New generation high performance in situ polarized $^3$He system for time-of-flight beam at spallation sources

C. Y. Jiang, 1 X. Tong, 1,a) D. R. Brown, 1 A. Glavic, 2,3 H. Ambaye, 1 R. Goyette, 1 M. Hoffmann, 1 A. A. Parizzi, 1 L. Robertson, 1 and V. Lauter 2

1Instrument and Source Division, Neutron Sciences Directorate, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393, USA
2Quantum Condensed Matter Division, Neutron Sciences Directorate, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393, USA
3Laboratory for Neutron Scattering and Imaging, Paul Scherrer Institute PSI, Villigen, Switzerland

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Modern spallation neutron sources generate high intensity neutron beams with a broad wavelength band applied to exploring new nano- and meso-scale materials from a few atomic monolayers thick to complicated prototype device-like systems with multiple buried interfaces. The availability of high performance neutron polarizers and analyzers in neutron scattering experiments is vital for understanding magnetism in systems with novel functionalities. We report the development of a new generation of the in situ polarized $^3$He neutron polarization analyzer for the Magnetism Reflectometer at the Spallation Neutron Source at Oak Ridge National Laboratory. With a new optical layout and laser system, the $^3$He polarization reached and maintained 84% as compared to 76% in the first-generation system. The polarization improvement allows achieving the transmission function varying from 50% to 15% for the polarized neutron beam with the wavelength band of 2–9 Angstroms. This achievement brings a new class of experiments with optimal performance in sensitivity to very small magnetic moments in nano systems and opens up the horizon for its applications. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4975991]

I. INTRODUCTION

Polarized $^3$He neutron spin filters have been widely used as neutron polarizers or analyzers at all major neutron facilities. 1–5 In particular, recently the in situ $^3$He system based on spin-exchange optical pumping (SEOP) which enables continuous pumping of $^3$He on a neutron instrument has gained interest among the neutron scattering community 6–10 because it is capable of overcoming the decay of $^3$He polarization present in a conventional ex situ pumped system. At Oak Ridge National Laboratory (ORNL), the first in situ system was commissioned in 2012 as a neutron polarization analyzer at the Magnetism Reflectometer (MR) 6 at the Spallation Neutron Source (SNS), which achieved 76% $^3$He polarization. This system gave an average analyzing efficiency of 98% and 25% polarized neutron transmission from 2 to 5 Å wavelength band, for neutrons that are spin parallel to the $^3$He. The overall size of the system, particularly the length (100 cm) along the neutron path, requires a sufficient space thus limits the use on other neutron instruments at ORNL. Later, a more compact 2nd generation in situ system (50 cm along the neutron path) was developed for the triple-axis spectrometer HB3 as a polarizer at the High Flux Isotope Reactor (HFIR) with 71% $^3$He polarization achieved. 7 Compared to the previous version, the 2nd generation system has a simplified optical layout and a smaller solenoid while still retaining the size of the maximally usable $^3$He cell. Encouraged by the results, a new generation of the high performance in situ polarized $^3$He system for the time-of-flight (TOF) beam at the MR was developed. The $^3$He polarization produced with the new optical system attained 84% and was maintained for 3 weeks. High $^3$He polarization resulted in the transmission function varying from 50% to 15% for the polarized neutron beam with the wavelength band of 2–8 Å. Here we report the design of the new optical system, construction, parameters, and neutron experimental data obtained with the new assembly. A distinct aspect of this system lies in its ability to be effectively used in the time-of-flight instruments with a broad wavelength band, which was not available until now.

II. $^3$He CELL OPTIMIZATION FOR BROAD WAVELENGTH BAND NEUTRONS

The main challenge of using the in situ polarized $^3$He system for time-of-flight (TOF) beam is achieving high neutron polarization and transmission over a broad wavelength band, and high $^3$He polarization is the essential key to this goal. The polarization and transmission for an unpolarized neutron beam passing through a polarized $^3$He cell can be described by the following two equations:

$$P_n(\lambda) = \tanh(O P_{He}),$$ (1)

$$T_n(\lambda) = T_e \exp(-n \sigma_0 \lambda) \cosh(n \sigma_0 \lambda P_{He})$$

$$= T_e \exp(-O) \cosh(O P_{He}),$$ (2)

where $T_e$ is the transmission through $^3$He cell windows ($T_e = 0.87$ is assumed for all calculations in this section 11), $P_{He}$ is

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*a)tongx@ornl.gov

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the $^3$He polarization, and $O$ is the opacity of the cell and has the relation of

$$O = n \sigma_0 l \lambda,$$  \hspace{1cm} (3)

where $n$ is the number density of $^3$He atoms, $l$ is the cell length, $\sigma_0$ is the absorption cross section for 1 Å wavelength neutron, and $\lambda$ is the neutron wavelength.

A trade-off between $P_n$ and $T_n$ occurs naturally and an optimization of $n * l$ (proportional to $^3$He cell pressure * length) is strongly dependent on the experimental conditions and requirements. Such an optimization is usually done by maximizing the figure of merit (FOM) defined by the following equation:

$$FOM(O, P_{^3He}) = P_n^2 T_n.$$  \hspace{1cm} (4)

Thus, the opacity $O$ that maximizes the FOM can be calculated depending on the $^3$He polarization and is given by the following equation:

$$O(P_{^3He}) = \frac{1}{2P_{^3He}} \ln \frac{2P_{^3He} - 1 + \sqrt{8P_{^3He}^2 + 1}}{2P_{^3He} + 1 - \sqrt{8P_{^3He}^2 + 1}}.$$  \hspace{1cm} (5)

Figure 1 shows a calculated behavior of $O(P_{^3He})$ as a function of $P_{^3He}$, unveiling that the opacity stays relatively flat around $O = 2$ for $P_{^3He} < 80\%$ with a breakthrough increase at $P_{^3He} > 80\%$.

A broad local minimum with $O_{\text{min}} = 1.865$ occurs around $P_{^3He} = 55\%$.

Using Eq. (5), the optimization of $n * l$ can be easily achieved for monochromatic neutron beam but becomes elusive for a neutron beam with broad wavelength band. To overcome this problem, we propose to use the following scheme for the optimization. First the $n * l$ is calculated based on $O = 2$ at a specific wavelength $\lambda_0$ within the band, then assuming a representative $^3$He polarization of 80%, the figure of merit $FOM_{av}$ averaged over the wavelength band is calculated as follows:

$$FOM_{av}(\lambda_0) = \int_{\lambda_{\text{min}}}^{\lambda_{\text{max}}} FOM(O = 2) s(\lambda) d\lambda,$$

where $\lambda_{\text{min}}$ and $\lambda_{\text{max}}$ correspond to the minimum and maximum of the wavelength band and $s(\lambda)$ represents the weight of neutrons at wavelength $\lambda_i$, which depends on the neutron spectrum. The $\lambda_0$ that maximizes the value of $FOM_{av}(\lambda_0)$ will then be used for the cell thickness optimization. An example is shown as following: Assuming a uniformly distributed neutron spectrum with wavelength band between 2 and 8 Å, Figure 2 shows $FOM_{av}(\lambda_0)$ as a function of wavelength, which maximizes at about 5.4 Å. Thus, the cell thickness will be optimized at 5.4 Å so that $n\sigma_0 l = 5.4 = 2$.

It is vital to achieve high $^3$He polarization for good performance of the polarizer, as demonstrated below. For the wavelength band mentioned above and optimized $^3$He cell thickness at 5.4 Å ($n\sigma_0 l = 2 = 3.7$), a series of plots of neutron transmission ($T$) and polarization ($P$) vs. $^3$He polarization (from 60% to 85% in 5% increment) are shown in Figure 3 which clearly demonstrates increasing $^3$He polarization improves both the polarization and transmission for all neutron wavelengths, but more dramatically higher $^3$He polarization increases neutron transmission at longer wavelengths and neutron polarization at shorter wavelengths.
The FOM used in these considerations is a good measure for experiments where the spin-flip is comparable to the non-spin-flip signal. However, if the intensities of the signals of the spin-flip and non-spin-flip channels are different by several orders of magnitude, the spin-flip signal is extremely difficult to measure even with a good neutron transmission through the analyzer. The measurement requires a very high neutron polarization with 1-P being comparable to the signal ratio. In such a case, it is advantageous to choose a better average polarization and tolerate the increased counting times needed due to the lower transmission, as is common in reflectometry and off-specular scattering measurements.

III. DESIGN AND CONSTRUCTION

The goal of the in situ ³He system is to provide a high performance ³He analyzer for the MR beamline with high and stable ³He polarization. Development of high performance features such as the laser safety interlock, real-time ³He polarization monitoring, NMR flipping, and motorized movement of the system are essential for effective operation during experiments.

A schematic picture of the system is shown in Figure 4. The outer frame in Figure 4(a) is manufactured from stainless steel with sections welded together to provide a rigid and sturdy structure. The system is enclosed using laser safety panels (Figure 4(b)), with all but one side being attached with security Torx screws to prevent unauthorized access as well as limiting the need for interlock switches on every panel of the enclosure. The front main access panel has two interlock switches that will shut down the laser system and lock it out of operation if the panel is detached. All connections pass through bulkhead connectors, including the heated air connections for the oven and the cooling water that keeps the µ-metal within its operating temperature range. The enclosure also incorporates cooling fans that maintain airflow inside the assembly and serves to maintain a reasonable temperature (below 50 °C) for the optics and other electronics associated with the laser system. A heater box is mounted on top of the frame and supplies heated compressed air into the ³He cell. The heater is interlocked with the airflow for safe operation at the beamline. Both vertical side panels have openings for 0.5 mm thick silicon windows for incoming and outgoing neutron beam to pass through.

The base structure of the system is a cut-to-fit optical breadboard that has 1/4 in.-20 threaded holes in a 1 in. spacing pattern across the surface of the breadboard for easy installation and adjustment of the optical layout. The system implements a one-layer optical layout as shown in Figure 5. A 200 W fiber-coupled laser with FWHM = 0.35 nm is used to polarize the ³He cell. The laser beam is split into two linearly polarized beams by the polarizing beam splitter. Two 3 in. diameter liquid crystal retarders serve as quarter wave plates and circularly polarize the two laser beams before they are directed to the ³He cell. These retarders have a 500 W/cm² laser damage threshold and a reflectance less than 0.5%, therefore they are well suited for our application. The cell named “Hokie” is fabricated in house using GE180 glass and it has an outer diameter of 7 cm and an overall length of 8 cm. The cell is filled with a mixture of 2.3 bars of ³He gas, 0.12 bar of N₂ gas, and traces of rubidium and potassium. It should be noted that the cell was not specifically designed and pressure optimized for the wavelength band 2–4.5 Å, but was the best choice of available cells. New cells that are optimized for a broader wavelength band of 2–8 Å are currently being fabricated. A µ-metal enclosed solenoid provides a 15 G magnetic field at the cell position, details about the solenoid can be found in our previous paper.

The optical layout is shown in Figure 5. The stray light intensity in the system is monitored using a photodiode. This additional photodiode is interlocked with the laser. If the amplitude of the signal from this photodiode was to drop below a preset level due to breakage of the laser fiber, the laser will be shut off automatically to prevent the fiber from overheating and to avoid a safety hazard. Such a feature is essential to meet the safety regulations in user facilities.

The magnitude of the ³He polarization is monitored in situ using a nuclear magnetic resonance (NMR) technique called free induction decay (FID) which provides a voltage readout that is proportional to the ³He polarization. In addition, the photodiode (different from stray light photodiode) shown in Figure 5 provides the electron paramagnetic resonance (EPR) capability which measures the absolute value of ³He polarization. Finally, to measure all four neutron spin cross sections during experiments with polarization analysis, the ³He polarization needs to alternate between two states, i.e., + and −, in a timely manner. We have installed the adiabatic fast passage (AFP) mechanism that flips ³He spins via a wave packet through electronics; such a control is scripted and

FIG. 4. (a) The scheme of the outer frame and the heater box; (b) the assembled system with laser safety panels installed. All the panels except the front main access panel use Torx screws to limit access; the positions marked with stars on the front panel have limit switches behind them.
FIG. 5. The optical layout of the system.

combined with data collection software which is directly accessible by users. The AFP flipping loss of 0.1% per flip is determined by fitting and flipping ³He spins 20 times.

The helicity of the circular polarization of the laser is controlled by adjusting the peak voltage of a square waveform applied to the liquid crystal retarders. The voltage value must also be reversed accordingly to continue to optically pump the cell in the new polarization direction after each AFP flip. The data acquisition control system (DAS) simultaneously sets pre-established new values for the liquid crystal peak voltages according to the desired final spin state as the analog I/O module is commanded to generate the RF voltage waveform for the AFP. This sequence is synchronized and automated from the data acquisition integrated Python scripting engine (PyDAS¹⁷), according to the desired spin state changes scheduled by the user through its associated graphical user interface (GUI). During the three-week run, the liquid crystal retarders maintained stability and performed as desired.

The entire system weighs approximately 110 kg and is mounted onto a stainless steel plate. The system can be lifted out of the neutron beam or lowered down in the beam (see Figure 6) by using a motor driven linear slide. The dimension of the system is 69 cm in length, 59 cm in width, and 54 cm in height.

FIG. 6. The overview of the mounted system. The system is mounted on a motorized linear vertical slide and can be reproducibly moved down in the neutron beam and up out of neutron beam without being switched off.

IV. COMMISSIONING AND PARAMETERS

To investigate the performance of the ³He system, we conducted a detailed study and series of measurements of the system in the experimental conditions on the Magnetism Reflectometer and commissioned it for operation in experiments.

First, the transmission measurements of the unpolarized neutron beam were conducted to derive the ³He polarization and the cell thickness; the polarized neutron transmission measurements were used to measure the flipping ratios.

The unpolarized neutron transmission is described by the following two equations:

\[ T_0(\lambda) = T_0 \exp(-n\sigma_0\lambda), \quad (6) \]

\[ T_n(\lambda) = T_0 \cosh(n\sigma_0\lambda P_{He}), \quad (7) \]

where \( T_0 \) is the transmission through unpolarized ³He and \( T_n \) the transmission through polarized ³He. The first fit of \( T/T_n \) (Figure 7) gives the product of \( n\sigma_0 P_{He} \) to be

\[ n\sigma_0 P_{He} = 1.098 \pm 0.001 \text{ Å}^{-1}. \]

The transmission through the cell glass windows and materials that are in the beam was measured previously to be \( T_e = 0.82 \).

From the fit to \( T/T_0 \) (Figure 8), the cell thickness can be readily obtained as follows:

\[ n\sigma_0 = 1.311 \pm 0.001. \quad (8) \]

Combining with the first fit result, we obtain the ³He polarization to be \((84 \pm 1)\%\). The fits were performed using a standard least square refinement on the data up to 7 Å and using the covariance matrix to estimate the errors on the resulting parameters. The upper plot in Figures 7 and 8 shows the difference between model and data divided by the individual standard deviations. We attribute the rather large \( X^2/\text{degrees of freedom} \) values to systematic deviations of data points below \( \sim 3.7 \) Å caused by the attenuation in the windows below a Bragg-edge. Fits performed on a limited data range above this wavelength yield \( X^2/\text{DoF} \) close to 1 with no changes to ³He polarization value but increased error bars. We have also

FIG. 7. Ratio of unpolarized neutron transmission through the polarized \( T_n \) and unpolarized \( T_0 \) ³He cell as a function of the neutron wavelength (bottom) and residuals used for refinement (top, \( X^2/\text{DoF} = 4.5 \)).
confirmed the stability of our result by performing logarithmic fits to the data and by also fitting \( T_e \), which all resulted in \(^3\)He polarization values between 82\% and 85\%.

EPR measurement was carried out to confirm and cross-check the \(^3\)He polarization. The EPR splitting between the spin up and the spin down states was measured to be 26 KHz.

V. EXPERIMENTAL TEST RESULTS

The \(^3\)He system is installed at the Magnetism Reflectometer at the Spallation Neutron Source of Oak Ridge National Laboratory\(^ {22} \) as the neutron polarization analyzer for polarized neutron reflectometry (PNR) experiments. PNR employs the polarization analysis method to unravel the magnetization arrangement in thin films and multilayers. In combination with the polarized neutron off-specular scattering, it provides information on the depth magnetization vector distribution, lateral domain arrangement, and interfacial magnetic roughness.\(^ {23-26} \)

Using PNR, the profile of magnetization vector distribution can be measured with an accuracy of a few kA/m with depth resolution \( \sim 1 \) nm. Off-specular polarized neutron scattering probes the in-plane magnetization distribution over the length scales from hundreds of nanometers to hundreds of micrometers and in combination with specular PNR provide information on the lateral long range fluctuations of the magnetization vector and magnetic domains.\(^ {25-27} \)

It was successfully applied to unravel the spiral magnetization vector distribution in exchange spring bilayers,\(^ {28} \) the domain structure in magnetic exchange-coupled multilayers\(^ {29} \) or even more complicated technologically relevant systems of laterally stripe-patterned exchange-biased films.\(^ {30} \)

Quantum-mechanical effects of Larmor pseudo-precession of neutron polarization vector at reflection can also be observed.\(^ {31} \)

To discriminate the magnetic and non-magnetic off-specular scattering, polarization analysis of the scattered neutrons is needed for the divergent neutron beam across a broad range of wavelengths with a high level of analyzing power and good transmission. Our in situ \(^3\)He analyzer is ideal for this purpose.

To examine the properties of the \(^3\)He assembly, we performed a test experiment on a multilayer structure grown with
molecular beam epitaxy on a sapphire substrate. The sample is exchange coupled single crystalline $^{57}$Fe/Cr $\times$ 12 superlattice in multi-domain state with four-fold symmetry.$^{32}$ The thickness of the chromium layer of 8 9 Å provides antiferromagnetic (AF) orientation of magnetization in the alternating Fe layers. The in-plane external magnetic field was applied along one of the axes of easy magnetization and induces a spin-flip phase in the system with layer magnetization turned perpendicular to field. The sample was studied in detail before$^{32}$ and was chosen here for the test due to its rich features in spin-flip off-specular scattering. The PNR experiment was performed in an external magnetic field of 30 mT applied in plane of the sample after saturation in a field of 1 T. Using the polarized neutron beam with polarization 98.5%$^{33}$ and a wavelength band from 2.6 to 8.6 Å, a resonance spin-flipper in front of the sample and flipping the $^3$He polarization as described in Section III, we measured the four “+ +,” “– +,” “+ –”, and “– –” neutron spin cross sections. First/second signs designate the polarization of the neutron spins before/after the sample, and “+” and “–” designate the neutron polarization with respect to the direction of the external field at the sample position.

The experiment is performed with two wavelength bands of 2.6 Å–5.6 Å and 3 Å–8 Å and several values of the incident angles to cover a broad band of momentum transfer $Q$. Figure 10 depicts the experimental two-dimensional intensity maps for “+ –” and “– –” neutron cross sections shown as functions of the perpendicular to the sample surface component of momentum transfer $Q_z$ and $p_f - p_i$. The $p_i$ and $p_f$ are the perpendicular components of the incident $k_i$ and scattered $k_f$ neutron wavevectors ($k_{zf} = 2\pi \sin(\theta_{zf})/\lambda$), as shown in the scheme at the right hand side of Figure 10 (see Ref. 34 for detailed explanation). The experiment confirmed the high quality of polarization analysis provided by the new $^3$He assembly.

In addition, to test the stability of the $^3$He analyzer, the system was left operating continuously for about 3 weeks. During this time, the $^3$He polarization remained stable as measured with NMR.

The $^3$He analyzer opens up new capabilities and enables us to perform experiments on the magnetic multilayers with non-collinear magnetization vector distribution in helical magnetic structures in Dy/Y superlattice, magnetic vector distribution in novel magnetic inherent nanolaminate Mn$_3$GaC films, magnetization depth profile in continuous and laterally structured FeRh films.

VI. DISCUSSION

In this paper, we present the results on the redesign and manufacturing of the new in situ $^3$He analyzer for the Magnetism Reflectometer. The $^3$He polarization improved from 76% to 84%, as a direct result from better laser alignment and the use of a new fiber delivered, narrower spectrum width laser source. Due to the two-layer structure and the use of stacked laser bars in previous system, the alignment and shaping of the laser light were incredibly difficult thus was by no means perfect in terms of uniform power distribution and parallel pumping, both of which is critical in achieving maximum $^3$He polarization.$^{35-37}$ In the current setup, we made the change such that all optics are on the same layer which has greatly simplified the laser alignment. In addition, we used a 200 W fiber coupled laser (FWHM $\sim$ 0.35 nm) in the new system which reduced the number of laser shaping lenses and offered a more uniform power distribution across the laser beam.

Our $^3$He system can accommodate cells up to 11 cm in diameter as compared to the current cell diameter of 6 cm. The use of a bigger cell will increase the solid angle.
coverage. Also, by shortening the distance between the sample stage and the $^3$He system, the solid angle can further be enlarged. The fabrication of bigger cells and the new system design are both in development but outside the scope of this paper. New updates will be shown in future works.

VII. CONCLUSION

The advanced high performance in situ polarized $^3$He system for the time-of-flight Magnetism Reflectometer at the SNS is developed. The $^3$He polarization obtained with the new-generation optical pumping system establishes a new record value of $(84 \pm 1)\%$. To our knowledge, this is the highest polarization ever reached in in situ systems worldwide. The high $^3$He polarization is achieved by using the new fiber delivered laser source and through an optimization of the laser alignment. The entire system is remotely controlled and monitored from the instrument control room including the NMR and EPR measurements. The increased $^3$He polarization resulted in further enhancement of the transmission of the $^3$He neutron polarization for several weeks of operation without interruption.

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15See https://neutrons.ornl.gov/mr for information about the polarized neutron capability.


