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Highlights from the magnetism reflectometer at the SNS

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ABSTRACT

The magnetism reflectometer at the SNS has passed the phase of commissioning and is in operation for users. The high power of the neutron source demands that special attention be paid to the optimization of the background in order to be able to measure the two-dimensional maps of reflected and scattered intensities with polarized neutrons in a broad range of momentum transfer. It implies an effective separation of the magnetic and non-magnetic reflectivity, off-specular scattering and the grazing incidence SANS in a high range of momentum transfer. Therefore the polarizing and the analyzing efficiencies are of particular importance on this time-of-flight instrument. At the beginning of July 2008 the world's first ^{3}He neutron analyzer with on-line pump-up polarization was successfully installed and the tests with a magnetic multilayer film showing a strong off-specular spin-flip scattering were made. The performance of the instrument is under constant improvement in order to make it an effective and optimal instrument for the applications in nanosciences.

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1. Introduction

Rapid advances in nanotechnology and nanoscience require development of various methods for the characterization of nanostructured materials [1]. Neutron scattering and in particular polarized neutron reflectometry serves this purpose and continuously develops [2,3]. The development goes in several directions. Amongst them the increasing power of the neutron sources plays a very important role [4]. Nonetheless, the effective use of the produced neutrons is a no less important element in this development. It includes the improvement of the background and with this the extending of the measured range of momentum transfer, raising the polarizing and analyzing efficiency of the specific elements of the instrument and increasing the resolution. This allows improving the sensitivity to specific parameters like the absolute value and direction of the layer magnetization vector across the film thickness, and interfacial magnetic and non magnetic roughness. Lateral nanostructures, magnetic and structural inhomogeneities create off-specular scattering in addition to the specular reflection. Therefore the registration and the polarization analysis of the off-specular scattering are of particular importance. Here we present the performance of the magnetism reflectometer at SNS which we obtained after a considerable and successful improvement of the background and of different elements of the instrument.

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2. Polarizer

The actual polarizer used on the reflectometer is a bender-type polarizer [5]. It is composed of 200 thin Si blades (each 0.15 mm wide) covered with a Fe/Si reflecting super-mirror coating ($m = 2.8$) and a Gd absorbing layer on the back. The bender is 75 mm long and covers a beam with a cross-section of $29 \times 30\,mm^2$. It is placed at the exit of the tapered guide and is aligned to the maximum intensity with the incident grazing angle of 0.4° to allow for polarized neutrons above a wavelength of 2 Å. The slits before and after the polarizer are used for the collimation of the beam. The profile of the polarized neutron beam after the polarizer is shown in Fig. 1a. It is registered with a position sensitive detector and is integrated over the time-of-flight band. It is measured with an entrance slit of 3 mm in front of the bender. A high efficiency RF-gradient spin-flipper [6] is used to reverse the incident beam polarization.

The registered intensity has a specific profile which results from the polarizer acting with the silicon blades as a Soller collimator; so several individual beams are created along the X-axis in Fig. 1a being reflected by several illuminated Si channels. The fine structure along the Y-axis is a detector effect. In Fig. 1b the time-of-flight spectra of each polarizer channel is shown. The brightest channel is selected by the slit after the polarizer as the incoming beam to the sample. In this case this would be the beam around the detector pixel X 125 in Fig. 1a, The spectral dependence of the polarized neutron beam integrated over the vertical direction of the beam (see Fig. 1) is depicted in Figs. 2a and b. It is measured for the time-of-flight intervals corresponding to 60 and 30 Hz repetition rate of the neutron pulse production of

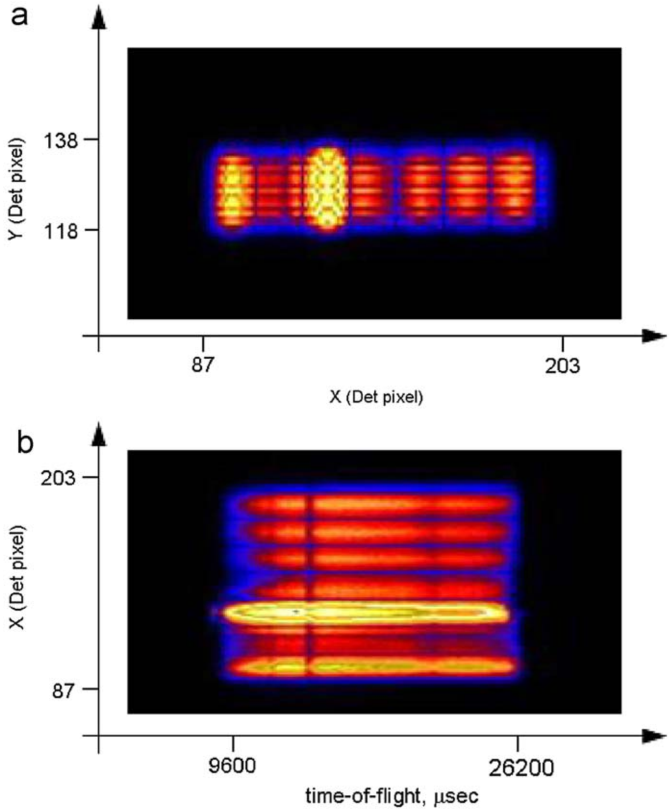

Fig. 1. Experimentally measured profile of the intensity distribution of the polarized neutrons after the polarizer bender with a slit of 3 mm in front: (a) cross-section of the polarized beam as a function of the detector pixels in horizontal (X) and vertical (Y) directions and integrated over the wavelength band and (b) spectral distribution of the polarized neutrons across the beam as function of the time-of-flight and the detector pixels.

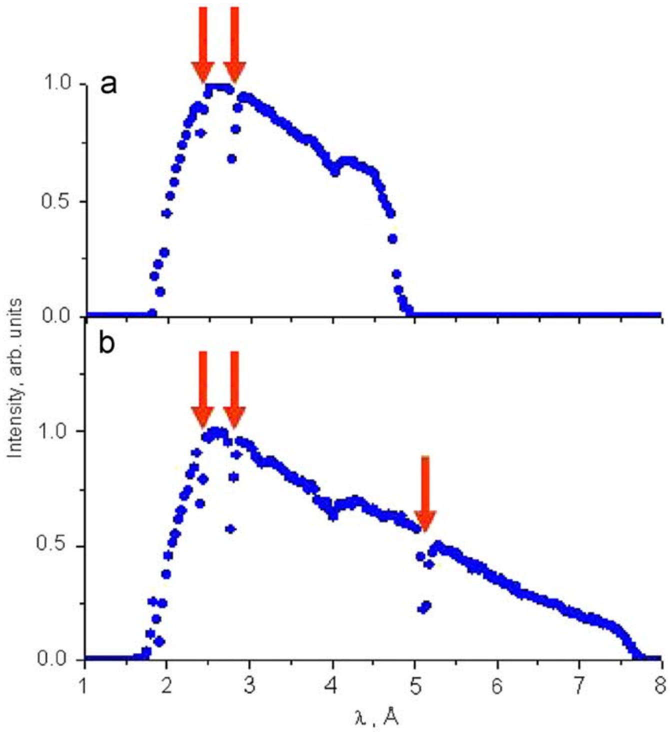

Fig. 2. The spectra of the polarized neutrons after the bender-polarizer as a function of the wavelength measured for the time-of-flight intervals corresponding to (a) 60 Hz and (b) 30 Hz repetition rate of the choppers, respectively. Arrows mark the intensity losses due to diffraction in the Si-blades of the polarizer.

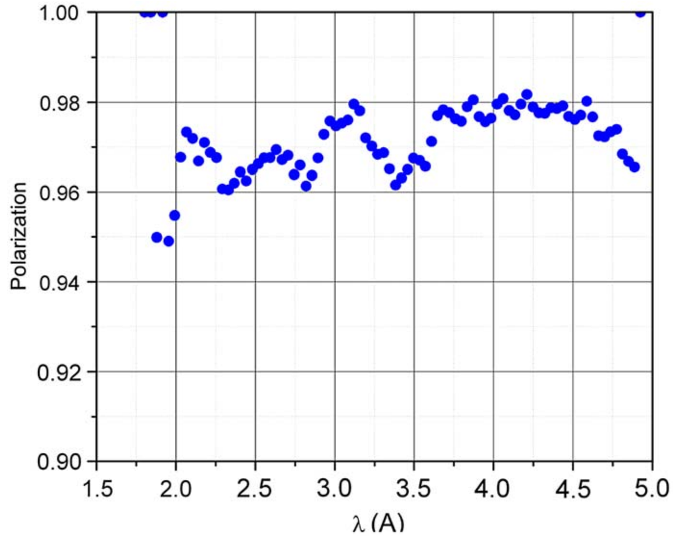

Fig. 3. Polarization of the bender-polarizer as a function of neutron wavelength.

the source, which correspond to wavelength intervals of 2–4.75 Å and 2–7.5 Å, respectively. The spectra of the polarized neutrons after the bender have peculiar features. Strong drops in intensity are marked with the arrows in Fig. 2. They are due to Bragg diffraction in the Si blades during the transmission of the thermal neutrons through the bender. This diffraction effect causes not only a loss of intensity but creates an additional background due to diffuse scattering.

The polarization function of the bender-polarizer is shown in Fig. 3. Although the mean value of the polarization efficiency of the bender is rather good (it varies between 96% and 98%) we plan to install a polarizer which does not have the above listed disadvantages and which is optimized to the experiments in grazing incidence geometry for our time-of-flight conditions and to the thermal neutron spectrum.

3. Magnetic off-specular scattering: measurement of the two-dimensional maps

The short wavelength band available on the magnetism reflectometer allows us to use the highest intensity of the incident beam and to obtain a high polarization. The disadvantage of the short wavelength band is the necessity to measure the reflected intensity at several incident scattering angles in order to gain a large range in momentum transfer. This requires the merging of several two-dimensional (2D) intensity maps of reflected and scattered neutrons measured at different incident scattering angles and with four spin states for each scattering angle. Further the different resolution functions for each angle have to be taken into account. This necessitates the development of dedicated software for the data reduction of the 2D intensity maps, which is currently under development.

We performed a test experiment using a $[^{57}Fe/Cr] \times 12/Al_2O_3$ multilayer with an anti-ferromagnetic inter-layer coupling and with an in-plane magnetic domain structure [7]. The polarized neutron reflection experiment was performed in an external magnetic field of 30 mT applied along the in-plane easy axis (0 0 1) after a saturation field of 0.5 T. The 2D pattern of specular reflection and off-specular scattering was measured with polarized neutrons in the wavelength band of 2 Å< λ <4.75 Å and a polarization of 0.97. The measurement was performed at 5 incident angles in order to obtain the range of momentum

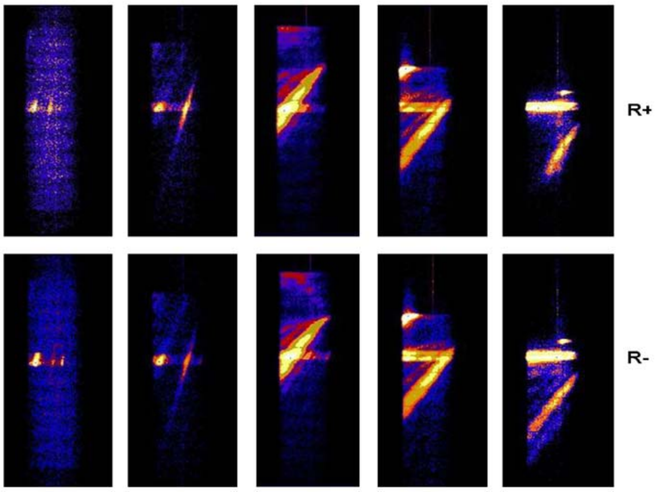

Fig. 4. Snap-shots of the intensity reflected and scattered by the sample measured at 5 incident angles for neutrons polarized along (R^{+}) or opposite (R^{-}) to the direction of the external magnetic field.

transfer Q_z from 0.008 to 0.3 Å^{-1}. The results of the PNR experiment are shown in Fig. 4 for the two spin states R^{+} and R^{-}.

After merging the 2D maps measured at different angles, we obtained the 2D intensity maps of reflected and scattered neutrons for $R^{+}=R^{++}+R^{+-}$ and $R^{-}=R^{--}+R^{-+}$, shown in Fig. 5.

The specular reflectivity is displayed by the horizontal line along the time-of-flight scale with the total reflecting region on the right side followed by Bragg-reflections from the bi-layer structure. The highest intensity is seen at the $\frac{1}{2}$ order magnetic reflection followed by the 1st order reflection and so on. Lower intensity oscillations originate from the total multilayer thickness. Off-specular scattering, which appears here as Bragg-sheet scattering, concentrates around the $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$,... order magnetic Bragg-peaks. It is created by spin-flip scattering on magnetic domains and its intensity is proportional to the component of the in-plane magnetization, which is perpendicular to the direction of the neutron polarization. The difference in the scattering of neutrons with spin-up (parallel to the direction of the external field H_{ext}) and of neutrons with spin-down (anti-parallel to H_{ext}) is seen in the asymmetry at the lower ends of the $\frac{1}{2}$ order Bragg sheet of off-specular scattering.

The reflectivity curves, extracted from the 2D intensity maps, are depicted in Fig. 5b. It is clearly shown that the half order Bragg-peak intensities are not influenced by the spin-state.

4. First test results of the online polarized ^{3}He neutron polarization analyzer

At the beginning of July 2008, we installed the online polarized-^{3}He neutron polarization analyzer. We did a first test of the system reported here.

The setup uses a cell called "Barbera", constructed by Wangchun Chen and Tom Gentile at NIST [8] It has a "thickness" $n\sigma l=4.12$ at 4.96 Å and was tested to have reached 76% ^{3}He-polarization. In the setup for the reflectometer, the cell was placed in a hot-air oven. The cell was heated to about 200 °C to control the alkali (Rb, K) vapor density in the cell, which determines the speed of the polarizing process. The nuclear magnetic resonance (NMR) process, called "adiabatic fast passage", was used to switch the ^{3}He polarization from parallel to antiparallel to the magnetic holding field and vice versa.

During the polarizing process (pump-up) of the system, the ^{3}He polarization was monitored by an NMR technique called

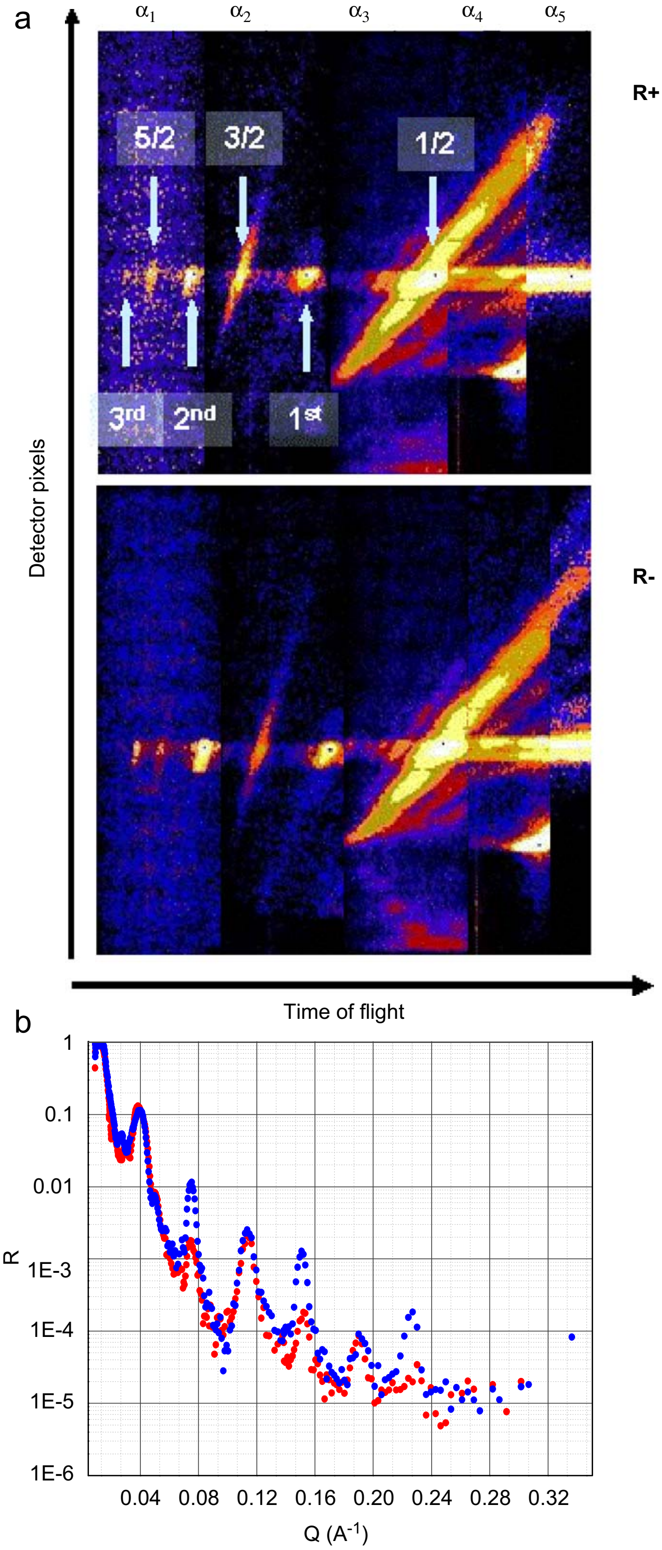

Fig. 5. (a) Experimental 2D intensity maps of neutrons reflected and scattered by a Fe/Cr multilayer film in an external magnetic field of 0.03 T parallel to the surface of the sample, (b) reflectivity curves R^{+} (blue) and R^{-} (red) extracted from the 2D intensity maps and normalized by the direct beam (shown as functions of momentum transfer).

"free-induction-decay" and for some part of the pump-up process, also monitored by neutron transmission measurements. The neutron transmission measurement, which provides an absolute measurement of both the neutron polarization and the ^{3}He

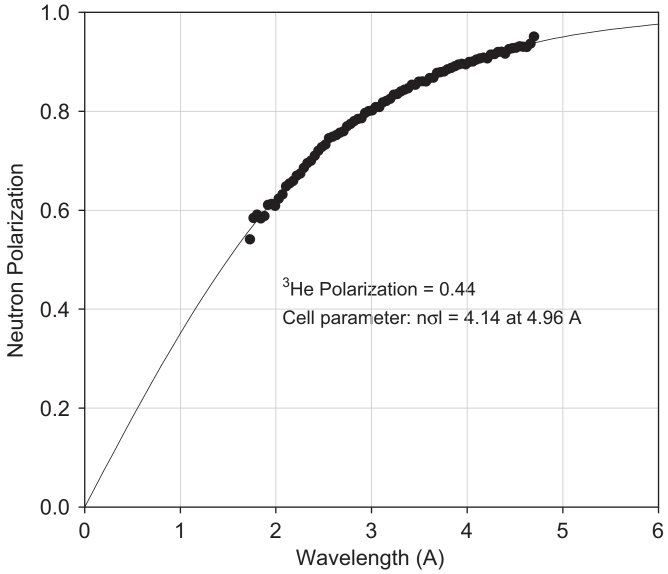

Fig. 6. Neutron polarization as a function of neutron wavelength taken at a ^{3}He polarization of 44%.

polarization, was then used to scale the NMR signal (see Fig. 6). This measurement was taken when the ^{3}He polarization was at 44% during one of the pump-up stages. The neutron polarization (P_{Neutron}) data was fitted to $P_{\text{Neutron}} = \tanh\,(P_{\text{He}}\ [\text{n}\sigma\text{l}]\ (\lambda/\lambda_0))$ to extract the ^{3}He polarization (P_{He}), where $\text{n}\sigma\text{l} = 4.14$ and $\lambda_0 = 4.96$ Å.

In order to test the ^{3}He-filter analyzer we performed an experiment with the Fe/Cr multilayer described above. The intensity maps of the four spin-state combinations around the $\frac{1}{2}$ order Bragg-peak are shown in Fig. 7. The spin-flip scattering is concentrated along the $\frac{1}{2}$ order Bragg-sheet. The intensity of the Bragg-sheet scattering in the non-spin flip channels R^{++} and R^{--} is due to an imperfect polarization efficiency of the polarizer and the analyzer. Likevise the non-spin flip specular scattering appears in R^{++} and R^{--} and traces due to not perfect polarization efficiency in R^{+-} and R^{-+}. Thus the results of Ref. [4] are verified but this time with a ^{3}He filter as the spin analyzer.

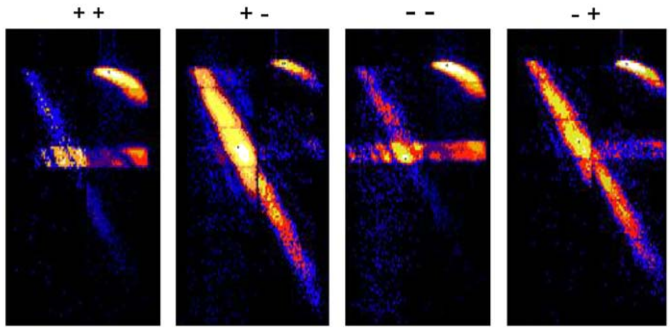

Fig. 7. (a) Experimental 2D intensity maps of neutrons reflected and scattered by a Fe/Cr multilayer film in an external magnetic field of 0.03 T parallel to the surface of the sample. Only two intensity frames are displayed (see Fig. 4 for comparison) to concentrate on the $\frac{1}{2}$ order Bragg-peak with its off-specular Bragg-sheet.

In summary we demonstrated a good performance of the new magnetism reflectometer at SNS. Additional planned modifications and improvements of the background and polarization in combination with a high power of the SNS will enable us to perform unique experiments on nanosystems.

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