Applications of ^{3}He neutron spin filters at the NCNR

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Abstract

At the NIST Center for Neutron Research (NCNR), we have applied ^{3}He neutron spin filters (NSFs) to the instruments where other standard polarizing techniques are inadequate, such as thermal triple-axis spectrometry, small-angle neutron scattering, and diffuse reflectometry. We present the status of our development and application of this method, including polarized gas production by spin-exchange optical pumping, magnetostatic cavities for storage of the polarized gas on the beam line, and NMR-based, on-line monitoring and reversal of the ^{3}He polarization. We present the status of developing user-friendly interfaces incorporated into the instrument software to handle these ^{3}He neutron spin filters while taking data and performing data analysis. Finally we discuss the status of development of a polarization capability on the Multi-Axis Crystal Spectrometer, which requires polarization analysis over a 220 degree angular range.

Key words: ^{3}He NSF; TAS; SANS; Wide-angle; Polarization analysis; ^{3}He flipping
PACS: 83.85.Hf; 61.12.Ld; 75.25. +z; 32.80.Bx

1. Introduction

Polarized neutron scattering is a powerful probe for a wide range of research fields from physics to biology. Nuclear-spin-polarized ^{3}He gas, produced by optical pumping, can be used to polarize or analyze neutron beams because of the strong spin dependence of the absorption cross section for neutrons by ^{3}He. Due to significant improvement in the performance of ^{3}He-based neutron spin filters (NSFs) during the last several years, including initial ^{3}He polarizations exceeding 70 %, the use of NSFs has grown in the neutron scattering community worldwide. Compared to commonly used polarizers such as supermirrors and Heusler crystals, NSFs have these advantages: 1) they can polarize a broad wavelength band of neutrons effectively; 2) they can polarize large area and widely divergent neutron beams; 3) they are add-on devices, requiring no beam alignment and convenient for evaluation of polarized beam corrections and 4) their polarization direction can be efficiently inverted by reversing the ^{3}He nuclear polarization[1].

The NIST Center for Neutron Research (NCNR) initiated a program in 2006 to support polarized neutron beam experiments using ^{3}He NSFs. The goal of the program is to polarize and/or analyze beams for neutron scattering instruments where other neutron-polarizing techniques are inadequate. So far we have applied ^{3}He NSFs to diffuse reflectometry[2], small-angle neutron scattering (SANS)[3,4], and thermal triple-axis spectrometry (TAS)[5].

For our current ^{3}He NSF applications, the ^{3}He gas is polarized off-line by the spin-exchange optical pumping (SEOP) method, transported to neutron scattering instruments, and stored on the beam line using a uniform magnetic field provided by a magnetostatic cavity. The development of on-line SEOP is underway. The key technical challenges in applying ^{3}He spin filters to neutron scattering are (1) producing a large volume of highly polarized ^{3}He gas and (2) minimizing the ^{3}He polarization decay. We have constructed two SEOP systems capable of producing 75 % polarized ^{3}He gas in 1-2 bar cells approaching 1 liter in volume[6]. These systems are now being employed to provide polarized ^{3}He gas to instruments at the NCNR. In this paper, we present applications of ^{3}He NSFs in thermal neutron triple axis spectrometry, small-angle neutron scattering and wide-angle polarization analysis.

2. Polarization analysis on the BT7 thermal triple axis spectrometer

Most of the newest thermal neutron TASs have used or plan to use large focusing monochromators to achieve higher on-sample neutron fluxes and higher useful detectable signal at the expense of increased beam angular divergences or wavelength bandwidths, but this has made it difficult to apply reflecting Heusler crystals for polarized beam production and analysis. A ^{3}He NSF decouples neutron polarization and energy selection, making it possible to optimize a thermal TAS in the unpolarized and polarized modes simultaneously over a wide range of neutron energies.

The BT-7 thermal TAS at the NCNR [7] features the choice of either a Cu(220) or PG(002) doubly-focusing monochromator, providing a useful continuous incident neutron energy range from 5 meV to 200 meV. Each doubly focusing monochromator has an active area 20 cm high and 20 cm wide. The diameter of the ^{3}He cell required to cover the entire neutron beam is 11 cm and a gas thickness between 11 bar-cm and 18 bar-cm yields reasonable neutron polarization and transmission for thermal neutrons (14.7 meV typically used)[5]. The ^{3}He pressure is typically 1.5 bar - 1.9 bar and the cell volume is close to 1 L. We typically refresh polarized ^{3}He gas which has an initial polarization of 75 % after 1 or 2 days on the beam line.

2.1. *Polarized beam setup and calibration on BT-7 TAS*

Fig. 1 shows a typical polarized beam setup using a ^{3}He polarizer and ^{3}He analyzer for routine user experiments on BT-7. The ^{3}He polarizer is located between the monochromator drum and the sample enclosure. Due to severe space constraints, we have developed an end-compensated variation of the "magic box" type of magnetostatic cavity[8] . This magic box, only 28.4 cm long, 40 cm wide, and 15 cm high, provides a homogeneous field for preserving ^{3}He polarization[9]. It provides a vertical field that is parallel to the guide field of the spin flipper that is located immediately after the ^{3}He polarizer. The magic box solved the neutron spin transport issue that existed in our previous setup due to a $\pi/2$ spin rotation within a distance of just 5 cm required from the solenoid to the flipper[5]. The transverse field gradient in the compact box has been determined to be 3.5×10^{-4} cm^{-1}. However, we have found that the open, compact nature of this device makes it difficult to shield magnetic fields along the neutron beam direction of ≈0.2 mT, which occur due to magnetization of parts of the instrument after use of a superconducting magnet. These stray fields have reduced the relaxation time of the polarizer cells, which typically have intrinsic relaxation times of 300 h, to the range of 40 h - 160 h. We are currently developing compensated magic boxes that operate at higher magnetic field (thus dominating external gradients) and methods to shield external fields.

For the ^{3}He analyzer, spin transport is facilitated by greater available space and stronger guide fields, hence we employ an 25 cm diameter, 35 cm long, magnetically shielded solenoid (MSS) located between the sample enclosure and the detector. The field gradient in the MSS has been determined to be 1.4×10^{-4} cm^{-1} using a combination of field mapping and relaxation measurements of both SEOP cells and low-pressure MEOP (metastability exchange optical pumping) cells. The MSS can shield stray fields of a few mT. We have found that employing a 7 mT vertical field located just before the MSS has significantly improved the spin transport. We now typically have spin transport efficiency of 0.99 and can achieve an initial instrumental flipping ratio of 18, independent of the beam size. Two sets of coils inside the sample enclosure are used to provide the horizontal and vertical field on the sample and also serve as a guide field for neutron polarization. The horizontal field corresponds to the $\mathbf{P}\|\mathbf{Q}$ configuration, while the vertical field corresponds to the $\mathbf{P}\perp\mathbf{Q}$ configuration, where $\mathbf{P}$ and $\mathbf{Q}$ are neutron polarization and wave vector transfer, respectively. Unlike supermirror and Heusler crystals, no beam alignment and tuning are required to set up the ^{3}He polarizer and analyzer.

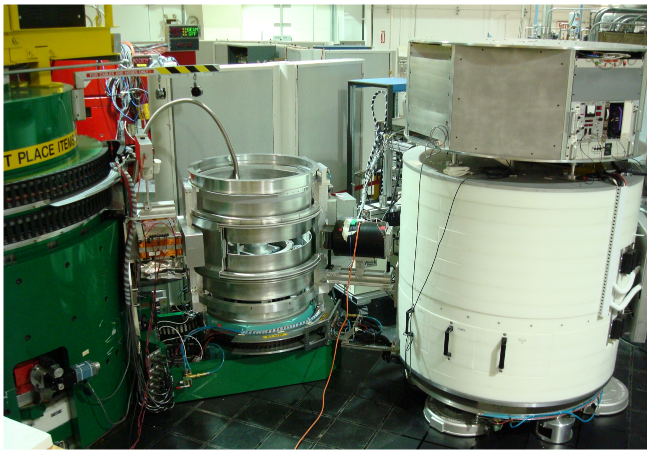

Fig. 1. (Color online) Polarized beam setup on the BT-7 Triple Axis Spectrometer. The ^{3}He polarizer is located between the monochromator drum (green) and the sample enclosure and the ^{3}He analyzer is located between the sample enclosure and the detector (white). Homogeneous magnetic fields for preserving the ^{3}He polarization are provided by an end-compensated magic box (polarizer) and a magnetically shielded solenoid (analyzer).

For quantitative data analysis it is essential to determine the polarizing efficiencies of the ^{3}He polarizer and analyzer, as well as the spin flipper and spin transport efficiencies. Evaluating these corrections typically requires measurement of four spin dependent cross sections with known samples[10]. Use of ^{3}He NSFs makes this calibration easier and more convenient since the polarizing efficiencies of the ^{3}He polarizer and analyzer can be independently determined by relative transmission measurements of unpolarized neutrons. However, the polarizing efficiency for the ^{3}He polarizer and analyzer is time-dependent due to decay of the ^{3}He polarization. Free induction decay (FID) nuclear magnetic resonance (NMR) is used to monitor the ^{3}He polarization decay on the neutron beam line. Periodic measurements of ^{3}He polarization from neutron transmission and flipping ratios are also used to confirm the ^{3}He polarization decay. The three methods are in general consistent.

Due to space constraints, we have avoided the use of a spin flipper after the sample by inverting the ^{3}He polarization using adiabatic fast passage (AFP) NMR[1]. There is no tuning required for the NMR-based ^{3}He flipper. However, there is a small loss of ^{3}He polarization for each flip, which we have determined to be 0.07% for the analyzer. This small loss can be easily added into the software for proper correction. For the incident beam, we employ a precession coil neutron spin flipper.

2.2. *Polarized beam experiments on BT-7 TAS*

We have successfully performed more than 15 polarized beam experiments on BT-7 using ^{3}He NSFs as polarizer and analyzer to polarize a 10 cm tall neutron beam. These included studies of magnetic structures in multiferroics and superconductor materials. Here we present an example. These experiments typically involve four cross section measurements at an applied sample field either perpendicular or parallel to the wave vector transfer.

The recent discovery of the rare-earth, iron-based, superconductor systems has raised much interest for studying the nature of the structure and magnetic order in the system. Magnetic peaks have been found from unpolarized data in the parent compound NdFeAsO at 30 K, well above the ordering temperature of the Nd to avoid any significant contribution from those moments[11]. Polarization analysis with neutron polarization both parallel and perpendicular to the wave vector transfer can clearly confirm if these peaks are magnetic in origin[10]. Fig. 2 shows the data for the (1,0,3) magnetic Bragg peak and the (0,0,2) nuclear Bragg peak[11]. The instrumental flipping ratios were monitored from the nuclear peak (Fig. 2a) and decreased from 18 to 8 as the ^{3}He polarization decays with time. The magnetic cross sections depend on the relative orientation of $\mathbf{P}$ and $\mathbf{Q}$. For $\mathbf{P}\|\mathbf{Q}$, all magnetic scattering goes into the spin-flip scattering channel. For $\mathbf{P}\perp\mathbf{Q}$, only half the magnetic Bragg scattering goes into the spin-flip channel, the other half goes into the non-spin-flip channel. The observed spin-flip scattering for $\mathbf{P}\perp\mathbf{Q}$ is reduced as expected compared to the $\mathbf{P}\|\mathbf{Q}$ configuration in Fig. 2b[11]. Fig. 2c shows the subtraction of the $\mathbf{P}\perp\mathbf{Q}$ intensity from the $\mathbf{P}\|\mathbf{Q}$ intensity, yielding a purely magnetic peak plus the Nd paramagnetic diffuse background.

3. Full polarization analysis for small-angle neutron scattering

Polarization analysis has not generally been implemented at SANS instruments due to lack of an adequate neutron spin analyzer to cover large solid angle scattered neutron beams with minimal small-angle scattering. The recent advances of polarized ^{3}He NSFs have made polarization analysis practical in SANS[3,12]. These initial experiments involved measurements of only two scattering cross sections, one with non-spin flip and the other with spin-flip, due to the absence of a spin flipper after the sample. In addition, the sample field was also used to maintain the ^{3}He polarization, which precluded high sample fields.

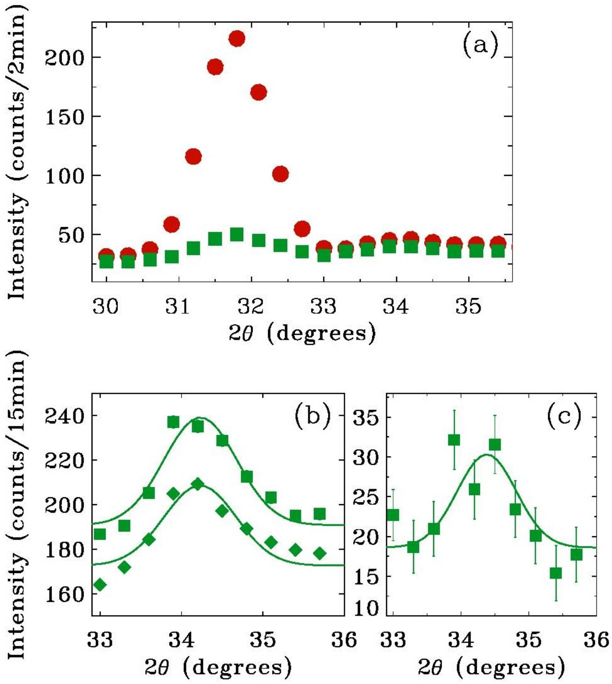

Fig. 2. Polarized neutron diffraction measurements from NdFeAsO on BT-7[11]. (a) shows the non-spin flip (solid circles) and spin flip scattering (solid squares) for the (0,0,2) structural peak at ≈32° and the magnetic peak at ≈34°. (b) shows spin flip scattering for the $\mathbf{P}\|\mathbf{Q}$ configuration (solid squares) and for the $\mathbf{P}\perp\mathbf{Q}$ configuration (solid diamonds). (c) shows the subtraction of the scattering in the $\mathbf{P}\perp\mathbf{Q}$ configuration from the scattering in the $\mathbf{P}\|\mathbf{Q}$ configuration.

For most of magnetic nanoparticle assemblies studied on SANS, a saturation field of a few Tesla is necessary. Here we discuss the operation of a compact ^{3}He spin analyzer that is equipped with an on-line ^{3}He spin flip capability and decoupled from the sample field. This apparatus has been used for full polarization analysis on the NG-3 SANS instrument[7]. A V-shaped Fe/Si supermirror in transmission geometry is used to polarize the incident neutrons, immediately followed by a precession coil spin flipper. However, we found that the aluminum wires in such flippers produces small angle scattering, hence such a flipper would be unsuitable after the sample. In addition, space constraints combined with a high strength sample field made a second flipper undesirable. Hence we used AFP NMR to invert the polarization in the ^{3}He spin filter analyzer. The compact, magnetically shielded solenoid for the ^{3}He analyzer is 20 cm diameter and 25 cm long, which just fits into the available space. The ^{3}He cell is located 50 cm away from the sample to reduce the additional ^{3}He polarization relaxation due to the external stray field from the 1.5 T electromagnet that we employed to saturate the magnetization of the magnetic nanoparticle assembly under study. The stray field from the electromagnet operating at 1.25 T was 2.5 mT upstream in front of the solenoid, but only reduced the relaxation time of the ^{3}He polarization from 140 h to 125 h. The relatively short relaxation time is mainly limited by the achievable field gradient for such as small solenoid. Nevertheless, compared to typical SANS experiment times of a few hours, such a relaxation rate is acceptable. The ^{3}He cells have a diameter approaching 12 cm, large enough to extend the area detector data to a reasonably high Q region. The polarizing efficiencies for the supermirror polarizer and the spin flipper have been extracted by measuring the four spin-dependent cross-sections (++, +-, -+, and −) without the sample[10]. The polarizer efficiency is determined to be 0.85 at 0.5 nm and 0.94 at 0.75 nm, and the flipper efficiency is typically above 0.96 (flipping probability of 0.98). We have found that accurate knowledge of these efficiencies is essential for analysis of the SANS data. The overall polarization efficiencies for the polarizer-spin flipper-analyzer system range from 0.79 to 0.89. With this apparatus we have investigated magnetic correlations in a 7 nm diameter magnetite nanoparticle assembly and are able to unequivocally separate 3-D magnetic from nuclear scattering[4]. This capability opens up new possibilities for characterizing magnetic nanoparticle systems showing promising for biomedical and data storage applications.

4. Progress toward wide-angle polarization analysis

We are currently developing apparatus for wide angle polarization analysis, with our first goal being application to the Multi Axis Crystal Spectrometer (MACS) under construction at the NCNR[13]. This instrument was designed for unpolarized beam experiments, as when the design was initiated it was not expected that polarized beam operation would be practical due to space constraints and the large double-focusing array monochromator. The polarizer, analyzer and any spin flippers must fit into a 40 cm diameter region surrounding the sample and be compatible with a typical cryostat tail of 13 cm diameter. We plan to locate a ^{3}He polarizer and analyzer just outside the cryostat tail diameter. A 40 cm diameter, 70 cm long, unshielded solenoid wound with aluminum wire will provide the uniform field for the ^{3}He cells and serve as the guide field for the polarized neutrons. This solenoid has been tested and found to have a field gradient better than 1.0×10^{-4} cm^{-1} over a cylindrical volume of 38 cm diameter and 12 cm long.

We have constructed a compact, radio frequency shielded solenoid that will allows us to efficiently invert the ^{3}He polarization in the polarizer without affecting the analyzer. A key challenge for this system is the cells for wide-angle polarization analysis. To cover the full angular range for the MACS instrument for all positions of the detector, we plan to use two fused quartz cells, on opposite sides of the neutron beam, that will cover a total of 240 degrees. These cells, as well as the simple, cylindrical, polarizer cell, will be polarized off-line directly by SEOP and installed on the instrument. Three analyzer cells have been constructed and two have been tested. Although we have obtained relaxation times of order 100 h for storage on the instrument, we are currently struggling with persistent orientation dependence[14] that reduces the relaxation time to about 25 h during SEOP. Improving the relaxation time and determining the achievable ^{3}He polarization for the analyzer are underway. A complete off-line test of the full system will occur soon, followed by integration into MACS.

5. Conclusions

We have developed polarized ^{3}He NSFs for routine polarized beam experiments on the thermal neutron triple-axis spectrometer, small-angle neutron scattering instrument, and diffuse reflectometer. We have provided ^{3}He NSFs to 30 user experiments at the NCNR since 2007. We have shown that ^{3}He NSF devices can be conveniently added into each instrument without affecting the geometry of neutron beams. In addition, use of ^{3}He NSFs does not require beam alignment and tuning procedure. We have shown that a thermal neutron triple axis spectrometer can be optimized in both the unpolarized mode and polarized mode using a ^{3}He polarizer and analyzer. We have developed a compact ^{3}He analyzer equipped with ^{3}He polarization flipping for full SANS polarization analysis at applied sample fields up to 1.5 T. We have reached ^{3}He polarization values as high as 78 % for the 11 cm diameter cells that are required to polarize such large neutron beams. We have made substantial progress in developing wide angle polarization analysis.

The authors thank J. Anderson and J. Fuller of the NIST Optical Shop for assistance with cell fabrication. The work at NIST was partially supported by Department of Energy.

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