Magnetic and magneto-optical properties of sputtered Co/Pd multilayers

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Co/Pd multilayer thin films consisting of Co layers between 2–25 Å and Pd layers of typically 14 Å were prepared by rf sputtering. Hysteresis loops and the magnetization were measured parallel and perpendicular to the film plane. From these measurements and torque curves it was found that the multilayers show a transition of the easy axis from an in-plane to a direction perpendicular to the film plane when decreasing the Co layer thickness below 10 to 6 Å depending on the sputter rate and sputter gas used (Ar or Kr). Below 4.5 Å Co layer thickness M_r/M_s is nearly one. The coercivity increases with decreasing Co layer thickness also depending on the sputter parameters used and the thickness of the seedlayer. The hysteresis loops measured with the Kerr-tracer did not show much difference with those measured with the VSM. Although TEM photographs show very clearly a layered structure when a seedlayer is used, low angle X-ray measurements seem to indicate sharper interfaces in multilayers where no or a very thin seedlayer is used. From the recording point of view the best results (high coercivity, squareness and anisotropy) were obtained by using Ar gas at high pressure.

1. Introduction

Metallic magnetic multilayers are of interest because of the possibility to use them in magneto-optical storage applications. In these multilayers the magnetic properties and the magneto-optical properties are strongly influenced by the thickness of the ferro-magnetic layer as well as the non-magnetic layer, the sharpness of the interfaces, the crystal structure, the texture and the number of layers. These properties in their turn are influenced by the process parameters. To study these effects samples of Co/Pd multilayers were prepared by rf sputtering using Ar and Kr gas. Hysteresis loops were measured by VSM and Kerr-tracer, anisotropy constants by Torque magnetometer, the structure by low angle X-ray and TEM measurements.

2. Experimental procedure

Co/Pd multilayers were rf sputtered using a Leybold Z-400 system in which one target position was used to fix two small targets of 4 cm diameter. The substrates were positioned below the targets on a rotating table and at a distance of 4 cm. Si(100) wafers were used as substrates. The sputtering chamber was evacuated with a turbo-molecular pump to a background pressure of typically 1×10^{-7} mbar. The thickness of the Co and Pd layers (t_{Co} and t_{Pd}) was determined by measuring the exposure time combined with Dek-tak or low angle X-ray measurements. Three different Ar pressures ($P_{\text{Ar}} = 1.2 \times 10^{-2}$, 5.2×10^{-2} and 7.5×10^{-2} mbar) were used with corresponding rates of 0.5, 0.9 and 1.2 Å/s for Co and of 0.8, 2.1 and 3.3 Å/s for Pd, while at 7.5×10^{-2} mbar another rate was applied (1.5 Å/s for Co and 4.4 Å/s for Pd) by changing the rf power. For Kr the pressure was kept constant ($P_{\text{Kr}} = 6.5 \times 10^{-2}$ mbar) and also the rate (1.0 Å/s for Co and 2.8 Å/s for Pd). The number of bilayers (N) was typically 25 for all films. Most of the films were deposited on a seedlayer of Pd, which is a

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Table 1

Data Co/Pd multilayer. In the table two values of M_s are given: $M_s(\text{Co-vol})$ which is the magnetization corresponding to the total Co volume and $M_s(\text{T-vol})$ which is the magnetization corresponding to the total multilayer volume

Sample	t_{Co} [Å]	t_{Pd} [Å]	$M_s(\text{Co-vol})$ [kA/m]	$H_{\text{c(perp)}}$ [kA/m]	M_r / M_s (perp)	$M_s(\text{T-vol})$ [kA/m]	K_{eff} [kJ/m ³]	θ_k [deg]
$P_{\text{Ar}} = 7.5 \times 10^{-2}$ mbar; Co-rate 1.23 Å/s and Pd-rate 3.33 Å/s, seedlayer 200 Å								
901203-1	2.1	13.6	2426	165	0.88	319	2000	0.06
910111-2	4.3	14.3	1581	88	0.97	366	1548	0.12
910128-2	6.7	14.3	1570	18	0.10	504	530	0.15
910129-2	8.2	14.3	1612	11	0.05	591	290	0.17
910130-2	10.4	14.3	1500	8	0.02	634	-74	0.20
910131-2	16.6	14.3	1385	12	0.02	745	-373	0.23
910201-2	22.7	14.3	1340	12	0.02	823	-486	0.22
$P_{\text{Ar}} = 7.5 \times 10^{-2}$ mbar; Co-rate 1.46 Å/s and Pd-rate 4.36 Å/s, seedlayer 200 Å								
910808-1	2.3	14.1	2445	152	0.96	341	1615	0.05
910807-1	4.5	14.1	2026	96	0.96	492	1006	0.09
910809-1	6.8	14.1	2055	40	0.11	669	213	0.13
910814-1	8.5	14.1	1850	36	0.10	703	42	0.14
910816-1	12.7	14.1	1759	24	0.05	826	-306	0.18
$P_{\text{Ar}} = 7.5 \times 10^{-2}$ mbar; Co-rate 1.46 Å/s and Pd-rate 4.36 Å/s, seedlayer 14 Å								
910808-3	2.3	14.1	2445	80	0.98	341	1471	
910805-3	4.5	14.1	2110	30	0.29	513	818	
910809-3	6.8	14.1	2055	8	0.02	669	131	
910814-3	8.5	14.3	1850	16	0.04	703	-35	
910816-3	12.7	14.1	1759	16	0.04	826	-370	
$P_{\text{Kr}} = 6.5 \times 10^{-2}$ mbar, Co-rate 0.99 Å/s and Pd-rate 2.81 Å/s, seedlayer 200 Å								
910618-1	2.2	14.1	2055	104	1.00	275	1330	0.05
910617-1	3.7	14.1	1876	61	1.00	392	778	0.09
910613-1	5.6	14.1	1639	44	0.28	465	246	0.11
910620-1	10.1	144	1478	16	0.07	611	-565	0.18

Fig. 1. Low angle X-ray reflection scan for a multilayer sputtered with Ar (a) or Kr (b) for two different seedlayers.

Fig. 2. TEM photographs of a Co/Pd film ($200 \text{ \AA} + 25(10.5 \text{ \AA} + 14.3 \text{ \AA})$): (a) bright field image; (b) dark field.

layer between the Si substrate and the multilayer. The crystallographic properties and the layered structure of the films were studied by low angle X-ray measurements and by TEM.

The magnetic fields applied at the vibrating sample magnetometer and at the torque magnetometer were at maximum respectively 800 and 1400 kA/m. The coercive field (H_c), the remanent magnetization (M_r) and the saturation magnetization (M_s) were calculated from the perpendicular hysteresis loops. From the torque measurements the surface anisotropy constant (K_s) and the volume anisotropy constant (K_v) could be calculated. The hysteresis loops were also measured with a Kerr-tracer. From these measurements the Kerr-rotation (θ_k) was determined. For one series also the reflectivity of the films was measured with the Kerr-tracer and together with θ_k a figure of merit could be calculated.

3. Results and discussion

3.1. Microstructure

Except for the X-ray measurements most of the figures shown here are derived from the data presented in table 1.

From the low angle X-ray measurements it seems (fig. 1) that the interfaces in films with a thin seedlayer of 14 Å (solid line) are sharper (higher Bragg peak) than with a thick seedlayer of 200 Å (dotted line). Only in a few films a clear layered structure can be detected by low angle measurements, even when no seedlayer is used, but the plots in figure 1 do give a trend. TEM photographs (fig. 2) of one of the films (200 Å + 25 × (10.5 Å + 14.3 Å)) clearly show the layered structure. A columnar structure (diameter 400 Å) exists, with a convex curvature of the surface with a height of approximately 50–75 Å, within the large columns, a fibre-like structure can be detected (see photograph). The layered structure seen in the large columns is following the surface curvature. It means that the interfaces are not flat but can deviate from the substrate plane as much as 14°, which is together with a larger surface roughness caused by the

Fig. 3. $K_{eff}t_{Co}$ vs. t_{Co} for different sputter gasses, pressures, seedlayers and rates.

seedlayer most likely the reason why the layered structure does not show in the low angle X-ray measurements. To study the layered structure with X-ray methods, high angle measurements will probably give better results, because the roughness of the surface and the interfaces is in that case a less disturbing factor. Similar results for Co/Au multilayers are mentioned by Folkerts et al. [9] and for Co/Pd and Co/Au by Hakkens [10]. Whether stress caused by the misfit of Co and Pd (10%) is the reason for the curvature or the growth process through nucleation and shadowing remains to be seen.

3.2. Anisotropy contributions

The effective anisotropy K_{eff} (per unit Co volume) is calculated from the torque curves and include also the shape anisotropy. With $t_{Co}K_{eff} = 2K_s + t_{Co}K_v$ both the surface and the volume anisotropy constants K_s and K_v can be obtained from fig. 3. It seems from these curves that in the case of films sputtered with Ar and for $t_{Co} > t_{crit}$ the surface anisotropy which is thought to be responsible for the perpendicular anisotropy is varying between $K_s = 0.40$ and $0.53 \times 10^{-3} \text{ J/m}^2$ depending on the thickness of the seedlayer and the sputter-rate. The thickness of the Co layer where the structure changes from coherent to incoherent is called t_{crit} [8]. The volume anisotropy for films sputtered with Ar is in all cases the same $K_v = -0.98 \times 10^6 \text{ J/m}^3$. For films

Fig. 4. VSM hysteresis loops for different Co thicknesses, sputtered with Ar at a higher rate on a seedlayer of 200 Å Pd. The dotted lines show the in-plane and the solid lines the perpendicular loops.

sputtered with Kr these values are $K_s = 0.50 \times 10^{-3} \text{ J/m}^2$ and $K_v = -1.1 \times 10^6 \text{ J/m}^3$. Comparing these results with what has been found by others [2,5,7] is difficult because the deposition conditions, which can have a large influence on the anisotropy, were not the same. Because the K_v for all the films sputtered with Ar is the same and independent of the sputter rate and of the thickness of the seedlayer, means that the change in the K_{eff} is due to a change in the K_s . According to den Broeder et al. [8] there are two contributions to K_s in the case of incoherent films ($t_{\text{Co}} > t_{\text{crit}}$), one due to the misfit strain anisotropy K_λ and the other K_N the Néel surface anisotropy. In the case of coherent films ($t_{\text{Co}} < t_{\text{crit}}$) only the K_N has a contribution to K_s . In our case (fig. 3) it seems that all our films have a K_N of approximately $0.12 \times 10^{-3} \text{ J/m}^2$. The films sputtered with the lower rate have the highest $K_\lambda = 0.44 \times 10^{-3} \text{ J/m}^2$ ($K_s = 0.53 \times 10^{-3} \text{ J/m}^2$), the films sputtered with the higher rate have independent of the seedlayer the same value for $K_\lambda = 0.28 \times 10^{-3} \text{ J/m}^2$ ($K_s = 0.40 \times 10^{-3} \text{ J/m}^2$) and in the films sputtered with Kr the strain anisotropy is higher again $K_\lambda = 0.37 \times 10^{-3} \text{ J/m}^2$ ($K_s = 0.5 \times 10^{-3} \text{ J/m}^2$). It means that the difference in the K_s for the different series is coming from strain mainly. The films sputtered with Ar seem to suggest a $t_{\text{crit}} = 4.5 \text{ Å}$ independent of the sputter rate and the seedlayer thickness, while the t_{crit} for the films sputtered with Kr tends to a lower value. Whether this value of 4.5 Å is a coincidence or really shows an agreement between the experiment and the theoretical value mentioned

for Co/Pd by den Broeder et al. [8] and van der Merwe et al. [11] remains to be seen.

3.3. Magnetic properties (H_c , M_s)

In fig. 4 a typical series of hysteresis loops is shown versus t_{Co} . Depending on the sputter parameters the hysteresis loops are square (M_r/M_s approximately 1) for t_{Co} less than 3.5 to 4.5 Å.

Fig. 5 shows the dependency of the H_c on the sputtering pressure for two different thicknesses of Co and Pd. The higher coercivity with higher sputtering pressures is probably due to the fact that the reflected neutral Ar atoms have less energy because of a shorter mean free path giving rise to less mixing of the Co and Pd atoms at the interfaces [6] and consequently a larger K_{eff} . A decreasing coercivity with an increasing thickness of the Co layer as shown in fig. 7 is also normally

Fig. 5. The H_c and K_{eff} as a function of P_{Ar} for two different t_{Co} and t_{Pd} , $N = 25$.

Fig. 6. K_{eff} and H_c vs. $t_{\text{Pd(seed)}}$ with $t_{\text{Co}} = 4.5 \text{ Å}$, $t_{\text{Pd}} = 14.1 \text{ Å}$, $P_{\text{Ar}} = 7.5 \times 10^{-2} \text{ mbar}$ and with the higher rate.

found. From the films sputtered with Ar it can be seen that the seedlayer has the largest influence (figs. 6 and 7). In fig. 6 it can also be seen that K_{eff} increases with the seedlayer thickness while in section 3.2 is shown that an increase of K_{eff} is probably caused by an increase in K_s . So the increase in coercivity is probably partly caused by an increase of the surface anisotropy K_s with increasing seedlayer thickness and partly by an increased roughness of the surface and interfaces of the film. Above a seedlayer thickness of approximately 200 Å H_c remains the same probably caused by a decreasing K_{eff} and an increasing surface roughness. The films sputtered with Kr cannot be compared properly with the Ar sputtered films because the deposition parameters (rate and gas pressure) are not the same but it

Fig. 7. The $H_{c(\text{perp})}$ vs. t_{Co} for different sputter gasses, pressures, seedlayers and rates.

Fig. 8. $M_s(\text{Co-vol})$ vs. t_{Co} for different sputter gasses, pressures and rates.

does not seem that the films sputtered with Kr show a tendency to higher coercivities as is the case with Co/Pt multilayers [6].

The magnetization for the films with perpendicular anisotropy is quite high, especially for the very thin t_{Co} region (fig. 8), but is comparable with the magnetization for evaporated films discussed in ref. [1] and probably due to an increase of the magnetic moment of the Co and a polarization of the Pd layers.

3.4. Magneto-optical properties

The hysteresis loops measured with the Kerr-tracer did not show much difference with the VSM loops. The coercivity and the squareness of the loops were the same. The Kerr rotation (fig.

Fig. 9. θ_k and the FOM vs. t_{Co} sputtered with Ar at a lower rate on a seedlayer of 200 Å Pd.

9) we found was in agreement with what others [5,6] have found. From one series of films also the reflectivity (R) was measured and the figure of merit was calculated $FOM = \theta^2 R$ and is plotted in fig. 9. The FOM found is also in agreement with the values found by others [5].

4. Conclusions

The differences in the anisotropy of our films, which are sputtered with Ar, are mainly caused by differences in the surface anisotropy. Films sputtered with Kr are not showing better properties than films sputtered with Ar.

In general it seems that the magnetic and magneto-optical properties of our films sputtered at high pressure are comparable with what is found for evaporated films. With evaporated films however it looks as if the interfaces are better defined.

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