Lifetime behaviour and polarization stability in $^3$He neutron spin filter cells

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Abstract. The successful use of $^3$He cells in polarized neutron applications demands that once polarized the gas has both a long relaxation time and high polarization. In spin-exchange optical pumping the cell characteristics determine the maximum polarization as well as the lifetime.

We report on our experience of neutron spin filter cells, showing results on the orientation dependence of such cells. We observe a previously observed lifetime hysteresis effect in $^3$He cell relaxation lifetime and include new measurements of the angular dependence of the relaxation lifetime, that are characteristic of a dipolar effect which diminishes upon repeated pumping/cooling cycles. We present demonstrations of magnetization effects in $^3$He and $^{129}$Xe cells and show the applicability of using sol-gel coatings on reducing the magnetization effect in $^{129}$Xe cells.

We report recent results of the stability of the polarization in new SEOP cells, which was evident for 2 states, one with high energy spin state, low $T_1$ orientation and the second with low energy spin state, high $T_1$ orientation. These results are suggestive of an abnormal relaxation and have only been observed for cells with short ($T_1 < 10$ Hours) lifetimes.

1. Introduction

For spin-exchange optical pumping (SEOP) [1] the maximum achievable polarization is limited by a balancing of the optical pumping rate and relaxation rate of the noble gas, as well as the individual “X-factor” [2] for a $^3$He cell. Hence it is important to minimise the noble gas relaxation for applications requiring high (75%+) polarization.

One such application is that of neutron spin filters [3, 4, 5], where it is essential to achieve high $^3$He polarization and maintain this for a long period of time. This is to achieve the highest attainable neutron polarization/analysing power and limit time dependent corrections for the changing neutron polarization. Wall relaxation can play a significant part in maintaining the gas polarization. Many extensive investigations into wall relaxation rates of $^3$He cells have produced key breakthroughs allowing vast improvements in the lifetimes ($T_1$) of cells [6, 7, 8, 9]. However, there is still a great deal of variation in the achieved relaxation rates of cells and many often exhibit magnetization and magnetic field orientation dependence effects [10]. The aim of this proceedings is to add to the existing observations of these effects.
In the case of continuously pumped $^3$He neutron spin filters [11, 12, 13], provided that the optical pumping rate is much greater than the relaxation rate ($\Gamma_{^3\text{He}} = 1/T_1$) of the cell, the cell should reach and maintain a steady-state polarization. However, we report on recent measurements of the $^3$He polarization stability of continuously pumped cells with high ($T_1 < 10$ hours) levels of relaxation.

2. $^3$He Neutron spin filter cells
SEOP $^3$He cells are generally produced from reblown GE180 glass tubing, an alumosilicate glass that has been shown to yield lifetimes of 100’s of hours that are dominated by dipole-dipole interactions only [9, 14]. Furthermore, the lack of Boron and other such materials with large neutron cross sections means that there is only a small amount of absorption and scattering from the neutron transmission through the cell itself and this is largely independent of energy. A typical neutron spin filter cell (figure 1) is cylindrical in shape, which provides a homogeneous area and known path length for the neutrons to pass through.

In order to make SEOP cells that achieve high gas polarization and have long gas relaxation times it is important to reduce the wall relaxation in the cell by having as few contaminants as possible in the system. To achieve this the cell production rig is based around a turbo molecular pumped vacuum system, obtaining a base vacuum of approx $1 \times 10^{-7}$ mbars. Furthermore the $^3$He and $^{129}$Xe, N$_2$ and alkali metals used to fill the cell are of very high purity. Full descriptions of the cell production procedure employed for the cells used in this work can be found here [15, 16, 17]. Upon production the cells are initially polarized in laboratory based SEOP systems and their room temperature lifetime ($T_1$) measured using Free Induction Decay (FID) NMR [18].

3. Observations of orientation dependence in neutron spin filter cells
The following section contains results showing the so called “orientation dependence” that has been observed previously in $^3$He SEOP cells used for medical imaging studies [10], as well as $^3$He neutron spin filters [16, 19, 20]. The effect is exhibited as a difference obtained for the $T_1$ values, dependent upon the orientation of the cell within the magnetic holding field. In table 1 the low ($T_{1A}$) and high ($T_{1B}$) values for $T_1$ are measured before and after a 180° rotation of the cell within the magnetic field, either by physical rotation of the cell, or by changing the polarity of the magnetic field. The difference in between the values in the two orientations varies from cell to cell and the effect is seemingly independent of the alkali metal element used and has been seen in cells coated with Rb, K and hybrid Rb-K mixtures [16, 21].

For the cells studied in this work the following behaviour was observed, firstly a new cell (i.e. no previous exposure to the magnetic field or laser) is polarized as normal and then a $T_1$, which we refer to as $T_{1A}$ is measured. The cell is then reversed by 180° and a second relaxation time,
Table 1. High and low orientation $T_1$’s for a selection of cells produced at ISIS and ORNL.

| Cell name  | Fill location | Alkali metal | $^3$He Pressure (bar) | $T_{1B}$ (High) | $T_{1A}$ (Low) |
|------------|---------------|--------------|-----------------------|-----------------|----------------|
| Llewellyn  | ISIS          | Rb           | 0.71                  | 124             | 65             |
| Sammy      | ISIS          | Rb           | 0.65                  | 96              | 56             |
| Bob        | ISIS          | Rb           | 0.65                  | 110             | 20             |
| Cooper     | ISIS          | Rb           | 2.32                  | 236             | 129            |
| Billy      | ISIS          | Rb           | 2.08                  | 119             | 50             |
| Dorothy    | ISIS          | Rb           | 0.8                   | 463             | 443            |
| Buttercup  | ISIS          | K            | 1.5                   | 94              | 58             |
| Blue Devil | ORNL          | RB-K         | 1.33                  | 151             | 36             |
| Chang      | ORNL          | RB-K         | 2.01                  | 230             | 150            |
| Messi      | ORNL          | RB-K         | 1.61                  | 151             | 67             |
| Smokey     | ORNL          | RB-K         | 1.98                  | 85              | 75             |
| Xavi       | ORNL          | RB-K         | 0.84                  | 90              | 60             |

$T_{1B}$ is measured. In this case it was always observed that $T_{1B} > T_{1A}$ which is shown clearly in figure 2.

3.1. $T_1$ hysteresis

Cells will usually show a further $T_1$ hysteresis effect, whereby the high/low orientation $T_1$’s will begin to decrease/increase and converge upon a value between the original two $T_1$’s. We investigated the cell Rb14, called Bob, further as with an initial high and low $T_1$ of 110 and 20 hours respectively this cell showed a large orientation dependence.

As illustrated in figure 3 the cell was firstly polarized and the $T_1$ measured in the low orientation ($T_{1A-A}$), then repolarized in the low orientation, turned through $180^\circ$ and measured in the high orientation ($T_{1A-B}$). It is then repolarized and measured in the high orientation ($T_{1B-B}$) and then finally it is repolarized in the high orientation, turned through $180^\circ$ and measured back in the low orientation ($T_{1B-A}$). The results show that as the cell is cycled through the orientations the $T_1$’s begin to converge, in this case to a value at roughly the mean of the relaxation rates.
3.2. Spherical cell investigation

The orientation and $T_1$ hysteresis behaviour can be seen in greater detail from an investigation of the effect on $T_1$ of a spherical cell, 5cm $\sigma$ containing 2 bar $^3$He, measured through a 360$^\circ$ cycle. The spherical cell was used to remove any geometrical effects, either through any holding field inhomogeneities or the resultant magnetic dipolar field that would arise from polarized $^3$He in a cylinder.

The cell was initially polarized in the low $T_1$ orientation, then the $T_1$ measured for approximately 50 hours before turning the cell through 45$^\circ$ and measuring the new $T_1$. It is then repeatedly turned through 45$^\circ$ and each subsequent new $T_1$ measured, the cell was not repolarized between orientations. Once a complete set (i.e. 360$^\circ$) of measurements had been obtained the cell was then repolarized in the high orientation and the measurements repeated.

Plots of the $^3$He relaxation rate as a function of angle can be seen in figure 4. In both plots the $0^\circ$ position refers to the low $T_1$ orientation. From figure 4(a) it is clear that the maximum lifetime is achieved when the cell is orientated at 180$^\circ$ from the initial polarizing position, regardless of which orientation the cell was pumped in. Furthermore, the difference between the high and low orientations decreased after the cell was repolarized. In this case the longest $T_1$ stayed at approximately the same value, whilst the lower values increased.

A polar plot of the data (figure 4(b)) reveals the behaviour to be like that of a dipole. It is clear that the high and low orientations are the dominant directions, with the $T_1$ scaling between these values as the cell is rotated. The suggestion of a hysteresis effect can also be seen clearly in the polar plot, as the cell is rotated through the magnetic field and then repumped. The effect appears to decrease as the $T_1$ for each orientation improves.

3.3. Cell magnetisation

The presence of magnetic impurities within the cell walls is further hinted at by the magnetisation effects seen by many groups [10, 22]. It was found that exposure to a high magnetic field (>1000 G) could cause a vast reduction in the $T_1$ of a cell, although this could often be recovered by degaussing the cell.

To investigate the effect further we took the cell Fred, which no longer showed any orientation behaviour and placed it between the poles of a 1500 G electromagnet. This caused a dramatic reduction in the $T_1$ of the cell from 40 to <1 hours, making it difficult to achieve any polarization within the cell. However, by using a degaussing procedure we were able to partially recover the $T_1$ of the cell, although with a reintroduction of an orientation dependence to 13 and 25 hours. To degauss the cell it was placed in the electromagnet and the polarity of the field was periodically reversed whilst the field strength was gradually decreased.

A further demonstration of the magnetisation effect occurred when a series of $^{129}$Xe cells were fabricated for MRI studies in Pyrex. They were polarized using an external cavity diode laser described in reference [23]. Polarimetry at high field was performed on a 1.5 T GE Signa HDx scanner using a home built birdcage coil of similar dimensions and construction to that
used previously for $^3$He [24]. Using this volume coil allows for uniform excitation of all the spins in the cell, care was taken to measure and minimise the flip angle and correct the magnetisation to account for losses in magnetisation from the RF to get an accurate determination of $T_1$. Low field NMR was performed using our home built NMR system [18].

At low field ($\approx 20$ G) a $T_1 \approx 50$ minutes [25] was found for sealed and valved uncoated cells. For bare Pyrex cells in the 1.5 T field the $^{129}$Xe $T_1$ measured was $\approx 2$ minutes. Further attempts to repolarise this cell resulted in negligible polarization and only a low voltage FID was observed from the cells. This is consistent with earlier investigations [20], which showed that magnetisation of alkali coated cells occurs after exposure to high magnetic field. To remove this problem a series of Pyrex cells were coated with a commercially available sol gel coating (similar to that used in [26, 27]). The coated cells were measured at 1.5 T and $^{129}$Xe $T_1$ of $156 \pm 10$ minutes and $183 \pm 10$ minutes were recorded for two orientations $180^\circ$ apart. In this case the coating appeared to work as a solution to the strong magnetisation effect. However, once this had been removed as a dominant relaxation effect, they appear to now exhibit orientation dependance.

4. Polarization decay during spin-up process

A further effect, observed at the Oak Ridge National Laboratory, occurred during the spin-up process for several new cells with a short ($< 10$ hours) lifetime. The cells have exhibited an unusual effect during $^3$He polarization build-up, whereby the polarization appears to reach saturation, but then begins to slowly decay. Figure 5 shows an example of this effect, measured in laboratory conditions using FID NMR.

Similar effects have been seen previously at the Los Alamos Neutron Science Center and the ILL [28, 29], however these cases occurred during in-situ pumping experiments in the presence of high neutron flux. The investigations revealed two effects: firstly, an immediate increase in alkali metal relaxation as the neutron flux was introduced; and a slow decrease in the optical transmission of the cell due to the build up of a white film on its inside walls (likely an Rb compound formed due to neutron interactions in the cell).

For the results shown in figure 5 a hybrid K/Rb spin filter cell containing 1.9 bar of $^3$He was polarized initially in the high energy (alkali metal optically pumped to ($J=1/2, m_J=1/2$))
Figure 5. Slow decay effect seen during $^3$He polarization build up. The cell was initially polarised in the high energy state.

state at $200^\circ\text{C}$ and the decay was observed. A $T_1$ was then measured, giving 3.6 hours. The cell orientation was then rotated by 180° and the laser helicity reversed, thereby polarizing the cell in the low energy (alkali metal optically pumped to $(J=1/2, m_J=-1/2)$) state. The $T_1$ for this second orientation was then measured, yielding 5.1 hours. The plots in figure 5 have been normalised to account for a change in amplitude observed between each spin-up state. This was likely to have been caused by movement of the NMR coil during the cell reversal and means that no reliable comparison of the amplitudes of the two spin-ups can be made. The normalised plots reveal that in this case the effect is more prevalent in the initial polarization state, i.e. high energy and low $T_1$ orientation. Fits to the early, i.e. pre-saturation section of each spin-up curve reveal a similar rate ($\approx 0.4$ hours$^{-1}$). However, fits to the post-saturation sections reveal a decay rate of 0.038 hours$^{-1}$ for the high energy, low $T_1$ state and 0.006 hours$^{-1}$ for the low energy, high $T_1$ state.

There are a number of potential causes for this observed effect. Before speculating over lifetime effects from the $^3$He itself, it is important to remove any potential instrumentation or measurement effects. For example drifts in the cell temperature or laser wavelength/power are likely to cause changes in the alkali metal polarization or density, which could lead to a change in the overall $^3$He polarization over time. More extensive investigations into the spin-up process, such as measuring laser absorption or the alkali metal polarization, relaxation or density [29] over time, are required to remove this as a possibility.

Another potential cause is radiation damping [30], an effect that can occur during the FID process. After the initial pulse that “tips” the $^3$He into the transverse plane, the precessing spins can induce a current in the NMR coil, creating a “counter” field that causes the magnetisation to rotate back into the longitudinal field direction. This will cause a change in the measured FID transverse relaxation time ($T_2^*$) that will scale with the cell polarization. The effect will also be different for each polarization state; in the low energy state $T_2^*$ will decrease, whereas for the high energy state $T_2^*$ will increase, sometimes showing a non-exponential form that will be unreliable for FID polarimetry [31]. Further analysis of the individual FID’s was not possible for the data shown in figure 5, although investigations of $T_2^*$ over time for each state would reveal any radiation damping effects. Furthermore in this case the position of the NMR coil relative to the cell was changed between measurements, which would also alter any radiation damping effects. Ultimately though, absolute polarization measurements, such as neutron transmission [5] or EPR, are required to reveal whether the observed decrease is a real decrease in $^3$He polarization, or an artifact of the NMR measurements.
5. Discussion

The orientation effect is clearly affecting the maximum achievable $T_1$ in $^3$He cells. The observations in this work show a history based effect, where high and low $T_1$ orientations are set in on the initial spin-up orientation, followed by a gradual decrease in the effect with subsequent spin-ups. Furthermore, as indicated by the spherical cell investigation the effect is dipolar and will change as a function of angle.

The existence of strong magnetisation and hysteresis effects suggests that there are ferromagnetic impurities present within the cell. GE quotes low impurity levels of 0.03% for $\text{Fe}_2\text{O}_3$, however the magnetisation effect has also been seen in cells produced from “iron free” glass [32]. Further investigations from Schmiedeskamp et al. identified dust like magnetite ($\text{Fe}_3\text{O}_4$) particles located close to, or at the inner glass surface as a potential source for magnetisation. It is unknown how these impurities may have entered the glass, although it is likely to be during the cell manufacturing process.

However, the existence of magnetic impurities alone does not fully explain the orientation effect, as the magnetic field strength used for SEOP is usually too low (in our case 1mT) for significant magnetization to occur. The overall mechanisms behind the orientation effect are clearly complex. Jacob et al. [19] speculated that orientation dependence could be dependent on interactions of the alkali metal with impurities within the cell (either from the cell walls, or introduced by the alkali itself). In this work, the addition of a sol gel coating appeared to remove, or reduce the strong magnetisation effect for pyrex $^{129}\text{Xe}$ cells, although it left the cells with an orientation dependence. This appears to agree with the predictions of Jacob et al., as the introduction of a coating will alter the interactions of the polarized gas with the cell walls, but not the alkali metal. However, although not widely reported, the orientation effect has been seen in uncoated cells [33], suggesting that this may not always be the case. It is worth noting however that an uncoated cell will usually be attached to a source of polarized gas using a valve, and that magnetic effects have in some cases been traced to valves [20].

The polarization stability results shown in section 4 demonstrate an undesirable effect that may occur when using continuously pumped in-situ $^3$He polarization devices. Although further investigations are required, if the effect is still present after instrumentation and measurement based factors have been discounted or removed then the effect could potentially be another demonstration of unusual lifetime behaviour in $^3$He cells.

Although the orientation effect clearly has a contribution to the $^3$He relaxation rates within the cell, it is still possible to produce cells where the $T_1$ is hundreds of hours, allowing high (>75%) $^3$He polarizations, thereby creating cells suitable for use in neutron experiments. However, care needs to be taken to not magnetise cells, which could affect the proximity of the cell to high magnetic field sample environment equipment, or even the field strength of the main holding field. The magnetisation effect has large implications in MRI and NMR applications due to the proximity of the high magnetic fields required. In all cases it is clear that either cell demagnetisation is needed, or that the processes or materials that contribute to the orientation dependence, or magnetization of, $^3$He cells must be isolated and removed from the cell making process.

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