Magnetic microstructures in Co-Cr sputtered thin films for perpendicular recording

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Abstract- The relation between deposition parameters, microstructural and magnetic properties are influencing the reversal mechanism of sputtered Co-Cr thin films having perpendicular anisotropy. Three Co-Cr films with perpendicular coercivities ($H_{c\perp}$) of 11, 90 and 170 kA/m are investigated. Besides the $H_{c\perp}$ several other parameters were studied by VSM, SEM, NMR, MFM, AFM, selective etching, such as the $H_{c\perp}/H_k$, Cr-segregation, domain structure, column sizes, etc.

The anomalous Hall effect (AHE) has been used to record the hysteresis curves of sub-micron Hall crosses. This very sensitive technique in combination with e-beam lithography and ion-beam etching resulted in the recording of AHE hysteresis loops with dimensions of the Hall crosses as small as $0.3 \times 0.3 \mu\text{m}^2$. The AHE-loops of three samples, with less than 60 columns, show different magnetic properties and the total number of steps measured in the hysteresis loop was 5 times the number of columns. The different reversal mechanism observed by the AHE are related to differences in microstructure, coercivity and domain size. The analysis of AHE hysteresis loops we conclude that magnetic units on the limit of superparamagnetism could be present in Co-Cr. The obtained results have been used as input for our model for micromagnetic simulations. Micromagnetic simulations shows that the initial layer and the exchange coupling of the initial layer with the columns has a large effect on the magnetisation reversal processes in the film.

Key words: perpendicular recording, Co-Cr sputtered film, initial layer, magnetisation reversal, magnetic structures, Anomalous Hall Effect, micromagnetism.

I. INTRODUCTION

Magnetic recording has been the dominant recording technology for information storage since the invention of the computer. The demand from the user is for more compact information carriers and more over smaller formats are useful and can be produced more cheaply. For such developments knowledge and expertise of the many relevant subjects are needed such as the mechanics of the recording system, the intelligent electronic system, the development of new materials and designs for heads and media and last but not least a better knowledge of the interface between head and medium. The combined approaches of all the research have led to the commercial

available densities $>1 \text{ Gbit/inch}^2$ using longitudinal magnetic recording (LMR) systems. Since 1992, the 10 Gbit/in^2 longitudinal recording, having bit areas less than $0.1 \mu\text{m}^2$, has been discussed [1]. To reach this goal drastic scaling-down of the track pitch, bit-cell length, head gap, medium thickness and head-medium spacing have been made. Although perpendicular recording has not been used commercially its potential is very promising. Based on the published results [2-5] one can conclude that it is still a potential candidate for the super high density recording application. Based on the trend of increasing densities for PMR an area density of more than 300 Gbit/in^2 will be available in the 21st century as predicted in [6]. This statement was based on computer simulation [7]. In fig.1 one track with written bits are given for the LMR and PMR modes.

Figure 1: An isolated recorded track with written bits for LMR (a) and PMR (b) in a medium having a thickness (δ) and a track width (tw), recorded wavelength (λ), linear bit length (bl), recorded transition and bitcell area [8].

In both cases the area density is given by the product of the linear bit density and the track density. Higher density means creating narrower tracks and more compact bits. For LMR the magnetisation vector forms a head-on state of transition which results, for thin film media in a saw-tooth shaped transition. Medium noise, mainly originating from the transition becomes larger at higher densities [9]. In the case of PMR the anti-parallel magnetisation in the adjacent bits are responsible for a significant reduction of the demagnetising field especially at higher densities. In an optimised single layer Co-Cr medium the signal to noise ratio has been increased [10] and it is also reported in general that the noise becomes lower in the case of high densities [9]. After searching the published results in the literature one can find that ratio bitlength/medium thickness (b_l / δ) for Co-Cr-(X) thin film media in LMR is about 10-20 and for PMR about 1/3-1/5. From this data it can be concluded that media with a perpendicular anisotropy can be made with larger layer thickness which are favourable for higher densities (higher signal, easier preparation conditions and smaller crystal size in relation to the critical volume for superparamagnetic behaviour). In the case of LMR densities around 10 Gbit/in² one of the problems will be the thermal stability of the recorded bits [11]. In the case of PMR the ratio KV/kT is much larger than for LMR media. This means that PMR is very stable in thermal fluctuation [12]. Typical values for LMR as well as PMR materials are given in [13]. The SNR of PMR with weak exchange interaction are much higher than those of LMR, despite the larger grain size of the PMR media. Also in the case of PMR the H_c is not a very important parameter for high densities because there is no strong relation between this parameter and the linear density as there is in the case of LMR. In [14], read/write experiments are given from the combination single-layered perpendicular media (Co-Cr-Pt; Co-Cr-Pt-Ta) and ring-type heads. In the case of contact recording using a narrow gap head (gap length < 0.2 μm) D50 (the linear recording resolution) 300 kFCI is confirmed (implying a bit length of 63 nm). Track density can be 25 kTPI. It is concluded [14] that taking into account medium noise and overwrite characteristics, signal processing, and bit stability, this single layered/ring head system indicates a possibility of 10 Gbit/in². This density is also claimed by [15] using a MIG ring head in combination with a double layered medium (hard Co-Cr layer with a soft magnetic back layer). High output and high linear density recording of over 300 kFRPI was obtained by using Co-Cr-(Nb, Ta, Pt) media in combination with a Single Pole Tip (SPT) head with pole thickness of 0.2 μm [16]. Also it is demonstrated that a high linear density of 1040 kFRPI (bit length: 24 nm) can be obtained and an areal density of 29.6 Gbit/in² has been verified. Before this can be used commercially better write and read performances of the head must be realised as well as further reduction of the medium noise. For the

predicted area density of more than 300 Gbit/in² the bit size will be reduced drastically and must be in the order of 50x50 nm². To achieve such densities the knowledge about the micromagnetic behaviour in relation to the measured macro-magnetic parameters and the microstructure of the medium becomes more and more important. Beside many others the magnetisation rotation on local scale plays an important role.

II. MACRO-MESO-MICRO MAGNETIC PROPERTIES

In order to understand the relation between the *macromagnetic properties* (e.g. hysteresis loop properties such as M_s , M_r , H_c measured with Vibrating Sample Magnetometer), *micromagnetic properties* (obtained by simulation and calculation) and *mesoscopic properties* (obtained by experimental methods (Lorentz microscopy, Magnetic Force Microscopy (MFM) and Anomalous Hall Measurements (AHM)) it must be realised that they are operating at a variety of length scales. Starting at the atomic level the interaction phenomena between spins and orbit, magnetic exchange interactions and dipole-dipole interactions must be considered (micromagnetic behaviour). Moreover the thin film media discussed here are composed of small crystallites consisting of a 3D periodic arrangement of (magnetic) atoms (mesoscopic behaviour). In the simplest case, each magnetic atom has the same local environment in which the neighbouring atoms (magnetic or otherwise) form a spatial configuration of a particular symmetry. A typical thin film medium for high density recording consists of individual magnetic crystals. When a VSM hysteresis loop from a typical sample (e.g. 5 x 5 mm²) is measured, a hysteresis curve of 15 000 interacting crystals is obtained assuming a size of 40 nm.

If a typical bit size (see fig.1) for such media is about 3 μm^2 the recorded information is obtained from 2500 crystals. Due to the complexity of the problem only a few hundred columns can be simulated even with the most advanced supercomputers. Consequently, there is a gap between micromagnetic simulations and the macromagnetic properties obtained by experimental methods (e.g. hysteresis loop). Therefore, is it necessary to develop experimental methods from which magnetic and microstructural information from smaller volumes can be obtained. Only then is it possible to find a more definitive relation between the microstructure, local chemical composition and the magnetic behaviour on a length scale more related to high density recording. Given 10 Gbits/in² the fundamental structural dimensions (crystal size and surface topology), intrinsic magnetic structures (domains) and the bit dimensions are approaching similar sizes. In the case of the 50x50 nm² bit size only about 25 crystals are available which means that micromagnetic simulations

become more and more realistic in relation with the recorded bitsize.

In 1987 we studied the reversal mechanism of Co-Cr sputtered films having a perpendicular anisotropy by calculating the $H_{c\perp}/H_k$ and observing the domain pattern of the samples with Kerr microscopy [17,18]. From this study it has been shown that samples with $H_{c\perp}/H_k = 0.02$ have long stripe domains. Short stripes occur for a values around 0.05 and short stripes or a so-called dotlike domain pattern are present in samples with $H_{c\perp}/H_k > 0.1$. The values used for this classification have been obtained from macroscopic magnetic measurements of large samples with VSM and torque-magnetometer. Micromagnetic simulations have also been carried out and especially the simulations in which there is exchange between Co-Cr crystals were in good agreement with the experimental hysteresis loops [19]. The micromagnetic approach to this problem is severely limited by the number of Co-Cr columns which can be simulated. The 'meso'-magnetic behaviour of the samples is studied by using a very sensitive technique: the anomalous Hall effect (AHE) applied on submicron Hall-crosses [20]. This technique offers us the possibility to bridge the gap between the macromagnetic and the micromagnetic behaviour responsible for the reversal mechanism. The AHE results are compared with simulated hysteresis curves obtained with an array of 5×5 columns. To get more inside about the reversal behaviour of small areas of Co-Cr we have selected three samples with different values for $H_{c\perp}/H_k$.

III. PREPARATION OF THE Co-Cr SAMPLES

Three Co-Cr samples with different deposition parameters are prepared to obtain various microstructures and morphologies. The major difference was the substrate temperature (T_s) during deposition as it is known that this parameter influences the film growth in such a way that large differences in structure and magnetic properties occur. This resulted in samples with low, medium and high coercivity, hereafter expressed as S1, S2 and S3 respectively. The low (S1) and high (S3) coercive films are prepared by rf-magnetron sputtering at a substrate temperature of respectively 40°C and 150°C. The medium H_c film was rf-sputtered at RT. All samples have been deposited on Si-substrates which were covered first with an oxide layer of about 70 nm (SiO_2). After deposition (and etching for the sub-micron samples) the wafers were cut into $1 \times 1 \text{ cm}^2$ samples for measurement.

A summary of the macroscopic magnetic parameters measured by VSM, AHE and torque, the thickness t and

the %Cr are determined from sputter time and target composition is given in Table 1.

IV. MACROSCOPIC MAGNETIC PARAMETERS

The VSM-hysteresis curves from the three samples both perpendicular and in-plane are given in fig. 2.

The differences in $H_{c\perp}$ can immediately be observed in the perpendicular curves.

The in-plane curves also show differences, especially near the crossing of the x-axis (at $H = H_c$) where for the S2 sample a large contribution from an initial layer (IL) can be observed (in-plane component of the magnetisation). The volume of the IL can be estimated by comparing the height of the jump with the height of the total loop. The IL of S1 and S3 is about 10% and for sample S2 about 16% of the total loop. The $H_{c\perp}$ can be determined both from the perpendicular VSM and the AHE loops, and as expected the results show similar values. The calculated $H_{c\perp}/H_k$ predicts according to the theory [18] for the low coercive sample S1 a stripe domain pattern and for the other two samples a dotlike domain pattern. The macroscopical parameters obtained by several techniques are listed in table 1.

Coercivity, magnetisation and reversal behaviour

The coercivity is a very important parameter for magnetic recording but it is very difficult to understand the physical background. First of all, the coercivity is an extrinsic parameter and is strongly influenced by the microstructural properties of the layer such as crystal size and shape, composition and texture. These properties are directly related to the preparation conditions. Material choice and chemical composition (and inhomogeneities) are responsible for the M_s of a material and this is also an influencing parameter of the final H_c . It is difficult to discriminate between all these parameters and to understand the coercivity origin in the different thin-film materials in detail. Ideal SDP which switches coherently, $H_c = H_k$, if a field is applied in the easy axis direction. Depending on various factors incoherent switching occurs and the H_c decreases. Even in a matrix of particles, with magnetostatic interaction, the coercivity will be influenced. Multidomain particles and thin films will switch by domain-wall motion and again the coercivity decreases in comparison with the H_k . In addition, for thin films surface and interface properties have an influence on H_c . The above leads to the assumption that H_c can only be determined by means of the macroscopic hysteresis loop in combination with the theory of micromagnetism [21].

Table 1: Macroscopical parameters measured by VSM, AHE and torque, the thickness t and the %Cr are determined from sputter-time and target composition

Sample	t [nm]	%Cr [target]	M_s [kA/m]	$H_{c\perp}$ (VSM) [kA/m]	$H_{c\perp}$ (AHE) [kA/m]	H_k [kA/m]	IL [vol.%]	$H_{c\perp}/H_k$
S1	320	22	236	13	11	244	11	0.04
S2	200	23	329	89	91	306	16	0.29
S3	320	22	428	173	169	501	10	0.34

Figure 2: The perpendicular and in-plane hysteresis curves of the three samples measured with a VSM

The reversal of the magnetisation is a basic principle of magnetic recording. The predominant mechanisms (domain wall motion and rotation) are strongly determined by the material and its dimensions. Knowledge of the microstructural properties of the material is indispensable. In the first place the size and shape of the magnetic unit (crystal, column) plays an important role. In SDP the H_c increases to a maximum at a critical particle-size diameter. If the particle size increases, incoherent switching mechanisms like curling, buckling and fanning are used when the single domain particle (SDP) diameter is decreased, it finally becomes super-paramagnetic. At this stage the reversal takes place by thermal agitation which again leads to a lower H_c . Further increase in the diameter results in a multidomain particle (MDP). Magnetic thin films with a polycrystalline structure are strongly exchange coupled and consequently their magnetisation reversal will

take place by domain-wall motion. In this case the H_c is determined by pinning mechanisms of the domain wall. These mechanisms are determined by the magnetically inhomogeneous regions like columnar boundaries, chemical inhomogeneities, stacking faults etc. The H_c for polycrystalline thin films have been discussed in detail in [22].

Critical size for a ferromagnetic entity

There are various critical dimensions for a ferromagnetic entity in relation with its magnetic behaviour. The precise size depends on the shape and the material. First of all the critical boundary between SDP and MDP (at a diameter of about 0.1 μm). The domain structure lowers the magnetostatic energy but increases the exchange energy caused by the domain walls. The reversal in such particles mainly takes place by domain-wall motion. At a certain smaller diameter the SDP switching mode changes from coherent into one of the incoherent modes (critical size in the order of 15 nm). Another critical dimension appears between the ferromagnetic and the superparamagnetic state of the particle; in other words the thermal activation energy kT exceeds the particle anisotropy energy barrier. When using the Néel-Arrhenius law for time decay in particle assemblies $1/\tau = 10^9 \exp(K_u V/kT)$ [23] the estimated superparamagnetic limit of Co-Cr is ($\approx 440 \text{ nm}^3$) when using a relaxation time τ of 100 s and an anisotropy constant K_u of $1.6 \cdot 10^5 \text{ J/m}^3$. For particulate media, it is also observed that the mentioned formula does predict the so-called activation volume, which is much smaller than the particle volume [24]. A typical critical diameter for materials mostly used in recording media (given a certain thickness) is about 5 nm. The reversal of the magnetisation in this type of particle is the result of the thermal motion. Consequently the intrinsic coercivity strongly depends on the particle diameter and can be described by a model given in [25]. For more detail description please refer to [26].

V. CHEMICAL MICROSTRUCTURE

To obtain high coercivity in a continuous thin film of Co-Cr the grains must be magnetically isolated from each other. The accepted mechanism is the compositional separation (CS) by a non-ferromagnetic Co-Cr composition (at % Cr > 26). Beside the columnar boundaries CS can also be found in the column itself. Maeda [29] first introduced the so-called chrysanthemum-like pattern (CP). A simple schematic structure (see fig.3) of sputtered Co-Cr films consisting of equal-sized columns is used as a model for showing the possible chemical structures. Figure 3a shows the microstructure with equal column size and shape and a columnar boundary. In this case, the interaction between the columns occurs by exchange and magnetostatic interactions. The boundary can only hinder the movement of the domain wall which means in principle

an increase in H_c . For higher H_c 's we need to break up the exchange forces between the columns. This can be realised by CS or separation (by voids) of the columns, which has been shown also in the case of Co-Cr-X media for LMR. In the case of CS a non-ferromagnetic Cr-rich composition is present between the columns (fig. 3b). The more complicated CP structure of one column in combination with a Cr enriched column boundary is shown in fig. 3c. This type of complicated structures have many consequences for the switching behaviour of the magnetisation and, moreover, for the coercivity. The spatial periodicity of CS in sputtered Co-Cr layers on top of soft magnetic Ni-Fe layers is in the range of 3-7 nm [29]. There are many different possible explanations of such a CS structure (which has never been observed in Co-Cr bulk alloys) in the literature. [30-38]. Most consistent work in this field has been published by [39-43]. Results obtained by APFIM (Atom Probe Field Ion Microscope) have shown how the concentration fluctuations in the grains (columns) are distributed [37, 44, 45]. Over a distance of 40 nm in the planar direction of a Co-Cr column differences of 30 at % Cr and 7 at % Cr have been measured. The latter composition is ferromagnetic while the composition above 26 at % Cr is paramagnetic. The fluctuations are less than 10 nm.

In order to determine the CS structures in more detail also extended polarised dependent x-ray absorption fine structure measurements of Co-Cr sputtered films have been performed [46]. Fourier transforms of the Co and Cr EXAFS data illustrate an unambiguous anisotropy in Co and Cr atoms local environments between the in-plane and out-plane structure.

A fitting analysis of the Fourier near neighbour peak of the Co and Cr EXAFS data indicates that a large percentage of the Co atoms exists in clusters, while the Cr environment is anisotropic with a greater number of Cr-Cr pairs parallel to the film plane. The EXAFS data were subjected to analysis procedure as given in [47]. The results can be summarised as follows. The first shell surrounding of the Co atoms shows Co-Co co-ordination numbers greater than that expected for a randomly mixed alloy. These results demonstrate the presence of Co enriched regions within the films. Similarly, analysis of the first shell surrounding the Cr atoms shows a Cr-Cr co-ordination number greater than expected for a randomly mixed alloy as well as an increase in the Cr-Cr co-ordination number in the plane of the film. This result is consistent with previous studies that suggested the presence of a thin Cr-rich region oriented parallel to the film plane, possibly between stacked Co-rich platelets which make up the columns.

Figure 3: Model of Co-Cr sputtered PMR medium in which the morphology is determined by the type of compositional separation (CS). Fig a) shows the ideal columnar morphology, b) the CS in the columns (CP structure) as well as at the boundaries c) CS only on the column boundaries (see also figure 4) [28]

The reversal of the magnetisation depends on the exchange and magnetostatic interactions between the columns or cluster of columns. For a reasonable S/N it is necessary to have at least 100-200 of these magnetic entities in a bit. Smaller bit sizes consequently need smaller individual switching units. In order to understand the magnetisation reversal in a small bit (e.g. 10 Gbits/in²) the question arises whether the hysteresis loop (measured with a Vibrating Sample Magnetometer; VSM) is still the most relevant magnetic characterisation for understanding reversals on this scale. New approaches are necessary to bridge the gap between micro- and macroscopic behaviour.

SEM and NMR results

The chemical inhomogeneities of the three samples were studied by the ⁵⁷Co spin-echo NMR technique [48] and by selective chemical etching of the Cr and successive SEM-observations [27]. In fig 4 SEM-images of sample S1 (a,b), S2 (c,d) and S3 (e,f) are given before and after chemical etching (right). The white stripes within each crystal are thought to correspond with dissolved highly Co-rich

a)

b)

c)

d)

e)

f)

Figure 4: SEM-images of sample S1 (a, b), S2 (c, d) and S3 (e, f): as deposited (left) and after chemical etching (right)

and the dark areas correspond with passivated Cr-enriched regions. In figure 3.d and 3.f there are faint stripes visible in S2 and S3 after seven hours of chemical etching of the samples. This is also an indication for compositional separation and the presence of a chrysanthemum pattern (cp-) structure. On the other hand sample S1 shows no chemical inhomogeneities inside the columns, even after 14 hours of etching. This confirms the NMR-results for this sample with a homogeneous Cr distribution. The NMR measures the hyperfine field interactions or the resonance frequencies of the material and is used here to study the compositional inhomogeneities. A spin-echo spectrum of Co-Cr consists of a main line frequency (M) and of satellites. The main line frequency arises from Co nuclei which have only Co atoms as nearest-neighbours (NN), for example, the hyperfine field of a Co nuclei in pure fcc Co has a main line frequency of 217 MHz and for pure hcp Co of 225.5 MHz, the exact frequency depends on the crystal structure, morphology and chemical inhomogeneities. If the Co is diluted by Cr (<1-2 at.% Cr), beside the main line frequency the first satellite peak (Sat1) appears in spectrum at 177 MHz and is caused by Co nuclei which have one Cr atom at the NN site. A second satellite line (Sat2) appears at about 130 MHz for Co nuclei with two Cr atoms at the NN sites (at about 5 at. % Cr), etc. To apply this technique the frequencies of the peaks in the measured spectrum are compared with those of powdered samples with a known composition. From figures 5 b/c it can be seen that both samples S2 and S3 show a main line frequency (M) of respectively 207 and 213 MHz, characteristic for the presence of highly enriched Co-areas. S2 has components with 90% Co and S3 with 97% Co. On the other hand sample S1 has no M-peak above the 150 MHz which indicates that there are no Co-enriched areas and that there is a homogeneous distribution of the Cr through the sample. Consequently, this observation of CS shows Co and Cr-rich regions in one crystal which means that such a film can consist of ferromagnetic regions smaller than one column. The Co and the Cr-rich components are strongly influencing the (micro) magnetic behaviour of the film.

In Table 2 together with some structural and magnetic properties also the NMR data is given. The domain size

determined by the MFM of S2 and S3 are respectively 147 and 258 nm. Together with the column size it can be estimated that the number of columns for one domain in the remanent state is about 5 and 14 for these two samples.

Unfortunately it was not possible to measure the domainsize of the low coercive sample S1. The magnetic tip influences the magnetisation beneath the tip too much, even when the tip was replaced by a magnetic tip with a soft magnetic layer of Ni/Fe. From this sample a Kerr image was recorded which showed long stripe domains.

VI. ANOMALOUS HALL MEASUREMENTS ON SUB-MICRON SAMPLES

With conventional techniques, such as VSM or Kerr it is not possible to observe the micromagnetic switching of individual columns in a hysteresis loop. The maximum sensitivity of a commercial VSM is typical in the order of 10^{-7} kA/m, whereas the magnetic moment involved in the switching of one Co-Cr column is in the order of 10^{-12} kA/m. With the AHE technique a sensitivity of 10^{-14} kA/m can be reached. The principle of AHE measurements on submicron Hall samples in Co-Cr was first used in 1987 by Webb and Schultz [49]. Their samples had thickness of 1-1.4 μm and a smallest Hall cross size of 0.7 μm .

Our samples were prepared by e-beam lithography and ion-beam etching. Using this process samples with a smallest Hall cross dimensions of $0.3 \times 0.3 \mu\text{m}^2$ could be prepared. A description of the anomalous Hall measurement set-up is given in [20]. Figure 6 shows a drawing of the Hall cross with a limited number of columns inside the measured area. We will discuss first some aspects of the recorded hysteresis loops which are the result of the smaller dimensions of the Hall cross. As can be seen from fig.4b relative large areas of the Co-Cr cross have been removed by etching which reduces the perpendicular component of the demagnetising factor (N_z) inside the Hall cross and therefore the recorded hysteresis loops will become less sheared. This problem has been discussed in [50]. To check the validity of this method samples with various dimensions have been analysed and also noise analysis of the measuring method have been carried out [50]. The total

Table 2: Summary of the microscopical parameters with t the film thickness, D the column size and M the main line frequency

Sample	t [nm]	D (SEM) [nm]	M (NMR) [MHz]	%Co enriched (NMR)	stripes visible after etching ?	domain size (MFM) [nm]
S1	320	60	<150	Bulk	no	-
S2	200	75	207	90	faint	147
S3	320	70	312	97	faint	258

Figure 5: NMR-spectra of the three samples S1 (a), S2 (b) and S3 (c)

number of measuring points was 4000 for each AHE-loop, resulting in a 9 hour recording time for each loop. Based on this extensive research all data given below can be considered as relevant. In figure 7 the AHE-loops are shown for the three samples with an enlargement of a typical area in the steep part of the loop. These graphs show that there are only steps in the AHE hysteresis curve for the S2 sample. The largest jumps correspond with the switching of one average sized column. Between the jumps there is an area in which the Hall voltage is almost constant ('plateau'). A closer look at this area (figure 8)

reveals that there is also an increase in the signal in this part of the curve. From an enlargement of this 'plateau' it can be calculated that the slope is 30 times the slope of the Hall curve in saturation.

This means that this effect is not caused by the normal Hall effect but by a very small increase in the AHE voltage for these very small field steps. The noise in the curves was measured by determining 4x the rms noise at saturation which contains about 95% of all data points and is about % of the total signal ($100\% = 2 \times M_s$) [50]. As the number of columns in the centre of the Hall cross in this sample is about 25, and most of the AHE-signal is derived from the Hall cross we expect that the number of jumps would be

Figure 6: The Hall pattern used for the sub-micron Hall structures: a) a $1.0 \times 1.0 \text{ cm}^2$ Hall sample with the contact points and the contact paths, b) a detail of the Hall cross, c) the centre of the Hall cross with Co-Cr columns and the active area (w is the size of the Hall cross)

about 25 if the reversal would take place by the switching of the individual columns independently of each other. To study the behaviour of the switching units in more detail several statistical analyses were carried out.

The hysteresis curves were recorded many times and the field values of the large steps were studied as a function of the applied field. This distribution of the stepsizes as a function of the applied field shows that there are significant differences in successive measurements with the same sample. A typical example shows that even for the two branches in the same loop that the distribution is different for the ascending and the descending branch and that most of the large steps occur around the coercive field. The largest steps around 2-3 % of $2M_s$ correspond with one column. As can be seen from the AHE curves in figures 7a,b and 7e,f there are no steps observed in the low and the high coercive samples. We think that the reason is that

e) **Figure 7: Anomalous Hall effect hysterises loops (left) and details of these loops (right) of the samples S1 (a, b), S2 (c, d) and S3 (e, f) respectively**

Table 3: Stepsizes for the Classes used to classify the 5 SPH-loops.

Class	stepsize [% of $2M_S$]	Specification
1	$\Delta M > 0.3$	large pos. step
2	$0.1 < \Delta M < 0.3$	small pos. step
3	$0.0 < \Delta M < 0.1$	very small pos. step
4	$-0.1 < \Delta M < 0.0$	very small neg. step
5	$\Delta M < -0.1$	small neg. step

that sample S1 has such a low coercivity that the domain wall motion takes place very gradually with a change in the field that no steps occur above the noise level. For the high coercive film S3 we propose that the reversal takes place by the rotation of very small units *cross* inside the Co-Cr columns and that those units are exchange decoupled and switch independently from each other.

Stepsize distinguished Partial AHE loops (SPAHE-loops). To study the AHE-hysteresis curves in more detail each AHE-curve was divided into 5 separate curves. Each curve contains only the steps with a certain stepsize and each of these Stepsize distinguished Partial Anomalous Hall Effect loops (SPAHE-loops) can be considered as the hysteresis loop of all the magnetic 'volumes' within a certain range [50]. Each range of jumpsizes is denoted by a Class, and those classes are defined in table 3, where ΔM is the change in magnetic moment of each step.

Figure 8: Enlargement of figure 7d, showing the increase in Hall voltage on the 'plateau'.

In figure 9 the division of an AHE curve of sample S2 in 5 SPH-loops is shown for the loop in figure 7c, with the division in classes as given in table 3. According to this table the loops of Class 1 contain the large steps, Class 2 the small steps, negative steps. Class 3 and 4 the small positive and negative steps and also noise, and Class 5 the small Class 1 contains the 'large' (irreversible) steps of volumes $> 30 \text{ nm}^3$. Table 4 gives the results of one classification of a typical hysteresis loop. From this table it can be seen that Class 5 has about 5%, this corresponds with steps in the 'reverse' direction, but Class 1 has the largest component with almost 60% (59.6% and 58%). As

Table 4: The % of $2M_s$ for a typical AHE-loop of sample S2 for the ascending branch and descending branch and the H_c of the individual SPH-loops

Sample S2	Class				
	1	2	3	4	5
Ascending branch [%]	59.6	28.1	40.6	23.8	4.8
Descending branch [%]	58.0	31.4	40.0	23.9	5.7
H_c [kA/m]	145	117	33	43	70

class 4 has only 24%, we can assume that the noise is a minor component in the hysteresis curve as class 4 contains both the noise and a reversible component of the magnetisation. The loop of class 1 has the largest coercivity and the H_c decreases for the loops which contain the smaller volumes but does not become zero, not even for Class 4 and 5 (see last row of table 4). This indicates that these loops have also a significant magnetic origin, as mere noise would not result in a coercivity for these loops.

Figure 9: a) SPH-loop of sample S2 (with $w = 0.3 \mu\text{m}$) with the classification according to table 4 and b) detail of a

VII. MICROMAGNETIC SIMULATIONS

Micromagnetic simulations are believed to be most suitable for understanding the magnetisation process on a micromagnetic level, because they start with basic principles (exchange, magnetisation, anisotropy and morphology) and yield hysteresis loops and magnetisation patterns. The reversal of a small number of columns which can be measured by the anomalous Hall effect gives us the opportunity to compare and evaluate these results with

computer simulations. The computer model has been

Figure 10: Model structure of 25 Co-Cr columns. Each column consists of small elements (cubes). The column have been modified with an initial layer (grey) at the bottom having an in-plane anisotropy

described in [19,51,52]. This model takes account of the external field, the anisotropy energy, the demagnetising energy and the exchange coupling. The Landau-Lifschitz equation is solved to obtain the dynamic magnetisation process. The Hall cross was simulated by an array of 5×5 exchange decoupled particles (columns) as given in fig 10. Each column was divided in $6 \times 6 \times 18$ cubic cells with cell dimensions of 8.5 nm^3 . The particles are magnetostatically coupled and in this case no exchange coupling between the columns was considered. The field step between two successive points is 1.6 kA/m . In these computer simulations we studied the influence of an initial layer (IL) of 0%, 2% and 8% of the total sample volume on the hysteresis loop. In the remaining of this paper we will call these samples A, B and C respectively. The initial layer was simulated by assigning a random in-plane magnetisation to a certain number of cubic cells in the lower 2 rows of the particles near the substrate. In real samples this component can be part of an initial layer and/or distributed throughout the sample. As macroscopical input parameters for our simulation model we used realistic parameters obtained by VSM and torque ($M_s = 400 \text{ kA/m}$ and $K_1 = 1.6 \times 10^5 \text{ J/m}^3$). Except for the IL the sample has a perpendicular anisotropy with a spread in easy axis of 1.8° from the film normal. In figure 11 the three simulated hysteresis curves are presented. Several changes can be observed with the

Figure 11: Simulated hysteresis loops for an array of 5×5 Co-Cr columns with an IL volume of 0% (a), 2% (b) and 8% (c)

increase in IL volume. Starting from the saturated state the drop in magnetisation with decreasing field takes place much earlier with the presence of the IL. The value of the reverse field at which the first large step occurs (corresponding with the switching of one or more columns) is also much lower for the samples with an IL. The total contribution of the small (reversible) parts of the magnetisation, before the first large step takes place is 2.5, 3.9 and 10.6 % respectively for sample A, B and C. Also can be seen from the size of the large steps that the number of columns which switch independently is 14 and 10 for samples B and C and only 3 for sample A. The largest number of columns switching together in one fieldstep is 5 for sample A and both 3 for samples B and C. Remarkable changes for the coercivity (410, 403 and 310 kA/m) and the saturation field (760, 750 and 620 kA/m) only occur for sample C with the large IL component of 8%. From these data we conclude that the presence of a large IL or in-plane component of the magnetisation facilitates the reversal, lowers the coercivity and results in the switching of smaller units (less switching of more than one column at a time). The results of all the simulations presented in [50].

The 'plateaux' in figure 12 which is an enlargement of a part of the curve of figure 11 (c) shows significant similarities with the 'plateau' in figure 8 of sample S2. The gradual decrease (or increase) of the magnetisation can be seen, only there are no 'reverse' steps and there is of course no measurement noise. For a larger IL component the increase of the magnetisation per unit of the applied field of

Figure 12: 'Plateau' in the simulated hysteresis curve of figure 11 (c)

the 'plateau' is larger. We also divided the curves into separate curves with different stepsizes in the same way as has been done in figure 9 for a real Co-Cr sample. A typical example is shown in figure 13 for sample C.

The curves with reserve steps (Class 4+5) as shown for the real AHE measurement are not present, resulting in only three SPH-loops. For all the three samples the coercivity of the Class 3 loops is much lower than for the Class 1 and 2 loops. This is in agreement with measurement results and an indication that the small in-plane components are responsible for the Class 3 contribution to the total hysteresis loop. Unfortunately the model is not sophisticated enough to stimulate the more complicated cp-structure of sample S2 and S3. Because the AHE measurements prove that the average magnetic unit size is not equal to the column size, each column can be modelled as a Co-poor matrix with 3 Co-rich platelets (Also indicated in fig.3b). Although the parameters used are not completely equal to the dimensions and distribution of the CS they show reasonable agreement with the experimental AHE loop [8].

VIII. DISCUSSION AND CONCLUSIONS

The reversal mechanism of Co-Cr sputtered films has been studied intensively during the last decades by micromagnetic simulations either by macroscopic magnetic measurements. From this it is very difficult to decide whether the reversal takes place by domain wall motion or by a rotation of the magnetisation. In this paper we studied three Co-Cr films with different coercivities (11, 90 and 170 kA/m) and different microstructural properties. The measurements on the three 'large area' samples and the

AHE measurements on the submicron samples reveal that the reversal mechanism is different on both a macroscopical and on a 'meso'-scopical scale. The low coercive sample S1 has a low H_{c1}/H_k value corresponding with a stripe domains in the remanent state. The AHE measurements show no jumps as the reversal takes place by a continues domain wall motion in very small steps.

Figure 13: SPH-loop of the simulated hysteresis curve of sample C ((1) = switching of more than 1 column, (2) = switching of one column, (3) = switching of units smaller than one column).

Sample S2 shows small jumps in the AHE-curve, the largest comparable to the reversal of one column, and also a very gradual increase in Hall signal which can be attributed to a combination of domain wall motion and rotation of (parts of) a column. The high coercive film S3 has probably a reversal mechanism by rotation of the magnetisation of very small volumes. This is in agreement with the microstructure as has been shown by NMR and SEM-imaging after selective chemical etching (cp-structure and high compositional separation (CS)). More evidence was also found in the literature by NMR [e.g. 39], APFIM [e.g.45], SANS [9] and EXAFS [e.g. 46]. They found that the magnetic unit for Co-Cr films with a high CS was smaller than that of a grain. From the results of the computer simulations we have seen that an initial layer (or in-plane) component lowers the coercivity and the saturation field and facilitates the reversal of smaller units [50]. If the real Co-Cr sample has a cp-structure the in-plane components (magnetic and non-magnetic) are also distributed inside the columns making the reversal of parts of the column more probable [8]. This makes it more likely that in the case of a CP structure the reversal can take place by the switching of small units, smaller than one column as we have seen for the AHE measurements. A more

sophisticated computer model is necessary to simulate the cp-structure. We have also seen that the 'plateau' in the real AHE measurement has much resemblance with the simulation and that the slope can be attributed to the reversal of very small magnetic volumes. Good agreement was also found for the hysteresis loops which were divided into loops with large medium and small steps (SPH-loops). Most significant result here was that the coercivity of the Class 3 loops was much smaller than for the Class 1 and 2 for both the measurement and the simulations. Sputtered Co-Cr crystals/columns are divided into smaller ferromagnetic structures than the columnar dimensions. Anomalous Hall measurements showed that the analysis of hysteresis loops of sub-micron Co-Cr samples on the basis of the measured magnetisation changes yields information on magnetic volumes that can be distinguished in the sample. The size of the measured discrete magnetisation changes shows that the average magnetic unit in Co-Cr is considerably smaller than one column. A density of 12 Gbit/in² for PMR hard disk has been observed and indicates that there is an interaction between the magnetisation fluctuations beside the track and the written bit. The magnetisation fluctuations measured with MFM are related to clusters of Co-Cr columns. The magnetic structures measured are always larger than the average columnar diameter. To obtain sharp tracks the size of the intrinsic magnetic structures beside the track should be smaller than the linear bit length. The microstructure and morphology are very important for the new class of high density recording media. Magnetic properties which should be optimised are the coercivity, the remanence, the film thickness and columnar size and shape. The sharpness and shape of the written transition are determined by the grain morphology, magnetic coupling of the texture of the grains. The particular behaviour will be finally limited by the superparamagnetism. With respect to perpendicular recording, high linear-bit densities have been demonstrated on laboratory scale, achieved by recording a medium consisting of Co-Cr-Ta on a soft magnetic underlayer with a special single-pole head. The ultimate density of a bit area (2500 nm²) is predicted with bit lengths smaller than 50 nm.

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REFERENCES

- [1] E.S. Murdock, R.F. Simmons, R. Davidson, IEEE Trans. Magn. 28, 3078 (1992).
- [2] Proceedings of the PMRC'94, J. Mag. Soc. Jpn., Vol.18, Suppl., No S1, 586 pages, (1994).
- [3] Proceedings Intermag 1996, Session Magnetic recording: Perpendicular Media, IEEE Trans. Magn. 32(5), pp. 3789-3857, (1996)
- [4] Special issue of the J.Magn.Soc.Jpn., Vol.21, No.6, (1997)
- [5] This proceeding.
- [6] Y. Nakamura, J.Magn. Soc. of Japan Vol.15, Suppl., No S2, 497-506, (1991).
- [7] I. Tagawa, Y. Nakamura, J.Magn.Soc.Japan, Vol.13, S1, 97, (1989).
- [8] J.C. Lodder, J.Magn.Magn.Mat., 159, 238-248, (1996).
- [9] N.R. Belk, P.K. George and G.S. Mowry, IEEE Trans. Magn. 23, 1350, (1985).
- [10] Y. Sonobe, Y. Ikeda, H. Uchida and T. Toyooka, J. Magn. Soc. Jpn., vol.21, No.6, 959-965, (1997).
- [11] P.-L. Lu and S.H. Charap, IEEE Trans. Magn. 30, 4230, (1990).
- [12] K. Yoshida, M. Hara, Y. Hirayama, Y. Sugita, J. Appl. Phys. 79(8), 5664-5666, (1996).
- [13] M. Futamoto, Y. Hirayama, Y. Honda and K. Itoh, J. Magn. Soc. Jpn., vol.21, no.6, 950-968, (1997).
- [14] M. Futamoto, Y. Honda, Y. Hirayama, K. Itoh, H. Ide, and Y. Maruyama, IEEE Trans. Magn., 32(5), 3789-3794, (1996).
- [15] S. Iwasaki, K. Ouchi and N. Honda, IEEE Trans. Magn., 32(5), 3795-3800, (1996).
- [16] N. Honda, K. Ouchi and S. Iwasaki, IEEE Trans. Magn., 32(5), 3804-3806, (1996).
- [17] J.C. Lodder, D. Wind, G.E. v. Dorssen, Th.J.A. Popma and A. Hubert, IEEE Trans. Magn. 23, 1, 214 (1987).
- [18] J.C. Lodder, D. Wind, Th.J.A. Popma and A. Hubert, IEEE Trans. Magn. Vol. MAG-23, No.5, 2055-2057, (1987).
- [19] J.H.J. van Opheusden and E.M.C.M. Reuvekamp, J. Mag. Mat. 88, 247 (1990).
- [20] S. de Haan, J.C. Lodder and Th.J.A. Popma, J. Mag. Soc. Jpn. Vol.15 Sup., No S2, 349 (1991).
- [21] W.F. Brown, "Micromagnetics", Interscience Publ., John Wiley, New York, p75, (1963).
- [22] H. Kronmüller, J. Magn. Soc. Jpn., Vol 17, Suppl.S1, 260, (1993).
- [23] L. Néel, Adv. Phys., 4, 191, (1955).
- [24] A.M. de Witte, K. O'Grady, G.N. Coverdale and R.W. Chantrell, J. Magn. Magn. Mat., 88, 183, (1990).
- [25] C.P. Bean, J. Appl. Phys. Vol 26, No.11, 1381, (1955).
- [26] S.D. Cullity, "Introduction to magnetic materials", Addison-Wesley, pp 399-418, (1972).
- [27] Y. Maeda, S. Hirono and M. Asahi, Jpn.J.Appl.Phys., Vol 24, L951, (1985).
- [28] J.C. Lodder, MRS bulletin, Vol. XX, No.10, 59-63, (1995).
- [29] Y. Maeda, K. Takei, S. Yamamoto and Y. Nakamura, J. Magn. Soc. Jpn., vol 15, Suppl., No S2, 457, (1991).

- [30] J.W. Smits, S.B. Luitjens and F.J.A. den Broeder, *J.Appl.Phys.*, 55, 2260, (1984).
- [31] A.K. Jinghan, *J.Magn.Magn.Mater.*, 54-57, 1685, (1986).
- [32] U. Hwang, Y. Uchiyama, K. Ishibashi and T. Suzuki, *Thin Solid films*, 147, 231, (1987).
- [33] J.N. Chapman, I.R. McFadden, J.P.C. Bernards, *J.Magn.Magn.Mater.*, 62, 358, (1986).
- [34] M. Sagoi, R. Nishikawa and T. Suzuki, *IEEE Trans.Magn.*, MAG-22, No.5, 1335, (1986).
- [35] F.F. Abraham and C.R. Bundle, *J.Vac.Sci Techn.*, 18(2), 506, (1981).
- [36] N.L. Peterson, *J. Vac. Sci. Techn. A*, Vol.4(6), 3066, (1986).
- [37] K. Hono, Y. Maeda, S. Babu, J. Li and T. Sakurai, *IEEE Trans.Magn.*, MAG 29, 3745, (1993).
- [38] K. Takei, J. Suzuki, Y. Maeda and Y. Morii, accepted for publication in *IEEE Trans.Magn.*, (1994).
- [39] Y. Maeda, D.J. Rogers and K. Takei, *J. Magn. Soc. Jpn.*, Vol 18, Suppl., No S1, 27-30, (1994).
- [40] M. Sagoi and T. Nishikawa, *J. Appl. Phys.*, 66, 3173, (1989).
- [41] M. Hasabe, K. Oikawa and T. Nishikawa, *J. Jpn. Inst. Met.*, 46, 577, (1982).
- [42] T. Takayama, M.Y.Wei and T. Nishizawa, *J.Jpn.Inst. Met.*, 45, 341, (1981).
- [43] Y. Maeda, K. Takei and D.J. Rogers, *Jpn. J. Appl. Phys.*, 32, 4540, (1993).
- [44] K.Hono, presentation during PMRC'94 Extended workshop on Co-Cr system alloy films, October 17-18th, Akita, Japan, (1994).
- [45] K. Hono, S.S. Babu, Y. Maeda, N. Hasagawa and T. Sukurai, *Appl.Phys.Lett.*, 62, 2504, (1993).
- [46] K.M. Kemmer, V.G. Harris, W.T. Elam and J.C. Lodder, *Mat. Res. Soc. Symp.Proc.*, Vol.375 21-23, (1995).
- [47] D.E. Sayeres and B.A. Bunker, in *Xray Absorption: Basic Principles of EXAFS, SEXAFS, and XANES*, edited by D.C.Koningsberger and R.Prins; Wiley, New York (1988).
- [48] K. Takei, Y. Maeda, *Jpn. J. Appl. Phys.*, Vol. 30, No. 6B, June 1991, pp. L1125-L1128.
- [49] B.C. Webb and S. Schultz, *IEEE Trans. Magn.*, Vol. MAG-24 (Nov. 1988), pp. 3006-3008,
- [50] S de Haan and J.C.Lodder, *J.Magn. Magn.Mat.*, 168, 321-335, (1997).
- [51] M. van Kooten, S. de Haan, J.C. Lodder, A. Lyberatos, R.W. Chantrell and J.J. Miles, *J. Magn.Magn.Mater.* 120, 145-148, (1993).
- [52] M. van Kooten, S. de Haan, J.C. Lodder and Th.A.J. Popma, *J.Appl.Phys.* 75 (10), pp 5508-5510, (1994).