Spin exchange optical pumping at pressures near 1 bar for neutron spin filters

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Motivated by applications to neutron spin filters and recent advances in spectrally narrowed laser diode arrays (LDAs), we are exploring spin exchange optical pumping of ^{3}He at pressures near 1 bar. Among our more interesting results has been the production of glass cells with extremely long relaxation times. The best of these has a lifetime of $T_1=840$ h [where the polarization decays versus time, t, as $\exp(-t/T_1)$], dominated by the dipole–dipole contribution of 950 h at a ^{3}He partial pressure of 0.85 bar. Using a broadband LDA, we have obtained 55% ^{3}He nuclear polarization in this cell. These results are particularly relevant to the application of ^{3}He-based neutron spin filters to neutron scattering and weak interaction experiments. Applications to magnetometry and polarized gas magnetic resonance imaging are also possible.
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Nuclear spin-polarized ^{3}He has been employed in several fields of physics, from magnetometry[1,2] and studies of nucleon structure[3–5] to more recent applications in polarized gas magnetic resonance imaging (MRI)[6,7] and polarized neutron research. The application of polarized ^{3}He-based neutron spin filters[8–10] to neutron scattering[11,12] and weak interaction experiments[13,14] places new demands on the production and storage of polarized ^{3}He. In particular, these applications require high ^{3}He polarization (50% or greater to obtain acceptable neutron transmission) in storage cells of a large cross-sectional area (to analyze divergent beams or polarize large area beams).

Two primary techniques exist to produce polarized ^{3}He: spin exchange optical pumping (SEOP),[15,16] whereby ^{3}He is polarized via spin exchange collisions with optically pumped Rb, and metastability exchange optical pumping (MEOP),[17,18] whereby ^{3}He is polarized via metastability exchange collisions with optically pumped ^{3}He metastable atoms. Each technique requires the minimization of polarization relaxation mechanisms, though for different reasons. For SEOP, the demand for high polarization requires slow depolarization rates, to compensate for the inherently long polarizing time constant (of the order of 10 h). In addition, many applications in neutron scattering can use neutron spin filters polarized off-line and transported to the neutron beamline. For MEOP, this manner of operation has been dominant, as the need for compression apparatus to achieve suitable ^{3}He pressures for neutron spin filters makes off-line operation preferable. For such operation, relaxation times of several days are desirable. We present results in SEOP and in cell development that have particularly important ramifications for neutron applications, and are potentially relevant to other applications of polarized ^{3}He.

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Newbury *et al.* demonstrated that dipole–dipole interactions in bulk ^{3}He limit the relaxation time of polarized ^{3}He to $807/P$ h (where P is the ^{3}He pressure in bar, for a cell temperature of 296 K).[19] This fundamental limitation on the relaxation time can only be approached if the contribution due to other interactions such as wall collisions[20] is negligible. Dipole–dipole dominated relaxation times have been observed in sealed SEOP cells made from aluminosilicate glass (lifetimes of 90 and 150 h observed in 8.9 and 5.0 bar cells, respectively, by Newbury *et al.*[19]), borosilicate glass (a lifetime of 300 h observed in a 2.5 bar cell by Smith *et al.*[21]), and sol-gel coated Pyrex[22] (a lifetime of 344 h observed in a 2.1 bar cell by Hsu *et al.*[23]). All of these cells contained Rb for the SEOP process, which has been shown to suppress wall relaxation as compared to bare glass cells.[9]

Studies of SEOP at pressures near 1 bar have not been vigorously pursued in the past, for three primary reasons. First, the electron scattering experiments that have driven the field require thick targets, and have operated near 10 bar to avoid excessive length. Second, the ratio of Rb density to ^{3}He density is largely irrelevant for SEOP, while the required laser power is set by the Rb volume. The greatest amount of polarized ^{3}He for a given laser power is then achieved at high pressures in small volumes. Third, the spectral overlap of the pressure-broadened Rb absorption line (around 18 GHz/bar) with the broadband emission from a laser diode array (LDA) (of order 1000 GHz) is maximized at higher pressures.[24] Recent developments in frequency narrowed LDAs[25,26] have inspired us to examine SEOP at pressures near 1 bar. We have constructed several cells with ^{3}He partial pressures as low as 0.85 bar, for which the dipole–dipole limit is 950 h. The best of these cells has a dipole–dipole dominated lifetime of 840 h (35 days), over twice as long as the longest relaxation time ever observed. Using a broadband LDA, we have obtained 55% ^{3}He polarization in this cell.

For this effort cells were constructed from GE180[27] (except where noted in Table I), an aluminosilicate glass chosen

TABLE I. Low pressure ^{3}He cell inventory, for SEOP (1–11) and MEOP (12–14) applications. Cylindrical cell dimensions in cm, length×diameter. The uncertainties provided reflect only the uncertainty in the fits to relaxation data. The lifetimes of cells 1, 7, and 11–14 were determined in our off-line FID apparatus, cells 2, 4, 5, and 9 on the neutron beamline, and cells 3, 6, 8, and 10 on our offline AFP apparatus. All cells constructed of reblown GE180 glass, except cells 10 (flat GE180 windows optically sealed to a Corning 1720 body), 11 and 12 (reblown fused silica).

Cell	Dim. (cm×cm)	^{3}He pressure (bar)	Lifetime (h)
1	4×5	0.85	840(16)
2	4×4.5	1.30	90 (2)
3	4×8.5	1.20	391(11)
4	4×10	0.85	350(12)
5	4×9.5	0.85	520(13)
6	4.5×9.5	0.85	185(10)
7	4.5×11	0.85	730(15)
8	3.5×11	1.25	55 (2)
9	4.5×11	0.85	98 (2)
10	4.5×5.0	0.85	270(20)
11	4.5×4.5	0.85	731(24)
12	10×7.0	Filled to 0.30	74 (1)
13	10×7.5	Filled to 0.30	240 (3)
14	10×7.5	Filled to 0.45	230 (7)

because it contains no boron (a strong neutron absorber), has sufficiently low ^{3}He permeability for long term operation at elevated temperatures, and is easier to work than other aluminosilicates. The largest GE180 tubing available is only 1.6 cm in diameter, requiring the glass to be completely reworked to produce large diameter cells. In our experience (and in the experience of others[19]), reblown glass has typically yielded the longest relaxation times. Our glass blower constructs and attaches the cells to a Pyrex tube (called the "string"). The string and cell surfaces are thoroughly rinsed: first with soapy water, then several times with distilled water, then with acetone, then several more times with distilled water, and finally with methanol. The string and cells are then attached to an oil-free, high vacuum system, where they are evacuated and baked at 400 °C for two days. The typical ambient pressure measured at the pump is 4×10^{-8} mbar. After this first bakeout, Rb is distilled from a side arm to a small reservoir (also baked) at the end of the string. The side arm is then removed ("pulled") from the string by fusing a glass constriction with a torch. The cells are then baked at 400 °C for two more days, the Rb is distilled into a second reservoir (very close to the cells), and the first reservoir is pulled from the string. The Rb is then distilled into the cells, typically as a vapor which condenses onto the cell walls. The cells are filled with approximately 65 mbar of N_2 gas (present to quench fluorescence of laser-excited Rb atoms during SEOP), followed by the desired amount of ^{3}He gas. Those cells filled to pressures greater than atmospheric were submerged in liquid N_2 to allow the cell to be pulled off. All gases were passed through a getter and filter stage to remove impurities before entering the cells.

The Rb (and thus the ^{3}He, in spin exchange collisions) is polarized by irradiating the cells, in ovens maintained at 140–180 °C, with laser light generated by a 32 W, 795 nm LDA with a bandwidth of ≤1.9 nm (900 GHz) (full width at half maximum).[28] For some tests, a second 16 W laser source (of similar bandwidth) was available. The laser light is split into two beams by a linearly polarizing beam splitter. The beams are independently circularly polarized and directed into the ^{3}He cell.

We have two stations where SEOP takes place, both equipped for adiabatic fast passage (AFP) nuclear magnetic resonance (NMR) to monitor the ^{3}He polarization (see Lorenzon *et al.*[29] for a description of NMR as applied to polarized ^{3}He). One of these stations is located on the NG6M 5 Å monochromatic neutron beamline at the National Institute of Standards and Technology (NIST) Center for Neutron Research.[8] This allows the performance of absolute neutron polarization measurements by neutron transmission, from which the ^{3}He polarization may be extracted and the NMR is calibrated.[8,10] At this station, laser power on the cell is limited to around 12 W, due to losses in optical components. At the second station, located off-line, incident laser power up to 37 W was available. Static magnetic fields of 2.0 mT are provided by 76-cm-diam Helmholtz coils.

Relaxation time measurements were also performed on an off-line apparatus[30] that is equipped for free induction decay (FID) NMR. For our best cell, we observe a relaxation time of 650 h on the neutron beamline, and 840 h on this off-line station (the difference has not been thoroughly investigated, but we suspect it is due to magnetic field gradients at the on-line system). Figure 1 shows the relaxation of the ^{3}He polarization for this cell in the off-line apparatus.

Table I lists an inventory of low pressure cells constructed for this study. A few cells are dominated by dipole–dipole relaxation and most have multihundred hour lifetimes.

We have performed extensive tests on cells 1 and 5 to determine the efficiency of SEOP near 1 bar with a broadband LDA. At the on-line station (12 W incident on the cell), we have achieved ^{3}He polarizations of 55(3)% in cell 1, determined by neutron transmission. The uncertainty in this measurement is dominated by the uncertainty in the neutron transmission through the cell windows, due to nonuniformities in their thickness. This result is comparable to results achieved on this system[8] and elsewhere[24] for higher pressure cells.

We find that with one half incident laser power (6 W), the relative decrease in polarization in cell 1 is only 5% (to 52%). The question of why higher power does not yield still higher polarizations arises. We note that models of SEOP often predict higher values of ^{3}He polarization than are actually observed.[31] To address this issue, studies to compare direct, independent measures of the ^{3}He and Rb polarizations are under way elsewhere.[31] Laser power is a more significant issue for larger cells, due to the increase in Rb volume. At the on-line station, we achieve a maximum ^{3}He polarization in cell 5 of 33(3)%. At the off-line station, we have observed 46(5)% ^{3}He polarization with 25 W incident laser power, and 50(5)% with 37 W. The off-line NMR is calibrated against neutron measurements on the cell transported between the two systems.

These results have the potential for profound impact in neutron scattering and tests of the weak interaction using cold neutrons. For mechanical stability, construction of large diameter cells is facilitated by operating pressures near 1 bar. The remarkably long relaxation times observed should allow a significant increase in the efficiency of off-line operation of optical pumping apparatus—the ^{3}He polarization in cell 1,

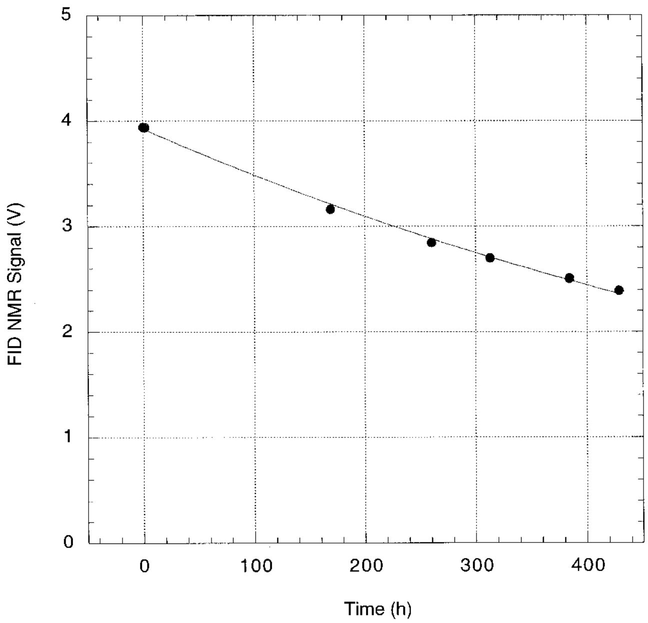

FIG. 1. Relaxation of the ^{3}He polarization in cell 1, measured in the offline FID NMR apparatus. The error bars, extracted from the fits of the individual FID measurements, are of the size of the data points. The data are fit to an exponential decay with a time constant of 840(16) h.

for example, would only decrease by 20% in one week. The high polarizations achieved are significant for weak interaction studies using cold neutrons, where continuous optical pumping is desirable due to the long time scales of such experiments. Operation near atmospheric pressure allows construction of cells with flat, optically sealed, windows.[32] Such cells are necessary for experiments that need uniform ^{3}He thickness so as to produce highly uniform polarization across the neutron beam, such as measurements of the polarized neutron beta-decay correlation coefficients A and B.[13] We have constructed one test cell (10) in this geometry.

While our results are most significant for neutron spin filter applications, we note in conclusion that the long lifetimes observed may be relevant to magnetometry and polarized gas MRI. For polarized gas MRI, long lifetimes are necessary if transporting the cells to remote imaging locations, but the cells cannot be sealed. Our results with valved cells (cells 12 and 13), tested on a MEOP compression apparatus,[30] indicate that long relaxation times are possible in this configuration.

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