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Physica B 397 (2007) 79–81
PHYSICA B
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# Multiple scattering in magnetic SESANS

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**Abstract**

Magnetic spin-echo SANS (SESANS) gives real-space information about correlations in a ferromagnetic material on a length scale 10–10 000 nm, inaccessible in conventional SANS. In *non*-magnetic SESANS correction for multiple scattering is done analytically. For *magnetic* SESANS we propose a modified formula and verify it by magnetic-SESANS measurements in stacks of copper plates with electro-deposited Ni. Such Ni film consists of domains magnetized perpendicular to the plane and length equal to its thickness. Perspectives of the technique are mentioned.


*PACS:* 78.70.Nx; 75.70.Kw

*Keywords:* Magnetic scattering of polarized neutrons; Thin films

Spin-echo small-angle neutron scattering (SESANS) is a novel method to determine the structure of materials in real space [1,2]. The method is based on the Larmor precession of polarized neutrons transmitted through special devices before and after the sample, which encode the scattering angle into a net precession angle. The advantages are: (1) high collimation of the beam is not needed, which allows high intensity; (2) a large length scale (10–$10^4$ nm) of correlations in the sample can be studied (inaccessible to ordinary SANS) and (3) multiple scattering can be taken into account analytically [1]. The principal difference from conventional SANS is that SESANS measures a real-space function. Until recently SESANS was applied to non-magnetic systems only, but in Refs. [3,4] we gave a conclusive demonstration of SESANS to magnetic systems.

The central feature in *magnetic* SESANS is the change of the polarization in the scattering process. As a consequence, to observe the spin-echo (SE) signal a flipper between the SE arms is in some cases not needed. So, by switching on or off the flipper and varying the precession plane, one can distinguish magnetic scattering from various magnetization directions from nuclear scattering and the unscattered beam. In a 2nd, 3rd... magnetic scattering event the spin is flipped again and so on. This modifies the analytical formula for multiple scattering (see Ref. [1], Eq. (19)). It is the aim of this paper to test this formula.

The polarization vector in the SE technique rotates in the plane perpendicular to the guide magnetic field; its orientation can be chosen. However, the local magnetization in the sample is a vector of arbitrary direction. Analysis of all 9 possibilities (3 components of magnetization × 3 precession plane of polarization) is similar to 3D analysis of the depolarization. This is the subject of a recent paper by Rekveldt [4]. Here we will restrict ourselves to a geometry where the local magnetization is along the beam axis $x$ and the polarization is in the $xz$-plane (Fig. 1). In this case the phase accumulation after one scattering event will be reversed.

## 1. Concept of spin echo SANS (SESANS)

SESANS is based on encoding the neutron fly direction through a precession device into an unique precession angle, when the device's front and end faces are inclined by an angle $\theta_0$ towards its main axis [1,2].

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doi:10.1016/j.physb.2007.02.048

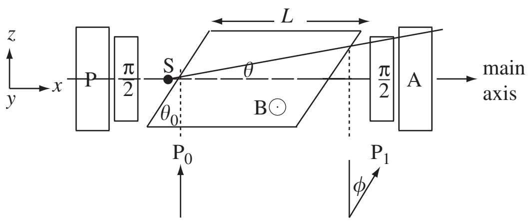


Fig. 1. Precession device with $\pi/2$-flippers (to orient and analyze the polarization perpendicular to the field $B$) between a polarizer $P$ and analyzer $A$. $\phi$ is the precession phase along the path after scattering in $S$, under the angle $\theta$ towards the main axis.

The polarization $P(\phi)$ after this device equals the cosine of the precession angle $\phi \approx c\lambda BL(1+\theta\cot\theta_0)$ (see Fig. 1). Two such devices in series with opposite magnetic fields $B$ (or parallel fields and a spin flipper in between) enable one to measure small-angle scattering of a sample positioned also in between. The net precession angle $\Delta\phi$, after passing the devices (length $L$) at transmission angles $\theta_1$ and $\theta_2$, successively, is

$$\Delta\phi = \phi_1 - \phi_2 = c\lambda BL\cot\theta_0(\theta_2-\theta_1) \equiv ZQ_z \tag{1}$$

with *SE length*

$$Z = \frac{c\lambda^2 BL\cot\theta_0}{2\pi} \quad \text{and} \quad Q_z = \frac{2\pi}{\lambda}(\theta_2-\theta_1). \tag{2}$$

The polarization $P_{\rm m}$ of the transmitted beam behind the analyzer with intensity $I_{\rm m}$ is composed as

$$P_{\rm m}I_{\rm m} = P_0 I_{\rm ns} + P_{\rm s} * I_{\rm s}, \tag{3}$$

where $P_0$ and $I_{\rm ns}$ are the polarization and intensity of the non-scattered part of the beam. $P_{\rm s} * I_{\rm s}$ is a convolution of the cross-section $\mathrm{d}\sigma/\mathrm{d}\Omega(Q)$ (probability for scattering) and polarization $P_{\rm s}$ of the beam scattered at some angle $\theta$, corresponding to momentum transfer $Q$:

$$P_{\rm s} * I_{\rm s} = I_0 x \frac{1}{k_0^2}\int \mathrm{d}Q_y\,\mathrm{d}Q_z P_{\rm s}(Q_z)\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(Q), \tag{4}$$

where $x$ is the sample thickness (seen as variable parameter), $P_{\rm s}(Q_z) = P_0\cos(Q_z Z)$ and $I_0$ the intensity of the incident beam with wave vector $k_0$. The total cross-section $\sigma$ is:

$$\sigma = (1/k_0^2)\int \mathrm{d}Q_y \mathrm{d}Q_z \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(Q).$$

In the single scattering approximation ($\sigma x \ll 1$), we have $I_{\rm ns} = I_0(1-\sigma x)$ and $I_{\rm m} = I_0$, so Eq. (3) becomes

$$\frac{P_{\rm m}}{P_0} = 1 - \sigma x + \frac{x}{k_0^2}\int \mathrm{d}Q_y\,\mathrm{d}Q_z \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(Q)\cos(Q_z Z).$$

To calculate the scattering by a sample of finite thickness we write for a sample of vanishing thickness $\mathrm{d}x$: $(P_{\rm m}-P_0)/P_0 \to \mathrm{d}P/P$. Then this equation becomes

$$\frac{\mathrm{d}P}{P} = -\mathrm{d}x\left[\sigma - \frac{1}{k_0^2}\int \mathrm{d}Q_y\,\mathrm{d}Q_z \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(Q)\cos(Q_z Z)\right].$$

Integrating over the thickness $l$ of the sample gives

$$\frac{P_{\rm m}}{P_0} = \exp\left(-l\sigma - \frac{l}{k_0^2}\int \mathrm{d}Q_y\,\mathrm{d}Q_z \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(Q)\cos(Q_z Z)\right). \tag{5}$$

As shown by Kruglov [5], the second term is the projection of the spatial pair correlation function along the beam axis. For isotropic samples it is

$$G(Z) = \int \mathrm{d}x\langle\rho(r)\rho(r+Z)\rangle, \tag{6}$$

where $\rho(r)$ is the scattering potential at point $r$.

## 2. Magnetic SESANS

The polarization $\mathbf{P}_{\rm s}$ of the *magnetically* scattered neutrons in a ferromagnetic sample is given by Ref. [6]

$$\mathbf{P}_{\rm s} = 2\mathbf{m}_\perp(\mathbf{m}_\perp\cdot\mathbf{P}_0) - \mathbf{P}_0, \tag{7}$$

where $\mathbf{m}_\perp = \mathbf{m} - (\mathbf{m}\cdot\hat{\mathbf{q}})\hat{\mathbf{q}}$ with $\hat{\mathbf{q}}$ the unit scattering vector, $\mathbf{m}$ the unit magnetization vector, and $\mathbf{P}_0$ the polarization just before scattering. So, for neutrons scattered at $\mathbf{Q}\perp\mathbf{m}$ the polarization component parallel to the local magnetization is unchanged, but the perpendicular component changes sign. In the single scattering approximation no polarization is lost. With Eqs. (3) and (4) we rewrite Eq. (5) for magnetic SESANS as

$$P_{\rm m}I_{\rm m} = P_0 I_0(1-l\sigma)\cos(\phi_1+\phi_2) - \frac{I_0 P_0 l}{k_0^2}\int \mathrm{d}Q_y\,\mathrm{d}Q_z\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega(Q)}\cos(\phi_2-\phi_1). \tag{8}$$

The first term (non-scattered part of the beam) has $\phi_1+\phi_2$ for argument in the cosine. It vanishes (amplitude of the precession signal $\downarrow 0$, no SE signal) because $\int \mathrm{d}\lambda\rho_\lambda(\lambda)\cos(\phi_1(\lambda)+\phi_2(\lambda)) \approx 0$. This is true even for the spectral density $\rho_\lambda$ in a so-called "monochromatic" instrument. In the second term (the magnetically scattered neutrons), the flipping in the scattering process gives $\phi_2 - \phi_1$ in the cosine, so a SE signal *is* produced. Finally Eq. (8) is rewritten

$$P_{\rm m}I_{\rm m} = -\frac{I_0 P_0 l}{k_0^2}\int \mathrm{d}Q_y\,\mathrm{d}Q_z\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(Q)\cos(Q_z Z), \tag{9}$$

which may be denoted $P_{\rm m}I_{\rm m} = -P_0 I_0 l\sigma G(Z)$ if we introduce the function $G(Z)$ in analogy with Eq. (6)—but with $\rho_m(r)$ the *magnetic* scattering potential—and if we express $\phi_1 - \phi_2$ in $Z$ using Eq. (2).

A *second* scattering event reverses the polarization again, so the term for scattering twice must be subtracted from the term for single scattering and so on. Hence, including multiple scattering, Eq. (9) becomes

$$\frac{P_{\rm m}}{P_0} = G(Z)\left[l\sigma - \frac{(l\sigma)^2}{2} + \frac{(l\sigma)^3}{2\cdot 3}\cdots\right] \equiv G(Z)F(l\sigma), \tag{10}$$

where $F(l\sigma) = l\sigma\exp(-l\sigma)$. This function is plotted in Fig. 2. The amplitude of the SE signal is proportional to

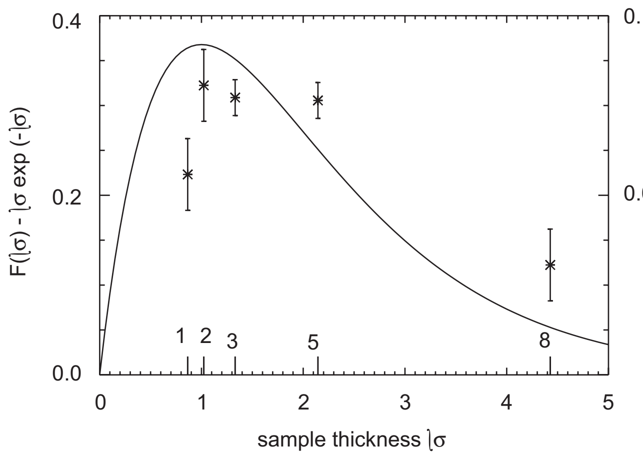


Fig. 2. Amplitude of the SE signal as a function of sample thickness $l$ in units of mean free path $l/l_{\mathrm{f}} = l\sigma$. The marks on the horizontal axis correspond to the total Cu/Ni thickness and the $*$'s to the minima at $Z = 2200\,\mathrm{nm}$ of the graphs in Fig. 3a.

$F(l\sigma)$ where $l\sigma = l/l_{\mathrm{f}}$ is the sample thickness in units of mean free path $l_{\mathrm{f}} = 1/\sigma$.

## 3. Magnetic SESANS measurements

The sample is a 15 μm Ni film electro-deposited on a copper substrate. Its "spaghetti" domain structure consists of rows of cylinders with length equal to the film thickness, magnetized perpendicular to the plane, and width (diameter of adjacent cylinders = spaghetti thickness) ≈ 3 μm. This structure was studied by angle dependent 3D neutron depolarization at $\lambda = 0.2\,\mathrm{nm}$. In perpendicular transmission we found $P_{\mathrm{m}}/P_0 = 0.6$ (±0.1 for different cut-outs) → $l\sigma = -\log(0.6) = 0.51$.

We used the monochromatic instrument for SESANS in Delft [3]—also laid out for $\lambda = 0.2\,\mathrm{nm}$—in *magnetic* mode (spin flipper between precession devices off). To vary sample thickness, we measured $P_{\mathrm{m}}/P_0(Z)$ in stacks of 1.8 plates cut from the sample. Fig. 3a gives the result, normalized to $P_0$, which we assumed to be the empty beam polarization in *nuclear* mode (spin flipper on) plotted as $P_0/2$ in Fig. 3a. The minima at $Z \approx 2200\,\mathrm{nm}$ (absolute value) are added in Fig. 2 (scale at right). In nuclear mode, with sample, we observed $P_{\mathrm{m}}/P_0 = 0.6$ independent of $Z$.

The data for small $Z$ depart with finite slope from $Z = 0$, confirming the cylinder- (not the sphere-model) [3] for the spaghetti domains. For $Z \downarrow 0$ we expect a similar behaviour as around 2200 nm. This is obscured by the scatter in data near $Z = 0$ due to the fact that the first term in Eq. (8) is not fully damped and interferes with the magnetic signal. This makes $P_{\mathrm{m}}$ critically dependent on experimental procedures.

After annealing the shape of the curve is strongly deformed (see Fig. 3b). This implies that the domain structure changes: part of the domains remains magnetized perpendicular to the plate, as before annealing and has size of order of 2 μm; part of the domains are magnetized in plane and has size ≈ 10 μm.

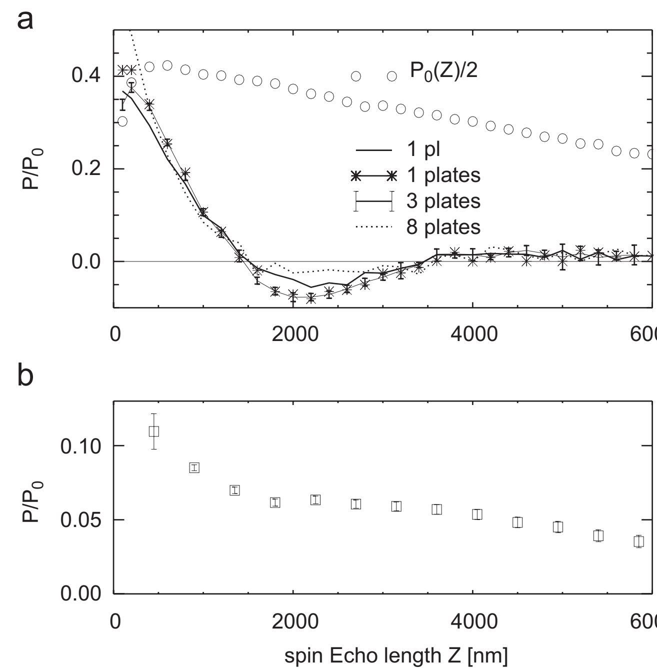


Fig. 3. (a) Depolarization measured on the SESANS instrument in Delft ($\lambda = 0.2\,\mathrm{nm}$) in 1...8 Cu/Ni plates as a function of $Z$. (For clarity the data for 5 plates, included in Fig. 2, are not shown); (b) idem in 1 plate after heating to 340 K.

We conclude that the formula for multiple scattering in magnetic SESANS is confirmed by the measurements. Comparing the present result with nuclear SESANS results in limestone in [1] shows the different characters of magnetic and nuclear SESANS.

A change in domain structure from perpendicular to in-plane magnetization after annealing clearly shows up in the measurements. This demonstrates the potential of magnetic SESANS for studies of ferromagnets in the critical regime.


## Acknowledgements

The work was supported by INTAS, Grant No. 03-51-6426, RFFR (project 05-02-16558) and partially by the EC, 6th Framework Programme "Strengthening the E R A", Research Infrastructures. Contract nr: HII3-CT-2003-505925.


## References


[1] M.T. Rekveldt, W.G. Bouwman, W.H. Kraan, O. Uca, S.V. Grigoriev, K. Habicht, T. Keller, in: Lecture Notes in Physics, Vol. 601, Springer, Berlin, 2003, pp. 87–99.

[2] M.T. Rekveldt, J. Plomp, W.G. Bouwman, W.H. Kraan, S.V. Grigoriev, M. Blaauw, Rev. Sci. Instrum. 76 (2005) 033901-9.

[3] S.V. Grigoriev, W.H. Kraan, M.T. Rekveldt, T. Kruglov, W.G. Bouwman, J. Appl. Crystallogr. 39 (2006) 252.

[4] M.T. Rekveldt, Rev. Sci. Instrum. 77 (2006) 073902.

[5] T. Krouglov, I.M. de Schepper, W.G. Bouwman, M.Th. Rekveldt, J. Appl. Crystallogr. 36 (2003) 117.

[6] Y.A. Izyumov, R.P. Ozerov, Magnetic Neutron Diffraction, Plenum Press, New York, 1970, pp. 171–176.