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Study of spin-exchange optically pumped ^{3}He cells with high polarisation and long lifetimes

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ABSTRACT

We present a detailed investigation into ^{3}He neutron spin filter cells polarised by spin exchange optical pumping (SEOP). We include measurements of the absolute ^{3}He polarisation using neutron transmission and characterisation of both the *X*-factor and ^{3}He relaxation times (T_1) for a number of cells. For one cell we calculated a maximum ^{3}He polarisation of 79% with a T_1 of 633 h. The measured *X*-factor of this cell, $X = 0.17 \pm 0.01$, is low. For all cells polarisations of $>71\%$ are observed. In addition we present ^{3}He relaxation data for a new design of magneto-static cavity with a field of high homogeneity $\Delta B/B_0 \approx 3.5 \times 10^{-4}\,\text{cm}^{-1}$. This compact device provides a magnetic field in an orientation suitable for in situ optical pumping that minimises the field inhomogeneity contribution to the T_1 to 930 h in a 1 bar cell, the longest reported on beam thus far. The results suggest that high ^{3}He polarisation with long relaxation times can now be routinely obtained with SEOP, enabling time independent incident beam polarisation to be easily implemented across many different neutron scattering instruments.

1. Introduction

Nuclear spin-polarised ^{3}He has uses in a diverse range of applications, including neutron spin filters (NSFs) [1], spin-polarised targets [2] and magnetic resonance imaging (MRI) [3].

For NSFs the preferential absorption of one neutron spin state makes ^{3}He an ideal candidate for broadband neutron polarisers [4] and analysers covering large solid angle [5,6]. For lung imaging the low proton density and short T_2^* (≈ 1 ms) makes traditional proton MRI problematic [7]. However, the use of inhaled spin-polarised gas as a contrast agent provides significant information on lung structure and function. Direct measurements of physical properties such as gas flow, diffusion and ventilation can be routinely obtained [8]. For both of these applications higher polarisations produce higher sensitivity for the same volume of gas polarised. In NSFs this increased ^{3}He polarisation translates to a higher neutron polarisation and transmission. For MRI increased ^{3}He polarisation allows increased signal to noise (SNR) in the images and hence lower volumes of gas are needed with the associated economic benefit of reduced gas consumption.

In this paper we use spin-exchange optical pumping (SEOP) [9] to polarise the ^{3}He, where Rb vapour is optically pumped to high polarisation and the ^{3}He polarised by spin-exchange via binary collisions with the Rb. Significant efforts are being employed to understand and increase the maximum ^{3}He polarisation obtainable using this method [10,11].

For SEOP the maximum polarisation obtainable is limited by a recently recognised form of relaxation [10], which has similar magnitude and dependence on alkali metal density as the spin-exchange process and varies for different cells. In the absence of this new form of relaxation, the steady state gas polarisation is given by

$$P_{\text{He}} = P_{\text{Rb}} \frac{k_{se}[\text{Rb}]}{k_{se}[\text{Rb}] + \Gamma_r} \quad (1)$$

where P_{He} and P_{Rb} are the ^{3}He and rubidium polarisations, respectively, [Rb] is the rubidium vapour density, Γ_r is the ^{3}He relaxation rate measured at $[\text{Rb}] = 0$ (in this case at room temperature) and k_{se} the spin exchange rate coefficient. However,

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recent work [10] has shown that there is a temperature dependent relaxation that modifies the total ^{3}He relaxation rate to $\Gamma_{\text{He}} = k_{se}[\text{Rb}](1+X)+\Gamma_r$, where X is a phenomenological parameter that represents the additional relaxation rate. With this additional relaxation term the total ^{3}He polarisation is given by

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

In the limit $P_{\text{Rb}} = 1$ [13] (which is achievable with high power narrowed lasers) and $\Gamma_r \ll k_{se}[\text{Rb}]$ the maximum ^{3}He polarisation is predicted to be $P_{\text{He}} = 1/(1+X)$, therefore in order to increase the maximum obtainable ^{3}He polarisation it is necessary to minimise this limiting quantity X, which differs from cell to cell. Typically cells have X factors of 0.25–0.45 [10]. In this work, we report on a detailed investigation of several cells, including details of the construction, measurements of the polarisation lifetime T_1 (where $T_1 = 1/\Gamma_r$), helium polarisation and X-factor along with a discussion of the limiting factors.

2. Cell preparation and characterisation

Three cells were investigated, namely J1, J7 and Lion, the cell geometry and fill pressures are summarised in Table 1. For the lifetime measurements the cells were polarised using either an external cavity diode laser (ECDL) [12] with either a 40 W or 100 W diode (n-Light, USA) as the source or a 30 W (Coherent FAP) laser with pulse-acquire NMR used to record T_1. For the X-factor measurements the cells were polarised using an ECDL with Faraday rotation diagnostics similar to those detailed in Ref. [13] used to measure [Rb] and pulse-acquire NMR used to record the spin up time constant.

2.1. Cell construction

The cell J1 is a cylinder of length 5 cm with curved hemispherical edges and a diameter of 5 cm, the cells Lion and J7 are cylinders both 5 cm diameter, 5 cm length, filled at NIST and Jülich, respectively. All cells were constructed from reblown GE180 glass and filled using procedures similar to those described by Rich et al. [14].

We will focus on the cell J1, which was constructed in the Jülich glass shop and filled at ISIS. The cell was baked for 4 days at 450 °C after which the vacuum measured at the base of the pump was 6.8×10^{-7} mbar. After distillation of the Rb with a hot air gun the cell was filled while immersed in liquid nitrogen first with nitrogen (Air products, 99.9% purity) to 36 mbar and then filled with ^{3}He (Spectra gases, 99.999% purity) to a total pressure of 308 mbar and sealed off. Assuming the ideal gas law we calculate the total ^{3}He pressure to be 1.05 bar at room temperature (296 K). Time of flight neutron transmission measurements [4] on the unpolarised cell showed the pressure to be 0.93 bar, this disagreement is not unexpected as the cell is sealed off with a flame above the liquid nitrogen.

Table 1
Cell geometry and filling pressures at room temperature.

Cell name	Length (cm)	Diameter (cm)	S/V (cm^{-1})	Fill pressure	
				N_2 (mbar)	^{3}He (bar)
J1	5.0	5.0	1.17	136	1.05
Lion	4.5	5.0	1.29	200	0.57
J7	5.0	5.0	1.20	100	1.04

2.2. Characterisation

Using a pulse-acquire NMR system similar to that described in Ref. [15] a T_1 of 663 ± 7 h was measured (Fig. 1). The NMR amplitude in volts is obtained via fitting of a short (10 ms) region of the FID decay with an exponential decay and the corresponding initial voltage recorded. Detailed information on the lineshape of the FID is not available and currently we are investigating additional effects which may arise if radiation damping is occurring in the NMR measurements as this may distort the measured spin-up and T_1 time constants [16]. This additional complication to the measurement can result in an effective flip angle which is dependent upon the total magnetisation (M_0). However, for these measurements we have a low-Q ($Q\approx10$) coil and cells of low pressures (see Table 1). We have checked this with simultaneous measurement of the polarisation by NMR and neutrons for a $Q=12$ coil. In this situation we find a linear relationship between the initial voltage and the polarisation measured by neutrons, which validates the approach used in this work.

The orientational dependence of the T_1 of the cell was also investigated. We observed a few percent difference in T_1 when measuring with different cell orientations in the magnetic field, indicating that this cell has a low level of paramagnetic impurities [17]. Moreover this represents the highest T_1 cell fabricated at ISIS although not at the maximum obtainable as determined in the limit by the dipole–dipole interaction ($T_1^{dipol} = 807\,(\text{bar h})/p\,(\text{bar})$). The magnetic field used for this measurement was a Helmholtz pair, diameter ≈ 0.9 m of sufficient uniformity to exclude contributions to relaxation from magnetic inhomogeneities and therefore we attribute the difference from the dipole–dipole limit to wall relaxation [18].

The X-factor for the cells were measured using the hot relaxation method [10]. Here Γ_{He} (the denominator in Eq. (2)) is determined as a function of [Rb] by measurement of the spin up time constant τ_{up}, where $\Gamma_{\text{He}} = 1/\tau_{up}$. As the spin-exchange rate coefficient k_{se} [19] is known then X can be determined from the slope of Γ_{He} vs [Rb] (Fig. 2).

The measured X-factor of 0.17 for cell J1 represents thus far one of the lowest value reported for a SEOP cell and adds to the evidence that it is possible to minimise this excess relaxation in certain cells. From these measurements the predicted ^{3}He polarisation is high, predicted $P_{\text{He}} = 1/(1+X) = 0.85$. Therefore in order to accurately determine the polarisation we used neutron transmission to provide an absolute measurement and the unambiguous determination of the pressure length product of the cell. We performed similar measurements on the other two

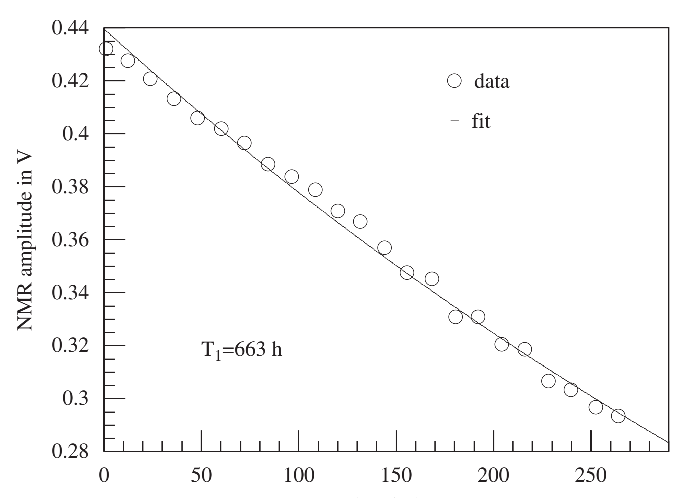

Fig. 1. NMR measurement of ^{3}He polarisation lifetime (T_1) of cell J1.