

Chapter 6

PREPARATION, MICROSTRUCTURE AND MAGNETIC PROPERTIES OF Co-Cr THIN FILMS

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1. Introduction

Sputtered Co-Cr films for perpendicular recording were first mentioned by Professor Iwasaki in 1975 [1]. Although in the meantime many alternative materials have been proposed, however, Co-Cr remains the medium mostly used for application in perpendicular recording.

Magneto-optic recording is the oldest mode of perpendicular recording having already been demonstrated in the literature in 1957 using a Mn-Bi medium with a perpendicular anisotropy [2]. Ten years later the concept of magnetic bubble memories was introduced [3]. Again about another decade later [1] the circular magnetisation mode and Co-Cr film having a perpendicular anisotropy were discovered and it is also interesting to know that this was based on magneto-optic recording research. Since then Co-Cr has been the most promising material for medium application in perpendicular recording although other media materials are still under development, such as Co-O [4], Co-CoO [5], Ba ferrite films [6], and Ba ferrite particles [7].

However, commercial use of Co-Cr, as a medium for perpendicular recording, was only started in 1989 [8] a co-operation between Censtor Cooperation and Northern Telecom in the USA. Current strategies on hard disk contact or quasi-contact perpendicular recording (on Co-Cr) have been discussed recently [9]. It is surprising that, although the research on perpendicular recording was completely localised in Japan, the start with commercialisation took place in the USA. In discussions with Japanese industrial representatives [10] it is not quite clear why Japan is not ready to produce the perpendicular recording system. Due to the total number of research activities and the published results [e.g. 11] there are no specific scientific problems for not producing a perpendicular recording system. Perhaps there are commercial reasons, or could it be the continuous improvement of the longitudinal recording technology? Although the recently published data about the ultra-high density recording from the IBM group in USA [12] and Hitachi in Japan [13] is referred to as a longitudinal recording mode it is questionable whether the vector of the resultant magnetisation is still in-plane or not.

Nevertheless, besides the commercial successes of the perpendicular mode, it has stimulated other modes of recording and has, of course, provided many new scientific and challenging

questions concerning new specific heads, contact between head and medium and, last but not least, the development of suitable thin-film media.

1.1 PERPENDICULAR RECORDING MODE.

The perpendicular recording mode has been described in several sections in this book. It should be understood that the total concept of this type of recording can only be realised as a complete success if the medium and head developments take place hand in hand. For instance, a combination of a perpendicular head and a so called double-layer medium, shown in fig.1, in which the bottom layer is prepared as a soft magnetic layer and the top layer is Co-Cr, was already in use for very high density recording performance in 1984 [14].

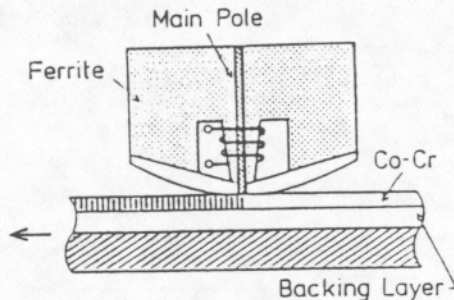


Figure 1. Main pole driven head in combination with a Co-Cr/Ni-Fe double layer medium [14].

A single-layer medium can also be used in combination with a ring head with a very narrow gap and a high saturation induction core [15]. It was clear from this research that satisfactory high density recording conditions are much more favourable for the single-layer medium using a ring head.

In general, a medium for perpendicular recording should have the following magnetic properties:

- The easy axis of magnetisation must be perpendicular to the film plane.
- Suitable coercivity for storing the information and reading it with a high S/N level.
- In connection with the above mentioned point, sufficient remanent magnetisation ($H_c = M_r$).
- A magnetisation reversal based on rotation instead of domain-wall motion.
- Chemical stable.
- Small surface roughness.

In order to realise this, a special microstructure should be obtained, namely a columnar morphology (small diameter), with a hcp [0001] texture and compositional separation to create a magnetic microstructure with less exchange and a high coercivity.

The relations between the magnetic and microstructural properties will be given in the following paragraphs.

1.2. CO-M AND THE CHOICE OF CO-CR LAYERS.

Co as a basic material for thin films is a reasonable choice because it shows an inherently high

uniaxial anisotropy based on magneto-crystalline anisotropy. In the case of perpendicular magnetisation, however, this anisotropy is smaller than the demagnetising field which is proportional to the saturation magnetisation. Consequently for a pure Co film the preferential direction of the magnetisation will be in-plane rather than perpendicular.

Therefore alloys of Co-M materials have been investigated [16,17], they include $M = \text{Rh, Pd, Mo, W, V, Ti, Cr, Mo, Pt and Mn}$. All the Co-M films exhibit a hcp crystal structure with the c-axis oriented perpendicular to the film surface. Since the actual deposition temperatures of the atoms were much higher than room temperature in these studies an important parameter was specially studied, namely the hcp/fcc transition temperature (T_{tr}). A stable hcp phase can only be formed if the film-growth temperature is close to T_{tr} . It can be seen in fig.2 that T_{tr} changes with the choice of M (two categories are present: those increasing with at. % M and those decreasing) and the composition of the Co-M alloys.

The data used in this figure concern bulk alloys and experiments in thermal equilibrium and that is completely different in the case of sputtered films. As will be discussed later the temperature which is relevant for phase transitions is not only difficult to determine for deposited films in general and sputtered films in particular, but also has a different thermodynamical meaning. Generally speaking the films are formed far from a thermal equilibrium state. Because a layer grows at its surface during deposition, it is predominantly the surface temperature that determines the crystal structure. Surface atoms are only partly surrounded by other atoms and therefore less bound, thus the surface properties are quite different from the bulk properties. In general the surface temperature may be said to be effectively higher than that of the value and the hcp to fcc transition of sputtered films will take place at lower temperatures than those determined for bulk alloys. The surface temperature depends strongly on the preparation method used for deposition of the layers and there is no experimental technique available for determination.

Using RF sputtering, Co-M alloyed films with excellent c-axis perpendicular orientation were obtained for $M = \text{Cr, Mo, W and V}$ with a sufficient M_s for recording. That is contrary to the alloys where T_{tr} decreases [19]. Because the perpendicular anisotropy field for Co-Cr is much higher than for the other three it is also, from this point of view, the most suitable choice. The H_k for Co-Cr solid solutions, in the area of interest, is about 520 kA/m. This increase, as a function of the composition, (H_k for pure bulk hcp-Co is about 520 kA/m) cannot simply be the result of an increasing ratio between K_1 and M_s ($H_k = 2K_1/\mu_0 M_s$) but must be caused by improvement of the c-axis orientation or stabilisation of the hcp structure.

Adding a second element to Co has important consequences for the Curie temperature (at which the spontaneous magnetisation disappears) of the alloy (T_c). The T_c for Co is very high namely 1131 °C and provides a reasonable margin for a number of additional elements by which T_c decreases. It is also true, at least for bulk material, that by adding Cr to Co the hcp structure can finally become unstable and at certain compositions it is also possible that two crystal structures (fcc and hcp) are present. Furthermore it has been shown that even two different phases, one with a high Co-rich composition (ferromagnetic) and the other with a high Cr-rich content (paramagnetic) can be formed [20].

Consequently, adding Cr to Co has two important effects, namely reduction of the T_c and M_s . For recording applications these parameters should have the right values. The T_c must not be too close to room temperature, because then the magnetic behaviour becomes too sensitive for temperature variations. M_s should have a certain value because otherwise the information cannot be read by the head. Comprehension of the physics behind the reduction of M_s and T_c is very complicated and not yet completely known. However, the most useful model in the case of Cr

being added to Co is to consider that the magnetic moment of Co atoms is reduced by electron transfer to the 3d band from Cr.

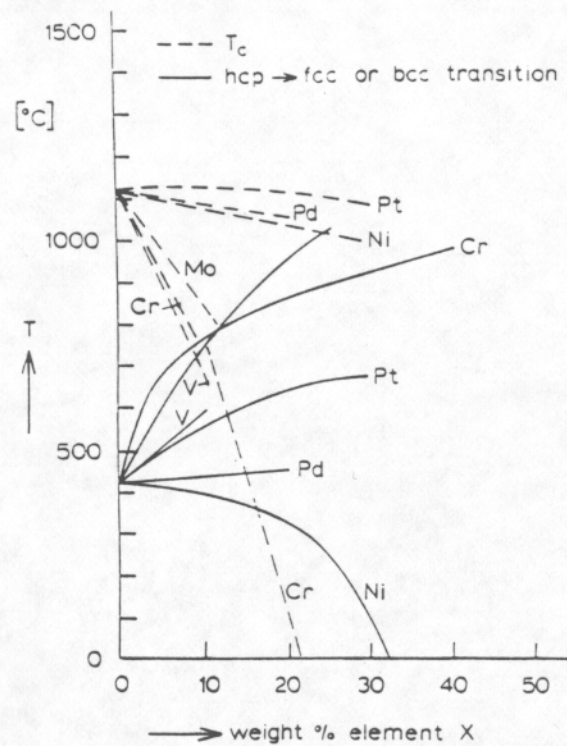


Figure 2. Phase diagrams of solid solutions of Co with various elements M [18]

For a rough indication how Cr is distributed in the Co matrix, we can use the data obtained on the basis of bulk-phase diagrams as shown in fig.2. Here the dependence of T_c vs. composition is also given as dashed lines in the figure. It can be seen that the T_c drastically decreases with the Cr content and becomes paramagnetic just above 22 wt% at room temperature. This is not expected when Cr only acts as a simple diluent. [18]. Also the transfer of 4s electrons from Cr to the 3d shell of Co may lower the magnetic moment [21]. Furthermore pure Cr is anti-ferromagnetic at room temperature and a ferromagnetic sublattice coupling also seems an acceptable explanation for the relatively strong decrease of T_c compared with other elements of M, which form a hcp phase with Co [22].

Another interesting point is that H_c can be adjusted by variation of the Cr content. Additional favourable influence on the film morphology is the reduction of column dimensions [23] and the appearance of the so-called segregation of Cr. (It is better to use compositional separation in this case). The latter has a great influence on the magnetic microstructure because it can lead to more or less exchange coupling between the columns. When the enhancement of Cr at the column

boundaries becomes higher than about 27 at% this type of coupling is completely broken but the so-called magneto static coupling is still active.

Smaller grain size gives the advantage of reduced surface roughness which results in a better head-medium interface. Last but not least the choice of Cr also improves the corrosion resistance and mechanical hardness of the Co-based medium, although in a few cases other elements have been added to increase the hardness and chemical resistance.

1.3. HYSTERESIS LOOP OF CO-CR FILM.

The following description is mainly based on the original one in Wielinga's thesis [18]. The hysteresis loop, in general, supplies information about the magnetic properties such as coercivity (H_c), saturation magnetisation (M_s), remanent magnetisation (M_r), preferred direction of magnetisation or anisotropy and the magnetisation process involved. In the following we shall concentrate on the loop of a thin film having a single domain state and a uniaxial anisotropy, which can be described simply by one single anisotropy constant K_1 . In this case the corresponding anisotropy field (H_k) is defined as: $H_k = 2K_1/\mu_0 M_s$. In fig.3 the two principle (theoretical) loops are given for the easy and hard directions.

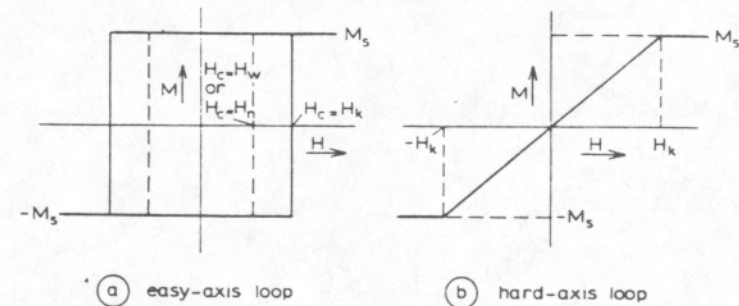


Figure 3. Easy (a) and hard (b) hysteresis loops of uniaxial anisotropic material.

The easy loop (measured parallel to the anisotropy direction) has a rectangular shape and exhibits irreversible changes of magnetisation while the hard-axis loop is linear between the saturation fields and hysteresis free. In this case, the magnetic field (H) will rotate the M according to $\sin\theta = H/H_k$ for $H \leq H_k$. Here θ denotes the angle between the magnetisation and the anisotropy axis. This loop is hysteresis free, i.e. no energy is dissipated by the process of rotation. The easy-axis loop is more complicated and has not the same character as the hard-axis loop. If M is uniform and switches by coherent rotation, H_c (the field where $M=0$) equals the H_k ($H_c=H_k$). As can be seen this description is similar as the one for a single domain particle in the case of coherent rotation. Depending of the size of the magnetic unit the magnetisation can be reversed by domain-wall motion or by rotation of the magnetisation. Domain walls are nucleated at a nucleation field (H_n) which is smaller than the anisotropy field (H_k). Since domain walls are usually hindered by imperfections, a minimum field H_w (=domain-wall movement field) is required for domain-wall movement. The two different cases to be considered are:

- $H_n > H_w$: at nucleation the magnetisation is reversed in one big jump by domain-wall motion.
- $H_n < H_w$: domains can be nucleated at fields $H_n < H_w$, but cannot grow very long, because the field H does not equal H_w . If $H=H_w$ the M is reversed by domain-wall motion.

Consequently only in the case where $H_c = H_k$ will the magnetisation process be an unambiguous coherent rotation. But if $H_c < H_k$ no conclusions can be drawn about the physical meaning of H_c in the case of a thin film having domain wall reversal, because the easy-axis loops are similar when $H_c = H_n$ or $H_c = H_w$. In the case of a small particle the mechanisms are clear defined by a number of incoherent rotation modes such as curling, buckling and fanning. In these cases the H_c is always smaller than H_k of the particle.

In the case of a perpendicular anisotropic Co-Cr thin film where non coherent switching and demagnetising fields are involved further complications arise.

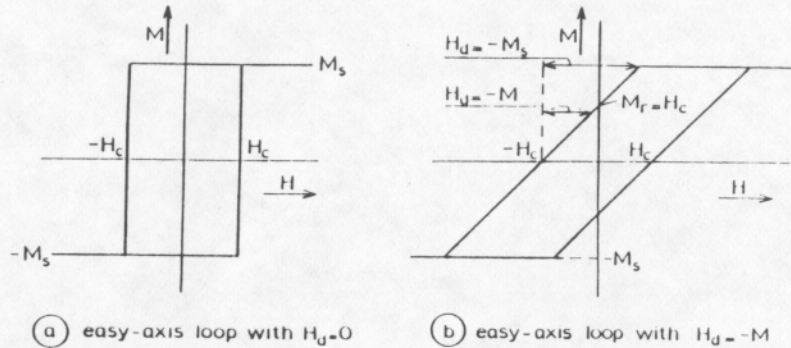


Figure 4. The easy axis loop for a single domain film with perpendicular anisotropy without H_d (a) and with (b).

Two perpendicular easy-axis loops are shown for the same film in fig.4; (a) without demagnetising field (H_d) and (b) with the perpendicular $H_d (= -M)$.

From a formal point of view it is simple if there is no physical meaning for H_c the rectangular easy-axis loop (a) can be formally transformed by the internal field ($H + H_d = H - M$). In the first order, most perpendicular magnetised layers obey this simple transformation rule quite well. But Co-Cr layers can show anomalous shearing of their perpendicular hysteresis loop depending on the magnetic microstructure (see later). This is in contrast for instance with MnBi films having a perpendicular anisotropy.

Here we are approaching a very difficult subject because the hysteresis loop is measured from the thin-film volume by a VSM and consequently gives a macroscopical "image" of the material, while the magnetisation process has a microscopical behaviour. Consequently the decision about the real type of magnetisation process cannot be based solely on the study of the hysteresis loop. Microscopical methods like Lorentz microscopy [18,24], anomalous Hall measurements [25], magnetic force microscopy [26,27], and micromagnetic simulations [e.g.28-30] should be taken into account. Continuing the discussion about the formal transformation of the easy-axis loop we met several physical implications. One important implicit assumption is that, in the case of domain wall reversal, the average magnetisation can have every value between the positive and negative saturation magnetisation. This is not possible for a single-domain film with coherent switching behaviour, consequently the magnetic microstructure must be of a multidomain type as shown in fig.5.

In this case the average M can have every value between $+M_s$ and $-M_s$ by variation of the domain width (d) under influence of a perpendicular field. The domains with a magnetisation parallel to the field will extend at the cost of the antiparallel orientated ones. Here, the (Bloch) domain walls, having a surface energy density exert a great influence on the magnetisation process, which is expected to be perceptible in the perpendicular hysteresis loop. This was first described for perpendicular films in [31] and further studied in [32].

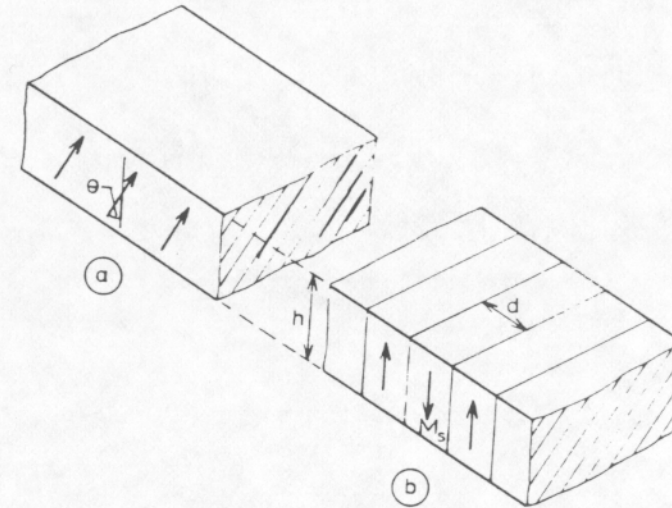


Figure 5. Magnetisation structures in a thin film (thickness h) having a perpendicular anisotropy. The uniform mode; M_s makes an angle θ with the normal (a) and the demagnetised mode with domain structure (b).

Another situation exists if the medium does not reverse by domain wall motion but by particle rotation. The average magnetisation is then built up from the sum of all particle magnetisations. One possibility is that each particle is a single domain and switches its magnetisation by rotation. Another possibility is that the particle only switches by domain-wall motion but has no stable domain configuration. The same type of easy-axis loop may be expected for both types of particles, only differing in H_c but not in shearing. If the particles do have stable domain structures, domain-wall energy is involved, which also influences the hysteresis loop. The shearing of the loop will be changed by the particulate character of the medium depending on the particle dimensions and their spacing. For further understanding please see [33-35].

Another important feature of the in-plane loop is the so-called in-plane remanence ($S_{//}$) described as the ratio between the remanence magnetisation ($M_{r//}$) of the in-plane direction and the saturated magnetisation. This parameter can be used as a measure of the perpendicular orientation of the magnetisation and it is assumed that a non-zero value of $S_{//}$ may be caused by the following [36]:

- Dispersion of the c -axis around the film normal

b) Orientations of the c-axis other than the film normal

c) Other magnetic phases with low magnetic anisotropy as for example fcc Co-Cr or magnetic oxides.

In fig.6 the in-plane hysteresis loops of a film sputtered under optimal conditions is compared with one having a different sputter power. $S_{//}$ is considerably increased from 5 to 14 %. A more detailed study was carried out recently [37] where different substrates have also been used and much thinner films have been analysed (see section 3.4).

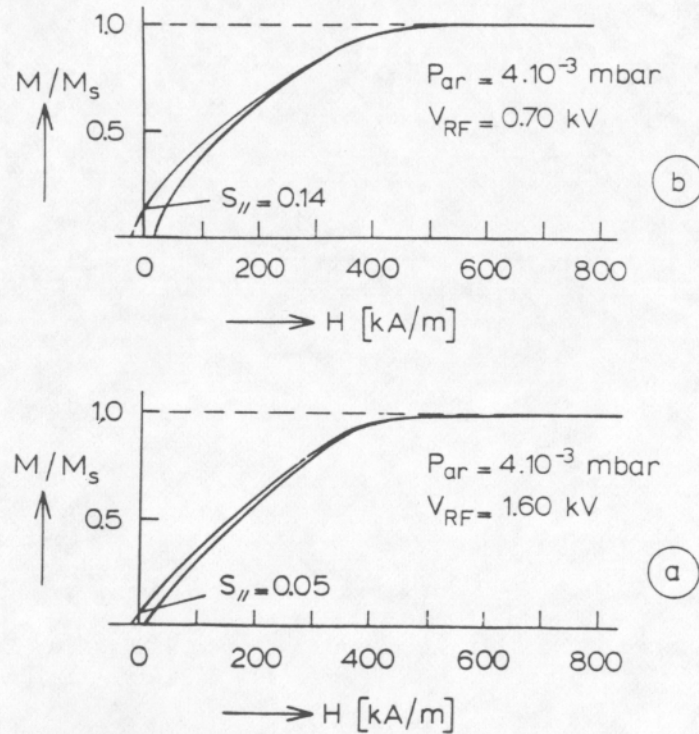


Figure 6. In-plane hysteresis loops of Co-Cr films sputtered under optimal (a) and other sputter conditions (b).

2. Preparation Methods

Nowadays many different thin-film technologies are commonly available for making thin layers in general. However Co-Cr thin-film media are only prepared by vacuum evaporation and sputtering processes.

2.1. VACUUM EVAPORATION

Evaporation of Co-Cr with perpendicular anisotropy is considered to be the best media from the point of view of production [37-40]. Continuous evaporation with the so-called roll-coater systems, for producing tapes at several hundred nm/s, have been in use since their introduction in 1984 [38]. In this case they use an electron gun as the source while the substrate is fed from a supply roll and transported around a drum during which the film is deposited. The drum is equipped with a heating stage. The rate is remarkably high and can be as much as 1000 nm/sec. Consequently, the power needed for feeding the guns will be around 20 kW. Good quality Co-Cr media have been made with this process and are described in [41]. In this process the substrate temperature (T_{sub}) plays an important role in determining the magnetic parameters (M_s , H_k and H_c).

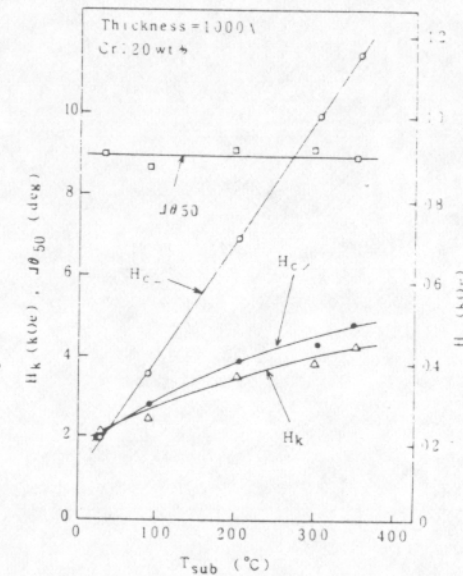


Figure 7. Magnetic and crystallographic parameters as a function of the substrate temperature for continuous evaporated $Co_{80}Cr_{20}$.

In Fig.7 magnetic parameters and the texture distribution ($\Delta\theta_{50}$) are given as a function of T_{sub} . In this case the deposition rate was 400 nm/sec. All films have a thickness of 100nm and an average at%Cr of 20. The latter shows a peak for H_k , the perpendicular coercivity (H_{cp}) and the lowest value for $\Delta\theta_{50}$ at a T_{sub} of 160 °C. The magnetic parameters all increase with the temperature, especially the perpendicular coercivity. Within the temperature range of 30-350 °C the $\Delta\theta_{50}$ is constant at about 9°, which is a relatively good perpendicular orientation of the hcp c-axis.

The effect of T_{sub} on the properties of the film is an ongoing study activity. The influence of argon-ion bombardment [42] and plasma-enhanced evaporation [43] have been studied to decrease the substrate temperature and at the same time retaining suitable magnetic properties. Again, lower temperatures can decrease the costs by using cheaper types of substrates.

Another approach to achieve suitable properties at reduced temperatures is the deposition on a seed layer of 30 nm Ge [44]. With e-beam evaporation of the Ge followed by the Co-Cr in a roll coater the $\Delta\theta_{50}$ and the relevant magnetic properties at a lower T_{sub} have been improved.

2.1.1. Incidence-angle effect. The incidence angle of the atomic flux plays an important role for the nucleation and growth processes. Atoms arriving under an angle at the substrate in general show different behaviour to those which approach perpendicularly. First of all the shadowing effect plays an important role in the film formation. If there is no adatom (physical adsorbed atom) mobility and the sticking coefficient probability equals one, an incoming atom is captured as soon as it touches the substrate or surface atom. Atoms already deposited and surface irregularities throw a shadow. No direct impingement is possible in this shadowed areas. When a non-zero adatom mobility occurs which is normally the case due to the kinetic energy of the atoms and the substrate temperature, they can then move to energetically favourable positions, including the shadowed areas. Therefore an increasing mobility will partly annul the shadowing effect. The degree of shadowing and mobility is determined by the deposition conditions. The direction of adatom movement is considered to have two contributors; namely the surface diffusion (responsible for the movement in all directions) and the angle of incidence effect which causes the atoms to have a momentum component parallel to the incidence plane. This is true for the situation where the substrate is fixed in relation to the incoming flux.

In the case of continuous evaporation a schematic view is given in fig. 8a.

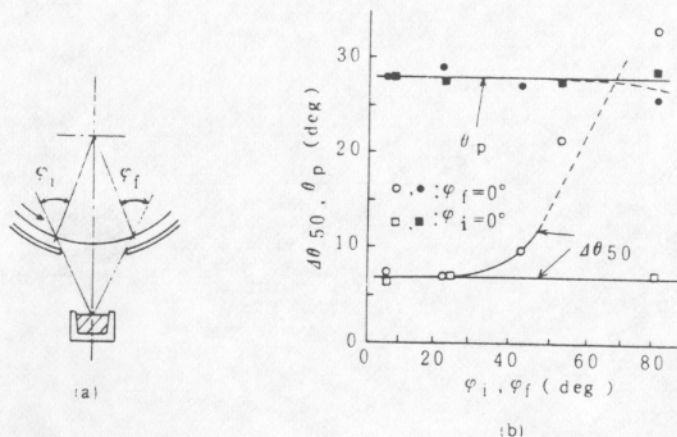


Figure 8. Schematic view of the incident angles of continuous deposition (a) and the relation between $\Delta\theta_{50}$ and the angles (b).

Here the incident angle varies continuously during the formation of the layer. This process has

been fully described by Sugita et al. [41]. The φ_i is the angle at the initial point while φ_f is the one at the final point of film formation. The dependence of $\Delta\theta_{50}$ and θ_p (which is the peak angle of the rocking curve of the (0002) planes) on the φ_i and φ_f is respectively shown in fig.8b.

The substrate transport velocity is adjusted according to the values of the two principle angles so that the thickness is about 100 nm. In this case the T_{sub} is 250 °C and the average film composition 22 at%Cr. When φ_f is fixed at 0° , the value of $\Delta\theta_{50}$ is constant at about 7° for every φ_i smaller than 30° . The $\Delta\theta_{50}$ value increases sharply with increasing φ_i over 40° , and θ_p is almost constant for every φ_i . Consequently the c-axis is oriented perpendicular to the film plane and the perpendicular anisotropy is obtained for values of φ_i smaller than 40° . However when φ_i is fixed at 0° there is no dependence of $\Delta\theta_{50}$ and θ_p on φ_f . If φ_i now remains constant, $\Delta\theta_{50}$ is constant for any value of φ_f and $\Delta\theta_{50}$ is smaller than 10° for φ_i less than about 40° . The authors also reported that in this process the tangent rule is valid. This means that the curve of the columns coincides with the change of the incident angle through the film growth. This indicates, according to the authors, that the growth of the film itself is epitaxial even at this very high rate of deposition. The most important deposition parameters are the transverse length of the evaporation source, the distance between the source and the substrate and the evaporation rate distribution in the transverse direction. The method described here has a material yield of more than 50%.

2.1.2. Co-evaporation. Using a predetermined source composition there will be degradation of composition as a function of the time.

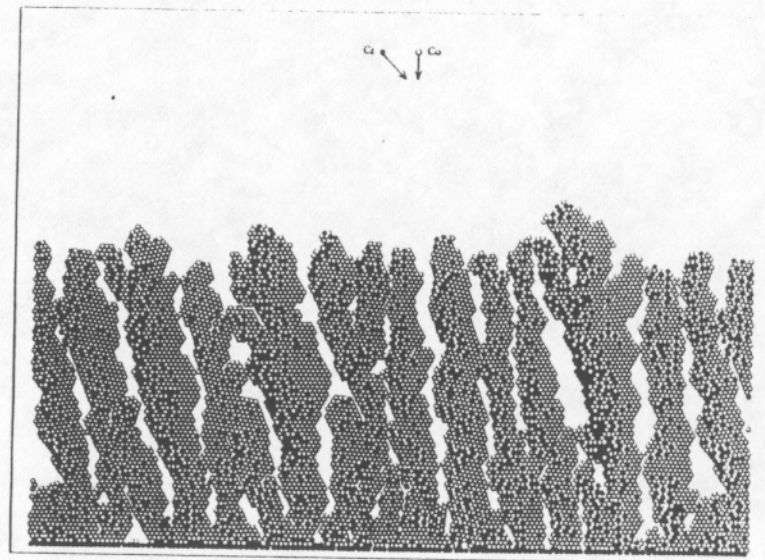


Figure 9. Simulated microstructure with 22 at% Cr and 78 at% Co. The arrows above the simulated structure indicate the vapour flux [50].

The loss of Cr as a function of the deposition time is due to the higher vapour pressure in relation to Co. The differential evaporation rates have been studied in [42] and [43]. Finally the progressive exhaustion of the alloyed source can lead to composition differences along the thickness of the film.

We tried co-evaporation of Co and Cr to overcome this problem [45]. At an intermediate incidence angle (deviating 30° from the normal) we found a tilt of the columns towards the Co source (the evaporation rate of Co was about 4 times larger than that of Cr) but also the hcp c-axis was slightly tilted in the same direction. Nevertheless the films have shown good properties for recording. The advantage of this method is that even at low temperatures high Hc's can be obtained which find their origin in the process-induced compositional separation of Co and Cr caused by the shadowing effect during the deposition process. Improvements have been shown by [46] where a seed layer of Ta was introduced for enhancement of the perpendicular anisotropy. In order to study the microstructural behaviour and the magnetic properties in more detail extended studies have been published [47-49] in which details of the compositional separation have been investigated by NMR, selective etching and electron-microscopical observation. The influence of the process variation on the magnetic and structural properties have also been studied in detail. The conclusion is clearly due to the effect of Cr separation by the shadowing mechanism, films with high coercivities and good perpendicular anisotropy can be achieved without heating the substrate. That is an advantage for deposition on plastic substrates. The nucleation and growth of this process have been simulated based on a 2D-Monte Carlo program and confirm the experimental results [50]. An example is given in fig. 9.

The only disadvantage of this co-evaporated method as performed in our laboratory is the very low deposition rate of our deposition apparatus. For commercial applications, studies as a function of high rates have to be carried out.

2.2 SPUTTERING

Sputtered Co-Cr films were first prepared by the group of Professor Iwasaki at the Tohoku University in 1975 [51]. The development of Co-Cr layers having a perpendicular anisotropy is strongly related to the application of the sputtering process. Although there are many parameters influence the final results of the layer such as: the sputtering mode (DC, RF, Magnetron, Ion Assisted, Target Facing Target Sputtering), the geometrical arrangements in the deposition unit (target/substrate distance, target size, substrate size), the type of target (alloyed or multitarget), we will concentrate here on the most important sputter parameters such as substrate temperature, argon pressure and power.

There are many excellent papers in the literature in which the optimisation process has been described for their own deposition processes and apparatus. Here our own results, using a commercial Leybold glow-discharge Rf sputtering unit (type Z-400) with the possibility to use the magnetron mode are mainly reported. Our data on sputtered Co-Cr films have been mainly described in [52-57].

In the case of RF sputtering an alloyed target was used (19 at%Cr), having a diameter of 10 cm and a purity of 99.99%. The target-to-substrate distance was fixed at 5 cm. The substrate holder was water-cooled but a heating element was installed to vary its temperature (T_{sub}). After a background pressure of 2.10^{-7} mbar was reached by pumping 10 hours with a turbo-molecular pump, the pressure was then further reduced to 10^{-7} mbar by using a liquid nitrogen Meissner

trap. The sputter gas (Ar) had a purity of 99.998%. Before deposition the target was sputter cleaned for 30 min and the substrates cleaned by means of glow discharge. The film composition appeared to be independent of the sputter parameters and close to the target composition. It was not known in advance how much the parameters were mutually dependent. This complicated the optimisation process and forced us to vary each parameter for various values of the others. In order to reduce the number of runs for a systematic variation of all parameters, we first fixed the sputter voltage (which is directly related to the sputterpower) at 1.60 kV (=VRF) when varying the P_{Ar} and used the water-cooled substrate holder. At this power a reasonable deposition rate R (=0.2 nm/sec.) was obtained.

2.2.1. *Sputter-gas pressure.* The $\Delta\theta_{50}$ and the in-plane remanence $S_{//}$ of a 150 nm thick Co-Cr layer are given as a function of P_{Ar} .

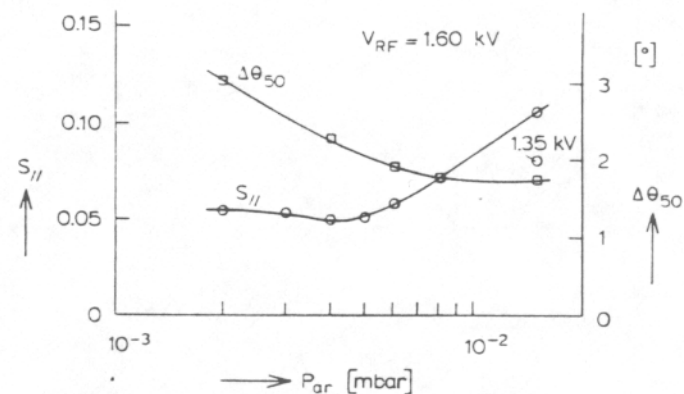


Figure 10. In-plane remanence and $\Delta\theta_{50}$ of a 150 nm thick sputtered Co-Cr film as a function of the Argon pressure.

The lower limit of P_{Ar} is determined by the requirements for stable plasma operation, which is about 2.10^{-3} mbar for our configuration. The $S_{//}$ exhibits a faint minimum at $P_{\text{Ar}} = 4.10^{-3}$ mbar and increases strongly at higher pressures. Surprisingly the $\Delta\theta_{50}$ shows the opposite behaviour, viz. an improving normal c-axis orientation at higher P_{Ar} . Obviously the $S_{//}$ originates from a not well oriented normal c-axis and the presence of other phases than hcp. However the (00.2) rocking curve exhibits no other orientations of the c-axis than the perpendicular one, but electron diffraction reveals the presence of the {10.1} and the (00.2) reflections in layers, sputtered at 2×10^{-3} and 1.5×10^{-2} mbar. These reflections are not observed in the layer sputtered at 4.10^{-3} mbar. No reflections of other magnetic phases are observed, such as fcc Co-Cr as was concluded by the absence of the (200) fcc reflections [56]. Furthermore the diffraction rings in the electron diffracted image of the sample sputtered at 1.5×10^{-2} mbar are more blurred, which indicates a defect-rich crystal structure. This is not accompanied by an increase of $\Delta\theta_{50}$, which improves at higher P_{Ar} .

The increase of $S_{//}$ for P_{Ar} which differs by 4.10^{-3} mbar, is apparently caused by the additional presence of a hcp compound with in-plane orientation of the c-axis. In the following

runs the P_{Ar} was therefore kept constant at 4.10^{-3} mbar. The V_{RF} was then varied between 0.70 and 2.0 kV. The results are given in fig.11.

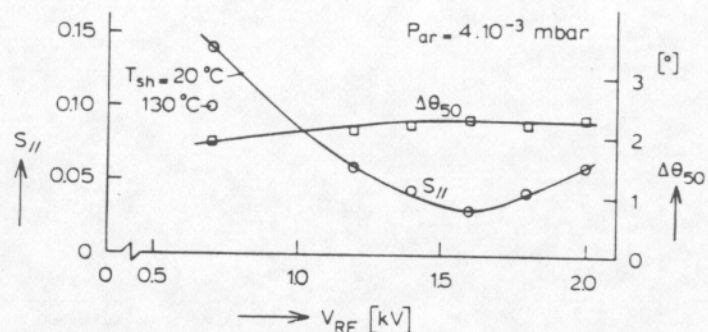


Figure 11. The relation between the in-plane remanence and the sputter power.

The $S_{//}$ has a clear minimum at $V_{RF}=1.60$ kV but again there is no correlation between $S_{//}$ and the c -axis dispersion. The $\Delta\theta_{50}$ is almost constant over the entire voltage range, showing only a slight increase in the lower part, while no other orientations of the c -axis are revealed by the (00.2) rocking curve. Layers sputtered at a different V_{RF} value than the optimal (1.60 kV), show the presence of a certain amount of hcp phase with in-plane orientation of the c -axis, as indicated by the {10.1} and (00.2) rings. Also the diffraction rings of the high voltage layer are more blurred, like those of the layer sputtered at high P_{Ar} .

2.2.2. *Substrate temperature.* The next step was to research the influence of the substrate temperature.

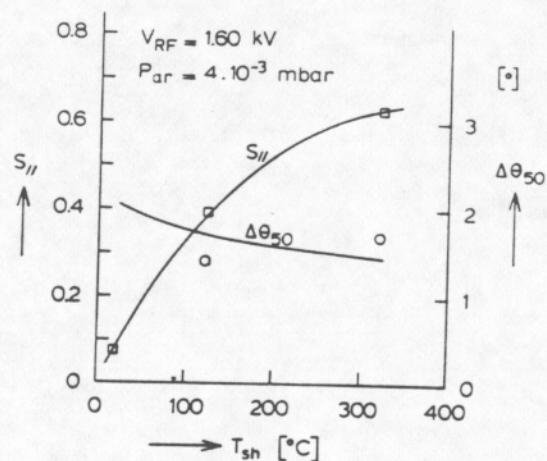


Figure 12. In-plane remanence and $\Delta\theta_{50}$ as a function of the substrate temperature.

In fig.12 the $S_{//}$ and the $\Delta\theta_{50}$ are given as functions of the T_{sh} . It can be seen that at an elevated temperature the magnetic orientation has strongly deteriorated. The increase of $S_{//}$ is, in fact, so steep that the lowest possible ambient T_{sh} of 20 °C seems to be very critical. Unfortunately no substrate cooling, e.g. by means of liquid nitrogen, was possible with our equipment. Nevertheless it appears that the perpendicular magnetic orientation depends strongly on T_{sh} . At elevated temperatures, $\Delta\theta_{50}$ hardly changes, but again the {10.1} and (00.2) diffraction rings become apparent and more blurred at increasing temperature [57]. The very strong relation between the in-plane c -axis orientation and the temperature may imply that the dependence of this parameter on the others is indirectly caused by T_{sh} variations. An increase of V_{RF} for example is accompanied by an increase of the heat flux to the film surface, not only by secondary electrons from the target, but also by the increased kinetic energy of the condensing atoms.

Although we know that the measured "bulk" temperature of the holder is not the same as the film-growth temperature we can only determine the first one. The latter will at least correlate to the actual temperature at the surface of the layer.

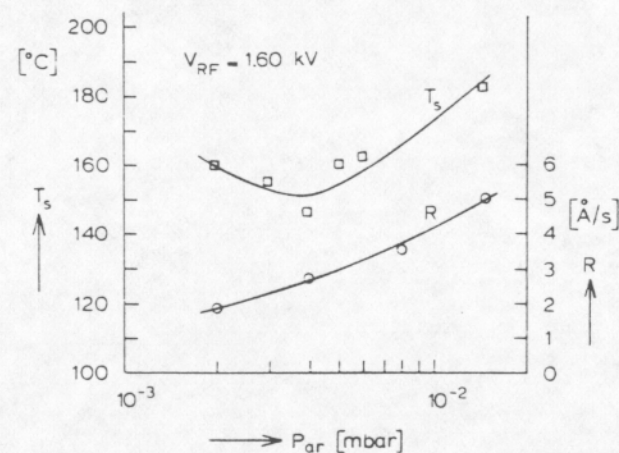


Figure 13. The substrate temperature and the deposition rate as function of the sputter power.

The substrate temperature T_s (see fig.13) is determined as a function of the P_{Ar} at a fixed power. Surprisingly T_s exhibits a minimum at $P_{Ar}=4.10^{-4}$ mbar, as was also found for $S_{//}$. This minimum temperature is probably caused by the competitive heating caused by the kinetic energy of the condensing atoms and the secondary electrons from the target. At lower P_{Ar} the heating of the secondary electrons is reduced, but the mean free path in the argon gas is simultaneously enlarged. As a result the sputtered atoms will arrive at the surface with a higher kinetic energy, less decelerated by collisions with argon atoms. The minimum of T_s is not caused by an anomalous minimum in the sputter power because R continuously increases in the entire Argon pressure range as can be seen in fig.13. In [58] a so-called disorder temperature T_d has been defined, which is the temperature at which the $\Delta\theta_{50}$ has the smallest value. It was found the lower the M_s (more average Cr content in the film) the higher the T_d . In fig.14 the T_d is given as a function of M_s together with T_{Tr} (the transition temperature at which the Co-Cr changes from

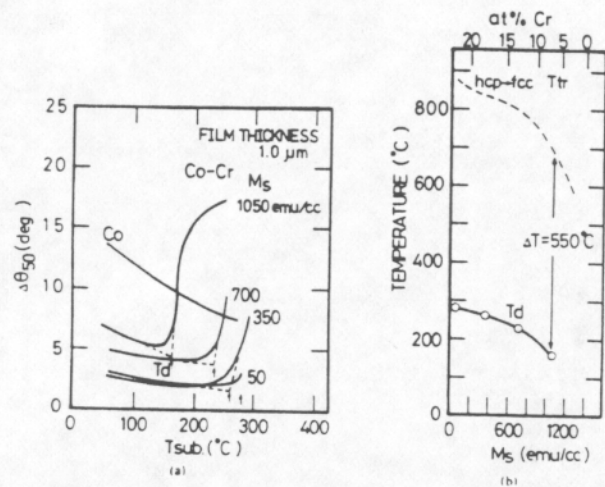


Figure 14. Relation between substrate temperature and $\Delta\theta_{50}$ for a pure Co and Co-Cr (a) and the disorder temperature (T_d) as a function of M_s (b).

hcp into fcc). It can be seen from this that the temperature difference (ΔT) is about constant ($=550^\circ$).

That means that the experimentally determined T_{sub} does not correspond directly to the real film-growing (surface) temperature. That can be understood by considering the following: In rf sputtering the surface temperature is predominantly determined by three factors which are in the same order of magnitude namely:

- the temperature of the substrate holder
- the temperature produced by heat from the condensing atoms
- the temperature mainly produced by the electrons out of the sputter plasma.

This complex heat mechanism not only makes it difficult to determine an accurate surface temperature (whatever this may mean) but also implies that this temperature will not be stationary during the sputter run. Consequently inhomogeneities can easily be obtained by this process.

2.2.3. Sputter power. In our sputter system we identify the Rf voltage directly with the sputter power because this is the parameter we can really vary.

In fig.15. the V_{RF} was varied and determined the T_s and the R at the P_{Ar} constant. There is a continuous increase of T_s and R with increasing power. This corresponds qualitatively with the rate of decrease of $S//$ (see fig.11)

2.2.4. Bias sputtering. As was already pointed out the properties of the growing film can be influenced by changing the flux and energy of the incident particles. It is difficult to control the neutral particles from the plasma but the charged ones can be influenced by the local electric field. A negative bias voltage can be applied to the substrate holder. Consequently a resputtering of the absorbed gasses as well as the atoms can take place depending on the binding energies and the level of the bias voltage.

Topography and chemical composition can be altered by this mode of sputtering. The influence on the structure and magnetic properties is given in [59]. In the system used a small bias voltage leads to a decrease of M_s and H_c which is attributed to a homogenisation of the Cr distribution.

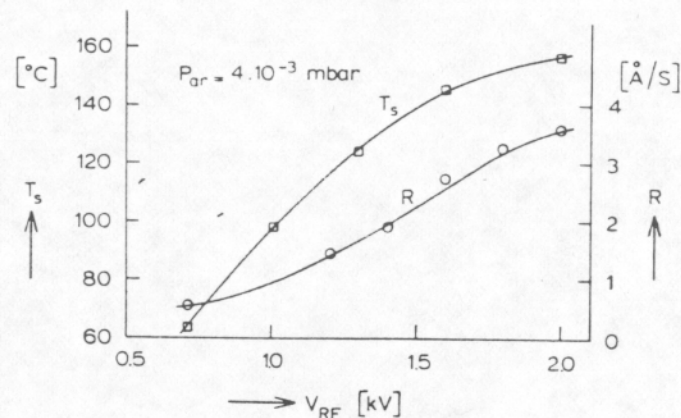


Figure 15. Substrate temperature and rate as a function of the sputter power.

Also $\Delta\theta_{50}$ is drastically influenced by the V_{bias} .

Similar M_s results are also mentioned in [60] and the authors also found a reduction in the number of defects and stacking faults due to the desorption of impurity gasses.

3. Microstructure, Texture And Morphology

3.1. CHEMICAL INHOMOGENEITIES OR COMPOSITIONAL SEPARATION.

Deposited Co-Cr layers show composition inhomogeneities composition in the growth direction as well as in the lateral direction. Deposition parameters such as the substrate-temperature and the use of a seed layer play an important role. In the literature the $\Delta\theta_{50}$, the effective anisotropy and the coercivity in the perpendicular direction are mostly used as indicators for optimising the properties.

There are, in principle, two driving parameters for obtaining the compositional separation namely temperature and deposition geometry. Detailed studies on the chemical inhomogeneities in Co-Cr are seldom found. Chemical separation of Cr in Co is very often called "segregation" and this can be found at the column boundaries or even in the column itself. The boundary segregation is also described as oxygen gettering [23] which means that during the growth the Cr mainly reacts with oxygen, thus increasing M_s of the ferro-magnetic composition.

Another explanation is that there is a recombination of atoms, molecules or clusters before they interact with the substrate [61]. The forming of Cr-Cr and/or Co-Co clusters gives a compositional change. The same result can be obtained by using non homogenous targets (in the case of sputtering).

Higher Cr concentration on the lattice faults present in the column have been mentioned in [62]. Other explanations, such as Cr migration to the boundary due to thermodynamic reasons, can be found in [63-65]. These explanations are based on the fact that the Co-Cr system strives for a low as possible surface energy by the enrichment of the boundaries with Cr atoms because Cr has a larger surface than Co and also the binding energy is lower. The Cr atoms are exchanged with Co atoms at the surface and also to the column boundaries. The Cr distribution is dependent on deposition parameters such as temperature and energy of the incoming particles.

Very often the phase diagrams of the bulk Co-Cr system have been used to explain the chemical composition although such a diagram is only valid at thermodynamic balance. There has been much literature published on the Co-Cr binary phase diagram; the most important so far being the diagrams discussed in [66-68]. Recently a complete overview and new data were published [69] and this one seems to be the most complete version at present and is used hereafter (see also fig.16).

With respect to the application of Co-Cr as a thin-film medium for magnetic recording the most interesting area of the phase diagram is around the temperature range from room temperature to 1000° C. Furthermore the interesting compositional area is ≤ 35 at% Cr. In this part of the diagram two phases, α -fcc and ϵ -hcp are represented.

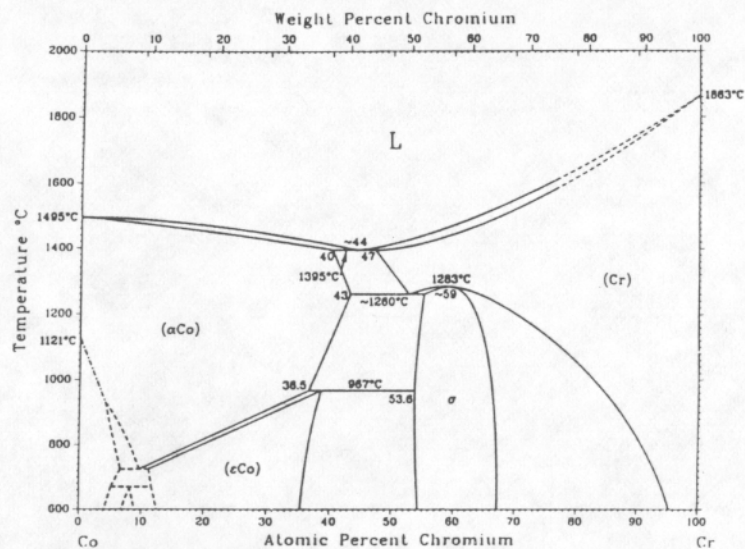


Figure 16. Solid phase diagram of Co-Cr [69].

It can be seen from fig.16 that even at 35 at% Cr a hcp ϵ -Co can exist. There can be two phases, one with a high Co-rich composition (ferromagnetic) and the other with a high Cr-rich content (paramagnetic). The phase diagrams are based on an equilibrium process and derived from thermo-dynamical calculations. The thin-film materials discussed here are made by deposition which is by definition a non-equilibrium process. In [69] a very important fact was assumed,

namely that the phase diagram at high Co concentration below 800°C is very complicated because sluggish diffusion inhibits the equilibrium.

It is known, for instance, that during sputtering the surface temperature of a growing film is quite different from that of the substrate [70]. The bombardment exerted by various particles from the plasma on the surface results in a much higher temperature at the surface.

Most of the authors explaining the magnetic behaviour of Co-Cr thin films have suggested that there is a Cr segregation at the columnar boundaries which can explain the higher coercivity and the magnetic reversal behaviour of the layers. However only a few research groups have experimentally shown the existence of such a compositional separation. Nevertheless the grain boundaries are assumed to be fast diffusion paths at which diffusion takes place by vacancies [71]. Besides the segregation at the boundaries Maeda et al. [72] were the first to have introduced the so-called chrysanthemum-like pattern (CP model), which is also related to the temperature as most important parameter. In this model the enhancement of Cr at the boundaries is a special case of the CP pattern (see fig.17).

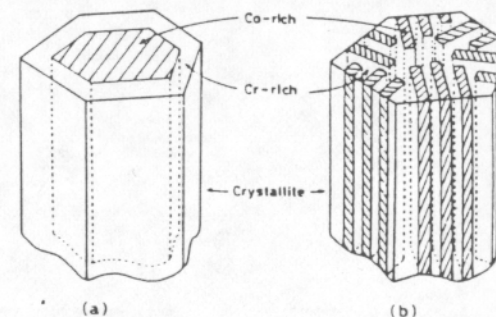


Figure 17. Compositional separation in a Co-Cr column consisting of Cr-rich boundaries and Co-rich core (a) and the CP structure of Maeda [72].

The CP structures are observed by TEM but also in the case of very thick samples by SEM [73] these observations are only possible in combination with selective etching techniques [74]. The main features of the CP structure are Co-rich stripes that tend to be perpendicular to the column boundaries. The CP structure also develops as a function of the thickness of the layer but mainly on the substrate temperature.

The Co-rich component and the Cr-rich component have a great influence on the magnetic properties. The spatial periodicity of compositional separation in sputtered Co-Cr layers on top of soft magnetic Ni-Fe layers is in the range of 3-7 nm [73].

3.2. COLUMNAR STRUCTURE AND "GRAIN" SIZE.

Nearly all deposited Co-Cr films mentioned in the literature have a so-called columnar structure. On various substrate materials (without any seed layer) we always found that the columnar diameter increases with the layer thickness (see fig.18.).

Using a seed layer a columnar diameter is more or less constant over the total thickness of the layer. The method of deposition also has a large influence on the development of the column

diameter. The diameter depends on the Par (lower pressure shows a smaller diameter [75]) and the Vbias (larger voltage show finer columns [76]). We found significant differences between RF and RF magnetron sputtered layers. In our RF sputtered layers it was observed that the columnar diameter increases almost linearly with the film thickness.

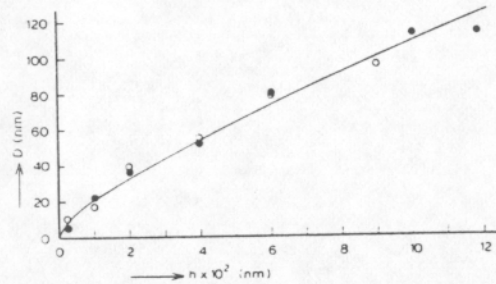


Figure 18. Columnar diameter (D) as a function of the layer thickness (h) for films sputtered with a multitarget (solid circles) and alloyed target (open circles).

Extraordinary column growth (nodular growth) in the case of Co-Cr deposited on polymer substrates was systematically examined in [77].

3.3. CRYSTAL STRUCTURE AND TEXTURE.

As mentioned above deposited Co-Cr films have an hcp crystal structure. The crystallographic data do not differ substantially from the well-known data of α -Co [54]. The lattice parameters of 15 at%Cr layer were determined as $a=0.252$ nm and $c=0.406$ nm. These give a c/a ratio of 1.62 which is almost the same as the ratio for hcp α -Co [ASTM Index File 5-727]. The 2θ values for our Co-Cr material show a small systematic shift with respect to those for α -Co.

The texture of a polycrystalline material can be simply defined as the crystallographic orientation distribution direction. In the case of a ferromagnetic thin film the orientation distribution of the column is of great importance because it determines the strength and the uniformity of the magnetic anisotropy. In the case of Co-Cr it is clear that we need to have hcp crystals with a very small deviation of the c -axes with the normal. The texture of our films has been measured with a Philips texture goniometer using Co $K\alpha$ radiation. The $\{00.2\}$ and $\{10.1\}$ pole distributions were measured with a pitch of 2.5 by means of the Schultz method [78]. Because these pole figures showed a high degree of rotational symmetry we decided to examine all other layers by the so-called rocking curve method where the sample is scanned in a curved line instead of spirally as in the previous case.

Fig. 19. shows three different rocking curves of sputtered Co-Cr layers on Si substrates having thicknesses of 25 nm (a) 800 nm (b) and one on glass (c) also with a thickness of 800 nm. From these measurements the full width at half maximum (FWHM) was determined as $\Delta\theta_{50}$. This angle represents the dispersion of the $[001]$ direction (hcp c -axis) around the normal of the film surface.

The relation between $\Delta\theta_{50}$ and the layer thickness is shown in fig.20 using various substrate materials.

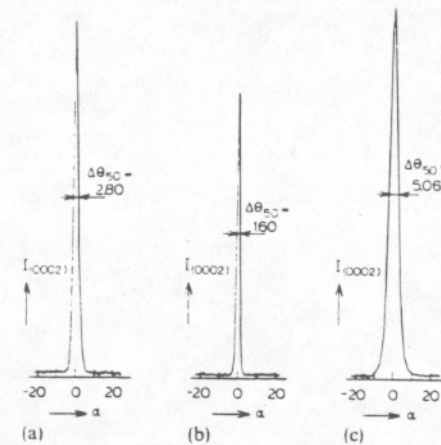


Figure 19. The $\Delta\theta_{50}$ values for $\{0002\}$ planes measured by XRD for Co-Cr sputtered films with thicknesses of 25 nm (a) and 800 nm (b) on Si and (c) 800 nm on glass.

Furthermore not only the type of substrate but also the cleaning procedure of the substrate is important for a well-oriented layer. Measurements of $\Delta\theta_{50}$ of a 25 nm thick Co-Cr sputtered layer on a glow-discharged cleaned Si substrate and an ordinary Si surface yielded values of 1.5° and 5.5° respectively. The power should be low to be sure that no etching of the Si occurs. Cleaning of the substrate means only the removal of impurities like H, H₂O and O₂, which is confirmed by SIMS studies.

Another influence on the orientation is caused by the sputter gas pressure and background pressure. It has been shown that a small amount of residual N₂ gas in the process chamber can cause the formation of the fcc phase which also destroys the well-oriented hcp structure [79] and [80]. In our RF sputtered system we also found an increase of $\Delta\theta_{50}$ caused by an increase of the sputter-gas pressure at a fixed power and target-to-substrate distance [56]. Higher Ar pressure means that the Co-Cr atoms are more scattered during their movement from target-to-substrate and consequently their kinetic energy is reduced. Therefore, for producing well-oriented films (i.e. $\Delta\theta_{50} < 5^\circ$), each sputter equipment has to be optimised for certain process parameters [56]. For depositing well oriented layers on polymer-like substrates many authors have proposed the deposition of a (non-magnetic) seed layer. The function of such a layer is to create a good surface for proper growth conditions as well as an intermediate layer between the plastic and Co-Cr film. The latter protects the film against gas evaporated from the plastic during production. Much data have been published about the deposition of non-magnetic underlayers or seed layers to improve the nucleation and growth of the Co-Cr layer and to decrease the $\Delta\theta_{50}$. The c -axis dispersion of evaporated layers on polyimide was discussed in [81] while for sputtered Co-Cr films a range of nucleation layers have been discussed in [82].

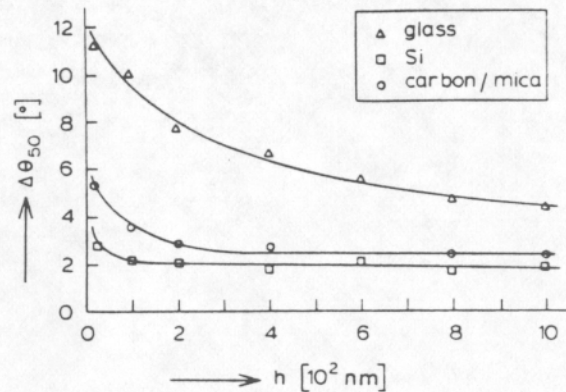


Figure 20. $\Delta\theta_{50}$ as a function of the layer thickness for various substrates.

In the case of evaporated layers the authors come to their conclusion that especially amorphous Ge lead to a decrease of $\Delta\theta_{50}$ and consequently an increase of the effective anisotropy field.

3.4 LAYER GROWTH AND INITIAL-LAYER EFFECTS.

The thickness of Co-Cr media has been reduced by a factor of 10 during the last decade and is nowadays about 150 nm thick. At this thickness it will become clear that the influence of top and bottom properties will be greater than the total layer behaviour. Several authors have reported on the existence and the properties of initial layers in Co-Cr films [45, 54, 81, 83-86].

For reasons of clarity the definition of an initial layer as it is used in this paper is given here. The initial layer in a Co-Cr film is the part of the layer that is closest to the substrate, where the crystal orientations are random in the sense that the hexagonal c-axis is randomly distributed in the plane of the film, while the well-known columnar structure has not yet developed (see fig.21).

It has been shown that magnetic and structural properties can be improved by the use of appropriate substrates or seed layers, such as Ti, Si, and Ge [54,54,81,83]. Several authors have adapted a so-called sub-layer (a division of one layer into several) model [84-86], by which they have explained the change in the thickness direction of the perpendicular coercivity, the saturation magnetisation and the hexagonal c-axis dispersion. This variation in the thickness direction was found to influence the magnetisation reversal mechanism, as concluded from magnetisation curves [86] and recording experiments [page 196 in 27].

Quite recently we studied in more detail [87] the existence of an initial layer in our films and several structural and magnetic properties of a thickness series of Co-Cr films in the range of 5-200 nm on different substrates and seed layers. First, the structures of the films are discussed from the viewpoint of their nucleations and growth mechanisms. This gives us a starting point to consider the influence of the initial layer on the macroscopic magnetic properties, as they are expressed by the in-plane remanence ratio (M_r / M_s)).

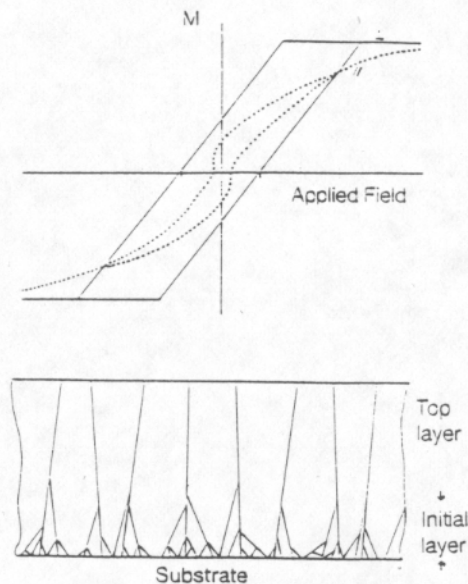


Figure 21. The perpendicular (solid) and in-plane (dotted) hysteresis loops for a Co-Cr film with an initial layer (top figure) and a schematic impression of the connecting microstructure (bottom).

The interaction between the substrate and the impinging atoms or clusters during the deposition process determines the subsequent growth mechanism. In general three growth modes are considered [88]. If the interaction energy (adhesion) between the atoms of the deposit and substrate is high compared to the cohesion of the deposit atoms, a layer-by-layer growth mechanism (2-D) will dominate (Frank-van der Merwe, or F-M). On the other hand, when the cohesion of the deposit atoms is much higher than the adhesion between the substrate and the deposit atoms, the growth will take place through island formation (3-D, Volker-Weber or V-W). If the interaction energy difference between the cohesion and adhesion is not that pronounced, growth takes place by formation of a layer (or several layers), followed by three-dimensional island growth. This growth mechanism is called Stranski-Krastanov (S-K) growth, and is often explained by including elastic lattice distortions in the consideration of the energy of the deposit layer. The initial stage of growth establishes a lattice relaxation of the deposit in order to reduce the misfit between deposit and substrate. The transition to island growth takes place because the total energy becomes smaller if the adhesive forces have a smaller range than the elastic forces [88]. In the case of sputtered Co-Cr films it is proposed that, under optimised conditions, with some substrates / seed layers a sort of reaction layer is formed [89], i.e. Ti, C, and amorphous Si or Ge. In [81] it is suggested that the interaction between a Co-Cr film and a Ge seed layer consists of a sort of epitaxial-like growth mechanism on the Ge(110) and Ge(111) planes, present in the form of micro-areas in an amorphous Ge matrix. However, the AES depth profile analysis shows the formation of silicides [89] (i.e. a region containing silicon, cobalt and chromium). In

[81] it is found that the solid solubility of Co in silicon is larger than that of Cr, as also stated in [89], but the diffusion coefficient of Cr in silicon is larger than that of Co. Also, at relatively low temperatures ($T < 900$ °C), the metal is the dominant diffuser in silicon, instead of it being otherwise. Furthermore, the heat of formation of $MSix$ (with $x = 1/2, 1, 2$) is larger with $M = Cr$ for $x = 2$ than with $M = Co$, but smaller for $x = 1/2$ or 1. Thus, it is reasonable to assume that the cobalt silicides and chromium silicides will form the reaction layer in about equal proportions. If the reaction layer is responsible for the observed improved oriented growth on silicon, the same improvement, found on Ti, Ge and C layers supports an extrapolation of the silicide formation to titanide, germanide and carbide formation. More evidence about this is given in a report by Schrauven et al. [82], excluding the possibility of a sort of epitaxial growth by considering the effect of lattice (mis)match on the $\Delta\theta_{50}$ values for different lattice planes of Ge. In the reaction layer these (silicide etc.) compounds would then be formed during the initial stage of the sputter deposition. The layer probably acts as a lattice-misfit relieving layer and also as a nucleation layer that does not exhibit any micro-roughness, on which subsequent oriented growth takes place. Experiments that did not incorporate this reaction layer, thus not compensating for the micro-roughness, resulted in a worse oriented layer (as shown by their respective $\Delta\theta_{50}$ values).

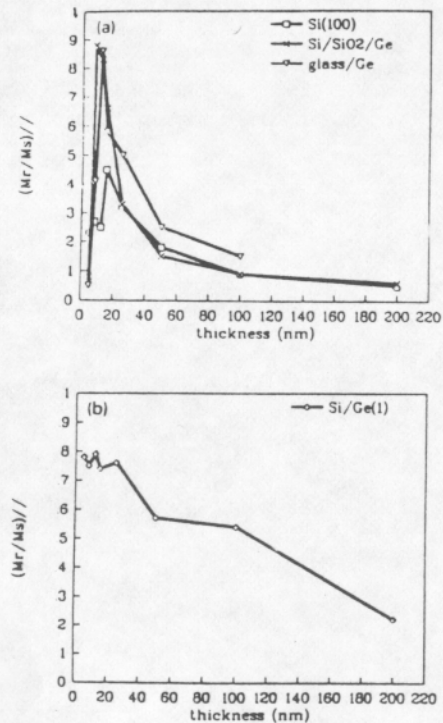


Figure 22. In-plane remanence ratio vs. Co-Cr layer thickness on different substrates. [89]).

This reaction layer-type of growth is a form of Stranski-Krastanov growth.

Although the $\Delta\theta_{50}$ values show the degree of orientation of the c-axis normal to the film plane, they do not supply complete information about the initial layer, which means that the crystals that are totally out of the normal orientation, e.g. the (100) and (101) orientations, have no effect on $\Delta\theta_{50}$ [91].

Thus, in addition to the crystallographic anisotropy information derived from the $\Delta\theta_{50}$ values, the in-plane remanence ratio gives a measure of the in-plane magnetic anisotropy in all orientations. In fig.22 the in-plane remanence ratio is shown as a function of thickness for several substrates and seed layers [87].

The in-plane remanence ratio shows the same general behaviour for all substrates / seed layers (fig.22a) except for the Si/Ge(1)-series (fig.22b). Initially it increases and after reaching a maximum it falls again. This indicates that the thinnest Co-Cr films ($d < 10$ nm) contain relatively more crystals that have their magnetic moments in the perpendicular direction than films just above 10 nm. Thicker films ($d > 50$ nm) show an improvement of the perpendicular orientation of the magnetic moment, as has also been shown in [54].

3.5. SURFACE PROPERTIES

For very high-density recording the surface becomes more and more important. On the one hand the surface smoothness and wearability are important for the fact that the head-to-medium distance is very close and on the other hand, for writing as well as for reading, the magnetic behaviour is a key factor [91-95].

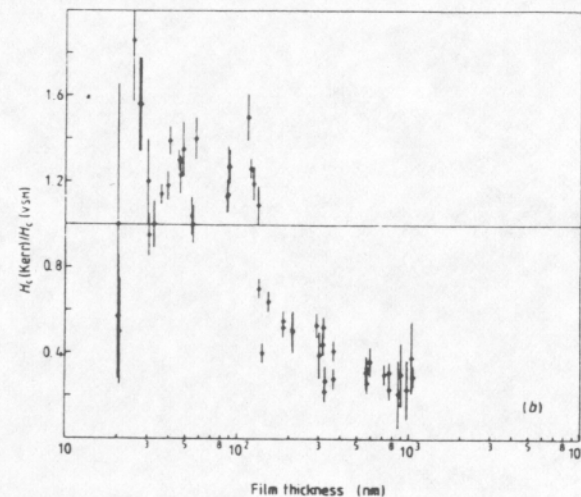


Figure 23. Surface/volume coercivity as a function of the layer thickness for RF sputtered Co₈₁C₁₉.

Therefore analyses of the chemical and structural properties (e.g. surface topology) in relation to

the magnetic properties are needed. Magnetic properties of the surface (from 5-50 nm) are important in relation to the Co-Cr films. The magnetic surface is characterised by the magneto-optical Kerr effect [96], the surface roughness with SEM and AFM/STM observations and the chemical state with AES and XPS studies [97]. The relation between these properties for Co-Cr has been studied in detail in [98]. Surface magnetic hysteresis, measured by the Polar M.O. Kerr effect on RF and Magnetron sputtered Co-Cr (81/19 at%) films in the range of 20-4000 nm is compared with the volume hysteresis measured by VSM, both with the external field perpendicular to the surface.

It can be concluded from these measurements that the surface coercivity H_{cs} is found to decrease below the volume coercivity at a critical thickness of 125 nm for RF films [57] and at about 1000nm for Magnetron sputtered samples [99].

The difference between the critical thicknesses of the two different sputter methods is difficult to explain because its origin is either due to the magnetic or microstructural properties which can originate from structural and chemical differences. One conclusion is obvious, namely that for films with different surface and bulk hystereses, the magnetisation cannot be homogeneous throughout the film thickness during all stages of the hysteresis curve.

In the case of the perpendicular recording mode the roughness and the oxidated/segregated top layer will redistribute the magnetic charges at the surface. Based on experimental data concerning chemical surface properties and roughness of the Co-Cr the reduction of the maximum readable information density have been calculated in [123].

4. Magnetic Properties And Microstructure

The basic magnetic properties such as magnetisation, coercivity and anisotropy depend on microstructural properties, compositional separation of Co and Cr and phase separation. These inhomogeneities are the main source for the deviation of the saturation magnetisation (M_s) from the expected value for the alloyed (bulk) composition.

4.1. PERPENDICULAR ANISOTROPY

The demagnetising field also plays an important role especially in the case where there are media with perpendicular anisotropy. For a thin continuous layer, having no anisotropy sources, the magnetisation prefers to lie parallel to the film plane, being the state of minimum energy. In the case of Co-Cr it is, in principle, a competition between the uniaxial anisotropy of the hexagonal structure and the demagnetising energy of the thin film. In other words there is competition between the perpendicular anisotropy field H_k and the demagnetizing field H_d .

Consequently if the morphology is not similar to a continuous film but more to a particulate structure (less magnetic exchange between the neighbouring columns; i.e. the columns are magnetically isolated) then even shape anisotropy can contribute to the perpendicular anisotropy. Mostly calculations on perpendicular films are started with a state of uniform perpendicular magnetisation and from this a so-called quality factor can be derived as $Q=2K_1/\mu_0 M_s^2$ (or H_k/H_d). In the case of a uniform magnetised layer, if $Q>1$, the perpendicular axis will thus be the direction of the preferred magnetisation. However the state of uniform perpendicular magnetisation is quite unusual; either the magnetisation of the layer is split up into domains by the demagnetising forces, or the layer is in the recorded state. In that case, shape anisotropy of a

single domain film loses its meaning and we will have to consider the demagnetising energy density (E_d) of a stripe-domain structure which is given by [100]. If the domain width (d) is smaller than the layer thickness (h) then we can write: $E_d = 0.136 \mu_0 d/h M_s^2$. The condition for stable perpendicular magnetisation ($K_1 > E_d$) expressed in Q becomes: $Q = 0.27 d/h$. If we consider a Co-Cr layer with values of $M_s = 400$ kA/m and $h = d = 1 \mu\text{m}$ we obtain $Q > 0.27$ or in this case $K_1 > 2.7 \cdot 10^4$ J/m. This is much less stringent than in the previous case where $Q > 1$ and $K_1 > 10^5$ J/m. This simple example shows that a substantial perpendicular magnetisation may be expected even if $Q < 1$.

Transferring the above-mentioned knowledge to the real recording (very small perpendicular bits) we know that the field near the transition is negligibly small and consequently the requirement for a high perpendicular anisotropy are, from that point of view, also not very severe.

4.2. MAGNETISATION

Frequently the relation between the magnetisation and composition is presented with the Slater-Pauling curve which gives the relation between the saturation magnetisation (M_s) and the homogeneous bulk Co-Cr [101,102]. In general most papers reported that the M_s of sputtered and evaporated films, deposited at higher substrate temperatures, is found to be larger than that for bulk alloys having the same average chemical composition. Although in the literature various origins have been proposed, the most likely explanation is the phase separation. Two hcp phases, which hardly occur in bulk Co-Cr material at low temperatures, cause compositional fluctuations along the grain boundaries as the film growth proceeds.

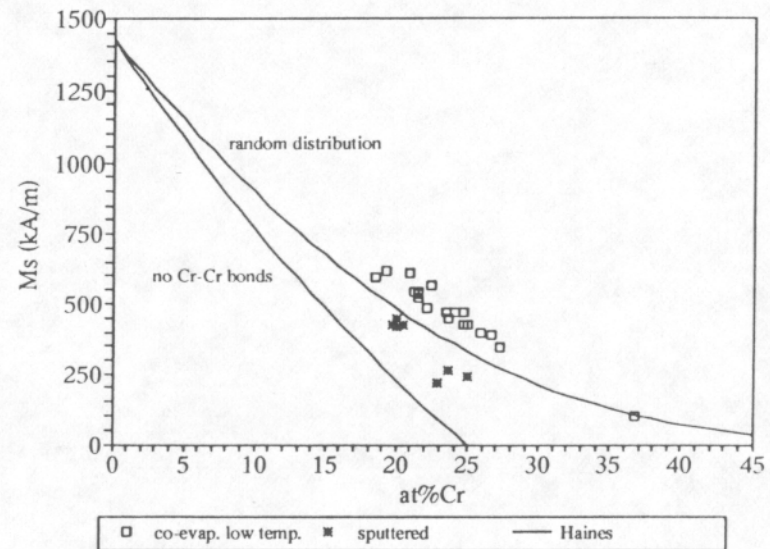


Figure 24. The theoretical and experimental relation, between M_s and the average Cr composition.

In fig. 24 we compared the theoretical data from Haines [103] with ours. In the random Co-Cr alloy (solid line "random distribution") the Cr atoms are not distributed in the most suitable way for reducing the Ms of the alloy. Therefore the maximum local content of Cr for this distribution is much higher than in the case where Cr-Cr bonds are not present. The "no Cr-Cr bonds" line in fig. 24 shows that the Ms becomes zero at 25 at% Cr based on the fact that for bulk material the measured Ms for this composition was zero. Consequently 4 Cr nearest neighbours in an hcp lattice make the final Ms zero. As a comparison we plotted a few values for sputtered Co-Cr samples and also from samples made by the low temperature co-evaporated films. During the latter the energy of the atoms is much lower than for sputtering and the so-called process-induced compositional separation occurs. From this it can be concluded that the Ms for deposited thin films is always higher than the theoretical line having no Cr-Cr bonds. This can be an indirect proof for compositional separation of the Cr in the Co for both types of deposition.

4.3. COERCIVITY.

The coercivity in ferromagnetic layers with perpendicular anisotropy is determined by two factors.

- The type of magnetisation process involved i.e. is the magnetisation reversed by (coherent) rotation or by domain-wall movement?
- Morphology of the layer i.e. shape and dimensions of the crystallites (columns), nature of the boundaries, surface and initial-layer properties.

In fact these two factors are strongly interrelated and their properties are strongly influenced by the deposition parameters such as temperature, P_{Ar} etc. Based on general knowledge about ferromagnetic films it can be concluded that the presence of stripe domains in Co-Cr must be quite common because the layer splits up into such a configuration by reducing its demagnetising energy. Theoretical support for reversed domains in uniaxial thin layers with perpendicular anisotropy is provided by [104,105]. Supporting arguments for the relation between coercivity and reversal mechanism obtained from research on permanent magnets, are given by [106].

In polycrystalline and/or columnar-like materials the critical domain size is difficult to estimate because it is influenced by the presence of neighbouring particles and their interactions. If the magnetization reversal takes place by coherent rotation the H_c must be equal to the H_k. In our case all the layers have H_c < H_k. An explanation for this effect can be made by introducing the so-called incoherent reversal modes such as fanning, curling and buckling. These can, in principle, only occur in layers in which the magnetic morphology consists of single domain particles. In the case of Co-Cr such a morphology can occur if the magnetic exchange interaction between the columns are absent. In the case of magnetically-separated columns an additional shape anisotropy can contribute to the total anisotropy. Higher coercivities in domainwall motion materials find their origin in hindering of the domainwall by grainboundaries, lattice faults, voids etc. Experimental work on high coercivity Co or Co-P layers [107-109] shows a variety of domain configurations independent of the source of the magnetic anisotropy.

The presence of a domain structure and the results of other measurements like hysteresis loss and H_c vs. the angle of the applied field, are not completely in line. This could perhaps be explained by the phenomena of interaction domains which were first introduced by [110]. Here particle exchange forces are negligible and the magnetic moments are mutually aligned by magnetostatic interactions [111].

The nucleation density is much lower for low H_c than for high H_c. It is clearly shown that nuclei stripe out at the tips of the domain after decreasing the applied field [34,112]. The

observable size of the dots (coming from saturation) in high H_c films is determined by the resolving power of the Kerr microscope. Nevertheless these dots do not expand very much due to the surrounding closeness of neighbouring dots. The reversal in one column could be, depending on its dimensions, initiated by a reversed domain. These small sizes cannot be resolved by Kerr microscopy and it can be accepted that such domains are no longer present in thinned TEM specimens. The particle behaviour without exchange coupling is suggested by Cr segregation at the column boundary. At present only preliminary results are available from Scanning Auger Microspectroscopy (SAM) and wet chemical etch procedures. From the latter we found only qualitatively a higher Cr content at the surface than in the bulk of all our samples. We came to the conclusion from a SAM measurement of a fractional surface of a high coercivity film that there is a Cr segregation at the column boundaries.

It can be concluded from the above that the coercivity (an extrinsic property) can only be determined by means of the macroscopical hysteresis loop in combination with the theory of micromagnetism [113]. Angle dependent H_c measurements are only valid over the whole angle area if they have been measured by a bi-axial VSM where the out-of-plane measurements of the H_c are corrected for the demagnetisation.

Knowledge about the microstructural properties of the material cannot be omitted. In the first place the size and shape of the material plays an important role. In SDP (single domain particle) the coercivity decreases with decreasing particle diameter and for an ideal material H_c becomes independent of the diameter until at a very small size (2 nm) the particle becomes super-paramagnetic and the reversal takes place by thermal activation which again leads to a lower H_c.

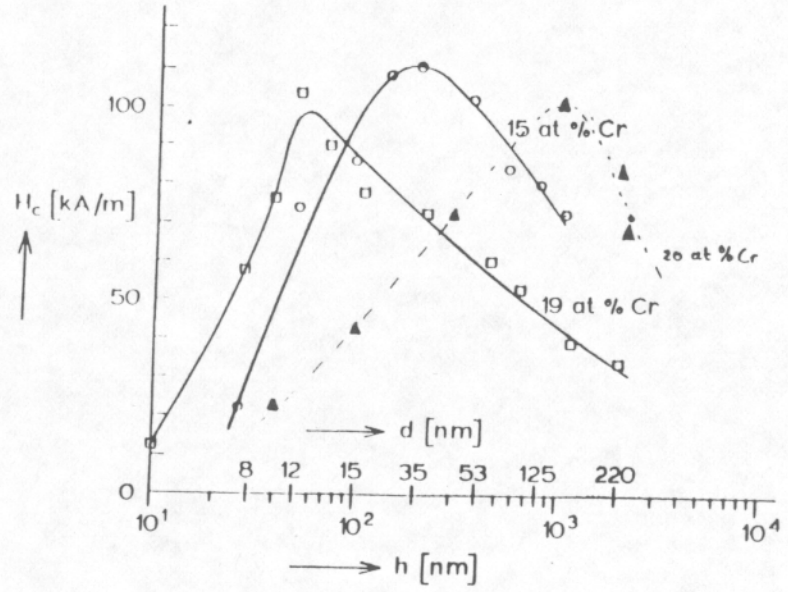


Figure 25. The coercivity as a function of the layer thickness for different compositions of Co-Cr films rf and magnetron sputtered.

The H_c for MDP (multi domain particle) is determined by pinning mechanisms of the domain wall. These mechanisms are determined by the magnetically inhomogeneous regions such as columnar boundaries, chemical inhomogeneities, stacking faults etc. The H_c has been measured as a function of the layer thickness for many different Co-Cr compositions and preparation methods [53,57,99]. As an example, in fig.25 the H_c vs. thickness is given for three different compositions. All exhibit a maximum H_c , but at different film thicknesses or columnar diameters as measured at the surface by SEM. Due to the difference in rf and magnetron sputtering a different behaviour can be seen between the two types of films. The differences in H_c depend on the average film composition [53] and the deposition method used [57]. However it is not easy to arrive at a conclusion because chemical composition, thicknesses, and deposition methods are all related to each other by diffusion and temperature effects etc.

It can be further concluded from this that the maximum H_c 's are remarkably smaller than the intrinsic H_k (for 15at%: $H_k=560$ kA/m and for 19 at% Cr: $H_k=320$ kA/m) and are about the same for both compositions namely about 100 kA/m. This can be qualitatively explained by domain-wall displacement for the reversal process [53]. It also contradicts the supposition that the H_c is related to rotational magnetisation. Thermal perturbations can also be explained as discussed in [33].

As a consequence of the difference between volume and surface H_c [57,99] the magnetisation cannot be homogeneous throughout the film during all stages of the hysteresis curve. This was proved by study of the chemical composition of our films using Auger and XPS depth profiling analysis [97]. This and the many stacking faults found by TEM can also explain the hindering of the domain-wall motion by an increase of the H_c .

4.5. MAGNETISATION REVERSAL PROCESS

The reversal mechanism of magnetisation and, associated with it, the domain structure in Co-Cr layers have still not been completely clarified. There are two main models, namely the particulate and the continuous model. In the first one the Co-Cr 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 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 [53]. Several methods have been used for studying the magnetisation process by experiments based on more microscopical observations like Magneto-optic Kerr microscopy [34,35,112]; Bitter technique [114]; Colloid-SEM method [e.g.115]; Neutron depolarisation [116-118]; Lorentz microscopy [24]; Magnetic Force Microscopy [27], micromagnetic simulations [e.g.28,29,30] but also with the more macroscopical approach using the VSM loop by studying the initial slope [53,33,119] and angle dependence of the applied field (H_a) [16] and changing the magnitude and direction of H_a [120,121].

Studying the intrinsic domain configuration can contribute to a comprehension of the magnetisation reversal. The magnetic properties of Co-Cr layers are strongly influenced by variations in microstructure, morphology and chemical homogeneity. Initial layer effects, Cr segregations at the column boundaries and surface are very important for the final behaviour of the domains. The morphology also depends on the sputtering method and parameters used (mainly substrate temperature and Ar gas pressure). From the perpendicular (easy axis) hysteresis loop, measured with the VSM we have, in some cases, observed a typical shoulder which indicated bubble domains which can stripe out over a long distance at a certain field. In order to understand more about the micromagnetics of such layers one has to correlate the morphology,

magnetic parameters and magnetic structures. Lorentz microscopy was studied for observation of the intrinsic magnetic-domain structure [24]. Different domain configurations with in-plane magnetisation were observed in thin Co-Cr layers. This can be explained by variations in the deposition methods and consequently differences in morphology, structure and chemical homogeneity. None of the samples used had a well-oriented hcp-axis. In the case of a good textured layer (Co₇₇Cr₂₃), having a thickness of 50nm, stripe domains with a width of 0.1 μm were observed [24,Grundy]. A maze-domain pattern, known from bubble-like materials, was observed in films of 0.1 μm thickness having a composition of Co₈₂Cr₁₈ [24, Hoffmann]. Very thin layers (30nm) with in-plane magnetisation show the so-called ripple structure in Co₉₀Cr₁₀ [18] and Co₇₉Cr₂₁ [24,Hoffman]. Different domain forms were observed in films with much better orientation and increased thickness (0.1 μm) [24]. A magnetic black-and-white dot structure with a distance between the dots of the same order as the columnar diameter has been shown by [24,Ohkoshi]. In this case the film morphology (one column consists of several crystallites) is totally different from the equiaxial crystal structure present in [24, Grundy] which shows a stripe-domain width in the order of the crystal size. Co-Cr films in the range between 17-30at%Cr with a Cr segregation into the column boundaries show dot-like Lorentz images which have the same size as the column diameter [24, Hoffmann]. Stripe-domain widths (<0.1 μm) having the same size as the column diameter are also observed in [24, Ouchi]. It is also shown that from relatively thick films (0.3 μm), the domain width is the same as the column size [24,Grundy] and it is suggested that the domain structure consists of a chain of crystals.

Recently, [24,Tsukahara] after an extensive study of single (Co-Cr) and double-layer medium (Co-Cr/Ni-Fe), it was concluded that magnetic images of Co-Cr observed by Lorentz microscopy could neither be explained by the particulate model nor by the domain-wall motion model. Magnetisation varied gradually from the up to the down direction and appeared as bubble-stripe and maze shapes in all the films examined. The stripe-type images show that there is ferromagnetic exchange coupling between the grains.

Based on neutron depolarisation experiments and magneto optic measurements of the surface of our Co-Cr films it can be concluded that the domain width is proportional to h , where x is about 0.8 for RF magnetron sputtered films [116-118].

Cross-sections of our films were studied with the SEM mode of the TEM and the average columnar diameter was determined from various samples having different coercivities. The parameter H_c/H_k was used as a classification parameter. The columnar-like morphology can be clearly seen in all the films independent of the H_c/H_k value. In combination with the domain studies using Kerr microscopy we came to the conclusion that one domain consists of 4-8 columns, for these films. This is in agreement with the observations by Tsukahara [122].

5. Micromagnetic calculations and simulations

Simulations of Co-Cr films having a continuous character were in good agreement with experimental data [29]. Particulate Co-Cr samples, having a homogeneous magnetisation inside the particle, showed strong dependence of the H_c on the exchange-coupling between the columns [28]. Recently we published the first results [30] on simulation studies of clusters of 9 and 25 magneto-statically coupled Co-Cr particles. The H_c is found to be much lower than the switching field of an isolated particle and furthermore the switching order leads to the conclusion that the magnetostatic interaction between neighbour facilitates reversal. The H_c in our calculations is still rather high, introduction of exchange-coupling should reduce it as was found by [28].