording (gap length G_r = 0.28 μm, track width T_w = 1.45 μm) and reproducing (shield- to-shield gap length $G_{s\text{-}s}$ = 0.2 μm, T_w = 0.9 μm). The linear velocity was 6.35 m/s. The medium noise was calculated by subtracting the system noise power from the total noise power and integrating up to 50 MHz.

3. RESULTS AND DISCUSSION

3.1 Magnetic properties

Table II shows the magnetic properties of the Co/Pd mulatilayered films (A-D) deposited on four kinds of conditions listed. A slope parameter in a *M-H* loop, α (= $4\pi(dM/dH)_{H=Hc}$), which is regarded as a parameter

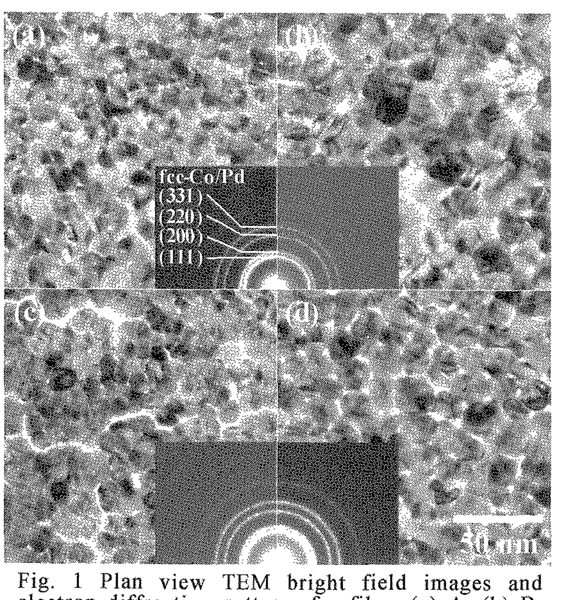

Fig. 1 Plan view TEM bright field images and electron diffraction patterns for films (a) A, (b) B, (c) C, and (d) D.

reflecting the magnitude of intergranular exchange coupling [6] and have been reported to be closely related to magnetic cluster diameter ($D_{cluster}$) [7], was measured by using a VSM. The squareness ratio ($SQR = M_r / M_s$, where M_r is remanent magnetization) was equal to unity within the experimental error and a nucleation field (H_n) exhibited a large negative value (< −2.3 kOe) for all samples used. The values of H_c, which is coercivity in the direction perpendicular to the film surface, for films B and C are small as compared with films A and D. The α value of film C is smaller than that of film A, which is thought to come from the decrease in not only the intergranular exchange coupling but also the inner-granular exchange coupling, because the value of M_s for film C is markedly reduced. Film D possesses the values of M_s and H_c as much as film A does, although it exhibits a smaller α value than film A. Thus, it was revealed that the intergranular exchange coupling of the Co/Pd multilayered film was successfully suppressed without a decrease in H_c by depositing the film upon the heated substrate under Ar gas containing N_2. Table II also shows the average and standard deviation of $D_{cluster}$ obtained from the MFM images at the ac demagnetized state. The smaller magnetic clusters are observed in the film exhibiting the lower α value. Namely, the sputtering technique proposed in this study does a lot for the decrease in $D_{cluster}$.

3.2 TEM observations

Figure 1 shows plan-view TEM bright field images and electron diffraction patterns for films A-D. The average and standard deviation of the Co/Pd crystal grain diameter (D_{grain}) for films A-D obtained from the TEM images shown in Fig. 1 is listed in Table II. Deposition upon the heated substarte enlarges D_{grain} in films B and D. The values of D_{grain} are not varied whether N_2 gas is added or not. In film C, broad grain boundaries are locally yielded by N_2 gas addition. The grain boundaries turned out to be voids with low density by EDS measurement. Combining the substrate heating process with the process of N_2 gas addition in film D, the voids uniformly disperse to the whole of the film and the crystal grains are physically isolated with the vacant grain boundaries. The decrease in intergranular exchange coupling for film D is considered to result from the enhancement in isolation of Co/Pd crystal grains. EDS experiments revealed that nitrogen was detected in both the crystal grains and grain boundaries for films B and D, but the two-dimensional distribution of nitrogen could not be determined quantitatively. In the electron diffraction patterns, the diffraction rings were completely identified as reflections of the fcc-Co/Pd structure, and no diffraction patterns of nitrides was observed. The values of $D_{cluster}$ are not necessarily interrelated with those of D_{grain}, as shown in

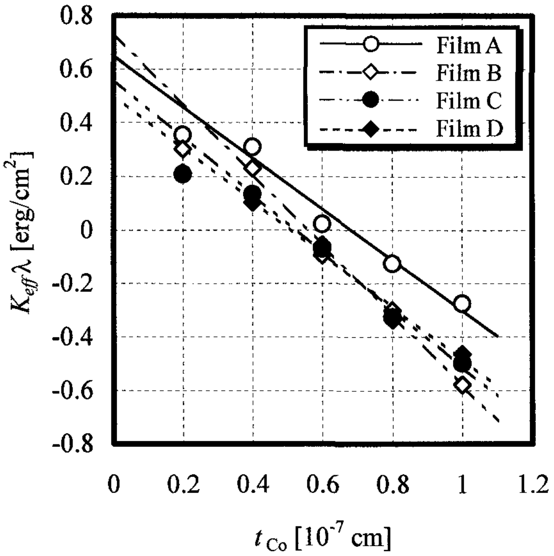

Fig. 2 Relationship between $K_{eff}\lambda$ and t_{Co} for films A-D.

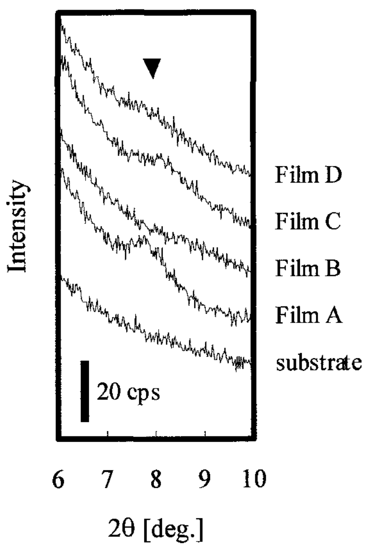

Fig. 3 XRD patterns in the range of 2θ = 6-10 deg. for films A-D together with that for glass substrate. The sign ▼ indicates the peak corresponding to the bi-layer periodicity of Co and Pd layers.

Table II. This result has us confirm that the isolation of crystal grains greatly contributes to the decrease of intergranular exchange coupling.

3.3 Evaluation of surface magnetic anisotropy

The sputtering techniques proposed in this study, *i.e.*, substrate heating and N_2 gas addition decrease the intergranular exchange coupling, but at the same time slightly degrade the perpendicular magnetic anisotropy, as shown in Table II. The perpendicular magnetic anisotropy of Co/Pd multilayered films attributes to surface magnetic anisotropy generated at the interfaces between Co and Pd sub-layers [1]. The surface magnetic anisotropy of films A-D can be evaluated from the relationship between $K_{eff}\lambda$ and t_{Co}, as shown in Fig. 2, by using the following phenomenological equation [1, 8]:

$$K_{eff}\lambda = 2K_s + K_v t_{Co}, \qquad (2)$$

where λ is the Co/Pd bi-layer period, K_s is surface magnetic anisotropy energy, K_v is volume magnetic anisotropy energy, and t_{Co} is the thickness of a constituent Co layer. In this experiment, t_{Co} was varied from 0.2 nm to 1 nm, whereas the thickness of a constituent Pd layer was fixed at 0.8 nm. Table III lists the values of K_s and K_v for films A-D, which are determined as the intercept with the ordinate and as the slope of the straight line obtained by the least-square fitting in Fig. 2, respectively. In the case of t_{Co} = 0.2 nm, the linearity between $K_{eff}\lambda$ and t_{Co} disappears, which is considered to be caused by the discontinuity of ultra-thin Co layers in the lateral direction and/or the decrease of magneto-elastic energy. Maesaka and Ohmori have reported that this variation of magneto-elastic energy is owing to the lattice strain relaxation based on the direct observations of lattice images with high-resolution TEM [9]. Therefore, the plots of t_{Co} = 0.2 nm of films A-D were excluded from the least-square fittings. The larger values of K_s and K_v in a positive sign serve for the higher perpendicular magnetic anisotropy. N_2 gas addition weakens the surface magnetic anisotropy in films C and D. On the contrary, film B deposited upon the heated substrate without N_2 gas shows larger values of K_s and $-K_v$ than film A does. Thus, it is considered that the degradations of perpendicular magnetic anisotropy in films C and D mainly result from the decrease in K_s and that in film B from the increase in $-K_v$.

Table III Values of K_s and K_v.

Film	K_s [erg/cm^2]	K_v [10^6 erg/cm^3]
A	0.325	-9.52
B	0.368	-13.2
C	0.282	-10.8
D	0.252	-9.89

An XRD measurement in the low angle region of 2θ is useful for evaluating the regularity of periodic bi-layer structure of Co and Pd layers. Figure 3 shows XRD patterns in the range of 2θ from 6 to 10 deg. for films A-D together with that for the glass substrate. The regularity of bi-layer structure contributes to an increase in peak intensity at the low angle region [10]. The peak intensity of film C decreases by adding N_2 gas, and that of film D becomes smaller than that of film C by combining substrate heating with N_2 gas addition. These variations are consistent with the decrease in the surface magnetic anisotropy shown in Table III. It may be said that the perpendicular magnetic anisotropy of films C and D is mainly influenced by the deterioration of the regular interfaces with the N_2 addition. The peak intensity of film B becomes smaller by heating the substrate than that of film A, although film B exhibits high surface magnetic anisotropy compared with film A. These results indicate that the magnetic anisotropy of

Table IV Magnetic properties and read-write characteristics for double-layered media.

Medium	T_{Sub} [°C]	P_{N2} [mTorr]	H_c [kOe]	α [-]	S_{p-p}/N_{rms} (200 kFRPI) [dB]
A'	20	0	6.1	5.3	0.7
D'	230	0.06	5.7	1.9	8.2

film B deposited upon the heated substrate without N_2 gas addition was not dominated only by the interfaces. It is considered that the increase of the values of K_s and $-K_v$ in film B is partly attributable to the lattice strain relaxation.

3.4 *R*/*W* characteristics

The above-mentioned results revealed that depositing upon the heated substrate with additive N_2 gas decreased the intergranular exchange coupling of Co/Pd multilayered films. Then, the double-layered media with SUL were fabricated by using the sputtering technique, and the *R*/*W* characteristics for the media were examined.

Table IV shows magnetic properties and *R*/*W* characteristics for the media composed of films A and D, which are designated as media A' and D', respectively. The values of H_c were measured by using a polar Kerr magnetometer. For the estimation of α values, the values of M_s for the single-layered films with the VSM shown in Table II were used. In the case of the double-layered media, the sputtering technique proposed in this study was also effective for suppressing the interglanular exchange coupling, which results in low α value for medium D'.

There was little difference between the reproduced voltage for media A' and D'. On the other hand, the medium noise for the medium D' was much lower than that for medium A', in particular at high linear recording density over 200 kFRPI. As a result, medium D' exhibited higher *S*/*N* ratio by 7-8 dB than medium A' over 200 kFRPI. This improvement of *R*/*W* characteristics in medium D' is attributed to the decrease in the intergranular exchange coupling leading to suppression of the formation of large magnetic clusters.

4. CONCLUSION

Effects of sputtering conditions, *e.g.*, substrate temperature during deposition and N_2 gas addition to Ar sputtering gas, on magnetic properties, especially intergranular exchange coupling, and crystalline microstructure of Co/Pd multilayered films were investigated. By depositing the Co/Pd multilayered film at 230 °C with N_2 partial pressure of 0.06 mTorr, the intergranular exchange coupling of the film was suppressed without a decrease in H_c, which came from Co/Pd crystal grains physically isolated with vacant grain boundaries. The recording medium used this film exhibited better *R*/*W* characteristics than that used the film deposited at room temperature without N_2 gas addition. From these results, the optimization of sputtering conditions is confirmed to be one of the most effective ways to fabricate the Co/Pd multilayered film for a high density magnetic recording medium.

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