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Towards a 3D magnetometry by neutron reflectometry

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Abstract

Specular polarised neutron reflectometry with polarisation analysis allows one to probe in-depth magnetic profiles of thin films (along the normal to the film). Off-specular reflectometry gives information about lateral structures (in the plane of the film) with typical lengthscales ranging from 5 to 100 μm. Furthermore, surface diffraction at grazing angle gives access to transverse dimensions between 10 nm and 300 nm with a resolution in that direction of a few nanometers. The combination of these three techniques applied to magnetic systems can lead to a 3D magnetic structure measurement. Such a technique is however not applicable to the study of a single magnetic dot, but it can generate unique results in several cases including patterns of domain walls in thin films with perpendicular anisotropy, arrays of magnetic dots, and patterned lines in magnetic thin films. © 1999 Published by Elsevier Science B.V. All rights reserved.

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1. Introduction

Magnetic thin films are now largely produced for fundamental studies and technological applications. Following the discovery of giant magnetoresistance in antiferromagnetically coupled multilayered films [1], there has been an extensive interest in the precise measurement of the magnetic moment directions in each layer and at the interface between layers. Owing to the large magnetic coupling between the neutron and the magnetic moment, neutron diffraction is a powerful tool to obtain information about magnetic configurations. Polarised neutron reflectometry (PNR) has been used for several years [2,3] to investigate such problems. Polarised neutron reflectometry with polarisation analysis (PNRPA) has proved to be a useful tool to probe in-depth vectorial magnetic profiles [4,5].

In the last two years, new classes of challenges for PNR have appeared: the first one is to investigate the dynamical properties of magnetic thin films

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through inelastic scattering [6]. The second one is to follow the evolution of the spin configuration as a function of field or temperature in a reasonable beam time [7]. The third is to obtain information on thin films which are not magnetically homogeneous in the plane of the film.

With the very high intensities available on synchrotron sources, it is possible to investigate lateral sizes of rough surfaces down to the nanometer. We will present here some off-specular results obtained by neutron scattering on magnetic thin films and mesoscopic structures patterned in thin films.

2. General ideas

A specular reflectivity curve consists in the measurement of the intensities R reflected by a film as a function of the scattering vector $\boldsymbol{q}$ for each polarisation state of the incident and reflected neutron spin (R^{++} (resp. R^{--}) referring to incident and reflected "up" (resp. "down") neutrons, $R^{+-} = R^{-+}$ referring to the spin-flip signals). For $\boldsymbol{q}$ smaller than a critical value $\boldsymbol{q}_c$, the beam is totally reflected; for larger $\boldsymbol{q}$ ($q > 3q_c$), the intensity decreases as the fourth power of q but presents oscillations whose amplitude is related to the composition and the magnetism of the thin film. Roughly speaking, R^{++} and R^{--} depend on the chemical and the magnetic profile along the applied magnetic field. In most cases, the spin-flip signals $R^{+-} = R^{-+}$ depend mainly on the magnetism perpendicular to the applied magnetic field.

In the case of homogeneous films, the neutron is sensitive only to the in-plane magnetisation and all the intensity is reflected in the specular direction. In the case of magnetically non-homogeneous films, all the directions of the magnetisation can be explored and intensity is scattered off the specular direction.

The incidence plane is defined by the incident wave vector and the perpendicular to the surface (Oz). Fig. 1 gives the corresponding geometry. The convention is to call "off-specular" the intensity measured in the incidence plane (x, z) and "surface diffraction" the intensity measured out of the reflection plane along the (Oy) direction.

The incident and reflected wave vector $\boldsymbol{k}_\mathrm{i}$ and $\boldsymbol{k}_\mathrm{o}$ are given by

$$\boldsymbol{k}_\mathrm{i} \begin{vmatrix} k_0 \cos\theta_\mathrm{i} \\ 0 \\ -k_0 \sin\theta_\mathrm{i} \end{vmatrix} \quad \text{and} \quad \boldsymbol{k}_\mathrm{o} \begin{vmatrix} k_0 \cos\theta_\mathrm{o} \cos\Delta\theta_y \\ k_0 \cos\theta_\mathrm{o} \sin\Delta\theta_y \\ k_\mathrm{o} \sin\theta_\mathrm{o} \end{vmatrix},$$

where $\boldsymbol{k}_0 = 2\pi/\lambda$ is the value of the neutron wave vector, θ_i is the incidence angle, $\theta_0 = \theta_\mathrm{i} + \Delta\theta_x$ is the reflection angle and $\Delta\theta_y$ is the angle of reflection relative to the incidence plane. These angles are defined in the Fig. 1.

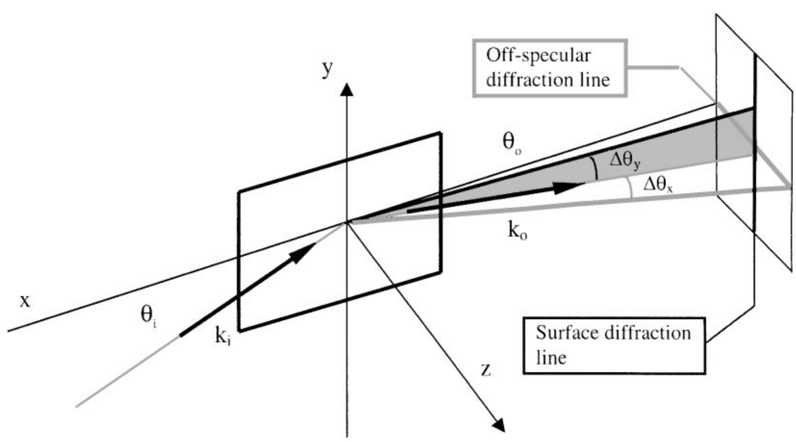

Fig. 1. Setup of the experiment. It is possible to study off-specular diffusion in the plane of incidence (along the "off-specular" line) and in the plane perpendicular to the plane of incidence (along the "surface diffraction" line).

The scattering vector wave vector $\boldsymbol{q}$ defined by $\boldsymbol{q} = \boldsymbol{k}_o - \boldsymbol{k}_i$ is given by

$$\boldsymbol{q}\left|\begin{array}{c} -k_0(\theta_i\Delta\theta_x + (\Delta\theta_x)^2/2 + (\Delta\theta_y)^2/2) \\ k_0\Delta\theta_y \\ k_0(2\theta_i + \Delta\theta_x) \end{array}\right|.$$

We have supposed that all the angles were small and performed a second-order Taylor expansion. It appears clearly that q_x is a second-order term and q_y is a first-order one.

The lateral sizes which are probed are typically given by $2\pi/q_x$ and $2\pi/q_y$ which are very different. For example, on the reflectometer PADA, which is monochromatic with a wavelength of 0.4 nm, we have $10\ \text{nm} < 2\pi/q_y < 400\ \text{nm}$ and $1\ \mu\text{m} < 2\pi/q_x < 50\ \mu\text{m}$. The upper limit is given by the resolution of the reflectometer and the lower limit by the lack of intensity. It appears that except for a region in the micron size range, due mainly to the lack of intensity, it is possible to probe a large range of lateral scales by reflectometry.

3. Non-specular neutron diffraction on periodic gratings

It is possible to produce periodic gratings using lithographic techniques. Optical lithography is especially suited for producing gratings in the micron size range. The different etching processes are dry etch by argon milling, reactive ion etching (RIE) or chemical etching.

A nickel grating with a 10 μm periodicity and 5 μm wide lines has been etched chemically in a solution of $FeCl_3$ in a nickel thin film (90 nm thick) deposited on a glass substrate. Off-specular diffusion has been measured on this sample using a time-of-flight reflectometer, by measuring a rocking curve around 0.75°, the detector being fixed at 1.5°. The intensity map is plotted in Fig. 2 in the (q_x, q_z) plane. One can observe different lines at constant q_x. The line $q_x = 0$ corresponds to the specular signal. The other lines (along $q_x = \pm 0.0006$, ± 0.0012, $\pm 0.0018\ \text{nm}^{-1}$) correspond to the different diffraction modes. Fig. 2 shows that it is possible to observe up to three diffraction orders.

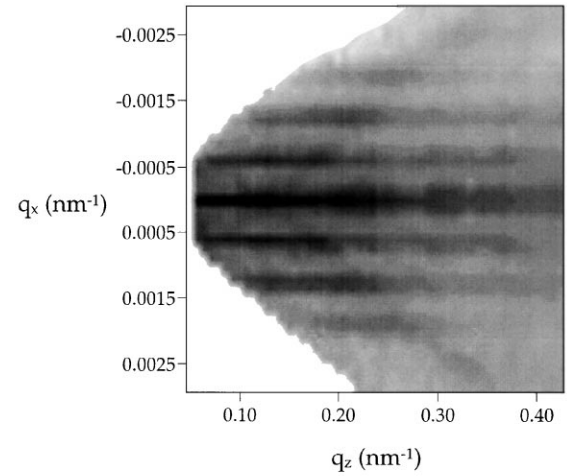

Fig. 2. Diffraction map in the (q_x, q_z) plane on a grating of nickel lines (width 5 μm, periodicity 10 μm, thickness 90 nm) deposited on a glass substrate. The line $q_x = 0$ corresponds to the specular reflection, the other lines correspond to diffraction modes.

In Fig. 3, the specular signal and the first two diffraction modes $+1$ and -1 have been plotted versus q_z. For clarity, the intensities of the mode -1 (resp $+1$) have been divided by a factor of 100 (resp. 10 000). Numerical fits based on a dynamical formalism [8] are plotted in black lines. They are in good agreement for the $+1$ mode but the intensity of the -1 mode is overestimated by a factor of 4. The typical reflectivity oscillations are not present because the resolution was lowered to increase the available neutron flux.

4. Diffraction on periodic magnetic stripe domains

We have measured magnetic surface diffraction on magnetic stripe domains appearing in $Fe_{0.5}Pd_{0.5}$ thin films. The sample was prepared by molecular beam epitaxy under ultra-high vacuum (10^{-7} Pa). A 2 nm seed layer of Cr was deposited onto a MgO (0 0 1)-oriented substrate in order to allow the epitaxial growth of the 60 nm single-crystal Pd buffer layer. After a 10 min annealing at 700 K, a 50 nm thick $Fe_{0.5}Pd_{0.5}$ alloy layer was deposited at room temperature using a monolayer (ML) by monolayer growth method in order to induce a chemical order similar to the one found in

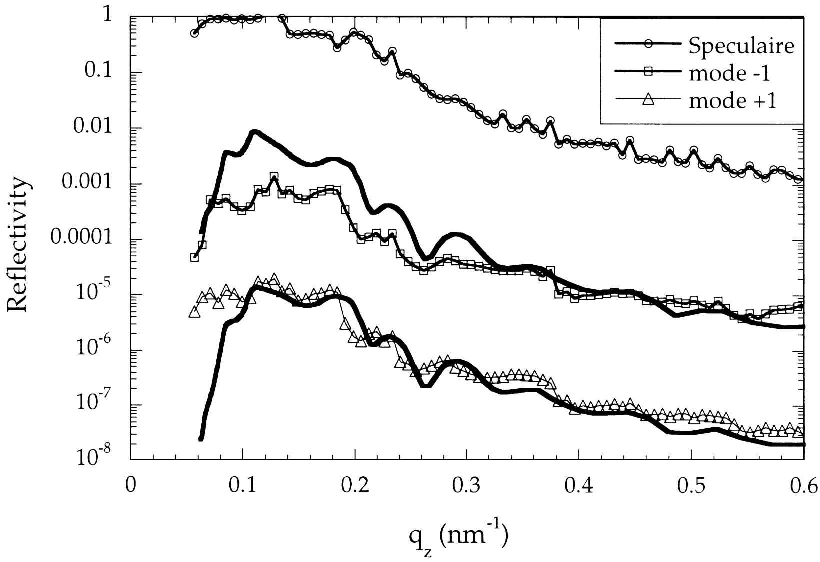

Fig. 3. Intensity of the diffracted modes + 1 and − 1 (triangles and squares) versus q_z. Numerical fits are plotted in black lines. For clarity, the intensities of the mode − 1 (resp. + 1) have been divided by a factor of 100 (resp. 10 000).

the tetragonal structure $L1_0$. This structure consists of alternate atomic layers of Fe and Pd on a body centred tetragonal lattice [9]. After a magnetisation along the easy axis, a stripe domain structure is observed (see bottom picture in Fig. 4) [10].

The diffraction measurement has been performed using a small angle neutron scattering spectrometer (the spectrometer PAPOL at the Laboratoire Léon Brillouin). In the experiment, the lines were aligned along the plane of incidence. An example of diffraction in shown in Fig. 4. One can observe a bright specular spot and two weaker (10^{-3}) off-specular peaks. The positions of these peaks along the (Oy) axis reflects the periodicity of the stripe domains (100 nm). The maximum intensity of these peaks is obtained in the direction correponding to the critical angle θ_c of the layer whatever the incidence angle is. The absolute maximum intensity of the diffraction peaks is obtained when the incidence angle is θ_c. These peaks have a behaviour similar to Yoneda peaks (or anomalous reflections) [11–13]. As a first approach, it is possible to explain these observations by using a DWBA approach. The considered unperturbed system is the flat FePd layer; the perturbation is the magnetic structure created by the stripes. In this case, the diffuse cross-section can be written as [14]

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{diff}} = (L_x L_y)\frac{|k_0^2(1-n^2)|^2}{16\pi^2} \times |T(\boldsymbol{k}_1)|^2\,|T(\boldsymbol{k}_2)|^2\,S(\boldsymbol{q}_t)$$

with

$$S(\boldsymbol{q}_t) = \frac{\exp(-[(q_z^t)^2 + (q_z^{t*})^2]\sigma^2/2)}{|q_z^t|^2} \times \int\!\!\int_S dX\,dY(\exp(|q_z^t|^2 C(X,Y)) - 1) \times \exp(i(q_x X + q_y Y)),$$

where $C(X, Y)$ is the magnetic roughness correlation function, $\boldsymbol{q}$ is the diffusion vector $\boldsymbol{k}_2 - \boldsymbol{k}_1$ and $\boldsymbol{q}_t$ is the wave-vector transfer in the medium. Maxima are obtained in the diffuse scattering when $\boldsymbol{k}_1$ or $\boldsymbol{k}_2$ makes an angle close to θ_c since in these positions, the Fresnel coefficients $\boldsymbol{T}$ reach a maximum.

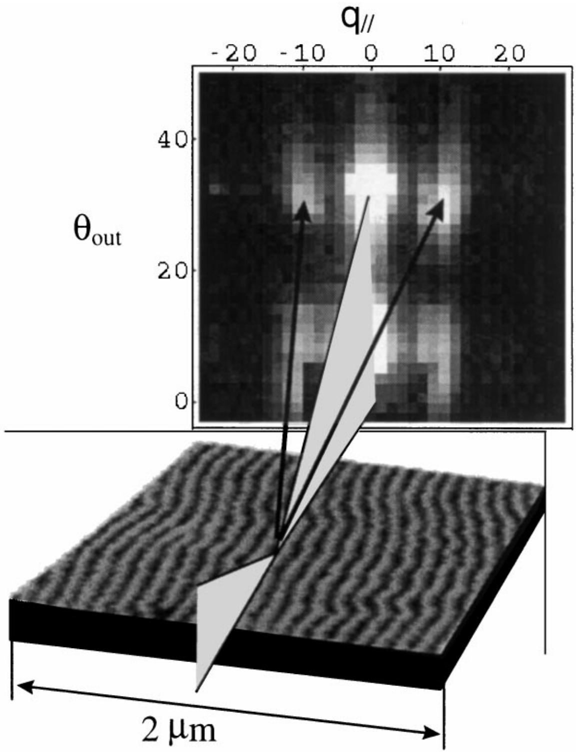

Fig. 4. Diffraction geometry and off-specular diffusion signal measured on a network of magnetic domains using a multidetector (top picture). The top peaks are the specular and off-specular peaks. The bottom signal is due to the refracted wave. The bottom picture is a MFM image of magnetic domains observed in the $Fe_{0.5}Pd_{0.5}$ thin films.

In the case of our magnetic lines, we define the correlation function of the magnetic roughness as:

$$C(X, Y) = \frac{1}{S_0} \int\int_{S_0} M(x, y)\, M(x + X, y + Y)\, \mathrm{d}x\, \mathrm{d}y.$$

The magnetic correlation function is a sawtooth function if we assume sharp interfaces between the domains. Fig. 4 shows an MFM picture of the magnetic stripe domains with the corresponding experimental diffusion pattern.

If q_z^t is small, $S(\boldsymbol{q}_t)$ reduces to the Fourier transform of the magnetic roughness correlation function:

$$S(q_t) = \int\int_S \mathrm{d}X\, \mathrm{d}Y\, C(X, Y)\exp(i(q_x X + q_y Y)).$$

The surface diffraction signal measures the Fourier transform of the magnetic correlation function. The Fig. 5 shows the off-specular signal calculated for two different incidence angles, the critical angle $\theta_c = 0.5°$ and $\theta_i = 0.7°$. The peak positions are unchanged whatever the incidence angle is. The maximum intensity is obtained when the incidence angle is equal to the critical angle θ_c.

However, the DWBA approach is not well suited to this problem since the magnetic roughness extends over the full thickness of the magnetic layer so that the flat layer as a basis state is very far from

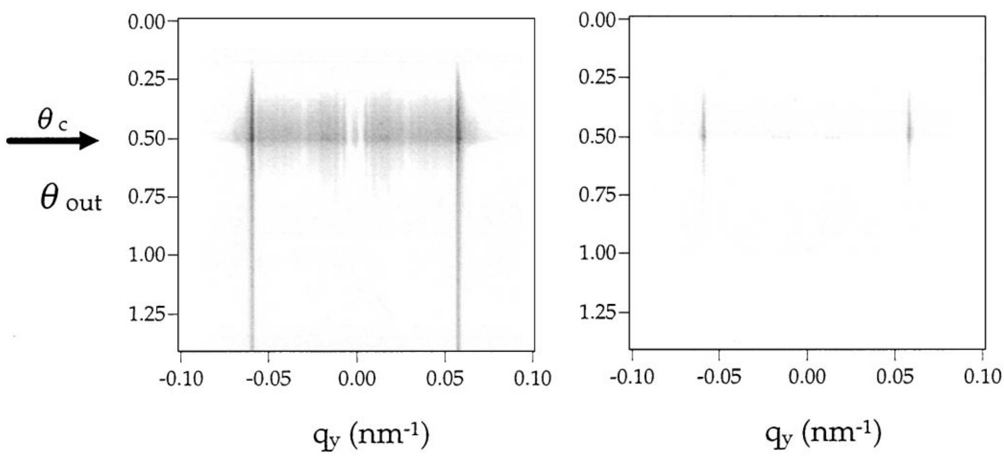

Fig. 5. Calculated off-specular signal as measured on a multidetector for two different incidence angles (a) $\theta_{inc} = \theta_c = 0.5°$ and (b) $\theta_{inc} = 0.7°$. The peaks maximum does not move but the intensity decreases as soon as the incidence angle is moved away from the critical angle θ_c.

the real eigenstates of the system. The determination of quantitative magnetic information will require a fully dynamical theory.

5. Conclusion

We have shown that it is possible to observe off-specular neutron diffusion on thin films in all the directions of the reciprocal space. We have observed surface diffraction from periodic magnetic domains. By a simple DWBA approach, it is possible to account qualitatively for the results. It should soon be possible to describe these results quantitatively by using a matricial dynamical approach. We hope that our efforts will pave the way for a new technique of 3D magnetometry; this will make it possible to measure quantitatively structures with an in-depth and in-plane resolution.

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