Development and demonstration of in-situ SEOP $^3$He spin filter system for neutron spin analyzer on the SHARAKU polarized neutron reflectometer at J-PARC

H Hayashida$^1$, T Oku$^1$, H Kira$^2$, K Šakai$^1$, M Takeda$^{1,3}$, Y Sakaguchi$^1$, T Ino$^4$, T Shinohara$^1$, K Ohoyama$^5$, J Suzuki$^2$, K Kakurai$^3$, M Mizusawa$^2$, N Miyata$^5$, D Yamazaki$^1$, R Maruyama$^1$, K Soyama$^1$ and M Arai$^1$

$^1$J-PARC Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
$^2$Comprehensive Research Organization for Science and Society, Tokai, Ibaraki 319-1106, Japan
$^3$Quantum Beam Science Directorate, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
$^4$High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan
$^5$Institute for Materials Research, Tohoku University, Sendai, Miyagi 980-8577, Japan

E-mail: hayashida.hiroshi@jaea.go.jp

Abstract. A new neutron reflectometer, SHARAKU, with a vertical sample-plane geometry was installed at beam line 17 at J-PARC Materials and Life Science Facility. Although a polarizing supermirror was previously installed as a neutron spin analyzer on SHARAKU, a $^3$He spin filter is advantageous because it can cover a large solid angle. An in-situ SEOP $^3$He spin filter system using a new compact laser unit has been developed for the analyzer. In this paper, we report a successful off-specular measurement with the new compact in-situ SEOP analyzer at SHARAKU.

1. Introduction

Polarized neutron-scattering methods are powerful tools to study magnetic materials, and these methods require some polarizing device for the neutron beam. $^3$He neutron spin filters (NSFs) are advantageous over magnetic supermirrors because they cover a large solid angle and produce polarized shorter neutron beam wavelengths. Hence, $^3$He NSFs become popular devices for polarized neutron scattering experiments in recent years. We have also been engaged with the development of $^3$He NSFs using the spin exchange optical pumping (SEOP) technique. The SEOP technique enables us to use a $^3$He NSF with in-situ pumping of $^3$He gas, which is a huge advantage for some experiments because the in-situ pumping technique allows us maintain a stable $^3$He gas polarization over long measurement times. In fact, the development of the in-situ SEOP $^3$He NSF systems in Japan has achieved a 73 % polarization of $^3$He gas [1,2].

A new neutron reflectometer, SHARAKU, with vertical sample-plane geometry was installed at the beam line 17 (BL17) at the J-PARC Materials and Life Science experimental Facility (MLF) [3]. Magnetic properties in thin magnetic films are one of the main targets for SHARAKU which provides
measurement capabilities of the specular and off-specular reflection. Polarizing supermirrors of reflection type (Fe/Si) have been installed as a neutron spin analyzer for SHARAKU. Some of the polarizing supermirrors are stacked vertically in order to cover a wide area (100 mm horizontal, 50 mm vertical), which analyze a polarized neutron beam with wavelengths longer than 0.2 nm [3,4]. However, the stacked supermirrors are not exactly aligned at equal angles. Therefore the analyzer gives an effect to off-specular patterns. The \(^3\)He NSF is able to overcome this problem. Moreover, the \(^3\)He NSF enables a large solid angle coverage and polarization of neutron beams with shorter wavelengths by controlling the pressure length of the \(^3\)He gas [5-12]. An in-situ \(^3\)He NSF has been developed for the analyzer on SHARAKU. A new compact laser unit has been installed inside the in-situ SEOP \(^3\)He NSF to simplify the laser beam adjustment. Off-specular measurement using the in-situ SEOP \(^3\)He NSF with the new compact laser unit was performed at SHARAKU. A Fe/Cr multilayered thin film with a giant magnetoresistance effect was used as a sample [13]. The experimental setup and performance of the in-situ SEOP \(^3\)He NSF is reported in Section 2, and the results for the off-specular pattern measurements are describe in Section 3.

2. Experimental setup and performance of the in-situ SEOP \(^3\)He NSF

Figures 1(a), (b) and (c) show a schematic diagram, picture of the experimental setup, and picture of the inside of a box of in-situ \(^3\)He SEOP NSF, respectively. A V-shape transmission polarizer, which consists of Fe/Si supermirrors and a Drabkin spin flipper, has been installed in front of the sample stage [14]. The polarizer can be moved in horizontally, and we can choose polarized or unpolarized modes for experiments. SHARAKU has nine slits in its beam path. The eighth and ninth slits are shown in Figs. 1(a) and (b). There is a large area, about 1500 mm in length and 500 mm in width, between eighth and ninth slits, and the in-situ \(^3\)He SEOP NSF is set to a wide area. A guide coil is set downstream of the eighth slit to avoid neutron spin depolarization. A two-dimensional position-sensitive detector is set downstream of the ninth slit. Figure 1(c) shows the inside of the box of the in-situ \(^3\)He NSF and the compact laser unit; the quartz mirror and solenoid coil are also shown. The \(^3\)He cell is set inside of the solenoid coil. The size of the \(^3\)He cell is 35 mm in diameter and 55 mm in length. The pressure length-product of the cell is 11 bar cm. The cell was fabricated using GE180, and Rb is doped inside the cell. The quality factor, \(Q = \frac{P_n^2}{T_r}\), where \(P_n\) is the polarization of the neutron beam and \(T_r\) is the transmission intensity of neutron beam, shows similar performance to a \(^3\)He cell because the \(P_n\) and \(T_r\) are in a trade-off relationship. The \(Q\) factor curve of the \(^3\)He cell with a pressure length of 11 bar cm exhibits a wavelength peak at 0.23 nm, which is evaluated with 70\% \(P_{\text{\(^3\)He}}\). Because a neutron intensity peak of the time-of-flight (TOF) spectrum for SHARAKU is 0.25 nm, which is nearly identical to the peak of the \(Q\) factor, the \(^3\)He cell with pressure length of 11 bar cm is selected for this experiment. The relaxation time of the \(^3\)He cell was 130 h, which was estimated from a measurement of the NMR signal of \(^3\)He. Because the diameter of the cell is 35 mm, only a square area of 25 mm by 25 mm can be covered, which is not sufficient for SHARAKU. Therefore we must develop a larger \(^3\)He cell, at least 150 mm in diameter, for the SHARAKU analyzer.

A diode laser element, volume holographic grating (VHG) element, and quarter-wavelength plate are contained inside the laser unit, whose dimensions are 135 mm (L), 135mm (W), and 100 mm (H). The laser unit is a joint-developed instrument by the authors and Toyoko Kagaku Co., Ltd. The laser unit is very compact, and its portability is drastically improved. The laser output power is 30 W with a peak wavelength of 794.7 nm. The unit is cooled with an air-cooled fan, and the temperatures of the diode laser element as well as the VHG element are controlled by a Peltier element, which enables us to control the peak of the output laser beam.
A performance test on the in-situ SEOP \(^3\)He NSF was performed without a sample. The wavelength dependence of the neutron beam transmission intensity through the \(^3\)He cell was measured. The polarization of \(^3\)He gas can be obtained from the neutron beam transmission intensity of the \(^3\)He cell filled with unpolarized \(^3\)He gas \(T_0\), and those with polarized \(^3\)He gas \(T\) as follows [15],

\[
\begin{align*}
T &= T_0 e^{-\sigma \rho t} \cosh \rho_3\sigma \rho t, \\
T_0 &= T_0 e^{-\sigma \rho t}, \\
P_{3\text{He}} &= \frac{1}{\sigma \rho t} \cosh \frac{T}{T_0} \\
\sigma \rho t &= 7.97 \times 10^{-2} p \ t \ \lambda
\end{align*}
\]

where \(P_{3\text{He}}\) is the polarization of \(^3\)He, \(\sigma\) is the neutron absorption cross section for the unpolarized \(^3\)He, \(\rho\) is the number density of \(^3\)He determined, \(t\) is the length of the \(^3\)He cell, and \(T_0\) is the transmission of an empty cell. \(p\) is the \(^3\)He pressure at 0°C, and \(\lambda\) is the neutron wavelength. The wavelength dependences of \(T\) and \(T_0\) were measured with the TOF neutron beam at BL17 and the polarization of \(^3\)He \(P_{3\text{He}}\) was obtained, as shown in Fig. 2. The result shows the polarization of \(^3\)He to be 68%. Moreover the in-situ SEOP system remained stable over 5 days and the \(^3\)He gas polarization of 68% was also stable over the duration of the experiment.

Figure 3 shows the wavelength dependence of the overall instrumental asymmetry \(A\). The \(A\) was obtained from \(I_{ON}\) and \(I_{OFF}\) as follows

\[
A = \frac{I_{ON} - I_{OFF}}{I_{ON} + I_{OFF}}
\]
where $I_{\text{ON}}$ is the TOF spectrum of the neutron intensity with the Drabkin spin flipper ON and $I_{\text{OFF}}$ is TOF spectrum with the flipper OFF. The $^3$He NSF was used as an analyzer. The instrumental asymmetry was more than 95% for wavelengths longer than 0.35 nm and was stable over the off-specular measurements, which are described in the next section. Polarization efficiencies of the polarizer and Drabkin spin flipper were included in the instrumental asymmetry $A$ shown in Fig. 3.

![Figure 2. Wavelength dependence of the polarization of $^3$He.](image)

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![Figure 3. Wavelength dependence of the overall instrumental asymmetry.](image)

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3. Results of the off-specular measurement
A demonstration of off-specular measurement using the in-situ SEOP $^3$He NSF was performed at SHARAKU. The Fe/Cr multilayered thin film with the giant magnetoresistance effect was used as a sample. A 10 kOe maximum magnetic field can be applied to the sample. For low magnetic field conditions, the Fe layers create antiferromagnetic correlations that produce a specular peak and off-specular reflection. On the other hand, in a strong magnetic field, antiferromagnetic correlations are
suppressed and both the specular peak and off-specular reflection disappear [13]. Because the polarizing supermirror on the SHARAKU is a transmission type, neutron spins that are antiparallel to a guide field can be produced. On the $^3$He NSF analyzer, neutron spins that are parallel to a guide field can be transmitted. Hence, specular and off-specular reflections containing a spin flip, ($-$, $+$), can be measured using the OFF mode of the Drabkin flipper. On the other hand, those reflections with no spin flip, ($+$, $+$), can be measured using the ON mode of the Drabkin flipper. In this experiment, measurements containing a spin flip ($-$, $+$) and non-spin flip ($+$, $+$) were performed with a 200 Oe (low magnetic field condition) and 10 kOe (strong magnetic field condition) magnetic field, respectively.

Figure 4 shows the results of this measurement indicated on an $Q_x$-$Q_z$ map. $Q_x$ and $Q_z$ show the momentum transfers of the in-plane and depth structures on the Fe/Cr multilayered sample, respectively. Polarization efficiency correction was performed for all results shown in Fig. 4. Specular reflection appears at $Q_x = 0$, while off-specular reflection appears at $Q_x = \neq 0$. Figures 4(a) and (b) show the results of the $Q_x$-$Q_z$ map with an applied magnetic field of 200 Oe. Figure 4(a) is the map with a spin flip ($-$, $+$), and (b) is the map with a non-spin flip ($+$, $+$). Off-specular reflections around $Q_z = 0.8$ nm$^{-1}$ are clearly observable for both the spin flip ($-$, $+$) and non-spin flip ($+$, $+$) conditions, which indicates the existence of antiferromagnetic correlations. In the spin flip condition ($-$, $+$), the off-specular reflection is much clearer than that of the non-spin flip condition ($+$, $+$). These features are consistent with the features reported in Ref. [3].

Figure 4(c) and (d) are the results of the $Q_x$-$Q_z$ map with an applied magnetic field of 10 kOe. Figure 4(c) is the map with a spin flip ($-$, $+$), and Fig. 4(d) is the map with a non-spin flip ($+$, $+$). Off-specular reflections around $Q_z = 0.8$ nm$^{-1}$ disappeared for both the spin flip ($-$, $+$) and non-spin flip ($+$, $+$) conditions, which indicates that antiferromagnetic correlations are suppressed. These features are also consistent with the features reported in Ref. [3].

For our demonstration of the off-specular measurement at SHARAKU using an in-situ SEOP $^3$He NSF, consistent results with the features reported in Ref. [13] were successfully observed. However, the size of the $^3$He cell was not large enough to cover the entire scattering area (100 mm horizontal and 50 mm vertical) at SHARAKU. High polarizations of $^3$He gas provide high transmission neutron beam intensities and high neutron beam polarization at short wavelengths of 0.2 nm. Hence, we plan to continue the development of a larger $^3$He cell and SEOP system with a high polarization of $^3$He gas.
4. Summary

A new compact laser unit for the SEOP $^3$He NSF was installed, and a performance test of the in-situ $^3$He SEOP NSF was performed at SHARAKU. In addition we were able to achieve the following: the polarization of $^3$He reached 68 %, polarization of neutron spin was more than 95 % for wavelengths longer than 0.35 nm, and the polarization remained stable for 5 days, which shows a good performance for practical use. Moreover, the portability of the in-situ SEOP $^3$He NSF was drastically improved.

Figure 4. Off-specular measurement results indicated in the $Q_x$-$Q_z$ map. (a) and (b) are the results with an applied magnetic field of 200 Oe. (a) is the map with a spin flip (-, +) and (b) is the map with a non-spin flip (+, +). (c) and (d) are the results with an applied magnetic field of 10 kOe. (c) is the map with a spin flip (-, +) and (d) is the map with a non-spin flip (+, +).
The first test of the compact in-situ SEOP $^3$He NSF for the measurement of an off-specular reflection was performed at SHARAKU. The Fe/Cr multi-layered thin film with a giant magnetoresistance effect was used as a sample. For the low magnetic field condition with a weak guide field of 200 Oe, the off-specular reflection was clearly observed, which indicates the existence of antiferromagnetic correlations. For the strong magnetic field condition with an applied field of 10 kOe, the antiferromagnetic correlations were suppressed and the off-specular reflection disappeared. These results are consistent with those reported in Ref. [13].

A demonstration of in-situ SEOP $^3$He NSF with the new compact laser unit and off-specular measurement using the in-situ SEOP $^3$He NSF was successfully performed at SHARAKU.

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