Some Additional Thoughts on the Hybrid Paper

Jaideep Singh
University of Virginia
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1   Introduction

The standard analysis of the spin-exchange optical pumping (SEOP) polarization rate equations yield the following equilibrium polarizations:

$$P_{\mathtt{He}} = \left[\frac{\gamma_{\mathtt{se}}}{\gamma_{\mathtt{se}} + \Gamma_{\mathtt{He}}} \right] P_{\mathtt{A}} \quad \& \quad P_{\mathtt{A}} = \frac{A_{\mathtt{op}}}{A_{\mathtt{op}} + \Gamma_{\mathtt{A}}} \tag{1}$$

The optical pumping rate is a convolution of the incident photon flux $\Phi(\nu)$ with the alkali absorption cross section $\sigma(\nu)$:

$$A_{\mathtt{op}} = \int_0^\infty \Phi(\nu)\sigma(\nu)\ d\nu \tag{2}$$

The spin-exchange rate (to the ^{3}He nuclei) is the product of the spin-exchange rate constant $k_{\mathtt{se}}$ and the alkali density [A]:

$$\gamma_{\mathtt{se}} = k_{\mathtt{se}}[\mathtt{A}] \tag{3}$$

Finally, the spin relaxation rates are traditionally given by the following sums:

$$\Gamma_{\mathtt{A}} = k_{\mathtt{sd}}^{\mathtt{A}}[\mathtt{A}] + k_{\mathtt{sd}}^{\mathtt{He}}[\mathtt{He}] + k_{\mathtt{sd}}^{\mathtt{N_2}}[\mathtt{N_2}] \tag{4}$$

$$\Gamma_{\mathtt{He}} = \Gamma_{\mathtt{wall}} + k_{\mathtt{dip}}[\mathtt{He}] \tag{5}$$

where $k_{\mathtt{sd}}^{\mathtt{i}}$ is the spin destruction rate constant between the alkali atom and some particle of type $\mathtt{i}$, $\Gamma_{\mathtt{wall}}$ is the wall relaxation rate, and $k_{\mathtt{dip}}$ is the ^{3}He nuclear dipolar self relaxation rate constant.

According to these equations, if one could provide enough laser power to keep a very high density of alkali metal polarized, then the ^{3}He polarization could be, in principle, made to equilibrate at unity:

$$P_{\mathtt{He}} = \lim_{[\mathtt{A}],\Phi(\nu)\to\infty} \left(\left[\frac{\gamma_{\mathtt{se}}}{\gamma_{\mathtt{se}} + \Gamma_{\mathtt{He}}} \right] P_{\mathtt{A}} \right) = \left[\lim_{\gamma_{\mathtt{se}}\to\infty} \frac{\gamma_{\mathtt{se}}}{\gamma_{\mathtt{se}} + \Gamma_{\mathtt{He}}} \right] \left[\lim_{A_{\mathtt{op}}\to\infty} \frac{A_{\mathtt{op}}}{A_{\mathtt{op}} + \Gamma_{\mathtt{A}}} \right] = 1 \tag{6}$$

In practice (with SEOP), the ^{3}He polarization has never been recorded higher than 0.85 and has rarely been made to exceed 0.80. The situation is even worse for SLAC/JLab style cells. With broadband lasers and pure Rb SEOP, polarizations have never exceeded 0.50. The situation improves using hybrid SEOP, but the polarization has yet to exceed 0.60.

Walker et al argue that the maximum possible ^{3}He polarization is about 0.85 because of a previously unaccounted for ^{3}He spin relaxation mechanism. This so-called 'X'-factor is either dependant on the alkali density or has an alkali vapor pressure-like temperature dependance. *What we do not know is why the target cells do not achieve even this level of performance.* More than likely, it is a combination of less than unity alkali polarization and Walker's 'X'-factor; but, as of yet, *no one has ever systematically measured either of these parameters for target cells.*

2 General Equations for Hybrid Alkali Polarization

Before going further, it's important to relax some of the assumptions made to derive the previous equations. In addition we must account for the modifications due to hybrid SEOP.

1. We'll introduce a parameter labeled by Λ: it is the probability that an optically excited alkali atom returns to the opposite ground state from which it started. Traditionally it is assumed that collisional mixing in the excited state results in $\Lambda = 1/2$.

2. We'll include the effects of pumping both the D1 and D2 transitions of Rb and K.

3. We'll allow the light to propagate at a skew angle to the magnetic field and also the possibility that it is not fully circularly polarized.

4. We'll explicitly acknowledge that the alkali polarization reaches equilibrium very fast relative to its diffusion time within the pumping chamber. Therefore the alkali polarization is position dependent with the largest gradient from the front of the cell to the back.

5. We'll allow for the possibility of a spin destroying process that is induced by the laser light. It is most natural to express these terms (X_R, X_K) as a fraction of the total optical pumping rate.

Putting this altogether, we get for the equilibrium alkali polarizations:

$$P_R = \frac{R_1 - R_2/2 + \eta_A \mathcal{D}\left[K_1 - K_2/2\right]}{R_0\left(1 + X_R\right) + \Gamma_R + \eta_A \mathcal{D}\left[\Gamma_K + K_0\left(1 + X_K\right)\right]} \tag{7}$$

$$P_K = \eta_A \left(P_R + \frac{K_1 - K_2/2}{A_{se}[Rb]}\right) \tag{8}$$

where R_0 & K_0 are the total optical pumping rates for Rb and K, $R_{1(2)}$ & $K_{1(2)}$ are the differential D1 (D2) optical pumping rates for Rb and K, η_A is the alkali spin-exchange efficiency for K, $\mathcal{D}$ is K to Rb density ratio, and Γ_R & Γ_K are the spin relaxation rates for Rb and K. The explicit forms of these terms are given as:

$$R_0, K_0 = 2\Lambda \int_0^\infty \left[\Phi_\mathcal{R}(\nu, \vec{r}) + \Phi_\mathcal{L}(\nu, \vec{r})\right]\left[\sigma_1^{R,K}(\nu) + \sigma_2^{R,K}(\nu)\right]\, d\nu \tag{9}$$

$$R_1, K_1 = 2\Lambda \int_0^\infty \left[\Phi_\mathcal{R}(\nu, \vec{r}) - \Phi_\mathcal{L}(\nu, \vec{r})\right]\cos(\theta)\sigma_1^{R,K}(\nu)\, d\nu \tag{10}$$

$$R_2, K_2 = 2\Lambda \int_0^\infty \left[\Phi_\mathcal{R}(\nu, \vec{r}) - \Phi_\mathcal{L}(\nu, \vec{r})\right]\cos(\theta)\sigma_2^{R,K}(\nu)\, d\nu \tag{11}$$

$$\eta_A = \frac{A_{se}[Rb]}{A_{se}[Rb] + \Gamma_K + K_0} \tag{12}$$

$$\mathcal{D} = \frac{[K]}{[Rb]} \tag{13}$$

where $\Phi_{\mathcal{R}(\mathcal{L})}$ is the photon flux for right (left) circularly polarized light, θ is the skew angle, $\sigma_{1(2)}^{R(K)}$ is the unpolarized absorption cross section for D1 (D2) transitions in Rb (K). The following equations account for attenuation of the photon flux as the beam propagates through the cell:

$$\frac{d\Phi_\mathcal{R}}{dz} = -\left[\left(1 - P_R\cos(\theta)\right)\sigma_1^R + \left(1 + \frac{P_R}{2}\cos(\theta)\right)\sigma_2^R\right][Rb]\Phi_\mathcal{R}$$
$$\qquad -\left[\left(1 - P_K\cos(\theta)\right)\sigma_1^K + \left(1 + \frac{P_K}{2}\cos(\theta)\right)\sigma_2^K\right][K]\Phi_\mathcal{R} \tag{14}$$

$$\frac{d\Phi_\mathcal{L}}{dz} = -\left[\left(1 + P_R\cos(\theta)\right)\sigma_1^R + \left(1 - \frac{P_R}{2}\cos(\theta)\right)\sigma_2^R\right][Rb]\Phi_\mathcal{L}$$
$$\qquad -\left[\left(1 + P_K\cos(\theta)\right)\sigma_1^K + \left(1 - \frac{P_K}{2}\cos(\theta)\right)\sigma_2^K\right][K]\Phi_\mathcal{L} \tag{15}$$

3 General Equations for Helium Polarization in a Target Cell

Now we are in a position to discuss the He polarization.

6. Because the He polarization reaches equilibrium very slow relative to its diffusion time within the pumping chamber, it is essentially uniform in the pumping chamber and only sensitive to the pumping chamber volume average of the alkali polarization.

7. We'll assume a relatively fast diffusion time between the pumping chamber and target chamber. This implies that the diffusion rate d_{tc} is usually much larger than all other rates in the He polarization process.

8. We'll allow for the possibility of a spin destroying process that is proportional to the alkali density. It is most natural to express this term X as a fraction of the total spin exchange rate.

$$P_{pc}^\infty = \frac{f_{pc}\left(\gamma_{se}^R \langle P_R \rangle_V + \gamma_{se}^K \langle P_K \rangle_V\right)}{f_{pc}\left(\gamma_{se}^R + \gamma_{se}^K\right)(1+X) + f_{pc}\Gamma_{pc} + f_{tc}\Gamma_{tc}\left[1+\Gamma_{tc}/d_{tc}\right]^{-1}} \tag{16}$$

$$P_{tc}^\infty = P_{pc}^\infty \left[1 + \Gamma_{tc}/d_{tc}\right]^{-1} \tag{17}$$

where the subscript pc(tc) refers to the pumping (target) chamber, d_{tc} is the probability per unit time that a He nucleus exits the target chamber and enters the pumping chamber, the brackets $\langle \cdots \rangle_V$ refer to a pumping chamber volume average, and $f_{pc(tc)}$ is the fraction of He nuclei in pumping (target) chamber. The time dependance of the He polarization is given as:

$$P_{pc}(t) = P_{pc}^\infty + \left[P_{pc}^0 - P_{pc}^\infty - c_{pc}\right]\exp\left(-\Gamma_s t\right) + c_{pc}\exp\left(-\Gamma_f t\right) \tag{18}$$

$$P_{tc}(t) = P_{tc}^\infty + \left[P_{tc}^0 - P_{tc}^\infty - c_{tc}\right]\exp\left(-\Gamma_s t\right) + c_{tc}\exp\left(-\Gamma_f t\right) \tag{19}$$

where $P_{pc,tc}^0$ are set by the initial conditions, $\Gamma_{s(f)}$ is the slow (fast) time constant, and the coefficients $c_{pc,tc}$ are given as:

$$c_{pc} = \frac{\Gamma_s\left(P_{pc}^\infty - P_{pc}^0\right) - (f_{tc}/f_{pc})\,d_{tc}P_{tc}^0 + \left[(\gamma_{se}^R + \gamma_{se}^K)(1+X) + \Gamma_{pc} + (f_{tc}/f_{pc})\,d_{tc}\right]P_{pc}^0 - \gamma_{se}^R\langle P_R\rangle_V - \gamma_{se}^K\langle P_K\rangle_V}{\Gamma_f - \Gamma_s} \tag{20}$$

$$c_{tc} = \frac{\Gamma_s\left(P_{tc}^\infty - P_{tc}^0\right) + (d_{tc} + \Gamma_{tc})\,P_{tc}^0 - d_{tc}P_{pc}^0}{\Gamma_f - \Gamma_s} \tag{21}$$

$$\Gamma_f = d_{tc}/f_{pc} + \left(\gamma_{se}^R + \gamma_{se}^K\right)(1+X) + \Gamma_{pc} + \Gamma_{tc} - \Gamma_s \tag{22}$$

$$\Gamma_s = f_{pc}\left(\gamma_{se}^R + \gamma_{se}^K\right)(1+X) + f_{pc}\Gamma_{pc} + f_{tc}\Gamma_{tc} - \delta\Gamma \tag{23}$$

Using the fact that the diffusion rate d_{tc} is usually much larger than all other rates in the He polarization process, we can reduce the above equations to the following:

$$P_{pc}^\infty = P_{tc}^\infty = \frac{f_{pc}\left(\gamma_{se}^R\langle P_R\rangle_V + \gamma_{se}^K\langle P_K\rangle_V\right)}{f_{pc}\left(\gamma_{se}^R + \gamma_{se}^K\right)(1+X) + f_{pc}\Gamma_{pc} + f_{tc}\Gamma_{tc}} \tag{24}$$

$$\Gamma_f = d_{tc}/f_{pc} \tag{25}$$

$$\Gamma_s = f_{pc}\left(\gamma_{se}^R + \gamma_{se}^K\right)(1+X) + f_{pc}\Gamma_{pc} + f_{tc}\Gamma_{tc} \tag{26}$$

$$P_{pc}(t) = P_{pc}^\infty\left[1 - \exp\left(-\Gamma_s t\right)\right] + \left[P_{pc}^0 f_{pc} + P_{tc}^0 f_{tc}\right]\exp\left(-\Gamma_s t\right) + f_{pc}\left[P_{tc}^0 - P_{pc}^0\right]\exp\left(-\Gamma_f t\right) \tag{27}$$

$$P_{tc}(t) = P_{tc}^\infty\left[1 - \exp\left(-\Gamma_s t\right)\right] + \left[P_{pc}^0 f_{pc} + P_{tc}^0 f_{tc}\right]\exp\left(-\Gamma_s t\right) + f_{tc}\left[P_{pc}^0 - P_{tc}^0\right]\exp\left(-\Gamma_f t\right) \tag{28}$$

During a spinup, the polarization starts from zero and there are two distinct time domains defined by the diffusion time scale:

$$t \ll 1/\Gamma_f \quad \left\{\begin{array}{c} P_{pc} = (\gamma_{se}^R\langle P_R\rangle_V + \gamma_{se}^K\langle P_K\rangle_V)\,t \\ P_{tc} = (\gamma_{se}^R\langle P_R\rangle_V + \gamma_{se}^K\langle P_K\rangle_V)\,d_{tc}t^2/2 \end{array}\right\} \tag{29}$$

$$1/\Gamma_f \ll t \ll 1/\Gamma_s \quad P_{pc}(t) = P_{tc}(t) = f_{pc}\left(\gamma_{se}^R\langle P_R\rangle_V + \gamma_{se}^K\langle P_K\rangle_V\right)t \tag{30}$$

$$t \gg 1/\Gamma_f \quad P_{pc}(t) = P_{tc}(t) = P_{pc}^\infty\left[1 - \exp\left(-\Gamma_s t\right)\right] \tag{31}$$

4 Interesting Plots

1. Alkali Polarization and Alkali Density vs. Laser Power: We would use 1, 2, 3, 4, or 5 lasers to "vary" the laser power or we could use a set of ND filters. We would do this at low temperature (low alkali density) and high temperature (high alkali denisty, essentially operating conditions). At low alkali density, we would have enough laser light to explore whether there is an X_R factor. This would be for one cell and it would be interesting to do the low alkali density test with broadband & narrow lasers.

2. Alkali Polarization and He polarization vs. Skew Angle: We would compare the center beamline with the offset beamline at a few different angles.

3. Alkali Polarization and He polarization vs. Beam Spot Size: We would vary the location of the second lens which more or less controls the size of the beam.

4. Alkali Polarization scanned across the cell: This would provide some information about the sensivity of the alkali polarization to the beam size.

5. Maximum achieved-Alkali Polarization and He polarization vs. Density Ratio: We would measure this for as many cells as possible.

6. Alkali Polarization and He polarization vs. Alkali Density: We would use this to compare the performance of the line narrowed with the broadband laser.

7. Density Ratio vs. Potassium Density: We would do this for one cell and include both pressure broadening data and faraday rotation data.

8. Density Ratio in the alkali mix vs. Density Ratio from pressure broadening measurements: We would measure this for as many cells a possible.

9. 'X' factor vs. S/V ratio: We would only have three different S/V ratios (because of the three different pumping chamber sizes for the target cells). In the Walker "Limits" paper, they see trends in there X data: (a) X seems correlated with S/V ratio (b) the lower limit of X is about 0.10. Our data would further explore the lower left region of the "triangle."

5 Measuring X

One can measure X with or without knowledge of the alkali density. In a double chambered cell, the measurement becomes tricker because one is typically measuring "weighted average" rates between the two chambers. The weights are fraction of atoms in each chamber and are temperature dependant:

$$f_{\mathrm{pc}} = \frac{v}{t+v} \tag{32}$$

$$f_{\mathrm{tc}} = \frac{t}{t+v} \tag{33}$$

$$t = \frac{T_{\mathrm{pc}}}{T_{\mathrm{tc}}} \tag{34}$$

$$v = \frac{V_{\mathrm{pc}}}{V_{\mathrm{tc}}} \tag{35}$$

where $T_{\mathrm{pc(tc)}}$ is the pumping (target) chamber temperature in Kelvin and $V_{\mathrm{pc(tc)}}$ is the pumping (target) chamber volume. There are three methods that require measurements at different alkali densities:

1. One can measure the ratio of the equilbrium He polarization to the alkali polarization as a function of alkali density. At sufficiently high alkali density, the ratio will become constant. The resulting consant will be equal to:

$$\lim_{[\mathrm{K}]\to\infty}\left[\frac{P_{\mathrm{pc}}^{\infty}}{\langle P_{\mathrm{K}}\rangle_{\ell}}\right] = \frac{\langle P_{\mathrm{A}}\rangle_{V}/\langle P_{\mathrm{A}}\rangle_{\ell}}{1+X} \tag{36}$$

where we've assumed that $P_{\mathrm{K}} = P_{\mathrm{R}}$ and allowed for the possibilty that the volume averaged alkali polarization is different than the line averaged alkali polarization that the farrot probe beam is sensitive to. The alkali density would be measured using polarization farrot.

2. One can do a "hot" spindown with the pump lasers off. In this scenario, you would have to wait for the cell to return to thermal equilibrium and for the "fast" diffusion exponential to decay away. Under those conditions, the polarization would decay with a time constant of Γ_{s}. The slope of the time constant vs. the alkali density would be given as:

$$\frac{\partial \Gamma_{\mathrm{s}}}{\partial [\mathrm{K}]} = f_{\mathrm{pc}} k_{\mathrm{se}}^{\mathrm{K}} \left(1 + \frac{k_{\mathrm{se}}^{\mathrm{R}}}{k_{\mathrm{se}}^{\mathrm{K}}} \left[\frac{B_{\mathrm{R}} - T_{\mathrm{pc}}}{B_{\mathrm{K}} - T_{\mathrm{pc}}} \right] \mathcal{D}^{-1} \right) (1 + X) \approx f_{\mathrm{pc}} k_{\mathrm{se}}^{\mathrm{K}} \left(1 + \frac{1.4}{\mathcal{D}} \right) (1 + X) \tag{37}$$

where $B_{\mathrm{R(K)}}$ is the pure vapor pressure temperature coefficient for Rb (K) and T_{pc} is the pumping chamber temperature. This technique requires knowledge of the cell temperatures and the alkali density measured using field farrot.

3. One can also do a spinup. This scenario is nearly identical to the previous one, except now the lasers are on. Therefore the alkali density would be measured using polarization farrot. The uncertainties in the cell temperature would be larger because of the ΔT effect due to the lasers. Once again, you would have to wait for the cell to return to thermal equilibrium after turning the lasers on and for the "fast" diffusion exponential to decay away. Under those conditions, the polarization would buildup with the same time constant Γ_{s} and the same equation would result for the slope of Γ_{s} vs. [K].

There are two methods that do not require a measurement of the alkali density, but do require a measuring a spinup and a cold spindown:

$$\frac{\Gamma_{\mathrm{s}} - \Gamma_{\mathtt{room}}}{f_{\mathrm{pc}} \left(\gamma_{\mathtt{se}}^{\mathrm{R}} + \gamma_{\mathtt{se}}^{\mathrm{K}} \right)} = 1 + X + \left[\frac{\Gamma_{\mathrm{pc}} + \Gamma_{\mathrm{tc}} \left(\frac{t}{v} \right) - \Gamma_{\mathtt{room}}/f_{\mathrm{pc}}}{\gamma_{\mathtt{se}}^{\mathrm{R}} + \gamma_{\mathtt{se}}^{\mathrm{K}}} \right] \tag{38}$$

The bracketed term on the right is very small when the spin exchange rate is very fast and can be neglecting when the spinup is done near operating temperatures. The trick is measuring the denominator of the left side of the previous equation:

1. One could use the appropriate combination of the spinup time constant and the equilibrium polarizations:

$$\frac{\Gamma_{\mathrm{s}} - \Gamma_{\mathtt{room}}}{P_{\mathrm{pc}}^{\infty} \Gamma_{\mathrm{s}} / \langle P_{\mathtt{A}} \rangle_{\ell}} = \left(\frac{\langle P_{\mathtt{A}} \rangle_{\ell}}{\langle P_{\mathtt{A}} \rangle_{V}} \right) (1 + X) \tag{39}$$

2. One could also track the polarization buildup while it is in the linear regime after the fast exponential has decayed away:

$$\frac{\Gamma_{\mathrm{s}} - \Gamma_{\mathtt{room}}}{\left[\frac{dP_{\mathrm{pc}}}{dt} \right] (1/\Gamma_{\mathrm{f}} \ll t \ll 1/\Gamma_{\mathrm{s}}) / \langle P_{\mathtt{A}} \rangle_{\ell}} = \left(\frac{\langle P_{\mathtt{A}} \rangle_{\ell}}{\langle P_{\mathtt{A}} \rangle_{V}} \right) (1 + X) \tag{40}$$

There last two methods are sensitive to ratio of the alkali polarization averaged over the volume of the cell to the alkali polarization average over the probe beam path length. It is possible, depending on the shape of the beam, to have a high probe beam path length average and a low volume averaged polarization. This is one reason why it would be nice scan the probe beam across the cell perpendicular to the magnetic field. We could do a crude mapping of the alkali polarization to insure that the probe beam path length average was representative of the volume average. Another way to study this problem is to do alkali polarization EPR spectroscopy using an amplitude modulated set of EPR coils. This would only require the same hardware as the He EPR polarization measurement. In this case however, the EPR coils are driven by an AM signal with a fixed EPR frequency. One would then record the D2 fluorescence signal from the EPR Lock-In as the magnetic field is swept. The "standard" analysis of the ratio of the areas under the peaks would result in the alkali polarization. *The benefit of this technique is that it provides an independant measurement of the alkali polarization over a different sample volume of the cell, without the need of a farrot apparatus.*