DOMAINS IN CoCr INVESTIGATED BY NEUTRON DEPOLARIZATION.

W.H. Kraan and M.Th. Rekveldt
Interuniversitair Reactor Instituut, Mekelweg 15, 2629JB Delft, The Netherlands.

K. Hemmes and J.C. Lodder
Twente University of Technology, P.O. Box 217, 7500AE Enschede, The Netherlands.

Abstract. Polarized neutrons (λ = 0.47 nm) are transmitted through RF and magnetron sputtered CoCr films (0.3-5 μm thick) with the polarization vector in the plane or perpendicular to the plane of the film. In the former case we can deduce from the depolarization the effective height h_{eff} of the domains and from the angular dependence of the depolarization the section width δ (which is proportional to the domain width) in the remanent states after perpendicular and after in-plane saturation.
As expected, h_{eff} appears to be larger after perpendicular saturation and for a film thickness h ~ 400 nm, h_{eff} approaches h. This is attributed to the disappearance of reversed spike domains in the thinnest films. The lower h_{eff} found in magnetron films with a lower surface/bulk coercivity ratio is also consistent with spike domain theory. The section width δ is found to be proportional to h^x with x depending on the preparation or magnetic history of the film between 0.6 and 0.8. For magnetron films δ is ~ 1.5 as large as in RF films of equal thickness, in qualitative agreement only with the fact that K_1 is twice as large as for RF films.

INTRODUCTION.

For CoCr sputtered films with their typical columnar morphology discussion remains about the particle vs continuous magnetic behaviour. It is well known that the morphology and crystal structure and therefore the magnetic properties of these films depend strongly on the deposition conditions [1]. In order to understand the magnetization process, domain studies are of great importance. For such studies Neutron Depolarization (ND) is a unique method, especially in studying the bulk of thick films (> 500 nm), complementary to Lorentz Microscopy which can only be applied to thin films (< 200 nm). Most other techniques are confined to the surface.
When a polarized neutron beam passes through a ferromagnetic specimen, the polarization vector precesses around the magnetic induction due to the magnetization M_s in the magnetic domains. In ND the polarization vector is adjusted by means of a "polarization turner" along one of the (x,y,z) directions of the laboratory system. By means of a second polarization turner after transmission the component of the polarization vector – as it emerges out of the sample – along any one of these directions can be analysed. Thus, a (3x3) depolarization matrix can be measured [2]. In this paper we only consider the diagonal elements D_{xx}, D_{yy}, D_{zz}.

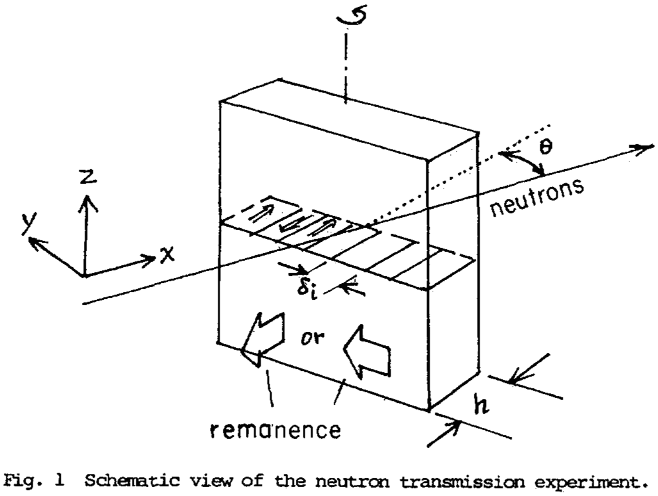


Fig. 1 Schematic view of the neutron transmission experiment.

Let us illustrate the relevance of ND with the model of Fig. 1 in which the domains extend through the thickness or "height" h of the film, defining (x,y,z) as indicated.
In perpendicular transmission the precession angle is + or $-\phi_m$, with

$$\phi_m = 5.72 \times 10^8 (M_s.h).\lambda \qquad (1)$$

where M_s is in A/m and the neutron wavelength λ and h are in meter. In non-perpendicular transmission, neutron trajectories passing a domain boundary occur, so the precession angle along them is smaller than ϕ_m, hence $\cos\phi$ and also $\langle\cos\phi\rangle$ is closer to 1. In an earlier analogous study [3] on thick electrodeposited Ni films we stated that $D_{zz} = \langle\cos\phi\rangle$ for all transmission angles θ. In case of domains of equal width we can deduce from [3], eq. (14) the following linear relationship between $\langle\cos\phi\rangle$ and $|\theta|$:

$$D_{zz} = \langle\cos\phi\rangle = \cos\phi_m + |\theta| f(\phi_m), \quad (0 \le \theta \le \arctan(\delta/h))$$

with

$$f(\phi_m) = \frac{h}{\delta}\left(\frac{\sin\phi_m}{\phi_m} - \cos\phi_m\right) \qquad (2)$$

From eq. (1) and (2) we obtain the following information: D_{zz} ($\theta = 0$) yields the height of the domains and the quantity $f(\phi_m)$; then, from the slopes of the measured D_{zz} for small $|\theta|$ we can determine δ.
When the domains have unequal widths, it is obvious that eq. (2) remains valid in good approximately provided $|\theta|$ is so small that contributions due to neutron trajectories passing 2,3,... domain walls are negligible. Then the quantity δ is defined by the formula $1/\delta = \langle 1/\delta_i \rangle$, with δ_i the domain section width as indicated in Fig. 1.
We realize that because of the "spaghetti" or "chain of column" like domain structure [4,5], the quantity δ may be larger than the actual domain width. We will refer to δ as "domain section width". This paper deals mainly with the quantities δ and h_{eff} for RF and magnetron sputtered CoCr films in the remanent states after perpendicular and after in-plane saturation.

EXPERIMENTAL.

The samples were sputtered from alloyed $Co_{81}Cr_{19}$ targets of 4" and 3" diameter respectively, on Si(100) substrates in a Leybold Hereaus RF-sputter apparatus (Z400) either without (so called 'RF-films') or with magnetron facilities (so called 'magnetron films'). Films were sputtered in batches of 16 (RF) or 23 (magnetron) on substrates with dimensions 10x10 mm^2. Each film was characterized by a VSM measurement yielding the volume coercivity H_{cv} and the product $M_s h$. Surface magnetization measurements on a set-up in which the polar Kerr rotation is determined as a function of a perpendicular applied field yield the surface coercivity H_{cs} [6].
The (magnetic) film thickness h is determined from the product $M_s.h$ and M_s = 460 kA/m.
Since sputtering conditions are less homogeneous in magnetron sputtering than in the RF mode, the former samples were divided into "centre" and "off centre" samples as described in [7]. Series B and D of the "centre" type have a higher CR = H_{cs}/H_{cv} and a higher thickness than series A and C belonging to the "off centre" type.

0018-9464/87/0100-0065$01.00© 1987 IEEE

All series consists of 7 films.
From torque measurements the value of K_1 was found to be 0.9 and $1.9\times10^5 J/m$ for RF and magnetron sputtered films, respectively.
The precession of the polarization vector to be expected on base of eq. (1) in a CoCr film with a thickness of 1 μm is only 7o, and hence $\cos\phi_m > 0.99$.
To make the difference of D_{ii} (i = x,y,z) from 1 better observable, we performed the ND experiments on stacks of 7, 12 or 14 films rather than on a single film, so in fact we measured $(D_{ii})^N$ (N = 7, 12, 14). Thus we were able to study films with thicknesses from 4,84 down to 0.31 μm. Table 1 gives a review of the films used in this study.

Table 1 review of CoCr films used.

A. RF sputtered films

ser. numb.	M_s h (A)	h (μm)	CR	numb. of films	ϕ_m (degr)
860325	0.144	0.31	0.4	14	2.22
031285	0.242	0.52	0.3	14	3.7
090384	0.421	0.95	0.3	14	6.48
080384	0.855	1.90	0.4	12	13.2
860314	2.22	4.84	0.7	14	34.2

B. Magnetron sputtered films

ser. numb.	M_s h (A)	h (μm)	CR	numb. of films	ϕ_m (degr)
851118	0.254	0.55	1.10	14	3.91
850911-A	0.719	1.56	0.53	7	11.7
850911-B	0.796	1.73	0.91	7	12.3
850912-C	0.799	1.74	0.44	7	12.3
850912-D	1.111	2.42	0.87	7	17.1

To measure the quantities D_{xx}, D_{yy} and D_{zz} as a function of θ (Fig. 2), we rotated the sample stepwise and measured in each setting the intensities denoted I_{xx}, I_{yy} and I_{zz}, respectively. The corresponding matrix elements are calculated by $D_{ii} = (I_s - I_{ii})/I_sP$ (i = x,y,z) with P (= 0.94) the polarizing power of the set-up and I_s the intensity of the fully depolarized beam.
Films were measured in the remanent states after applying an external field larger than 7 kA/cm (8 kOe) either perpendicular or parallel to the plane of the film. In the latter case the sample was mounted such that the remanence was perpendicular to the axis of rotation (Fig. 1).

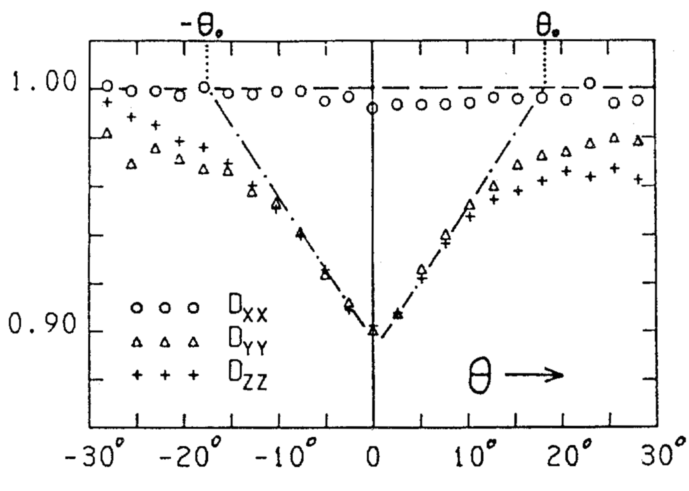


Fig. 2 Behaviour of the diagonal elements in sample 080384 after in-plane saturation as a function of transmission angle.

RESULTS.

(i) Domain height.
From D_{xx} 1 (Fig. 2) in all experimental results follows that the domain magnetization is perpendicular to the plane of the films [3].
The actual precession of the polarization vector is manifest only in D_{yy} and D_{zz} and appears to be less than the value calculated with eq. (1). Therefore we introduce the quantity h_{eff} defined by

$$h_{eff}/h = \arccos(\sqrt[N]{D}\,)/\,\phi_m \qquad (3)$$

with D the average of D_{yy} and D_{zz} for $\theta = 0$ and N the number of stacked films. The quotient h_{eff}/h is plotted in Fig. 3a and b as a function of h in both magnetic states for RF and magnetron sputtered films, respectively.

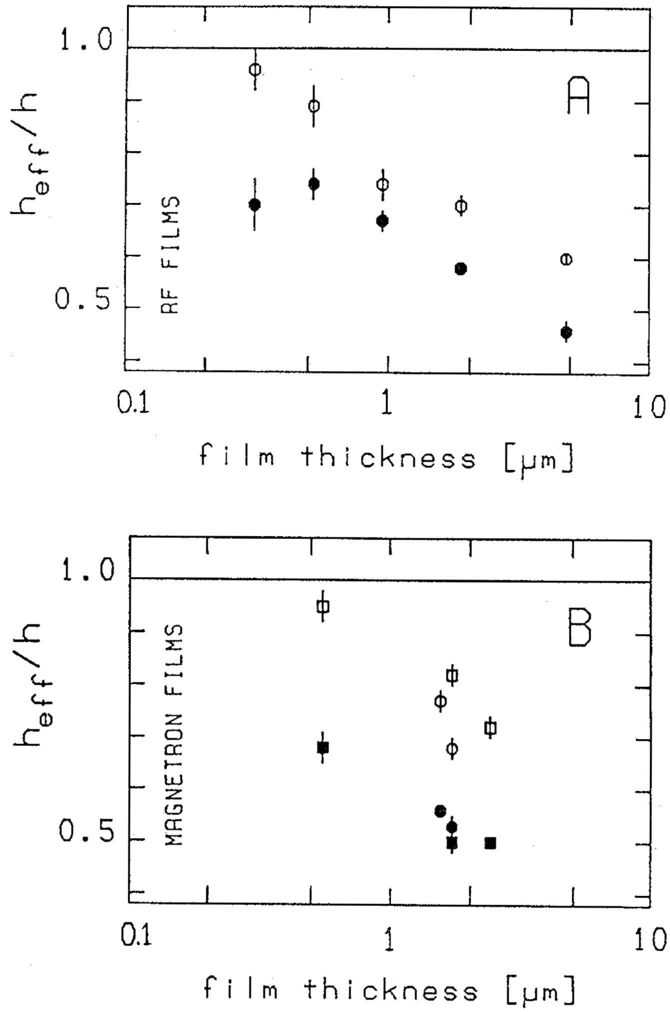


Fig. 3 Effective height measured in RF (a) and magnetron films (b) as a function of film thickness after perpendicular (open symbols) and in-plane (black symbols) saturation.
In (b) square and circular symbols correspond to films with high and low CR, respectively.

The behaviour of RF and magnetron sputtered films is qualitatively similar. In the remanent state after perpendicular saturation h_{eff} appears to approach h in thin films. In the in-plane remanent state h_{eff}/h also increases upon decreasing film thickness, but tends to remain well below 1. The low CR of the magnetron series A and C is found to correlated with a lower h_{eff} compared with series B and D with a CR close to 1 (Fig. 3b, table 1).

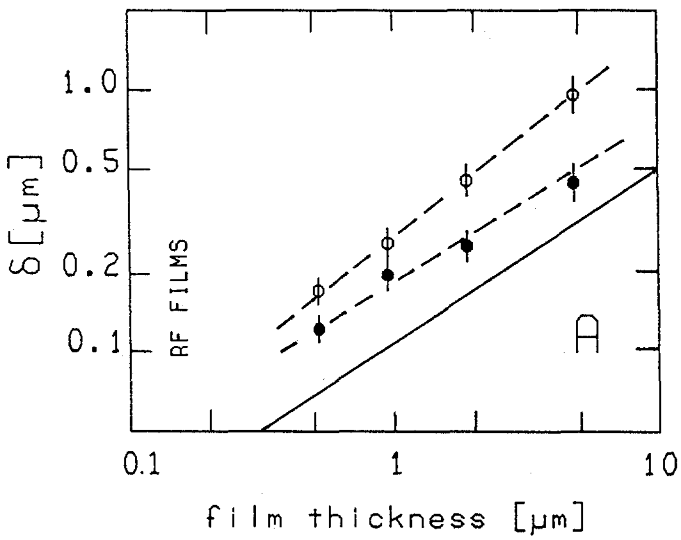
(ii) Domain section width.

Using the plots of D_{yy} and D_{zz}, δ is determined from the angle θ_o (Fig. 2). From eq. (2) we can deduce that

$$\delta = \frac{2}{3} \theta_o . h \qquad (4)$$

Setting h equal to h_{eff} in eq. (4) yields the behaviour plotted in Figs. 4a and b for δ as a function of thickness in RF and in magnetron sputtered films, respectively. We notice that δ is ~ 1.7 times as large after perpendicular saturation as after in-plane saturation. Moreover, for magnetron films δ is about 1.6 times as large as for RF-films. In the RF film of 0.31 μm hardly any dependence of D_{yy} and D_{zz} on θ was observed, so we can give no reliable value for δ .

In all plots δ can be described as ~ h^x, with for RF films x = 0.6 and 0.8 ($\pm$0.05) after in-plane and perpendicular saturation, respectively and for magnetron films x = 0.7 and 0.8 ($\pm$0.05) respectively.



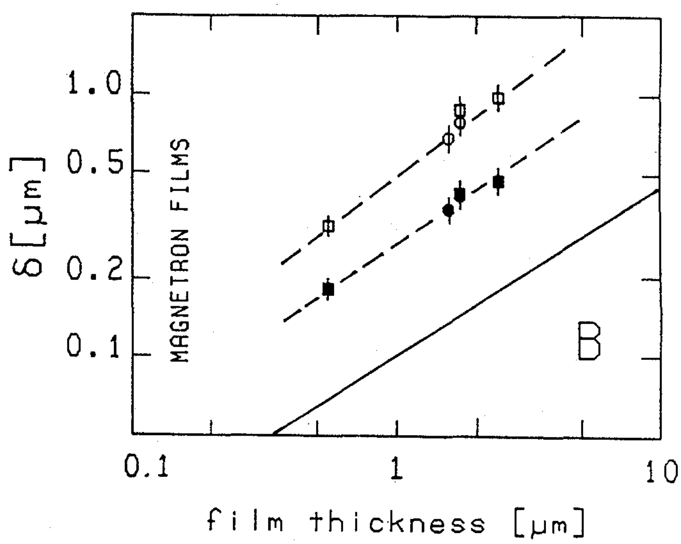


Fig. 4 Domain section width δ measured in RF (a) and magnetron films (b) as a function of film thickness. Symbols as in Fig. 3. The broken lines are a best fit to the observations; the full lines correspond to the domain width d according to "reversed spike theory". By the "curvature" of the domains d should be ~ δ/2.

DISCUSSION.

(i) Domain height, h_{eff}.

In an earlier [6] and in a parallel [7] study on CoCr films it is reported that the quantity CR in RF films abruptly decreases from 1.3 to 0.5 above a critical film thickness of 125 nm. The critical thickness for magnetron films was found to be 1.1 and 0.8 μm for centre and off-centre samples, respectively. The increase in CR is explained by the vanishing of reversed domains or spikes from the surface in films thinner than the critical thickness. An order of magnitude estimate [7] based on theoretical calculations of flux closure in uniaxial systems by "reversed spike domains" i.e. "branching" of the main domains [8,9,10] shows that spike domains in CoCr are expected to disappear at a films thickness near this critical value.

Also the present observation that h_{eff}/h after perpendicular saturation approaches 1 at a thickness near this value is consistent with spike domain theory. In this theory it is moreover expected that the magnetron series A and C with a "low" CR ("many spikes") should have a lower h_{eff} than the series B and D with a "high" CR value. This is indeed found after perpendicular saturation, as can be seen in Fig. 3b.

(ii) Domain section width δ and domain width d.

A sharp knife cross-section through a serpentine domain structure reveals the section widht distribution. Its average is larger than the shortest perpendicular "cross over" of a domain by a factor of ~2, depending on the "curvature" of the serpentines. We assume that δ is proportional to the domain width d. In continuous domain models with free poles or closure domains, e.g. the Kittel model [11] the dependence of d on h is described with a power 1/2: d = 0.19 $h^{1/2}$ and 0.21 $h^{1/2}$ in RF and magnetron films respectively. In spike domain theory one finds d = 0.11 $h^{2/3}$ and 0.10 $h^{2/3}$, respectively (d and h in μm). The present observations that the exponent of h has values between 0.6 and 0.8 is in favor of the spike domain model and in agreement with measurements by Gemperle and coworkers [12] on Co platelets.

The Kittel model predicts a weak dependence of d on K_1. However, we observe that δ in magnetron films is 1.6 times as much as in RF films, whereas K_1 differs by a factor of 2.

So this model does also not describe the observed K_1 dependence of δ . It is concluded from this and other observations that RF and magnetron films differ in more respects (e.g. internal stress and coercivity [7]) than merily the quantity K_1.

The difference of h_{eff} and δ after perpendicular and after in-plane saturation, found in all our films is probably correlated with domain nucleation, which proceeds in different ways after both saturated states. It proves that the columnar morphology is not the only factor defining the domain structure, but that also the magnetic history is important.

CONCLUSION.

We have studied the effective height and the domain section width in sputtered CoCr films. In the same film both quantities are different after in-plane or after perpendicular saturation. The increase of h_{eff} with decreasing film thickness is explained by the vanishing of "reversed spike domains" below a critical thickness. These phenomena and the 2/3 power dependence of the domain width on film thickness are more consistent with the spike model than with the Kittel model for a continuous medium. The lower h_{eff} found in magnetron films with a lower surface/bulk coercivity ratio is also consistent with spike domain theory.

REFERENCES.

[1] e.g. P.J. Grundy and M. Ali, J.M.M.M. 40, p154, (1983)
[2] M.Th. Rekveldt, J.F. van Schaik and W.H. Kraan, Nukleonika 24, p809, (1979)
[3] W.H. Kraan and M.Th. Rekveldt, J.M.M.M. 5, p247, (1977)
[4] This conf., contr. by J.C. Lodder and A. Hubert
[5] W.H. Kraan, M.Th. Rekveldt, K. Hemmes and J.C. Lodder, Physica 136B, p440, (1986)
[6] K. Hemmes et al., J. Phys. D:Appl.Phys.19, p1311, (1986)
[7] This conf., contr. by K. Hemmes et al.
[8] I. Privorotskii, "Thermodyn. Theory of Domain Structures", J. Wiley & Sons, New York-Toronto,(1976)
[9] A. Hubert, IEEE Trans. on Magn. MAG-21, p1604, (1985)
[10] W.A. Barker and G.A. Gehring, J. Phys. C 19, p259, (1986)
[11] C. Kittel, Phys. Rev. 70, p965, (1946)
[12] R. Gemperle, A. Gemperle and I. Bursuc, Phys. Stat. Sol 3, p2101, (1963)