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Remanent Multilayers for Polarized Neutron Instrumentation

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ABSTRACT

Polarizing multilayers and supermirrors for applications in neutron instrumentation have been produced using DC-magnetron sputtering. The chosen material combination $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ offers the possibility to adjust the scattering length density of the 'contrast' material Ti to the value being effective on neutrons in the $|-\rangle$-state in the FeCoV alloy magnetized to saturation, by loading Ti with the suited amount of nitrogen. Neutron reflectometry data for supermirrors give flipping ratios of 40 and more and reflectivities better than 85% at twice the critical angle of Ni. The production process induces a magnetic anisotropy for directions lying within the layer plane. $Ti_{1-v}Gd_v$ absorbing subcoatings have been used to suppress the remaining reflectivity of the unwanted spin component from the substrate-mirror interface. SQUID and MOKE magnetometry accompanied the experiments performed with polarized neutrons.

1. Introduction

Besides polarizing filters and field gradient devices neutron beam polarization can be accomplished by reflection at a monochromator or a mirror which has different scattering cross sections for the two spin states of the neutron. The term neutron mirror includes thin film multilayers, single layer coatings and polished surfaces. Mirror coatings have the advantage of easy availability and relatively low costs, and to a certain measure they can be designed to the needs of the instrument they are aimed for. Their application is in particular advantageous for the polarization of cold neutrons. Thus, mirrors serve for this purpose since 1951 [1]. In particular the properties of FeCo alloys have been exploited for effective

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Keywords: Polarizer, Supermirror

polarizers. This alloy can be easily saturated and the composition can be varied over a wide range to give the desired optimum scattering parameters either for a single layer [2,3] or a multilayer when combined with an adequate spacer material [4]. A giant step forward to make thin film polarizers useful for the large beam cross sections that are often required in neutron experiments has been made by the emergence of two ideas, one to extend the regime of total reflection by using multilayers and the other to cover the whole cross section with an array of bent mirrors on thin substrates, and thereby forcing the neutrons to be reflected at least once.

The first improvement originates from the work of Turchin [5] and Mezei [6] and consists of a stack of alternating layers of two materials made in such a way that constructive interference of the partial waves reflected at each interface takes place in a q-band ($q = 4\pi\frac{\sin\vartheta}{\lambda}$) adjacent to the edge of total reflection at q_c as one finds it for a bulk material. To achieve this, the layer thicknesses have to increase in a way that for the nth layer

$$d(n) = \frac{d_c}{4\sqrt[4]{n/2}} .$$

Herein, the scattering parameters for the material combination A-B are included through

$$d_c = \left(\sqrt{\left(\rho_{at}^{(A)} b^{(A)}/\pi\right)^2 - \left(\rho_{at}^{(B)} b^{(B)}/\pi\right)^2} \right)^{-1} .$$

$\rho_{at}^{(X)}$ denotes the atomic density and $b^{(X)}$ the scattering length of each material, respectively. Often, it is useful to calculate the layer sequence with a slightly increased d_c. Due to refraction the layer thickness for each material has to be corrected to give an optical thickness [6,7]:

$$d_{opt}^{(X)}(n) = \frac{d(n)}{\sqrt{1 - \frac{d(n)^2}{\pi/\rho_{at}^{(X)} b^{(X)}}}} .$$

The reflectivity in this band can be increased by selecting a material combination with a high contrast in scattering length density $\Delta\rho_{sc} = \rho_{at}^{(A)} b^{(A)} - \rho_{at}^{(B)} b^{(B)}$. Mezei and coworkers succeeded in producing a first Fe-Ag supermirror.

The second invention is the curved Soller guide [8] that has been equipped with thin films of a $Fe_{40}Co_{60}$ alloy to polarize the neutrons which pass through the narrow channels by reflection from the magnetized mirrors [9]. Consequently, a short time later supermirrors have been introduced into Soller guides. By thermal evaporation of a Gd-Ti multilayer that serves as antireflection coating and subsequent deposition of a sequence of Co-Ti bilayers O. Schärpf and co-workers produced thousands of polarizing supermirrors [10]. A large part of them have been mounted into 'superbender' Soller guides.

We aimed for a polarizer coating with a high $|+\rangle$ -('spin up')-reflectivity $R^{|+\rangle}$ on the basis of a commercially available $Fe_{0.50}Co_{0.48}V_{0.02}$ alloy. The scattering length densities for this material are $\rho_{sc}^{|+\rangle} = 1066\ \mu m^{-2}$ and $\rho_{sc}^{|-\rangle} = -64\ \mu m^{-2}$. To minimize the $|-\rangle$ -reflectivity $R^{|-\rangle}$ of the multilayers, we combined the ferromagnetic alloy with titanium as the spacer material, loaded with nitrogen to give $\rho_{sc}(Ti) \cong \rho_{sc}^{|-\rangle}(Fe_{0.50}Co_{0.48}V_{0.02})$.

2. Experimental

A Leybold Z600 DC-magnetron sputtering plant has been used to produce multilayers [11] for neutron reflection experiments and magnetometry. Supermirror layer sequence consisting of 150 single layers were calculated according to the formalism of Hayter and Mook [12]. Single layer $Fe_{0.50}Co_{0.48}V_{0.02}$ samples were produced under different sputtering conditions to perform stress and magnetometric measurements. As substrates we selected 2 mm float glass and 0.5 mm polished silicon wafers because of their very good surface microroughness [13]. The Ti:N layers were realized by reactive sputtering. The variation of the partial pressure of nitrogen p_{N_2} allows to regulate the amount of nitrogen embodied in the titanium layer. Neutron reflectometry experiments have been done mainly on V6 at BENSC/HMI and on the T3 mirror testing facility at the ILL. Both instruments work in the constant wavelength mode. An electromagnet on V6 makes it possible to reflect polarized neutrons from mirrors in a variable magnetization field.

Magnetometric measurements were conducted on a SQUID and a MOKE magnetometer at the KFA Jülich. Whereas the SQUID signal comes from the entire sample (5×10 mm^2 surface area), MOKE (magnetooptic Kerr effect) is only sensitive to the magnetization in a volume penetrable to light. The rotation of the polarization axis of the light is a measure for the component of the magnetization vector in the surface plane. The results from magnetometry are extracted from the diploma work of one of the authors [14]. It represents only a small fraction of his extensive characterization of magnetostriction in $Fe_{0.50}Co_{0.48}V_{0.02}$ layers. Additionally, TEM micrographs were taken at HMI to visualize the grown multilayer.

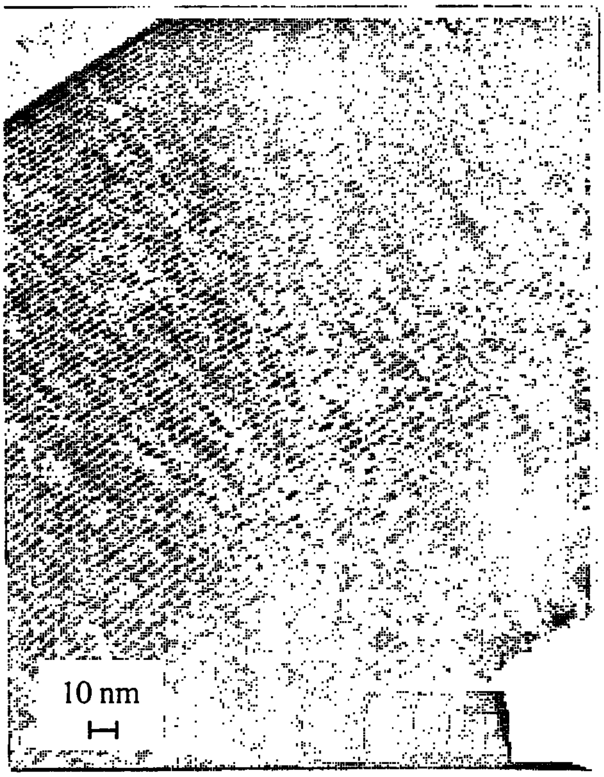

Fig. 1. Transmission electron microscopy (TEM) micrograph taken from an ultramicrotomy cut of a 150 layer $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ supermirror (sample R1285). The substrate side is in the top left corner. The picture was taken with a Philips 120 keV microscope.

3. Results

Reflectivity measurements on a series of 'monochromator'-type $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ multilayers showed that an increased p_{N_2} during the sputtering of Ti brings with it an increased $\rho_{sc}(Ti)$. The results led us to the deposition conditions under which we achieve the matching of the scattering length densities [15], i.e. $\rho_{sc}(Ti) \cong \rho_{sc}^{|-\rangle}(Fe_{0.50}Co_{0.48}V_{0.02})$. For this p_{N_2} the detected $R^{|-\rangle}$ was not discriminable from the remaining 1.5% $R^{|+\rangle}$–signal that originates from the imperfect efficiency of polarizer and flipper. We could not find any evidence for 'magnetically dead' layers at the interfaces although $R^{|-\rangle}$ is very sensitive to their appearance in the case of $\Delta\rho_{sc} = 0$ [16].

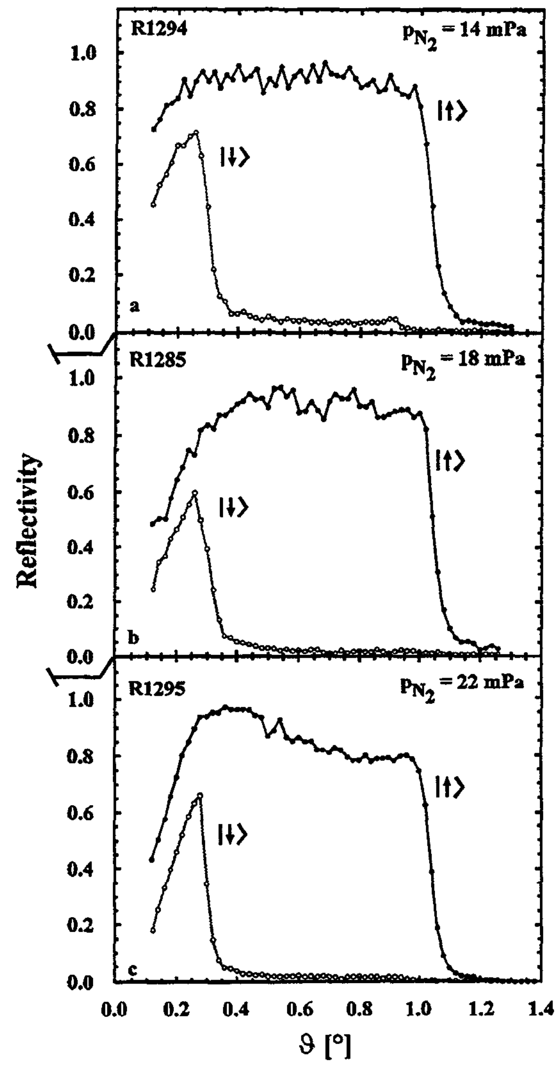

Fig. 2. Polarized neutron reflection from 150 layer $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ supermirrors produced with nitrogen partial pressures of 14 mPa (a), 18 mPa (b) and 22 mPa (c). The data was taken at the V6 reflectometer at BENSC/HMI ($\lambda = 0.47$ nm, $B_{ext} = 79$ mT).

Consecutively, we produced supermirrors under the same conditions as for the monochromators. A copy of a TEM micrograph of such a supermirror is displayed in Fig. 1. Fig. 2 compiles the reflection data for the mirrors magnetized to saturation in an external field $B_{ext} = 79$ mT. It is obvious that for $p_{N_2} = 18$ mT a good matching has been achieved also giving a high $R^{|+\rangle} \cong 87$ %. The data was taken at $\lambda = 0.47$ nm, so that the measured cut-off of $R^{|+\rangle}$ at 1.04° corresponds to $m = 2.23$. Here, given by $\vartheta_c = m \cdot \vartheta_c(Ni)$, m characterizes the supermirror cut-off in terms of multiples of the critical angle of bulk nickel

$\vartheta_c(\mathrm{Ni}) = 0.99 \cdot \lambda[\mathrm{nm}]$. The $R^{|-\rangle}$–curves in Fig. 2 exhibit a cut-off that comes from the reflection of neutron transmitted through the supermirrors coating but reflected from the interface of the glass substrate. To avoid this effect which decreases the q-regime that is of use for beam polarization we followed the path of other groups and coated the substrate with an antireflecting layer of $\mathrm{Ti}_{1-v}\mathrm{Gd}_v$ before depositing the supermirror sequence. The adequate Gd concentration and thickness of the layer could to be found experimentally in an easy way. Because the sputtering in DC-magnetron mode is preferentially from a ditch defined by the arrangement of the magnets on the back of the target we only needed to insert 3 mm Gd pellets into this ditch. In three series for which we implemented an increasing number of pellets into the Ti target we coated antireflecting layers of 30 nm, 50 nm and 70 nm underneath the $\mathrm{Ti{:}N\text{-}Fe_{0.50}Co_{0.48}V_{0.02}}$ supermirror. 50 nm Ti:Gd with the highest Gd concentration reduced the remaining $R^{|-\rangle}$–signal to 10%. Although a better damping of unwanted neutron reflection from the substrate can be achieved by depositing a Ti-Gd multilayer with decreasing Gd layers thicknesses [7] we assessed the single layer solution to be sufficient for multireflection devices as bent Sollers. Fig. 3 shows the performance of such a polarizing mirror.

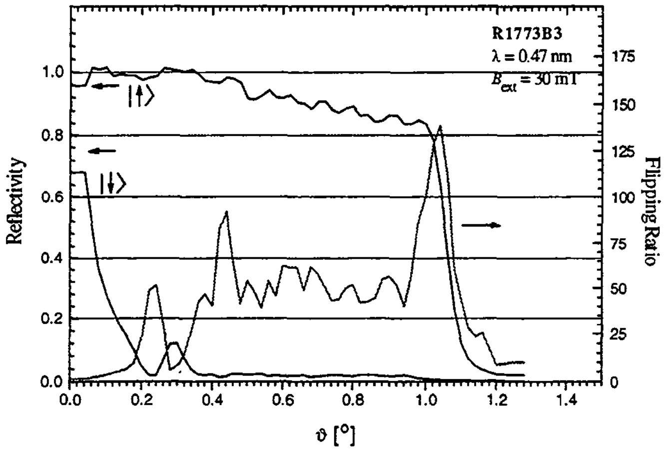

Fig. 3. Polarized neutron reflection from a 150 layer $\mathrm{Ti_{1-u}N_u\text{-}Fe_{0.50}Co_{0.48}V_{0.02}}$ supermirror coated onto a 50 nm Ti:Gd antireflection layer. The magnetic layers are saturated in B_{ext} = 30 mT.

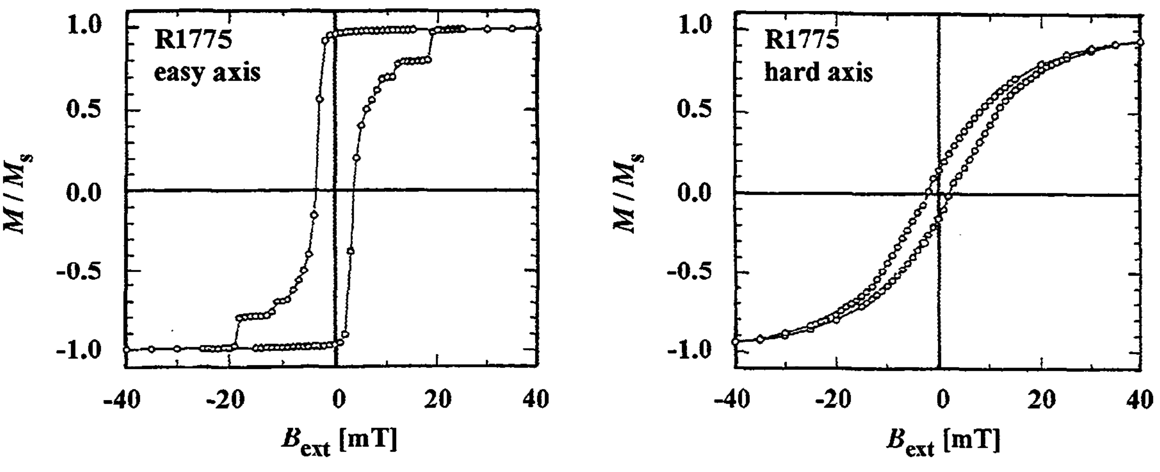

Fig. 4. Hysteresis loops for in-plane magnetization of a $\mathrm{Ti_{1-u}N_u\text{-}Fe_{0.50}Co_{0.48}V_{0.02}}$ supermirror along the easy axis (left) and the hard axis (right) as measured with a SQUID magnetometer.

Additionally we can take advantage of the deposition procedure in the Z600. It gives rise to special characteristics of the $Fe_{0.50}Co_{0.48}V_{0.02}$ layers when they are produced under $p_{Ar} = 0.6$ mPa. The substrate table moves through underneath a rectangular target of approximately $88 \times 500\ \mathrm{mm}^2$ with a speed determined by the desired layer thickness. A diaphragm is used to get a homogeneous thickness in the direction perpendicular to the movement. These anisotropic deposition conditions give rise to a stress anisotropy in our films. The stress is high enough that in the extreme case of very thick multilayer (600 layers and more) the coatings rip off themselves in small strips from the substrates. As sputtered films especially those of Ti stick extremely good to our substrates, a part of the glass is torn away from the rest and stays connected to the dismantled films. Nevertheless this mechanical problem is not encountered for the coatings under investigation in this work. Moreover, the stress anisotropy implies a magnetic anisotropy caused by magnetostriction. This is revealed by the magnetometry data taken from the supermirrors (Fig. 4). An easy axis for saturation can be found in the direction perpendicular to the movement of the table. Along this axis the material can be saturated in fields as low as 25 mT. The remanent magnetization of more than 90% of the saturation level is sufficient to find a useful polarization in $B_{ext} = 0$. We measured coercitive fields in the range of $B_{coerc} \cong 4$ mT with a SQUID. Polarized neutron reflection still showed the remanent behavior when this field has been applied in opposite direction to a saturated supermirror (Fig. 5a). Then at $B_{ext} = -10$ mT the magnetization flipped within 20 min. and almost saturated again (Fig. 5b).

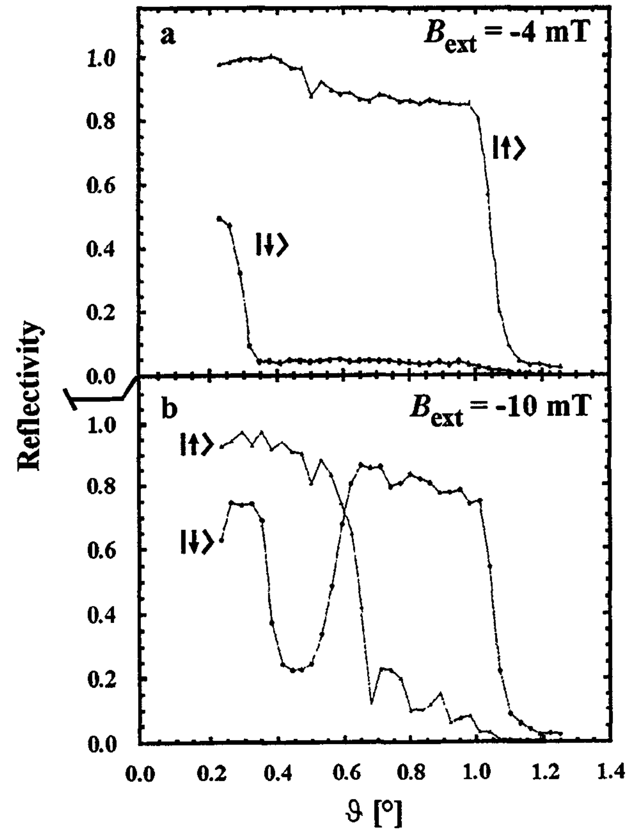

Fig. 5. Polarized neutron reflection from 150 layer $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ supermirrors produced with a nitrogen partial pressure of 18 mPa in two external fields oppositely directed to the previously saturated magnetization ($\lambda = 0.47$ nm).

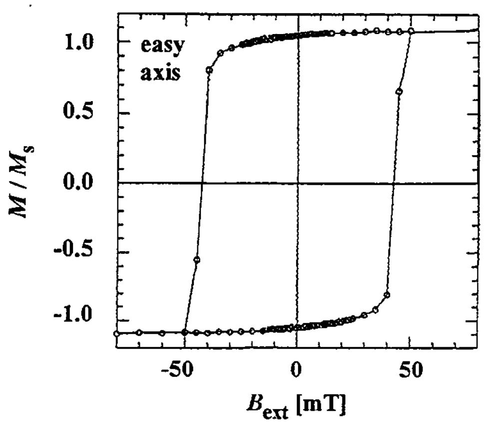

Fig. 6. SQUID hysteresis loop for in-plane magnetization along the easy axis of a 28 nm $Fe_{0.50}Co_{0.48}V_{0.02}$ film produced by stationary sputtering.

In Fig. 6 the hysteresis loop of a 28 nm single layer of $Fe_{0.50}Co_{0.48}V_{0.02}$ is displayed. This sample has been produced under conditions that are different to the previous examples. It is made in the static mode, i.e. the table did not move relative to the target. The substrate position was located 80 mm away from the center of a circular 75 mm target. Thus, only sputtered particles under oblique incidence arrive on the substrate. As can be deduced from Fig. 6 a substantially higher coercitive field can be achieved in the easy axis direction than for the 'dynamically' sputtered multilayers or single films.

The performance of the Ti:N-$Fe_{0.50}Co_{0.48}V_{0.02}$ multilayer supermirrors does not change after $2^1/_2$ hours heat treatment at 230°C. The flipping ratios $\tilde{R}$ for a saturated mirror calculated from the ratio of the neutron reflection spectra for the two spin states is $\tilde{R} \geq 40$ over a q-range exceeding from $m = 0.77$ to the cut-off (Fig. 3). In the remanent state we find $\tilde{R} \geq 25$, which gives a polarization $P \geq 0.92$. We also succeeded in producing a polarizing multilayer monochromator consisting of 251 layers on an antireflecting sublayer that reflects a wavelength band $\frac{\Delta\lambda}{\lambda} = 10\%$ (Fig. 7).

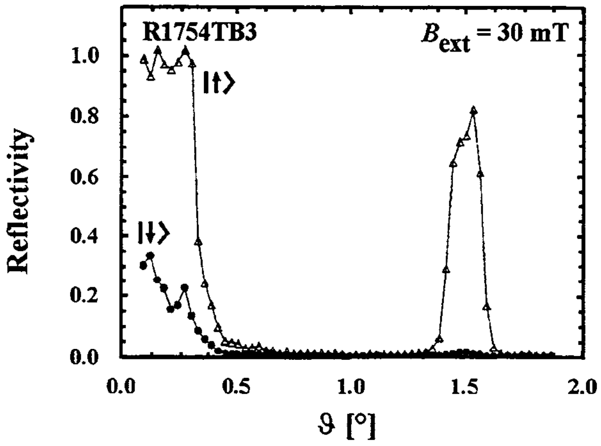

Fig. 7. Polarized neutron reflection from a 251 layer $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ multilayer magnetized to saturation ($\lambda = 0.47$ nm).

4. Conclusions and Outlook

By matching the scattering length density of $Ti_{1-u}N_u$ with the one for $|-\rangle$-neutrons in magnetized $Fe_{0.50}Co_{0.48}V_{0.02}$ we succeeded to manufacture reproducibly a supermirror coating with the following useful properties:

(i) a high $|+\rangle$-reflectivity (87% at 2.1 times the critical angle of bulk Ni),

(ii) flipping ratio $\tilde{R} \geq 40$,

(iii) operational in zero field (important for neutron spin echo),

(iv) no clear evidence for 'magnetically dead' layers and

(v) stable with regard to temperature ($T \leq 230C$), therefore we expect that the layers hardly interdiffuse.

Presently, we are in the course of producing polarizing mirrors for focusing Soller ‘superbenders’ that will be operated in the DNS spectrometer and the neutron spin echo spectrometer at the Jülich neutron scattering facility, and for a triple axis spectrometer for polarized neutrons at the SINQ neutron source at PSI. The reflection spectrum for one of the representative mirror of the many that will be inserted is shown in Fig. 8.

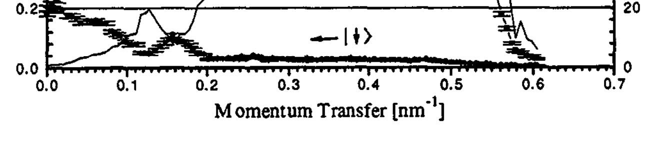

Fig. 8. Polarized neutron reflection from a 150 layer $Ti_{1-u}N_u$-$Fe_{0.50}Co_{0.48}V_{0.02}$ supermirror on a 50 nm antireflecting Ti:Gd sublayer supported by a 0.2 mm glass substrate.

Other applications are in a project state. An especially interesting application is considered for the reflectometer at SINQ/PSI [17]. This instrument can operate in the white beam time-of-flight mode. As the remanent mirrors can withstand an oppositely oriented magnetic field of the order of magnitude of a typical guide field, i.e. $B_{ext} = -5$ mT, they could be used as spin selecting device [18]. An external field pulse sufficient to saturate the layers can be applied by several small electromagnetic coils, and then set to zero in order to avoid any change in the direction of the guide field. By flipping the direction of the applied external magnetic field in a short pulse one can select the desired beam polarization.

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