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Perspectives of polarized-neutron reflectometry: magnetic domains and off-specular scattering

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Abstract

Specular reflectometry of polarized-neutrons was developed in the 1980s as a tool for measuring magnetic depth profiles in flat films, which were laterally uniform. When the lateral uniformity breaks down in an assembly of domains, off-specular grazing incidence scattering takes place. This review discusses this new frontier of reflectometry, describing the advances that are taking place in linking the observations of the scattering at grazing incidence with the size, the statistics, and the magnetic orientation of the domains. The article discusses also the progress made in linking the domain distribution thus found with the transport properties of these nanomagnetic systems. © 2001 Published by Elsevier Science B.V.

Keywords: Polarized-neutron reflectivity; Off-specular scattering; Magnetic thin films; Multilayers; Domains

1. The image of antiferromagnetic domains

Polarized-neutron reflectometry (PNR) was developed in the 1980s as a tool for measuring magnetic depth profiles in flat films [1]. Basically an optical technique, it measures the intensity of the specular reflection from the surface: this can be accomplished by a single counter, poised at an angle θ with the reflecting surface, and 2θ with the primary beam. However, in several instruments a one-dimensional, position sensitive detector is used, with the geometry sketched in Fig. 1. Such detectors measure not only neutrons that are specularly reflected, but also those scattered at grazing incidence. In the geometry of Fig. 1 a one-dimensional detector discriminates only neutrons exiting the surface at an angle θ_f different from θ_i; however the scattering takes always place in the plane of reflection. Neutrons scattered out of the reflection plane at an angle φ will be counted in the same counter set at the same θ_i.

A special case in which such counters have turned out to be useful is that in which the reflecting film was a multilayer, with the magnetization of subsequent magnetic layers alternating parallel and opposite to a given axis within the plane of the films (Fig. 2). These kinds of materials were made popular by their giant magnetoresistive properties, stemming from the destruction of the antiferromagnetic state by laboratory magnetic fields [2]. If the repeat distance of the multilayer is D, the chemical Bragg reflections occur at $q_z = 4\pi \sin\theta/\lambda \approx 2\pi n/D$, where q_z is the momentum transfer perpendicular to the surface, λ the neutron wavelength, and n is a positive integer. The relation is only approximate, because it does not take into account effects due to the refractive index. The AF Bragg reflections appear at $q_z \approx 2\pi(n + 1/2)/D$.

Often the AF Bragg reflections are not sharp, but are made of two components, one sharp and one diffuse. This phenomenology has been presented in several ways. In Fig. 3 a contour plot is presented [3] as it

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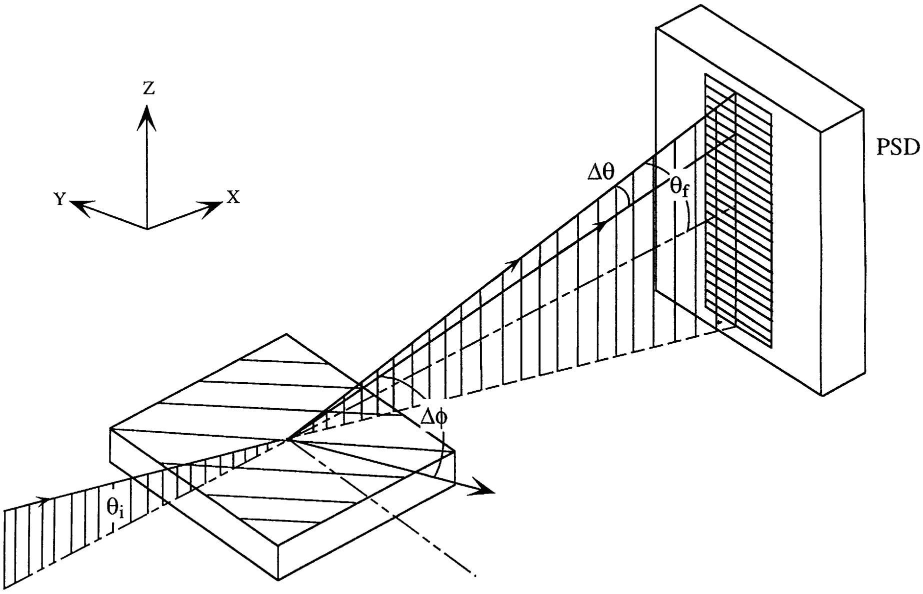

Fig. 1. Geometry of scattering at grazing incidence: specular reflection ($\theta_i = \theta_f$), scattering in the specular plane ($\varphi = 0$) and off the specular plane.

appears at a reflectometer making use of a broad band of neutron wavelengths: the sample is set at a fixed angle with respect to a polychromatic beam. Fig. 4 presents a typical contour plot [4] obtained in a reflectometer using a monochromatic beam: to span an adequate momentum range the sample is rotated. Fig. 5 gives a contour plot [5] in terms of the momentum transfers q_z and q_x:

$$q_x = \frac{2\pi}{\lambda}(\cos\theta_f - \cos\theta_i) \tag{1a}$$

$$q_z = \frac{2\pi}{\lambda}(\sin\theta_f + \sin\theta_i) \tag{1b}$$

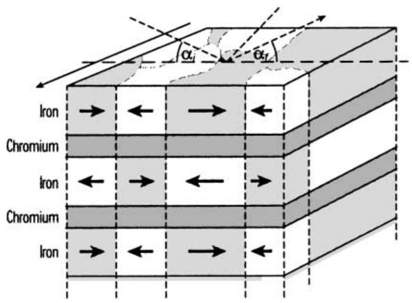

Fig. 2. An AF superlattice of Fe/Cr, and its breakdown into AF domains.

As it can be seen the diffuse scattering appears at the value of q_z corresponding to the AF peak (π/D), and spreads out along q_x. The patterns of Figs. 3 and 4 can easily be interpreted as the line $\theta_f + \theta_i = (\lambda/2\pi)(\pi/D)$. In Fig. 3, the ridge of maximum intensity follows the straight line obtained by keeping θ_i fixed and varying λ; in Fig. 4, the ridge appears for $\theta_f + \theta_i =$ constant. The two methods seem exactly equivalent. However, the resolution of the two

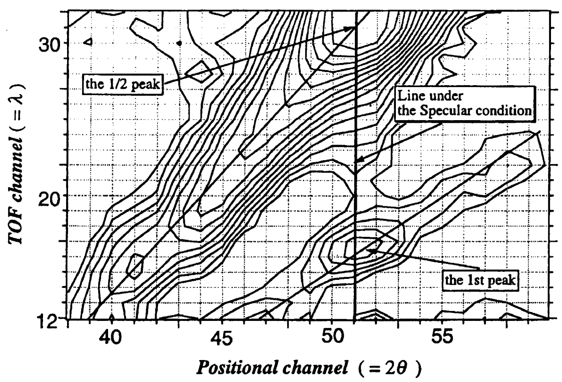

Fig. 3. Off-specular scattering from an AF Fe/Cr multilayer: the contours are plotted vs. neutron wavelength and angle of scattering. Sizeable scattering occurs around the AF (1/2) peak [3].

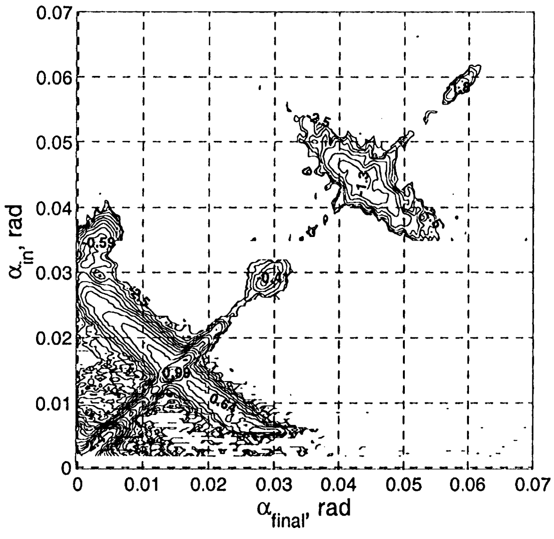

Fig. 4. Contour plot of an AF Fe/Cr multilayer, as a function of the angles of incidence α_{in} ($=\theta_i$) and scattering α_{final} ($=\theta_f$). The large peaks on the diagonal are AF [4].

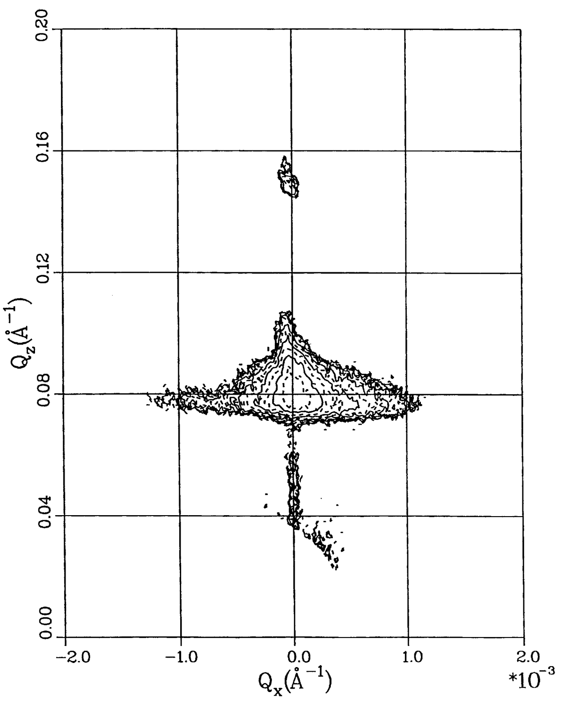

Fig. 5. Contour plot of an AF Fe/Cr multilayer, as a function of q_x, q_z. The strong innermost peak along the line $q_x = 0$ is AF; the outer weak one is structural [5].

techniques is different, as can be seen by differentiating Eqs. (1a) and (1b).

2. Antiferromagnetic domains: the Born approximation

For the multilayer system correlation between roughness at each interface from bottom to top layer has to be taken into consideration, as seen in Fig. 2. Coherent interfacial roughness and roughness without correlation between interfaces coexist in real multilayers. Although at present there is no complete theory for neutron (or X-rays) scattering from interfacial roughness, scattering from coherent roughness appears only in the profile of a transverse scan at the q_z appropriate for a Bragg reflection [6]. On the other hand, neutrons reflected at the interfaces with incoherent roughness are diffusely scattered in all directions in the reciprocal space with no structure. As shown in Fig. 2, AF magnetic domains represent a peculiar kind of "roughness", which give rise to scattering only at the values of q_z of the AF Bragg reflections.

Following the lead of Sinha et al. [7], the cross section for magnetic scattering from an AF multilayer can be written after making some drastic assumptions:

1. The scattering is treated in the first Born approximation.
2. Non-magnetic scattering is neglected.
3. The AF structure is represented by an Ising system, with the magnetic moments either parallel or opposite to a unique direction. Domains can just have two states: black and white. On the surface, the domain structure appears with a checkerboard or a striped image.
4. The domain walls are perpendicular to the surface.

With these assumptions the cross section for a solid is:

$$\frac{d\sigma}{d\Omega} = \int_V d\vec{r} m(\vec{r}) \int_{V'} d\vec{r}' m(\vec{r}') \exp[-i\vec{q} \cdot (\vec{r} - \vec{r}')] \quad (2)$$

where m stands for a magnetic scattering amplitude. The cross section can be written explicitly for one of the domains shown in Fig. 2, as the square of an amplitude

$$A_T = \frac{m_T}{q_z}\left[\sum_{N_z}\exp(-\mathrm{i}n_z q_z d_z)\right] \times \int_{S_T}\exp[-\mathrm{i}(q_x x + q_y y)]\,\mathrm{d}x\,\mathrm{d}y \quad (3)$$

m_T may take the value $+m$ or $-m$, depending on the direction of magnetization of the surface layer; the summation is over the N_z AF layers of thickness d_z, and it is a constant at the q_z value appropriate for the AF peak. The integral in Eq. (3) plays an important role in defining the width of the diffuse peak. The cross section for the entire sample is obtained by summing and squaring over all domains that are positioned on the surface at the coordinates X, Y

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \sum_{TT'} A_T A_{T'} \exp[-\mathrm{i}\{q_x(X_T - X_{T'}) + q_y(Y_T - Y_{T'})\}] \quad (4)$$

This expression becomes simple only when there is no correlation between the domains. In that case the terms with $T = T'$ cancel each other, and the cross section is just equal to the sum of the squares of the amplitudes A_T.

This treatment is equivalent to that given by Sinha et al. [7]. In its most simple form, the scattering object they considered was a homogeneous body with a rough surface. After transforming the volume integral (Eq. (2)) into a surface integral, the diffuse intensity reflects the scattering by the various Fourier components that represents the roughness and the strength of these components. These in turn reflect correlation in roughness, as the lateral height–height correlation function $C(X, Y)$:

$$I(q) = \frac{I_0 \exp(-q_z^2\sigma^2)}{q_z^2}\int\int \exp[q_z^2 C(X,Y)] \exp[-\mathrm{i}(q_x X + q_y Y)]\,\mathrm{d}X\,\mathrm{d}Y \quad (5)$$

The roughness here is step-like and corresponds to the thickness of one antiferromagnetic layer (a structure may be accommodated within the volume, if it gives rise to a term that contains a simple summation along z). This roughness can be correlated along the surface; but, at least in the Ising model, its appearance is stepwise, and its correlation is an oscillating function of X and Y.

3. Evolution of the methods of analysis

Sizeable scattering was first observed around the AF peaks of Co/Ru [8]. The scattering had the shape of a ridge centered around a value of $|q|$ equal to the value of the maximum of the AF peak. No corresponding broadening was found at the first structural Bragg reflection. The lateral dimensions of the domains observed for Co/Ru with the help of Eq. (4) was found to be $L_x \sim 4$ μm.

Takeda et al. [3] investigated surface and interfacial roughness in single crystal Fe/Cr multilayers prepared by molecular epitaxy on (1 0 0) MgO. The profile was found to be very sensitive to the interfacial roughness originating from the substrate, condition of crystal growth and external magnetic fields. Their observations were made in zero field, where the system is AF, and at saturation. At zero field the magnetic AF showed broadening, but not the first structural Bragg reflection. At saturation, the AF peak disappeared but the first structural Bragg reflection — this time having a magnetic contribution — acquired a diffuse component. In analogy with non-magnetic multilayers, the magnetic disorder at the interface was taken as a height distribution of the magnetic scattering length rising or dipping from the average flat interface. The correlation function, $C(X,Y) = \langle z(x-X, y-Y)z(x,y)\rangle$, where $z(x, y)$ is the vertical displacement of the surface from its average height, was treated as an exponential

$$C(R) = I_0\sigma^2\exp\left(-\frac{|R|}{\xi}\right) \quad (6)$$

characterized by a correlation length ξ. The term $\exp[q_z^2 C(X,Y)]$ in Eq. (5) can then be expanded and each term Fourier transformed analytically. If the correlation function (6) is truncated after a certain length, the intensity can be separated into the sum of two terms, a delta function and a diffuse term

$$I(q_x,q_y) = \frac{2\pi I_0 \exp(-q_z^2\sigma^2)}{q_z^2} \times \left[2\pi\delta(q_x) + \sum_m \left\{\frac{2\xi(q_z^2\sigma^2)}{m!}\right\} \times \left\{\frac{1}{1+q_z^2\sigma^2/m^2}\right\}\right] \quad (7)$$

The diffuse term is a sum of Lorentzians, which converges rapidly for a sufficiently small $q_z\sigma$ product. This expression has the advantage of identifying a sharp component and a diffuse background of the Bragg reflection; the latter can be fitted easily with a lateral correlation length ξ.

A further development of these concepts has recently taken place [9], in the course of the analysis of off-specular scattering from an AF Co/Cu multilayer. Here the AF peak was found to be diffuse but the structural Bragg reflection was sharp and the low structural roughness was consistent with the results of a X-ray study. When at saturation the diffuse scattering was very weak over the entire q_z range. To analyze the results Langridge et al. [9] pointed out that the diffuse scattering contains components arising from the structural roughness, the magnetic domain disorder and the interference between the two. The magnetization profile $m(r)$ was constrained to lie in the plane by the shape anisotropy, and it was defined by a vector of fixed size and direction $\phi(r)$ varying from domain to domain. $\phi(r)$ was treated as a random variable, characterized by the correlation function $M(|r|) = \langle\phi(r)\phi(0)\rangle$. In analogy with the local height variation in the structural model of Sinha et al. [7], $M(i)$ was parametrized as $M(r) = \sigma_{\rm m}^2 \exp(-r/\xi_{\rm m})$. Structural roughness is included following the formalism of Sinha. With this formulation Langridge et al. [9] were able to analyze the angular variation of the diffuse magnetic scattering in terms of $\sigma_{\rm m} = \langle\phi^2\rangle$, which is the width of the angular distribution and therefore characterizes the magnetic domain disorder, and $\xi_{\rm m}$, the lateral correlation length, which is a measure of a typical domain size.

A totally different approach was followed by Toperverg, in collaboration with several research groups [4,10]. He considered the scattering from ''magnetic roughness'' in the distorted wave Born approximation (DWBA), where basically the amplitude of the incident and reflected waves at each depth of the sample are obtained by solving the reflectivity equations, and scattering of these waves off the specular peak may occur as a single scattering process. The novelty of Toperverg's approach consisted in combining DWBA with an elegant formalism to express the interaction of the neutron spin with an arbitrary magnetic configuration.

The supermatrix formalism [10] allows an easy recasting of the reflectivity in terms of total polarimetry: it basically suffices to substitute the nuclear and magnetic structure factors with the scalar and vectorial components of the reflectance:

$$\hat{R} = \frac{1}{2}(R_0 + \boldsymbol{R}\hat{\boldsymbol{\sigma}}) \quad (8)$$

Here $\hat{\boldsymbol{\sigma}}$ is the vector of Pauli matrices. The reflectance itself is obtained by calculating

$$\begin{pmatrix} \hat{T}|t_0\rangle \\ \mathrm{i}k_{\rm S}\hat{T}|t_0\rangle \end{pmatrix} = \hat{S}_{\rm tot}\begin{bmatrix} (1+\hat{R})|t_0\rangle \\ \mathrm{i}k_0(1-\hat{R})|t_0\rangle \end{bmatrix} \quad (9)$$

for the supermatrix $\hat{S}_{\rm tot}$ of the entire film, which has neutron transmittance $\hat{T}$. Here $|t_0\rangle$ is the incident beam vector with wavenumber k_0, and $k_{\rm S}$ the wavenumber in the substrate. In principle, measurements have to be taken with the incident neutron polarized along one of the principal axes, and the polarization of the scattered neutrons has also to be analyzed in three components. Fortunately the experiments do not always require such amount of information. This method of analysis provides a framework by which to calculate the reflectivity starting from a model. This then can be compared with experimental results, and the fitting of the images may give a clue on how satisfactory the model is. In spite of such seemingly cumbersome treatment, calculated images were obtained quite similar to the experimental ones [4].

The experimental determination of the polarization of the scattered beam offered a real challenge. Conventionally magnetic mirrors are used, but these cover only a small angular range of the scattered beam. A novel solution of this problem was to use a transmission polarization filter, consisting of polarized ^{3}He gas. The first experiment (on a Fe/Cr superlattice) permitted the simultaneous investigation of sections of the reciprocal space while exploiting spin sensitivity, and provided a better mapping of the data to be interpreted in terms of the supermatrix formalism [11].

4. Domain size and transport properties

A robust effort has taken place to associate the domain size distribution — even if imperfectly determined — with transport properties such as the magnetoresistance of multilayers. An early paper already pointed out that, when a Fe/Cr AF multilayer was subjected to a magnetic field, the magnetoresistance was proportional to the size of the AF peak [12]. It was later shown that a similar relationship existed when a Fe/Cr sample was annealed at different temperatures [5]. Are the *domain sizes* also linked to the magnetoresistance [13]? Experiments gave evidence that, for two different systems, some correlation of this type exists. Antiferromagnetic $Ni_{80}Fe_{20}$/Ag multilayers exhibit, upon annealing at temperatures of ~300°C, an increased magnetoresistance. Neutron reflectivity measurements indicated [14] a considerable broadening of the AF peaks upon annealing. This was associated with diffusion of silver in the Fe layers, that promotes the formation of planar ferromagnetic domains of micron size with each $Ni_{80}Fe_{20}$ layer AF correlated along the growth axis. Co/Cu multilayers have an altogether different phenomenology. The as-prepared sample presents a sizeable magnetoresistance, but his drops irreversibly after cycling the sample in a suitable magnetic field. Neutron measurements [15] showed that in the as-prepared sample the Co domains (of μm size) are strongly AF correlated across the Cu. Fields as low as the coercive field destroy the AF correlation.

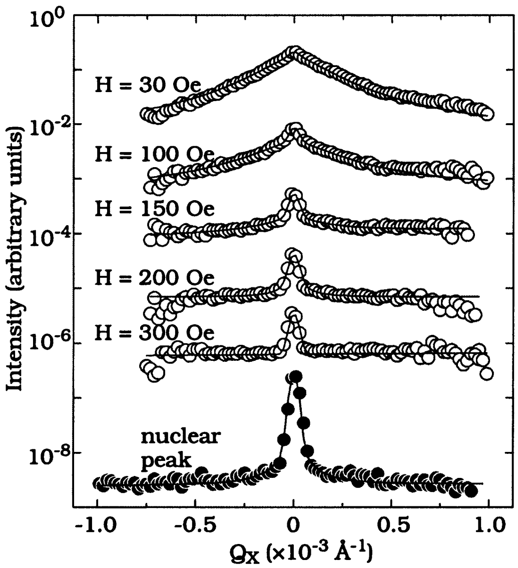

Fig. 6. The diffuse scattering observed at the AF peak of a Co/Cu multilayer as a function of applied field. The lines are fits to the data [9].

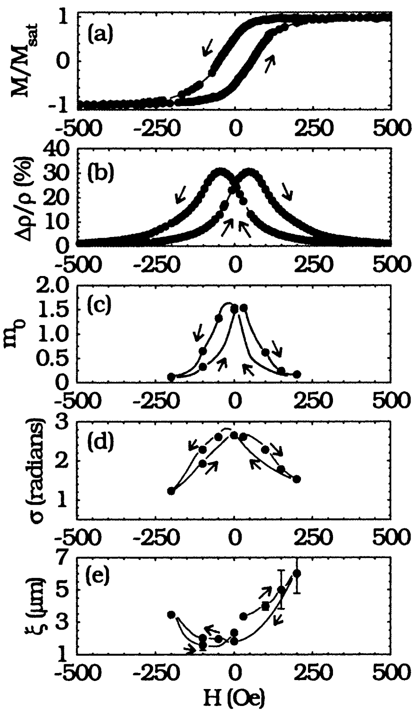

Fig. 7. Field dependence of the magnetization, the magnetoresistance and the domain fitting parameter for a Co/Cu multilayer [9].

A more systematic correlation between domain size and magnetoresistance is put in relief by following the behavior of a sample in a magnetic cycle. Figs. 6 and 7 shows the field dependence of the domain size and the magnetoresistance for a sample of Co/Cu [9]. The magnetic domains become coarser when the magnetic field is applied — as indicated by the increase of ξ. How the coarsening takes place, and how dependent is on the magnetic prehistory of the sample, is the subject of a new fascinating study [16].

In conclusion, in the past 10 years polarized-neutron reflectivity has given birth to a new off-specular technique capable of observing magnetic domains. The extent of the excitement and of the interests that are being spawned is hard to compress in a short article but become more vivid by a survey of the articles published, following a recent successful workshop [17].

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