

# **Field Ionization and Field Emission with Intense, Single-cycle THz Pulses**

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by

© Yunxiao Wang

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## Abstract

Nuclear-polarized  $^3\text{He}$  targets have been widely used in electron-scattering experiments in Thomas Jefferson National Accelerator Facility (JLAB) since mid 1990s. It is of great importance to produce large amounts of  $^3\text{He}$  gas with high polarization.

The latest experiments run in JLAB prior to the 12GeV upgrade have been using cells polarized with Spin-Exchange Optical Pumping (SEOP). These cells were made of the GE180 glass and use a two-chambered design. The top chamber, known as the pumping chamber, is where  $^3\text{He}$  is polarized through SEOP. The bottom chamber, known as the target chamber, is where electron scattering occurs. Great effort has been made in our lab to develop this generation of cells. Alkali-hybrid SEOP together with narrowband laser diode arrays have increased the  $^3\text{He}$  polarization from 37% to 65%. Among other things, we also carefully studied an additional spin relaxation mechanism that limits the maximum achievable  $^3\text{He}$  polarization.

The 12GeV upgrade makes the future experiments much more demanding in terms of target cell performance. One challenge it brings is the high relaxation due to electron beam. We have designed and tested a new style cell that uses convection instead of diffusion to increase the rate at which the polarization in the target chamber is being replenished by gas from pumping chamber. We have obtained over 50% polarization with controllable convection speed so far.

An additional problem that comes with higher beam current is that the glass end windows of traditional design are not likely to survive the experiments. Our group started exploring the option of using metal end windows from a decade ago. The first problem to solve is to find out the correct material and the proper technique

to incorporate metal without introducing significant spin relaxation and still being able to hold high pressure gas (12 atm) inside. This is a brand new technique that may have a profound impact of future cell designs once fully developed. Although no metal end windows have been tested so far, multiple glass cells with different kinds metal tubes (much larger in area compared to the end windows that will be used in JLAB experiments) attached were examined and were enough to convince us the extra spin relaxation is not likely to cause significant problems. The metals tubes were connected to Pyrex glass with knife-edge (housekeeper) seals and stayed intact through high pressure tests. After exploring options such as pure copper, gold coated copper, titanium, stainless steel, gold coated titanium, we have established that electroplating gold on copper substrate yields the best result so far. Further tests are planned before attaching metal end windows to GE180 glass and using them in electron-scattering experiments.

## Acknowledgements

Put your acknowledgements here...

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— MUN School of Graduate Studies

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# Chapter 1

## Spin-Exchange Optical Pumping

### 1.1 Overview

Spin-polarized noble gases have been widely used for various purposes. In JLAB, polarized  $^3\text{He}$  is used as target for electron-scattering experiments. This is because a  $^3\text{He}$  nucleus has a pair of protons with paired spins and a single neutron that contributes the most of the nuclear spin. In MRI, polarized  $^3\text{He}$  has seen uses such as detecting structural damage in the lungs.

There are generally two ways of polarizing  $^3\text{He}$ : Metastability-Exchange Optical Pumping (MEOP) and Spin-Exchange Optical Pumping (SEOP). Our group focuses on SEOP as MEOP polarizes gas at relatively low pressure, thus further compression is required to produce target cells.

In SEOP, alkali metal is polarized by circularly polarized laser that's tuned to the D1 transition of it.  $^3\text{He}$  obtains polarization from alkali metal through spin exchange process. With the combination of hybrid alkali mixtures (typically Rb and K) and

spectrally narrowed lasers, more than 70% polarizations have been produced.

## 1.2 Optical pumping

### 1.2.1 Rb for SEOP

In optical pumping, Rb is often the alkali metal that's optically pumped by circularly polarized laser light. The angular momentum of a polarized photon is transferred to the valence electron of Rb atom. In the case of hybrid mixture of Rb/K, Rb is still the alkali metal that's pumped by laser light directly while K serves as a more efficient medium to transfer the polarization from Rb to  $^3\text{He}$  compared to direct transfer from Rb to  $^3\text{He}$ . Rb is so widely used mainly for its low melting point, the relative ease of acquiring laser at the Rb D1 line wavelength and the clear separation between D1 (794.7nm) and D2 line (780nm).

The Rb melting point is at 39.5°C, so it's easy to achieve enough Rb vapor without having to drive the oven temperature too high. In our lab, depending on if the cell contains pure Rb or Rb/K mixture, the oven temperature can be between 85°C to as high as 255°C. Perhaps the most used oven temperature for hybrid is 235°C which has empirically been a good temperature to produce Rb/K mixture vapor, while 85°C is enough for pure Rb.

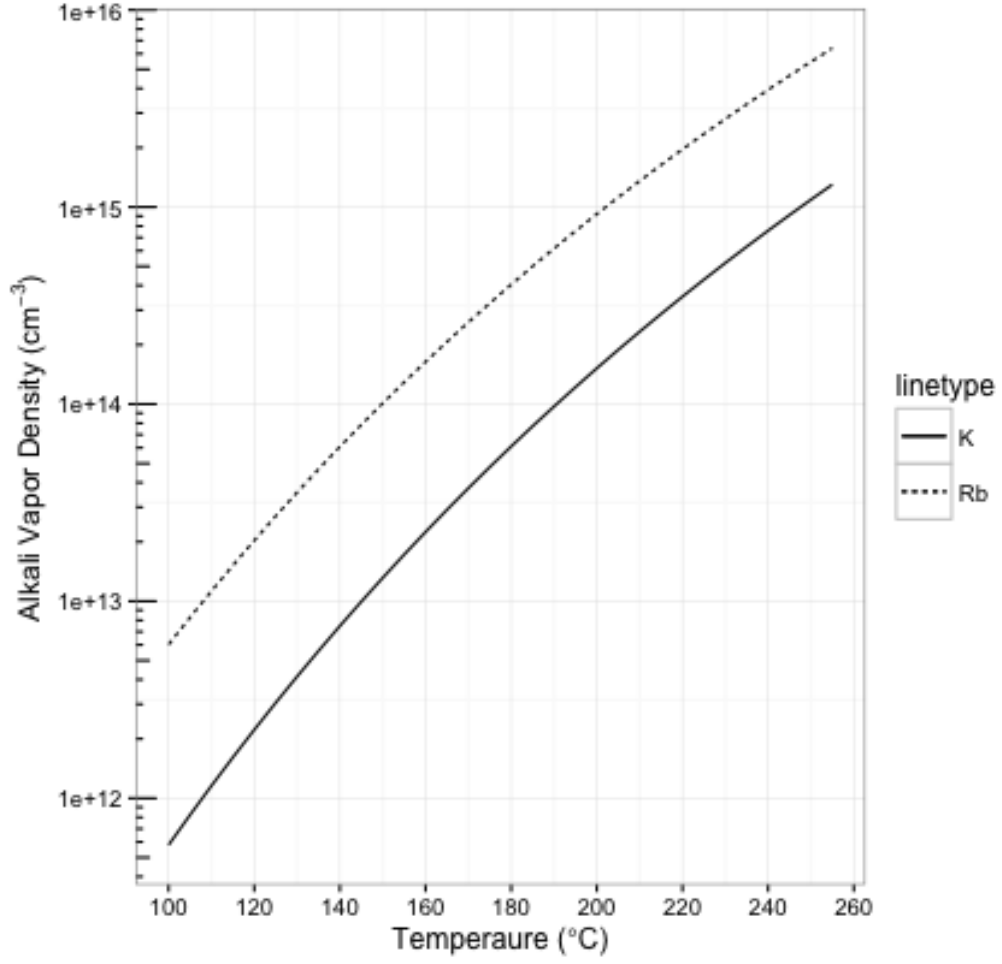


Figure 1.1: Rb And K Number Density Curves

### 1.2.2 Energy Levels of Alkali Metal in External Magnetic Field

The Hamiltonian for ground state ( $L=0$ ) alkali metal atoms in external magnetic field is:

$$\mathbf{H} = A\mathbf{I} \cdot \mathbf{S} + g_e\mu_B S_z B_z + g_N\mu_N I_z B_z \quad (1.1)$$

The first term  $A\mathbf{I} \cdot \mathbf{S}$  describes the coupling of the nuclear spin  $\mathbf{I}$  with the electron spin  $\mathbf{S}$  and is key to spin exchange, where  $A$  is the isotropic magnetic-dipole coupling coefficient. The resulting energy levels are referred to as hyperfine structure. The second and third terms describe the Zeeman splitting due to the presence of a weak external magnetic field.  $\mu_B = 9.274 \times 10^{-24} J/T$  and  $\mu_N = 5.051 \times 10^{-27} J/T$  are the bohr and nuclear magnetons.  $g_e \approx 2$  and  $g_N \approx 5.59$  are the electronic and nuclear Lande g-factors.

The linear relationship between energy levels and magnetic field only holds for weak magnetic fields as is the case with our lab where 13 Gauss is used most of the time. When the Zeeman splitting grows relative to the hyperfine energy difference one would have to take into account the quantum mixing of the states, the result is described by Breit-Rabi Formula. With 13 Gauss, the hyperfine term dominates the total Hamiltonian. The energy levels of  $^{87}\text{Rb}$  are shown in figure ??.

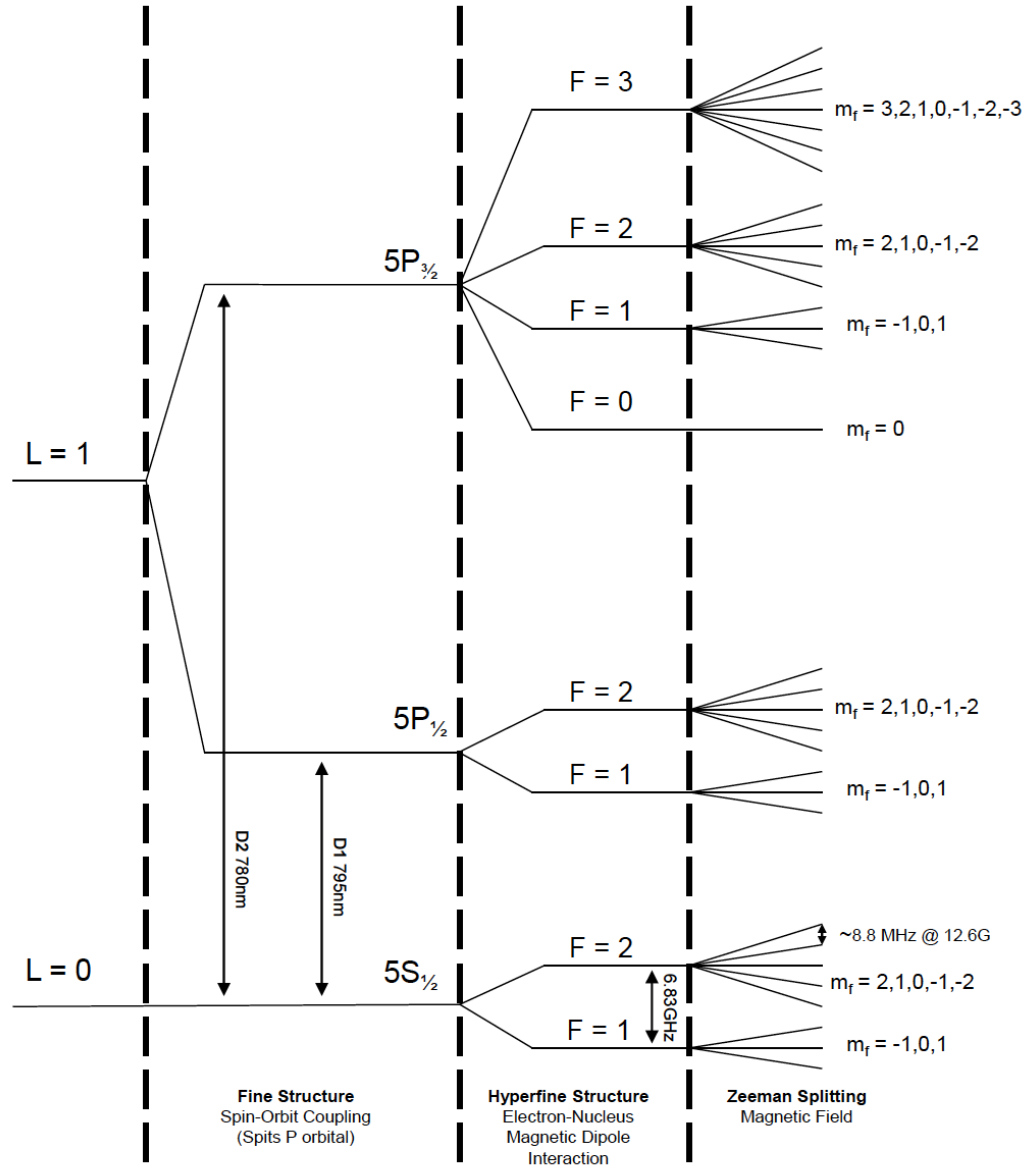


Figure 1.2: Level Diagram of  $^{87}\text{Rb}$ . The splittings are not to scale.

Adapted from Dolph's PhD thesis.

### 1.2.3 Optical Pumping Process Overview

For simplicity, the following discussion will ignore the nuclear spins for now. The inclusion of nuclear spins will increase the number of energy states but the optical pumping mechanism remains the same. In our typical setup, circularly polarized laser light is tuned to the D1 line of Rb and excites valence electrons of Rb from  $5S_{1/2}$  state to  $5P_{1/2}$  state as shown in figure 1.3 ( $2S_{1/2}$  and  $2P_{1/2}$  states are used in the figure for the simple model described below).

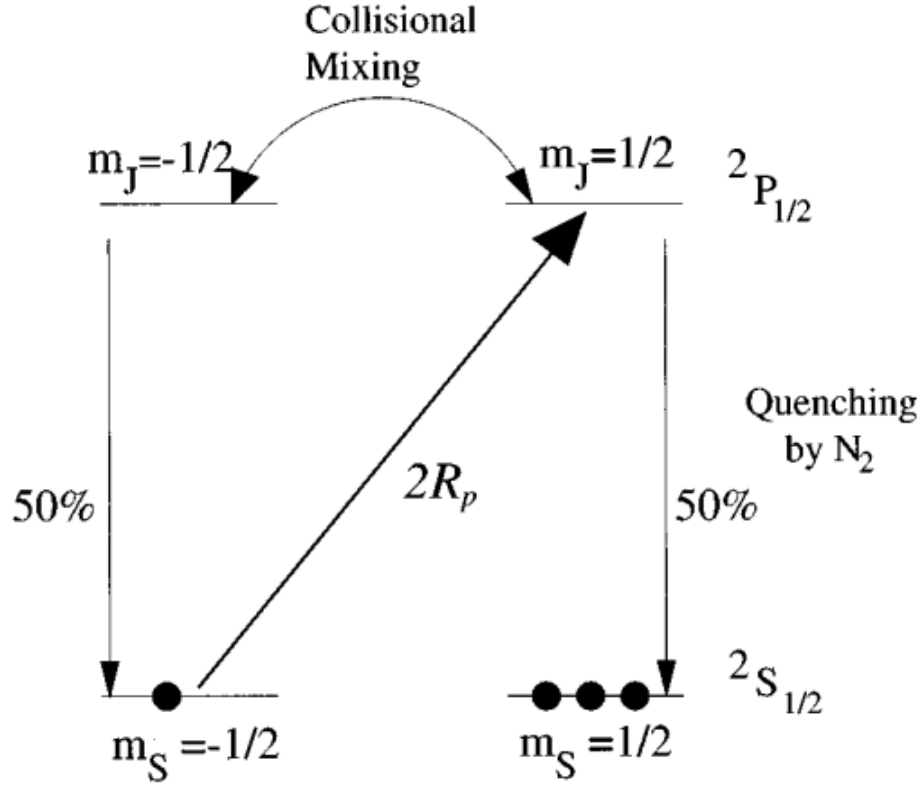


Figure 1.3: The interaction of alkali-metal atoms with left-circularly ( $\sigma^+$ ) polarized light. (from Ref. [?])

Although left-circularly polarized light is assumed in the figure, either circular polarization works the same. Conservation of angular momentum requires  $\Delta m = +1$  as the figure show. Through collisions with other Rb atoms, excited electrons will be mix and evenly distribute on the two  $2P_{1/2}$  states. Electrons then decay to the two ground states with equal probabilities. The selection rule for the decay process is  $\Delta m = 0$  or  $\pm 1$ . Even though both grounds receive electrons from the decay, only the  $m = -1/2$  state absorb the circular polarized photons, so atoms are in effect pumped to the  $m = +1/2$  state. When we consider Rb with nuclear spins, both  $5S_{1/2}$  and  $5P_{1/2}$  states are split into more energy levels, but the net effect is still that the ground state with highest  $m$  accumulate atoms from those with lower  $m$ .

When the excited electrons decay back to the ground state, they emit unpolarized photons which can depolarize the gas. A small amount of  $N_2$  gas is added into the cell (typically around 0.1 Amagats) to non-radiatively quench the excited electrons as  $N_2$  molecules can absorb the released energy of spontaneous decays into their rotational and vibrational modes of oscillation. With an appropriate amount of  $N_2$ , the photon-emitting decays can be reduced to less than 5%.

### 1.2.4 Optical Pumping Rate

The optical pumping rate at position  $\vec{r}$  can be described by

$$R = \int \Phi(\nu, \vec{r}) \sigma(\nu) d\nu \quad (1.2)$$

where  $\Phi(\nu, \vec{r})$  is the position dependent photon spectral flux density and  $\sigma(\nu)$  is the photon absorption cross section. The cross section has a natural Lorentzian

lineshape which is broadened by Doppler effect and pressure broadening. The pressure broadening effect dominates the lineshape as our cells normally have densities well above one amagat.  $\sigma(\nu)$  follows the sum rule:

$$\int \sigma(\nu) d\nu = \pi r_0 c f \quad (1.3)$$

where  $r_0 = 2.82 \times 10^{-13}$  cm is the classical electron radius and  $f=0.337$  is the transition oscillator strength. Thus the photon absorption cross section can be described by Lorentzian lineshape:

$$\sigma(\nu) = f r_e c \frac{\frac{\Gamma_A}{2}}{(\nu - \nu_0)^2 + (\frac{\Gamma_A}{2})^2} \quad (1.4)$$

where  $\Gamma_A$  is the pressure dependent FWHM. At the front of the cell, the photon spectral flux density is the product of a Gaussian spatial distribution and a Gaussian spectrum.

$$\phi(\nu, \vec{r}) = \phi_0(\vec{r}) G(\nu) \quad (1.5a)$$

$$\phi_0(\vec{r}) = \frac{P}{h\nu} \frac{2}{\omega^2 \pi} e^{2r^2/\omega^2} \quad (1.5b)$$

$$G(\nu) = \frac{1}{\sqrt{2\pi}\sigma_l} e^{-(\nu-\nu_l)^2/2\sigma_l^2} \quad (1.5c)$$

where  $P$  is the laser power;  $\omega$  is the beam waist;  $\sigma_l$  is the Gaussian width of the laser and  $\nu_l$  is the central laser frequency.



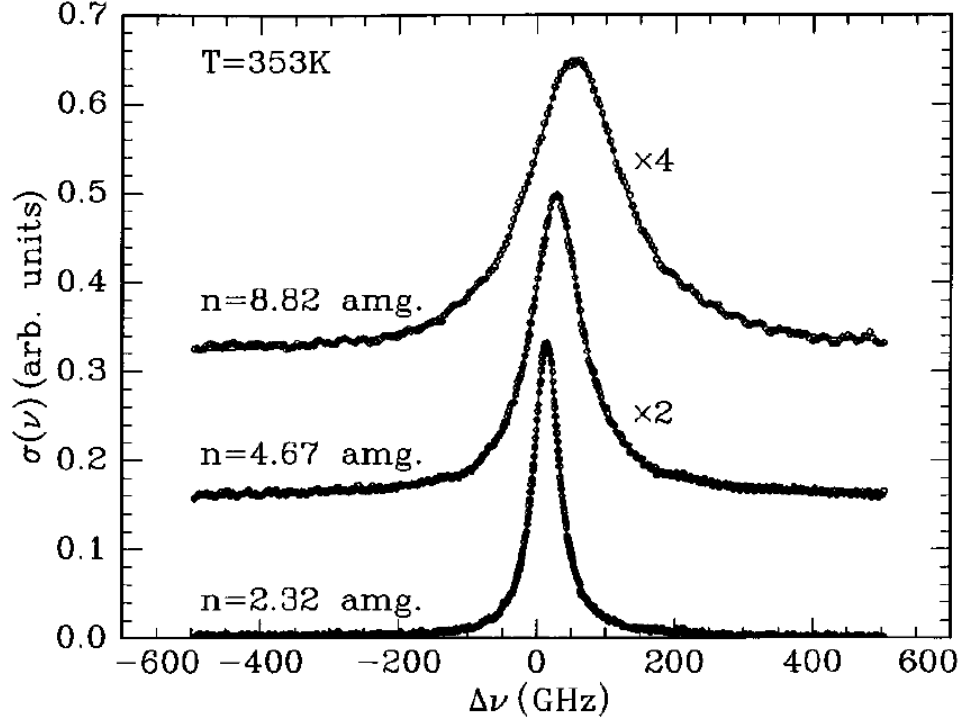


Figure 1.4: Absorption cross section for Rb  $D_1$  line in the presence of three different densities of  $^3\text{He}$ . (from Ref. [1])

### 1.2.5 Polarization Time Evolution

Although polarizations of Rb electrons are more complex,  $^3\text{He}$  nuclei have an intrinsic nuclear spin of  $1/2$ , and it's simpler to explain the math with spin of  $1/2$ , let's define the polarization as the asymmetry between  $+1/2$  state and  $-1/2$  state:

$$P = \frac{\rho_{+1/2} - \rho_{-1/2}}{\rho_{+1/2} + \rho_{-1/2}} = \rho_{+1/2} - \rho_{-1/2} \quad (1.6)$$

where  $\rho_{\pm 1/2}$  is the population in the  $\pm 1/2$  state.

The time evolution of polarization for both Rb electrons and  $^3\text{He}$  follows the equation:

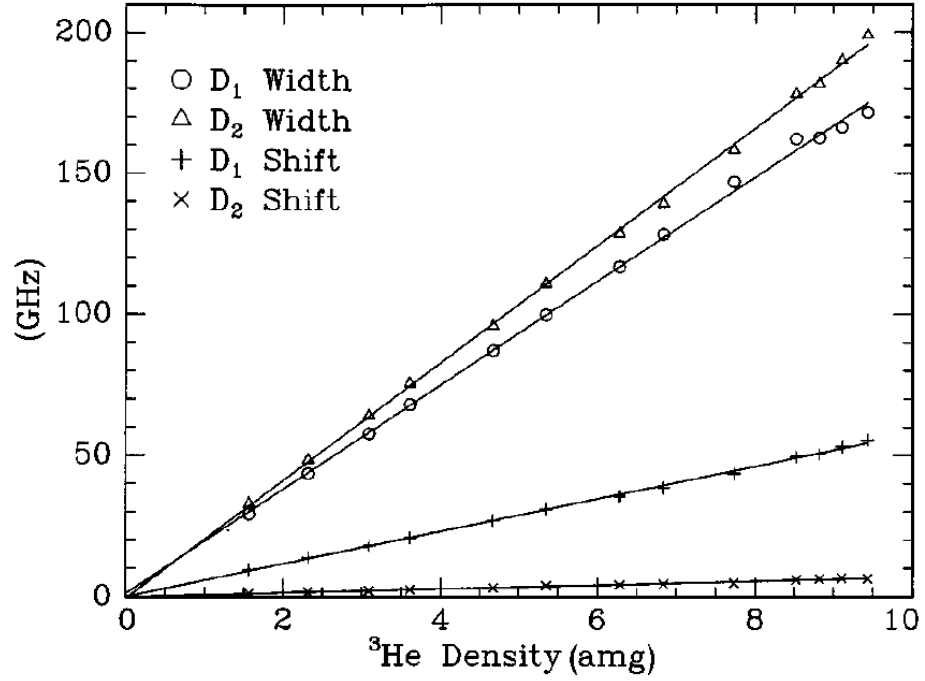


Figure 1.5: The shift and the broadening due to presence of  $^3\text{He}$  for Rb  $D_1$  and  $D_2$  lines. (from Ref. [1])

$$\frac{dP}{dt} = \gamma(1 - P) - \Gamma \cdot P \quad (1.7)$$

$\gamma$  is the polarization rate and  $\Gamma$  is the depolarization rate in the above differential equation. The solution has the simple form of:

$$P(t) = Ce^{-(\gamma+\Gamma)t} + \frac{\gamma}{\gamma + \Gamma} \quad (1.8)$$

Note the depolarization rate also contributes to the speed at which  $P$  approaches saturation. The saturated polarization is defined as the value of  $P$  in the limit  $t \rightarrow \infty$ :

$$P_{\infty} = \frac{\gamma}{\gamma + \Gamma} \quad (1.9)$$

The initial polarization is defined as the value of P at t = 0:

$$P_0 = C + \frac{\gamma}{\gamma + \Gamma} = C + P_{\infty} \quad (1.10)$$

Thus, P(t) can be expressed as:

$$P(t) = (P_0 - P_{\infty})e^{-(\gamma+\Gamma)t} + P_{\infty} \quad (1.11)$$

In the case of polarizing Rb with a pump laser.  $\gamma$  is the pumping rate R and  $\Gamma$  is the Rb spin relaxation rate  $\Gamma_{Rb}$ . There is typically a small angle  $\theta$  between the pump laser and the magnetic field that separates the different spin states of Rb valence electrons even though great effort has been made to minimize the angle. Thus P(t) can be rewritten as:

$$P(t) = P_0 e^{-(R+\Gamma_{Rb})t} + P_{laser} \cos\theta \frac{R}{R + \Gamma_{Rb}} (1 - e^{-(R+\Gamma_{Rb})t}) \quad (1.12)$$

$P_{laser}$  is the circular polarization of the pump laser which is above 99.5%. Rb close to the front side of the cell can reach above 97% (depends on the laser power and other factors) on the order of 100's of microseconds. As the laser propagates through the cell, power is attenuated by Rb vapor. Therefore Rb polarization at the back side of the cell is lower than that at the front side. One way to overcome the problem is to shine pump laser from both sides of the side which would lead to higher overall Rb polarization and  $^3\text{He}$  polarization.

Spins are thermally polarized with the presence of a magnetic field even without external pumping source. The probability for a spin to be in state  $s$  is:

$$Prob. = \frac{e^{-E_s/k_B T}}{\sum_{si} e^{-E_{si}/k_B T}} \quad (1.13)$$

where  $E_s$  is the energy of the state,  $k_B$  is the Boltzmann constant and  $T$  is the temperature. Using the thermal distribution, under typical operating conditions,  $^3\text{He}$  polarization is  $10^{-9}$  and Rb polarization is  $10^{-5}$ . Both are negligible without active pumping.

### 1.2.6 Rb Spin Destruction Rate

There are two main mechanisms of Rb depolarization: the binary collisions with Rb,  $^3\text{He}$  and  $N_2$  and the formation and breakup of van der Waals molecules, while the second mechanism is negligible for  $^3\text{He}$  cells. The Rb spin destruction rate can then be expressed as

$$\Gamma_{Rb} = k_{Rb-Rb}[Rb] + k_{Rb-^3He}[^3He] + k_{Rb-N_2}[N_2] \quad (1.14)$$

where  $k_{Rb-X}$  is the spin destruction rate constant and  $[X]$  is the density of  $X$ . Dolph has summarized these constants based on measurements from various groups:

$$k_{Rb-^3He}(T) = 55.9(9) \left( \frac{T}{473.15K} \right)^{3.31(12)} \text{ Hz}/\text{amg} \quad (1.15a)$$

$$k_{Rb-N_2}(T) = 290(30) \left( \frac{T}{473.15K} \right)^{2.0(25)} \text{ Hz}/\text{amg} \quad (1.15b)$$

$$k_{Rb-Rb} = 4.813(48) \times 10^{-13} \text{ Hz} \cdot \text{cm}^3 \quad (1.15c)$$

For a pure Rb cell at 170°C with the following densities in the pumping chamber:

$$[{}^3He] \approx 8.0 amg \quad (1.16a)$$

$$[N_2] \approx 0.08 amg \quad (1.16b)$$

$$[Rb] \approx 6.0 \times 10^{14} cm^{-3} \quad (1.16c)$$

The approximate spin destruction rates due to various gases are:

$$\Gamma_{Rb-{}^3He} \approx 360 Hz \quad (1.17a)$$

$$\Gamma_{Rb-N_2} \approx 20 Hz \quad (1.17b)$$

$$\Gamma_{Rb-Rb} \approx 289 Hz \quad (1.17c)$$

The total spin destruction rate is 669 Hz.

### 1.3 Spin Exchange

Following equation 1.12, the time evolution of  ${}^3He$  polarization can be expressed as:

$$P_{{}^3He}(t) = P_0 e^{-(\gamma_{se} + \Gamma)t} + P_{Rb} \frac{\gamma_{se}}{\gamma_{se} + \Gamma} (1 - e^{-(\gamma_{se} + \Gamma)t}) \quad (1.18)$$

The saturation polarization is

$$P_{\infty} = P_{Rb} \frac{\gamma_{se}}{\gamma_{se} + \Gamma} \quad (1.19)$$

where  $\gamma_{se}$  is the spin exchange rate between  ${}^3He$  and Rb, and  $\Gamma$  is the spin relaxation rate.

### 1.3.1 Spin-Dependent Interactions

The key process in spin-exchange optical pumping is collisional transfer of polarization between optically pumped alkali-metal atoms and the nuclei of the noble gas atoms. As in Fig. 1.7, the transfer of angular momentum occurs either while the atoms are bound in van der Waals molecules or in simple binary collisions. For  $^3\text{He}$ , binary collisions dominate, and the contribution from van der Waals molecules is negligible. The time scale for binary collisions is on the order of  $10^{-12}$  sec, so the collision can induce both  $\Delta F = \pm 1$  and  $\Delta F = 0$  transitions between hyperfine sublevels. For heavier noble gases like  $^{129}\text{Xe}$  at pressure of a few tens of Torr, the contributions of van der Waals molecules can greatly exceed that of binary collisions. At several atmos which is the typical operating pressure for SEOP, the time scale of van der Waals molecules is greatly limited by collision so that the binary collisions dominate.

Spin-dependent interactions produce the spin transfer and relaxation. For SEOP, spin-rotation interaction between  $\vec{S}$  and the rotational angular momentum  $\vec{N}$  and the isotropic hyperfine interaction between  $\vec{S}$  and the noble-gas nuclear spin  $\vec{I}$  dominate the spin-exchange process:

$$V_1(\vec{R}) = \gamma(R)\vec{N} \cdot \vec{S} + A(R)\vec{I} \cdot \vec{S} \quad (1.20)$$

$\vec{I}_a$  and  $\vec{I}_b$  are the nuclear spins of the atomic pair.

The spin-rotation interaction is caused by the magnetic fields from relative motion of the charges of the colliding atoms, and the isotropic hyperfine interaction comes from the magnetic field inside the nucleus of the noble-gas atom.

An alkali-metal atom and a noble-gas atom interact via both a large spin-independent

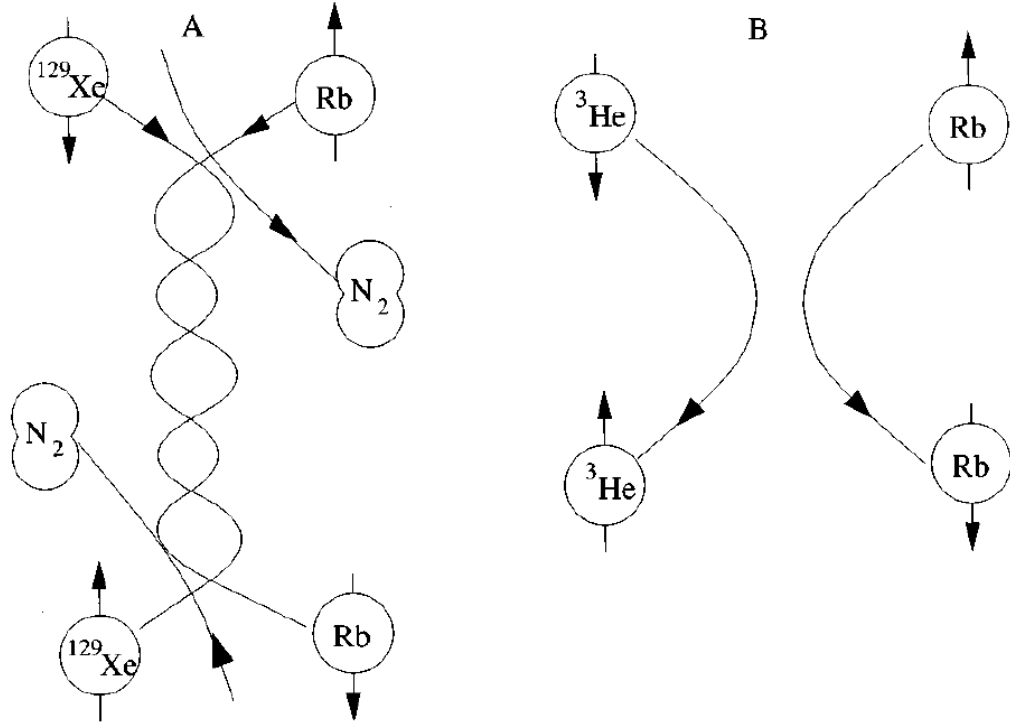


Figure 1.6: A. Formation and breakup of alkali-metal/noble-gas van der Waals molecule. B. Binary collision between an alkali-metal atom and a noble-gas atom. (from Ref. [?])

interaction  $V_0(R)$  and a small spin-dependent interaction  $V_1(R)$ . At the high operating temperatures,  $V_0$  determines classical collision trajectories, while  $V_1$  acts as a small perturbation. We'll focus on  $V_1$  below since it is responsible for spin exchange.

Including a few more terms that were neglected in Eq. 1.20, the spin-dependent interaction  $V_1(R)$  can be expressed as:

$$\begin{aligned}
V_1(\vec{R}) = & \gamma(R)\vec{N} \cdot \vec{S} + \sum_k A_k(R)\vec{I}_k \cdot \vec{S} \\
& + \sum_k B_k(R)\vec{I}_k \cdot (3\vec{R}\vec{R} - 1) \cdot \vec{S} \\
& + \sum_k C_k(R)\vec{I}_k \cdot (3\vec{R}\vec{R} - 1) \cdot \vec{I}_k
\end{aligned} \tag{1.21}$$

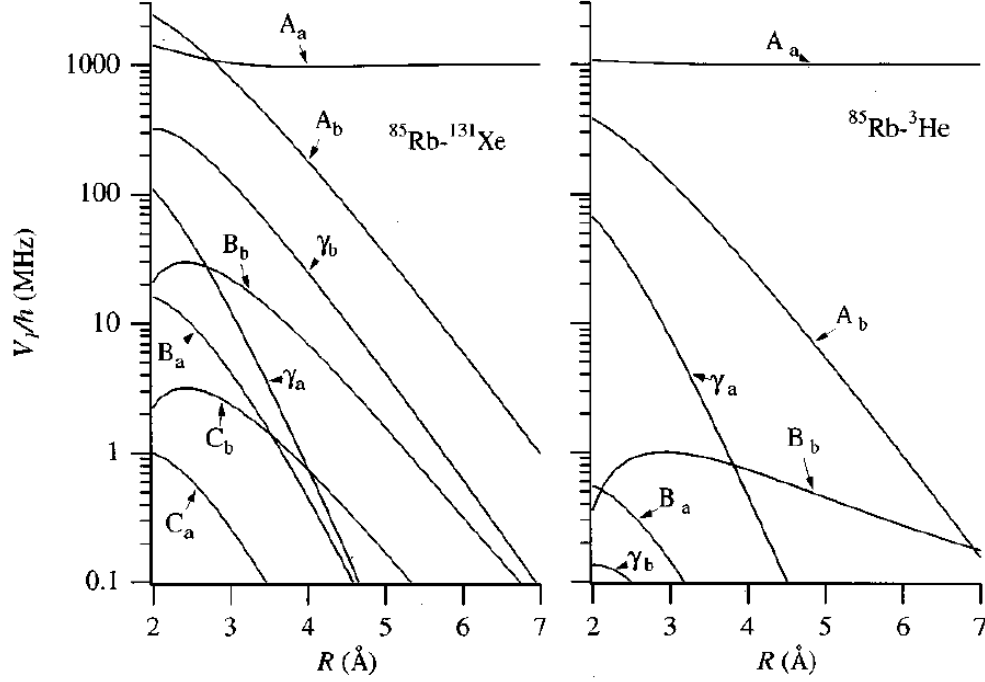


Figure 1.7: Strengths of various spin-dependent interactions as functions of separation (from Ref. [?])

where  $\gamma$  is the coefficient of the spin-rotation interaction, while  $A_k$ ,  $B_k$ ,  $C_k$  are the coefficients for isotropic magnetic-dipole hyperfine interactions, anisotropic magnetic-dipole hyperfine interactions, and electric quadrupole interactions, respectively.  $A_a$



greatly exceed other coefficients as the separations between atoms increase.

The isotropic hyperfine interactions come from the Fermi-contact magnetic fields of the nuclei pair.

$$A_b(R) = \frac{8\pi g_s \mu_B \mu_b}{3I_b} |\eta \phi_0(R)|^2 \quad (1.22)$$

where  $\eta$  is the enhancement factor which equals to the ratio of the perturbed wave function at the noble gas nucleus to that without the noble gas atom. The isotropic hyperfine interaction also introduces a frequency of the magnetic resonance lines for alkali-metal and noble gas atoms. The frequency shift of alkali-metal electrons due to the polarized noble gas nuclei is used in the technique Electron Paramagnetic Resonance (EPR) to calculate the polarization of noble gas nuclei.

The isotropic magnetic-dipole coupling polarizes the noble gas nuclei parallel to the electron spin polarization, while the anisotropic magnetic-dipole coupling polarizes in the opposite direction. Fortunately, the anisotropic interaction is negligible compared to isotropic interaction.

### 1.3.2 Spin-Exchange Rate

# Bibliography

- [1] E. M. M. V. Romalis and G. D. Cates. Pressure broadening of rb d<sub>1</sub> and d<sub>2</sub> lines by <sup>3</sup>he, <sup>4</sup>he, n<sub>2</sub>, and xe: line cores and near wings. *Phys. Rev. A*, 56(6), 1997.

# Appendix A

## Appendix title

This is Appendix A.

You can have additional appendices too (*e.g.*, `apdxb.tex`, `apdxc.tex`, *etc.*). If you don't need any appendices, delete the appendix related lines from `thesis.tex` and the file names from `Makefile`.