Polarized Neutron Reflection at LANSCE

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ABSTRACT: The integration of polarized neutron reflection (PNR) with the surface profile reflectometer (SPEAR) at LANSCE is described. To achieve a PNR capability comparable to that of CRISP at ISIS, a Co-Ti super-mirror, a flat-coil spin-flipper, and magnetic guide fields can be introduced into the SPEAR neutron beam line by simple computer command. Instrumentation, particularly with regard to the use of a flat-coil spin-flipper at a pulsed spallation source, and computer software for the optimization of such a spin-flipper and the analysis of polarized neutron reflection data are described. Recently, the ability to measure the reflection of polarized neutrons from thin-film and multilayers samples as they are manufactured *in situ,* was demonstrated. Results from such an experiment are presented.

1. Introduction

During 1992, polarized beam handling equipment was added to SPEAR. The purpose of this equipment is to facilitate studies of magnetic thin-films and multilayers using the polarized-neutron-reflection technique [1]. The motivation for these studies is the need to quantify the magnetization profiles of thin-films and multilayers, and correlate their magnetizations with detailed understandings of their atomic structures. Particularly in multilayer systems like those exhibiting giant-magneto-resistance (GMR), the magnetic coupling of neighboring magnetic layers is sensitive to roughness and inter-diffusion at their interfaces [2]. The attraction of PNR is that the technique can characterize the magnetizations of surfaces and interfaces in thin-films and multilayers *and* determine details about their nuclear structures with a depth resolution typically on the order of 5Å. When this technique is combined with an intense neutron source, such as LANSCE, efficient polarization devices, and an ultra-high vacuum thin-film and multilayer *in situ* fabrication capability, the surfaces and interfaces of thin-film systems can be observed in their pristine state. The magnetic structures of these spatially-limited systems can also be studied at intermediate steps during their manufacture. Not only does this capability facilitate model fitting of complicated structures, but the origin(s) of unusual magnetic properties, such as those which produce spin-flip scattering, can be identified during the fabrication of the multilayer. In §2 of this paper, details about the polarization equipment at SPEAR are described. In §3 an application of this equipment to the study of the magnetization of a thin Fe film on an MgO substrate is discussed, and in the last section, the recently installed fabrication facility in the SPEAR neutron beam line is discussed.

2. Polarization Equipment and its Computer Control

Neutrons with wavelengths ranging from 2 to 8Å are provided by preferentially reflecting one neutron polarization state from Co-Ti polarizing super-mirrors [3]. The super-mirrors are magnetized in a direction perpendicular to the reflection plane by a 1kG field produced by permanent magnets. The polarization of the neutron beam is maintained from the super-mirror (Fig. 1) to the sample position by magnetic guides which provide a magnetic field of 50G in the same direction as that applied to the super-mirror.

In order to measure the spin-up (ω_+, neutron spin aligned parallel to the magnetic field on the polarizer) and spin-down (ω_-) reflectivities of a sample, the neutron spin must be either maintained or flipped relative to the magnetization of the super-mirror. Spin-flipping at LANSCE is obtained by adiabatically precessing the neutron spin through a flat-coil spin-flipper [4]. The neutron spin can be made to precess an odd multiple of π, if the magnetic

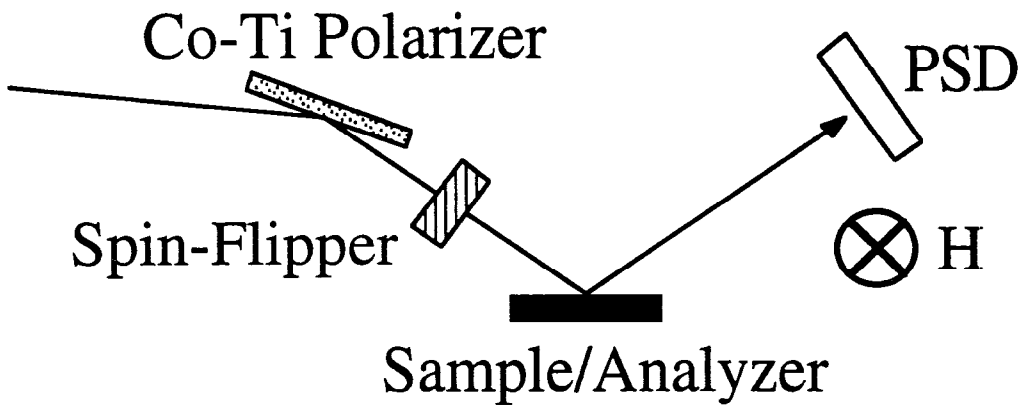

Fig. 1 Side view of the polarization components that can be introduced into the SPEAR beam line. The unpolarized neutron beam becomes polarized after reflection from the Co-Ti polarizer. The polarization state can be changed from spin-up, i.e. aligned with the magnetic field (into the figure), to spin-down by the spin-flipper. A magnetic guide field (not shown) maintains the polarization state of the neutrons from the polarizer to the sample. The spin-up and spin-down reflectivities, ω_+ and ω_-, are measured by a position sensitive detector (PSD).

field within the spin-flipper is:

- normal to the direction of the magnetization of the super-mirror, and
- of the correct strength for the velocity (or wavelength) of the neutron.

In the case of SPEAR, the direction of the magnetic field in the spin-flipper was chosen to lie in the vertical plane and normal to the neutron beam. The field is produced by passing current through a coil (called the flipping coil), which has a rectangular cross-section. Since the spin-flipper lies within the confines of the magnetic guide field, current passing through a second outer coil (called the compensating coil) is needed in order to cancel the magnetic field from the guide; thus, the neutron beam passes through a region with a purely vertical magnetic field, when reversal of the neutron beam polarization is desired.

In order to assure that every neutron precesses equally regardless of its wavelength, the strength of the magnetic field inside the flipping coil must be changed as a function of time so that the integral of magnetic field strength over time is constant for every neutron. This requirement is accomplished by ramping the current through the flipping coil, I_f, as a function of time, t, according to the relation:

$$I_f(t) = \frac{a}{t} + b, \tag{1}$$

The parameter a is related to the precession angle of a neutron (a constant), and b is related to the distance between the spin-flipper and the spallation target. Since the magnetic field due to the magnetic guide is constant with time, only a DC current is required in the compensating coil. Past experience (R.P.) with other flat-coil spin-flippers suggests that optimum performance is achieved when the compensating coil current, I_c, is somewhat time dependent. The relation used to change I_c with time is,

$$I_c(t) = r + st, \tag{2}$$

where r represents the current required to cancel the magnetic field from the magnetic guide, and st is a small perturbation on the order of 10% of r.

The degree to which a spin-flipper is properly "tuned", i.e. the spin-flipper flips the spin of a neutron from spin-up to spin-down, can be determined by measuring the ω_+ and ω_- reflectivities of a second polarizing super-mirror (the analyzer), when it is placed at the sample position. If the polarizing super-mirror and spin-flipper are functioning perfectly,

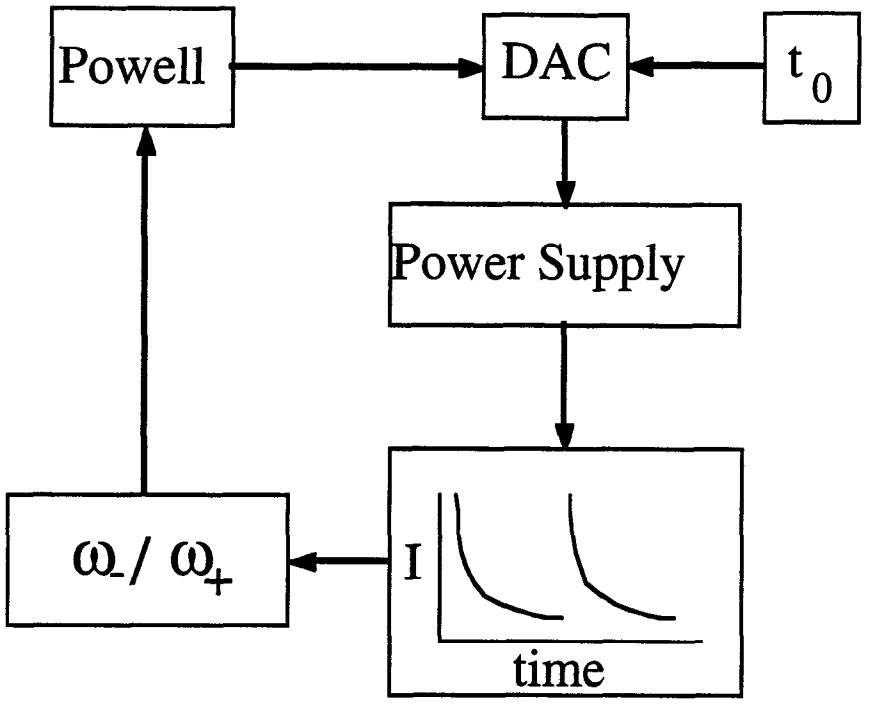

Fig. 2 The procedure for optimizing the flipping efficiency of the spin-flipper consists of loading the coefficients of a polynomial into a digital-to-analog converter (DAC), which drives a power supply to produce a time dependent variation of current through a spin-flipper. The DAC is reset when a new spallation event is detected (every 50ms). The reflectivities ω_+ and ω_- are measured and used to compute the integral of the flipping ratio over time, φ, whose maximum is desired. An algorithm, called "Powell" in the diagram, perturbs the polynomial coefficients until φ^{-1} is minimized (or the integrated flipping ratio is maximized).

then the spin-down reflectivity, ω_-, of the analyzer should be zero. In other words, the ratio ω_+/ω_-, called the flipping ratio, should diverge for all neutrons with wavelengths in the range of 2 to 8Å. The optimum values of *a, b, r* and *s* are those which maximize the flipping ratio. The values of these parameters were determined from the procedure shown in Fig. 2.

This procedure first calculates $I_f(t)$ and $I_c(t)$ for an initial set of guess parameters over a 50ms (the duration between successive spallation events or proton pulses at LANSCE) time range. These values are loaded into a BiRa 12 bit digital-to-analog (DAC) CAMAC module [5], which produces two analog signals (one for each coil). The signals are triggered during every proton pulse and used to drive two Kepco programmable bipolar power supplies [6], which operate in current control mode. The currents pass from the power supplies through the flipping and compensating coils producing a magnetic field inside the spin-flipper that changes with time as prescribed by equations (1) and (2). When properly tuned, every neutron precesses in the changing magnetic field through the same angle so that the polarization of the neutron beam is anti-parallel to the direction of the magnetic field in the guide and that which is applied to the sample (spin-down). When no precession is desired, the DAC produces a constant zero voltage signal which results in no current to the spin-flipper. In this situation the magnetic field inside the spin-flipper is produced by the magnetic guide; thus, the polarization of the neutron beam is the same as that which is reflected from the polarizing super-mirror (spin-up). Since the spin-down neutron reflectivity of the analyzer (or sample) is much smaller than for the spin-up reflectivity, spin-down measurements are accumulated for longer periods of time (typically five times longer) than the spin-up measurements in order to obtain roughly equal statistical quality for both measurements.

After normalizing the ω_+ and ω_- reflectivities to their respective exposures, which are measured by an incident beam monitor, the variation of the flipping ratio with time-of-flight

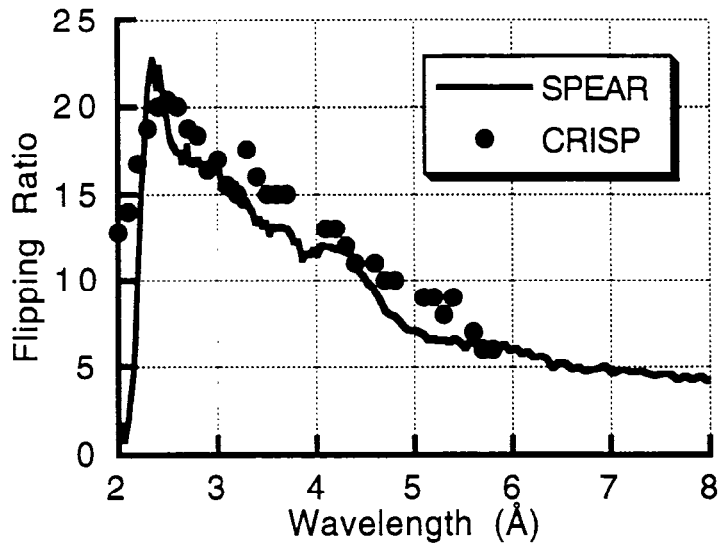

Fig. 3 Flipping ratio plotted versus wavelength as observed with SPEAR (solid curve) and CRISP (•).

(TOF) for a particular set of parameters *a, b, r* and *s*, is calculated. The problem of determining optimal values for *a, b, r* and *s*, is now one of maximizing the integral of the flipping ratio over the time-of-flight of all polarized neutrons (in practice, φ^{-1} is minimized),

$$\varphi(a,b,r,s) = \int \frac{\omega_+}{\omega_-} dt \ . \tag{3}$$

The location of the minimum of φ^{-1} can be efficiently determined by utilizing any number of computational algorithms which are designed to minimize a function of several arguments. The algorithm chosen for use at LANSCE is Powell's Quadratically Convergent Method [7]. While this algorithm does not converge as quickly as those that use information about the gradient of a function to locate its minimum, the Powell search algorithm can be relatively easily constrained. This is a practical consideration, since the current limits of each power supply are chosen to protect the spin-flipper coils from damage. Gradient information used in conjugate gradient and variable metric minimization methods often test parameter sets which result in currents that exceed the limits of the power supplies, consequently, causing the more sophisticated methods to fail.

The Powell procedure involves perturbing *a, b, r* and *s*, along orthogonal directions that eventually lead to the location of an extremum. After each perturbation, ω_+ and ω_- are measured, and a new φ^{-1} , φ'^{-1}, is calculated. The change of φ'^{-1} from φ^{-1} is used to calculate new parameters, *a', b', r'* and *s'*, representing a new set of directions, and the process is repeated until φ'^{-1} differs from φ^{-1} by about 1%. The optimization procedure requires approximately six hours to complete when the neutron source runs reliably at 60μA. Interestingly, the optimal value of *a* that was obtained for SPEAR, corresponded to a precession angle of 3π. This angle was found to yield better flipping ratios than a precession angle of π. A plot of the flipping ratio versus wavelength measured after the optimization routine was completed is shown in Fig. 3 as the solid curve. This flipping ratio is very comparable to that obtained at CRISP (•'s in Fig. 3), which utilizes a Drabkin spin-flipper rather than a flat-coil spin-flipper [8].

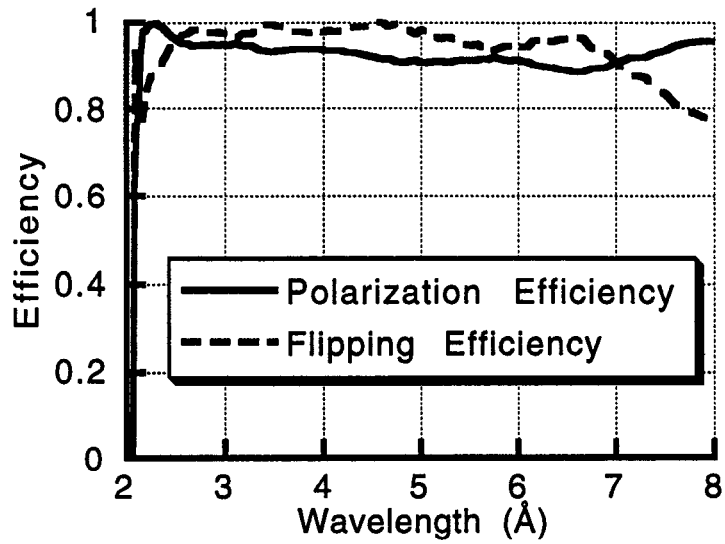

Fig. 4 The variation of polarization (solid curve) and flipping (dashed curve) efficiencies versus wavelength measured for SPEAR.

The flat-coil spin-flipper used at LANSCE is considerably larger than those in use at the ILL. Both are about 12cm by 12cm in cross-sectional area, but the LANSCE version is 2.5cm long while the ILL version is about half as long. A second much shorter (only 0.6cm long) flat-coil spin-flipper was also tested at LANSCE; however, the shorter design obtained flipping ratios only half as large as those shown in Fig. 3. The poorer performance of the shorter flipper might be attributed to an inability to obtain precession angles of 3π, since the power supplies could not supply currents large enough to generate a flipping field that could precess fast neutrons through the larger angle. Based on this test the optimal flat-coil flipper design would seem to be one utilizing a relatively long flight path and high currents to precess neutron spins by at least 3π.

After the spin-flipper was optimally tuned, the polarization and flipping efficiencies of the polarization components were measured in a procedure requiring four different reflection measurements of the analyzer [9]. The measurements included spin-up and spin-down measurements with and without a depolarizing Fe foil (shim) inserted between the polarizing mirror and spin-flipper. By comparing the ratios of these measurements, the polarization and flipping efficiencies, $P(\lambda)$ and $F(\lambda)$, respectively, were obtained (Fig. 4). Since the polarizer and spin-flipper are imperfect, mixtures of polarization states are actually reflected from a sample; therefore, P and F are used to correct the observed reflectivities ω_+ and ω_- to obtain the "true" spin-up and spin-down reflectivities, R_+ and R_-. R_+ and R_- can be compared directly to model calculations. The relationships between R_+ and R_- with the observed reflectivities are given in equation (4).

$$R_{\pm} = \omega_+ \pm \frac{1 \mp P}{2PF} (\omega_+ - \omega_-) \tag{4}$$

3. Application of PNR to Measure the Magnetization Profile of a Thin Fe Film on a MgO Substrate

As an example of using PNR with SPEAR, results from measurements of a thin Fe film on MgO are presented. The thin-film (ca. 267Å thick) was epitaxially grown on a polished $4cm^2$ single-crystal wafer of MgO heated to a temperature of 500°C. The ω_+ and ω_- reflectivities were accumulated after alternating between many spin-up and spin-down

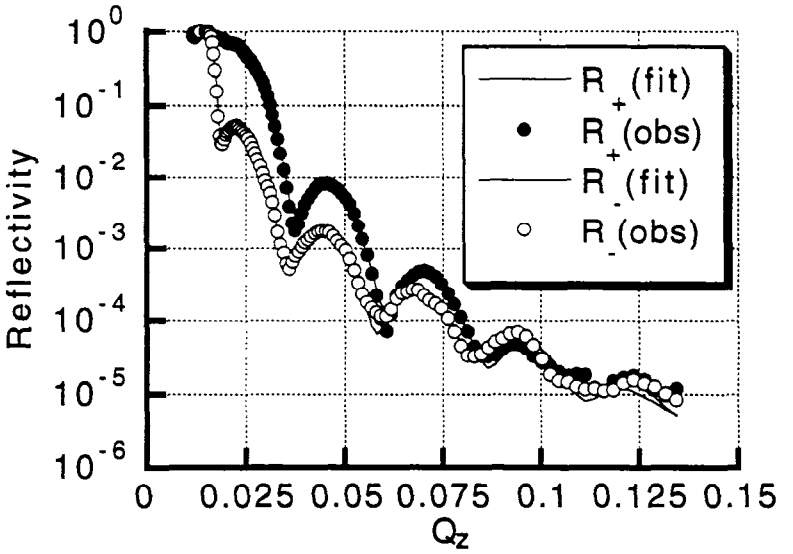

Fig. 5 Polarized neutron reflectivities, R_+ (•) and R_- (○), from an Fe thin-film epitaxially deposited on a single-crystal MgO substrate. The solid curves are calculated reflectivities from a model structure discussed in the text.

measurements— each lasting ca. 5 and 15 minutes, respectively. In order to obtain reflectivities for momentum transfers as large as 0.15Å^{-1} within the polarized wavelength range of 2 to 8Å, the data collection required three measurements at different angles of incidence. Each measurement was corrected for the variations of the incident beam spectrum and polarization and flipping efficiencies to yield R_+ and R_- by software adapted from that which is already used to analyze unpolarized reflection data collected with SPEAR. Additional software was written which concatenates an arbitrary number of smaller reflection measurements to produce reflectivity curves, R_+(•) and R_-(○), like those shown in Fig. 5 for the Fe/MgO sample. The data in this figure required a total of eight hours to collect.

The software package used to reduce data from polarization experiments also affords the user the opportunity to optimize parameters of model structures so as to find a model that best fits both reflectivity curves (spin-up and spin-down) simultaneously. Parameters that can be optimized include: layer thickness, nuclear and magnetic scattering length densities, and interfacial roughness. This software was used in the following analysis to determine the magnetization profile as a function of depth into the Fe on MgO sample.

In the model, the Fe film was represented by three layers, consisting of a thin (25Å thick) native oxide (γ-Fe_2O_3, a ferrimagnet), an Fe interior, and a phase boundary region with a reduced density compared to that of the film interior. The oxide layer is motivated by Mössbauer studies of similarly grown Fe single-crystals which determined the stochiometry, phase and thickness of the native oxide [10]. The thickness of the oxide layer determined in the present study is in good agreement with that determined by the Mössbauer work. The magnetic moments of Fe atoms in the oxide ($1.6\mu_B$) and Fe interior ($1.8\mu_B$) are both somewhat smaller than those measured for bulk materials (cf. $2.3\mu_B$ and $2.2\mu_B$, respectively [11]). These reductions may be due to a property of the thin-film geometry or an inability to fully magnetize the sample in the 1.17kG field. If the sample had not been fully magnetized, then some spin-flip scattering from the sample may have occurred. Since the current instrument can not analyze the polarization state of the scattered radiation, spin-flip scattering is not detected and assumed not to have occurred. During 1993, polarization analysis equipment will be installed so that spin-flip scattering can be detected when it occurs.

A separate layer representing the interface between the Fe film and MgO substrate— a phase boundary, with a much reduced density (66% of the Fe interior) and enhanced magnetic moment ($2.5\mu_B$) was required in order to obtain a good fit to the data. The notion of a phase boundary region is motivated by X-ray diffraction observations taken at grazing incidence of a reconstructed interface between the Fe film and MgO substrate [12]. Like grain boundaries, which often have densities 20% less than the bulk [13,14], phase boundaries are also expected to be less dense than the bulk. The enhancement of the magnetic moments of Fe atoms in the phase boundary may be a consequence of its reduced density, since the magnetic moments of transition metal atoms, like Fe, are known to increase as their densities are reduced [15]. The present study of the Fe/MgO system is one example of the importance of interfacial structure on the magnetic character of interfaces.

4. Future Plans

The magnetic native oxide on the Fe film illustrates a difficulty encountered in studies of surface and interface magnetism— how can the magnetism of surfaces and interfaces be observed in their pristine state? While passivating materials can be used to prevent oxidation, these materials can also profoundly change the magnetism system they are trying to protect. For example, the diffusion of Al or V into Fe can depress the Curie temperature of the Fe film [16] so that a magnetic Fe surface may become non-magnetic. The best approach to the study surface and interfacial magnetism is to avoid oxidation in the first place by fabricating samples *in situ*. Not only is oxidation avoided, but changes in the magnetic structure of a sample can be observed during its fabrication, e.g. magnetism can be monitored as layers of different materials are deposited to form multilayer structures. This approach has been implemented at LANSCE with the construction of a fabrication system with the following characteristics:

- samples can be fabricated under ultra high vacuum conditions,
- heating filaments can raise the temperature of the sample to greater than 700°C,
- sapphire windows allow polarized neutrons to enter and exit the chamber for reflection studies, and
- magnetic fields in excess of 1T can be applied to the sample during its fabrication (if desired), and while PNR measurements are collected.

During 1992, this equipment was used to manufacture and study thin films of Fe and Ni on Si substrates *in situ*. The polarized neutron reflectivity from a thin Fe film prepared *in situ* is shown in Fig. 6. The data required one change of incident angle, and seven hours of collection time. This collection time is sufficiently long that for the vacuum in the chamber ($1 \cdot 10^{-6}$Pa), a monolayer of oxygen probably formed on the sample surface. For the 1993 experiment, an ion pump will be used to improve the vacuum by one to two orders of magnitude. Combined with improvements in the polarization optics, which decrease data collection time, studies of surface magnetism of thin-films and multilayer samples in their pristine state should be possible. In particular, the onset of spin-flip scattering like that observed in Fe/Cr multilayers [17], can be monitored after each deposition step in the fabrication of a multilayer. This experiment is planned for completion in the summer of 1993. At a next generation advanced spallation source where the incident intensity might be 25-50 times greater, these PNR measurements could be made during the actual deposition of a thin-film sample, rather than after the completion of each step in the deposition process. This capability would permit studies of dynamic processes during the fabrication of magnetic and non-magnetic thin-films and multilayers.

The construction of the polarization equipment for neutron reflection studies was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under contract W-7405-Eng-36. Funding for PNR measurements of the thin-film samples and construction of the "*in situ* fabricator" was obtained from Laboratory Discretionary Research and Development resources (project XA62).

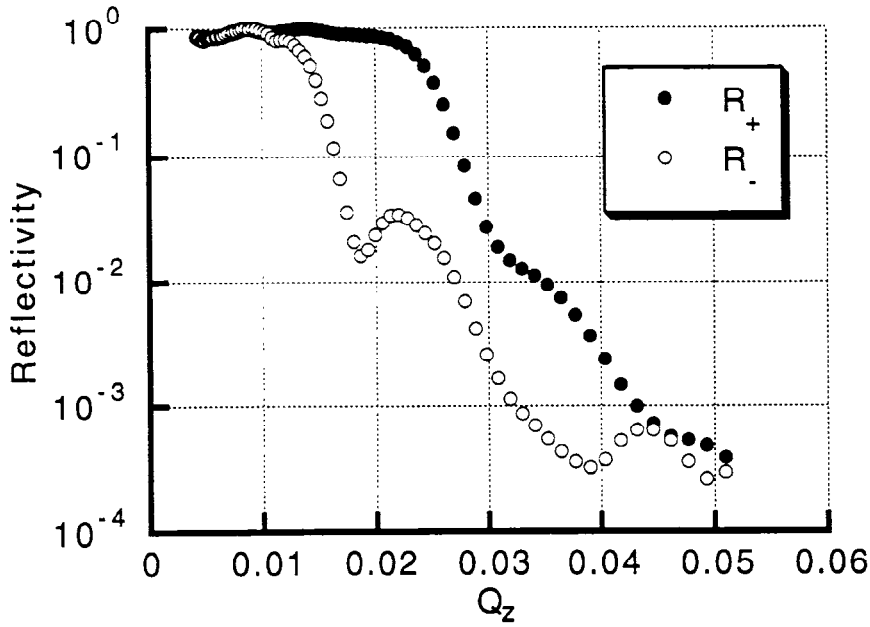

Fig. 6 Polarized neutron reflectivities, R_+ (•) and R_- (○), measured from an Fe thin-film epitaxially deposited *in situ* on a single-crystal Si substrate.

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