Measurement of laser heating in spin exchange optical pumping by NMR diffusion sensitisation

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Introduction : We detail in-situ measurement of the temperature/pressure of alkali metal spin-exchange optical pumping (SEOP) cells containing ^{3}He [1] using NMR techniques. Typical spin exchange optical pumping setups consist of a cell containing the noble gas to be polarised, alkali metal vapour (typically rubidium) and a buffer gas (nitrogen in the case of ^{3}He and nitrogen-helium mixture in the case of ^{129}Xe). In this process circularly polarised photons at the D_1 alkali metal valence electron transition are absorbed and then de-excited non-radiatively via collisions with a nitrogen buffer gas. These non radiative transitions deposit energy from the photons into heat. This heat can cause changes in the alkali metal density ([Rb]), which in turn can affect the polarisation processes, having a direct bearing on the polarisation and production rate achieved. The aim of the work was to use NMR to develop tools to probe and further understand the physics during laser optical pumping by monitoring of the noble gas temperature. For SEOP of ^{3}He and ^{129}Xe, current efforts are focused on achieving both high polarisation and volume production rate [2-4]. For ^{129}Xe-Rb the faster spin relaxation rate [1] than that for ^{3}He-Rb results in a higher absorption of the pumping light. Nitrogen heating was first identified [5] by Raman scattering measurements of the nitrogen showing that elevated temperatures of several hundred degrees above the oven temperature can occur in cells. Modelling using finite element methods has predicted the existence of convection currents driven by temperature gradients across the cell. These simulations also predict a resultant 'gradient' or spatial variation in the ^{129}Xe nuclear polarisation across the pumping cell [6], due to increased wall contact. This heating gives rise to an increase in Rb density which in turn increases the absorption creating a runaway effect [7]. In optical pumping, low field NMR is usually used to monitor of the build up of polarisation. We use additional diffusion weighted MR pulse sequences to measure the gas diffusion constant (D) which allows determination of the gas pressure/temperature and imaging methods to demonstrate spatial distribution of polarisation in the cell.

Materials and Methods :. The cells were polarised with SEOP using an external cavity diode laser (ECDL) [3] using a 100 W diode. This was narrowed with emission (FWHM ~ 0.1 nm) using a holographic grating. A home built NMR spectrometer was adapted to output the gradient waveforms in addition to the RF excitation pulse. A linear amplifier and used to drive a homebuilt pair of 15 cm radius gradient coils (10 turns of 0.7 mm diameter coaxial with B_0). Two types of gradient waveform were used, a gradient recalled echo and diffusion weighted bipolar pair. The gradient echo waveforms were of variable length with a ramp time of 0.5 ms, typical durations of 8 ms. A volume RF coil was wound to act as a combined transmitter/receiver - two rectangular coils, placed either side of the cell with the B_1 axis orthogonal to B_0. The cell pressure was also measured using neutron transmission [8].

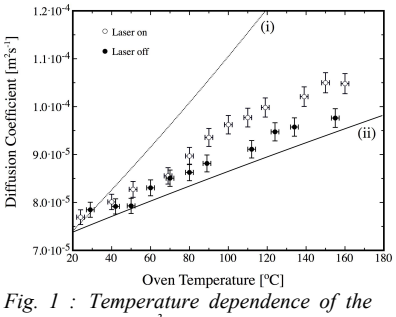

Fig. 1 : Temperature dependence of the measured D for ^{3}He measured in situ in the oven with the laser off (closed points) and on (open points). Lines - calculations from Chapman-Enskog theory [9], (i) taking into account only the explicit temperature dependence and (ii) the temperature and cell pressure dependence. Calculation assumes cell fill pressures of ^{3}He 2.16 Bar and N_2 135 Bar at room temperature.

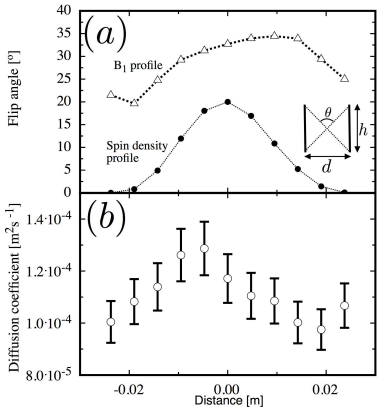

Fig. 2 : (a) Flip angle profile (open triangles) of the spherical cell with cell profile from the gradient echo (closed circles). (b) Spatially localised measurement of diffusion coefficient of cell at 160° C.

Results : Diffusion measurements were made in a ^{3}He optical pumping cell as a function of oven temperature, the results are shown in figure 1. A clear increase in the diffusion coefficient was observed as a function of oven temperature with the laser off (open circles). In order to better quantify this effect, D was calculated for this cell based on the Chapman-Enskog theory [9]. Shown in figure 1(line (i)) is the expected behaviour of D considering only temperature dependence. Good agreement is found at room temperature if the nitrogen pressure is assumed to be 135mBar, a small deviation from the fill pressure measured at 150±2 mBar. This deviation is attributed to differences in gas mixing during filling. The pressure at room temperature is known from the neutron transmission measurements [8], therefore under constant volume assuming an ideal gas law we can rewrite the current pressure ($p_2=T_1/T_2p_1$) and substitute for p_2. This results in the line (ii), which aside from a small constant offset fits well with the experimental gas diffusion data. The effect of laser heating was then investigated. At low oven temperature (20-70° C, [Rb]=0) the measured D shows no change when the laser is on or off. At higher temperatures where [Rb] > 0, the increased laser absorption results in an increase in D, which we attribute to heating of the gas. This small change in D corresponds to ~30º±15ºC higher temperature in the cell than that prescribed by the oven thermocouple. In order to further assess the spatial sensitivity of the setup to diffusion we performed a B_1 map of the spherical cell shown in figure 2(a). The map shows a slight asymmetry which we attribute to the non-optimised nature of the Helmholtz coil transmit field. Finally the pulse sequence was further modified to combine a gradient echo with diffusion sensitisation, to map the spatial profile of D, shown in figure 2(b). A higher D is found in the cell centre suggesting that there is a distribution of temperatures within the cell.

Conclusions : A means of measuring cell temperature and laser heating with NMR is demonstrated using a simple 1-D gradient imaging system. These methods may be used to better understand laser heating in Rb-Xe SEOP and, for example also be applied to study flow dynamics in a flow cell using phase contrast with bipolar gradients. Studies with ^{129}Xe where laser heating is a problem are more challenging due to differences in NMR sensitivity (lower γ_{Xe}) and lower gas concentration/pressure, however with signal averaging, the use of enriched ^{129}Xe and higher sensitivity coils this should be feasible. In addition, the low diffusion coefficient of typical Xe mixtures 2Bar (3%^{129}Xe, 10%N_2, 87%^{4}He) D~8x10^{-6}m^{2}s^{-1} and lower γ will require stronger gradients.

References :1.Rev. Mod. Phys. 69, 629 (1997), 2.Phys. Rev. Lett. 88, 123003 (2003), 3.Phys. Rev. Lett. 96, 053002 (2006), 4.Phys. Rev. A 75, 013416 (2007), 5.Phys. Rev. Lett. 86, 3264 (2001), 6.Phys. Rev. A 72, 053411 (2005), 7.J. Magn. Reson. 159, 175 (2002), 8.Physica B, 297, (2007) 179, 9. E.L.Cussler, 'Diffusion: Mass Transfer in Fluid Systems, p108 Cambridge University Press (1984)

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