Development of a $^3$He nuclear spin flip system on an in-situ SEOP $^3$He spin filter and demonstration for a neutron reflectometer and magnetic imaging technique

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Abstract. We have been developing a $^3$He neutron spin filter (NSF) using the spin exchange optical pumping (SEOP) technique. The $^3$He NSF provides a high-energy polarized neutron beam with large beam size. Moreover the $^3$He NSF can work as a $\pi$-flipper for a polarized neutron beam by flipping the $^3$He nuclear spin using a nuclear magnetic resonance (NMR) technique. For NMR with the in-situ SEOP technique, the polarization of the laser must be reversed simultaneously because a non-reversed laser reduces the polarization of the spin-flipped $^3$He. To change the polarity of the laser, a half-wavelength plate was installed. The rotation angle of the half-wavelength plate was optimized, and a polarization of 97% was obtained for the circularly polarized laser. The $^3$He polarization reached 70% and was stable over one week. A demonstration of the $^3$He nuclear spin flip system was performed at the polarized neutron reflectometer SHARAKU (BL17) and NOBORU (BL10) at J-PARC. Off-specular measurement from a magnetic Fe/Cr thin film and magnetic imaging of a magnetic steel sheet were performed at BL17 and BL10, respectively.

1. Introduction
Polarized neutron scattering methods are effective for studying magnetic materials, and magnetic imaging techniques are also powerful tools to investigate and visualize magnetic structures in magnetic materials [1-5]. A $^3$He neutron spin filter (NSF) is an effective device to provide a polarized neutron beam. A $^3$He NSF has some advantages compared with a polarizing supermirror, another commonly used neutron polarizing device, because the $^3$He NSF produces a polarized neutron beam with shorter...
wavelengths and cover a large solid angle [6]. The polarizing supermirror can also be made to cover a large solid angle by stacking multiple supermirrors. However the stacking precision of the supermirrors often creates a non-uniform distribution of neutron intensity, which is not an issue for the \(^3\)He NSF. Moreover the \(^3\)He NSF can be used as a \(\pi\)-flipper for a polarized neutron beam by simply flipping the \(^3\)He nuclear spin using the nuclear magnetic resonance (NMR) technique. A \(^3\)He NSF can thus provide a compact SEOP system which functions as both a neutron polarizing device and neutron spin flipper. Hence we attempted to combine \(^3\)He nuclear spin flip system with the in-situ SEOP \(^3\)He NSF that has been developed at J-PARC [7-12]. With the in-situ SEOP technique, the polarization of the laser must be reversed simultaneously with the \(^3\)He spin, because a non-reversed laser destroy the polarization of the spin-flipped \(^3\)He. The change of the polarity of the laser was accomplished using a half-wavelength plate installed after the circularly polarized laser. The details of the development of the \(^3\)He nuclear spin flip system for the in-situ SEOP system is reported in section 2, and the demonstration of the system for off-specular reflection and magnetic imaging measurements are described in sections 3 and 4, respectively.

2. Nuclear spin flip system

Figure 1 (a) and (b) show the schematic diagram and photograph of the nuclear spin flip system using a half-wavelength plate. Circularly polarized laser light was emitted from the laser unit [12] and reflected by the quartz mirror, and a \(^3\)He cell was placed after the quartz mirror. The laser unit consists of an air-cooled laser diode array (LDA) with output power of 30 W. Since the laser spectrum of the LDA is broad, a volume holographic grating (VHG) element is used to narrow the laser spectrum and match it to the Rb absorption line [12]. A half-wavelength plate (Thorlabs Inc., AHWP10M-980) was installed just downstream of the laser unit. The half-wavelength plate was set on an optical rail, and could be moved orthogonally to the laser beam direction, allowing us to change the polarity of the laser by inserting or removing the half-wavelength plate. A spectrometer (Thorlabs Inc., PAX5710IR1-T) was set after the \(^3\)He cell position in order to measure the circularly polarized light incident on the \(^3\)He cell. At first we measured the circular polarization without the half-wavelength plate, and we observed a right-handed polarized laser with a polarization of 97 %. Next, we inserted the half-wavelength plate, and after optimizing the rotation angle of the half-wavelength plate, we obtained a 97 % left-handed polarized laser light.

![Figure 1](image)

Figure 1. (a) the schematic diagram and (b) photograph of the \(^3\)He nuclear spin flip system for the in-situ SEOP system.
After finishing the optimization of the half-wavelength plate, an in-situ SEOP pumping test was performed with the $^3$He cell. An Adiabatic Fast Passage NMR (AFP-NMR) system was also installed to flip the $^3$He polarization, and the polarization of $^3$He nuclear spin was monitored via the voltage of the NMR signal $V_{NMR}$. The AFP-NMR system has two measurement modes, spin-flip mode and non-spin-flip mode, by changing the sweep function of current for the static field [11]. In this test, the $^3$He polarization was monitored every 3 hours. Figure 2 shows the pumping result for the $^3$He nuclear spin with the SEOP technique. A $^3$He cell with a pressure length of 11.1 atm cm was used in this demonstration [11]. At about 40 hours after the start of laser pumping, the $^3$He polarization was almost saturated, and the first $^3$He nuclear spin flip on the in-situ SEOP was performed, shown as “Right → Left” in Fig. 2. The change to AFP-NMR spin-flip mode and the insertion of the half-wavelength plate were performed manually and not synchronized with each other, in the present state. $V_{NMR}$ reversed with the $^3$He nuclear spin as shown in Fig. 2, and $V_{NMR}$ remained stable over 30 hours. After the first $^3$He nuclear spin flip, the spin flip was performed an additional four times, as indicated by “Left → Right” and “Right → Left” in Fig. 2. In each spin flip test, $V_{NMR}$ ($^3$He polarization) did not decay, and the $^3$He nuclear spin flip system on the in-situ SEOP worked stably over 180 hours (7.5 days). These results show that the installed $^3$He nuclear spin flip system provides good performance for the in-situ SEOP system. The maximum value of $|V_{NMR}|$ (absolute value of $V_{NMR}$) was about 34 mV, and it corresponds to a $^3$He polarization $P_{^3He}$ of about 70 %. The $P_{^3He}$ was estimated using a calibration factor obtained by simultaneous measurement of neutron transmission and AFP-NMR in another experiment, which was previously reported in ref. [11].

3. Off-specular measurement

As a demonstration for neutron scattering, we installed the system in BL17 SHARAKU which is a neutron reflectometer at J-PARC. Normally, BL17 has a neutron spin polarizer and analyser consisting of polarizing supermirrors, and experiment modes using non-polarized or polarized neutrons can be selected [13]. Drabkin-flippers are installed before and after the sample position [14], and four patterns of neutron spin combination before and after the sample, $(+, +), (−, −), (+, −)$ and $(−, +)$, can be measured. Here, “+” and “−” are neutron spin states parallel or anti-parallel to a guide magnetic field, respectively. The analyser is fabricated from supermirrors (Fe/Si, $m = 4$) with a stack structure, covering a beam area of 50 mm in height and 100 mm in width with compact size (350 mm in length) and enabling off-specular and grazing incidence small-angle neutron scattering (GISANS) measurements. In such an analyser, however, it is impossible to stack the supermirrors at exactly equal angles, producing a small
error that affects the off-specular and GISANS patterns. On the other hand, the $^3$He NSF gives a smaller effect to the scattering patterns compared with stacked supermirrors. Hence BL17 requires the $^3$He NSF as a neutron spin analyser.

The previous test of in-situ SEOP $^3$He NSF has been performed and reported in ref. 10. However the $^3$He nuclear spin flip system had not been installed in the in-situ SEOP system used in the past test. Therefore we could not select neutron spin states after the sample and could only measure two patterns of neutron spin states ($-$, $-$) and ($+$, $+$) as reported in ref. [10].

Figure 3 shows the photograph and schematic diagram of the current demonstration. In this demonstration, the pair of the Drabkin flipper and the analyser mirror that are normally placed between the $8^{th}$ slit and $9^{th}$ slit were exchanged for the in-situ SEOP $^3$He NSF system with $^3$He nuclear spin flip system. This system then enabled us to measure four patterns of neutron spin states ($+$, $+$), ($+$, $-$), ($-$, $+$) and ($-$, $-$) as shown in Fig. 3.

The $^3$He cell with a pressure length of 11.1 atm cm with 35 mm in diameter and 55 mm in length was used. The cell was fabricated using GE180, and Rubidium is doped inside the cell. The $^3$He nuclear polarization was monitored by AFP-NMR, and $V_{NMR}$ was about 29 mV which corresponds to 60 % of polarization. That is lower than that of the off-line test (without neutron beam). The reason is thought to be due to the 25 Oe guide field generated from the guide coil placed about 300 mm upstream of the $^3$He cell. However, the SEOP $^3$He NSF system worked stably over the experiment time, and off-specular measurements could be performed.

A Fe/Cr multilayer thin film with the giant magnetoresistance effect was used as a sample. In low magnetic field, the Fe layers create antiferromagnetic correlations that produce an off-specular reflection around $Q_z = 0.8 \text{ nm}^{-1}$. This was the same sample used in the past experiment using in-situ SEOP at BL17 [10], and its features have already been reported in ref. [15]. In this experiment, the magnetic field applied to the sample was 200 Oe, and the off-specular reflections with four patterns ($+$, $+$), ($+$, $-$), ($-$, $+$) and ($-$, $-$).
and (-, -) were measured. Figure 4 shows the results of the off-specular measurements indicated on $Q_x$-$Q_z$ map. $Q_x$ and $Q_z$ show the momentum transfers of the in-plane and depth structures on the sample, respectively. Off-specular reflections appear at $Q_x \neq 0$. In all off-specular patterns shown in Fig. 4, off-specular reflections are observed at about $Q_z = 0.8 \text{ nm}^{-1}$. Moreover the off-specular patterns of (+, -) and (-, +) are almost identical. These features are consistent with the features reported in Ref. [15].

BL17 requires a large $^3$He cell of about 150 mm in diameter, because the detector on BL17 has a 100 mm $\times$ 100 mm area. An improvement of the $^3$He nuclear polarization is also necessary. We have recently started to develop a potassium-doped hybrid-cell to improve the $^3$He nuclear polarization. Moreover we must adjust the pressure length of $^3$He gas to match the peak intensity of the neutron beam at BL17. However, the $^3$He nuclear spin flip system on the in-situ SEOP was demonstrated successfully and worked stably over 4 days of experiment time.

Figure 4. The results of the off-specular scattering measurement from Fe/Cr multilayer thin film with four neutron spin states.

4. Magnetic imaging

As another demonstration of the $^3$He nuclear spin flip system using in-situ SEOP, we applied the system to a magnetic imaging technique at BL10 NOBORU (NeutrOn Beamline for Observation and Research Use) at J-PARC. When the neutron spin is depolarized entirely, it becomes impossible to obtain any information in a magnetic sample. For a magnetic sample with a strong magnetic field, the neutron spin becomes increasingly easy to depolarize. Hence using polarized neutrons with shorter wavelengths is very important, because shorter wavelength neutrons are not as easy to depolarize compared with longer wavelengths. The $^3$He NSF is effective to provide polarized neutron with shorter wavelengths compared with a polarizing supermirror, therefore we started to apply the $^3$He NSF to the magnetic imaging technique.

The photograph and schematic diagram of the experiment setup are shown in Fig. 5 (a). The in-situ SEOP $^3$He NSF with the $^3$He cell with pressure length of 17 atm cm was used as a neutron spin polarizer,
and the ex-situ (not in-situ) one with that of 11.1 atm cm was used as a neutron spin analyser. The $^3$He cell size was 35 mm in diameter and 55 mm in length for both. There was insufficient space for setting two in-situ SEOP systems in the BL10 experiment room, so in-situ SEOP was used only for the polarizer, including the $^3$He nuclear spin flip system. An RPMT, a two-dimensional position sensitive detector consisting of a scintillator and position-sensitive photomultiplier tube, was placed after the analyser [16]. The sample was placed between the polarizer and the analyser.

A polarized neutron beam parallel/antiparallel to the guide field can be obtained without/with application of the $^3$He nuclear spin flip on the in-situ SEOP polarizer. After rotating around a sample magnetic field $B$, the neutron spins are analysed by the ex-situ SEOP. Then $I_+$ and $I_-$ can be measured without and with application of the $^3$He nuclear spin flip system, respectively. Neutron spin polarization $P_{NS}$ can then be obtained from $P_{NS} = (I_+ - I_-) / (I_+ + I_-)$.

A magnetic steel sheet in the shape of a ring with a thickness of 0.35 mm was used as the sample as shown in Fig. 5 (b). About three-fourths of the sample was wrapped with a solenoid coil to apply and control a magnetic field to the sample. A neutron beam with a size of 15 mm $\times$ 15 mm was irradiated to the unwrapped portion of the magnetic steel sheet.

The $^3$He polarization of the polarizer was 40 %. Our laser unit has air cooling system [12], but in this experiment, the experiment room of BL10 was very hot and the air cooling system could not provide sufficient cooling. To prevent overheating, the laser power had to be reduced, leading to the lower $^3$He polarization. However the $^3$He nuclear polarization was kept stable over the experiment time. The $^3$He NSF for the analyser was polarized before starting the magnetic imaging measurements. However the analyser was not in-situ SEOP, so the $^3$He nuclear polarization decayed with time. The $^3$He nuclear polarization before the start of the experiment was about 60 %, and during the 30 hours of the experiment, the polarization was reduced to about 40 %, where these percentages were estimated from the AFP-NMR measurements. The polarization of the direct neutron beam (without sample) was also reduced. Figure 6 shows the change of the neutron beam polarization, where the circles and triangles show the polarization before and after the magnetic imaging measurements.

Figure 5. (a) Photograph and schematic diagram of the experiment setup, and (b) the ring shape of magnetic steel sheet.
Figure 6. The polarization of neutron beam before (circle) and after (triangle) the magnetic imaging measurements.

Figure 7 shows the results of the magnetic imaging measurement of the magnetic steel sheet. Figure 7 (a) and (c) are two-dimensional (2D) images of neutron polarization at a neutron wavelength of 0.27 nm (1 pixel = 0.5 mm). Initially, we applied a current of $I = 1.42$ A to the solenoid coil on the sample, and the 2D image of the neutron polarization is shown in Fig. 7 (a). Next, the current was reduced to 0 A, and the resulting 2D image is shown in Fig. 7 (c). In both of these images, the neutron polarization $P_{NS}$ is normalized by the polarization of the direct beam for each measurement time $P_{ND}(t)$, where $t$ is the measurement time. Then we obtain the normalized neutron polarization from $P_N = P_{NS}/P_{ND}(t)$. Figure 7 (b) shows the wavelength dependence of $P_N$ extracted from the square region indicated in Fig. 7 (a). Similarly, figure 7 (d) shows $P_N$ from the square region indicated in Fig. 7 (c). The square regions in Fig. 7 (a) and (c) are at the same position on the sample. The wavelength dependence of $P_N$ is caused by a rotation of neutron spin around a magnetic field. The oscillation cycle shows an integral of magnetic field of a neutron path, and depolarization of neutron spin leads to a decay of the amplitude of $P_N$ [3, 11]. Both the 2D images and the wavelength-dependent polarization show drastically different patterns for the $I = 1.42$ A and $I = 0$ A cases. In this measurement, only the magnetic component perpendicular to the quantization axis of the neutron spin was measured, therefore it is impossible to determine the intensity and direction of magnetic field quantitatively. However, these results clearly show that the distribution of magnetization in the magnetic steel sheet changed with different applied magnetic field.
Figure 7. The results of the magnetic imaging measurements. (a) and (c) are 2D images of $P_N$ from $I = 1.42$ A and $I = 0$ A at $\lambda = 0.27$ nm, respectively. (b) and (d) show the wavelength dependence of $P_N$ of same regions shown in (a) and (c). The pixel size of the 2D maps (a) and (c) is 0.5 mm.

In this demonstration, the $^3$He nuclear spin flip system on the in-situ SEOP worked stably during the entire experiment time of 30 hours, and magnetic imaging using polarized neutrons at shorter wavelengths was successfully performed. However neutron polarization of the direct beam was not high, for example $P_N = 0.2$ at $\lambda = 0.1$ nm. We are planning to use the polarized neutron with $\lambda = 0.05$ nm for a strong magnetic field sample, for example a stacked magnetic steel sheet and electrical motor. Therefore big improvements are necessary for all components of the in-situ SEOP $^3$He NSF system as follows: 1) a high $^3$He nuclear polarization of more than 80%, 2) a high pressure length of $^3$He cell (of more than 45 atm cm), and 3) a laser unit with power greater than 100 W in a compact size of less than 150 mm in length, 150 mm in width and 100 mm in height. Moreover, as excessive distance between the sample and detector leads to blurring of the 2D image due to beam divergence, an extremely compact in-situ SEOP system (with a length less than 300 mm) for the neutron spin analyser is needed. Therefore the extremely compact analyser system is very important for the magnetic imaging technique. Accomplishment of these requirements is not easy, but we must continue to develop each component.

5. Conclusion
A $^3$He nuclear spin flip system for an in-situ SEOP $^3$He NSF has been developed using a half-wave plate. Pumping and nuclear spin flip tests were performed, and the system worked stably over 180 hours (7.5 days) with several $^3$He nuclear spin flips.

A demonstration of the $^3$He nuclear spin flip system for the in-situ SEOP was performed at the neutron reflectometer SHARAKU (BL17) at J-PARC. The in-situ SEOP $^3$He NSF was used as a neutron spin analyser. A $^3$He cell with a pressure length of 11.1 atm cm, a diameter of 35 mm, and a length of 55 mm was employed, and a Fe/Cr multi-layered thin film was used as a sample. Off-specular measurements were performed, and the $^3$He nuclear spin flip system enabled us to measure four neutron spin states, (+, +), (+, -), (-, +) and (-, -). The applied magnetic field was 200 Oe, and off-specular patterns were observed at $Q_z = 0.8$ nm$^{-1}$. The off-specular patterns with neutron spin flipped (+, -) and
(-, +) were almost the same. These results are consistent with features reported in ref. [15]. The demonstration was successfully performed, however we need to continue some development to optimize the system for use at BL17, including improvement of the $^3$He nuclear polarization to 80%, enlargement of the cell size to cover 100 mm $\times$ 100 mm areas, and optimization of the $^3$He gas pressure length to the neutron wavelength distribution at BL17.

Next, we applied the in-situ SEOP to a magnetic imaging technique. The in-situ SEOP using a $^3$He cell with pressure length of 17 atm cm was used as a neutron spin polarizer, and the ex-situ SEOP with pressure length of 11.1 atm cm was used as a neutron spin analyser. By using the $^3$He nuclear spin flip system, the intensities of both parallel and anti-parallel spin polarized neutrons could be measured, and we could obtain the neutron spin polarization $P_N$. A magnetic steel sheet with solenoid coil was used as the sample. By changing the current in the solenoid coil, magnetic field applied to the sample could be controlled, and magnetic imaging measurements with two current conditions, $I = 1.42$ A and $I = 0$ A, were performed. 2D images of $P_N$ were obtained, and different $P_N$ patterns between the two conditions were observed. Moreover the observed wavelength dependences of $P_N$ for a specific area of the sample were also obviously different for each condition, showing again that the magnetization patterns between the two conditions were altered. This demonstration was also successfully performed, however many improvements are still needed including a higher $^3$He nuclear polarization of more than 80%, a high pressure length of more than 45 atm cm, and an extremely compact in-situ SEOP system with length less than 300 mm for the neutron spin analyser.

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