SANS EXPERIMENTS ON THIN CoCr FILMS 
WITH PERPENDICULAR DOMAIN STRUCTURE

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The application of small angle neutron scattering in the study of thin magnetic layers used in perpendicular recording media has been demonstrated. The domain structure in these materials is characterized by the domain height perpendicular to the film and the domain width in the plane of the film. These quantities have been deduced from the two dimensional scattering pattern. The domain height is found from the momentum dependence of the anisotropy of the scattering where the film normal makes an angle of \( \theta \) degrees with the beam direction. The domain width in the plane of the film has been analysed from the scattering profile itself. In principle also angular variations of the domain orientation may be analysed from the momentum dependence of the anisotropy.

1. Introduction

Deposited films of CoCr on several substrates have been studied for applications in perpendicular recording. The magnetic domain structure in this kind of thin layers consists of domains in the bulk with their magnetization directed perpendicular to the plane of the film and an initial layer where the domain structure is unknown. The bulk domains are at first characterised by their height \( t \) perpendicular to the film and secondly by the domain width \( \delta \) in the plane of the film (fig. 1). The precise domain structure and the magnetization reversal mechanism in these films are still unsolved problems and depend very strongly on the morphology of the layers. The most common techniques used to study the films are summarized below.

Magnetization measurements yield the coercive field and the hysteresis curve from which information about the average domain magnetization direction can be obtained. Transmission electron microscopy (TEM) [1] enables one to obtain a photographic picture of the surface and the cross section of the sample, showing clearly a crystalline columnar structure of the film. The magneto-optic Kerr effect [2,3] yields both the magnetic domain structure and the magnetization hysteresis at the surface. The resolution of this method for measuring the domain size in the plane of the film is about 0.2 \( \mu \)m. More recently the neutron depolarization technique (NDT) [4,5] has been introduced which enables one to determine the domain height together with the domain width in two orthogonal directions. The limitations of this last method are in the film thicknesses which can be studied, unless a large number of films in one packet is used. Domain sizes in the plane of the film are de-
M. Th. Rekveldt et al. / SANS experiments on thin CoCr films

111

In this paper the application of small angle neutron scattering (SANS) will be discussed using samples with different magnetic history as a demonstration of the possibilities of this method. The next sections deal with a description of the experiments, a theoretical model to describe the results, the measuring results and a discussion.

2. Description of the experiment

For the experiments use is made of the small angle scattering instrument D11 of the ILL in Grenoble which enables one to use a neutron beam with angular divergence $\Delta \theta$ of 0.5 mrad and a neutron wavelength of 1.2 nm, resulting in a momentum resolution $\Delta Q$ of $2\pi\Delta \theta/\lambda = 2.5 \times 10^{-5}$ nm$^{-1}$. The samples are successively positioned in the neutron beam according to fig. 2. The cross section of the neutron beam was confined to a circular opening of 8 mm diameter for all of the samples except for the larger sample no. 1 which was measured with a square diaphragm of 1 cm$^2$. The detector was positioned at a distance of 35 m behind the sample. The spatial resolution on the detector was 1 cm in both directions. The samples were rotated over various angles $\theta$ around the vertical $z$-direction. At each angle $\theta$ a two dimensional spectrum was measured. Table 1 summarizes the total number of experiments. The measured intensities corrected for background scattering were normalized for the different sample diaphragms used. As a result the intensity profiles from the different samples could be compared also in absolute sense.

Table 1

<table>
<thead>
<tr>
<th>Sample</th>
<th>Layer thickness $t_0$ (µm)</th>
<th>Magnetic history</th>
<th>Angular settings (°)</th>
<th>Domain height $t$ (µm)</th>
<th>Domain width $\delta$ (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
<td>⊥ magn.</td>
<td>0,5,7,10</td>
<td>1.2</td>
<td>0.25</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>⊥ demagn.</td>
<td>0,10</td>
<td>1.6</td>
<td>0.20</td>
</tr>
<tr>
<td>3</td>
<td>2</td>
<td>⊥ magn.</td>
<td>0,10</td>
<td>1.2</td>
<td>0.27</td>
</tr>
<tr>
<td>4</td>
<td>2</td>
<td></td>
<td></td>
<td>magn.</td>
<td>0,2,4,8,10,15</td>
</tr>
<tr>
<td>5</td>
<td>2</td>
<td>as sputtered</td>
<td>0,10</td>
<td>1.1</td>
<td>0.12</td>
</tr>
</tbody>
</table>

The Co–Cr films of 2 µm thickness are deposited on a Si single crystal substrate of 0.3 mm thickness. A sketch of a possible magnetic domain structure of these films is shown in fig. 1, where characteristic parameters as domain height $t$, domain width $\delta$ and orientation variation $\alpha$ are defined. Sample 1, a disk of 5 cm diameter, was produced separately from the other samples which have been manufactured in successive series under the same conditions and of equal size of 1 cm$^2$. Table 1 summarizes the specific properties of the samples investigated, the magnetic history and the angular settings at which intensity profiles were collected. Samples 1 and 3 were magnetized perpendicularly, sample 2 was demagnetized perpendicularly with an ac field, sample 4 magnetized parallel to the plane and the $y$ direction of the reference system in the neutron scattering machine (see fig. 1). The as-sputtered sample 5 has never been exposed to a magnetic field.

3. Model of neutron scattering from a perpendicular domain structure. Angular dependence

The initial purpose of the experiments was to look for total reflection of neutrons on the domain...
wells which are nearly perpendicular to the film. However, considering the typical length of the walls along a neutron path of only 2 μm, it appears that the phase change difference between two neutron paths in neighbouring domains is only of the order of 10⁻², which is too small to expect a considerable contribution of total reflection and refraction in the domain walls [6]. On the other hand, it can be shown that the total scattering amplitude per domain is very small which makes the Born approximation applicable. This is supported by the half width of the scattering profile $ΔQ = 10^{-2} \, \text{nm}^{-1}$ which roughly agrees with the expected domain width of about 0.5 μm expected from normal scattering theory. So we will continue to find an expression for the scattered neutron intensity based on the master equation for magnetic elastic scattering of an unpolarized neutron beam [7],

The integral is split into an integral over the domain height $t$ and over the plane $s$ of the film probed by the vector $R$. By rotating the film and the reference system over an angle $θ$ around the $z$-axis and remembering that in SANS the momentum transfer is always perpendicular to the initial neutron beam, it follows that $Q_z = Q_x \tan θ$ and $μ_p(R)$ is given by

$$μ_p(R) = (1 - k_{γ}^2 \sin^2 θ)^{1/2}.$$  

The integral develops quite easily into,

$$\frac{dσ}{dΩ} = \frac{\sin^2 Q_x t/2}{(Q_x t/2)^2} (1 - k_{γ}^2 \sin^2 θ) \times (Nt)|s(Q_x)|^2,$$

and the anisotropy is given by the ratio between the scattering in the $y$ and $z$ directions,

$$A = \frac{\sin^2 Q_x t/2}{(Q_x t/2)^2} (1 - k_{γ}^2 \sin^2 θ) \frac{S(Q_y)}{S(Q_z)}.$$  

Here $S(Q)$ represents the scattering function describing the structure of domain widths in the plane of the film and $p_0$ the magnetic coherent scattering amplitude of each atom. By measuring the scattering along the $y$ and $z$ direction, the measured anisotropy in this scattering is directly proportional to the first two factors in eq. (5) and therefore this anisotropy enables one to determine the magnetic domain height in a direct way. Possible anisotropy in $S(Q)$ is eliminated for this purpose by measuring also at $θ = 0°$ where $Q_x$ vanishes.

3.1. Domain height

For the evaluation of eq. (1) we use Cartesian axes $(x, y, z)$ fixed to the film according to figs. 1 and 2. Considering only the scattering of the perpendicular domains, $p(r)$ is a constant along the $x$-direction and eq. (1) can be rewritten as,

$$\frac{dσ}{dΩ} = \frac{\sin^2 Q_x t/2}{(Q_x t/2)^2} (1 - k_{γ}^2 \sin^2 θ) \times (Nt)|s(Q_x)|^2,$$

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Variation in the orientation $α$ of the perpendicular domains can be taken into account by averaging eq. (5) over the angular interval $α$. It is clear that this averaging affects the anisotropy at different $θ$ values in a different way. This is shown in fig. 3 where at an angular spread of $α = 7°$ the anisotropy has been plotted for $θ = 5°, 10°$ and
Fig. 3. Theoretical anisotropy as a function of $Q_x$, calculated with a domain height $t = 1.12$ μm at various $\theta$ and $\alpha$ values as indicated in the figure.

$15^\circ$ and at $\alpha = 0^\circ$ and $\theta = 5^\circ$, using a domain height of $t = 1.1$ μm.

Variations in the domain height can be taken into account in the same way. However it seems not possible to deduce the interval $\alpha$ and the $t$ variation separately from the experimental results.

### 3.2. Domain width structure of the film

The structure of the domain widths is given by $p(R)$ or $S(Q)$ in eq. (4). To interpret the measured data in terms of physical quantities, we will try to find a mathematical expression for $p(R)$ in terms of well defined parameters. After transformation of this $p(R)$ to $S(Q)$ the result will be compared with the measurements to find the optimal values of the parameters used in $p(R)$.

In first approximation the perpendicular domains can be described as a stripe domain structure with a width $\delta$ and a random orientation of the stripes in the $yz$ plane. The magnetic scattering amplitude $p(R)$ introduced in eq. (1) and which has alternating values in crossing the stripes, is considered to be a periodic function and has been approximated by averaging the first Fourier term $\cos(Q_0 R)$ of this periodic function with $Q_0 = 2\pi/\delta$ between a lower and an upper limit $Q_1$ and $Q_2$, which delivers,

$$p(R) = \frac{\sin Q_1 R - \sin Q_2 R}{(Q_1 - Q_2)R} \exp(-cR).$$

with $Q_1 = +Q_1$, $Q_2 = -Q_1$, $Q(3) = -Q_2$, $Q(4) = +Q_2$, and $i =$ imaginary number. The function $S(Q)$ is similar to a distribution of Lorentz curves with a mean maximum value at $Q = 2\pi/\delta$. The width of the total curve depends on the damping factor $c$ in eqs. (6) and (7).

Here the factor $\exp(-cR)$ with $c$ a damping constant has been added to account for the nonideal periodicity of the domain structure in any direction. Substituting eq. (6) in eq. (4a) the scattering function $S(Q)$ from eq. (4b) can be calculated and appears to be,

$$S(Q) = \frac{4}{\pi} \frac{1}{\delta} \sum_{j=1}^{4} \int_{0}^{\pi/2} d\phi \frac{(-1)^j}{c + i(Q \cos \phi - Q(j))}.$$

with $Q(1) = +Q_1$, $Q(2) = -Q_1$, $Q(3) = -Q_2$, $Q(4) = +Q_2$, and $i =$ imaginary number. The function $S(Q)$ is similar to a distribution of Lorentz curves with a mean maximum value at $Q = 2\pi/\delta$. The width of the total curve depends on the damping factor $c$ in eqs. (6) and (7).

Fig. 4 shows the results of the calculated $S(Q)$ with $Q_1 = 10$ μm$^{-1}$, $Q_2 = 15$ μm$^{-1}$, for various values of the parameter $c$, which may be compared with the experimental results. The quantities $Q_1$ and $Q_2$ are chosen more or less arbitrary to fit with the experimental data.

It should be realized that also the nonperpendicular domains present in the initial layer on the substrate may influence the $Q$ dependence of

Fig. 4. Simulation of $S_2(Q)$ using the parameters $Q_0 = 10^{-2}$ nm$^{-1}$, $Q_0 = 1.5 \times 10^{-2}$ nm$^{-1}$ and values of the damping constant $c$ as indicated in the figure.
the scattering. In the discussion we will return to this point only qualitatively.

4. Measuring results

Fig. 5 gives iso-intensity plots for sample 1 ($\theta = 0^\circ$, $5^\circ$ and $10^\circ$), sample 4 ($\theta = 0^\circ$) and of sample 5 ($\theta = 0^\circ$ and $10^\circ$). These plots indicate very clearly the presence of anisotropic scattering with increasing angle $\theta$ in all samples and also at $\theta = 0$ in sample 4. As qualitative conclusions are drawn from these figures only, the levels of the lines are of no importance and have been omitted.

The difference in anisotropy of the samples is striking and has been presented for the different $\theta$ values as a function of the momentum transfer $Q_x$ in fig. 6. For this purpose the measured data were analysed with the program ANCOS2 belonging to the D11 software packed, which splits the measured spectra according to

$$ S(Q) = S_1(Q) + B(Q) \cos^2 \phi $$

Fig. 6. Anisotropy of the scattering in sample 1 at various angles $\theta$ as indicated divided by the anisotropy at $0^\circ$ as a function of $Q_x$. For samples 2, 3 and 5 only the anisotropy at $\theta = 10^\circ$ is given. The calculated anisotropy fitted with $t$-values as given in table 1 is given as a solid line in the figures.
with
\[ \phi = \tan^{-1}(Q_x/Q_y) \]
and which describes the anisotropy of eq. (5) reasonably well. In eq. (8), \( S_1(Q) \) represents the symmetric part and \( B(Q) \) the anisotropic part of the scattering, respectively. The anisotropy factor \( A \), as used in the figures can be written as,
\[ A = \frac{(S_1(Q_x) + B(Q_y))/S_1(Q_y)}{Q_y}. \] (9)

Together with the experimental results the theoretical anisotropy of eq. (5) has been added in the figures with an adapted value of the domain height and without spread in the orientation \( \theta \) of the perpendicular domains. Fig. 7 shows the anisotropy in sample 4 for a number of \( \theta \) values together with a theoretical fit. The anisotropy in sample 4 already present in the \( \theta = 0^\circ \) measurement has been presented separately in fig. 8 together with the isotropic and anisotropic scattering. The symmetric scattering \( S_1(Q) \) of the other samples has been presented in fig. 9.

5. Discussion

5.1. Anisotropy

As shown in the previous section the anisotropy in the scattering at \( \theta \) unequal zero yields the

![Fig. 7. Anisotropy of the scattering in sample 4 at various \( \theta \) values divided by the anisotropy at \( \theta = 0^\circ \) as a function of \( Q_x \). The calculated anisotropy fitted with \( t = 1.22 \mu m \) has been added in the figure.](image)

![Fig. 8. (a) Anisotropy at \( \theta = 0^\circ \) of sample 4 as a function of \( Q_x \); (b) plot of the isotropic \( S_1(Q_y) \) and anisotropic part \( B(Q_y) \) of the scattering at \( \theta = 0^\circ \) as a function of \( Q_y \). Note the reversal in the anisotropic scattering \( B(Q_y) \) with increasing \( Q_y \).](image)

domain height. In first approximation the data with different \( \theta \) values fit reasonably to the calculated anisotropy. Especially sample 1 demonstrates that this is satisfactory for all \( \theta \) values. From the fits of the calculated anisotropy in eq. (5) to the experimental ones in fig. 6 the domain heights of the samples have been determined and collected in table 1. It appears that these values, 1.25 \( \mu m \) in sample 1 and 1.63 \( \mu m \) in sample 2, depend strongly on the magnetic history of the sample. In particular the difference found between sample 2 and 3 is striking. The small difference between the domain heights in samples 3, 4 and 5 where the magnetic history is quite different, is also surprising.

The differences in the \( Q_x \) dependence of \( A \) at different \( \theta \) values should be ascribed to different relative \( \theta \) spreads as discussed in the theoretical
section and shown in fig. 3. These relative spreads are largest at low \( \theta \) values. From a rough comparison of the data in fig. 6 of sample 1 with the model expectation of the anisotropy in fig. 3 we conclude that an orientation spread of about 7° roughly explains the different \( A(Q_1) \) curves at different \( \theta \) values.

The anisotropy of sample 4 is more complicated. At \( \theta = 0^\circ \) the anisotropy \( A(Q) \) varies nearly quadratically in \( Q \) from a value of 0.5 for \( Q = 0 \) to 2.5 for \( Q = 3.0 \times 10^{-2} \) nm\(^{-1} \) (see fig. 8). The anisotropy at small \( Q \) values is explained by a preferred direction of the perpendicular domain walls parallel to the \( y \) direction, which causes a preferential scattering of the domains in the \( z \) direction for \( Q \) values in the range 0.5–1.5 \( \times 10^{-2} \) nm\(^{-1} \). Above this \( Q \) range the anisotropy is larger than one, which means that a preferential scattering in the \( y \) direction occurs, which is most probably due to scattering from the "closure domains" in the initial layer which short circuit the magnetic flux of the perpendicular domains (see fig. 1). Because the latter have their domain walls mostly in the \( y \) direction, the magnetic flux for short circuiting is essential in the \( z \) direction. Magnetic scattering is perpendicular to the magnetization, therefore scattering in the \( y \) direction from these "closure" domains is expected. This picture is consistent with Kerr observations and neutron depolarization results which also showed the existence of domain walls parallel to the \( xy \)-plane and to the magnetic field in the \( y \)-direction of the film plane.

5.2. Domain structure in the plane of the film

As shown in the theoretical section \( S(Q) \) itself gives information about the domain width in the plane of the film. From the maximum values of \( S(Q) \) for the different samples in fig. 9 the domain width in the plane is estimated to be of order \( 2\pi/2Q_{\text{max}} \approx 0.2 \) \( \mu \)m for the samples 1, 2, 3 and 4 and 0.1 \( \mu \)m for sample 5. The difference between 1–4 and 5 can only be attributed to the difference in magnetic history (see table 1). The results suggest that by applying a magnetic field, the initial domain size of 0.1 \( \mu \)m grows more than a factor 2, an effect which cannot be cancelled anymore by demagnetization or magnetization in any direction.

The shape of the experimental \( S_1(Q) \) qualitatively agrees with the simulation in fig. 4, using a simple model for the perpendicular domain structure.

The differences in maximum value of \( S(Q) \) for the samples do not agree with the ratio of the squares of the domain height as predicted by formula (4).

Another interesting feature is found by considering the scattering results of sample 4 in fig. 8. As discussed already in the previous section, the anisotropy exceeds 1 at some \( Q \) value, but in the scattered intensity, the anisotropic scattering expressed in \( B(Q) \) shows the opposite extreme values at \( Q_1 \) and \( Q_2 \). The minimum at \( Q_1 \) agrees very well with the average domain width as found also in the maximum of \( S_1(Q) \), which fits to the considerations in the previous section. The maximum in \( B(Q) \) at \( Q_2 \) however occurs at a \( Q \) value which agrees very well with the \( Q \) value of the maximum of sample 5, which corresponds to the domain size of the initial domain structure without treatment. Because this second peak was attributed to "closure" domains in the initial layer, it may be
concluded that the size of these “closure” domains is the same as the perpendicular domain size in the magnetically virgin film (sample 5).

Although the scattering on the initial layer is present in all the samples, only the anisotropy of sample 4 enables us to separate this scattering from the perpendicular domain scattering.

6. Conclusions

Small angle neutron scattering gives detailed information on the domain structure in thin films, especially when the angular dependence of the scattering in such mainly uniaxial domain structures is used. In particular, it appears possible to determine the mean magnetic domain sizes perpendicular to the film and in the plane of the film in two orthogonal directions. It also seems possible to determine the orientation variation of the perpendicular domains together with the variation in the domain heights. However these last two quantities cannot be determined separately from this kind of SANS experiments.

Perpendicular domain structures in thin films appear to be simple model systems for neutron scattering which can satisfactorily be described with a computer simulation.

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References