Production of highly polarized ^{3}He using spectrally narrowed diode laser array bars

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We have produced 70%–75% ^{3}He polarization by spin-exchange optical pumping in cells $\approx 100\ \mathrm{cm}^3$ in volume. The polarization achieved is consistent with known spin-exchange and spin-relaxation rates, but only when the recently discovered temperature dependence of ^{3}He relaxation is included. Absolute ^{3}He polarization measurements were performed using two different methods in two different laboratories. The results were obtained with either a spectrally narrowed laser or one type of broadband laser. Based on tests of several larger cells at pressures near 1 bar, we find that the power required to reach the same polarization is typically three times lower for the spectrally narrowed laser. This last result indicates that spectrally narrowed lasers will be important for obtaining the highest polarization in large volume neutron spin filters. Polarization in excess of 55% as obtained in cells up to 640 cm^3 in volume and 70% polarization is anticipated with available increases in spectrally narrowed laser power.
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I. INTRODUCTION

Polarized ^{3}He and ^{129}Xe gases are currently being applied to a wide variety of scientific and medical problems. They include neutron spin filters,[1–3] magnetic resonance imaging,[4] spin-polarized targets,[5] surface science,[6] probing of biological systems,[7] polymer science,[8] precision measurements,[9] and quantum computation.[10] In this article we focus on producing highly polarized ^{3}He gas, in particular for applications such as neutron spin filters that require large volume cells. In addition, we focus on the spin-exchange optical pumping (SEOP) method,[11,12] which is currently better suited for continuous long-term operation than the metastability-exchange method[13,14] because the gas can be polarized directly at the required pressure. In the SEOP method, electronic polarization is produced by optical pumping of Rb atoms, and the polarization is transferred to the ^{3}He nuclei by the hyperfine interaction during collisions.

^{3}He polarizations of 72%–79% have been reported for 17 cm^3 cells that were optically pumped by a Ti-sapphire laser.[15] However, in that work only 55% was reported for 35 cm^3 cells of similar geometry. In an early test of a ^{3}He-based neutron spin filter, 70% polarization was reported for a 3 cm^3 cell that was optically pumped with a dye laser.[16] Most applications currently employ broadband high power diode laser arrays and cells one to two orders of magnitude larger in volume, and under such conditions we are not aware of any published reports of ^{3}He polarization that exceeds 60%.

Motivated by our recent results with long lifetime cells[17] and spectrally narrowed high power diode laser arrays,[18] we report polarization results for a variety of cells with volumes between 40 and 640 cm^3. Although some of these tests have been performed for conditions under which traditional rate balance theory would predict polarization values of nearly 100%, the maximum polarization we observed is 75%. Our polarization values are based on two independent measurement techniques in two different laboratories. This observation is consistent with recent measurements of temperature-dependent ^{3}He relaxation which were discovered in the course of new measurement of the spin-exchange rate coefficient.[19] While we are currently investigating the source of this limitation, in this article we focus on practical results, in particular SEOP of large volume cells at near atmospheric pressure for neutron spin filter applications.[17] Blown glass cells and cells with optical quality windows were tested using both narrowband and broadband lasers. We found that the narrowband laser and the broadband laser typically yield the same polarization for a broadband laser to narrowband laser power ratio of 2.5–3. Given that our results indicate that obtaining the highest polarization in very large cells would otherwise require well over 100 W of broadband light, this result is of great practical value for SEOP near 1 bar.

The use of high power diode laser bars rather than Ti:sapphire or dye lasers for spin-exchange optical pumping of Rb greatly reduces the cost and increases the reliability of spin-exchange optical pumping experiments.[20] In order to partially compensate for the broad frequency spectrum of such lasers (≈ 1 THz) it is common to operate at high gas pressures, typically between 5 and 10 bar, so that the pressure-broadened Rb linewidth (18 GHz/bar) more closely matches that of the laser. In optically thick spin-exchange

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TABLE I. ^{3}He cells used in this work and the maximum values of ^{3}He polarization, P_{He}, obtained in each cell with either 14 W of light from a spectrally narrowed laser (labeled 14N) or 50 W of light from the two broadband lasers (labeled 50B) used at NIST. All cells except Joe Cool are cylindrical, with diameter D and length L in cm, volume V in cm^3, and ^{3}He partial pressure P in bar. The cells and optical pumping conditions are described in the text.

Cell name	D	L	V	P	T_1 (h)	P_{He} (14N) (%)	P_{He} (50B) (%)
Joe Cool	6.5	1.4	44	3.5	110	63	63
Betty	4.7	4.9	81	0.87	140–300	71	70
Wilma	4.9	4.9	91	0.80	840	76	75
Sulu	4.8	6.2	110	0.90	110	62	⋯
Mars	6.0	8.0	227	0.85	105–270	65	69
Bullwinkle	6.9	7.1	268	1.22	550	63	64
BamBam	9.4	4.7	327	0.85	80–120	52	⋯
Dino	10.6	5.1	450	0.85	700	⋯	61
Astro	11.3	6.4	640	0.89	730	54	58

cells, however, it is still difficult to attain uniformly high polarization over the full cell volume because the relatively small fraction of resonant light is strongly absorbed as it propagates through the cell. In addition, whereas high gas pressures are well matched to some applications such as targets for electron scattering and polarized gas magnetic resonance imaging, cold neutron spin filters are best operated near 1 bar, primarily to avoid explosion of large diameter glass cells, especially cells with flat windows. In addition, operation at 1 bar allows convenient dimensions for typical gas path lengths between 4 and 8 bar cm. Using large volume cells at 1 bar not only decreases the utility of broadband light, it also increases the need for high optical pumping efficiency because of the increased volume. We have recently reported that SEOP at 1 bar can be effective,[17] but in this article we demonstrate higher efficiency using narrowband light and assess the potential for further increases in polarization for large volume cells.

II. APPARATUS

The cells for this work were constructed at the National Institute of Standards and Technology (NIST), Gaithersburg, Maryland, using techniques recently described elsewhere.[17] We report results with two different spectrally narrowed laser systems, one located at the University of Wisconsin and the other at NIST, both of which are based on the University of Wisconsin system.[18]

A. Cells

An important advantage of operating at 1 bar is the reduced contribution of dipole–dipole relaxation, which places a fundamental limit on the ^{3}He relaxation time of 807/P h (hours), where P is the partial pressure of ^{3}He in bar,[21] at a temperature of 295 K. We have recently reported cells blown from GE180 (Ref. 22) glass with polarization lifetimes (T_1) approaching the dipole–dipole limit of 950 h at 0.85 bar.[17] Although this success has not yet been achieved for cells with optical quality windows, we have still obtained relaxation times over 100 h in four such cells. The parameters of the cells used in this article are listed in Table I. The cells, denoted Betty, Mars, and BamBam, are nearly perfect right circular cylinders that were constructed by optically sealing flat windows of either GE180 glass (Betty and Mars) or ^{10}B-depleted Corning 1720 glass[23] (BamBam) to stock cylindrical Corning 1720 tubing. The cell Sulu is a slightly less perfect right circular cylinder, constructed by optically sealing flat GE180 windows to reblown GE180 tubing. The cells Wilma, Bullwinkle, Dino, and Astro are roughly cylindrical cells completely blown from GE180 glass. The cell Joe Cool is shaped like a meniscus lens so as to provide curvature for mechanical stability at a pressure of 3.5 bar, while still maintaining a reasonably uniform path length.

Whereas we obtain relaxation times of 150 h or longer with reasonably good reproduciability for blown cells, this is not yet true for cells made with flat, polished windows and either stock or blown cylindrical tubing. We have typically obtained lifetimes of between 10 and 80 h upon the first fill of such cells. A 240 h relaxation time was obtained in the cell Betty after repeated refilling of the cell; each refill involves opening the cell and cleaning out the oxidized Rb, and then repeating the filling process. For the cells Mars and BamBam improvement of the relaxation time was only obtained by performing a nitric acid rinse[21] in the course of a refill. Each of these three cells exhibit dependence of its relaxation time on its orientation with respect to the magnetic field, which has been recently reported both for cells that have been exposed to high magnetic fields[24] and for cells that have not been exposed to high fields.[25] To date we have found this effect to be more prevalent in cells that are not fully blown, but we have also observed it in blown cells. We have also found that the relaxation times can change over time, and this is possibly linked to when the cell is heated and cooled. In Table I the range of relaxation times that have been observed for Betty, Mars, and BamBam is listed.

B. University of Wisconsin apparatus

The work at the University of Wisconsin has primarily been done with a 30 W diode laser array bar specially selected for low array curvature ($<3\ \mu$m "smile").[18,26] The bandwidth is narrowed from 700 to $\approx$125 GHz using an external cavity with a 4× telescope and 2400 lines/mm grating. To limit the intracavity power to 30 W, the diode laser current is operated at a reduced value that yields 22 W from the laser in the absence of feedback. The resultant output power in the spectrally narrowed beam is 14 W, which yields three times higher spectral power (power per unit bandwidth) compared to the original unnarrowed laser. One cylindrical and one spherical lens convert the highly asymmetric multispatial mode laser output to a roughly square shape. For some tests, light from two additional broadband lasers was added. The linewidth of each broadband lasers was measured to be 1200 GHz.

The University of Wisconsin apparatus is equipped with several diagnostics[19] for measuring absolute Rb polarization (and polarization imaging), Rb density, Rb relaxation rates, light absorption, and ^{3}He adiabatic fast passage (AFP) and free-induction decay ^{3}He nuclear magnetic resonance (NMR). The cell is heated with hot air and its surface temperature is stabilized to better than 1 °C, measured by observing a small piece of blackened metallic tape with an optical pyrometer. Absolute ^{3}He polarization is deduced by the Rb electron paramagnetic resonance (EPR) frequency shift method,[27] with small corrections made for the nonspherical cell geometry. All of the EPR-based measurements of ^{3}He polarization presented in this article were on right circular cylindrical cells, which allows accurate calculation of these small corrections. We estimate the relative standard uncertainty in this measurement at 3%, and it is dominated by uncompensated drifts in the magnetic field. Employing this method requires knowledge of the density of the ^{3}He gas in the cell, which we determined by neutron transmission measurements[28] and by measurement of the Rb absorption profile, with relative standard uncertainties of 3% and 10%, respectively. For Betty, the pressure at 295 K was determined by the neutron and Rb absorption methods to be 0.876 ± 0.026 bar and 0.82 ± 0.08 bar, respectively. The value from neutron measurements, which also agreed well with the value of 0.872 bar recorded on the pressure gauge at the time the cell was filled, was used in determining ^{3}He polarization. The uncertainty in the pressure yields a total relative standard uncertainty of 4% for the EPR-based determinations of ^{3}He polarization.

C. NIST apparatus

At NIST, the holding field is provided by a pair of 75 cm diam Helmholtz coils operated at a field of 2.7 mT (27 G). The cell is heated in a cubical, hot air oven that is 16 cm on each side, and constructed of Teflon and glass. The laser beams enter through two 15 cm × 15 cm diam, 0.15 cm thick Pyrex windows that were masked to 13 cm diameter and are spaced 1.0 cm. Data were obtained with either 14 W of narrowband light delivered through one side of the oven or with 50 W of broadband light (900 GHz linewidth) split between opposite sides of the oven. The spectrally narrowed laser has an output power of 15 W, while the broadband laser light was provided by two 30 W lasers; the lower power delivered to the oven is due to loss in optical elements. All of the lasers were tuned by a low resolution spectrometer[29] to observe the absorption of laser light through a hot cell. For the narrowed laser, the temperature of the laser was adjusted such that the unnarrowed peak was on the atomic resonance, and the feedback grating was adjusted to place the narrowed peak on resonance. Proper tuning of the narrowed peak was occasionally checked using a photodiode to establish that the light transmitted by the cell was at a minimum. The wavelength of the narrowed peak was found to be quite stable.

The spectrally narrowed laser at NIST is essentially a duplicate of the University of Wisconsin design, but has not yet been characterized as well. The laser employed is a 20 W diode bar with a fast axis collimating lens and was specially selected for smile $<1\ \mu$m.[30] This laser was operated with less feedback than the Wisconsin system, resulting in higher output power (15 W) but also with incomplete suppression of the broadband peak. The linewidth without feedback is ≈ 900 GHz; aside from verification of the narrowing with the low resolution spectrometer,[29] the bandwidth with feedback has not been measured. A 100 mm focal length cylindrical lens was followed by a telescope formed by two spherical lenses with focal lengths of 100 and 300 mm, thereby allowing the range of rectangularly shaped beams needed for testing a variety of cells. The typical distance from the second spherical lens to the cell is 110 cm and the total distance from the laser diode to the cell is 240 cm.

The unpolarized beams from each broadband laser[26] diverged from fiber optic cables and were collimated to a diameter of either 2 or 4 cm. The collimating lenses were adjusted to provide sufficient beam divergence so as to overfill each cell. Two polarized beams were produced for each broadband laser by a 5 cm diam polarizing beamsplitter. Since there were four beams, it was convenient to deliver the broadband light to two sides of the cell. For each broadband laser, the two beams were separated 10 cm at the location of the 7.5 cm diam mica quarter-wave plates that were employed to circularly polarize the laser light. For one laser, the beams travel a direct distance of 160 cm between the plates and the cell, hence each beam was at an angle of 1.8° relative to the magnetic field axis. For the other laser, the beams traveled 270 cm to a 10 cm diam mirror on the opposite side of the Helmholtz coils, which redirected these beams for an additional 120 cm to the cell. Since the angle of incidence for this reflection was only 7°, the effect on the circular polarization was expected to be negligible.

While for the flat windowed cells (Betty, Sulu, Mars, and BamBam) and Joe Cool the laser light is sent along the axis of the cylinder, for the blown cells the light is sent through the side. This approach is employed because the thickness of the glass is fairly uniform on the sides but is rather nonuniform on the ends. Hence despite the curvature of the glass on the side, optical pumping from the side results in less lensing of the light and hence more uniform illumination of the cell volume. For Astro, side pumping presented an elongated cell profile, hence additional weak diverging cylindrical lenses were employed. For both lasers and all of the optical pumping conditions in this work, we estimate that skew light effects are minor.[31]

Temperature measurement was less accurate in the NIST apparatus compared to the careful University of Wisconsin measurements. The typical oven temperatures employed at NIST were between 160 and 170 °C, as measured by a resistance temperature device located in free space near the cell. Based on a recent measurement of the spin-exchange rate coefficient[19] and standard vapor pressure curves the spin-exchange time constants measured at NIST indicate that the true temperature is typically about 5 °C lower, with variations from test to test of a few degrees centigrade.

The polarization is monitored during optical pumping (or relaxation) with AFP NMR. The NMR drive field is produced by a pair of 45 cm Helmholtz coils, and the AFP signal is detected by a pair of 13.5 cm diam, 300 turn coils that are located just outside the sides of the oven. The NMR signal is calibrated by transmission measurements on the NG6M monochromatic beam line at the NIST Center for Neutron Research (NCNR).[28] This is accomplished by cooling the cell, transferring it to a battery-operated solenoid that provides a field of 1.3 mT, transporting it to the neutron beam line, and transferring it to another holding field on the beam line. A small correction is applied to account for the typical polarization loss of 2% (fractional) observed in the transfer. We estimate the relative standard uncertainty in the values of ^{3}He polarization to be $\pm 4\%$ for flat-windowed cells. For blown cells the thickness of the glass windows was estimated, increasing the uncertainty to $\pm 5\%$.

Given the time constraints on optimizing conditions for two different lasers and many cells, we consider the NIST results to be general indicators for a variety of cells, not careful comparisons for a specific cell. The most precise comparison of narrowband and broadband light would be obtained by simply operating the narrowband laser without feedback (see Sec. III A), because in such a comparison the beam size and shape is the same for both spectral conditions. However, the NIST tests were meant to address the more practical question of the relative performance of the spectrally narrowed laser compared to typical fiber-coupled broadband lasers.

III. RESULTS

Our results include a variety of tests to address different issues in achievable polarization. For the most part the University of Wisconsin and NIST results are complementary, but in a few cases were deliberately redundant so as to establish reproducibility between different apparatuses with different measurement techniques. The diagnostics available at the University of Wisconsin, in particular the capability of direct measurement of the Rb polarization, permitted a comparison of the absolute ^{3}He polarization achieved to that expected by simple rate-balance theory. At NIST the availability of a large variety of cells and a ^{3}He polarization measurement method that is more easily applicable to blown cells permitted practical tests of the polarization achievable under a variety of conditions. At each laboratory results with a spectrally narrowed laser were compared to those obtained with broadband lasers.

A. University of Wisconsin experiments

In Fig. 1 we present studies of the absolute ^{3}He polarization obtained in cell Betty as a function of the rubidium density. With 14 W of narrow-band pumping over the Rb density range between 1×10^{14} and 3×10^{14} cm^{-3} (corresponding to oven temperatures of between 150 and 175 °C) we consistently obtain 70% ^{3}He polarization. At higher Rb densities, the ^{3}He polarization drops because the laser power is insufficient to maintain 100% Rb polarization. Measurements of the Rb polarization are shown in Fig. 2.

The results shown in Fig. 1 were determined by fitting the rise in polarization for EPR measurements taken for typically two time constants. Scatter in these results is primarily due to the stability of the EPR measurement, with a smaller contribution from the uncertainty in fitting the final polarization. An additional consideration that may have produced some scatter in the data in Fig. 1 is the time and orientation dependence of Betty's room temperature relaxation time (see Sec. II A). These effects on the relaxation time were not discovered until after the University of Wisconsin data were obtained.

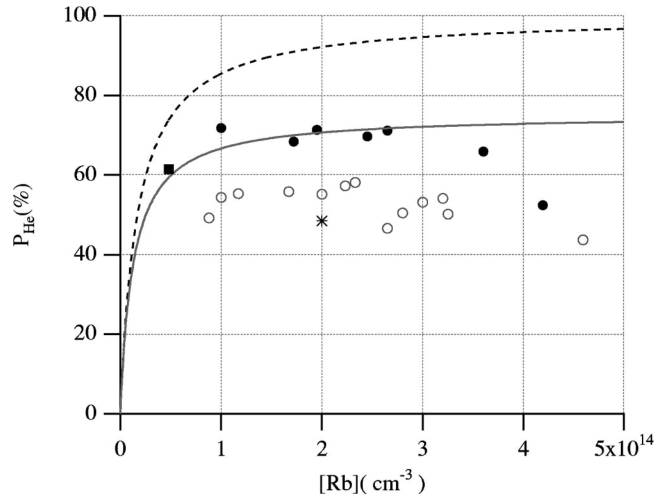

FIG. 1. Measured ^{3}He polarization vs the Rb density deduced from the Rb EPR frequency shift. The temperature range of the data shown is between 150 and 187 °C. Data obtained with 14 W of optical power from the narrowband laser are shown by closed circles, while data obtained with 42 W of broadband light are shown by open circles, except for the point at 2×10^{14} cm^{-3} (168 °C) shown by an asterisk, which was obtained with 28 W of broadband light. One measurement that was performed at NIST is shown by a closed square. (Since Rb density measurements were unavailable at NIST, the Rb densities were estimated based on the time constant for the rise in polarization.) The solid line shows the calculated polarization including the excess relaxation. The dotted line shows the predicted polarization assuming 100% Rb polarization and ^{3}He relaxation solely due to spin-exchange and room-temperature wall relaxation.

The advantage of using frequency-narrowed diode arrays for spin-exchange optical pumping is illustrated by the points at 2×10^{14} cm^{-3} in Fig. 1. After pumping the cell to 68% polarization with the frequency narrowed laser, we replaced the diffraction grating with a gold mirror, thereby returning the laser to its unnarrowed frequency spectrum of 700 GHz but increasing the output power by a factor of 2. The ^{3}He polarization so obtained is only 49%.

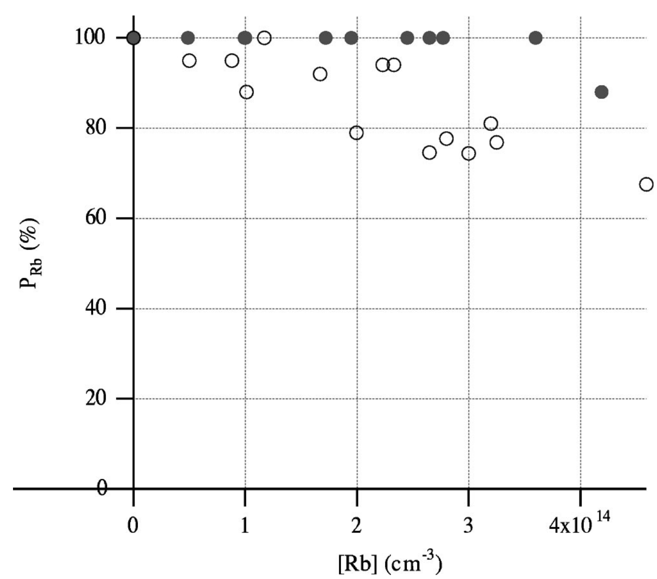

FIG. 2. Measured Rb polarizations vs the Rb density for optical pumping with either 14 W of spectrally narrowed laser light (closed circles) or 42 W of broadband light (open circles).

Figure 1 also shows polarization obtained by adding the output from two broadband diode arrays to the rear of the cell, bringing the total pumping power to 42 W. This increased the ^{3}He polarization to just under 60%, still substantially lower than that obtained with the frequency narrowed laser of only 14 W.

Despite the increased ^{3}He polarization obtained with the narrowband laser, there remains an important discrepancy between our measured values of the ^{3}He polarization P_{He} and the value we expect from independent measurements of the volume-averaged Rb polarization, P_{Rb}, the spin-exchange rate γ_{se}, and the room temperature wall-relaxation rate Γ_r. The ^{3}He polarization P_{He} is determined by a balance between spin-exchange and spin-relaxation rates:

$$P_{\mathrm{He}}=P_{\mathrm{Rb}}\frac{\gamma_{\mathrm{se}}}{\Gamma_{\mathrm{He}}}, \qquad (1)$$

where Γ_{He} is the total ^{3}He relaxation rate. (In Eq. (1), anisotropic spin-exchange[32] is neglected.) The dotted line in Fig. 1 shows the expected variation of the ^{3}He polarization with Rb density using known spin-exchange rates[19] and assuming $\Gamma_{\mathrm{He}}=\gamma_{\mathrm{se}}+\Gamma_r$. Using rf spectroscopy of the Rb, we have made absolute, position-dependent determinations of the Rb polarization and find that the volume-average Rb polarization exceeds 95% at Rb densities below $3.5\times10^{14}\ \mathrm{cm}^{-3}$ (180 °C). Hence if this simple rate balance theory were correct, we would expect our data to fall close to the dotted line shown in Fig. 1, whereas our data are typically 20% lower (fractional) than the dotted curve.

In the course of measuring spin-exchange rates, we recently measured the ^{3}He spin-relaxation rate as a function of the Rb density for several cells. We found, as expected from the argument in the previous paragraph, a linear dependence on the Rb density, but with the slope of the curve, $9.1\times10^{-20}\ \mathrm{cm}^3/\mathrm{s}$, 33% higher than the spin-exchange rate coefficient of $k_{\mathrm{se}}=6.8\times10^{-20}\ \mathrm{cm}^3/\mathrm{s}$.[19,33] This implies that Eq. (2) should be written in more detail as

$$P_{\mathrm{He}}=P_{\mathrm{Rb}}\frac{k_{\mathrm{se}}[\mathrm{Rb}]}{k_{\mathrm{se}}[\mathrm{Rb}](1+X)+\Gamma_r}, \qquad (2)$$

with $X=0.33$. If we use these values in Eq. (1) along with the measured room temperature relaxation time of 240 h for cell Betty, we obtain the solid line in Fig. 1. Hence the polarization data are consistent with the measurements of ^{3}He spin-relaxation rate and support the existence of an unknown excess ^{3}He relaxation that limits the attainable polarization to $P_{\mathrm{He}}<1/(1+X)$. Understanding the origin of this excess relaxation is critical to further advances in the polarization achievable with spin-exchange optical pumping.

When pumping with large quantities of broadband light, we find ^{3}He polarizations 15%–20% lower than for narrowband pumping. Our Rb polarization studies, presented in Fig. 2, show that most of this is due to reduced Rb polarization. The Rb polarization declines from 90% to 75% as the Rb density is increased and also shows variations that are due to sensitivity to issues such as beam alignment. This explains the greater scatter for the broadband data in Figs. 1 and 2. However, we find some interesting effects that we cannot yet explain. For example, employing more than about 30 W of laser power does not lead to an increase in either Rb or ^{3}He polarization.

The cell BamBam, which has over four times the volume of Betty and a average relaxation time of only 100 h, was also tested at the University of Wisconsin. After careful optimization of the beam size and oven temperature, 63% polarization was obtained with 17 W of spectrally narrowed light. The polarization measured is consistent with the value expected given the measured room temperature relaxation time, but again only if recent measurements of the spin-exchange rate coefficient and temperature dependent ^{3}He relaxation rate are utilized.[19]

B. NIST results

The University of Wisconsin results indicate that 70% polarization is achievable in a flat-windowed, long lifetime, relatively small cell, and that this polarization is limited by the temperature-dependent ^{3}He relaxation. At NIST we confirmed this result with a different method for measuring ^{3}He polarization and evaluated the polarization in a variety of cells with either shorter lifetime, curved windows, larger volume, or some combination of these characteristics. These tests are important for a variety of applications, but here we focus on issues for neutron spin filters: (1) constructing cells with lifetimes of greater than 200 h is not always guaranteed and the lifetimes of spin filter cells may be reduced because of operation in magnetic field gradients from high field magnets. (2) We and others[21] have found that long lifetimes can be obtained with greater reliability in blown cells. Blown glass causes optical effects such as skew light[31] and degradation of the circular polarization of the laser light, which may reduce the ^{3}He polarization achievable compared to flat-windowed cells. (3) Cells with both large diameter and volume are needed for neutron applications. The results are listed in Table I.

The time constants for these tests were typically between 10 and 20 h, which sometimes makes it difficult to wait long enough to observe the final polarization. The values reported in Table I are actual observed polarizations; whereas these are essentially the final values, we have noted in the text cases where we extrapolated to slightly higher final values at infinite time. When calculated polarizations are stated, they were determined by measured room temperature relaxation times, and recent measurements of the spin-exchange rate coefficient and temperature-dependent ^{3}He relaxation rate.[19]

For Betty we obtained 72% polarization using the spectrally narrowed laser, in agreement with the University of Wisconsin results. In contrast with the University of Wisconsin broadband results, we obtained 71% (68%) polarization with 50 W (25 W) of broadband light. The reason for the difference in the NIST and Wisconsin results in the tests with broadband light is not completely clear, but may be due to the larger linewidth of some of the broadband lasers that were employed at the University of Wisconsin.

The results with the blown cell, Wilma, indicate that there are no significant issues with pumping through the curved cell sides, at least for this relatively small cell. The slightly higher polarization obtained in Wilma compared to that obtained in Betty is most likely due to Wilma's very long lifetime, which is close to the dipole–dipole limit. Similarly, the lower polarization in the cell Sulu is consistent with its shorter lifetime.

With the cell Mars, we also compared the polarization obtained with 26 W of broadband light delivered to one side of the cell to that obtained with 13 W delivered to each side of the cell. Aside from some minor differences in optics, the total power is the same, hence this experiment tests whether a higher volume-averaged Rb polarization is achieved by pumping from both sides of the cell. Since Mars has flat windows, lensing effects are not an issue. We found that pumping from both sides (one side) yielded 60% (57%) polarization.

The effect of limited laser power is more pronounced in BamBam. The polarization of only 52% is lower than the value of 63% obtained at the University of Wisconsin, which we attribute to lower laser power and less attention to optimization. We planned to refill BamBam in an attempt to improve its lifetime and then perform more careful measurements at NIST, but the seal of one of BamBam's windows was lost in this attempt and has not yet been repaired.

The polarization achievable in the larger cells, Dino and Astro, were clearly limited by laser power and thus provided the best tests of the relative efficiency of spectrally narrowed and broadband light. For Astro polarization of 54% was obtained with spectrally narrowed light, slightly lower than the value of 58% obtained with broadband light. For Astro there was an error in the temperature measurement; based on a time constant of ≈ 60 h, we expect that the true temperature was just over 140 °C. For both the spectrally narrowed and broadband cases, the extrapolated polarization was 60%.

The results with the cell Joe Cool indicate that the improved efficiency of the spectrally narrowed laser is also evident at 3.5 bar. To increase the sensitivity of the test, we decreased each power, resulting in 60% for 7.5 W of narrowed light and 55% for 25 W of broadband light.

Systematic tests of the temperature dependence of the polarization have not been performed, but a few general features have been established. In the larger, long lifetime cells such as Bullwinkle and Astro, typically $\approx 15\%$ higher polarization is obtained at 160 °C than at 170 °C, indicating that imperfect Rb polarization limits the ^{3}He polarization. For these large, long lifetime cells, the best polarization for long-term operation may be obtained at quite low temperature (such as in the Astro test), but time considerations did not allow us to fully investigate this regime. In the case of BamBam, the relatively short lifetime does not allow operation at such low temperature, and careful optimization was required to balance the conflicting needs of high Rb polarization and short spin-exchange time, a familiar characteristic of SEOP. While cells for which the laser power is insufficient to maintain 100% Rb polarization throughout the cell provide the best test of the relative efficiency of narrowband and broadband light, these same properties make the achievable polarization sensitive to temperature and beam size and shape.

A survey of the results indicates that slightly higher polarization is generally obtained with 50 W of broadband light compared to 14 W of spectrally narrowed light. In the course of our development, we performed some additional tests with 38 W of broadband light and 10 W of spectrally narrowed light; based on all of our data, we conclude that the spectrally narrowed laser can produce the same polarization as the broadband laser for a broadband laser to narrowband laser power ratio of 2.5–3.

Based on other data not shown it appears that the achievable polarization in Bullwinkle is still rising with increasing laser power, with an asymptotic value of 75% in the limit of infinite power. This is consistent with the observed limit for Wilma and with the existence of excess relaxation.

IV. CONCLUSION

We have optically pumped a variety of cells and reported the polarization achievable for both narrowband and broadband pumping. We observe a maximum of 75% polarization, consistent with the recent discovery of excess ^{3}He relaxation. Understanding of this process is essential in order to realize the full potential of spin-exchange optical pumping and thereby attain ^{3}He polarization of over 90%. We have shown that spectrally narrowed lasers are substantially more efficient than broadband lasers. In situations in which fiber-coupled, turn-key lasers are needed, we still observed that the highest polarization could be achieved with substantially higher power provided by broadband lasers, but such values are not necessarily always guaranteed.

We expect that with doubling of the narrowband laser power from 14 to 28 W, along with careful evaluation of the best optical conditions to maximize the use of that power, it should be possible to approach 70% in large neutron spin filter cells. Such doubling could be possible with a single 40 W diode bar, which we are currently pursuing. In contrast, such doubling would require 120 W of broadband power, which at the very least would be expensive, as well as inconvenient because of the multiple beams needed. Hence these results illustrate that narrowband lasers will be of great utility in reaching 70% polarization in large cells.

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