SEOP program for neutron spin filters at the Jülich Centre for Neutron Science

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Abstract. The Jülich Centre for Neutron Science is developing spin-exchange optical pumping systems capable of polarizing the $^3$He gas for use as neutron spin filters with developments focused on individually optimized solutions for each application. When possible steady polarization achieved with in-situ optical pumping will provide high time averaged neutron performance and time-stability, however some applications are foreseen to use a “local filling station” approach. In-situ polarizers are being developed for polarization analysis on reflectometry and small angle neutron scattering instruments. This overview of the current status of our SEOP program describes the motivation of in-situ polarization, the design and first results from the polarizer for analysis on reflectometry, and discusses the designs for a SANS analyzer and plans for wide angle analysis.

1. Introduction: Polarized $^3$He used for neutron spin analysis
Polarized $^3$He has been shown to be advantageous when used as an analyzer for polarized neutron scattering, an area of physics research that is growing quickly. These sorts of experiments are normally conducted in large scale facilities for neutron scattering research where even at the highest flux neutron sources these experiments can require long data collection times. This is partly because of the losses in neutron counting rates caused by the addition of neutron polarizers and analyzers, but also because one now normally must measure two to four combinations of the incident beam polarization and analyzer polarization, essentially performing the same experiment several times. Further factors such as small sample size in the case of magnetism studies or low angular coverage of the scattered beam further lengthen these types of experiments.

Much work has been done to optimize the use of polarized $^3$He for use as neutron spin filters ($^3$He NSF). Routinely experiments using $^3$He NSF are conducted at many neutron sources worldwide. These experiments include polarized neutron diffraction, polarized neutron reflectometry, neutron interferometry, polarized small angle neutron scattering and fundamental particle physics [1–6].

Two methods are typically used to polarize $^3$He gas, spin-exchange optical pumping (SEOP) and metastable-exchange optical pumping (MEOP) [7,8]. In recent years both methods have been achieving ever higher understanding of the physical processes limiting the magnitude and rates of $^3$He polarization. This along with technological achievements have produced $^3$He polarizations of up to 80% in $^3$He NSF cells that have been verified with neutron measurements.
[1,9–11]. These levels of performance make $^3$He NSFs highly applicable in many areas of polarized neutron instrumentation.

This overview of the current status of the Jülich Centre for Neutron Science (JCNS) SEOP program will describe the motivation of in-situ polarization, the design and first results from our polarizer designed for reflectometry/grazing incidence small angle neutron scattering (GISANS), the designs for a soft matter SANS analyzer currently under construction, and instrumentation planning to use the “local filling station” approach.

2. Advantages of in-situ $^3$He polarization

The polarization of $^3$He gas, being an atomic physics process, is normally performed in the controlled conditions of an atomic-molecular-optical physics laboratory because conditions involving high power class IV lasers, highly uniform magnetic fields and isolation from electronic and magnetic interference are required. However $^3$He polarization is a dynamic process involving spin-polarization and $T_1$ decay of nuclear spin, thus once the $^3$He is polarized and in an appropriate container, and the polarization from optical pumping is no longer taking place, it begins to undergo $T_1$ relaxation to 0 polarization. This polarization decay places certain limits on experiments using off-line polarized gas.

In practical terms, $T_1$ lifetimes of 100 hours to 300 hours are the achievable goal in neutron experiments using $^3$He NSFs. The neutron energy dependent absorption cross section for $^3$He is nearly entirely spin dependent and the neutron polarization efficiency $P_n$ is related to the $^3$He polarization, $P_{He}$, and the product of the $^3$He density-length and neutron cross section, $\Theta$, by $P_n = tanh(P_{He} \Theta \lambda_n)$ where $\lambda_n$ is the neutron wavelength. The increase in neutron transmission relative to the unpolarized transmission of the $^3$He NSF is given by $T_n/T_0 = cosh(P_{He} \Theta \lambda_n)$ where $T_0$ is $e^{-\Theta \lambda_n}$. Since the neutron spin analyzing efficiency or quality can be shown to be on the order of $P_n^2 T_n$ [12,13] when the $^3$He is allowed to undergo $T_1$ decay of polarization its quality as an NSF will also continually decay with time. Thus we can see the importance of maintaining as high a degree of time-averaged $^3$He polarization as possible.

To take advantage of the full potential performance of a $^3$He NSF one must actively counteract the $^3$He $T_1$ decay through periodic refreshing of the polarized gas or in-situ polarization. Commonly gas is polarized either with SEOP or MEOP in a laboratory away from the neutron instrument and then transported to the location of use. In this case, depending on the on instrument $^3$He relaxation time, the cells are then exchanged every one to two days, with calibration measurements of the instrument necessary for each cell, or cell filling, to correctly normalize to the time dependence of the $^3$He polarization [14,15]. Additionally, refreshing of the polarized gas periodically in a pulsed mode has been proposed to maintain polarizations closer to those achieved in laboratory conditions in a pseudo-steady state. However, the only way to truly obtain the full potential for a $^3$He NSF, is to develop a system capable of polarizing the $^3$He gas in place, maintaining it at the steady state maximum polarization one obtains in the controlled laboratory conditions. In this case one further benefits by having time stability of the instrument.

We will assume that $P_n^2 T_n$ is the relevant quality factor for a fixed neutron wavelength [12,13] and that since both $P_n$ and $T_n$ are functions of $^3$He polarization, the achieved instrument performance or quality factor $Q$ will be proportional to the time integral of $P_n^2 T_n$. When normalized to flux, $Q$ can be thought of as the fraction of neutrons recorded on the the detector that can be used to obtain counting statistics. Assuming a neutron polarizer and polarization analysis (PA) system consisting of an ideal supper mirror (SM) incident beam polarizer (i.e. neutron transmission of 50% and neutron polarization of 100% for the unpolarized incident beam through the SM) and a $^3$He analyzer operating at high $^3$He polarization and optimal $P_n^2 T_n$ ($P_n^2 T_n \approx 0.2 - 0.25$ typically) one would need to count for 4-5 times as long as without PA to obtain the same counting statistics. In addition to the lowering of count rates caused by the
addition of PA, experiments using PA commonly measure the four possible combinations of the polarizer and analyzer spin states. Thus with PA one would measure a total of 16 to 20 times as long to obtain similar statistical errors for each of the four states as one would require for a single unpolarized measurement.

To show the effects of the time dependence of $Q$ we have created plots of the time required to measure $Q = 10n_0$ as a function of $^{3}\text{He}$ polarization for several different cases of time decaying and steady $^{3}\text{He}$ polarization. Here we define $n_0$ as the number of counts recorded in one hour from the scattered beam without PA installed on the instrument. Then the time to reach $Q = 10n_0$ would be the time required to record $2.5n_0$ counts for each of the four states giving a statistical accuracy of $1/\sqrt{2.5n_0}$ for each state if one sample/set of conditions were measured. We simply note that $n_0$ as defined above is a function of the incident beam intensity, the neutron detector, and the neutron scattering cross section of the sample, thus the amount of time required to obtain a relevant level of statistical accuracy is highly variable and is dependent on all of these parameters. But for example, if one has a detector count rate of 111 Hz for the scattered beam without PA, it would take 2.5 hours to measure $1 \times 10^6$ counts, i.e. to obtain a statistical accuracy of 0.1%. Thus given our definition of $n_0$, $Q = 10n_0$ would correspond in time to the time required to obtain $1 \times 10^6$ counts in each of the four states if the detector count rate for the unpolarized measurement on the same sample with the same instrument were 111 Hz. Or for another example, to perform 10 measurements on a series of samples/sample conditions, when the count rate is 1.11 kHz without PA installed, and maintain statistical errors of 0.1% for all four states with PA.
Figure 1 thus shows how the quality factor $Q$ evolves as a function of the maximum achievable $^3$He polarization. One can see that to obtain $Q=10n_0$, one must count for about 38 hours with a constant $P_{^3\text{He}}=80\%$ and about 50 hours with time decaying $P_{^3\text{He}}$ starting at 80\% with a 100 hour $T_1$ that has the polarization refreshed daily. 50 hours is the same time as required to obtain this level of statistical accuracy using a cell polarized constantly to $P_{^3\text{He}} = 70\%$. A cell with a 100 hour $T_1$ initially polarized to $P_{^3\text{He}} = 80\%$ that does not have the polarization refreshed would require nearly double the counting time, about 70 hours, as the cell continuously polarized to 80\% to reach $Q=10n_0$.

Also shown in figure 1 is a cell with a 400 hour $T_1$, a value similar to what we have obtained in tests on a SANS instrument discussed in Ref. [16]. Here the difference in time integrated quality becomes less over time scales short compared to $T_1$. As a rule of thumb, one can use a $^3$He NSF cell for approximately one day per 100 hours of on-beam $T_1$. In this case, over long time scales, one would obtain a relative time averaged quality, $Q/n_0$, about 3/4 of that obtained with constant polarization. This ratio can be seen by comparing the green and red line in Fig. 1 and is relatively independent of starting polarization or $T_1$ in limit where the measurement time is longer than the time one can use each cell filling, i.e. one day per 100 hours of $T_1$. Clearly one could chose to refresh the polarization more often to approach the levels obtained with constant polarization if the resources are available, as can be seen comparing the blue line to the red line in Fig. 1, however time dependent corrections and calibrations of each cell’s polarization would still be required. From this one can see the advantage of higher time averaged polarization, and the optimal advantage of in-situ optical pumping. Because of this gain in performance, plus the additional advantages of time-stability and lower daily maintenance, first work at the JCNS will focus on the development of in-situ $^3$He polarizers using the SEOP method.

3. SEOP $^3$He Polarizer developed for a reflectometry polarization analyzer

The polarizer we developed for the new JCNS magnetism reflectometer, called MARIA, allows us to take advantage of the positive traits of an in-situ $^3$He polarizer [17]. The MARIA reflectometer has a large area, 40 cm by 40 cm position sensitive detector that makes a $^3$He spin filter cell an ideal choice for analysis, in accord with instrumentation developed at other centers for reflectometer applications [1, 14, 18, 19].

The polarizer device has been described previously in [11] and is shown in Fig. 2. The system is based on a “magic box” style magnetostatic cavity which provides a wide aperture opening for the scattered neutron beam and optical access perpendicular to the axis of neutron propagation through the system. Further, the $\mu$-metal of this magnetic cavity provides good shielding from external magnetic fields and thus gives stable $^3$He $T_1$ relaxation times independent of external factors or instrument settings.

Since we polarize the $^3$He gas in-situ, we are able to achieve high stability and time averaged performance of the $^3$He NSF. This device recently maintained a high level of $^3$He polarization of $80.4\%\pm1.5\%$ in test measurements using the cell called J1 which has a diameter D=6 cm and is optimized for a cold neutron spectrum [11]. Such levels of polarization maintained over time provide high $^3$He NSF performance for this application, and the stability over time simplifies data analysis and instrument calibration.

When used with a larger diameter cell, D=9 cm, this device was capable of maintaining the same saturation polarization as when this cell is polarized in laboratory conditions. For over 4 days during a polarized GISANS experiment we maintained a $^3$He polarization of 73\% while using the sample magnet and performing routine AFP spin reversal of the $^3$He polarization to be discussed below. In this experiment we used the cell called Orvieto on loan from NIST [20], that provided approximately 5 degrees of angular coverage in the configuration of this particular experiment. During the week of operation the system allowed full flexibility of the MARIA sample environment and sample magnet which was operated at up to 1.2 T without causing any
Figure 2. The $^3$He polarizer developed for the MARI A magnetic reflectometer. This polarizer is based on a 76 cm long magic box style magnetostatic cavity with a 40 cm high by 30 cm wide cross section. This dimension allows full acceptance of the scattering angle allowed by the MARI A detector and laser access perpendicular to the scattered neutron beam for the optical pumping. The lasers are two frequency narrowed diode array bars that are collimated and expanded as shown in the diagram. They are then directed into the cell with mirrors.

noticeable effect on the $^3$He polarization, or on the measured cell $T_1$ in the absence of optical pumping. The sample magnet on MARI A is a yoked electro-magnet, the return field of which couples weakly to the $B_0$ holding field of our magic box, changing the Larmor frequency of the $^3$He atoms by about 50 Hz when its field is varied from 0 T to 1.2 T. During this experiment the $^3$He cell was about 100 cm away from the center of the magnet bore, i.e. the front of the magic box was about 60 cm away from the center of the magnet and the $^3$He cell was centered longitudinally in the 76 cm long magic box. A photo of the in-situ polarizer is shown in Fig. 3.

Another feature of this in-situ polarizer is to use the $^3$He NSF as an ideal wide area neutron spin filter by performing adiabatic fast passage (AFP) spin reversal of the $^3$He gas polarization. The AFP system is similar to that described in Ref. [21]. However for this application the AFP must also be synchronized to circular polarization reversal of the optical pumping lasers which we accomplish by using liquid crystal variable wave retarders whose control is written into the program that performs the AFP RF pulse generation discussed below. This allows us to perform continual optical pumping of the $^3$He gas while the $^3$He NSF spin state, or analyzing direction, is controlled by the MARI A instrument with a simple TTL interface.

Our AFP system is based on a digital 1.2 MHz computer data acquisition card and a high power (200 W), high voltage ($\pm 75$ V$_{pp}$), power amplifier driving an off-resonant solenoid coil
Figure 3. A picture of the $^3$He polarizer developed for the MARIA magnetic reflectometer. When in operation the entire frame is covered with aluminum plates for laser radiation security. These outer aluminum plates have been removed to show the mounting of the laser optics with respect to the magic box.

wound directly onto the SEOP oven to provide the transverse RF field. A digital waveform is generated by a computer program that allows us to easily change the parameters. The program produces a frequency chirped wave from 0.25 to 1.5 times the Larmor frequency of the $^3$He atoms at the chosen $B_0$ holding field which is normally about 10 gauss (corresponding to a Larmor frequency of 32 kHz). This chirped wave has a duration of 0.5 s and is amplitude modulated with a gaussian profile of width equal to 1/8 of the pulse duration to insure a fully adiabatic reversal of the $^3$He. The house built power amplifier that allows us to drive a non-resonance coil is important because we can freely chose any $B_0$ field without modifying the coil tuning or circuit impedance matching as would be required with a resonant coil. The 0.5 s long sweep helps make the AFP robust against small changes in the $B_0$ holding field caused by slight field drifts due to power supply stability or coupling of the SEOP system’s magnetic cavity to the sample magnet.

4. Design of an ultra compact polarization analyzer for SANS and plans for wide angle analysis on TOPAS and DNS

While the MARIA system focuses on coverage of a wide angle scattered beam in conjunction with a high sample field, there are other applications that will not require the use of high sample fields and thus can be made more compact, placing the cell very near to the sample in order to obtain high angular coverage with a relatively small cell and less laser power. Separation of incoherent background for soft-matter studies with small angle neutron scattering is one such application [22]. Initial testing of an off-line polarized cell showed the feasibility of this technique for the JCNS. Consequently we have designed and begun prototyping an ultra compact SEOP polarizer to be used for SANS. The system will use an 18 cm long $\mu$-metal end-capped and shielded solenoid. The system will have a small entrance aperture and a large exit aperture in the end caps for the scattered neutron beam. The effects of these holes in the $\mu$-metal end caps on the field homogeneity are then corrected with additional coils. This design will allow the
sample to be placed within 3-4 cm of the \(^{3}\)He cell while using standard sample environment. Ultra-wide angle applications, i.e. involving angular coverage >90 degrees in the horizontal and or vertical planes, such as DNS [23] or the future TOPAS spectrometer [24], are foreseen to use a filling station approach. The possible system for DNS could be similar to the PASTIS approach [25] and use \(^{3}\)He gas polarized off line which is transported to a single wide-angle cell in the DNS detector housing using the local filling technique [14]. The system would have the \(^{3}\)He NSF close to the sample and given the cold energy neutrons, i.e \(\lambda \approx 4.5\ \text{Å}\), it would not require more than 1-2 bar liters of polarized \(^{3}\)He per cell filling. This system could serve as a prototyping stage for the much more demanding application on the TOPAS spectrometer currently under construction. TOPAS will use thermal neutrons with wavelength \(\lambda \approx 1\ \text{Å}\) requiring on the order of 30 bar-cm of gas thickness in the NSF and an array of cells with a surface area of 0.7 m\(^2\) to obtain full detector coverage. This extremely large volume of polarized \(^{3}\)He will necessitate the development of new techniques and larger amounts of polarized gas per day than what is currently in use at any neutron research center. The polarizer will most likely be a large scale SEOP polarization station designed to have a throughput of 100 bar liters per day. This large scale station would then be connected to the array of cells via polarization preserving transport tubes and an automated vacuum pump/gas recycling system as in the local filling approach.

5. Summary
The \(^{3}\)He spin filter program at the JCNS is making steady progress towards application of \(^{3}\)He neutron spin filters on a variety of neutron instruments. Initial devices have been constructed and tested with positive results. Further developments will focus on refining the prototypes and making new novel designs customized to the particular application.

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