SURFACE AND VOLUME HYSTERESIS OF SPUTTERED CoCr

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Abstract. Surface magnetic hysteresis, measured by Polar M.O. Kerr effect on RF and Magnetron sputtered CoCr (81/19 at%) films in the range 20 - 4000 nm is compared with volume hysteresis measured by VSM, both with external field perpendicular to the surface. The surface coercivity H_{CS} was found to decrease below the volume coercivity at a critical thickness of 125 nm for RF films [1] and at about 1100 and 800 nm for centre and off-centre Magnetron samples respectively. The slope of the Kerr loop at $\theta_K = 0$ increased slightly again deviating from the VSM slope at a film thickness of about 100, 300 and 500 nm for RF, centre and off-centre Magnetron CoCr samples respectively. These results are consistent with the formation of small spikes from the surface for films thicker than a critical value. Order of magnitude calculations do not oppose the formation of such small reversed domains and the higher critical thickness for Magnetron samples is also in qualitative agreement with these calculations, due to a higher perpendicular anisotropy by a factor 2 for these films. If spike domains do exist in CoCr the reversal mechanism is most likely one in which the reversed domains grow at the expense of the main domains.

INTRODUCTION

The reversal mechanism and associated with it the domain structure of CoCr thin films for perpendicular magnetic recording is not clear at present. There are two main models, namely the particulate and the continuous model. In the first one the CoCr columns that are formed during film deposition are believed to interact only through magnetostatic interaction. No exchange force acts over the column boundaries due to e.g. a higher concentration of Cr there. In the continuous model the reversal mechanism is thought to take place by Bloch walls as in stripe domains, hindered by the column boundaries [2]. On the base of Neutron Depolarisation and Magneto-Optic Kerr measurements a third intermediate model has been proposed, in which the magnetization reversal takes place by spike domain wall motion within the columns [1,4].

Using the Magneto-Optic Kerr Effect (MOKE) the hysteresis loop of the surface of CoCr can be measured, since the penetration depth of light ($\lambda = 632$ nm) into CoCr is about 15 nm. Comparison of surface and volume hysteresis could reveal a possible inhomogeneity in the magnetization distribution throughout the thickness of the film and thus more insight can be gained in the domain structure and reversal mechanism of the CoCr films.

Earlier we reported on the surface coercivity H_{CS} and the coercivity of the total film H_{CV} measured with a VSM as a function of film thickness [1]. A rotating analyser apparatus was used to measure the Kerr rotation as a function of the applied field. The experimental set up is described in [1]. Using a multiple-reflection sample holder the Kerr rotation could be measured with an accuracy of 0.002 deg. A maximum H_C was found at $t = 80$ nm for RF-films and at about 1000 nm for Magnetron films (both 81/19 at%). The coercivity ratio CR is defined by H_{CS}/H_{CV} . For RF-films a critical film thickness $t_c = 125$ nm was found, so that if $t < t_c$ then $CR > 1$ and if $t > t_c$ then $CR < 1$.

The presence of a critical thickness in Magnetron films was not clear from the reported data, therefore additional samples were Magnetron sputtered, also to cover the thicker films regions, and results are reported here. Secondly we will compare the slope of the easy-axis hysteresis loop measured with the Kerr effect and VSM at $\theta_K = 0$ and $M = 0$ respectively.

The functional dependence of the VSM slope on film thickness for RF-films was explained by Wielinga using a continuous domain model [2]. Hubert expanded the theoretical basis of this analysis to branched domain structures in CoCr [3].

SAMPLE PREPARATION AND CHARACTERIZATION

The CoCr samples were sputtered from alloyed CoCr (81/19 at%) targets of 4" and 3" inch diameter respectively, on Si (100)

substrates in a Leybold Heraeus RF-sputter apparatus (2400) either with or without Magnetron facilities. We will distinguish here between RF- and Magnetron films. RF-films were sputtered in batches of 4 samples under the optimized sputter parameters $P_{Ar} = 4 \times 10^{-3}$ mBar and $V_{RF} = -1.6$ kV [5] while for Magnetron films the optimum parameters for obtaining good perpendicular anisotropy were determined at $P_{Ar} = 6 \times 10^{-3}$ mBar and $V_{RF} = -250$ V. The target substrate distance was 5 and 3 cm respectively. As is well known from literature sputter rate and film properties in Magnetron sputtering depend on the position of the sample on the substrate. Two homogeneity groups were formed based on the film thickness and coercivity that coincided with a substrate position near the centre and off-centre. Here they will be referred to as group 1 and 2. The previously reported [1] Magnetron samples all belong to group 1 (near the centre). In addition to these, 5 series of 23 samples were Magnetron sputtered to cover a thickness range of 500 to 4200 nm. Of each series 8 samples fell in group 1 and 15 samples in group 2. X-ray fluorescence was used to check the composition and film thickness. The saturation magnetization M_s could be determined at 460 kA/m from these measurements together with VSM results, and was found to be almost independent of film thickness. Also a constant Kerr rotation of about 0.1 degree was found for all films, indicating that M_s and composition do not significantly depend on film thickness. Here the (magnetic) film thickness is determined from the magnetic volume of the sample ($M_s \cdot V$) determined by VSM ($\pm 3\%$), the sample surface (1 cm^2) and $M_s = 460 \text{ kA/m}$. The perpendicular anisotropy constant K_1 corrected for the sheet demagnetization $1/2 \mu_0 M_s^2$ was determined from torque measurements at 90 kJ/m^3 for RF-films [6] and now found to be 190 kJ/m^3 for magnetron films. No dependence on film thickness was found nor did K_1 differ significantly for Magnetron samples from group 1 or 2. In preparing thick Magnetron samples we were faced with the problem that the CoCr spontaneously peeled off the Si substrate due to internal stress. Presputtering the substrates at $V_{RF} = 1$ kV for 10-15 minutes prevented this problem. The thickest films however had to be measured quickly after preparation and still only 8 samples could be measured with both MOKE and VSM. Before peeling off occurred the metallic appearance of the CoCr disappeared probably due to the formation of microcracks. VSM hysteresis curves were measured both with applied field in-plane and perpendicular to the surface. For all films $S_{//}$ defined as $(M_C/M_S)_{//}$ was smaller than 0.12 and typically 0.09 for Magnetron and 0.06 for RF-films, indicating good perpendicular anisotropy of the CoCr films.

COERCIVITY

The surface coercivity H_{CS} for RF-films was found to decrease almost discontinuously at a critical film thickness of 125 nm after a maximum was reached of 120 kA/m at a film thickness of 80 nm. For Magnetron films the results are blurred due to the intrinsic spread in the samples (depending on substrate position). But also for Magnetron films it is found that H_{CS} decreases more rapidly with increasing film thickness than the volume coercivity H_{CV} after a maximum is reached. In Fig. 1a H_{CS} (Kerr) and H_{CV} (VSM) are plotted as a function of film thickness. Symbols without error bars were previously published [1] each representing a single sample. The symbols with error bars indicate the average values of film thickness and coercivity per group and the bars show the standard deviations of the coercivity. A maximum coercivity of 100 kA/m (1250 Oe) is found for group 1 samples at a film thickness of about 1000 nm. The maximum coercivity H_{CV} for group 2 is about 90 kA/m (1100 Oe) at a film thickness of 700 nm approximately. Again the surface coercivity H_{CS} decreases faster than H_{CV} .

This can be seen more clearly in Fig. 1b where the coercivity ratio (CR) is plotted as a function of film thickness. Here the averages of the samples from group 1 and 2 are plotted with

different symbols to distinguish them from the previously reported samples. A critical thickness is found at about 1100 nm for group 1 and for group 2 (off-centre) samples the critical thickness must certainly lie between 400 and 1000 nm, but probably between 700 and 1000 nm, after the maximum H_c is reached. So also for Magnetron films a critical thickness (defined by the coercivity ratio) is found. The maximum coercivity is about the same for RF and Magnetron samples, only the thickness at which it is reached is shifted to thicker films together with the critical thickness for Magnetron films.

Fig. 1a. H_{cv} (VSM) and H_{cs} (Kerr) as a function of film thickness for Magnetron sputtered CoCr. (see text).

Fig. 1b. The Coercivity Ratio (C.R) versus film thickness calculated from the data in Fig. 1a (see text).

SLOPE OF THE PERPENDICULAR LOOP.

RF-sputtered CoCr films.

The slope of the perpendicular hysteresis curve at $M = 0$ as a function of film thickness for RF-films was explained by Wielinga [2] using the Kooy & Enz model. Also in the particulate model an increased slope was predicted for the thinnest films ($t < 100$ nm) but this increase was not large enough to fit the experimental results. In the light of this discussion it is interesting to note, that the slope of the perpendicular hysteresis curve measured with MOKE deviates from the bulk value. In determining the Kerr slope the physical state of saturation $\theta_K(\text{sat})$ is set equal to M_s . In Fig. 2 both the slope of the Kerr and VSM curves are shown as a function of film thickness for RF-films. The VSM slope reproduces the results by Wielinga, but the Kerr-slope again increases, deviating from the VSM slope, for films thicker than about 100 nm.

Fig. 2. The slope of the perpendicular surface (Kerr) and volume (VSM) hysteresis curve near the origin for RF-films as a function of film thickness. Each point represents the average of 4 samples.

Fig. 3a. The slope of the perpendicular surface (Kerr) and volume (VSM) hysteresis curve for Magnetron films as a function of film thickness. (see text).

Fig. 3b. The slope ratio calculated from the data Fig. 3a versus film thickness (see text).

Magnetron sputtered CoCr films.

Results for the Magnetron films are similar. In Fig. 3a Kerr and VSM slopes are plotted as a function of film thickness. Again the symbols without error bars refer to single samples published earlier [1] and the symbols with error bars represent the average values plus standard deviations for group 1 and group 2 samples. The VSM slope of the two groups are comparable and are both represented by a 'x'. The Kerr hysteresis curve becomes significantly steeper (say 10%) than the VSM curve for films thicker than about 800 and 500 nm for group 1 and 2 type samples respectively, as can be deduced more clearly from Fig. 3b. In this figure the ratio between the Kerr and the VSM slopes calculated from the data in Fig. 3a is plotted as a function of film thickness.

DISCUSSION AND CONCLUSIONS

In Table 1 experimental values for the critical film thickness based on the coercivity ratio t_c (C.R.) and on the Slope Ratio t_c (S.R.) of Kerr and VSM hysteresis loops are summarized together with the film thickness t_{max} at which a maximum coercivity is reached. This t_{max} is approximately the same for H_{CV} and H_{CS} . After the maximum is reached H_{CS} decreases more rapidly than H_{CV} . For RF-films this decrease is very abrupt. From t is 100 to 150 nm H_{CS} decreases from 120 to 60 kA/m. These results indicate that the decrease in H_{CV} after the maximum is reached probably originates from the top surface.

At a film thickness near the maximum coercivity the slope of the Kerr hysteresis loop becomes larger than the VSM slope. These results hold as well for the RF-films as for the group 1 and 2 type Magnetron samples. So a relation between the three phenomena could exist.

Table 1

Film thickness (in nm)	CoCr 81/19 at%		
	RF	Magn.	
		G-1	G-2
t_c (C.R.)	125	1100	800
t_c (S.R.)	100	800	500
t_{max} (H_c)	80	1000	700

One conclusion is obvious, namely that for films with different surface and bulk hysteresis, the magnetization cannot be homogeneous throughout the film thickness during all stages of the hysteresis curve. The cause could be a subdivision of the domains by small reversed domains at the surface (spikes), as proposed by Privorotskii [8], or a deviation of the magnetization from the perpendicular orientation as described in the μ^* -effect. The theory must explain the different behaviour of films near the critical value. For RF-films this behaviour is almost discontinuously. Spike-domain theory is most promising, since it predicts a critical thickness at which these domains can be created to lower the total energy. According to Privorotskii these spikes penetrate to about 10% of the film thickness or not at all. The remanent magnetization M_r is lowered at the surface only by the formation of spikes. Together with M_r the coercivity H_{CS} is decreased. The two are related by the slope T according to: $M_r = T \cdot H_{CS}$. Hence a strong decrease in H_{CS} accompanied by a more gradual decrease of H_{CV} can be explained by the formation of spikes. Also an increase of the Kerr slope for films thicker than t_c is consistent with spike domain theory. It is well known that basically the slope is equal to one, due to the sheet demagnetization; $H_d = -M$. The formation of spikes lowers the demagnetizing field, hence the slope of the hysteresis curve becomes steeper. Recently Hubert [3] renewed the interest in branched domain structures in thin films with uniaxial anisotropy, and also calculated the (VSM) slope of the perpendicular hysteresis curve for such a branched domain structure. Based on this

calculations and the agreements with the experimental (VSM) results by Wielinga [2] (reproduced here) he concluded that his analysis confirmed the domain model of CoCr layers. Spike domains are commonly found in uniaxial bulk materials like Co and Magnetoplumbite [7], but these domains observed by bitter powder and Kerr microscopy are several orders of magnitude larger than expected in CoCr thin films. However, the parameter that determines the material length $\lambda_m = \sigma_w / \mu_0 M_s^2$ ($\sigma_w = 4(A \cdot K_1)^{1/2}$) is indeed very small for CoCr (81/19 at%), namely 3.5 nm for RF-films and 5 nm for Magnetron films. These values have been calculated assuming that the exchange constant A determined for RF films at $A = 6 \times 10^{-13}$ J/m³ [2] also holds for Magnetron films. However, the value of λ_m as calculated above is comparable with the dimensions of the discontinuities in the morphology of CoCr.

Using Privorotskii's theory the critical film thickness for the formation of spikes is calculated between 25 and 40 nm, but strictly speaking this cannot be correct since the length of the spikes would become smaller than the wall thickness. Probably the weak anisotropy assumption and the approximation of no interaction between top and bottom surface made by Privorotskii does not hold for CoCr in this case.

In the particulate model the critical radius r_c for a spherical single domain particle is also very small. According to Chicazumi [7] $r_c = 4.5 \lambda_m$. The column diameter d and film thickness t for (RF) CoCr films are roughly related by $d = t^{2/3}$ (both in nm) [6,9]. Here we measured by (S)TEM that about the same relation holds for Magnetron films. So by simply putting $2r_c = d$ the critical film thickness would be $t_c = (9 \lambda_m)^{3/2}$ or 180 nm for RF and 300 nm for Magnetron films. Hence order of magnitude calculations do not oppose the formation of such small spike domains in CoCr thin films. Also the larger critical film thicknesses found for Magnetron films are qualitatively predicted.

Due to the discontinuous character of the reported results, especially the surface coercivity of RF-films, it is not likely that the μ^* -effect will be able to explain all the results. However, a possible contribution of this effect cannot be disregarded.

Concluding, it can be stated that present results of comparing surface and bulk magnetic behaviour are consistent with a spike domain theory in CoCr thin films. If spike domains do exist in CoCr, then the reversal mechanism is most likely one in which the reversed domains grow at the expense of the main domain.

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