# Some notes on Alkali-$^3$He SEOP

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## 1 Cross Sections and Rates

A particle passes through a finite target region filled with target points. When the particle encounters a target point, an interaction takes place. The probability that a single interaction occurs $p_1$ is dependant on the target point density [N], the length of the target region $l$, and some factor $\sigma$ that is dependant solely on the nature of interaction:

$$p_1 = \sigma[\mathrm{N}]l \quad \rightarrow \quad \sigma = \frac{p_1}{[\mathrm{N}]l} = \frac{p_1}{\phi} \tag{1}$$

The factor $\sigma$ is called the interaction cross section and has units of area. It is the probability of a single interaction per unit target point density per unit target region length. A cross section is useful because it isolates the part of the interaction probability that is due to the mechanism of the interaction. An equivalent definition of cross section is the probability of interaction per unit flux, where flux $\phi$ is the number of target points per unit cross sectional area.

The interaction rate $\gamma$ is defined as the probability of an single interaction per unit time:

$$\gamma = \frac{p_1}{t} = \frac{\sigma[\mathrm{N}]l}{t} = \sigma v[\mathrm{N}] \tag{2}$$

where $v$ is the relative velocity between particle and the target points. Alternatively, this can rewritten using the flux per unit time $\Phi \equiv \frac{\phi}{t}$:

$$\gamma = \frac{p_1}{t} = \frac{\sigma\phi}{t} = \Phi\sigma \tag{3}$$

Both forms of the interaction rate will be used.

## 2 Alkali Polarization

The *local* equilibrium alkali polarization is calculated by:

$$P_A^\infty(\vec{r}) = P_{\mathrm{light}}(\vec{r})\frac{\gamma_{\mathrm{op}}(\vec{r})}{\gamma_{\mathrm{op}}(\vec{r}) + \gamma(\vec{r})} \tag{4}$$

where $\gamma_{\mathrm{op}}$ is the optical pumping rate, $\gamma$ is the total alkali metal relaxation rate. Note that we can essentially ignore the motion of the alkali atom during the optical pumping cycle because the alkali polarization reaches equilibrium on a time scales much faster than that determined by it's thermal velocity. The alkali polarization can be thought of as spatially "frozen." [Phys. Rev. A 58, 2282-2294 (1998)]

The optical pumping rate for monochromatic light with a frequency of $\nu$ is:

$$\gamma_{\mathrm{op}}(\vec{r}) = \Phi(\vec{r})\sigma(\nu) \tag{5}$$

where $\Phi$ is the number of photons per unit cross sectional area per unit time and $\sigma$ is the absorption cross section. When both the photon flux and the absorption cross section have a frequency dependence, the optical pumping rate is generalized to:

$$\gamma_{\rm op}(\vec{r}) = \int_0^\infty \Phi(\nu,\vec{r})\sigma(\nu)\mathrm{d}\nu \tag{6}$$

$$\Phi(\nu,\vec{r}) = \frac{\text{number of photons at position }\vec{\mathrm{r}}}{\text{unit cross sectional area} \quad\times\quad \text{unit time} \quad\times\quad \text{unit frequency interval}} \tag{7}$$

$$\sigma(\nu) = \text{photon absorption cross section at }\nu \tag{8}$$

It is useful to express $\gamma$ in terms of $\gamma'_{\rm se}$, the spin exchange rate from the noble gas to the alkali metal. These quantities are related through the spin exchange efficiency $\eta$ which is simply the ratio of the spin exchange rate with the noble gas to the total rate of alkali relaxation [Phys. Rev. Lett. 80, 2801-2804 (1998)] and [Phys. Rev. Lett. 91, 123003 (2003)]:

$$\eta = \frac{\gamma'_{\rm se}}{\gamma} \quad\rightarrow\quad \gamma = \frac{\gamma'_{\rm se}}{\eta} \tag{9}$$

Note that this $\gamma'_{\rm se}$ is not to be confused with $\gamma_{\rm se}$ which is the spin exchange rate from the alkali metal to the noble gas. These are calculated in the following way:

$$\gamma'_{\rm se} = \langle\sigma v\rangle_{\rm se}\,[\mathrm{N}] \tag{10}$$

$$\gamma_{\rm se} = \langle\sigma v\rangle_{\rm se}\,[\mathrm{A}] \tag{11}$$

where $\langle\sigma v\rangle_{\rm se}$ is the thermal velocity averaged spin exchange cross section (also labeled as $k_{\rm se}$) and [N] & [A] are the noble gas and alkali metal densities.

# 3 $^3$He Polarization

## 3.1 Particle Rate Equations

Neglecting the transfer tube, a cell is composed of a pumping chamber and a target chamber. The $^3$He is polarized only in the pumping chamber by contact with polarized alkali atoms in vapor form. The number of $\pm$ helium nuclei in the pumping and target chambers, labeled by subscript, is $N^{\pm}_{p,t}$. The total number of helium nuclei in the pumping and target chamber, labeled by subscript, is $N_{p,t} = N^+_{p,t} + N^-_{p,t}$. The total number of helium nuclei is $N = N_p + N_t$. The fraction of nuclei in either chamber, labeled by subscript, is $f_{p,t} = N_{p,t}/N$.

Assuming that the alkali polarization reaches equilibrium on time scales much faster than the $^3$He polarization build-up, then the change in the number of $\pm$ nuclei in either chamber is governed by the following rate equations:

$$\frac{dN^+_p}{dt} = \langle\sigma v\rangle_{\rm se}[A^+]N^-_p - \langle\sigma v\rangle_{\rm se}[A^-]N^+_p + \left(\frac{N_p}{2} - N^+_p\right)\Gamma_p + N^+_t D_t - N^+_p D_p \tag{12}$$

$$\frac{dN^-_p}{dt} = \langle\sigma v\rangle_{\rm se}[A^-]N^+_p - \langle\sigma v\rangle_{\rm se}[A^+]N^-_p + \left(\frac{N_p}{2} - N^-_p\right)\Gamma_p + N^-_t D_t - N^-_p D_p \tag{13}$$

$$\frac{dN^+_t}{dt} = N^+_p D_p - N^+_t D_t + \left(\frac{N_t}{2} - N^+_t\right)\Gamma_t \tag{14}$$

$$\frac{dN^-_t}{dt} = N^-_p D_p - N^-_t D_t + \left(\frac{N_t}{2} - N^-_t\right)\Gamma_t \tag{15}$$

The relaxation rates $\Gamma_{p,t}$ represent interactions which show no preference for either state and therefore push equilibirum towards equal amounts of $\pm$ nuclei. The diffusion rate $D_{p(t)}$ is the probability per unit time per nucleus that a nucleus will exit the pumping (target) chamber and enter the target (pumping) chamber.

The rate of change of the total number of particles in the two chambers is given by:

$$\frac{dN_p}{dt} = \frac{dN_p^+}{dt} + \frac{dN_p^-}{dt} = N_t D_t - N_p D_p \tag{16}$$

$$\frac{dN_t}{dt} = \frac{dN_t^+}{dt} + \frac{dN_t^-}{dt} = N_p D_p - N_t D_t \tag{17}$$

$$\frac{dN}{dt} = \frac{dN_p}{dt} + \frac{dN_t}{dt} = 0 \tag{18}$$

At particle number equilibrium, this implies:

$$N_t D_t = N_p D_p \qquad \rightarrow \qquad f_t D_t = f_p D_p \tag{19}$$

## 3.2 Polarization Rate Equations

Polarization is defined as:

$$P_{p,t} = \frac{N_{p,t}^+ - N_{p,t}^-}{N_{p,t}^+ + N_{p,t}^-} = \frac{N_{p,t}^+ - N_{p,t}^-}{N_{p,t}} = f_{p,t}^+ - f_{p,t}^- \tag{20}$$

$$N_{p,t}^{\pm} = \frac{1}{2}\left(1 \pm P_{p,t}\right) \tag{21}$$

Combining the particle rate equations in the the appropriate way and, to reiterate, assuming that the alkali polarization reaches equilibrium very early in the $^3$He polarization build-up, then the polarizations of nuclei in the two chambers of the cell are governed by:

$$\frac{dP_p}{dt} = \gamma_{\rm se}\left(P_A - P_p\right) - \Gamma_p P_p - D_p\left(P_p - P_t\right) = aP_p + bP_t + B \tag{22}$$

$$\frac{dP_t}{dt} = D_t\left(P_p - P_t\right) - \Gamma_t P_t = cP_p + dP_t \tag{23}$$

where the following substitutions are made:

$$a = -\left(\gamma_{\rm se} + \Gamma_p + D_p\right) \tag{24}$$

$$b = D_p \tag{25}$$

$$c = D_t \tag{26}$$

$$d = -\left(\Gamma_t + D_t\right) \tag{27}$$

$$B = \gamma_{\rm se} P_A \tag{28}$$

Note that we are essentially averaging over the motion of the $^3$He atom during the spin exchange cycle because the $^3$He polarization reaches equilibrium on a time scale much slower than that determined by it's thermal velocity. The $^3$He atoms essentially sample all parts of pumping chamber during the spin-exchange process and is therefore sensitive to the volume averaged alkali polarization:

$$P_A \equiv \int_{\rm pc} P_A^{\infty}(\vec{r})\, \mathrm{d}^3 r \tag{29}$$

The equilibrium ($t \rightarrow \infty$) polarizations are found by setting the rate equations to zero:

$$P_p^{\infty} = \frac{B}{bc - ad} \tag{30}$$

$$P_t^{\infty} = -\left(\frac{c}{d}\right) P_p^{\infty} \tag{31}$$

The above can be written in a more illuminating form by noting that $f_p D_p = f_t D_t$ and by some algebra:

$$P_p^{\infty} = P_A \left[\frac{\gamma_{\rm se} f_p}{\gamma_{\rm se} f_p + \Gamma_p f_p + \Gamma_t f_t \left(1 + \frac{\Gamma_t}{D_t}\right)^{-1}}\right] \tag{32}$$

$$P_t^{\infty} = P_p^{\infty}\left[1 + \frac{\Gamma_t}{D_t}\right]^{-1} \tag{33}$$

The coupled rate equations can be rewritten as a matrix equation:

$$\mathbf{M} = \begin{bmatrix} a & b \\ c & d \end{bmatrix} \tag{34}$$

$$\vec{P} = \begin{bmatrix} P_p \\ P_t \end{bmatrix} \tag{35}$$

$$\vec{B} = \begin{bmatrix} B \\ 0 \end{bmatrix} \tag{36}$$

$$\frac{d\vec{P}}{dt} = \mathbf{M}\vec{P} + \vec{B} \tag{37}$$

This equation is solved by finding the eigenvalues of the rate matrix $\mathbf{M}$. These eigenvalues are the characteristic rates of the system and are labeled as slow and fast:

$$\Gamma_s = -\frac{1}{2}\left[a + d + \sqrt{(a-d)^2 + 4bc}\right] \tag{38}$$

$$\Gamma_f = -\frac{1}{2}\left[a + d - \sqrt{(a-d)^2 + 4bc}\right] \tag{39}$$

The solutions to the coupled rate equations are given by:

$$P_p(t) = P_p^\infty + \left[P_p^0 - P_p^\infty - c_p\right]\exp\left(-\Gamma_s t\right) + c_p \exp\left(-\Gamma_f t\right) \tag{40}$$

$$P_t(t) = P_t^\infty + \left[P_t^0 - P_t^\infty - c_t\right]\exp\left(-\Gamma_s t\right) + c_t \exp\left(-\Gamma_f t\right) \tag{41}$$

where $P_{p,t}^0$ are set by the initial conditions. Finally, the coefficients $c_{p,t}$ can be obtained by satisfying the coupled rate equations and after some algebra:

$$c_p = \frac{\Gamma_s\left(P_p^\infty - P_p^0\right) - bP_t^0 - aP_p^0 - B}{\Gamma_f - \Gamma_s} \tag{42}$$

$$c_t = \frac{\Gamma_s\left(P_t^\infty - P_t^0\right) - dP_t^0 - cP_p^0}{\Gamma_f - \Gamma_s} \tag{43}$$

## 3.3 Fast Diffusion Limit

In the limit where the diffusion is very fast compared to all other rates, then the characteristic rates of the system become to lowest order:

$$\Gamma_s = \gamma_{\text{se}} f_p + \Gamma_p f_p + \Gamma_t f_t + \mathcal{O}\left(\frac{1}{D_t}\right) = \langle\gamma_{\text{se}}\rangle + \langle\Gamma\rangle \tag{44}$$

$$\Gamma_f = \frac{D_t}{f_p} + \gamma_{\text{se}} f_t + \Gamma_p f_t + \Gamma_t f_p + \mathcal{O}\left(\frac{1}{D_t}\right) \tag{45}$$

where brackets $\langle\rangle$ refer to an average over all atoms. In the limit $D_{p,t} \to \infty$, the equilibrium polarizations and coefficients become:

$$P_p^\infty \to P_A\left[\frac{\gamma_{\text{se}} f_p}{\gamma_{\text{se}} f_p + \Gamma_p f_p + \Gamma_t f_t}\right] = P_A\left[\frac{\langle\gamma_{\text{se}}\rangle}{\langle\gamma_{\text{se}}\rangle + \langle\Gamma\rangle}\right] \tag{46}$$

$$P_t^\infty \to P_p^\infty \tag{47}$$

$$c_p = f_p\left(P_t^0 - P_p^0\right) \tag{48}$$

$$c_t = f_t\left(P_p^0 - P_t^0\right) \tag{49}$$

which gives for the polarizations in the two chambers:

$$P_p(t) = P_p^\infty + \left[P_p^0 f_p + P_t^0 f_t - P_p^\infty\right]\exp\left(-\Gamma_s t\right) + f_p\left[P_t^0 - P_p^0\right]\exp\left(-\Gamma_f t\right) \tag{50}$$

$$P_t(t) = P_p^\infty + \left[P_p^0 f_p + P_t^0 f_t - P_p^\infty\right]\exp\left(-\Gamma_s t\right) + f_t\left[P_p^0 - P_t^0\right]\exp\left(-\Gamma_f t\right) \tag{51}$$

After the fast exponential has decayed away, the polarization in the two chambers evolves identically as if the initial polarization in the two chambers was a volume of average of the true initial polarizations in the two chambers:

$$\begin{aligned}
P(t) &= P_p(t) = P_t(t) && (52)\\
&= P_p^\infty \left[1-\exp\left(-\Gamma_s t\right)\right] + \left[P_p^0 f_p + P_t^0 f_t\right]\exp\left(-\Gamma_s t\right) && (53)\\
&= P^\infty \left[1-\exp\left(-\Gamma_s t\right)\right] + \left\langle P^0\right\rangle \exp\left(-\Gamma_s t\right) && (54)
\end{aligned}$$

### 3.4 Slow Diffusion Limit

In the limit where the diffusion is very slow compared to all other rates in the system, then the characteristic rates of the system become to lowest order:

$$\begin{aligned}
\Gamma_s &\rightarrow \gamma_p = \gamma_{\mathrm{se}} + \Gamma_p + D_p + \mathcal{O}\left(D_t^2\right) && (55)\\
\Gamma_f &\rightarrow \gamma_t = \Gamma_t + D_t + \mathcal{O}\left(D_t^2\right) && (56)
\end{aligned}$$

where we have relabeled the rates with subscripts $p, t$ instead of slow and fast which are no longer relevant. In the limit $D_{p,t} \rightarrow 0$, the equilibrium polarizations and coefficients are to lowest order:

$$\begin{aligned}
P_p^\infty &= P_A \left[\frac{\gamma_{\mathrm{se}}}{\gamma_{\mathrm{se}}+\Gamma_p}\right]\left[1 - \frac{D_p}{\gamma_{\mathrm{se}}+\Gamma_p} + \mathcal{O}\left(D_t^2\right)\right] && (57)\\
P_t^\infty &= P_p^\infty \left[\frac{D_t}{\Gamma_t} + \mathcal{O}\left(D_t^2\right)\right] && (58)\\
c_p &= \frac{P_t^0 D_p}{\gamma_{\mathrm{se}}+\Gamma_p-\Gamma_t} && (59)\\
c_t &= P_t^0 + \left(\frac{D_t}{\Gamma_t}\right)\left[\frac{P_A\gamma_{\mathrm{se}} - P_p^0\Gamma_t}{\gamma_{\mathrm{se}}+\Gamma_p-\Gamma_t}\right] && (60)
\end{aligned}$$

which gives for the polarizations in the two chambers:

$$\begin{aligned}
P_p(t) &= P_p^\infty\left[1-\exp\left(-\gamma_p t\right)\right] + P_p^0 \exp\left(-\gamma_p t\right) + D_p\left[\frac{P_t^0}{\gamma_{\mathrm{se}}+\Gamma_p-\Gamma_t}\right]\left[\exp\left(-\gamma_t t\right) - \exp\left(-\gamma_p t\right)\right] && (61)\\
P_t(t) &= P_t^0 \exp\left(-\gamma_t t\right)\\
&\quad + \left(\frac{D_t}{\Gamma_t}\right)\left(P_p^\infty\left[1-\exp\left(-\gamma_p t\right)\right] + \left[\frac{P_A\gamma_{\mathrm{se}} - P_p^0\Gamma_t}{\gamma_{\mathrm{se}}+\Gamma_p-\Gamma_t}\right]\left[\exp\left(-\gamma_t t\right)-\exp\left(-\gamma_p t\right)\right]\right) && (62)
\end{aligned}$$

# 4 Theoretical Maxiumum Helium-3 Polarization

### 4.1 Introduction

To recap, using the fast diffusion limit for a two chambered cell:

$$\begin{aligned}
P_A^\infty(\vec{r}) &= P_{\mathrm{light}}(\vec{r})\left[\frac{\gamma_{\mathrm{op}}(\vec{r})}{\gamma_{\mathrm{op}}(\vec{r}) + \gamma(\vec{r})}\right] && (63)\\
P_H^\infty &= P_p^\infty = P_t^\infty = P_A\left[\frac{\gamma_{\mathrm{se}} f_p}{\gamma_{\mathrm{se}} f_p + \Gamma}\right] && (64)
\end{aligned}$$

where:

$$\begin{aligned}
P_A &\equiv \int_{\mathrm{pc}} P_A^\infty(\vec{r})\ \mathrm{d}^3 r && (65)\\
\Gamma &\equiv f_p\Gamma_p + f_t\Gamma_t && (66)\\
f_{p,t} &= \frac{\text{number of } ^3\text{He atoms in chamber } p,t}{\text{total number of } ^3\text{He atoms}} && (67)
\end{aligned}$$

Note that the optical pumping rate and spin-exchange rate are dependent on the laser intensity and density of alkali atoms respectively:

$$\gamma_{\text{op}}(\vec{r}) = \int_0^\infty \Phi(\nu,\vec{r})\sigma(\nu)\mathrm{d}\nu \tag{68}$$

$$\gamma_{\text{se}} = \langle\sigma v\rangle_{\text{se}}[A] \tag{69}$$

In the limit of infinite laser intensity and infinite alkali density, the theoretical maximum $^3$He is:

$$\begin{aligned}
P_H^{\max} &= \lim_{\Phi,[A]\to\infty} P_H^\infty && (70)\\
&= \lim_{\Phi,[A]\to\infty}\left(\int_{\text{pc}} P_{\text{light}}(\vec{r})\left[\frac{\gamma_{\text{op}}(\vec{r})}{\gamma_{\text{op}}(\vec{r})+\gamma(\vec{r})}\right]\,\mathrm{d}^3r\left[\frac{\gamma_{\text{se}}f_p}{\gamma_{\text{se}}f_p+\Gamma}\right]\right) && (71)\\
&= \left[\lim_{\gamma_{\text{op}}(\vec{r})\to\infty}\int_{\text{pc}} P_{\text{light}}(\vec{r})\frac{\gamma_{\text{op}}(\vec{r})}{\gamma_{\text{op}}(\vec{r})+\gamma(\vec{r})}\,\mathrm{d}^3r\right]\left[\lim_{\gamma_{\text{se}}\to\infty}\frac{\gamma_{\text{se}}f_p}{\gamma_{\text{se}}f_p+\Gamma}\right] && (72)\\
&= \int_{\text{pc}} P_{\text{light}}(\vec{r})\,\mathrm{d}^3r && (73)
\end{aligned}$$

which is the volume averaged laser light polarization. From this one would expect that with sufficient amounts of laser intensity and alkali density, the $^3$He polarization should saturate at $\approx 1$.

However, from many years of practical experience, anecdotal evidence, and detailed optical pumping simulations, this is simply not the case. Typically it is assumed that there are no relaxation mechanisms that are dependant on the laser intensity $\left(\Phi_0(\vec{r}) = \int \Phi(\nu,\vec{r})\,\mathrm{d}\nu\right)$ and alkali density ($[A]$):

$$\gamma = \langle\sigma v\rangle_{\text{sd}}^{\text{A}-\text{N}_2}[\text{N}_2]_p + \langle\sigma v\rangle_{\text{sd}}^{\text{A}-{}^3\text{He}}[{}^3\text{He}]_p + \langle\sigma v\rangle_{\text{sd}}^{\text{A}-\text{Rb}}[\text{Rb}]_p + \langle\sigma v\rangle_{\text{sd}}^{\text{A}-\text{K}}[\text{K}]_p + \gamma_{\text{wall}-p}(\vec{r}) \tag{74}$$

$$\Gamma_p = \langle\sigma v\rangle_{\text{sd}}^{{}^3\text{He}-\text{N}_2}[\text{N}_2]_p + \langle\sigma v\rangle_{\text{sd}}^{{}^3\text{He}-{}^3\text{He}}[{}^3\text{He}]_p + \Gamma_{\text{wall}-p} \tag{75}$$

$$\Gamma_t = \langle\sigma v\rangle_{\text{sd}}^{{}^3\text{He}-\text{N}_2}[\text{N}_2]_t + \langle\sigma v\rangle_{\text{sd}}^{{}^3\text{He}-{}^3\text{He}}[{}^3\text{He}]_t + \langle\sigma v\rangle_{\text{se}}^{{}^3\text{He}-\text{Rb}}[\text{Rb}]_t + \langle\sigma v\rangle_{\text{se}}^{{}^3\text{He}-\text{K}}[\text{K}]_t + \Gamma_{\text{wall}-t} \tag{76}$$

$$\tfrac{\partial\gamma(\vec{r})}{\partial\Phi_0(\vec{r})} = \tfrac{\partial\Gamma}{\partial[A]} = 0 \tag{77}$$

where $\gamma$ and $\Gamma$ are the total alkali and $^3$He relaxation rates, respectively. Note that there is a very small vapor pressure (at most less than one percent relative to the pumping chamber) of alkali atoms in the target chamber under operating conditions. These alkali atoms are essentially unpolarized because they are not exposed to the laser light and are very unlikely to diffuse into the pumping chamber. Therefore spin-exchange collisions with these alkali atoms are (to a good approximation) *relaxing* for the helium atoms in the target chamber.

Although we do not understand the physical mechanism underlying these extra relaxation mechanisms, we can add phenomenological terms to account for their potential effects. It is most natural to express them as fractions of the optical pumping and spin-exchange rates, since those rates set a useful scale to measure against:

$$X_\alpha \equiv \frac{\partial\gamma(\vec{r})}{\partial\gamma_{\text{op}}(\vec{r})} \tag{78}$$

$$X_\beta \equiv \frac{\partial\Gamma_p}{\partial(\gamma_{\text{se}})} \tag{79}$$

Taking into account these terms gives:

$$P_A^\infty(\vec{r}) = P_{\text{light}}(\vec{r})\left[\frac{\gamma_{\text{op}}(\vec{r})}{\gamma_{\text{op}}(\vec{r})[1+X_\alpha(\vec{r})]+\gamma_0(\vec{r})}\right] \tag{80}$$

$$P_H^\infty = P_p^\infty = P_t^\infty = P_A\left[\frac{\gamma_{\text{se}}f_p}{\gamma_{\text{se}}f_p[1+X_\beta]+\Gamma_0}\right] \tag{81}$$

$$P_H^{\max} = \lim_{\Phi,[A]\to\infty} P_H^\infty = \frac{\int_{\text{pc}} P_{\text{light}}(\vec{r})\,\mathrm{d}^3r}{[1+\langle X_\alpha\rangle][1+X_\beta]} \tag{82}$$

Note that $X_\alpha$ may have a position depedence. In the following sections, we'll discuss what is known about $X_\alpha$ and $X_\beta$.

## 4.2 Helium Laser Interaction

It is hard to imagine that there is a direct coupling between the laser light and the helium atoms. The wavelengths that are typically used are far from any known He optical transitions. Even if there was such an interaction, it could only effect the *nuclear spin polarization* of the helium through the subsequent recoupling of the helium nucleus and helium electrons via the hyperfine interaction. In the ground state of helium, this interaction is zero because the total angular momentum of a closed shell is zero.

## 4.3 Inefficient Optical Pumping

Inefficient optical pumping refers to either (1) optical pumping with unpolarized light or (2) optical pumping unwanted transitions ($D_2$). If we include the possibilty of pumping both the $D_1$ and $D_2$ transitions, then the *local* equilibrium alkali polarization is calculated by:

$$P_A^\infty(\vec{r}) = \frac{P_{\mathrm{D1-light}}(\vec{r})\gamma_{\mathrm{opD1}}(\vec{r}) - \frac{1}{2}P_{\mathrm{D2-light}}(\vec{r})\gamma_{\mathrm{opD2}}(\vec{r})}{\gamma_{\mathrm{opD1}}(\vec{r}) + \gamma_{\mathrm{opD2}}(\vec{r}) + \gamma(\vec{r})} \tag{83}$$

If we pump with monochromatic $D_1$ light and there is no light dependent relaxation, then the maximum alkali polarization is 1.0. On the other hand, the maximum polarization pumping with pure monochromatic $D_2$ light is $-0.5$. Note that given the *same* sense of circular polarizaton, $D_2$ light acts to polarize the alkali into the state *opposite* relative to that of $D_1$ pumping. Skew pumping essentially reduces the light polarization by a factor of $\cos(\Theta) = \hat{k}_{\mathrm{light}} \cdot \hat{B}_{\mathrm{holding}}$ in the reference frame of the alkali atom. It also results in more light absorption and therefore less light penetrates to the back end of the cell [Phys. Rev. A 66, 033406 (2002)]. Assuming that the $D_2$ optical pumping rate is proportional to the $D_1$ optical pumping rate and that the "extra" relaxation proportional to laser intensity comes purely from inefficient optical pumping, then in the limit of infinite laser intensity:

$$\lim_{\Phi\to\infty} P_A^\infty(\vec{r}) = P_{\mathrm{D1-light}}(\vec{r})\left[\frac{\cos(\Theta) - \frac{P_{\mathrm{D2-light}}(\vec{r})}{2P_{\mathrm{D1-light}}(\vec{r})}\frac{\gamma_{\mathrm{opD2}}(\vec{r})}{\gamma_{\mathrm{opD1}}(\vec{r})}}{1 + \frac{\gamma_{\mathrm{opD2}}(\vec{r})}{\gamma_{\mathrm{opD1}}(\vec{r})}}\right] = P_{\mathrm{D1-light}}(\vec{r})\left[1 + X_\alpha\right]^{-1} \tag{84}$$

If we assume that the skew angle $\Theta$ is small, the degree of circular polarization of both $D_1$ and $D_2$ light is the same, and that the $D_2$ pumping rate is small relative to the $D_1$ pumping rate, then $X_\alpha$ due to inefficient optical pumping is:

$$X_\alpha \approx \frac{\Theta^2}{2} + \frac{3}{2}\left[\frac{\gamma_{\mathrm{opD2}}(\vec{r})}{\gamma_{\mathrm{opD1}}(\vec{r})}\right] \tag{85}$$

In practice, it is very difficult to get $\Theta < 1.0^o$. On the other hand, $\Theta \approx 10^o$ is a very conservative upper estimate for the skew angle, the effect on the *local* alkali polarization is:

$$0.00015 < X_\alpha \approx \frac{\Theta^2}{2} < 0.015 \tag{86}$$

Note that the "global" effect of skew optical pumping on the helium polarization is larger because it is sensitive to the volume averaged alkali polarization (or equivalently the cumulative effect of many "local" inefficiencies). The "global" effect can only be understood by a full optical pumping simulation. This only increases amount of laser intensity required to maintain a "high" alkali polarization. It limits the highest attainable alkali polarization only through the $\cos(\Theta)$ term.

To estimate the effect of $D_2$ pumping, we'll assume that the laser light distrubution is gaussian:

$$\begin{aligned} \Phi(\nu,\vec{r}) &= \Phi_0(\vec{r})G(\nu) && (87)\\ G(\nu) &= \frac{1}{\sigma_l\sqrt{2\pi}}\exp\left(-\frac{(\nu-\nu_l)^2}{2\sigma_l^2}\right) && (88)\\ \lim_{\left(\frac{\sigma_l}{\nu_l}\right)\to 0}\int_0^\infty G(\nu)\mathrm{d}\nu &= 1 && (89) \end{aligned}$$

The absorption cross section is lorentzian when dominated by pressure broadening:

$$\sigma_{1,2} = f_{1,2}\sigma_0 L(\nu) \tag{90}$$

$$L(\nu) = \frac{1}{\pi}\frac{\frac{\Gamma_{1,2}}{2}}{(\nu-\nu_{1,2})^2+\left(\frac{\Gamma_{1,2}}{2}\right)^2} \tag{91}$$

$$\lim_{\left(\frac{\Gamma_{1,2}}{\nu_{1,2}}\right)\to 0}\int_0^\infty L(\nu)\mathrm{d}\nu = 1 \tag{92}$$

where $f_{1,2} = \frac{1.5\mp 0.5}{3}$ is the oscillator strength. For the $D_1$ pumping, we'll assume that the emission bandwidth of the laser is much larger that the absorption linewidth, $\sigma_l \gg \Gamma_1$:

$$\begin{aligned}
\gamma_{\mathrm{opD1}}(\vec{r}) &\approx \lim_{\frac{\Gamma_1}{\sigma_l}\to 0}\int_0^\infty \Phi(\nu,\vec{r})\sigma_1(\nu)\mathrm{d}\nu && (93)\\
&\approx \Phi_0(\vec{r})f_1\sigma_0\int_0^\infty\left[\lim_{\frac{\Gamma_1}{\sigma_l}\to 0}L(\nu)G(\nu)\right]\mathrm{d}\nu && (94)\\
&\approx \Phi_0(\vec{r})f_1\sigma_0\int_0^\infty G(\nu)\delta(\nu-\nu_1)\mathrm{d}\nu && (95)\\
&\approx \Phi_0(\vec{r})f_1\sigma_0 G(\nu_1) && (96)
\end{aligned}$$

For the $D_2$ pumping, the absorption peak is in the tail of the emission bandwidth and vice versa. Therefore if we make the assumption that the separation of the two peaks is much larger than either width , $\frac{\Gamma_2}{\nu_2 - nu_l} \ll \frac{\sigma_l}{\nu_2-\nu_l} \ll 1$ , then we can split the integral, $\nu_l < \nu_a < \nu_2$:

$$\begin{aligned}
\gamma_{\mathrm{opD2}}(\vec{r}) &\approx \lim_{\frac{\Gamma_2}{\sigma_l}\to 0}\int_0^\infty \Phi(\nu,\vec{r})\sigma_2(\nu)\mathrm{d}\nu && (97)\\
&\approx \Phi_0(\vec{r})f_2\sigma_0\int_0^\infty\left[\lim_{\frac{\Gamma_2\ \text{or}\ \sigma_l}{\nu_2-\nu_l}\to 0}L(\nu)G(\nu)\right]\mathrm{d}\nu && (98)\\
&\approx \Phi_0(\vec{r})f_2\sigma_0\left(\int_0^{\nu_a}\left[\lim_{\frac{\sigma_l}{\nu_2-\nu_l}\to 0}L(\nu)G(\nu)\right]\mathrm{d}\nu+\int_{\nu_a}^\infty\left[\lim_{\frac{\Gamma_2}{\nu_2-\nu_l}\to 0}L(\nu)G(\nu)\right]\mathrm{d}\nu\right) && (99)\\
&\approx \Phi_0(\vec{r})f_2\sigma_0\left(\int_0^{\nu_a}L(\nu)\delta(\nu-\nu_l)\mathrm{d}\nu+\int_{\nu_a}^\infty\delta(\nu-\nu_2)G(\nu)\mathrm{d}\nu\right) && (100)\\
&\approx \Phi_0(\vec{r})f_2\sigma_0\left[L(\nu_l)+G(\nu_2)\right] && (101)
\end{aligned}$$

Therefore, an estimate of the effect of inadvertent $D_2$ pumping is given as:

$$X_\alpha \approx \frac{3f_2}{2f_1}\left(\frac{G(\nu_2)+L(\nu_l)}{G(\nu_1)}\right) \tag{102}$$

We will take the laser emission to be centered on the $D_1$ line, $\nu_l = \nu_1$. Using typical values for optical pumping of Rb:

$$\begin{aligned}
\nu_2-\nu_1=\nu_{\mathrm{so}} &= c\left(\frac{1}{780\ \mathrm{nm}}-\frac{1}{795\ \mathrm{nm}}\right)\approx 7\times 10^3\ \mathrm{GHz} && (103)\\
\Gamma_2 &= \langle\sigma v\rangle_{\mathrm{pb}}[^3\mathrm{He}]\approx\left(20\ \frac{\mathrm{Ghz}}{\mathrm{amg}}\right)(7\ \mathrm{amg}) = 140\ \mathrm{GHz} && (104)\\
\sigma_l &\approx 1\ \mathrm{nm}\approx 500\ \mathrm{GHz} && (105)
\end{aligned}$$

gives the following estimates (in units of $\mathrm{GHz}^{-1}$):

$$G(\nu_1) = \frac{1}{\sigma_l\sqrt{2\pi}}\approx 8\times 10^{-4} \tag{106}$$

$$G(\nu_2) = \frac{1}{\sigma_l\sqrt{2\pi}}\exp\left(-\frac{(\nu_2-\nu_l)^2}{2\sigma_l^2}\right) \approx 10^{-32} \quad (107)$$

$$L(\nu_1) = \frac{1}{\pi}\frac{\frac{\Gamma_2}{2}}{(\nu_1-\nu_2)^2+\left(\frac{\Gamma_2}{2}\right)^2} \approx 5\times 10^{-7} \quad (108)$$

$$X_\alpha \approx 3\left(\frac{G(\nu_2)+L(\nu_l)}{G(\nu_1)}\right) \approx \frac{3\sigma_l\Gamma_2}{\nu_{\rm so}^2\sqrt{2\pi}} \approx 2\times 10^{-3} \quad (109)$$

where $\nu_2 - \nu_1$ is just the spin orbit splitting $\nu_{\rm so}$. Note that the contribution from of the laser light intensity at the $D_2$ resonance ($G(\nu_2)$) is very small compared to the contribution of the $D_2$ absorption cross section at the center of laser emission spectrum $L(\nu_l)$.

Another source of inefficiency comes from the possible reabsorption of $D_1$ and $D_2$ fluorescence. The rate equations that govern the populations of the $S_{\frac{1}{2}}$, $P_{\frac{1}{2}}$, and $P_{\frac{3}{2}}$ are given by:

$$\dot{s} = -\gamma_{\rm op}s + (\gamma_{\rm rad}+\gamma_{\rm non})(p_1+p_2) \quad (110)$$

$$\dot{p}_1 = +\gamma_{\rm op}s - (\gamma_{\rm rad}+\gamma_{\rm non})p_1 + \gamma_{\rm mix}(p_2-p_1) \quad (111)$$

$$\dot{p}_2 = -(\gamma_{\rm rad}+\gamma_{\rm non})p_2 + \gamma_{\rm mix}(p_1-p_2) \quad (112)$$

where $\gamma_{\rm op}$ is the $D_1$ pumping rate, $\gamma_{\rm rad}$ is the radiative quenching rate, $\gamma_{\rm non}$ is the non-radiative quenching rate, $\gamma_{\rm mix}$ is the collisional mixing rate, and $s, p_1, p_2$ are the relative populations of the $S_{\frac{1}{2}}$, $P_{\frac{1}{2}}$, and $P_{\frac{3}{2}}$ states respectively. Note that we have dropped the position dependence, assumed that there is little $D_2$ pumping, and assumed that both quenching rates and the collisional mixing rates are the same for both $P$ states. At equilibrium, this gives:

$$s = \frac{\gamma_{\rm rad}+\gamma_{\rm non}}{\gamma_{\rm rad}+\gamma_{\rm non}+\gamma_{\rm op}} \quad (113)$$

$$p_1+p_2 = \frac{\gamma_{\rm op}}{\gamma_{\rm rad}+\gamma_{\rm non}+\gamma_{\rm op}} \quad (114)$$

$$\frac{p_1-p_2}{p_1+p_2} = \frac{\gamma_{\rm rad}+\gamma_{\rm non}}{\gamma_{\rm rad}+\gamma_{\rm non}+2\gamma_{\rm mix}} \quad (115)$$

$$\frac{p_1}{p_2} = 1+\frac{\gamma_{\rm rad}+\gamma_{\rm non}}{\gamma_{\rm mix}} \quad (116)$$

$$(117)$$

Assuming that every fluorescense photon is unpolarized, the local relaxation rate due to reabsorption is estimated by:

$$\gamma_{\rm f} = \int \Phi_{\rm f}(\nu)\sigma(\nu){\rm d}\nu \quad (118)$$

$$= \Phi_{\rm f}^0\frac{\int\sigma^2(\nu){\rm d}\nu}{\int\sigma(\nu){\rm d}\nu} \quad (119)$$

$$\Phi_{\rm f}^0 = \gamma_{\rm rad}[A]\left[\frac{\gamma_{\rm op}}{\gamma_{\rm rad}+\gamma_{\rm non}+\gamma_{\rm op}}\right]\left[\frac{\gamma_{\rm rad}}{\gamma_{\rm rad}+\gamma_{\rm non}}\right] l \quad (120)$$

$$\approx \gamma_{\rm op}\frac{\int\sigma(\nu){\rm d}\nu}{\int\sigma^2(\nu){\rm d}\nu}\left[\frac{\gamma_{\rm rad}}{\gamma_{\rm rad}+\gamma_{\rm non}}\right]^2 \quad (121)$$

$$\langle l\rangle^{-1} \equiv [A]\frac{\int\sigma^2(\nu){\rm d}\nu}{\int\sigma(\nu){\rm d}\nu} \quad (122)$$

where the fluorescence intensity is given as the radiative decay rate times the alkali density times the fraction of atoms in the excited state times the fraction of atoms that undergo radiative decay times the average absorption length ($l$) and we've taken advantage of the relative sizes of the different rates [RMP 44 p169-249

(1972)]:

$$\gamma_{\rm rad} = \frac{\gamma_{\rm rad-D1}+\gamma_{\rm rad-D2}}{2} = \left\langle \tau_{\rm spon}^{-1}\right\rangle = (27\ {\rm ns})^{-1} \approx 40\ {\rm MHz} \tag{123}$$

$$\gamma_{\rm non} \approx \left[\frac{\sigma_{\rm qD1}+\sigma_{\rm qD2}}{2}\right] \langle v_{\rm N2-Rb}\rangle\, [{\rm N}_2] = \left(50\times 10^{-20}\ {\rm m}^2\right)\left(700\ \frac{\rm m}{\rm s}\right)(0.08\ {\rm amg}) \approx 750\ {\rm MHz} \tag{124}$$

$$\gamma_{\rm mix} \approx \left[\frac{\sigma_{\rm mD1}+\sigma_{\rm mD2}}{2}\right] \langle v_{\rm N2-Rb}\rangle\, [{\rm N}_2] = \left(20\times 10^{-20}\ {\rm m}^2\right)\left(700\ \frac{\rm m}{\rm s}\right)(0.08\ {\rm amg}) \approx 300\ {\rm MHz} \tag{125}$$

$$\gamma_{\rm op} \approx 1\text{ to }10^3\text{ kHz depending on initial laser power and position in cell} \tag{126}$$

Note that the wavelength distribution of the fluorescence light is same as the absorption cross section. Therefore we can estimate the effect of fluorescence reabsorption:

$$\gamma_{\rm f} = \gamma_{\rm op}\left[\frac{\gamma_{\rm rad}}{\gamma_{\rm rad}+\gamma_{\rm non}}\right]^2 \to X_\alpha \approx \left[\frac{\gamma_{\rm rad}}{\gamma_{\rm rad}+\gamma_{\rm non}}\right]^2 \approx 2.5\times 10^{-3} \tag{127}$$

This is admittedly a very crude estimate, but it is almost certainly good to within one order of magnitude. From skew pumping, inadvertent $D_2$ pumping, and fluorescence reabsorption, this gives a total estimate of:

$$X_\alpha \approx \frac{\Theta^2}{2} + \frac{3\sigma_l\Gamma_2}{\nu_{\rm so}^2\sqrt{2\pi}} + \left[\frac{\gamma_{\rm rad}}{\gamma_{\rm rad}+\gamma_{\rm non}}\right]^2 \tag{128}$$

$$0.002 < X_\alpha < 0.02 \tag{129}$$

In comparison to experiment, [Phys. Rev. Lett. 91, 123003 (2003)] gives $X_\alpha \approx 0.0027$ in some hybrid cells they tested.

## 4.4 Extra Spin-Exchange Related Couplings

Normally the alkali atom-helium nucleus interaction is written as (in SI) [Walker & Happer, RMP 69, p629 (1997)]:

$$\mathcal{H}_{\rm se} = -\left[\frac{\mu_0}{4\pi}\frac{8\pi}{3}\delta(\vec{R})\right] g_{\rm e}\mu_B g_{^3{\rm He}}\mu_N \vec{S}\cdot\vec{K} \tag{130}$$

where $\vec{R}$ is the separation between the alkali atom valence electron and the helium nucleus. The spin exchange cross section is proportional to the modulus square matrix element of this interaction. Since the alkali atom is much larger than the helium atom, we can evaluate the matrix element of the interaction using the alkali atom wave functions:

$$\sigma_{\rm se} \propto \left[\frac{\mu_0}{4\pi}\frac{8\pi}{3} g_{\rm e}\mu_B g_{^3{\rm He}}\mu_N\right]^2 |\langle\psi|\,\delta(\vec{R})\,|\psi\rangle|^2 = \left[\frac{2\mu_0}{3} g_{\rm e}\mu_B g_{^3{\rm He}}\mu_N\right]^2 |\psi(0)|^2 \tag{131}$$

This form of the interaction hamiltonian is incomplete. In principle, the "full" interaction hamiltonian is [Jackson, Classical Electrodynamics, 3rd ed, pages 188-90 (1999)]:

$$\mathcal{H}_{\rm full} = -\vec{\mu}_{^3{\rm He}}\cdot\left(\vec{B}_{\rm spin}+\vec{B}_{\rm orbital}\right) \tag{132}$$

$$\vec{B}_{\rm spin} = \frac{\mu_0}{4\pi}\left[\frac{3\hat{n}\left(\hat{n}\cdot\vec{\mu}_{\rm electron}\right)-\vec{\mu}_{\rm electron}}{R^3} + \frac{8\pi}{3}\vec{\mu}_{\rm electron}\delta(\vec{R})\right] \tag{133}$$

$$\vec{B}_{\rm orbital} = \frac{\mu_0}{2\pi}\frac{\mu_B}{R^3}\vec{L} \tag{134}$$

The interaction hamiltonian now contains three terms:

$$\mathcal{H}_{\rm full} = \mathcal{H}_{\rm se}+\mathcal{H}_{\rm ani}+\mathcal{H}_{\rm oam} \tag{135}$$

$$\mathcal{H}_{\rm ani} = -\frac{\mu_0}{4\pi} g_{\rm e}\mu_B g_{^3{\rm He}}\mu_N\left[\frac{3\left(\vec{n}\cdot\vec{S}\right)\left(\vec{n}\cdot\vec{K}\right)-R^2\vec{S}\cdot\vec{K}}{R^5}\right] \tag{136}$$

$$\mathcal{H}_{\rm oam} = -\frac{\mu_0}{2\pi}\frac{\mu_B g_{^3{\rm He}}\mu_N}{R^3}\vec{L}\cdot\vec{K} \tag{137}$$

The anisotropic term is the interaction between the long range dipolar field of the electron spin with the helium nucleus. In the literature [D. K. Walter, W. Happer, and T. G. Walker, Phys. Rev. A 58, 3642-3653 (1998)], it is referred to "Anisotropic Spin Exchange." It acts to polarize the the nuclear spin in an orientation opposite to that of the usual or "isotropic" spin-exchange interaction. The effect of this interaction can be written as [RMP 69]:

$$X_\beta \approx \frac{\int \left[\frac{3\vec{R}'\cdot\hat{R}\hat{R}\cdot\vec{R}'-R'^2}{R'^5}\right]|\psi(R')|^2 \mathrm{d}^3R'}{\frac{16\pi^2}{9}|\psi(0)|^2} \leq 0.025 \tag{138}$$

where the estimate comes from [PRA 58, 3642 (1998)]]. This point is discussed in [PRA 66, 032703 (2002)]. Essentially they argue that since the calculation in [PRA 58, 3642 (1998)] correctly estimates the size of the "isotropic" spin exchange rate constant (within 20 percent), their estimate of the "anisotropic" spin exchange rate constant must be roughly as good. Therefore it contributes only a small amount to $X_\beta$.

For the ground state atoms, only the isotropic and anisotropic spin exchange terms contribute because $\vec{L} = 0$ in the ground state. However, the excited states have $\vec{L} > 0$ and therefore the orbital angular momentum term has a contribution. As was mentioned in the previous section, a small fraction (at the level of 1-1000 ppm) of the alkali atoms are in the excited state. These excited states atoms are probably not polarized due to collisional mixing. Therefore any interaction between these excited atoms and helium nuclei are probably spin relaxing. The effect of orbital angular momentum interaction can be written as:

$$X_\beta \approx \left[\frac{\gamma_{\mathrm{op}}}{\gamma_{\mathrm{rad}}+\gamma_{\mathrm{non}}}\right] \frac{\int |\psi_{\frac{1}{2}}(R)|^2 + |\psi_{\frac{3}{2}}(R)|^2 \frac{\mathrm{d}^3R}{2R^3}}{\frac{16\pi}{9} g_e^2 |\psi(0)|^2} \tag{139}$$

where we assume that the excited atoms are equally distributed between $P_{\frac{1}{2}}$ and $P_{\frac{3}{2}}$ states.

## 4.5 Helium Spin-Rotation Interaction

The helium nuclear spin can couple to the angular momentum of the alkali atom helium atom pair rotating about a common axis:

$$\mathcal{H}_{\mathrm{rot}} = f(R)\vec{N}\cdot\vec{K} \tag{140}$$

For He-alkali collisions, the rotational angular momentum $\vec{N}$ couples much more strongly to the electron spin than to the helium nuclear spin [Phys. Rev. A 56, 2090-2094 (1997)]. Therefore it is assumed that this effect is negligible.

## 4.6 Wall Interactions

A series of studies have been done to understand the wall relaxation in helium cells. In studies where the lifetime of cell have been measured, it has been found:

1. Alkali coated cells are better than bare wall cells. [PLA 201, p337-43 (1995)]
2. Sol gel coatings appears to increase the lifetimes as well as reduce their variation. [Appl Phys Lett, 77, p2069-71 (2000)]
3. Bare wall cells do not exhibit magnetic hysterisis. [PRL 87, 143004 (2001)] [J App Phys, 92, p1588-97 (2002)] [Chem Phys Lett, 370, p261-7 (2003)] [PRA 69, 021401 (2004)] [Physica B, 356, p 91-95 (2005)]
4. Alkali coated cells exhibit hysterisis. [ibid]
5. Alkali coated cells can be degaussed. [ibid]

Recently [Jour. App. Phys., 94, pp. 6908-14 (2003)] and [Phys. Rev. Lett. 96, 083003 (2006)] have shown:

1. The alkali polarization is quite often $\approx 1$, therefore $X_\alpha \approx 0$.
2. There is an unaccounted for helium relaxation rate that appear proportional to the alkali density and surface to volume ratio.

3. They found the range $0.15 < X_\beta < 1.3$.
4. For small surface to volumes ratios, there is less scatter in $X_\beta$.
5. For large surface to volumes ratios, there is larger scatter in $X_\beta$.
6. For the cells with the largest $X_\beta$ for a particular value of surface to volume ratio $\frac{S}{V}$:

$$0.15 \leq X_\beta \leq X_\beta^{\max} \approx (0.4 \text{ cm}) \frac{S}{V} \tag{141}$$

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