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Polarized ^{3}He spin filters for neutron science

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The large spin dependence of the absorption cross section for neutrons by ^{3}He gas provides a method to polarize neutron beams. For certain applications, such polarized ^{3}He-based neutron "spin filters" have advantages over conventional neutron optical polarizing methods. Spin filters operate at all neutron wavelengths, can cover a large angular range and/or a large energy range, and decouple neutron polarization from energy selection. Both spin-exchange optical pumping (SEOP) and metastability-exchange optical pumping (MEOP) are currently being employed to polarize ^{3}He spin filters at various neutron facilities worldwide. We focus on the development and application of SEOP-based neutron spin filters at the National Institute of Standards and Technology (NIST), Center for Neutron Research (NCNR). The combination of long relaxation time spin filter cells, high power spectrally narrowed diode lasers, and the use of Rb/K mixtures have allowed us to reach ^{3}He polarizations up to 85 % in spin filter cells $\approx$1 liter in volume. Studies have revealed limits to the achievable polarization from temperature-dependent relaxation and unexplained magnetic field dependence for relaxation in SEOP cells. Applications include neutron scattering methods such as triple-axis spectrometry and small angle neutron scattering, and fundamental neutron physics. In most neutron scattering applications, cells are transported to the beam line and stored in a magnetically shielded solenoid or box. A recent focus has been apparatus for wide-angle neutron polarization analysis. A measurement of the spin-dependence of the neutron-^{3}He scattering length was performed with a small, polarized ^{3}He cell located inside a neutron interferometer. A precision measurement of the neutron polarization for this experiment was also performed with a ^{3}He spin filter. Use of spin filters in high flux neutron beams have revealed beam-induced alkali-metal relaxation and long term effects on SEOP spin filter cells.

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1. Introduction

The large spin dependence of the absorption cross section for neutrons by ^{3}He gas provides a method to polarize neutron beams [1]. For certain applications, such polarized ^{3}He-based neutron "spin filters" have advantages over conventional neutron optical polarizing methods [2, 3, 4, 5]. Spin filters operate at all neutron wavelengths, can cover a large angular range and/or a large energy range, and decouple neutron polarization from energy selection. Both spin-exchange optical pumping (SEOP) [6, 7] and metastability-exchange optical pumping (MEOP) [8, 9] are currently being employed to polarize ^{3}He spin filters at various neutron facilities worldwide [10, 11, 12, 13, 14, 15, 16, 17]. I will focus on the development and application of SEOP-based neutron spin filters at the National Institute of Standards and Technology, Center for Neutron Research (NCNR). In the SEOP method, electronic polarization is produced by optical pumping of alkali-metal atoms and transferred to ^{3}He nuclei in spin-exchange collisions. Optical pumping of Rb or Rb-K mixtures [18] is performed with high power diode lasers at a wavelength of 795 nm.

A ^{3}He spin filter with sufficient gas thickness and 100 % ^{3}He polarization would provide 100 % neutron polarization with 50 % absolute transmission of the beam. For the typical achievable value of 75% ^{3}He polarization for current spin filters an optimal thickness can be chosen based on a trade-off of neutron polarization and neutron transmission. Since the cross section has a simple linear dependence on the neutron wavelength, polarized ^{3}He can polarize a broad range of wavelengths and the required product of cell pressure and cell length varies inversely with the wavelength. For cold neutrons (wavelength of 0.5 nm) a pressure-length product of 7 bar-cm of 75 % polarized ^{3}He yields 96 % neutron polarization with 27 % absolute transmission. By measuring the ratio of the neutron transmission through the spin filter with the ^{3}He polarized and unpolarized, one can determine the neutron polarization produced by the spin filter [19]. If the absolute transmission of the unpolarized spin filter and empty cell is also measured, the ^{3}He polarization can be determined. SEOP-based neutron spin filter cells are generally constructed from a boron-free aluminosilicate glass [20]. The ^{3}He polarization can also be monitored with nuclear magnetic resonance (NMR) [21, 22] and determined with electron paramagnetic resonance (EPR) [23, 24].

2. Spin filter development

The basic equation that governs the achievable polarization in SEOP cells [25] is

$$P_{\mathrm{He}} = P_{\mathrm{Rb}} \frac{k_{\mathrm{se}}[\mathrm{Rb}]}{k_{\mathrm{se}}[\mathrm{Rb}](1+X)+\Gamma_{\mathrm{r}}} \tag{2.1}$$

Here P_{He} and P_{Rb} are the ^{3}He and Rb polarizations, respectively, k_{se} is the spin-exchange rate coefficient, [Rb] is the Rb density, and Γ_{r} is the room temperature ^{3}He relaxation rate. X is a phenomenological parameter that describes the observation that the ^{3}He wall relaxation rate increases with alkali-metal density with a slope that exceeds k_{se}. Eq. (2.1) can be generalized for Rb/K mixture cells [25].

Until about a decade ago, it was assumed that the ^{3}He polarization would approach unity if the alkali-metal polarization were unity and the spin-exchange rate $\gamma_{\mathrm{se}} = k_{\mathrm{se}}[\mathrm{Rb}]$ were much faster than the ^{3}He wall relaxation rate. However, improved measurements of the spin-exchange rate

coefficient [26] revealed a linear increase in the ^{3}He wall relaxation rate with alkali-metal density for SEOP cells under operating conditions. The combination of SEOP cells with relaxation times of hundreds of hours [27], optical pumping with spectrally narrowed diode array lasers [28], alkali-metal polarization measurements [26], and accurate ^{3}He polarization measurements by neutron transmission [19] and electron paramagnetic resonance [23, 24] allowed for a detailed study of the limits of SEOP set by this relaxation phenomenon [25]. According to Eq. (2.1) P_{He} is limited to a maximum value of $1/(1+X)$. X has been found to vary from cell to cell, indicating that the source of the excess relaxation is primarily a surface phenomenon. Nevertheless, there must be also be some contribution from bulk anisotropic spin-exchange (ASE) [29], which would also vary linearly with alkali-metal density. The hyperfine interaction between the alkali-metal atom and the ^{3}He nucleus is dominated by the isotropic spin-exchange that arises from the Darwin term, but there is also a small contribution from the anisotropic part that arises from the dipole-dipole interaction. Whereas the isotropic part aligns the two spins, the anisotropic part anti-aligns them. Theoretical estimates of this effect yield a limiting value of $P_{\mathrm{He}} = 0.96$ [29, 30], and a direct experimental measurement yielded 0.90 ± 0.11 [31]. The highest ^{3}He polarization value reported in Ref. [25] and elsewhere [32] is $P_{\mathrm{He}} = 0.81$. More recently, we have obtained higher values, up to $P_{\mathrm{He}} = 0.85$, with diode lasers spectrally narrowed with chirped volume holographic gratings (VHGs) [33]. Details of these results and implications for the limits of SEOP will be discussed in an upcoming publication.

The achievable polarization and efficiency of SEOP has been greatly increased in the last decade by the use of spectrally narrowed lasers [28] and hybrid SEOP [18, 34]. Spectral narrowing from the typical free-running linewidth of 2 nm for diode bar lasers down to 0.25 nm has been performed with feedback from diffraction gratings [35]. More recently volume holographic gratings [36] have been employed in commercial [37, 38] or laboratory systems [33]. It has recently been shown that spectral narrowing avoids reduction in achievable polarization due to optical pumping with off-resonant light [39, 40]

In the typical pressure range of between 1 bar and 2 bar for spin filter cells, the alkali spin relaxation rate is dominated by alkali-alkali collisions. The operating alkali density and hence the polarizing rate is set by a balance between maximizing the spin-exchange rate γ_{se} while still maintaining P_{Rb} near unity. In the hybrid SEOP method, a mixture of Rb and K is employed to improve the efficiency of SEOP. The Rb is optically pumped and rapid electronic spin-exchange transfers the polarization from Rb atoms to K atoms. Because the spin relaxation rate for K is lower than that of Rb while the spin-exchange rates are comparable, a larger volume can be polarized for the same laser power or a given volume can be polarized more rapidly. These same advantages have been demonstrated for practical SEOP with pure K using 770 nm lasers, but Rb-K mixtures have three advantages: 1) the same laser can be used for both pure Rb and Rb-K mixture cells and the range of available lasers and output powers is greater for 795 nm lasers, 2) the small fine structure splitting for K may in some cases reduce the achievable polarization, and 3) empirically Rb-K cells have yielded better performance than pure K [34]. However, hybrid cells do require the additional complexity of distilling Rb and K with the appropriate mixture ratio into such cells. Typical vapor pressure mixture ratios $D = [\mathrm{K}]/[\mathrm{Rb}]$ are between $D = 2$ and $D = 6$ [34]. Whereas lower ratios simply result in less gain in efficiency, ratios above $\approx$10 yield decreased alkali-metal polarization for reasons that are only partially understood [41].

The apparatus and laser systems we employ have been described elsewhere [33]. Currently in each system we employ two lasers, each based on a 100 W diode bar [42] and spectrally narrowed with a chirped VHG. The VHGs [43] are 14 cm tall (dimension along the 10 mm bar length), 14 cm wide, and 1.75 mm thick, with a "chirp" of 0.15 nm/mm in the feedback wavelength along the width. Hence tuning of the wavelength is accomplished by simply translating the VHG across the laser beam. With the use of hybrid SEOP we obtain 80 % polarization in 1 liter cells with typical pumping time constants of between 4 h and 10 h.

3. Applications

3.1 Neutron scattering

Neutron spin filters have applications in neutron scattering and fundamental neutron studies. Both the magnetic scattering of neutrons by unpaired electrons and the scattering of neutrons by nuclei of non-zero spin can be strong functions of the neutron spin. The pioneering work of Moon, Riste, and Koehler [44] showed that polarized neutron scattering is a powerful technique to uniquely separate and identify the relevant cross sections. We have demonstrated the use of spin filters for small angle neutron scattering [45], reflectometry [46, 47] and triple-axis spectrometry [48]. In 2007 a program [49] to provide neutron spin filters for user experiments at the NCNR began and currently over 110 experiments conducted over 350 days of beam time have been supported. The primary applications have been small angle neutron scattering [50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61], triple-axis spectrometry [62, 63, 64, 65, 66, 67], and polarized neutron reflectometry. In general these experiments are conducted with ^{3}He cells that are polarized off-line, transported to the neutron beam line in battery-operated solenoids, and stored on the beam line in compact, magnetically shielded solenoids or other magnetic field apparatus [49, 68]. Since most experiments only last a few days, cells can be replaced every day or two as the ^{3}He polarization decays. Besides minimizing relaxation from magnetic field gradients, other key issues are neutron spin transport and manipulation. Once neutrons are polarized, they must always be in a "guide"field to preserve the polarization. Rotations of the field direction may be necessary as the neutron moves between the holding field for the ^{3}He spin filter, fields associated with neutron spin flipping devices, and potentially strong fields that may be applied to the samples under study. The ^{3}He polarization is often inverted on the beam line when space and/or field constraints make this approach preferable over neutron spin flippers.

Space constraints on neutron scattering instruments that were not necessarily designed for spin filters, as well as versatility to serve different instruments, have often made off-line SEOP preferable over polarizing on neutron beam lines. However, compact apparatus have been developed for in-situ optical pumping [69] and developed for the magnetism reflectometer the Spallation Neutron Source [70, 71].

3.2 Fundamental neutron physics

Neutron spin filters and polarized ^{3}He targets have also been applied in fundamental neutron physics, in particular few body nuclear physics and weak interaction studies [72]. At the NIST Neutron Interferometry and Optics Facilty [73] several experiments have been performed to measure the n-H, n-D, and n-^{3}He scattering lengths [74, 75]. These experiments are motivated by recent

advances in theoretical approaches, which now predict few nucleon properties such as scattering lengths with a precision on the order of 10^{-3}. A polarized ^{3}He cell located within the neutron interferometer was employed to determine the spin dependence of the n-^{3}He scattering length [76, 77]. The incident neutron beam was polarized with a supermirror and the polarization was inverted with a neutron spin flipper. This experiment had different systematics than those of the only other measurement, which was based on pseudo-magnetic precession in a spin echo apparatus [78]. Four 25 mm diameter, 42 mm long, optically sealed cells [79] filled with between 1.7 bar and 2 bar of ^{3}He were prepared for the experiment. These target cells were polarized off-line and transported to the interferometer. The bulk of the data was acquired with one particular cell that had an operational relaxation time of 115 h in the interferometer (reduced from its 330 h intrinsic relaxation time due to magnetic field gradients). The experiment required highly accurate knowledge of the neutron polarization, which was determined with an accuracy of less than 0.1 % using ^{3}He analyzer cells.

A ^{3}He spin filter [80] was employed at the Los Alamos Neutron Science Center (LANSCE) to provide a large area polarized neutron beam for the NPDGamma experiment. In this experiment, a series of parity-violating asymmetries in the emission of gamma rays in the absorption of neutrons by compound nuclei [81] and hydrogen [82] were determined. The spin filter was operated on the LANSCE beam line for nearly a year, which revealed effects for SEOP performed in neutron beams. Both a decrease in the achievable ^{3}He polarization correlated with neutron beam exposure and slow degradation of the optical transmission of the ^{3}He cells were observed. Whereas it has long been known that ionization produced by charged particle beams interferes with SEOP, such effects were not expected to be significant for neutron beams. However, neutron absorption by ^{3}He yields an energetic triton and proton that produce ionization in the gas. Studies revealed that the decrease in ^{3}He polarization was due to decreased alkali-metal polarization [83]. In later experiments with two orders of magnitude higher neutron flux obtained on the PF1b beam line [84] at the Institut Laue-Langevin (ILL), the alkali-metal spin relaxation rate was found to be proportional to square root of the neutron flux density, consistent with expectations for relaxation induced by ionization [85]. However, the detailed nature of the ionization mechanisms are not understood. Following the standard method for charged particle beams [86, 87] a double cell to decouple the optical pumping volume from the spin filter volume was tested, and found to be unaffected by the neutron beam. In further experiments the influence of gas composition was studied [88], in particular the role of the N_2 gas that is added to SEOP cells to suppress radiative decay of the 5p state of Rb [89]. The presence of two time scales for neutron induced alkali-metal relaxation was established. The fast component occurs on a time scale of less than 1 s and seems relatively constant with respect to [N_2] or captured power density. The slow component is increasing strongly with respect to captured power density or [N_2].

The slow decrease in optical transmission is associated with a white film that develops on the cell walls as it is used for SEOP in the neutron beam. Its origin is unknown but speculated to be due to the formation of RbH or alkali azides. It was observed that less whitening occurs for Rb-K cells [85]. Finally we note that all of these effects are associated with SEOP performed in neutron beams, not with cells that are polarized off-line by SEOP or MEOP and employed in neutron beams.

For the neutron capture flux density on the order of 10^8 s^{-1} for the LANSCE neutron beam, the increase in the alkali-metal relaxation rate was relatively minor. However, for the higher values

of up to 10^{10} s^{-1} available on PF1b at the ILL [84], the new NG-C beam line at the NCNR [90], and the Fundamental Neutron Physics Beam line at the Spallation Neutron Source [91], the beam induced spin relaxation may dominate the total alkali-metal spin relaxation rate. Primarily for this reason the continuation of the NPDGamma experiment at the SNS [92], where the flux density is about 30 times higher as compared to that obtained at LANSCE, will utilize a supermirror polarizer [93]. ^{3}He spin filters will continue to be employed for accurate neutron polarimetry [94]. We have initiated tests of double cells for large area neutron beams.

4. Wide-angle polarization analysis

Neutron polarization analyzers that can accommodate a large range of scattering angles are of interest for many neutron scattering instruments. Surmounting the challenge of wide-angle polarization analysis would substantially increase the impact of spin filters on polarized neutron scattering and thus is currently being pursued with spin filters based on MEOP [95, 96, 12] and SEOP [97]. The Multi-Axis Crystal Spectrometer (MACS) [98] at the NCNR is able to simultaneously detect neutrons from twenty independent channels covering an angular range of 220 ° around the sample region, yielding more than an order of magnitude gain in detection efficiency as compared to a conventional triple axis spectrometer. To perform polarized neutron scattering experiments with the MACS and other instruments, analyzers covering wide scattering angles are required.

A compact system based on polarized ^{3}He neutron spin filters to retrofit MACS with neutron polarization analysis has recently been reported [97]. The concept and techniques will be applied to other instruments at the NCNR and other neutron facilities. Our apparatus for wide-angle neutron polarization analysis consists of a vertical, neutron-compatible, solenoid that contains spin filter polarizer and analyzer cells close to the tail of a sample cryostat. The polarizer cell is contained in a shielded radio-frequency solenoid to permit inversion of the ^{3}He polarization via adiabatic fast passage (AFP) NMR with negligible loss of polarization in the analyzer cell. The wide-angle cells cover an angular range of up to 110 ° on either side of the neutron beam. Free induction decay NMR is used to monitor the ^{3}He polarization in the spin filter cells. Using conventional cylindrical cells [33], the apparatus was employed for a study of superconducting $Fe_{1+y}Te_{0.62}Se_{0.38}$ [99] on MACS. Recently we reported several upgrades to this apparatus [100], in particular the development of GE180 [20] wide-angle cells with relaxation times between 100 h and 400 h, and P_{He} between 0.65 and 0.80.

5. Relaxation studies

A variety of effects of magnetic fields on the ^{3}He relaxation time T_1 in sealed SEOP cells have been reported. The first effect observed was labelled "T_1 hysteresis"to describe the observation that the wall relaxation rate, measured in the typical magnetic field of 3 mT for SEOP, could be increased by a factor of between 2 and 20 by exposure of the glass ^{3}He cell to magnetic fields of a few tenths of a Tesla. However, the original T_1 could be recovered by demagnetization with an AC (alternating current) field [101]. Soon thereafter it was reported that even the weak magnetic fields typically used for SEOP could lead to orientation dependence, ie. the observation that the

room temperature relaxation time can depend on the orientation of the cell with respect to the magnetic field. In general the T_1 may depend on the direction, strength and history of the magnetic field [102, 103]. More recently, it was found that the T_1 can be shortened in fields of order 0.01 T, although exposure to such fields would not necessarily shorten the T_1 if the cell were simply exposed to this field and returned to 2 mT [33]. These effects have only been observed in cells with alkali-metal and are not understood. Magnetic field effects have also been studied in valved, cesium-coated, quartz cells filled with polarized gas produced by MEOP [104].

Wall relaxation in ^{3}He cells continues to be studied but remains difficult to understand [105, 106, 107]. In quartz SEOP cells an increase in relaxation at relatively low temperatures [108] and high values of X [100] have been reported. Recent studies for SEOP Pyrex cells [109] indicate that they may also exhibit higher than average X values, in particular for pure Rb cells.

Both the longitudinal (T_1) [110, 111] and transverse (T_2) [111, 112] relaxation time in polarized ^{3}He cells have been employed to set limits on new particles with masses in the sub-eV range. More recently, better limits have been obtained by determining the precession frequency shift of polarized ^{3}He and ^{129}Xe nuclei in the presence of an unpolarized mass [113, 114, 115].

6. Conclusions

Polarized ^{3}He spin filters are currently being applied worldwide to neutron scattering and fundamental neutron physics. A program to provide ^{3}He spin filter at the NCNR has been in place for several years, with increasing applications to neutron scattering. Future applications include wide-angle neutron polarization analysis. Continued development of SEOP-based spin filters has led to polarization values up to 85 % in large spin filter cells. However, the limits to the achievable polarization as well as various relaxation phenomena are not well understood. Whereas most application to date has been with remotely polarized spin filter cells, the use of in-situ SEOP will be increasing. For very high flux beams such as those used for fundamental neutron physics experiments, alkali spin relaxation and cell degradation due to the neutron beam will need to be addressed, perhaps with double cells as used for electron scattering.

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