HEP

RAL-93-018
Science and Engineering Research Council

Rutherford Appleton Laboratory

Chilton DIDCOT Oxon OX11 0QX
RAL-93-018

The Use of Polarisation Analysis in Polarised Neutron Reflection Studies

J Penfold J Webster and D Bucknall

March 1993

THE USE OF POLARISATION ANALYSIS IN POLARISED NEUTRON REFLECTION STUDIES

J. Penfold, J. Webster and D. Bucknall.

ISIS Science Division, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 OQX, UK.

Abstract

The application of polarisation analysis to polarised neutron reflection studies is discussed. Its implementation on the reflectometer CRISP is described, and evaluated for a model system comprising of a thin nickel film.

Introduction

The specular reflection of neutrons gives information on the neutron refractive index profile normal to an interface, and hence can provide important information about the composition of surfaces and interfaces (1,2). Due to the magnetic interaction; magnetic materials have a neutron spin dependant refractive index. The refractive index, n, in a magnetic material contains a nuclear part n_n and a magnetic part $\pm n_m$, where the (±) signs depend on whether the neutron spin polarisation is parallel (+) or antiparallel (-) to the magnetisation direction. The spin dependence of n is responsible for a number of optical phenomena, the most widespread of which is the use of total reflection from magnetised mirrors to produce polarised neutron beams (3), However, since the pioneering work of Felcher (4) the technique has developed into a sensitive method for measuring magnetisation profiles in thin films (5-7). The application of the technique has benefited from the construction of dedicated polarised neutron reflectometers (8,9)

In conventional polarised neutron scattering the use of polarisation analysis, if not in common use, is well established and can be powerfully applied to seperate spin flip from the non-spin flip scattering. However, in polarised neutron reflectometry it has not been extensively considered. Bland et al (12,13) have discussed the use of polarisation analysis to measure the spin orientation of Larmor precessing polarised neutrons critically reflected from the ferromagnetic films. We will discuss here the use of polarisation analysis in conventional polarised neutron reflection experiments.

On a polarised neutron instrument the principle components which interact with the neutron spin are the polariser and spin-flipper. The efficiencies of these components are in practice always less than 100%. It is essential to correct experimental data to allow for these imperfections, and obtain reliable estimates of the spin-dependent reflectivities, R_+ and R_-. It is straightforward to design polarising mirrors with efficiencies ≥ 0.99; but it is difficult to obtain spin-flippers with high efficiency over the wide wavelength band encountered on time of flight neutron reflectometers (14). Indeed the Drabkin non-adiabatic spin-flipper on CRISP has a strong wavelength dependence (15). Although alternative spin flippers exist, such as current sheets (16) and single coil spin turners (17), they have other disadvantages for neutron reflectometers and do not garantee high efficiency.

It is, therefore, essential to obtain an accurate estimate of the polariser (P) and spin-flipper (f) efficiencies and a number of methods have been proposed (18). On CRISP we have used the "depolarising shim" method (15); and this provides an adequate estimate of P and f. In common with the experience of polarised neutron diffraction experiments it has proved difficult to obtain P and f with sufficient accuracy to adequately correct data for large sample "flipping ratios" (R+/R-), for flipping ratios ≥ 10. We will demonstrate in this paper that the incorporation of polarisation analysis in polarised neutron reflection experiments offers the opportunity to measure the efficiency factors with greater accuracy. More importantly it offers the opportunity to obtain a greater discrimination against the "unwanted" spin contractions in the contamination of R_+ and R_-.

Polarisation Analysis In Polarised Neutron Reflection

The spin dependence of the neutron refractive index gives rise to a spin dependent reflectivity R_+ and R_-; where the (+-) signs refer to the neutron spin polarisation direction paralled (+) or anti parallel (-) to the magnetisation direction. The object of a polarised neutron reflection experiment is to measure the parallel and antiparallel reflected intensities and relate them to the calculable spin reflectivities R_+, R_-. R_+, R_- are directly related to the magnetisation profile in the film.

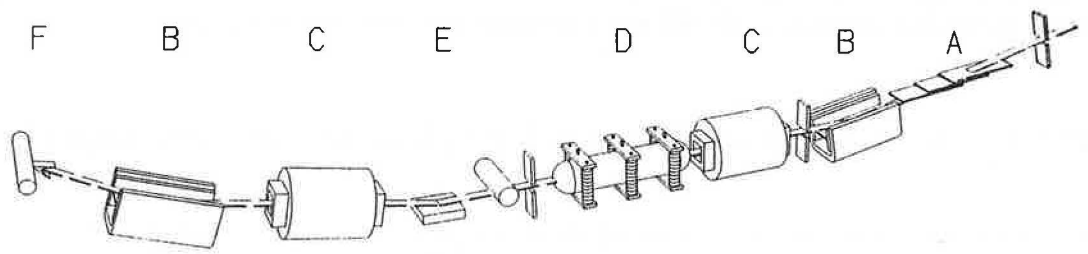


Figure 1 Schematic diagram of the polarised mode of the CRISP reflectrometer, where B is the polarised analyser, C the spin flipper, D the magnetic guide field, A the frame overlap mirrors E the sample position and F the neutron detector.

A general schematic layout of the CRISP time of flight polarised neutron reflectometer installed at the ISIS pulsed neutron source, is shown in figure 1. The main components of the reflectometer are the cobalt-titanium polarising supermirrors (B) and the Drabkin two coil non-adiabatic spin flippers (C). The polarising supermirror provides a high polarising efficiency (P~0.99) over a wide wavelength range (2 to 6.5 Å). The Drabkin spin flipper reverses the neutron beam polarisation direction over the whole wavelength band; with a wavelength dependent efficiency which decreases from 98% at 2Å to 87% at 6.5 Å (15)

For a conventional polarised neutron reflection measurement the intensities for the spin parallel (flipper off) and spin antiparallel (flipper on) cases are given by,

$$I_{off} = \tfrac{1}{2} I_0 k \quad [(1+P)\ R_+ + (1 - P) R_-] \qquad - (1)$$

$$I_{on} = \tfrac{1}{2} I_0 k \{[(1 + P) (1 - f) R_+ + (1 - P) f R_+]$$

$$+ [(1 + P) fR_- + (1 - P) (1- f) R_-] \qquad - (2)$$

where I_0 is the incident beam intensity and k is a constant which includes instrumental factors such as polariser reflectivity and detector efficiency.

For perfect polariser and spin flipper efficiences I_{off} and I_{on} give an accurate estimate of R_+ and R_-. For efficiencies less than unity the measured intensities have a finite contribution from the opposite spin state; table 1 gives a indication of the level of the contamination in I_{off} and I_{on} for a polarising efficiency of 0.98 and a range of spin flipper efficiencies.

TABLE 1 Relative R_+,R_- contributions to I_{off}, I_{on} for P=0.98 and f in the range 1.0 to 0.85

	I_{off}		I_{on}	
Spin flipper efficiency, f.	R_+	R_-	R_+	R_-
1.0	0.99	0.02	0.01	0.99
0.95	0.99	0.02	0.06	0.94
0.90	0.99	0.02	0.11	0.90
0.85	0.99	0.02	0.15	0.84

It is clear from table 1 that the spin flipper efficiency does not need to vary from unity by very much before the measurement of R_- (using the I_{on} intensity) contains a significant contribution from R_+. For the measurement of large flipping ratios it is essential to have an accurate estimate of P, and f for a good evaluation of R_-; and this will be discussed in more detail in a later section.

The addition of a second spin flipper and polariser (analyser) allows the spin direction after reflection by the sample to be analysed; and so provides polarisation analysis. There are now four experimentally measureable intensities compared to two in equations (1 and 2) depending on the state of both spin-flippers,

$$I_{off-off} = \tfrac{1}{4} I_o k \{(1+P1)(1+P2)R_+ + (1-P1)(1-P2)R_-\} \quad — (3)$$

$$I_{off-on} = \tfrac{1}{4} Io\, k \{(1+P1)(1+P2)(1-f2)R_+ + (1+P1)(1-P2)f2\,R_+$$
$$+ (1-P1)(1+P2)f2\,R_- + (1-P1)(1-P2)(1-f2)R_-\} \quad — (4)$$

$$I_{on-off} = \tfrac{1}{4} I_o k \{(1+P1)(1+P2)(1-f1)R_+ + (1-P1)(1+P2)f1\,R_+$$
$$+ (1+P1)(1-P2)f1\,R_- + (1-P1)(1-P2)(1-f1)R_-\} \quad — (5)$$

$$I_{on-on} = \tfrac{1}{4} I_o k \{(1+P1)(1+P2)(1-f1)(1-f2)R_+ +$$
$$(1-P1)(1+P2)f1(1-f2)R_+ + (1+P1)(1-P2)(1-f1)f2\,R_+$$
$$+ (1-P1)(1-P2)\, f1f2\, R_+ + (1+P1)(1+P2)f1f2\,R_-$$
$$+ (1-P1)(1+P2)(1-f1)f2\,R_- + (1+P1)(1-P2)f1(1-f2)R_-$$
$$+ (1-P1)(1-P2)(1-f1)(1-f2)R_-\} \quad — (6)$$

If we assume that P1 = P2 = P and f1 = f2 = f then equations 3-6 simplify to;

$$I_{off-off} = \tfrac{1}{4} Io\, k [(1+P)^2 R_+ + (1-P)^2 R_-] \quad - (7)$$

$$I_{off-on} = \tfrac{1}{4} Io\, k [(1+P)(1+P(1-2f))R_+ + (1-P)(1-P(1-2f))R_-] \quad - (8)$$

$$I_{on-off} = \tfrac{1}{4} Io\, k [(1+P)(1+P(1-2f))R_+ + (1-P)(1-P(1-2f))R_-] \quad - (9)$$

$$I_{on-on} = \tfrac{1}{4} Io\, k [(1+P(1-2f))^2 R_+ + (1-P(1-2f))^2 R_-] \quad - (10)$$

In principle equations 3-6 can be used to determine Pl,P2,f1 and f2. However, in normal circumstances P1 ~ P2 and f1 ~ f2; from equations 8 and 9 I_{off-on} and I_{on-off} are almost identical, and it will be difficult to get an accurate determation of the four efficiency factors. An inspection of equations 7 - 10 show that the addition of polarisation analysis does provide a significant increase in the discrimation against the "unwanted" spin state and $I_{off-off}$ and I_{on-on} provide a much improved representation of R_+ and R_- for typical values of P and f, see table 2.

TABLE 2 Relative R_+, R_- contributions to $I_{off-off}$ and I_{on-on} for,

TABLE 2 Relative R_+, R_- contributions to $I_{off-off}$ and I_{on-on} for,

(a) P=0.98 and f in the range 1.0 to 0.85

	$I_{off-off}$		I_{on-on}	
f	**R+**	**R-**	**R+**	**R-**
1.0	0.98	0.0001	0.0001	0.98
0.95	0.98	0.0001	0.0035	0.98
0.90	0.98	0.0001	0.012	0.80
0.85	0.98	0.0001	0.025	0.71

(b) f=0.9, and P in the range 0.99 to 0.90

	$I_{off-off}$		I_{on-on}	
P	R_+	R_-	R_+	R_-
0.99	0.99	0.000025	0.01	0.80
0.98	0.98	0.0001	0.012	0.80
0.95	0.95	0.0006	0.012	0.77
0.90	0.90	0.0025	0.02	0.74

The contribution of R_- to $I_{off-off}$ is small; but shows a strong dependance on P. Whereas I_{on-on} is dominated by the contribution from R-, and exibits a weak dependence on both P and f. The R_+ contribution to I_{on-on} is by contrast small, but has a strong dependance on f.

Experimental Details

The polarised mode of the CRISP reflectometer has been described in detail elsewhere (8) and the measurements presented in this paper have been made with an identical Drabkin non-adiabatic spin flipper and cobalt-titanium polarising supermirror placed after the sample position.

The measurements have been made with the polariser and analyser supermirrors at angles of 0.35°; giving a maximum usable wavelength range of 2.0 to 6.5Å. The measured counts have been corrected for background, detector efficiency and normalised to the incident spectral shape and absolute run time (using a low efficiency monitor immediately before the sample) and designated I_{off} and I_{on} for measurements without polarisation analyisis, and $I_{off-off}$, I_{on-on}, I_{off-on} and I_{on-off} for measurements with polarisation analysis.

Measurements have been made on a 1800Å nickel film deposited on a glass optical flat at a glancing angle of 0.5° and in an applied field of 2.6 kilogauss

For measurements with polarisation analysis implemented the standard nomalisation procedures described above do not take into account the reflectivity of the analysing mirror. A measurement of $I_{off\text{-}off}$ without a sample is used as an estimate of the response function of the analyser.

Experimental Results

Figure 2 shows the normalised intensity $I_{off\text{-}off}$, $I_{on\text{-}on}$, $I_{off\text{-}on}$ and $I_{on\text{-}off}$ with no sample. The similarity of $I_{off\text{-}off}$ and $I_{on\text{-}on}$ suggest (see equations 7-10) that P and f are close to unity, and the similarity of $I_{on\text{-}off}$ and $I_{off\text{-}on}$ indicating that P1~P2 and f1~f2. In subsequent analysis $I_{off\text{-}off}$ will be used as a estimate of the analyser reflectivity.

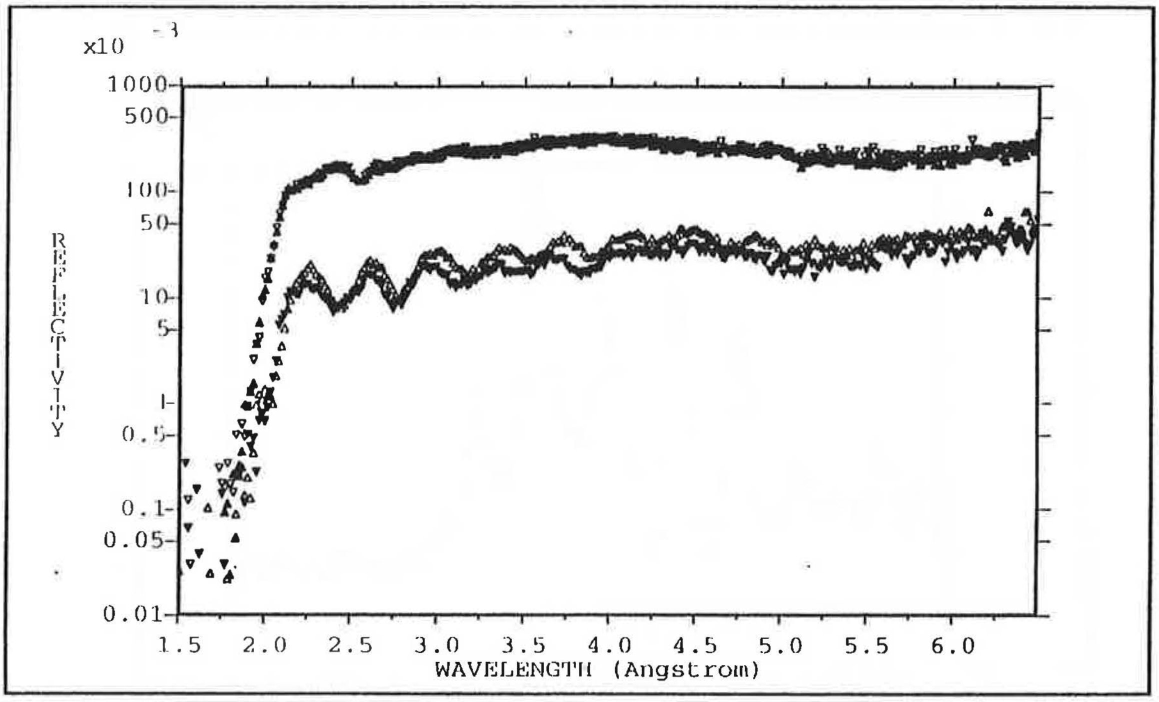
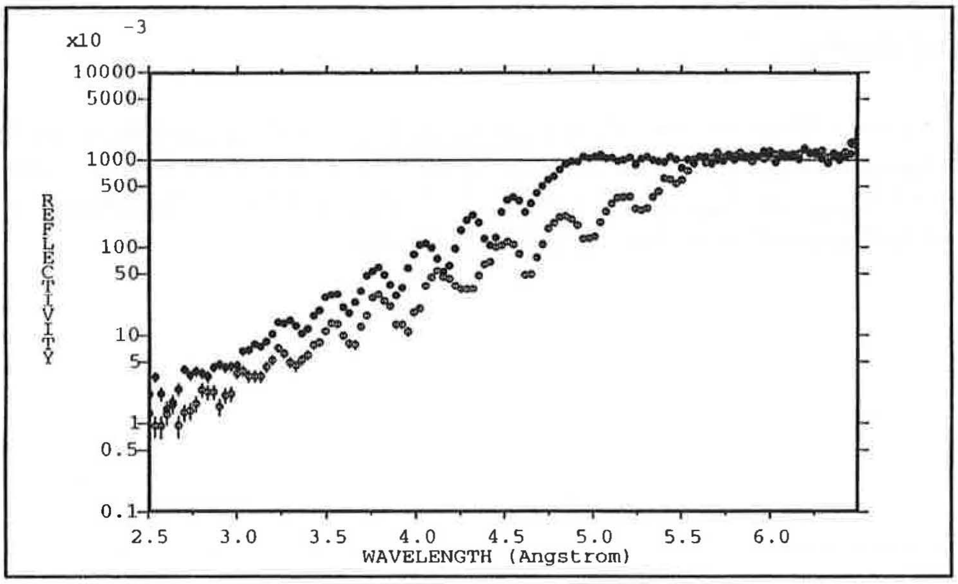


Figure 2: Normalised intensities $I_{off\text{-}off}$ (▽) $I_{on\text{-}on}$ (▲) and $I_{off\text{-}on}$ (Δ) and $I_{on\text{-}off}$ (▼) for no sample

Figure 3 shows the flipper off (I_{off}) and flipper on (I_{on}) reflectivities for a 1800Å nickel film measured without polarisation and with no corrections for P and f applied.



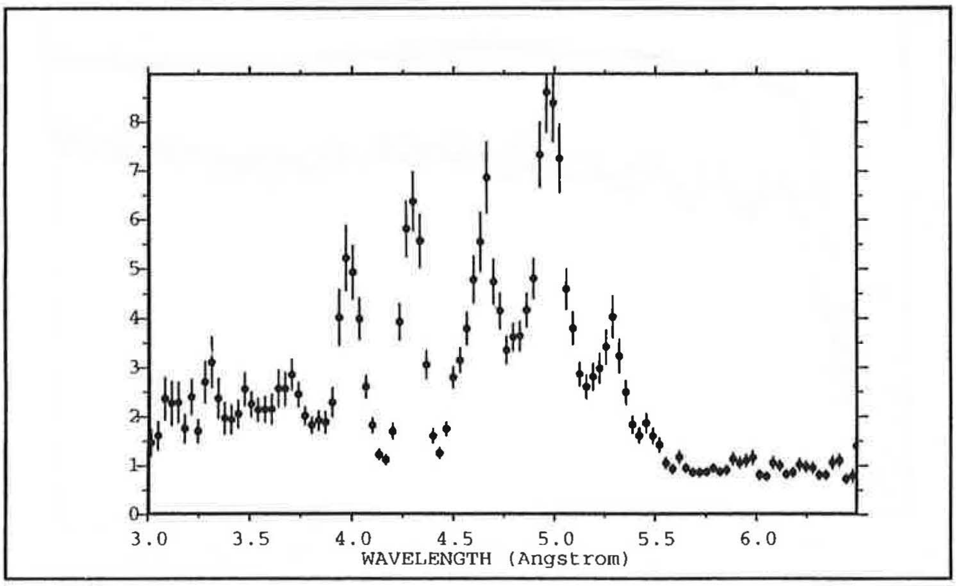
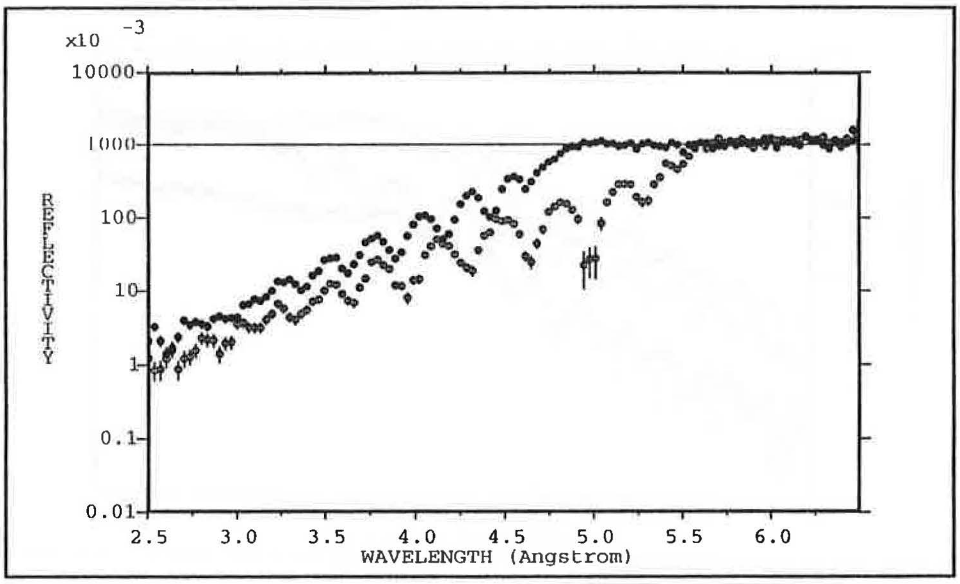


Figure (3a) Normalised reflectivity I_{off} (•) and I_{on} (o) for a 1800 Å nickel film at $\theta = 0.5°$ and in a field of 2.6 kgauss

Figure (3b) Reflectivity ratio I_{off}/I_{on} for data in (3a).

Figure 4 shows the same measured spin dependent reflectivities; but with corrections made for P and f following the proceedure described reference. Although P and f have been determined quite accurately using the "shim method" (15) the R_- reflectivity, in the region of 5Å, is clearly being overcorrected. This provides a clear demonstration of the need for a very good estimate of P and f in order to correct data where the "flipping ratio" (R_+/R_-) is large.



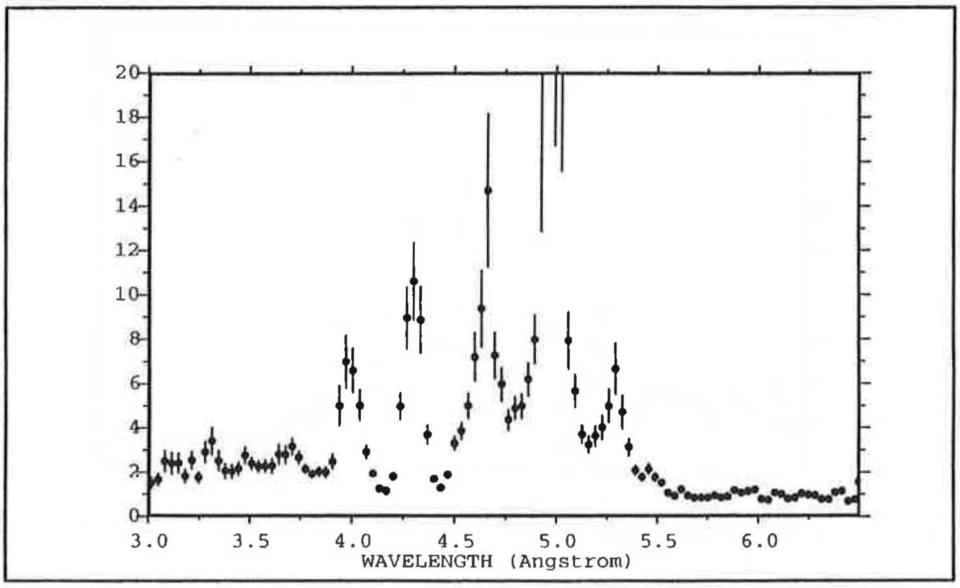
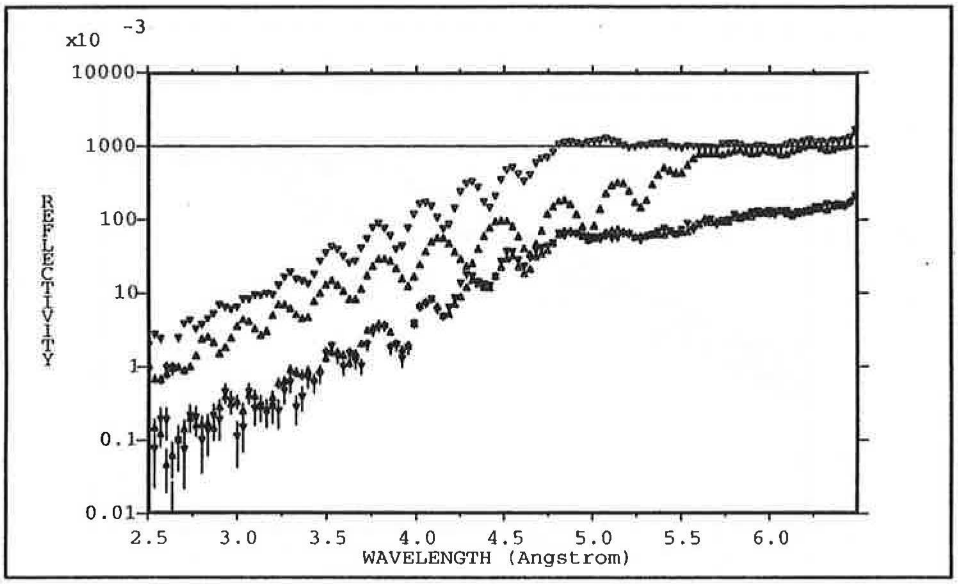


Figure 4 a, b as figure 3, but with data corrected for P and f.

Figure 5 shows the normalised intensities $I_{off\text{-}off}$, $I_{on\text{-}on}$, $I_{off\text{-}on}$ and $I_{on\text{-}off}$ for a 1800Å nickel film at $\theta = 0.5°$ and in a field of 2.6 kgauss. This data has not been corrected for the analyser reflectivity, and hence $I_{off\text{-}off}$, $I_{on\text{-}on}$ do not reflect correctly the R_+ and R_- reflectivities. The ratio does however now give an accurate representation of R_+/R_- (see figure 5b). This should be compared with figure 3b and figure 4b, and clearly provides a much improved estimate of R_+/R_- in the region of its maximum value.



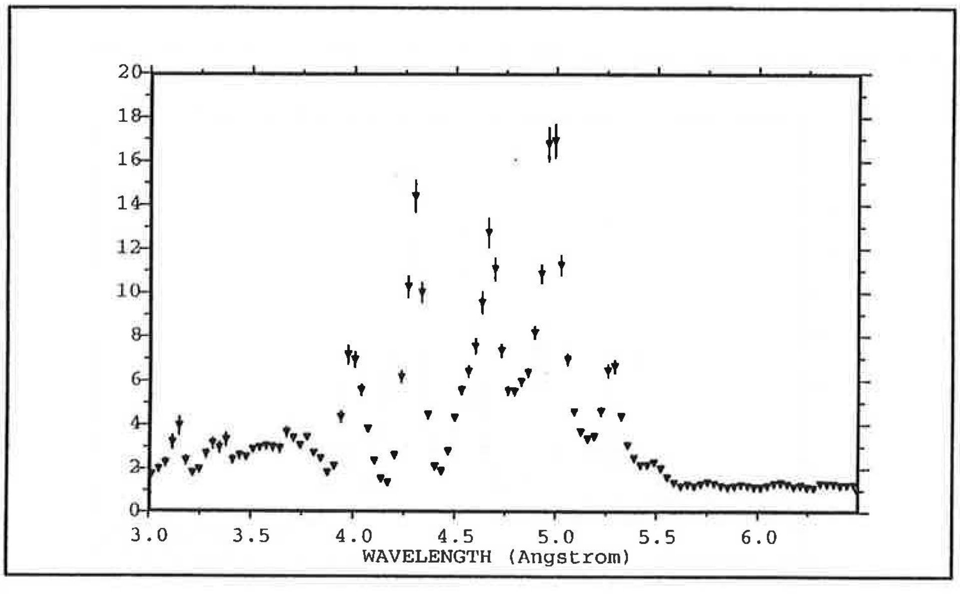


Figure 5 (a) Normalised reflections $I_{off\text{-}off}$ (∇), $I_{on\text{-}on}$ (▲), $I_{off\text{-}on}$ (Δ) and $I_{on\text{-}off}$ (▼) for a 1800 Å nickel film on glass at θ=0.5° and in a field of 2.6kgauss.

(b) Intensity ratio $I_{off\text{-}off}/I_{on\text{-}on}$ from (a)

The normalised intensities $I_{off\text{-}off}$ and $I_{on}\text{-}I_{on}$ shown in figure 6 have been corrected for the analyser reflectivity and provide an accurate measured of R_+, R_-.

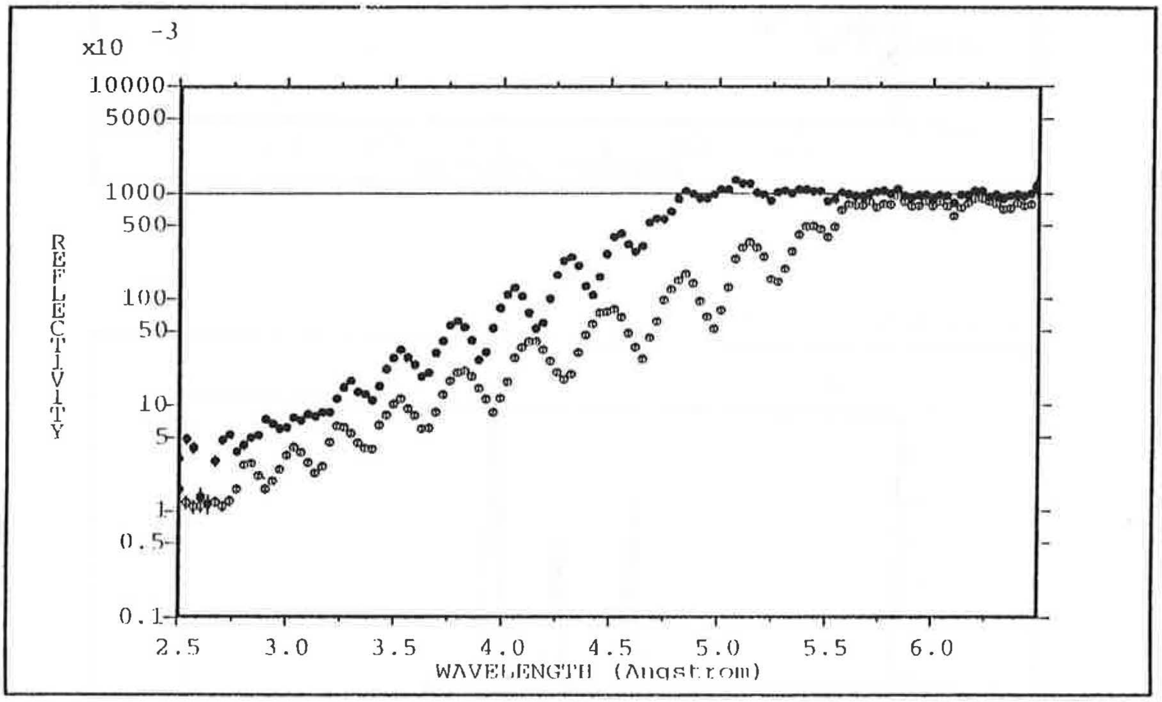
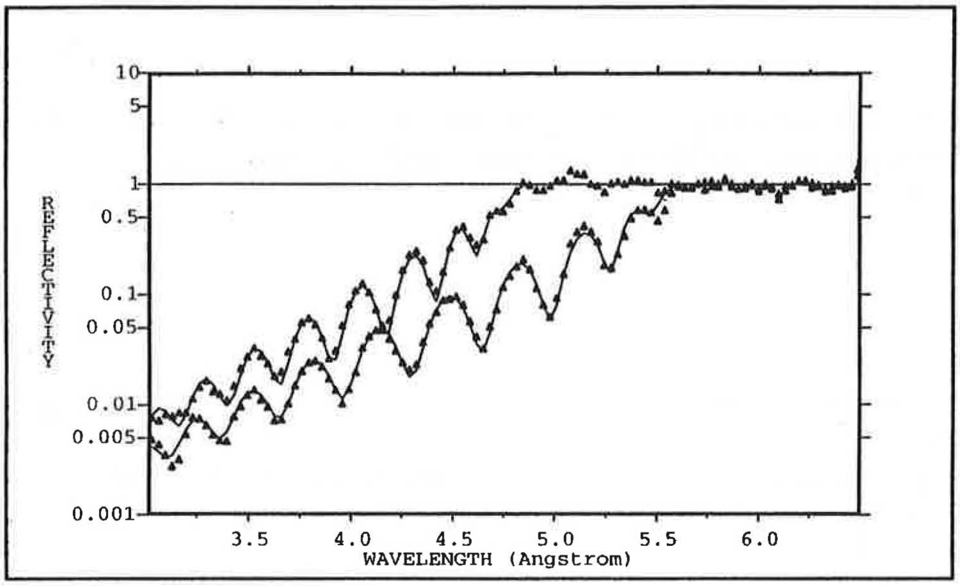


Figure 6 As figure 5(a) but with data corrected for the analyser reflectivity.

We have made a detailed analysis of the corrected data, and compared with earlier work (8). A least squares fit to a single uniform magnetised film is shown in figure 7, and the fitted parameters are summarised in table 3.



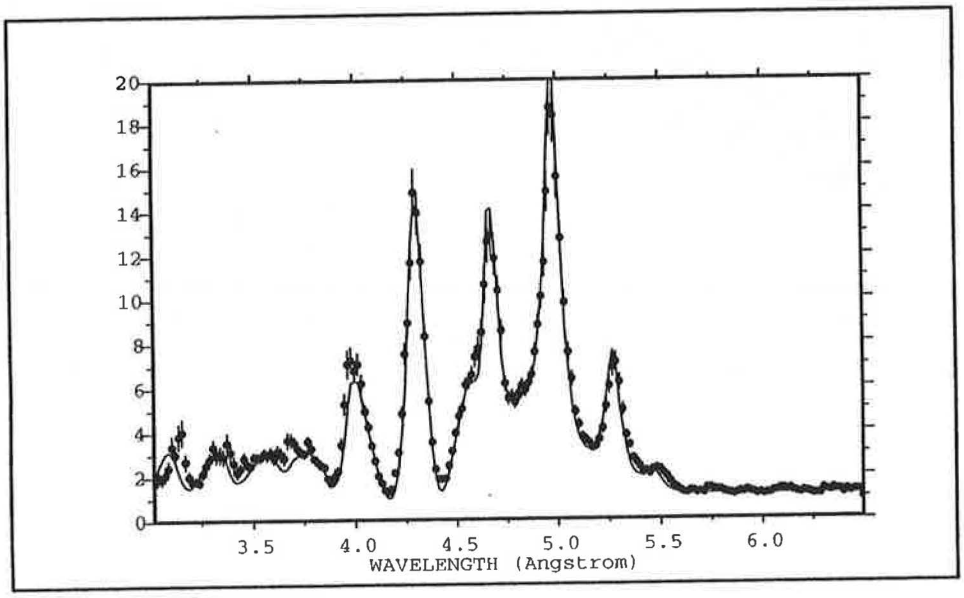


Figure 7 (a) R_+ (Δ) and R-(Δ) for 1800Å nickel film on glass at θ=0-5° and in a field of 2.6kgauss

(b) R_+/R_- , solid lines are the model fit for the parameters in table 3.

The sample was previously measured in 1988 and it is interesting to compare the results to establish the role or the ageing of the film. The fitted resolutions (Δθ) of 1.6% is well described by the instrumental resolution, and shows that the contribution from the sample is negligible. The interfacial roughness at the air-film and film-substrate interfaces are in good agreement with the previous data.

Table 3 Model fit parameter for 1800Å nickel film on glass at 0=0.5° and applied field of 2.6 kgauss.

	Film Thickness d (Å)	Scattering Length density (Å^{-2} x 10^{-6})	z1	z1	$\Delta\theta$
R+	1931	10.34$^{(1)}$	30	60	1.6
R-	1944	7.75$^{(2)}$	30	60	1.6

(1) N(b + p), (2) N(b - p)

where z1, z2 are the interfacial roughnesses at the air-film and film-substrate interfaces, and $\Delta\theta$ is the convolution of the instrumental resolution and the sample contribution.

The mean value of the film thickness is 1938Å , compared to the original value of 1850Å, (8) a change of some 5% in $\Delta d/d$. From the values of N(b+p) and N(b-p) from R_+ and R_- the nuclear scattering length density is 9.05 x 10^{-6} Å^2 and the magnetic scattering length density is 1.3 x 10^{-6} Å^{-2} From the original data the nuclear density has decreased from 9.4 x 10^{-6} Å^{-2}by ~4%. This is probably associated with an oxidation of the film. However, the data and model fits suggest that the change is uniform throughout the film and not confined to a thin surface layer. This is consistent with the observed thickness change. Furthermore the value of Np gives a moment/atom of 0.538μ_B compared to the saturation value of 0.606μ_B. The sample when originally measured in 1988 gave the saturation magnetisation in a field ~ 2.0kgauss. In an applied field of 2.6kgauss the film is now only 89% saturated.

Summary

We have demonstrated the advantages of polarisation analysis in conventional polarised neutron reflection experiments. It provides a method for the accurate determination of R_+,R_- profiles without the need to know the polariser and spin flipper efficiencies accurately.

Acknowledgements

We wish to thank Peter Phillips for his technical assistance, and Gavin Williams for valuable discussions on the details of polarisation analysis.

References

1. J. Penfold and R.K. Thomas, J Phys: Condensed Matt 2 (1990) 1369

2. T.P. Russel, Materials Science Reports 5 (1990) 171

3. J.B. Hayter, J. Penfold and W.G. Williams, J Phys E 11 (1978) 454

4. G.P. Felcher Phys Rev B24 (1981) 1995

5. J.A.C. Bland, A.D.Johnson, R.D. Bateson, S.J. Blundell, H.J. Lauter, C. Shackleton and J. Penfold J. Mag and Mag Mat 93 513 (1991)

6. G.P. Felcher, R. Felici, R.T. Kampwirth and K.E. Gray J Appl Phys 57 (1985) 3789

7. S.S.P. Parkin, R. Sigsbee, R. Felici, G.P. Felcher (1985) appl phys 57 3771

8. R. Felici, J. Penfold, R.C. Ward and W.G. Williams Appl Phys A45 (1988) 174

9. G.P. Felcher, R.D. Hilleke, R.K. Crawford, J. Haumann, R. Kleb, G. Ostrowski, Rev Sci Inst 58 609 (1987)

10. R.M. Moon, T. Riste and N.C. Koehler, (1969) Phys Rev 181 920

11. R.M. Moon, W.C. Koehler, J.W. Cable and R. Child, Phys Rev (1972) 65 997

12. J.A.C. Bland, R. Bateson and G. Hird, J Phys: Condens Matt 1 (1989) 4399

13. S.J. Blundell and J.A.C. Bland; Phys Rev B 46 (1992) 3391

14. T.J.L. Jones and W.G. Williams; RL-77-079

15. R. Felici, J. Penfold, R.C. Ward and W.G. Williams; RAL-87-025 (1987)

16. W.H. Kraan,; M The Rekveldt, and PTPor, Nucl Inst and Meth A300 (1991) 35

17. F. Mezei; Z Physik, 255 146 (1972)

18. See references in ref 15.