

## Effect of Pd Seed Layer on Magnetic Anisotropy in Co Films Evaporated at Oblique Incidence

Kikuo ITOH, Mitsutoshi TOMITA, Shuhei TAKAHASHI, Keita AKASAKI,  
Fusao ICHIKAWA and Yoshie NOGUCHI

*Department of Physics, Kumamoto University, Kumamoto 860-8555*

(Received September 30, 2010)

The effect of Pd seed layer on the magnetic anisotropy in Co films evaporated at oblique incidence was investigated. The thickness of Pd was 500 Å and that of Co was varied from 100 to 600 Å. The substrate temperatures were  $-60$ ,  $-30$  and  $20$  °C. Negatively large anisotropy of the squareness ratio appeared in the films prepared at low substrate temperatures  $-60$  and  $-30$  °C though the anisotropy of the reflection coefficient and the magnetic anisotropy field were positive. The degree of the  $c$ -axis orientation perpendicular to the incidence plane seems to be a little larger than that of parallel one. From these results, it is considered that the positive in-plane magnetic anisotropy may originate from the shape anisotropy of the columnar grains and/or magnetocrystalline anisotropy, and the anisotropy of the squareness ratio does not reflect the in-plane magnetic anisotropy. In the films with negatively large anisotropy of the squareness ratio, large coercive force perpendicular to the film plane appeared. The  $MH$  loop in those films can be explained to be superposition of two loops with small and large coercive forces. We consider that the coercive force of under Co layer is small and that of over Co layer large.

### §1. Introduction

Systems like thin films and multilayers of Pd/Co with Co thickness of few monolayer have been intensively studied<sup>1)</sup> due to their special magnetic properties such as perpendicular magnetic anisotropy (PMA) and enhanced magnetic moment at surface and interfaces.<sup>2)</sup> However, the correlation between magnetic properties and the structure of the system has not been understood completely yet.

We have so far investigated the uniaxial magnetic anisotropy in obliquely deposited films.<sup>3)</sup> There, the easy axis of the anisotropy in the film plane is either perpendicular or parallel to the incidence plane and the anisotropy has been expressed with positive or negative sign in the former or latter case respectively. The magnetic anisotropy in obliquely deposited films depends on deposition parameters, such as deposition rate,<sup>4)</sup> pressure during deposition,<sup>5)</sup> substrate temperature,<sup>6)</sup> incidence angle,<sup>7)</sup> seed layer<sup>8)</sup> and so on. From the data accumulated so far, the most effective parameter seems to be the substrate temperature.

On the basis of the results of our preliminary experiment it is expected that the PMA may appear in Pd/Co films with larger thickness of Co layer. We prepared [Pd/Co]<sub>2</sub> films with Co layer thickness of 100 ~ 600 kÅ at the substrate temperatures of  $-60$ ,  $-30$  and  $20$  °C. In this paper, we present the results of the magnetic measurements, ellipsometry and X-ray analysis, and discuss the Pd seed effect on the magnetic anisotropy in Co films.

## §2. Experimental details

The Pd/Co films were vapor deposited on glass microscope-slide substrates at the substrate temperatures  $T_S$  of  $-60$ ,  $-30$  and  $20$  °C. The deposition rate of Pd layer  $R(\text{Pd})$  was  $20$  Å/s and that of Co layer  $R(\text{Co})$  was  $0.6$  Å/s. The nominal thickness of Pd layer  $t(\text{Pd})$  was  $500$  Å and that of Co  $t(\text{Co})$  was varied from  $100$  to  $600$  Å. The incidence angle of Pd vapor beam was  $30^\circ$  and that of Co  $60^\circ$ . The vacuum system used an oil diffusion pump with a liquid-nitrogen-cooled trap and its base pressure was about  $2 \times 10^{-4}$  Pa. The pressure during evaporation was  $4.0 \times 10^{-3}$  Pa, which was maintained by introducing argon gas automatically through a leak valve. The magnetic measurement was carried out using a vibrating sample magnetometer (VSM) and an automatic balancing magnetometer. The geometrical alignment of Co columnar grains was followed by ellipsometry<sup>9)</sup> where the light source was an He-Ne gas laser and its wavelength was  $633$  nm. The crystallographic preferred orientation was analyzed by drawing the pole figures, the pole densities of which were determined using X-ray diffraction with Cu  $K\alpha$  radiation (SmartLab, Rigaku).

## §3. Results and discussion

Figure 1 is the schematic representation of the vapor beam direction, film normal ( $z$  axis), incidence plane ( $yz$  plane) and film plane ( $xy$  plane). In Fig. 2, the anisotropies of the squareness ratio  $\Delta M_r/M_S$  of  $[\text{Pd}/\text{Co}]_2$  films deposited at the substrate temperatures of  $-60$  (■),  $-30$  (●) and  $20$  °C (▲) are shown as a function of Cobalt thickness  $t(\text{Co})$ , where  $\Delta M_r$  is the difference between the remanences in the directions perpendicular and parallel to the incidence plane and  $M_S$  is the saturation magnetization. The anisotropy  $\Delta M_r/M_S$  is considered to be a measure of the magnetic anisotropy in cobalt films.<sup>10)</sup> The anisotropies  $\Delta M_r/M_S$  are negative in all  $t(\text{Co})$  range irrespective of the substrate temperature. The observed magnetic anisotropy in Cobalt films has so far been analyzed by taking account of the following two anisotropies: (1) The anisotropy of the demagnetizing field due to the shape anisotropy derived from the columnar grain structure.<sup>11)</sup> (2) The magnetocrystalline anisotropy through the textural structure.<sup>12)</sup> The shape anisotropy due to the columnar grain structure can be detected by ellipsometry. or the geometrically aligned columnar grains the reflection of polarized light depends on the direction of the electric vector. The reflection coefficient is highest when the vector is parallel to the alignment. Figure 3 is a plot of the anisotropy of the reflection coefficient  $\Delta r/r$  of  $[\text{Pd}/\text{Co}]_2$  films deposited at the substrate temperature of  $-30$  °C (●) as a function of  $t(\text{Co})$  where  $\Delta r$  is the difference between the coefficients in the directions perpendicular and parallel to the incidence plane and  $r$  is the average coefficient. The positive and negative signs of  $\Delta r/r$  indicate the alignment in the perpendicular and parallel to the incidence plane, respectively. The anisotropy  $\Delta r/r$  in Fig. 3 is almost positive though  $\Delta M_r/M_S$  in Fig. 2 is negative. This result suggests that the negative  $\Delta M_r/M_S$  is not due to the shape anisotropy but the magnetocrystalline anisotropy.

In order to discuss the contribution of the magnetocrystalline anisotropy to the

magnetic anisotropy the  $\{0002\}$  pole figure of  $[\text{Pd}/\text{Co}]_2$  films marked by A in Figs. 2 and 3 ( $t(\text{Co}) = 0.3 \text{ k}\text{\AA}$ ) is represented in Fig. 4. The center of the plot is the film normal and the solid circle indicates the Co vapor beam direction. The  $\{0002\}$  pole is in the vicinity of the film plane and the degree of its orientation perpendicular to the incidence plane seems to be a little larger than that of parallel one. This result suggests that the magnetocrystalline anisotropy through the  $c$ -axis orientation may contribute positively to the in-plane magnetic anisotropy. Therefore it is considered that the anisotropy  $\Delta M_r/M_S$  does not reflect the in-plane magnetic anisotropy. The in-plane magnetic anisotropy can be estimated precisely by the torque measurement. Figure 5 shows the magnetic anisotropy field  $H_k = 2L/M_S$  of the same films as in Fig. 3 as a function of  $t(\text{Co})$ , where  $L$  is the amplitude of torque curve in the film

plane and  $M_S$  is the saturation magnetization. The field  $H_k$  is positive in all  $t(\text{Co})$  range suggesting that the in-plane magnetic anisotropy originates from the shape anisotropy of the columnar grains.

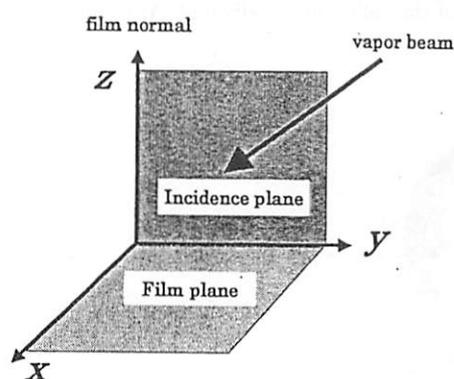


Fig. 1. The schematic representation of the vapor beam direction, film normal ( $z$  axis), incidence plane ( $yz$  plane) and film plane ( $xy$  plane).

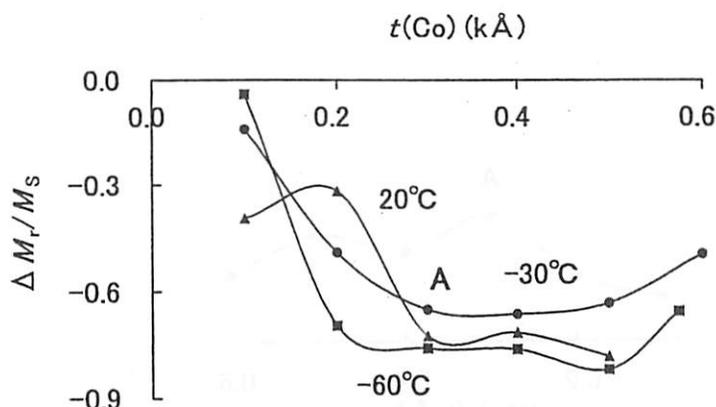


Fig. 2. The  $t(\text{Co})$  dependences of the anisotropy of the squareness ratio  $\Delta M_r/M_S$  for  $[\text{Pd}/\text{Co}]_2$  films deposited at the substrate temperatures of  $-60$  ( $\blacksquare$ ),  $-30$  ( $\bullet$ ) and  $20$  ( $\blacktriangle$ )  $^\circ\text{C}$ .

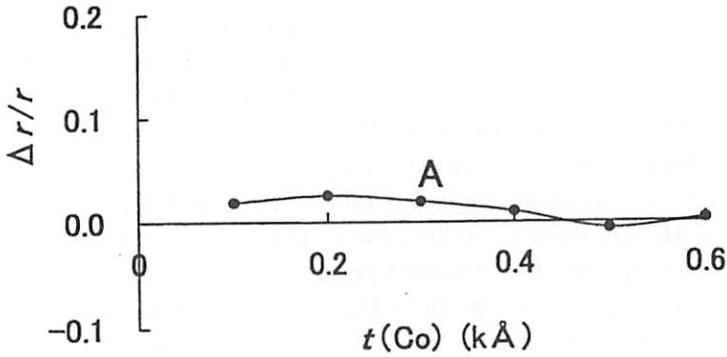


Fig. 3. The  $t(\text{Co})$  dependence of the anisotropy of the reflection coefficient  $\Delta r/r$  for  $[\text{Pd}/\text{Co}]_2$  films deposited at  $-30^\circ\text{C}$ .

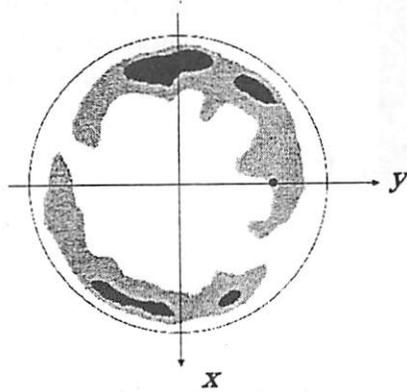


Fig. 4. The  $\{0002\}$  pole figure of  $[\text{Pd}/\text{Co}]_2$  films marked by A in Figs. 2 and 3 ( $t(\text{Co}) = 0.3 \text{ kÅ}$ ).

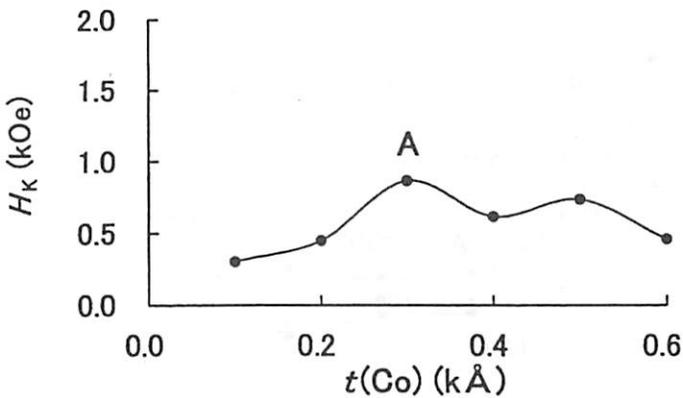


Fig. 5. The  $t(\text{Co})$  dependence of the magnetic anisotropy field  $H_k$  for the same  $[\text{Pd}/\text{Co}]_2$  films as in Fig. 3.

Finally a short comment on the shape of the  $MH$  loop perpendicular to the film plane will be made. Figure 6 shows the coercive forces  $H_C(z)$  perpendicular to the film plane of the same films as in Fig. 2. The values of  $H_C(z)$  for  $-60$  and  $-30$  °C are beyond 1.2 kOe and much larger than that for  $20$  °C. The value of  $H_C(z)$  beyond 1.2 kOe can not be seen in the pure cobalt films. Figure 7 shows the  $MH$  loop for the same  $[\text{Pd}/\text{Co}]_2$  film as in Fig. 4 which is marked by A in Figs. 2, 3, 5 and 6. As is seen, there are sharp bends in the loop. From the shape of the minor loop we supposed that the loop in Fig. 7 is superposition of two loops with small and large coercive forces. In Figs. 8 (a), (b) and (c) two calculated loops and superposition of these loops, which are denoted by A1, A2 and A1+A2, respectively, are represented. The loop in Fig. 8 (c) very resembles that in Fig. 7. We consider that the  $H_C(z)$  of under Co layer is small and that of over Co layer large.

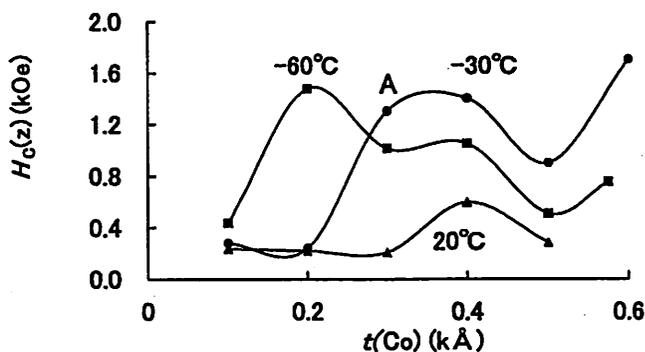


Fig. 6. The  $t(\text{Co})$  dependences of the coercive forces  $H_C(z)$  perpendicular to the film plane for the same  $[\text{Pd}/\text{Co}]_2$  films as in Fig. 2:  $-60$  (■),  $-30$  (●) and  $20$  °C (▲).

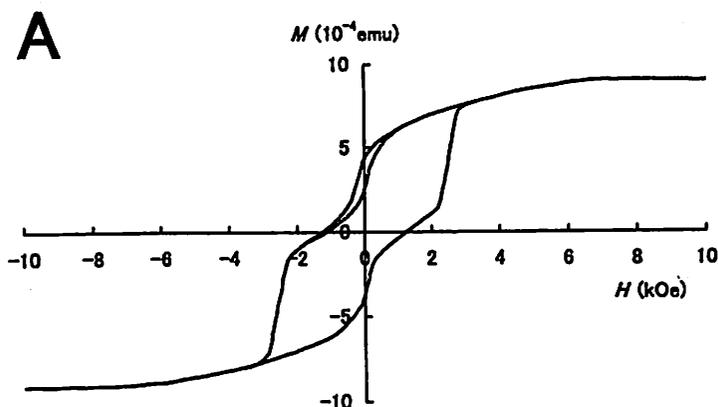


Fig. 7. Observed  $MH$  loop for the same  $[\text{Pd}/\text{Co}]_2$  film as in Fig. 4 which is marked by A in Figs. 2, 3, 5 and 6 ( $t(\text{Co}) = 0.3$  kÅ).

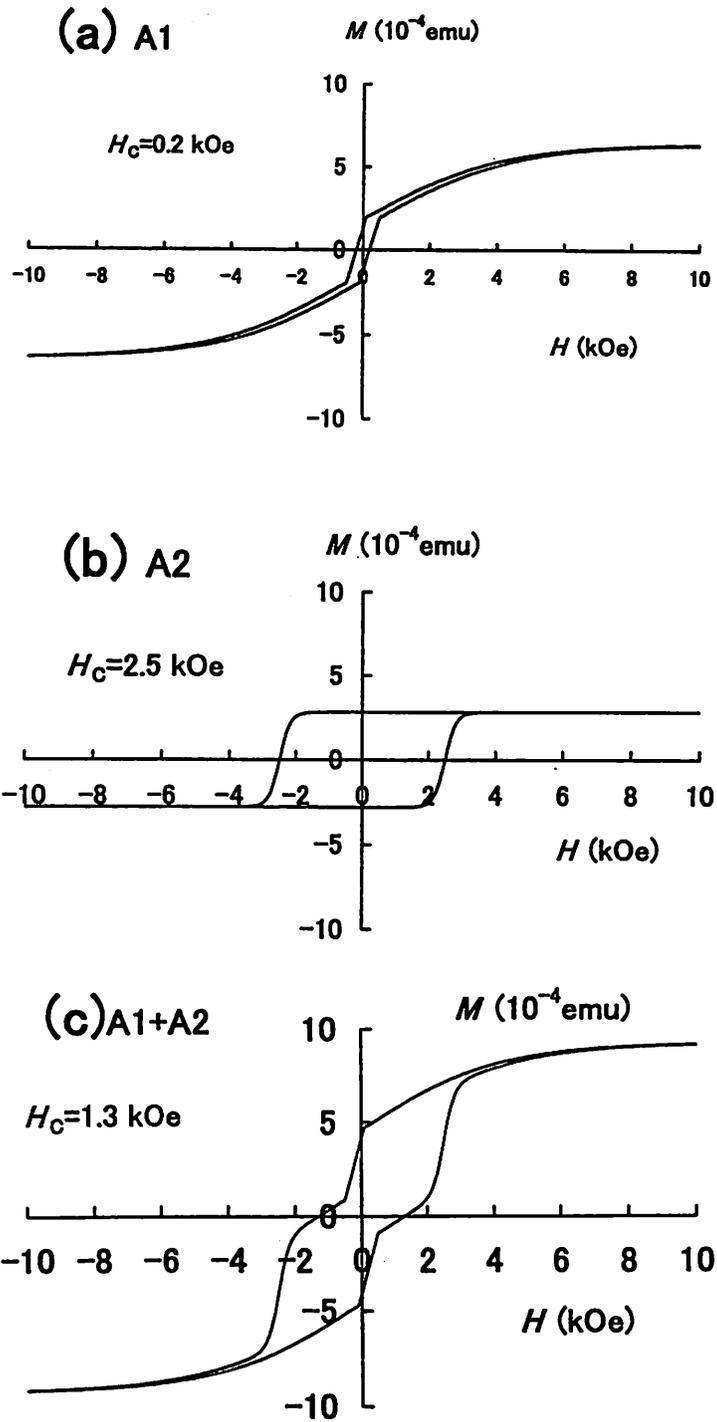


Fig. 8. Calculated  $MH$  loops: (a)  $H_C(z) = 0.2$  kOe, (b)  $H_C(z) = 2.5$  kOe and (c)  $H_C(z) = 1.3$  kOe.

#### §4. Conclusions

The effect of Pd seed layer on the magnetic anisotropy in Cobalt films evaporated at oblique incidence was investigated. The deposition rate of Pd layers  $R(\text{Pd})$  was 20 Å/s and that of Co layers  $R(\text{Co})$  was 0.6 Å/s. The thickness of Pd  $t(\text{Pd})$  was 500 Å and that of Co  $t(\text{Co})$  was varied from 100 to 600 Å. The incidence angle of Pd vapor beam was 30° and that of Co 60°. The substrate temperatures were -60, -30 and 20 °C. Results obtained are as follows.

- (1) Negatively large anisotropy of the squareness ratio  $\Delta M_r/M_S$  appeared in the films prepared at low substrate temperatures -60 and -30 °C though the anisotropy of the reflection coefficient  $\Delta r/r$  was positive.
- (2) The {0002} pole in the films with negatively large  $\Delta M_r/M_S$  was in the vicinity of the film plane and the degree of its orientation perpendicular to the incidence plane seems to be a little larger to that of parallel one suggesting that the magnetocrystalline anisotropy through the  $c$ -axis orientation may contribute positively to the in-plane magnetic anisotropy.
- (3) The magnetic field  $H_k$  is positive in these films suggesting that the in-plane magnetic anisotropy originates from the shape anisotropy of the columnar grains and/or the magnetocrystalline anisotropy.
- (4) In the films with negatively large  $\Delta M_r/M_S$ , large coercive force  $H_C(z)$  perpendicular to the film plane appeared. The  $MH$  loop in those films is explained to be superposition of two loops with small and large coercive forces. We consider that the  $H_C(z)$  of under Co layer is small and that of over Co layer large.

From the results (1) and (3) it is considered that the anisotropy of the squareness ratio  $\Delta M_r/M_S$  does not reflect the magnetic anisotropy in the film plane. In order to discuss the difference between the signs in  $\Delta M_r/M_S$  and  $H_k$  further investigation is necessary.

#### References

- 1) Y. P. Lee, S. K. Kim, J. S. Kang, Y. M. Koo, J. I. Jeong, J. H. Hong and H. J. Shin, *J. Magn. Magn. Mater.* **126** (1993), 316.  
A. M. Baker, A. Cerezo, A. K. Petford-Long, *J. Magn. Magn. Mater.* **156** (1996), 83.  
T. Onoue, J. Kawaji, K. Kuramachi, T. Asahi and T. Osaka, *J. Magn. Magn. Mater.* **235** (2001), 82.  
T. Asahi, K. Kuramochi, J. Kawaji, T. Onoue and T. Osaka, *J. Magn. Magn. Mater.* **235** (2001), 87.  
H. S. Lee, S. B. Choe, S. C. Shin, C. G. Kim, *J. Magn. Magn. Mater.* **239** (2002), 343.  
S. Nakagawa and H. Yoshikawa, *J. Magn. Magn. Mater.* **287** (2005), 193.  
F. Luo, L. Yan, M. Przybylski, Y. Shi and J. Kirschner, *J. Magn. Magn. Mater.* **316** (2007), e342.
- 2) B. Heinrich and J. A. C. Bland, *Ultrathin Magnetic Structure* (Springer, Berlin, 1994), p. 21.
- 3) M. Prutton, *Thin Ferromagnetic Films* (Butterworths, London, 1964), p. 223.  
R.F. Soohoo, *Magnetic Thin Films* (Harper & Row, New York, 1965), p. 119.  
K. L. Chopra, *Thin Film Phenomena* (McGraw-Hill, New York, 1969), p. 629.
- 4) K. Okamoto, T. Hashimoto, K. Hara, M. Kamiya and H. Fujiwara, *Thin Solid Films* **147** (1987) 299.
- 5) K. Okamoto, T. Hashimoto, K. Hara, M. Kamiya and H. Fujiwara, *Thin Solid Films* **129** (1985), 299.

- 6) K. Itoh, F. Ichikawa, Y. Takahashi, K. TsuTsumi, Y. Noguchi, K. Okamoto, T. Uchiyama and I. Iguchi, *Jpn. J. Appl. Physics*, **45** (2006), 2534.
- 7) K. Okamoto and K. Itoh, *Jpn. J. Appl. Physics*, **44** (2005), 1382.
- 8) T. Hashimoto, K. Okamoto, K. Hara, M. Kamiya and H. Fujiwara, *Thin Solid Films* **169** (1989), 289.
- 9) M. Kamiya, K. Hara, T. Hashimoto, K. Okamoto and H. Fujiwara, *J. Phys. Soc. Jpn.* **52** (1983), 3885 [Erratum; **53** (1984), 468].
- 10) K. Hara, K. Itoh, M. Kamiya, K. Okamoto and T. Hashimoto, *J. Magn. Magn. Mater.* **161** (1996), 287.
- 11) K. Okamoto, T. Hashimoto, H. Fujiwara, K. Hara and M. Kamiya, *J. Magn. Magn. Mater.* **81** (1989), 374.
- 12) K. Okamoto, K. Itoh and T. Hashimoto, *J. Magn. Magn. Mater.* **87** (1990) 379.