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Polarized ^{3}He spin filters in neutron scattering

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Abstract

We report progress in the development and application of neutron spin filters based on transmission through polarized ^{3}He gas. Tests and application of these devices are underway at the National Institute of Standards and Technology (NIST) Center for Neutron Research (NCNR) and the Intense Pulsed Neutron Source (IPNS). The results of application to polarization analysis for diffuse reflectometry is discussed, along with an example of the issues in choosing the flipping ratio for such applications. The status of our development of polarized gas production using both spin-exchange (SEOP) and metastability-exchange (MEOP) optical pumping will be presented. For SEOP, we currently obtain 70–75% ^{3}He polarization in cells up to 500 cm^3 in volume.
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1. Introduction

Polarized ^{3}He spin filters are under development for neutron scattering at several neutron facilities in Europe [1]. In this paper, we review the status of a collaborative program in the US that includes contributions from the National Institute of Standards and Technology (NIST), Indiana University, Hamilton College, the University of Wisconsin, Argonne National Laboratory, and the Spallation Neutron Source. We are currently

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focusing on two key areas for which ^{3}He spin filters have notable advantages: analyzers for divergent beams and broadband polarizers for thermal energy beams. In Section 2 we discuss polarization analysis for diffuse reflectometry at the NIST Center for Neutron Research (NCNR) and the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory, along with an example of the issues in choosing the flipping ratio for such applications. A compact, continuously operating polarizer developed by Hamilton College and recently tested on the Single Crystal Diffractometer (SCD) at IPNS is described in another paper in these proceedings [2]. Section 3 reviews our developments in polarized gas production and we conclude in Section 4.

Polarized ^{3}He spin filters are based on the spin dependence of the ^{3}He–neutron capture cross section. For the ^{3}He spin parallel to the neutron spin, the thermal capture cross section is essentially zero, whereas for the spins anti-parallel, the thermal capture cross section is 10,666 b [3]. The cross section is directly proportional to the neutron wavelength. A cross section independent of wavelength would be ideal, but nevertheless a ^{3}He spin filter can serve well as a broadband neutron polarizer. This feature is particularly relevant to spallation neutron sources. The basic equations for ^{3}He polarizers are described elsewhere [4,5]. The similarities between the formulae for transmission and polarization allow the polarization of an initially unpolarized, monochromatic, neutron beam to be determined by measurements of transmission alone. At a spallation source, time-of-flight analysis allows for analysis at each wavelength as well as analysis using the known linear dependence of the cross section on wavelength.

2. Analyzers for reflectometry

2.1. Progress at the NCNR

Recently we reported tests of ^{3}He spin filters on the NG1 polarized neutron reflectometer [4]. In that work, we carefully compared the results obtained using a supermirror analyzer to those obtained using a ^{3}He analyzer, for the simple case of a specular reflection. Using a linear position sensitive detector (PSD) to simultaneously obtain data for a range of scattering angles, we have conducted a similar study for a diffuse reflection on the NG1 reflectometer. By obtaining data from the PSD at different sample angles, a polarized ^{3}He analyzer in conjunction with a PSD provides a two-dimensional map of the wave vector transfer in reciprocal space without the need to scan the detector. This increased efficiency will become important for the measurements of weak off-specular scattering. We obtained such maps for a Co antidot sample, and also compared results obtained with the ^{3}He/PSD apparatus with selected one-dimensional data obtained with a supermirror analyzer. A detailed description has been published [6].

For this off-specular experiment, we employed the same 7 cm diameter, 280 cm^3 volume cell (identified as "Bullwinkle") and magnetically shielded solenoid that had been used in our earlier specular experiment. The ^{3}He gas was polarized off-line by SEOP using 14 W of spectrally narrowed laser light [7] incident on one side of the cell and 25 W of broadband laser light incident upon the other side. During the course of the 65 h experiment, the ^{3}He polarization decayed from 70% to 50%, corresponding to a decrease in ^{3}He analyzing efficiency from 0.97 (flipping ratio of 70) to 0.90 (flipping ratio of 20), and a decrease in transmission of the desired spin state from 36% to 20%. (For reference, the transmission of an unpolarized beam would have decreased from 18% to 11%. All of these transmission values include a loss factor of 0.88 due to scattering by the glass cell itself [6].) An exponential fit to the ^{3}He polarization yielded a relaxation time of 195 ± 4 h. Since the relaxation time for the cell Bullwinkle had previously been measured to be 520 h, the relaxation time during the experiment was dominated by a contribution from magnetic field gradients [8]. A known gradient in this solenoid had previously limited the relaxation time for the cell Bullwinkle to 350 h; we believe that the lower value observed in this experiment may be due to gradients produced by guide fields that were improvised for transport of the neutron spin.

In the summer of 2004, we plan to employ ^{3}He spin filters on the Advanced Neutron Diffractometer/Reflectometer (AND/R) [9] at the NCNR for further applications in diffuse reflectometry. This instrument is being equipped with a 20 cm × 17 cm two-dimensional PSD. To make full use of this PSD and allow for long relaxation times in larger cells, we have constructed a 28 cm diameter, 46 cm long shielded solenoid with 8.9 cm apertures. Ten compensation turns at each end of the solenoid were necessary to optimize the homogeneity, which was found to be just under $\Delta B/B = 2\times10^{-4}\,\mathrm{cm}^{-1}$ from both magnetic field mapping and from measurements of the relaxation times of low pressure (~1 mbar) cells polarized by MEOP. For an 11 cm diameter, 6 cm long cylindrical cell with a relaxation time of 730 h, we measured a relaxation time of 420 h in this solenoid, corresponding to a gradient of $2.2\times10^{-4}\,\mathrm{cm}^{-1}$. This gradient is still larger than expected and we are investigating whether more precise compensation and/or shaking [10] can reduce it to $1\times10^{-4}\,\mathrm{cm}^{-1}$.

2.2. Progress at the IPNS

In our first test of a ^{3}He spin filter at the POSY polarized neutron reflectometer at IPNS, we reported polarization analysis of a Zeeman-split reflection from a magnetized iron film [11]. ^{3}He was polarized on an MEOP compression apparatus [12] and transported to IPNS. Since then, we have constructed an SEOP apparatus at Indiana that will soon be relocated to IPNS. For an experiment conducted in December 2004, ^{3}He gas was polarized with this SEOP system and transported to IPNS. Data were obtained on a film of Cr(2 nm)/Co(20 nm)/SiO_2/Si that consists of circular antidots (holes) on a square lattice. The analysis is underway. Here we report preliminary results for the performance of the spin filter analyzer, which includes a magnetic shield to avoid depolarization from the sample magnet.

For this IPNS experiment, the Indiana SEOP system utilized 15 W of spectrally narrowed diode laser light using a system similar to the NIST laser described in Ref. [7]. Two blown glass cylindrical cells that were constructed and filled at NIST were employed in this experiment: "Peabody" (7 cm diameter, 300 cm^3 volume) and "Yeoman Rand" (6 cm diameter, 175 cm^3 volume). Both were measured to have relaxation times of 650 h, close to the dipole–dipole limit of 700 h for their ^{3}He partial pressure of 1.16 bar. For Yeoman Rand, the overall instrumental flipping ratio varied between 5.2 for a wavelength of 0.3 nm up to nearly 20 at 0.8 nm. The flipping ratio vs. wavelength was well fit using a ^{3}He polarization of 62% and a combined efficiency of the supermirror polarizer and non-adiabatic spin flipper of 0.91. The fractional loss during the 6 h transport time from Indiana Univ. to IPNS is estimated to be $\approx 5\%$, based on the known relaxation time of the cell and an average loss of 1% that has been observed when transferring the cell between holding fields.

The ^{3}He spin filter cell was stored on the beam line in 2 mT magnetic field provided by a 31.4 cm diameter, 53 cm long solenoid that is surrounded by a double layer magnetic shield. The inner (outer) cylindrical shield is 32.4 cm (40.6 cm) diameter and 56.5 cm (60.0 cm) long; both are made from 1.2 mm thick mu-metal with a specified maximum magnetic permeability $\mu = 3000$ and a saturation field of 2.2 T [13]. For the relevant range of applied and stray fields, we expect the μ to be near the maximum value, which leads to an estimated shielding factor for transverse fields of 60 [14]. The homogeneity of the shielded solenoid was optimized by adding several compensation turns to each end of the solenoid. Using an automated field mapping system, the average field gradient was measured to be $3\times10^{-4}\,\mathrm{cm}^{-1}$ in the volume occupied by the cell. When the ^{3}He analyzer was on the beam line, the sample magnetic field was along the beam line (thus parallel to the ^{3}He analyzer holding field) and operated between 20 and 80 mT. The stray field was typically ~1 mT at the end of the shielded solenoid closest to the sample magnet. The typical relaxation time for the cells on the beam line was 220 h.

2.3. Choosing the analyzer's flipping ratio

^{3}He spin filters differ from neutron polarizing devices such as supermirrors and Heusler crystals

in that there is a necessary compromise between flipping ratio and transmission. A "thick" spin filter, i.e. one with a large product of ^{3}He gas pressure and length, will provide a high flipping ratio, but due to imperfect ^{3}He polarization the transmission will be low. (Improving the ^{3}He polarization increases both the neutron polarization and transmission.) Although this issue has been addressed in the literature [15–22], most of these analyses have considered the optimization of a ^{3}He spin filter for polarizing the beam (or for both polarizing and analyzing). In addition, some analyses have been quite complex because the optimum choice depends on the type of experiment, the backgrounds present and other variables. In this section, we present a simple approach to provide some general guidance and rationale for the design of spin filter analyzers. We shall see that modest flipping ratios are generally desirable.

As a specific example, we assume that the goal is to measure a small fraction of spin-flip scattering in the presence of a larger amount of non-spin-flip scattering. We also assume that the neutron beam is polarized with a supermirror (or any device for which the polarizing efficiency is not linked to its transmission) and analyzed with a polarized ^{3}He spin filter. This is a likely scenario for most cold neutron applications in the near future, as supermirrors are simple, reliable devices that provide high polarizing efficiency for collimated beams. For simplicity, we further assume that the spin-flip efficiency is unity. The fraction of spin-flip scattering, f_{sf}, is given by

$$f_{\mathrm{sf}} = \frac{A_0 - A_{\mathrm{s}}}{2A_0}, \tag{1}$$

where A_0 is the transmission asymmetry for the direct beam (or a sample with pure non-spin-flip scattering), and A_{s} is the measured asymmetry for the scattered beam to be analyzed, where we assume that both asymmetries are measured for neutrons passing through the polarizer, spin flipper, and analyzer [23]. The asymmetry for the scattered beam is given by

$$A_s = \frac{N_{\mathrm{off}} - N_{\mathrm{on}}}{N_{\mathrm{off}} + N_{\mathrm{on}}}, \tag{2}$$

where N_{off} and N_{on} are the number of counts for the flipper off and on, respectively. A_0 is defined similarly for the direct beam, and is equal to the product of the polarizing efficiency of the supermirror polarizer, P, and the analyzing power of the ^{3}He spin filter, A_{a}. The analyzing power is related to the flipping ratio $F_{\mathrm{a}} = N_{\mathrm{off}}/N_{\mathrm{on}}$ by $A_{\mathrm{a}} = (F_{\mathrm{a}} - 1)/(F_{\mathrm{a}} + 1)$. The analyzing power of a ^{3}He spin filter is given by $A_{\mathrm{a}} = \tanh(n\sigma l P_{\mathrm{He}})$, where n is the density of ^{3}He atoms in the cell, σ is the wavelength-dependent neutron absorption cross section, l is the length of the cell, and P_{He} is the ^{3}He polarization.

We assume that A_0 can be determined from measurements with high count rates and that this same instrumental analyzing power is relevant to the scattered beam. We further assume that it can be measured accurately, and thus, consider only statistical uncertainties in measuring A_{s}. The fractional uncertainty in f_{sf}, denoted by $\Delta f_{\mathrm{sf}}/f_{\mathrm{sf}}$, is related to the uncertainty in A_{s}, ΔA_{s}, by

$$\frac{\Delta f_{\mathrm{sf}}}{f_{\mathrm{sf}}} = \frac{\Delta A_{\mathrm{s}}}{A_0 - A_{\mathrm{s}}}. \tag{3}$$

If we assume counting statistics, i.e. that the uncertainties in N_{off} and N_{on} are equal to $N_{\mathrm{off}}^{1/2}$ and $N_{\mathrm{on}}^{1/2}$, respectively, then ΔA_{s} is given by

$$\Delta A_{\mathrm{s}} = \left(\frac{4N_{\mathrm{on}}N_{\mathrm{off}}}{(N_{\mathrm{on}} + N_{\mathrm{off}})^3}\right)^{1/2} = \left(\frac{1 - A_{\mathrm{s}}^2}{N}\right)^{1/2}, \tag{4}$$

where $N = N_{\mathrm{off}} + N_{\mathrm{on}}$. N is proportional to the transmission through the ^{3}He spin filter averaged over both spin states, which is given by $T_{\mathrm{n}} = T_0 \cosh(n\sigma l P_{\mathrm{He}})$, where $T_0 = T_{\mathrm{E}} \exp(-n\sigma l)$ is the transmission through the unpolarized cell and T_{E} is the transmission through the empty cell. Making use of Eq. (1), we finally obtain

$$\frac{\Delta f_{\mathrm{sf}}}{f_{\mathrm{sf}}} = \frac{1}{2A_0 f_{\mathrm{sf}}}\left(\frac{1 - A_0^2(1 - 2f_{\mathrm{sf}})^2}{N}\right)^{1/2}. \tag{5}$$

Fig. 1 shows the variation of $\Delta f_{\mathrm{sf}}/f_{\mathrm{sf}}$ with the spin filter opacity $n\sigma l$ for a supermirror polarizing efficiency $P = 0.987$, a ^{3}He polarization $P_{\mathrm{He}} = 0.6$, and an assumption of 10^4 neutrons incident on the ^{3}He spin filter. The locations where the flipping ratio of the analyzer, F_{a}, is equal to 2, 10, 100, and 1000 are also shown. For $f_{\mathrm{sf}} \approx 0.5$, we obtain the

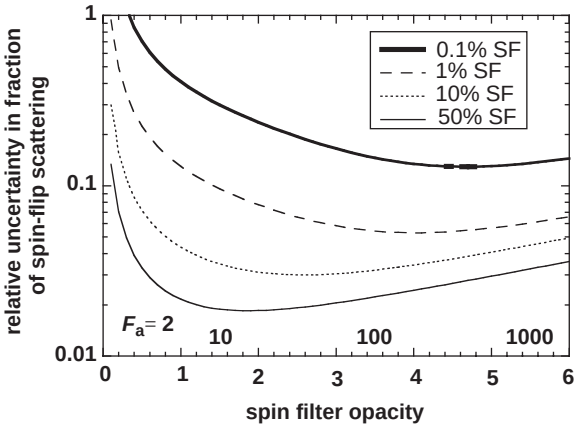

Fig. 1. The variation of the relative uncertainty in measuring the fraction of spin-flip scattering, $\Delta f_{sf}/f_{sf}$, with the opacity $n\sigma l$ of the ^{3}He analyzer. The locations where the flipping ratio of the analyzer, F_a, is equal to 2, 10, 100, and 1000 are also shown. The assumptions for the calculation are described in the text.

small asymmetry limit ($A_s \approx 0$), which leads to the often quoted figure of merit $\Delta f_{sf}/f_{sf} \approx (A_0^2 N)^{-1/2}$. In this small asymmetry case, essentially any flipping ratio above about 3 is acceptable. For $f_{sf} = 0.1$, the optimum flipping ratio is 20, but any value between 5 and 100 yields only a small increase in uncertainty. For $f_{sf} = 0.01$, the optimum flipping ratio increases to 150, but any value between 20 and 1000 yields only a small increase in uncertainty. Only for $f_{sf} = 0.001$ do very high flipping ratios become desirable, and even in this case, the uncertainty only increases by 30% if a flipping ratio of 30 is employed. Hence a flipping ratio of 30, for which the transmission of the desired spin state will be 0.28 for $P_{He} = 0.6$ (including the typical loss factor of 0.88 due to glass transmission), should satisfy the needs of most experiments. In many cases it may be preferable to employ lower flipping ratios. Although this treatment is idealized, the effects of background, imperfect spin transport, and less efficient polarizing or spin-flipping elements would be to make even lower flipping ratios preferable.

The results in Fig. 1 are for a monochromatic beam. It is more difficult to choose a flipping ratio that is optimal for a wide range of wavelengths such as would be encountered at a spallation source. Since the opacity is directly proportional to wavelength, one can readily determine from Fig. 1 the wavelength range over which $\Delta f_{sf}/f_{sf}$ is within some acceptable range. For example, for $f_{sf} = 0.1$, the wavelength can vary by about a factor of five with no more than a 35% increase in $\Delta f_{sf}/f_{sf}$ at the ends of the range ($n\sigma l$ between 1 and 5). If the spin filter is not continuously optically pumped, a similar issue exists because of the decay of the ^{3}He polarization. One will want to choose an initial flipping ratio such that the final flipping ratio is still acceptable.

3. Polarized gas production

We are currently developing two optical pumping methods for polarizing ^{3}He gas: spin-exchange (SEOP) [24,25] and metastability-exchange (MEOP) [26].

In the SEOP method, electronic polarization in alkali atoms (usually Rb) is produced by optical pumping, and the polarization is transferred to the ^{3}He nuclei through the hyperfine interaction during collisions. The Rb vapor density, and thus the rate at which ^{3}He gas is polarized, is controlled by varying the temperature of the cell. Recently it has been found that the ^{3}He polarization in an SEOP cell is limited to $1/(1+X)$, where X accounts for the observation of a linear dependence of the ^{3}He relaxation rate on the Rb vapor density with a slope that exceeds the spin-exchange rate constant [27].

We recently reported a typical value for X of 0.33 [7], which limits the maximum ^{3}He polarization to 75%. Recently we have found that although $X = 0.33$ is a typical value for cells with surface area to volume ratios (S/V) near 1 cm^{-1}, X can vary between 0.2 and 1.2 for cells with larger S/V. Although larger values of X have only been observed for large S/V, there is no clear correlation between X and either S/V or room temperature relaxation time. Further studies to understand the origin of this temperature-dependent relaxation are in progress. Fortuitously, the relatively large cells that will be needed for many neutron spin filter applications have moderate S/V ratios.

Recently, we implemented a 40 W diode array bar that allows us to deliver 25 W of spectrally

narrowed laser light [28] to a cell. In conjunction with 25 W of broadband laser light entering the opposite side of the cell, we now routinely obtain 75% ^{3}He polarization for long lifetime cells up to 500 cm^3 in volume. In general, we determine the ^{3}He polarization from neutron transmission measurements and estimate the relative standard uncertainty in the ^{3}He polarization obtained in blown cells to be ±5% [7]. We have not necessarily optimized the use of the light for each cell. The use of two spectrally narrowed diode array bars on each side of a cell should allow for 75% ^{3}He polarization in cells approaching one liter. However, the hybrid spin-exchange method discussed below may be a better approach for very large cells. If the cell is continuously optically pumped, the hybrid approach may also be useful for cells with relatively short lifetimes or reduced lifetime due to magnetic field gradients [8].

While Rb has been the traditional choice for SEOP, K and Na have similar spin-exchange rates [29], but much lower alkali spin-destruction rates [24,30–32]. However, convenient high-power lasers are not available for direct optical pumping of either K or Na. Recently, a hybrid approach has been demonstrated [33], in which a small amount of Rb and much larger amount of K was distilled into a cell. The Rb is optically pumped and its electronic spin is rapidly transferred to the K. This method has been shown to yield a higher optical pumping rate than for pure Rb, which can either be used to lessen laser power demand or offset a non-optimal ^{3}He polarization relaxation time. Based on these promising results, we have begun preparing large Rb–K hybrid neutron spin filter cells to evaluate the potential improvement. At NIST, we have prepared Rb–Na cells that will be studied at the University. of Wisconsin, where the diagnostics for measuring Na density are currently under development.

A detailed paper on the piston compressor for MEOP at Indiana University has recently been submitted for publication [34]. During a 2 h fill of a 480 cm^3 storage cell (Orion) to a pressure of 1.22 bar with a 33% ^{3}He, 67% ^{4}He mixture, we determined that the apparatus preserved 81% of the polarization produced in the optical pumping cell. The storage cell ^{3}He polarization was determined to be 0.46 based on a neutron-based measurement of 0.42, corrected for the transport from Indiana University to IPNS. We determined the optical pumping cell polarization to be 56% by analysis of the circular polarization of the 668 nm light emitted by the discharge [35]. Although the compressor itself has been found to have essentially no loss of polarization, there are comparable losses due to relaxation in the storage cell region and the buffer cell region between the two compression stages. Whereas the relaxation time of the Rb-coated storage cell Orion is over 200 h when its valves are closed [36], the relaxation in this cell has been found to be pressure-dependent when left open to the compression system. This suggests a contribution due to diffusion into Pyrex tubing and other more relaxing regions. Although the relaxation time of the buffer cell itself has not been measured, data suggests that a similar issue exists in the buffer cell region. Improvements in the buffer cell and storage cell regions may be sufficient to yield an operationally lossless device. Improving the optical pumping cell polarization will require increasing the laser power and improving the gas purity. At present, two 1300 cm^3 optical pumping cells are polarized using 4 W of laser light from an arc lamp pumped Nd:LMA laser.

4. Conclusion

We are pursuing several directions to further the impact of ^{3}He spin filters in neutron scattering. For large solid angle polarization analysis, we are focusing on diffuse reflectometry and have demonstrated the use of ^{3}He spin filters at the NCNR and IPNS. We have used a simple example to provide guidance in choosing a flipping ratio for the case of a supermirror polarizer used with a ^{3}He analyzer. We find that a flipping ratio of 30 should cover the needs of most experiments and that in many cases, lower flipping ratios may be desirable. Large, long lifetime, boron-free spin filter cells along with spectrally narrowed diode lasers for SEOP have made continuous operation of neutron spin filters with 75% ^{3}He polarization a realistic possibility. Further improvement in the achievable ^{3}He

polarization will require reducing or eliminating temperature-dependent ^{3}He relaxation. Maximizing the ^{3}He polarization in large cells and/or cells with reduced lifetime can be addressed with hybrid SEOP.

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