

Presented at the International Conf.
on Neutron Scattering in the 90s.
14-18 January 1985, Jülich, W. Germany
(Invited)

BNL 36077

MAGNETIC SCATTERING AND POLARIZED NEUTRONS

CONF-850110--6

by

BNL--36077

G. Shirane and C. F. Majkrzak
Brookhaven National Laboratory
Upton, New York 11973

DE85 009823

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MAGNETIC SCATTERING AND POLARIZED NEUTRONS

ABSTRACT

The last few years have witnessed considerable progress in the use of polarized neutron beam techniques for the study of condensed matter physics. Among the methods actively pursued at present are neutron spin echo as pioneered by Mazel for ultra high resolution and the energy integrated magnetic scattering measurement technique developed by Brown and Ziebeck. We have concentrated on a medium resolution range utilizing 5 to 100 meV polarized neutrons. In this review we discuss recent work at Brookhaven on the development of the triple axis neutron scattering technique with polarization analysis. We have now reached the stage where quantitative characterization, in absolute units, can be done for a wide range of energy and momentum transfers in paramagnetic scattering. We will discuss some examples of recent inelastic measurements on 3d ferromagnets as well as diffraction studies of multilayer thin film structures.

INTRODUCTION

The polarized beam technique has been widely utilized for the study of spin density distributions in ferromagnets, following the pioneering work of Shull and Nathans¹ in the 1950's. This technique requires only a double axis configuration, without analyzer, and the magnetic scattering length p is scaled to the nuclear length b by the flipping ratio R :

$$R = \frac{(b + p)^2}{(b - p)^2} \quad (1)$$

For this purpose the flipper F (see Fig. 1) is placed between the monochromator M and the sample S . Then in 1969, the classic paper by Moon, Riste and Koehler² appeared which advocated the use of polarization analysis after scattering. In particular, when a magnetic field \vec{H} is applied parallel (HF) to the scattering vector \vec{Q} , then all magnetic scattering is spin-flip. In the configuration shown in Fig. 1, this scattering appears in the flipper ON channel.

This flipper ON channel also contains the nuclear spin incoherent scattering (NSI) and all of the background. The straightforward way to eliminate these unwanted components and to obtain the magnetic scattering intensity I_M is to take the difference between the intensity I for HF and that for a vertical field (VF), both with flipper ON (see Table 1):

$$I(HF - VF) = I_M/2.$$

(2)

This is the method used extensively by Ziebeck, Brown and their collaborators⁴ in a series of pioneering studies of magnetic scattering from 3d metals and compounds. They used, intentionally, very broad energy resolution so that the analyzer essentially integrates over ω giving $S(Q)$ directly. The measured intensities were put on an absolute scale by comparison with powder intensities. Among the magnetic materials which have been studied are Fe, Ni, and MnSi.

Recent work^{4,5} at Brookhaven has focused on the development of a polarized beam spectrometer with sufficient energy resolution to give direct information about $S(Q,\omega)$. So far we have centered our effort in the neutron energy range between 13 and 130 meV, with Heusler monochromator and analyzers. For future experiments at lower neutron energies, an extensive effort has been made to develop multilayer polarizers⁵.

The use of a Drabkin⁶ energy-dependent flipper in conjunction with polarizing supermirrors or multilayers makes it possible to decouple beam angular divergence from the energy resolution. Such a flipper has now been built and successfully tested at Brookhaven and is expected to be used routinely in the near future.

So far we have concerned ourselves with the use of polarized neutrons for studies of spin dependent cross sections. However, in 1972 Mezei⁷ invented a novel application of a polarized beam, namely, neutron spin echo. This gives an extremely high energy resolution by measuring changes in the modulation of the neutron beam polarization caused by inelastic scattering. So polarized beams can be used to perform spin dependent measurements as well as to obtain ultrahigh resolution in non-magnetic inelastic scattering.

II. POLARIZATION ANALYSIS

Fig. 1 depicts our standard triple axis spectrometer converted into one utilizing polarized incident beams as well as polarization analysis. This work was carried out as part of the U.S. Japan collaboration on neutron scattering and a few key components, such as the Sm-Co permanent magnets for the analyzer, were supplied by the Japanese collaborators⁸. Large Heusler crystals, in the (111) transmission geometry, were kindly provided by the Institut Laue-Langevin⁹. Among the many technical problems to be solved is the production of polarized neutron beams with adequate intensity. In fact,

this is the principal reason why the powerful techniques of polarization analysis has not yet realized its full potential. For energy range between 13 and 60 meV, we are now down about a factor of 20 in intensity using a fully polarized configuration as compared with our focused pyrolytic graphite triple axis instrument. This has allowed us to study several interesting magnetic systems.

Fig. 2 shows one of our earlier studies on spin waves in solid oxygen¹⁰. The spin-flip scattering is mainly of magnetic origin and characteristic of antiferromagnetic spin waves, in their powder averaged configuration. The non-spin-flip scattering is mainly nuclear consisting of Bragg peaks and phonons. The next example we chose is the recent study of paramagnetic scattering in pure Fe¹¹. Fig. 3(b) demonstrates again the clear separation of magnetic (ON) and nuclear (OFF) cross sections. Fig. 3(c) shows the "difference" data given by Eq.(2), where a magnetic field of 20 Oe was applied parallel and perpendicular to the scattering vector to separate out the magnetic scattering, thereby eliminating the background subtraction problem. This type of measurement sheds light¹² on the puzzle of propagating spin waves above T_c.

Encouraged by these successes we have very recently investigated¹³ the weak magnetic scattering from YFe at 1200K. The results shown in Fig. 4 confirm the ferromagnetic correlations reported in recent publications¹⁴. In addition, our study has provided direct information on the scattering function S(Q,ω) and the unusual Q-dependence of the diffusive line width. There are now a large number of interesting magnetic materials which appear within the technical capability of our spectrometer; these include heavy fermion super conductors, UPt₃ and UBe₁₃, and weak ferromagnets such as MnSi and Ni₃Al.

So far we have given examples of "bulk" magnetic scattering. There is the new and exciting area of study of layered magnets.

III. POLARIZED NEUTRON DIFFRACTION FROM THIN FILM MULTIBILAYERS

Considerable theoretical and experimental efforts to understand the electronic and magnetic structures of surfaces and interfaces have been undertaken in recent years^{15,16}. Nevertheless, theories describing such fundamental physical phenomena as the critical behavior at the surface or interface of a ferromagnet¹⁷⁻¹⁹ have remained largely untested although

a number of measurements of the magnetization at an interface in thin film multibilayer structures by polarized neutron diffraction have been reviewed by Endch²⁰.

Polarized neutron scattering is a powerful technique for studying such problems. As an example, we will discuss the investigation of an Fe-Ge thin film multibilayer structure²¹. The multilayers were prepared by radio frequency sputter deposition of alternating thin films of Fe and Ge onto a glass substrate. X-ray diffraction scans with the scattering vector \vec{Q} parallel and then perpendicular to the plane of the substrate reveal that the Ge films are amorphous while the Fe films are "textured", that is an Fe film is made up of microcrystallites each with a $[110]$ direction normal to the plane of the substrate but with orthogonal directions in the plane randomly rotated about the normal $[110]$ axis. The dimension of an Fe microcrystallite parallel to the plane of the substrate is of the order of 100\AA . A rocking curve at a fixed scattering angle corresponding to the Fe(110) atomic plane spacing shows that the $[110]$ directions normal to the plane of the substrate have an angular distribution with a full width at half maximum of several degrees. Thus, a

diffraction scan as a function of $|\vec{Q}|$ and with the direction \vec{Q} perpendicular to the substrate plane shows structure not only at low scattering angles where the principal diffraction peaks occur at values of $Q = n 2\pi/D$ where n is an integer and D is the thickness of a single Fe-Ge bilayer, but also at high angles with peaks at multiples of $2\pi/d$ where d is the Fe(110) plane spacing.

If a beam of polarized neutrons of intensity $I_{0\pm}$ (where \pm denotes the spin eigenstate of the beam) is incident on a thin film multibilayer where one of the film materials is ferromagnetic with its magnetization aligned perpendicular to Q , then two scattered intensities I_{\pm} and the corresponding "flipping ratio" $R \equiv I_{\pm+}/I_{\pm-}$ can be measured. According to the kinematic theory of diffraction (where extinction is taken to be negligible), if Q is perpendicular to the plane of the multilayer films (and fluctuations in bilayer thickness are neglected for now), then

$$I_{\pm}/I_0 = \left(\frac{4\pi}{Q}\right)^2 \left| \sum_{j=1}^N (f_{N_j} \pm f_{M_j}) e^{iQZ_j} \right|^2 \cdot \left| \frac{\sin(MQD/2)}{\sin(QD/2)} \right|^2 \quad (3)$$

where f_{Nj} and f_{Mj} are the nuclear and magnetic scattering length densities, respectively, M is the number of bilayers and Z_k is the position of the j th atomic plane within a bilayer (assuming the bilayer is composed of N discrete, parallel atomic planes). By measuring the integrated intensities about each $Q = m 2\pi/D$ and/or the diffraction profile about $Q = 2\pi/d$ at high angles, it is possible to determine, in principle, the magnetization profile across the thickness of the bilayer. For large enough M , however, extinction effects can become significant, particularly for the first order superlattice peak and a dynamical theory of diffraction must be applied. For sufficiently small values of Q where mirror reflection and refraction effects are important, the neutron is treated as a plane wave incident upon a layered but continuous medium.

Polarized neutron diffraction measurements were made on a number of Fe-Ge multilayer samples. In Fig. 5 the scattered intensity is plotted versus Q for a multilayer consisting of 509 thin film bilayers of Fe and Ge. \vec{Q} is perpendicular to the film planes and $D = 108\text{\AA}$. Seven diffraction peaks with periodicity $Q = m 2\pi/D$ are shown beginning with $m = 1$. The "ON" ("OFF") data points correspond to incident neutrons in the "+" ("-") spin eigenstate. The fact that the flipping ratio R is not always >1 implies that the magnetization is not constant across the thickness of the bilayer. Both high- and low-angle polarized neutron diffraction data were fitted to various models for the structure factor. A consistent fit is obtained for a model where each bilayer is composed of three distinct regions; the first of pure amorphous Ge, the second of a random FeGe alloy with the same structure and orientation as the pure bcc Fe which constitutes the third region. An Fe moment in the pure Fe region has the full "bulk" moment while an Fe atom in the FeGe alloy region has a diminished moment depending upon the concentration of Ge about the Fe atom's location in the layer. The Ge concentration is highest next to the pure Ge region and decreases to zero adjacent to the pure Fe region. It was therefore concluded that although a fraction of the Fe atoms have a moment whose magnitude is reduced from that in the bulk, this reduction is due primarily to the interdiffusion of Fe and Ge across the interface of the thin films which occurs during the deposition process. As shown in Ref. 21, the flipping ratios of some of the higher-order superlattice peaks can change by an order of magnitude or more for relatively small changes in the Fe moment in the interfacial region. Thus it is shown that polarized neutron diffraction can be an extremely sensitive technique for measuring such magnetization profiles.

Similar studies are now underway involving multilayer structures consisting of ferromagnetic and superconducting thin films (e.g. Ni and V or Ni and Nb) of various thicknesses. The idea is to investigate any interaction between the ferromagnetic and superconducting states which are in spatial proximity to one another.

ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the contributions of our fellow collaborators at Brookhaven, namely, J. D. Axe, P. Böni, L. Passell, O. Steinsvoll and J. Wicksted. Part of this research was supported by the U.S.-Japan Collaboration in Neutron Scattering. Research at Brookhaven was also supported by the Division of Materials Sciences, U.S. Department of Energy under contract No. DE-AC02-76CH00016.

REFERENCES

- [1] See for example, R. Nathans, C. G. Shull, G. Shirane and A. Andressen, J. Phys. Chem. Solids 10, (1959) 138.
- [2] R. M. Moon, T. Riste and W. C. Koehler, Phys. Rev. 181, (1969) 920.
- [3] K. R. A. Ziebeck and P. J. Brown, J. Phys. F10, (1980) 2015.
- [4] C. F. Majkrzak and G. Shirane, Jour. de Physique, Colloque C7, (1982) 215.
- [5] C. F. Majkrzak and L. Passell, Acta Cryst., A40, (1984) in press.
- [6] G. M. Drabkin, JETP (USSR) 43, (1962) 1107.
- [7] F. Mezel, Z. Physik 255, (1972) 146.
- [8] Y. Ito and H. Yoshizawa have made substantial contribution for many aspects of the set up.
- [9] Courtesy of R. Pynn and A. Freund, Institut Laue-Langevin, Grenoble, France.
- [10] P. W. Stephens, R. J. Birgeneau, C. F. Majkrzak, and G. Shirane, Phys. Rev. B 28, (1983) 452.
- [11] J. Wicksted,
- [12] See a review article by G. Shirane, O. Steinsvoll, Y. J. Uemura and J. P. Wicksted, J. Appl. Phys. 55, (1984) 1887.
- [13] P. Böni, G. Shirane, J. P. Wicksted and C. Stassis, Phys. Rev. B, to be published.
- [14] P. J. Brown, H. Capellman, J. Déportes, D. Givord, and A. Murani, K. R. A. Ziebeck, J. Magn. Magn. Mat. 30, (1983) 335, and preprint.
- [15] A. J. Freeman, T. Jarlborg, H. Krakauer, S. Ohnishi, D. S. Wang, E. Wimmer and M. Weinert, Proc. of the Yamada Conf. VII, Muon Spin Rotation, Shimoda, 1983. Hyperfine Interactions 17-19, (1984) 413.
- [16] C. Rau, J. Magn. and Magn. Mat. 30, (1982) 141.

- [17] T. Wolfram, R. E. Dewames, W. F. Hall and P. W. Palmberg, Surf. Sci. 28, (1971) 45.
- [18] H. W. Diehl, J. Appl. Phys. 53, (1982) 7914.
- [19] K. Binder and D. P. Landau, Phys. Rev. Lett. 52, (1984) 318.
- [20] Y. Endoh, Jour. de Physique 43, (1982) C7-159.
- [21] C. F. Majkrzak, J. D. Axe and P. Böni, J. Appl. Phys., in press.

Figure and Table Captions

- Fig. 1 The schematic on the right represents a triple axis neutron spectrometer where the monochromating crystal M and the analyzer crystal A are also polarizers. α_0 , α_1 , α_2 and α_3 are collimations while S, F, and C denote sample, flipper and detector respectively. As shown, the sample is placed in a horizontal magnetic field (HF) directed along the scattering vector \vec{Q} . The data on the left show magnon cross sections for ^{60}Ni obtained by the technique described in the text.
- Fig. 2 Evidence of antiferromagnetic spin waves (spin-flip scattering) in solid O_2 as obtained using a polarized beam triple axis spectrometer with polarization analysis. After Stephens et al.¹⁰.
- Fig. 3 Measurement of magnetic scattering in Fe using polarization analysis: (b) demonstrates the clear separation of magnetic (ON) and nuclear (OFF) cross sections while (c) is a plot of $I(\text{HF-VF})$ for flipper ON (see Eq. 2). After Wicksted et al.¹¹.
- Fig. 4 Scattering due to ferromagnetic correlations in γ iron. After Boni et al.¹³.
- Fig. 5 Polarized neutron diffraction scan of an Fe-Ge multibilayer with \vec{Q} perpendicular to the film planes and to the magnetization. Seven diffraction peaks with periodicity $Q = n 2\pi/D$ are shown beginning with $n = 1$ ($D = 108\text{\AA}$). The "ON" ("OFF") data points correspond to incident neutrons in the "+" (" - ") spin eigenstate. The fact that the flipping ratio R as defined in the text is not always >1 is indicative of the fact that the magnetization is not constant across the thickness of the bilayer. After Majkrzak et al.²¹

Table I. This table illustrates how various cross sections σ can be obtained by taking the difference of measured scattered intensities with an applied magnetic field parallel and perpendicular to \vec{Q} . σ_{NSI} is the nuclear spin incoherent cross section, σ_{NC} is the nuclear coherent cross section, σ_{NCM}^z is a cross term involving the product of nuclear coherent amplitudes and the z-component of magnetic amplitudes, and σ_{MAG} is the magnetic scattering cross section.

Table 1

Spin Flip	Non Spin Flip
$\vec{H} \parallel \vec{Q} \text{ (HF)} \quad \sigma_{\text{MAG}}^{xx} + \sigma_{\text{MAG}}^{yy} + \frac{2}{3} \sigma_{\text{NSI}}$	$\frac{1}{3} \sigma_{\text{NSI}} + \sigma_{\text{NC}}$
$\vec{H} \perp \vec{Q} \text{ (VF)} \quad \sigma_{\text{MAG}}^{xx} + \frac{2}{3} \sigma_{\text{NSI}}$	$\sigma_{\text{MAG}}^{zz} + \frac{1}{3} \sigma_{\text{NSI}} + \sigma_{\text{NC}} + \sigma_{\text{NCM}}^z$

Table I. This table illustrates how various cross sections σ can be obtained by taking the difference of measured scattered intensities with an applied magnetic field parallel and perpendicular to Q . σ_{NSI} is the nuclear spin incoherent cross section, σ_{NC} is the nuclear coherent cross section, σ_{NCM}^z is a cross term involving the product of

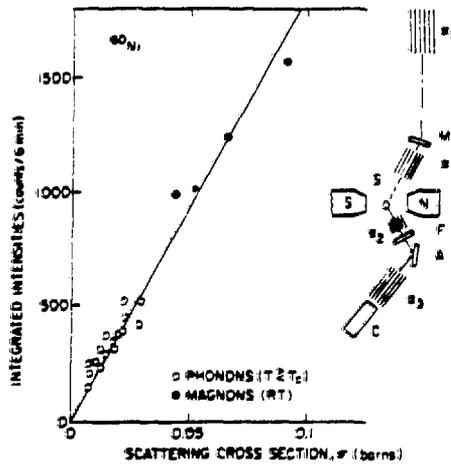


Fig. 1 The schematic on the right represents a triple axis neutron spectrometer where the monochromating crystal M and the analyzer crystal A are also polarizers. a_0 , a_1 , a_2 and a_3 are collimators while S, F, and C denote sample, flipper and detector respectively. As shown, the sample is placed in a horizontal magnetic field (HF) directed along the scattering vector Q. The data on the left show magnon cross sections for ^{60}Ni obtained by the technique described in the text.

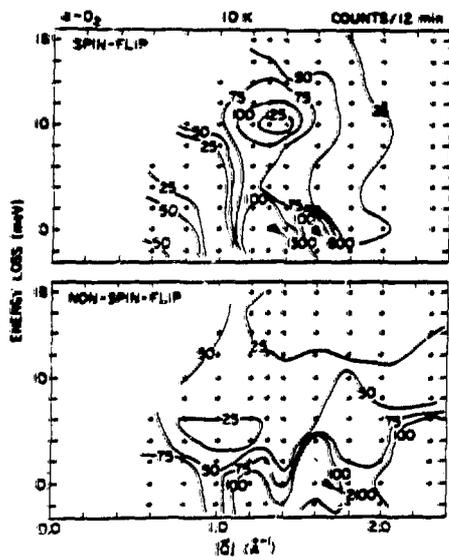


Fig. 2 Evidence of antiferromagnetic spin waves (spin-flip scattering) in solid O_2 as obtained using a polarized beam triple axis spectrometer with polarization analysis. After Stephens et al¹⁰.

REPRODUCED FROM
BEST AVAILABLE COPY

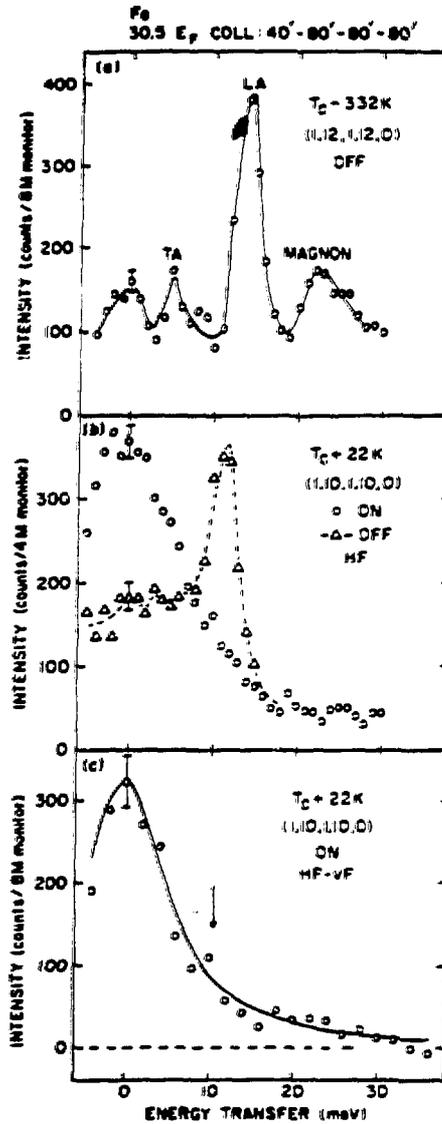


Fig. 3 Measurement of magnetic scattering in Fe using polarization analysis: (b) demonstrates the clear separation of magnetic (ON) and nuclear (OFF) cross sections while (c) is a plot of I (HF-VF) for flipper ON (see Eq. 2). After Wicksted et al.¹¹.

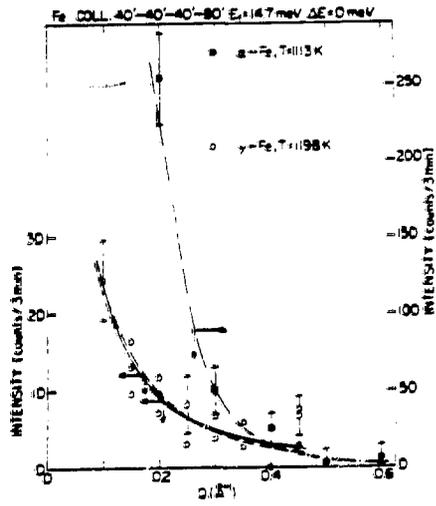


Fig. 4 Scattering due to ferromagnetic correlations in γ iron. After Boni et al.¹³

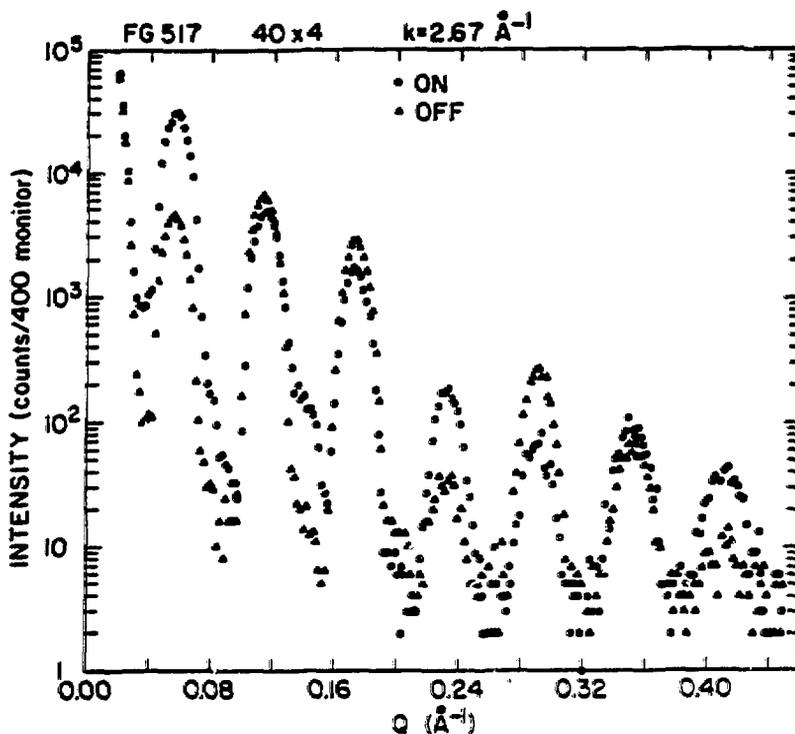


Fig. 5 Polarized neutron diffraction scan of an Fe-Ge multibilayer with \hat{Q} perpendicular to the film planes and to the magnetization. Seven diffraction peaks with periodicity $Q = m 2\pi/D$ are shown beginning with $m = 1$ ($D = 108\text{\AA}$). The "ON" ("OFF") data points correspond to incident neutrons in the "+" ("-") spin eigenstate. The fact that the flipping ratio R as defined in the text is not always >1 is indicative of the fact that the magnetization is not constant across the thickness of the bilayer. After Majkrzak et al.²¹