Laser absorption and photon efficiency of a spin exchange optical pumping 129Xe polariser

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Target audience: Hyperpolarised gas MR; ^{129}Xe SEOP polarisation.

Purpose: Hyperpolarised ^{129}Xe gas can be inhaled to non-invasively study lung structure and function with MRI [1]. The technique used to polarise the nuclei of ^{129}Xe gas atoms is spin exchange optical pumping (SEOP), whereby Rb valence electrons are optically pumped with circularly polarised laser light resonant on the D_1 transition line (794.77 nm in air), resulting in a highly polarised electron spin Zeeman ground state. Within a SEOP cell, collisions between ^{129}Xe and Rb atoms transfer spin polarisation from the Rb electrons to the ^{129}Xe nuclei. The photon absorption rate within the SEOP cell and the spin-transfer efficiency (photon efficiency) can be used to determine the production rate of hyperpolarised of ^{129}Xe and are thus important parameters in the performance of SEOP polarisers. In this study, these two parameters were evaluated for a SEOP polariser operating at a mid-range cell pressure of 2 bar with gas under flow.

Methods: SEOP polariser components (see Fig. 1): cylindrical Pyrex cell (25 cm length, 5 cm diameter) filled with < 1 gram of molten rubidium, 1.25 amg of He, 0.14 amg of N_2 and 0.04 amg of Xe; a Helmholtz B_0 coil (diameter 80 cm, B_0 ~ 2.7 mT); and a 25 W external cavity diode laser (emission profile with FWHM ~ 0.1 nm). The photon absorption rate, $\Delta\phi$, was calculated for a range of gas flow rates by measuring the transmitted laser power at the back of SEOP cell when the cell was cold, P_{cold}, (~ 293 K) and hot, P_{hot} (~ 373 K) and using $\Delta\phi(\lambda,T) = (P_{\text{cold}} - P_{\text{hot}})/E_p$, where E_p is the energy of a single photon carried in the laser beam with an emission centre wavelength of 794.77 nm. Theoretical and experimental photon efficiencies, η_{theory} and η_{exp}, were calculated using Eqs. (1) and (2) below [2]:

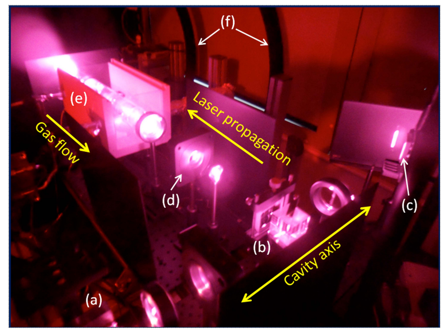

Figure 1: SEOP apparatus. (a) laser diode bar; (b) beam-splitter cube (1/3 feedback along cavity axis, 2/3 transmission along cell axis); (c) holographic grating; (d) λ/4-wave plate; (e) ceramic oven for housing cell (lid removed); (f) B_0 coils

$$\eta_{\text{theory}} = \frac{\kappa_{\text{SE}}^{\text{BC}} + \kappa_{\text{SE}}^{\text{vdW}}}{(\kappa_{\text{SD}}^{\text{Rb-Xe}} + \kappa_{\text{SD}}^{\text{vdW}}) + f(\kappa_{\text{SE}}^{\text{BC}} + \kappa_{\text{SE}}^{\text{vdW}})} \quad (1); \quad \eta_{\text{exp}} = \frac{[\text{Xe}]V\, P_{\text{Xe}}/\tau_{\text{emp}}}{\Delta\phi} \quad (2).$$

Eq 1: $\kappa_{\text{SE}}^{\text{BC}}$ and $\kappa_{\text{SD}}^{\text{BC}}$ are the spin exchange and destruction cross sections for two-body Rb-Xe binary collisions, which have been previously calculated as 2.17×10^{-16} cm^{3}s^{-1} [3] and 1.09×10^{-14} cm^{3}s^{-1} [2]; $\kappa_{\text{SE}}^{\text{vdW}}$ and $\kappa_{\text{SD}}^{\text{vdW}}$ are the cross sections for short-lived three-body Rb-Xe vdW complexes (with either N_2 or He acting as the third body), which have been calculated as 3.46×10^{-16} cm^{3}s^{-1} and 1.79×10^{-16} cm^{3}s^{-1}, respectively [2]. f is the enrichment factor of (0.86 ^{129}Xe). Eq 2: [Xe] is the number density of Xe atoms in the cell volume, V, P_{Xe} is the ^{129}Xe polarisation measured for a gas cell residency time equal to an empirically determined spin exchange time, τ_{emp}. The spin exchange time, τ_{emp}, was extrapolated from the relationship between ^{129}Xe polarisation, P_{Xe}, and gas flow, Q, through the optical pumping cell, which was fit to $P_{\text{Xe}}(t) \propto (1-e^{-(\gamma+\Gamma)t})$. $t = Q/V$ is the gas residency time in the cell, Γ is the Xe-Rb spin relaxation rate, which was estimated to be $1/T_1$, where T_1 is the ^{129}Xe relaxation time in the cell (calculated to be 48 minutes) and $\gamma = 1/\tau_{\text{emp}} = 0.022$ Hz is the spin exchange rate, which was determined using a least-squares fitting routine (Levenberg-Marquardt method) [2].

Results and discussion: The number of photons being absorbed in the cell decreased linearly with increasing gas flow rate (Fig. 2). To our knowledge, such an observation has not previously been reported and may be attributed to an increase in cell cooling at higher flow rates causing a lower Rb vapour density, thus resulting in a decrease in photon absorption. The theoretical photon efficiency, η_{theory}, was calculated (Eq. (1)) to be 0.049, which is higher than the previously reported efficiency of 0.043 [3]. The difference in efficiencies may be attributed to the inclusion of vdW interactions in the efficiency calculation in this study; previously, only binary collisions were considered. Furthermore, whereas the spin transfer efficiency was previously considered to be temperature-independent [3], both the binary and vdW spin destruction cross sections calculated in this study depend on cell temperature [2]. Using Eq. (2), an experimental photon efficiency was calculated for a gas mixture flowing through the cell at a rate of 650 sccm (Fig. 2), the flow rate for which $t_{\text{res}} = \tau_{\text{emp}} = 1/\gamma = 45$ seconds. The polarisation at this flow rate was measured as 8.6 % and substituted into Eq. (2). Using a calculated photon absorption rate, $\Delta\phi$, of 2.36×10^{19} photons s^{-1}, a ^{129}Xe enrichment fraction, f, of 0.86 and a cell volume of 491 cm^3, gives an experimental photon efficiency of $\eta_{\text{exp}} = 0.046$, which is in reasonable agreement with the theoretical value, $\eta_{\text{theory}} = 0.049$. Experimentally and theoretically, therefore, $1/\eta_{\text{exp}} = 22$ and $1/\eta_{\text{theory}} = 20$ photons are required per ^{129}Xe spin flip.

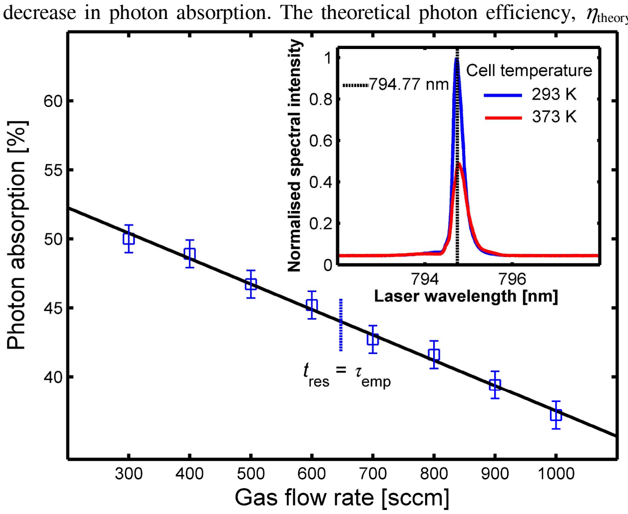

Figure 2: Inverse linear dependence of photon absorption with gas flow rate. The black line is a linear fit to experimentally measured photon absorption percentage using $(P_{\text{cold}}-P_{\text{hot}})/P_{\text{cold}}$ and the vertical blue line indicates the absorption for which the cell residency time, t_{res} is equal to the spin exchange time, τ_{emp}. Inset shows absorption spectrum (red) for gas flowing at 300 sccm through a cell at T = 373 K.

Conclusions: It has been reported here for the first time a gas flow rate dependent photon absorption in a SEOP cell. This observation has implications on the choice of SEOP polariser running parameters, namely gas flow rate and laser power, for the production of large volumes of polarised ^{129}Xe gas. With 20–22 photons required to induce each ^{129}Xe spin flip and a photon absorption rate of 2.36×10^{19} photons s^{-1}, this indicates that it is possible on this system to produce 120–130 cm^3 of 100% polarised ^{129}Xe per hour at STP.

References:

[1] Mugler JP, et al. Simultaneous magnetic resonance imaging of ventilation distribution and gas uptake in the human lung using hyperpolarized xenon-129. Proc. Natl. Acad. Sci. U. S. A. Dec 14 2010;107(50):21707-21712. [2] Norquay G, et al. Optimized Production of Hyperpolarized 129Xe at 2 bar for In Vivo Lung MRI. J Appl Phys. 2012; (in press). [3] Jau YY, et al. High-field measurement of the Xe-129-Rb spin-exchange rate due to binary collisions. Phys Rev A. Nov 2002;66(5). [4] Bhaskar ND, et al. Efficiency of Spin Exchange Between Rubidium Spins And Xe-129 Nuclei in a Gas. Phys Rev Lett. 1982;49(1):25-28.