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To cite this article: Thad G Walker 2011 *J. Phys.: Conf. Ser.* **294** 012001

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Fundamentals of Spin-Exchange Optical Pumping

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Abstract. The standard model of Spin-Exchange Optical Pumping of ^3He has come under close scrutiny as researchers try to maximize the quantity and polarization of ^3He . This paper will review four important effects that have come to light: 1) pumping with frequency narrowed diode lasers significantly improves the polarization and production rate as compared to off-the-shelf broadband sources 2) Rb-K mixtures again improve the production rate due to reduced collisional loss in K-He collisions, 3) a unexplained ^3He wall relaxation mechanism that scales closely with alkali vapor pressure limits the attainable ^3He polarization to about 80%, 4) a small collisionally-induced breakdown in the selection rules for optical pumping explains why narrowband lasers are so advantageous, and may explain some observed limitations for Rb-K pumping as well.

1. Introduction

The Spin-Exchange Optical Pumping (SEOP) method for producing spin-polarized ^3He is in principle relatively simple and inexpensive to implement experimentally, yet still provides excellent performance. The basic processes governing SEOP were reviewed a number of years ago [1], and are summarized by the following equations:

$$\frac{dP_{\text{He}}}{dt} = k_{\text{SE}}[A](P_{\text{A}} - P_{\text{He}}) - \Gamma_{\text{He}}P_{\text{He}} \quad (1)$$

$$P_{\text{A}} = \frac{R(z)}{R(z) + \Gamma_{\text{A}}} \quad (2)$$

$$\Gamma_{\text{A}} = (k_{\text{SE}} + k_{\text{SR}})[\text{He}] + \Gamma_0 \quad (3)$$

$$\frac{dR(z)}{dz} = -[A]\sigma_L\Gamma_{\text{A}}P_{\text{A}} \quad (4)$$

Equation 1 states that polarization is transferred back and forth by spin-exchange collisions between the alkali atoms of polarization P_{A} and the ^3He nuclei of polarization P_{He} . The transfer rate, proportional to the alkali density $[A]$, is typically 0.1/hr. The ^3He polarization is also lost to the walls of the container at a rate Γ_{He} which at room temperature is now routinely less than 0.005/hr [2]. Assuming no temperature dependence to the wall relaxation rate, Eq. 1 predicts that the steady-state polarization should be $P_{\text{He}} > 0.95P_{\text{A}}$.

Equation 2 states that the alkali polarization at position z in the cell is a competition between optical pumping (rate $R(z)$) and alkali spin-relaxation processes (rate Γ_{A}). The polarization is high as long as $R(z) \gg \Gamma_{\text{A}}$. The relaxation processes, Eq. 3, include spin-exchange (k_{SE}) and spin-relaxation (k_{SR}) collisions with ^3He , and other processes such as relaxation in alkali-alkali collisions and diffusion to the walls.

Equation 4 states that the pumping rate, which is proportional to the light intensity, decreases as the light propagates through the optically pumped vapor. As long as the alkali polarization is nearly 1, the attenuation is linear with distance. Absent the optical pumping to the dark state, the absorption length of $1/[A]\sigma_L \approx 50 \mu\text{m}$ for an atomic absorption cross section of $\sigma_L \approx 5 \times 10^{-13} \text{ cm}^2$ in a $[\text{He}]=1 \text{ atm}$ cell implies that the light would be completely absorbed less than a mm after entering the cell.

Now let us estimate the performance that should be attained. We assume $P = 10 \text{ W}$ of light entering a 10 cm diameter, 4.5 cm long cylinder with 1 atm of ^3He . The vapor is Rb, at a density of $4 \times 10^{14} \text{ cm}^{-3}$. The pumping rate entering the cell of area A is $R(0) = \sigma_L P / (\hbar \omega A) = 2.6 \times 10^5 / \text{s}$. The estimated spin-relaxation rate is $\Gamma_A = 270 / \text{s}$, mostly from Rb-Rb collisions [3]. At the exit of the cylinder, numerical solution of Eq. 4 finds $R(4.5) = 0.05R(0)$, with $P_A = 0.978$. The cell-averaged polarization is in excess of 99%. The ^3He polarization should be in excess of 95%.

In practice, in order to successfully pump a cell of this size requires 50 W or more of frequency narrowed diode laser light [3], and the ^3He polarization is found to be at most in the high 70% range. When Ref. [1] was written, the polarizations obtained with high power diode arrays were typically in the mid-50% range or lower. In this paper I will summarize some of the effects that have been discovered and some of the experimental techniques that have been introduced in our laboratory at Wisconsin, working closely with our colleagues at NIST, to improve this situation.

2. Frequency-narrowed lasers

Wagshul and Chupp [4] first showed that it is possible to use high power semiconductor lasers for SEOP of ^3He . Previously, ion-laser pumped dye lasers had been used. Comparatively, semiconductor lasers offer greatly reduced price and greatly increased reliability, crucial for stable operation over the 10s of hours required to produce the polarized ^3He . The principal drawbacks were poor temporal and spatial mode structures of these lasers. In particular, the linewidths of these lasers tended to be several nm corresponding to 1 THz or greater in frequency units. When compared to the 20 GHz pressure-broadened linewidth at 1 atm ^3He pressure, this appeared to be a significant disadvantage. However, since SEOP cells are extremely optically thick at operating temperatures, light in the spectral wings of the laser is still absorbed, albeit at a smaller rate [5]. Even when high power arrays were used, it was found that polarizations were at most 60%.

In the late '90s, use of frequency narrowed semiconductor lasers began to be pursued. Levron *et al.* [6] used a custom laser for CsXe SEOP. In our laboratory, Ian Nelson and Bien Chann showed that off-the-shelf diode arrays [7] and diode-array bars [8] could be frequency narrowed, and with Tom Gentile's group at NIST showed that the polarizations dramatically improved, into the mid 70% range [9], as shown in Fig. 1.

The linewidth of these sources, typically 50-100 GHz is well-matched to the lineshape for optical pumping at 5-10 atm, but is still broader than would be best for neutron physics applications that prefer on the order of 1 atm. Thus the recent demonstration of Knize's group [10] of $< 2 \text{ GHz}$ linewidth for an external cavity laser with a high quality AR coated laser chip is intriguing for future improvements.

3. Hybrid Spin-Exchange Optical Pumping

In 1998, Baranga *et al.* [11] showed that the fundamental spin-exchange and spin-relaxation mechanisms for K- ^3He are significantly favorable as compared to Rb- ^3He . In practice, however, diode lasers for K are generally more difficult to obtain and are less developed than for Rb. For this reason, we demonstrated a hybrid optical pumping scheme in 2003 [12] that potentially allows for the best of both worlds: a high density K vapor is polarized by efficient spin-exchange collisions with a low density optically pumped Rb vapor.

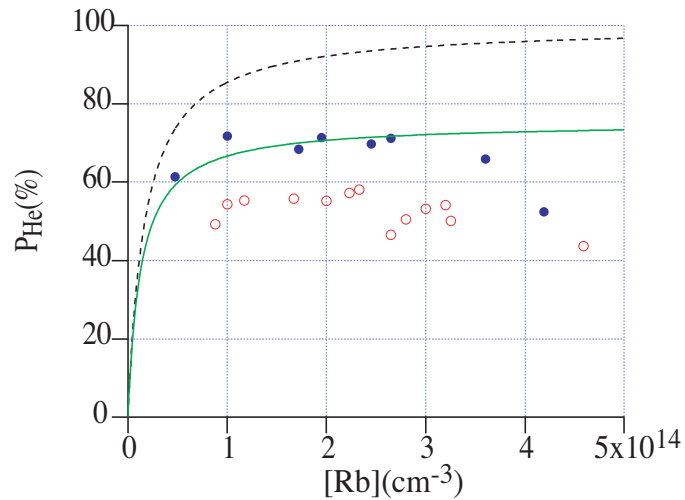


Figure 1. Comparison of ^3He polarizations obtained with broadband (42 W, open circles) and frequency narrowed light (14 W, closed circles) for ^3He SEOP. The solid line shows the calculated polarization including an unknown excess relaxation (see Sec. 3). The dotted line shows the predicted polarization assuming 100% Rb polarization and ^3He relaxation solely due to spin-exchange and room-temperature wall relaxation. From Ref. [9].

The advantages of hybrid spin-exchange can be understood with the following argument. The polarization achieved in the Rb is as always a balance between the optical pumping rate and the spin-relaxation rate. But in a hybrid cell each Rb atom has an effective spin-relaxation rate that is the sum of its direct relaxation rate and the relaxation of $\mathcal{D} = [\text{K}]/[\text{Rb}]$ K atoms. Thus the spin-polarization (Eq. 2) for hybrid spin-exchange becomes

$$P_A = \frac{R}{R + \Gamma_{\text{Rb}} + \mathcal{D}\Gamma_{\text{K}}} \quad (5)$$

The photon demand, number of photons scattered per second per alkali atom, is

$$\phi = \frac{R(1 - P_A)}{1 + \mathcal{D}} = \frac{\Gamma_{\text{Rb}} + \mathcal{D}\Gamma_{\text{K}}}{1 + \mathcal{D}} P_A \quad (6)$$

which is less than the value $\Gamma_A P_A$ for pure Rb pumping. With increasing K density, the additional relaxation increases more slowly than the atom density.

The increased capacity of hybrid spin-exchange can in principle be used to increase the spin-exchange rate (Fig. 2) or increase the polarized volume. However, it was discovered in the original hybrid experiments [12] that P_{He} begins to decrease for large \mathcal{D} . This was speculated to be a result of the K atoms having a non-negligible probability for absorbing the optical pumping light. This highly-off-resonant absorption is likely insensitive to the K spin-polarization, and therefore acts as an effective spin-relaxation mechanism. Experiments to quantify this possibility are currently being made by Brian Lancor.

A careful study of the gains from hybrid spin-exchange was led by Wangchun Chen and Tom Gentile [3]. Taking into account light propagation effects and tradeoffs between polarized volume and speed, it was found that the range $\mathcal{D} \sim 2 - 6$ generally gives the best performance.

4. He polarization limits

Models of the SEOP process, using known spin-exchange rates and measured room-temperature wall-relaxation rates, consistently predict $P_{\text{He}} > 0.9$, whereas the highest values measured to

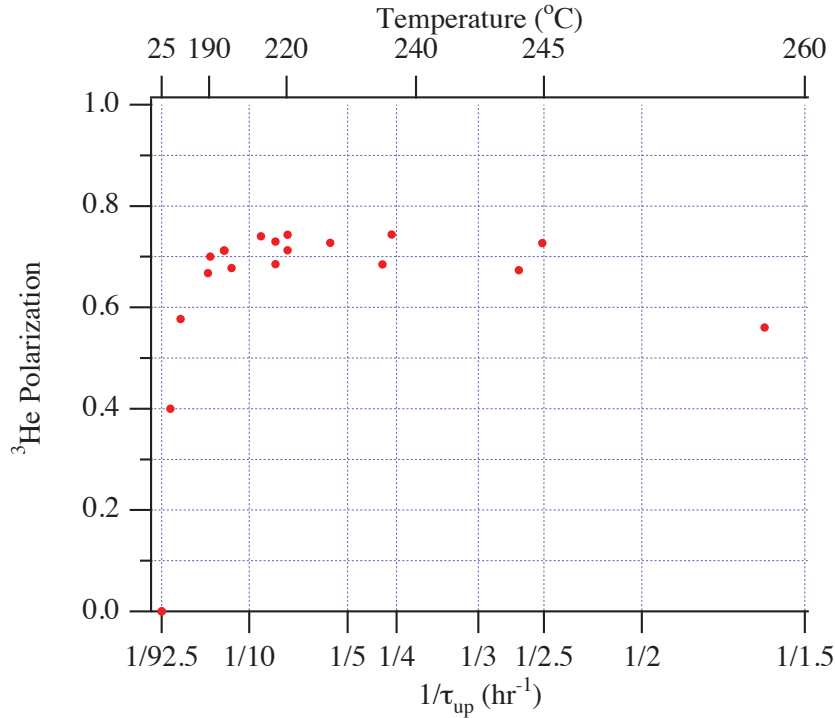


Figure 2. Fast spin-exchange optical pumping of a 25 cm³, 3.3 amagat hybrid $\mathcal{D} = 1.5$ Rb/K cell using a frequency-narrowed diode laser. From Ref. [12].

date are about 0.8. From Eq. 1, the ^3He polarization should be

$$P_{\text{He}} = \frac{k_{\text{SE}}[\text{A}]P_{\text{A}}}{k_{\text{SE}}[\text{A}] + \Gamma_{\text{He}}} \quad (7)$$

At high alkali densities, this should approach P_{A} unless Γ_{He} also increases at high $[\text{A}]$. The first indication that Γ_{He} has a strong temperature dependence was uncovered in Bien Chann’s measurements of k_{SE} for Rb [13], where the values of k_{SE} deduced from wall-independent methods were found to be consistently smaller than indicated from the traditional method of studying the temperature dependence of spin-exchange time constants.

A systematic study of this “X-factor” effect was undertaken by the NIST and Wisconsin groups [14]. The wall-relaxation rate of ^3He was indeed found to vary as

$$\Gamma_{\text{He}} = \Gamma_0 + k_{\text{SE}}[\text{A}]X \quad (8)$$

where X is a parameter that was found to be 0.2 or greater, but to fluctuate wildly from cell to cell. The fluctuations scale with surface-to-volume ratio S/V , as seen in Fig. 3:

$$X = X_0 + \chi \frac{S}{V} \quad (9)$$

where χ is a poorly controlled parameter, varying randomly from cell to cell. Large cells have smaller fluctuations than small ones. At high alkali density, the polarization limit becomes

$$P_{\text{He}} < \frac{1}{1 + X} \quad (10)$$

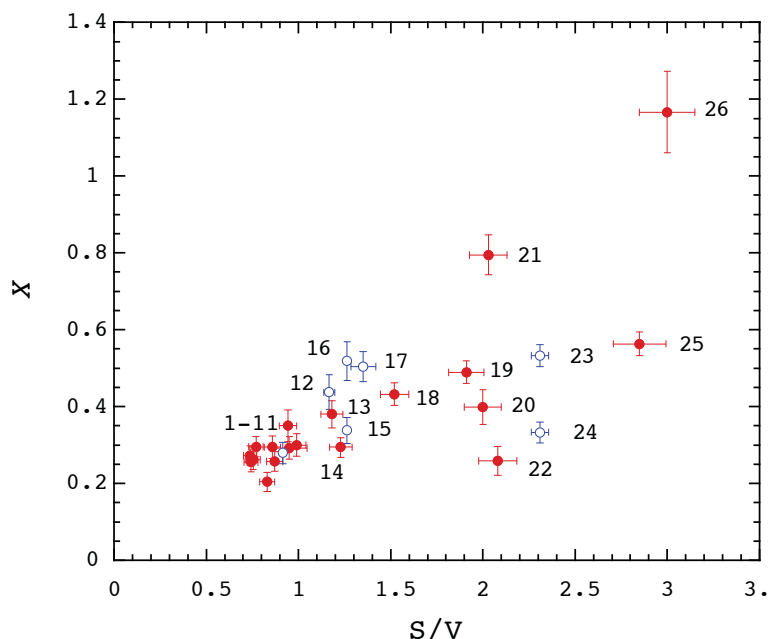


Figure 3. Measured X-factors for a variety of SEOP cells, done at NIST. From Ref. [14], where the characteristics of the various cells, identified here by number, are listed.

thus explaining why the X-factor causes the He polarization to reach a hard limit at high alkali densities.

A histogram of the data from Ref. [14], reparameterized in terms of χ , is shown in Fig. 4. The distribution is approximately Gaussian, with a mean of $\bar{\chi} = 0.31$ cm and a standard deviation of $\sigma_{\chi} = 0.08$ cm. This seems to support the hypothesis that the X-factor is linearly correlated with surface-to-volume ratio.

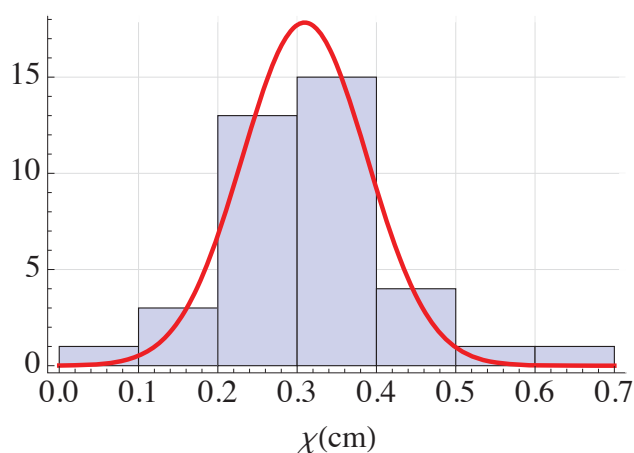


Figure 4. Distribution of χ -values, where $X = \chi S/V$, for the 38 cell sample reported in Ref. [14]. The solid line is a best-fit Gaussian distribution with a mean of 0.31 cm and a standard deviation of 0.08 cm.

The lack of cells with $X < 0.2$ brings up the question of whether or not there might be a fundamental process contributing to X . The only known such process is anisotropic spin-

exchange [15]. Experimentally demonstrating anisotropic spin-exchange is difficult, as the effects are difficult to disentangle from wall effects when making measurements on ^3He relaxation, or appear simply as increased spin-relaxation rates when measuring alkali relaxation. A recent experiment [16] introduced a method that in principle can unambiguously isolate the effect, but was not sensitive enough to make a measurement of the effect: $X_0 = 0.10 \pm 0.13$.

5. Light-Induced Relaxation

In the first hybrid experiment [12], we observed that for broad-band pumping it was impossible to fully polarize the alkali atoms, even at low alkali density and very high laser intensities. This brought up the possibility that in the presence of high pressure He fully polarized alkali atoms might still absorb the circularly polarized pumping light, contrary to the selection rules for free atoms. Due to rapid excited-state spin-relaxation, such a process would act to *de-pump* the atoms and lead to a maximum alkali polarization P_∞ at high pumping rates. Even if P_∞ is very close to 1, the extreme optical thickness in SEOP cells can make this process very damaging. This is because the light-induced relaxation with cross-section σ_∞ adds to Eq. 4 a standard Beer's law type term:

$$\frac{dR(z)}{dz} = -[A]\sigma_L\Gamma_A P_A - [A]\sigma_\infty R(z) \quad (11)$$

High optical depths and alkali polarizations require $R \gg \Gamma_A$, causing $\sigma_\infty R$ to be comparable to or even greater than Γ_A .

Initial measurements of this effect were made for off-resonant pumping light by Earl Babcock in his thesis [17], with a careful, complementary on- and off-resonant study being done by Brian Lancor [18, 19]. Such measurements are equivalent to studying the spin-dependence of pressure-broadening, and much of the molecular physics had been studied before in the context of fine-structure changing collisions [20]. Nevertheless, to our knowledge the effect of pressure-broadening collisions on the angular momentum selection rules had never been observed experimentally before.

Figure 5 shows how the light-induced absorption affects light propagation. The power and polarization through the cell are shown. As the light traverses the cell, the off-resonant pumping increasingly favors σ_∞ as compared to σ_L . The Rb polarization drops much more quickly with the dichroism effect than would be expected just from normal off-resonant pumping. We believe that this explains for the first time why such large polarization increases were observed when implementing narrowband pumping; the narrowband pumping light not only increased the pumping rate but also decreased the relative importance of σ_∞ as compared σ_L .

6. Conclusions

While considerable progress has been made in improving both the performance and understanding of SEOP over the past decade, important problems remain. Gauging from the response of the attendees at the JCNS Workshop, a solution to the X-factor problem would be most desirable. It would be nice to know if the observed X_0 arises from anisotropic spin-exchange. On this problem we are encouraged to note that the ITAMP group has become quite interested in *ab-initio* calculations of spin-dependent interactions of van-der-Waals molecules [21] and they may soon have a theoretical prediction for X_0 . In my opinion it is unlikely that the result will be larger than 0.1. We would also like to improve on the experiment of Ref. [16] to at least get a measurement of X_0 for KHe. The much larger spin-relaxation rates for Rb-He make such an approach extremely difficult for RbHe.

Concerning the surface contribution to X , we note that the materials that have been studied to date have all been glasses (really, glasses coated with alkali films) such as pyrex, quartz, GE180, 1722, and sol-gels. Of course it is necessary to have glass surfaces to allow the light into

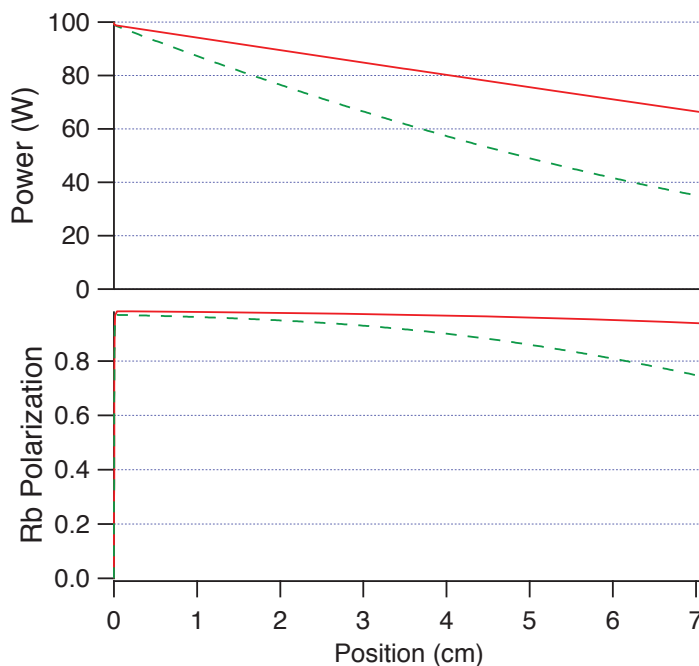


Figure 5. Optical pumping by a broad pump laser without (red, solid) and with (green, dashed) dark-state absorption. The upper graph shows that the dark state absorption produces a faster attenuation of the light. The lower graph shows how the dark state absorption reduces the Rb polarization at the cell entrance, with further reductions as the spectral profile of the light becomes increasingly off-resonant deeper inside the cell. From Ref. [18].

the cell. An example of an interesting experiment to try would be to coat the cell interior with a sub-skin-depth layer of gold or copper to see what differences that might make. Unfortunately, there is no model for the surface physics that gives rise to the X-factor, so it is difficult to know what particular metals might be promising.

We are currently engaged in a new assessment of the “photon budget” for SEOP. Given what we now know about light-induced relaxation and hybrid spin-exchange, along with other topics such as excited-state relaxation [22], radiation trapping [22], and cell heating [23], we hope that we are close to being able to model the SEOP process sufficiently accurately to account for the amount of light required to polarize ^3He .

Acknowledgments

I would like to thank I. Ioffe, E. Babcock, and T. Gutberlet for organizing the excellent JCNS Workshop on “Modern Trends in Production and Applications of Polarized ^3He ”. The work summarized here was performed by a long series of excellent students and collaborators. At Wisconsin, S. Kadlecik, I. Nelson, B. Chann, E. Babcock, and B. Lancor have all made extremely important contributions to SEOP of ^3He . We have enjoyed a long and fruitful collaboration with T. Gentile and his group at NIST. Our early work on these problems was supported by the National Science Foundation, with the recent work supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under award DE-FG02-03ER46093.

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