Temperature Optimization for Spin-Exchange Optical Pumping of ^{3}He Target Cells

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Abstract

ABSTRACT

Free neutrons have a lifetime of approximately fifteen minutes, so experiments investigating neutron structure must utilize an alternative target. The polarized ^{3}He target offers a viable effective neutron target characterized by its short spin-up time and its high neutron polarization. The spin-exchange optical pumping (SEOP) technique is a robust method of polarization which is well suited for long term operation with low spin destruction rates. For this study, circularly polarized laser light was used to atomically polarize a mixture of Rubidium and Potassium. The alkali metals then interact with the ^{3}He nuclei, producing a polarized effective neutron target. Several factors effect the polarization and spin-up time including laser power, relative densities of the cell, and pumping chamber temperature. The temperature of the pumping chamber is dictated by an oven. The oven plays a large role in determining the maximum polarization of the ^{3}He, as well as the spin-up time. By controlling the temperature, the oven is able to dictate the amount of Rb vapor within the pumping chamber. The purpose of this study was to optimize the target oven temperature to provide maximum ^{3}He polarization. Nuclear magnetic resonance (NMR) techniques were used to measure the relative target polarization. An absolute calibration was obtained by monitoring the electron paramagnetic resonance signal of the Rb valence electron. During the experiment, periodic measurements of the polarization using NMR were taken. Once maximum polarization was achieved, EPR was used to calibrate the NMR and the measurements were adjusted, accordingly. Analysis of the data shows the optimal polarization, as well as the spin-up time. It was found that the optimal temperature for minimizing spin-up time and maximizing polarization is 216°C. The optimal temperature is dependent upon the laser power used for optical pumping. This result shows the preferred temperature for 69.2 W of laser power. This will benefit future ^{3}He target scattering experiments by minimizing the down time between target swaps, while maintaining the highest possible level of polarization.

1. Introduction

The Helium three target group is responsible for optimizing polarization of ^{3}He targets for use in scattering experiments at Jefferson lab (Jlab). ^{3}He is an effective Neutron target meaning that it simulates a target of free neutrons. The reason a free neutron target is not viable is because the lifespan of free neutrons in approximately fifteen minutes.

The target lab focuses on studies involving polarization of ^{3}He targets using the spin-exchange optical pumping (SEOP) technique. Over the course of the 2010 summer target lab studies were performed on the target cell called Boris.

Target cells are hand blown glass which are then filled with ^{3}He and an alkali mixture to a pressure of approximately 10 ATM. The ^{3}He target cell consists of three distinct volumes; the pumping chamber, transfer tube, and target chamber (see figure 1). The pumping chamber is where the interaction with the incident laser light takes place. The transfer tube is in place to promote spin exchange. The geometry of the target chamber is designed to be placed in the Jlab beamline. This study will examine the range of oven temperatures that will allow for optimal SEOP. We will examine 180-250 degrees Celsius at ten degree intervals.

1.1 Polarization

When subjected to a magnetic field, the magnetic moment, μ, of a particle will align itself with the field. It will either be aligned parallel or anti-parallel to the direction of the field. The polarization of a sample is defined as the ratio of the difference of particle spin directions over the total number of particles. Under normal thermal distribution, the number of particles with spin either aligned or anti-aligned is given by the Botlzmann distribution of the spin states. This is possible because spin ½ particles have only two possible quantum mechanical orientations, spin up and spin down by

convention. We utilize SEOP to overcome the thermal distribution and polarize the majority of spins in one direction. Here N_+ is the number of spins aligned with the holding field, N_- is the number of spins anti-aligned, and E_+ and E_- are the energies of the states respectivly.

$$
\begin{aligned}
N_+ &= N \cdot e^{-\frac{E_+}{kT}} \\
N_- &= N \cdot e^{-\frac{E_-}{kT}} \\
P &= \frac{N_+ - N_-}{N_+ + N_-}
\end{aligned}
$$

In the case of the ^{3}He target we are measuring the polarization of the magnetic moment of the ^{3}He nucleus which is a composite of two protons and a neutron. Looking at the definition of polarization we can see that if the spins are equally distributed between the aligned and anti-aligned states there will be zero polarization whereas if all the spins are in one state there will be a polarization of one.

1.1.1 Spin-up

Polarization as a function of time is defined to be exponential and follows the curve

$$
P(t) = P_{max}(1 - e^{-t/\tau}) \tag{2}
$$

Pmax is the maximum attainable polarization and τ is the spin-up constant. The spin-up constant τ dictates the time required to reach maximum polarization less the maximum polarization over e. An example of Spin-up can be seen in figure 8.

1.1.2 Spin-Down

Spin down describes the natural polarization decay rate. It is polarization as a function of time without any external influences such as laser or oven. In other words there is no polarization process underway when describing the spin-down curve.

$$P(t) = P_{max} e^{-t/\Gamma} \tag{3}$$

Pmax is still the maximum polarization but in the case of decay it is also the initial polarization. Γ is the spin-down constant and describes the time it takes for the sample to be reduced by a factor of e^{-1}. The spin-down constant is not equal to the spin-up constant.

1.2 Spin-Exchange Optical Pumping

The definition of polarization tells us that the polarization will be proportional to the magnetic field and inversely proportional to the temperature. The usual brute force method for polarizing a sample is to increase the field strength while decreasing the temperature of the target cell. The target lab utilizes a more elegant approach to polarization known as optical pumping. Optical pumping depends on an alkali metal and spin-exchange to successfully polarize the sample. Within the target cell there is a Rubidium-Potassium mixture, Nitrogen to act as a buffer, and ^{3}He for the effective neutron source.

Alkali metals are chosen to interact directly with the incident laser light because they carry one valence electron. By tuning the frequency of the incident light we are able to excite the valence electron up a level and manipulate its spin.

The process begins by placing the pumping chamber within the ceramic oven and heating. This is essential because the Rb must be in vapor form if it is to interact with the laser light. In other words, the hotter the oven, the more Rb vapor and consequently the more absorption that takes place. This is only true to a certain optimal density and is what we determined with in this study.

Next laser light must circularly polarized carrying. We then tuned the frequency of the laser light to match the energy splitting of Rb. The Rb will be able to absorb the light bringing its valence electron from the $5S_{1/2}$ state to the $5P_{1/2}$ state. Circularly polarized light carries ± 1 unit of angular momentum. This restricts the states the Rb valence electron can absorb a photon from. Only from the

$5S_{-1/2}$ state can it absorb the photon taking the electron to the excited $5P_{+1/2}$ state. From the $5P_{+1/2}$ it will decay to the $5S_{1/2}$ emitting a photon. If the electron drops to the m=-1/2 state the process will repeat until it eventually gets stuck in the m=+1/2 state (see figure 3).

This process will allow the valence electrons of the Rb to collect in the $5S_{1/2}$ state. The Rb valence electron then interacts with the ^{3}He nucleus, and through spin exchange collisions dictates, the spin of the nucleus. Because of the Pauli Exclusion Principle the ^{3}He protons must have opposite spins, therefore canceling. This results in the net spin of the nucleus being determined solely by the neutron. Therefore by controlling the spin of the Rb valence electrons, we are able to manipulate the spin of the ^{3}He nucleus.

2. Materials

2.1 Optics

The current optics setup in the ^{3}He target lab contains three tunable broad bandwidth Coherent diode lasers. Each laser has been tuned to maximize absorption in the Rb. This means that we have adjusted the temperature of the photo diode and the current so that the output light is as near to 795nm, (known as D1 light) as possible. D1 light is the frequency required for the Rb valence electron to absorb the photon moving from the $5S_{1/2}$ to $5P_{1/2}$. When correctly tuned, the absorption spectrum will appear with a large dip in the center, these are the wavelengths the Rb are absorbing.

This dictates the need for high power output lasers. After tuning each laser and taking power measurements, we were operating at 30A each of diode current, and found the total laser power incident on the pumping chamber to be 69.2W (see table 1).

The optics setup is shown in figure 2. Every laser is connected via a fiber optic cable to a mount, where it enters an optical line. A set of optics for a singular laser is known as a line. We used three lasers which passed through separate optical lines. Each line contains an identical optic setup.

The first element is a focusing lens which directs the light into a beam splitter. This linearly polarizes the light and splits the beam into two separate beams. One beam passes through to a quarter wave-plate circularly polarizing the light. The other beam passes through a quarter wave-plate, reflects off a mirror, and back through the quarter wave-plate. This puts the beam 180 degrees out of phase, and therefore allows it to pass back through the beam splitter undeterred and through a quarter wave-plate, circularly polarizing the beam. The net result is two beams per laser, each circularly polarized, for a total of six beams.

2.2 Polarized Target System

The target cell is positioned so the pumping chamber is located within the ceramic oven and pick up coils placed on either side of the target chamber. Around the oven were placed two perpendicular sets of Helmholtz coils allowing the holding field to be directed in any direction along the horizontal plane. Perpendicular to the Helmholtz coils are the NMR RF coils positioned so the field is vertical. The EPR coil is located in the horizontal plane of the Helmholtz coils with the EPR diode positioned above the pumping chamber. Setup is shown in figure 4.

The current configuration of the target setup is a longitudinal one. This means that the holding field generated by the Helmholtz coils is directed in the same direction as the incident laser light.

3. Methods

3.1 Polarimetry

3.1.1 Nuclear Magnetic Resonance

Nuclear Magnetic Resonance (NMR) is one technique used for measuring the polarization of

the target. Due to the presence of the holding field generated by the Helmholtz coils the magnetic

moment of the nuclei will precess about the holding field at the Larmor frequency (see figure 5). The

Larmor frequency is directly proportional to the strength of the holding field and is described by

$$\omega = \gamma * H \qquad (4)$$

where γ is the gyromagnetic ratio constant and H is the holding field.

Perpendicular to the holding field is the magnetic field generated by the RF coils. The strength

of the RF field is periodic at a radio frequency dictated by the RF coils. The power supply of the RF

coils is connected to a function generator, which allows for the frequency of the RF field to be ramped.

Resonance occurs when the frequency of the RF field is equal to the frequency the magnetic

moment is precessing about the holding field, the Larmor frequency. The occurrence of resonance

induces a spin-flip in the nuclei. They will flip from the aligned state to the anti-aligned state or vice

versa.

3.1.1.1 Spin-flip Classical Description

The equation of motion of spin, J in a magnetic field, H is

$$d\vec{J}/dt = \vec{M} \, x \, \vec{H}_o \qquad (5)$$

where M=γJ. M is precessing about the holding field at the Larmor frequency ω=γH.

$$d\vec{M}/dt = \vec{M} \, x \, \gamma \, \vec{H}_o \qquad (6)$$

When we introduce the vertical RF field, the equation for the magnetic moment's motion becomes

$$d\vec{M}/dt = \vec{M} \, x \, \gamma \, (\vec{H}_o + \vec{H}_1) \qquad (7)$$

where the field H_1, broken into vector components, is rotating about the z axis at frequency ω.

$$\vec{H}_1 = H_1(\hat{i} \cos(\omega t) + \hat{j} \sin(\omega t)) \qquad (8)$$

By placing our coordinate system in the rotating frame, dictated by the magnetic moments

natural frequency about the z axis, we can determine the effective field.

$$d\vec{M}/dt = (d\vec{M}/dt)_r + \vec{\omega} \times \vec{M} \qquad (9)$$

$$d\vec{M}/dt = \vec{M} \times \gamma(\vec{H}_o + \vec{H}_1) + \vec{\omega} \times \vec{M} \qquad (10)$$

$$d\vec{M}/dt = \vec{M} \times ((\omega_o - \omega)\hat{k} + \omega_1 \hat{i}') \qquad (11)$$

This tells us our effective field is

$$H_{eff} = ((\omega_o - \omega)\hat{k} + \omega_1 \hat{i}')/\gamma \qquad (12)$$

It also tells us the angle the magnetic moment is precessing about the z axis at

$$\tan\theta = \omega_1/(\omega_o - \omega) \qquad (13)$$

There are two methods to satisfy the resonance criteria, the field sweep and the frequency sweep. The field sweep holds the frequency of the RF coils constant and sweeps the strength of the holding field up and then down. By changing the strength of the holding field the Larmor frequency is changing. At a point on the sweep up the Larmor frequency will equal the RF frequency and the spin flip will occur sending, angle θ from 0 degrees to 180 degrees. The strength of the field then sweeps back down, through resonance and another spin flip occurs.

The frequency sweep involves keeping the strength of the holding field constant and changing the frequency of the RF coils. The frequency of the RF coils is swept up until it matches the Larmor frequency, and a spin flip occurs.

To measure the spin flip, pick up coils are located on either side of the target chamber. When a spin flip occurs, an EMF will be created, inducing an EMF current in the pick up coils. This current is sent to a pre-amplifier to enhance the signal and then to a lock-in amplifier to pick the signal out from the noise. The strength of the induced current is proportional to the number of spin flips taking place which is in turn proportional to the polarization of the ^{3}He.

The benefits of NMR include a low loss rate per measurement and fast results. Unfortunately NMR only gives a relative polarization. It must be calibrated by either using an EPR result or doing a

water cell calibration.

3.1.2 Electron Paramagnetic Resonance

Electron Paramagnetic Resonance (EPR) is absolute. EPR is a really a three step process involving the EPR coil, photo diode, and the NMR setup. Where NMR works by measuring an electrical signal, EPR works by looking at the light emitted by the Rb's valence electrons. The light's frequency is dependent upon the Zeeman splitting taking place between electron energy levels. Zeeman splitting occurs in the presence of a constant magnetic field, therefore the holding field is the major contributor to the Zeeman splitting. The alignment of the polarized ^{3}He nuclei also create a magnetic field. Although its contribution is smaller than that of the holding field, it is measurable. The greater the Zeeman splitting the greater energy required to move between energy levels. The light emitted by the electron as it moves between levels is a comosite of, the D1 light, as well as the separation due to the Zeeman splitting. The Zeeman splitting may then be split into the portion due to the holding field and that due to the polarized ^{3}He nuclei. We start by looking at the energy of the photon. E is the photon's energy and ν is it's frequency.

$$E = h\nu_i \rightarrow E = h(\nu_{D1} + \nu_z) \tag{14}$$

The energy is the energy to travel between energy levels plus the energy to travel across the Zeeman splitting.

$$\nu_z \propto \nu_B + \nu_{3\text{He}} \tag{15}$$

$$\nu_{3\text{He}} \propto P_{3\text{He}} \tag{16}$$

The goal is to isolate the contribution to the frequency due to the ^{3}He. To accomplish this we have to take a closer look at the energy levels of the Rb valence electron (see figure 6). For right circularly polarized light the majority of the Rb valence electrons work their way into the F=3 M=3 state.

While in the M=±3 state the Rb valence electron is unable to absorb a photon because of the angular momentum selection rules. To manipulate the electron an EPR coil is used which is simply an RF coil. An FM sweep is performed where the EPR coil scans until the proper frequency is found which will incite the electron from the M=3 state to the M=2 state. From the n=5P, F=3, M=2 state it can drop down to the n=5S, F=2, M=2 state emitting a photon.

This light is measured by a photo-diode. When a max intensity is found the EPR coil will lock in at this frequency allowing for a strong signal as the electrons transititon between states.

In order to find the frequency of the light due to the polarized ^{3}He nuclei, the diode measures the light's frequency at max intensity. The NMR process is then utilized to induce a spin flip in the electrons. By flipping the spin the magnetic field due to the ^{3}He is now detracting from the total magnetic field contributing to the Zeeman splitting whereas before it was adding to it (see figure 7).

After the spin flip takes place, the Diode then measures the new frequency of the light. The new frequency is now

$$E = h\nu_f \rightarrow E = h(\nu_{DI} + \nu_z) \tag{18}$$

where

$$\nu_z \propto \nu_B - \nu_{3He} \tag{19}$$

By taking the difference between the initial frequency and the final, we can isolate the frequency due to the ^{3}He.

$$\nu_i - \nu_f = \nu_{DI} + \nu_B + \nu_{3He} - \nu_{DI} - \nu_B + \nu_{3He} \tag{20}$$

$$\nu_i - \nu_f = 2\nu_{3He} \tag{21}$$

Because the frequency is proportional to the polarization of the ^{3}He, we can now know absolutely the number of polarized ^{3}He nuclei within the target cell.

4. Experiment - Temperature Optimization

We examined the effects oven temperature had on the maximum polarization of the target cell as well as the spin-up time. The optimum temperature for these characteristics depends on the laser power used for optical pumping. We were operating at 30A on all three Coherent lasers with a total laser power of 69.2 W. This puts us in a lower spectrum of temperatures where the maximum polarization will occur than if we had more laser power. To find this value in conjunction with its spin-up time, we looked at the range of temperatures from 180°C-240°C.

Temperature has a large effect on the maximum polarization of the cell. Optical pumping is used to polarize the Rb which then interacts with the ^{3}He, the more vapor Rb present in the cell the more absorption that takes place. This would lead us to believe the higher the temperature the higher the polarization. This is only true to a certain point however. There comes a temperature where all of the laser light is being absorbed by the Rb and depolarizing effects begin to play a larger role.

Higher temperature means the atoms have more energy, increasing the number of interactions taking place within the cell. At higher temperatures the depolarizing effects due to wall collisions, interactions between ^{3}He, Rb, and K, and dipolar interactions become significant. Once that point is reached and the temperature continues to rise the maximum polarization will decrease. Our task is to find the optimum temperature where 69.2W of laser power will allow for high polarization before the depolarizing effects become too significant.

We began by setting the oven to the desired temperature and depolarizing the cell. With the polarization of the cell at a minimum we would then turn on the lasers and take NMR measurements periodically as we polarized the cell (spin-up).

4.1 Data Point Corrections

4.1.1 EPR Calibration Constant Correction

Before we can use the NMR signal and consider it accurate we need to make two essential corrections, the EPR calibration and the Adiabatic Fast Passage (AFP) loss. Because the NMR measurements are relative, we need to perform an EPR calibration for each oven temperature. We performed the calibration after we took all the data points and the polarization was near max. This gives us a new calibration constant was then applied to each point giving us the correct measured polarization for the temperature.

$$P_n^m = C_t / C_o P_n \tag{22}$$

where C_t is the calibration constant for the specific temperature and C_o is the calibration constant used to take the measurement.

4.1.2 EPR Calibration Constant Temperature Correction

The calibration constant correction may be theoretically calculated, if we exploit the fact that the number of atoms in the cell is a constant. We can use the ideal gas law to find the number of atoms in a particular portion of the cell.

$$PV = nRT \tag{23}$$

$$n_i = PV_i / RT_i \tag{24}$$

The sum over the three section of the cell, pumping chamber, transfer tube, and target chamber, will yield the total number of atoms in the cell.

$$N = \sum_1^3 n_i \tag{25}$$

Then we can isolate the number of atoms in the target chamber.

$$P = NR / (\sum_1^3 V_i / T_i) \tag{26}$$

$$n_i = PV_i / RT_i = NV_i / (T_i \sum_1^3 V_i / T_i) \tag{27}$$

Because our pick-up coils are located over the target chamber, the calibration constant is
proportional to the number of atoms in the target chamber.

$$C_t = D n_i \qquad\qquad (28)$$

$$P_n^m = D n_i / D n_{io} P_n \qquad\qquad (29)$$

$$n_i / n_{io} = (1/(T_i \sum_1^3 V_i / T_i)) / (1/(T_{io} \sum_1^3 V_{io} / T_{io})) \qquad\qquad (30)$$

Because the summations are over the volume of the cell, the temperature terms are the
temperatures in the specific volumes of the cell. This gives us a scaling factor relating the actual
calibration constant to the one used to take the measurement.

4.1.3 AFP Loss Correction

Adiabatic Fast Passage is the process of the nuclei's magnetic moment flipping from zero to one
hundred eighty degrees. Everytime this happens a certain percent of the total polarization is lost.
Before we could apply the correction we needed to find out what our AFP loss was per-measurement.
To accomplish this we turned off the lasers when we were near max polarization and took ten NMR
measurements, one every ten minutes. We then re polarized to near max and took a single NMR
measurement. Ninety minutes later we took another NMR measurement. The line designated by these
two points is known as the base line. It is the polarization loss due to natural spin-down without the
influence of measurements. Fitting the ten points we found a loss per measurement. By subtracting the
slope of the base line from the fitted slope we found the polarization we lost per measurement.

The goal is to find a percentage lost per-measurement so we know how much to correct each
individual measurement by. To do this we took an average of our loss per-measurement divided by the
measurement.

$$AFP = 1/N \sum_0^N 1/P_n * (dP_n/dn - m_{base\,line}) \qquad\qquad (31)$$

This value of AFP is a percentage of the polarization that was lost by taking the NMR measurement. For example, on our first measurement we lost AFP percent of the actual polarization.

To correct the nth measurement we utilized the formula.

$$P_n^A = P_n^m / (1 - AFP) + \sum_0^{n-1} (AFP) * P_n^A \qquad (32)$$

Here P^a is the actual polarization P^m is the measured polarization. By using these two corrections we were able to find the actual polarization for each data point. Plotting the data points as a function of time, we performed a nonlinear regression to equation 2.

The constants in this equation tell us everything of interest from the particular oven temperature. We then repeated this process for each oven temperature until we were able to get a curve of Pmax as a function of temperature and spin-up as a function of temperature.

5. Results

Throughout the course of the experiment all NMR measurements were taken with incorrect EPR calibration constants. Although the process of obtaining calibration constants was utililzed, only one successful EPR calibration was performed. The successful calibration was performed at 180°C.

Using the wrong calibration constant resulted with inaccurate polarization readouts during the NMR measurements. To correct the measurements we utilized two separate corrections. The primary correction was made by using the calibrated constant ratio and scaling all polarizations to one single constant. The 180°C constant was the only reliable C_t. The process of bringing all of the polarizations to the 180°C constant means a loss of absolute polarization. In other words only differences in polarizations are meaningful.

After correcting the polarizations to be relative, the temperature correction was used. As previously stated, the ratio of nuclei in the target chamber at the temperature the measurements were

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made at over the temperature the calibration constant was taken at was applied.

Finally the AFP loss was taken into account to correct for the polarization lost during the spin-flips of the NMR measurements.   The three corrections in conjunction with each other corrected each data point properly so that they could be plotted and analyzed using a nonlinear regression. The data can be seen in table 2, and figure 9.

It is apparent that 210°C was able to reach the highest relative maximum polarization. To find a rough approximation of the behavior of the data a quadratic fit was applied. The fit resulted in

$$Relative\ P_{max}(T) = -260.52 + 2.5588T - 0.0059095T^2 \tag{33}$$

Setting the derivative equal to zero and solving reveals that the optimal temperature for polarizing ^{3}He using SEOP at 69.2W of laser power is about 216°C.

6. Conclusion

While conducting spin-exchange optical pumping utilizing 69.2W of laser power we found that the temperature best suited for attaining maximum polarization is 216°C. From the data, it also appeared that increasing the temperature decreases the spin-up time. However, a compromise must be struck between the maximum achievable polarization, and the time it takes to reach that polarization.

This experiment introduced several further questions which should be looked into more extensively in the future. How does the laser power impact the temperature where maximum polarization is found? Does the technique explored in this study for correcting calibration constants offer a theoretical alternative to using the EPR system to calibrate the NMR system everytime a change is made to the setup?
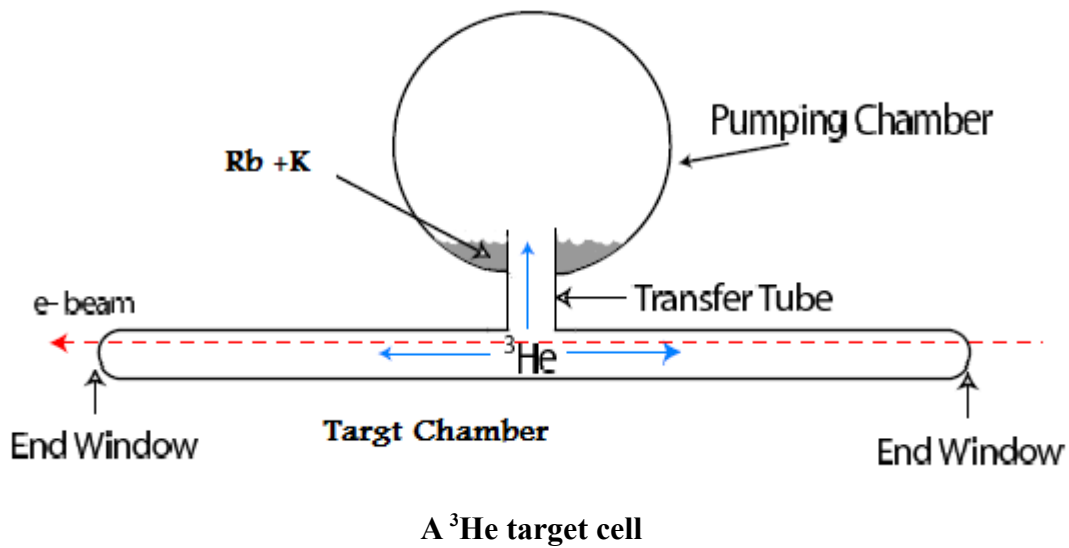
7. Figures
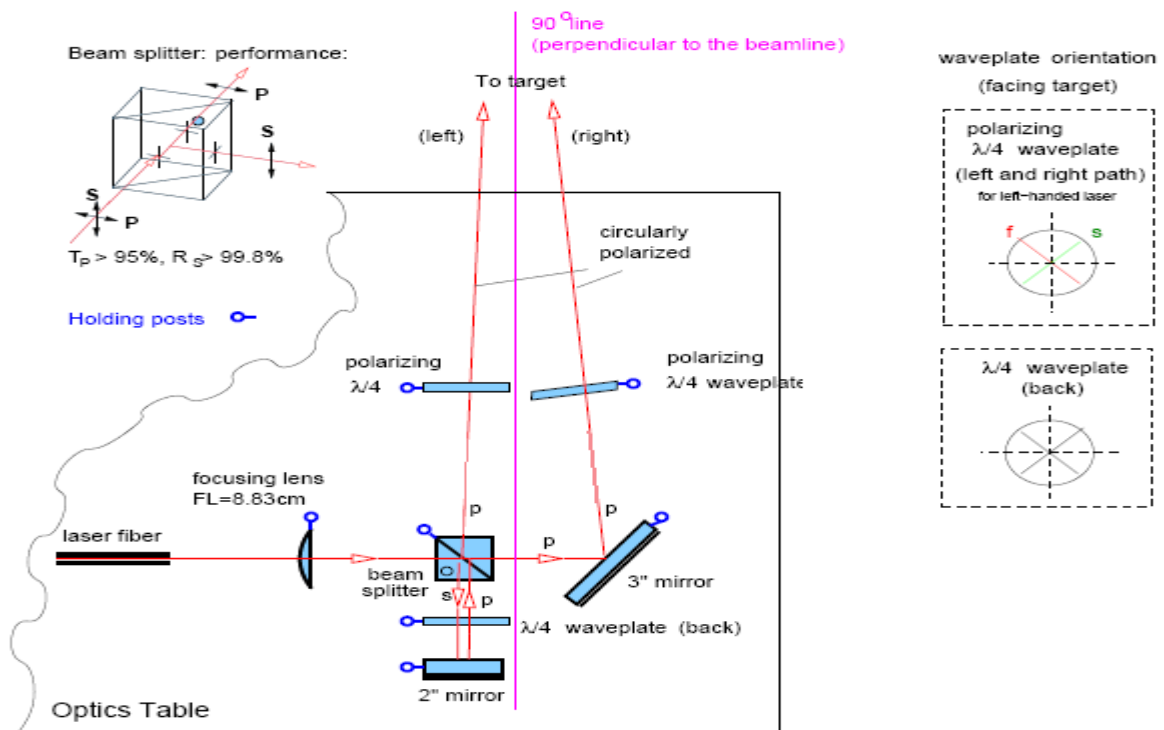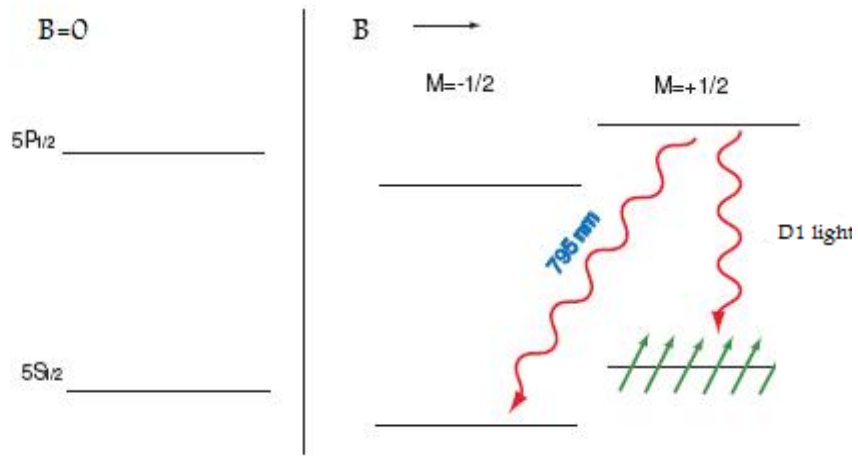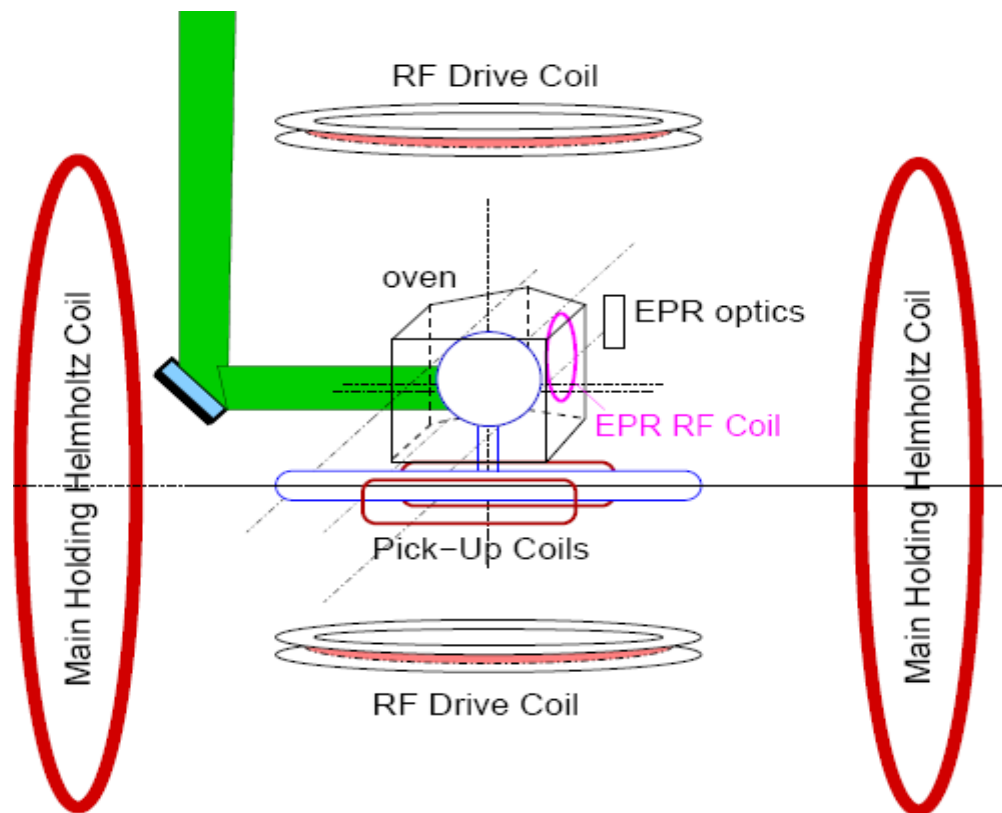
Figure 1

A ³He target cell

Figure 2

Figure 2 shows the optical components of one line from a birds eye view. The three lines are stacked vertically on top of each other.

Figure 3

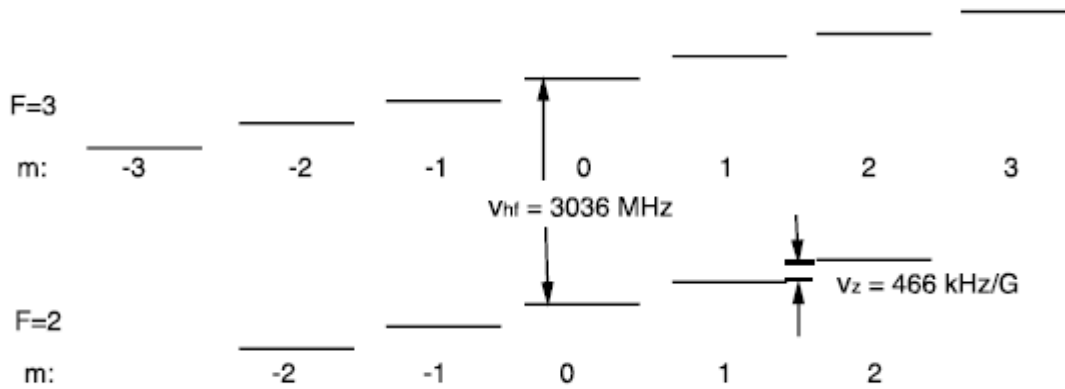**Splitting of the energy levels of the Rb valence electron**

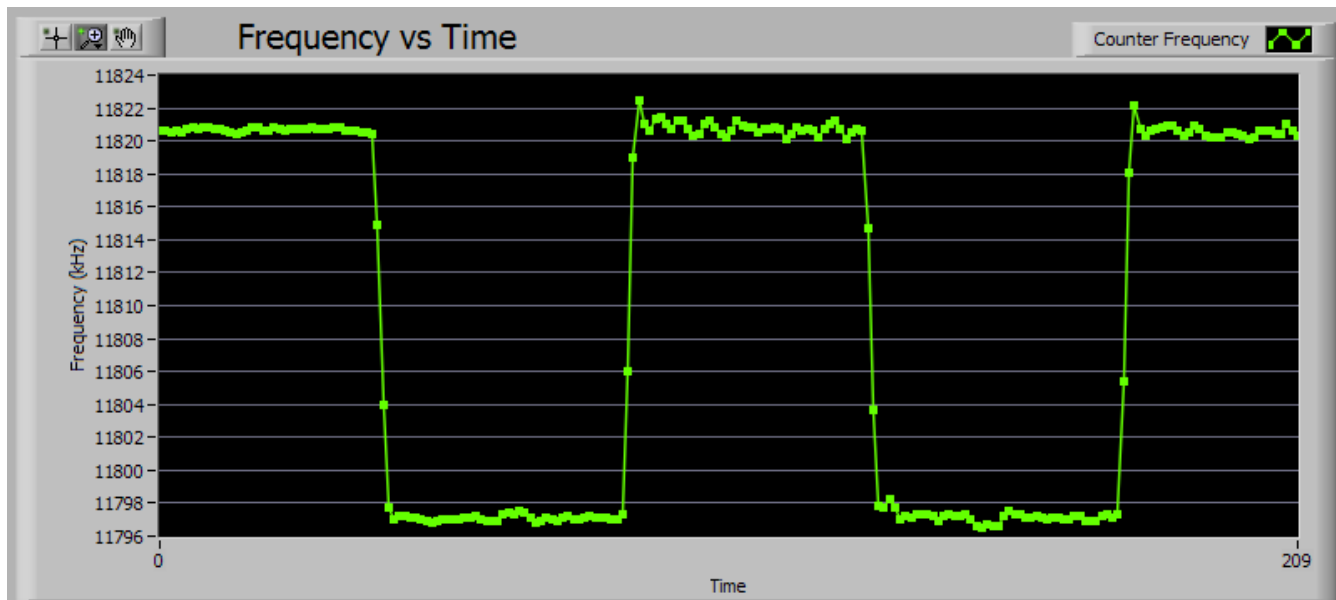**Figure 4**

Target area setup

Figure 5

**magnetic moment of a particle precessing about the holding field**

Figure 6

Splitting of the F sub-levels of Rb valence electron including separation due to Zeeman splitting

Figure 7

$$\nu_B + \nu_{3He} \qquad \nu_B - \nu_{3He} \qquad \nu_B + \nu_{3He} \qquad \nu_B - \nu_{3He} \qquad \nu_B + \nu_{3He}$$
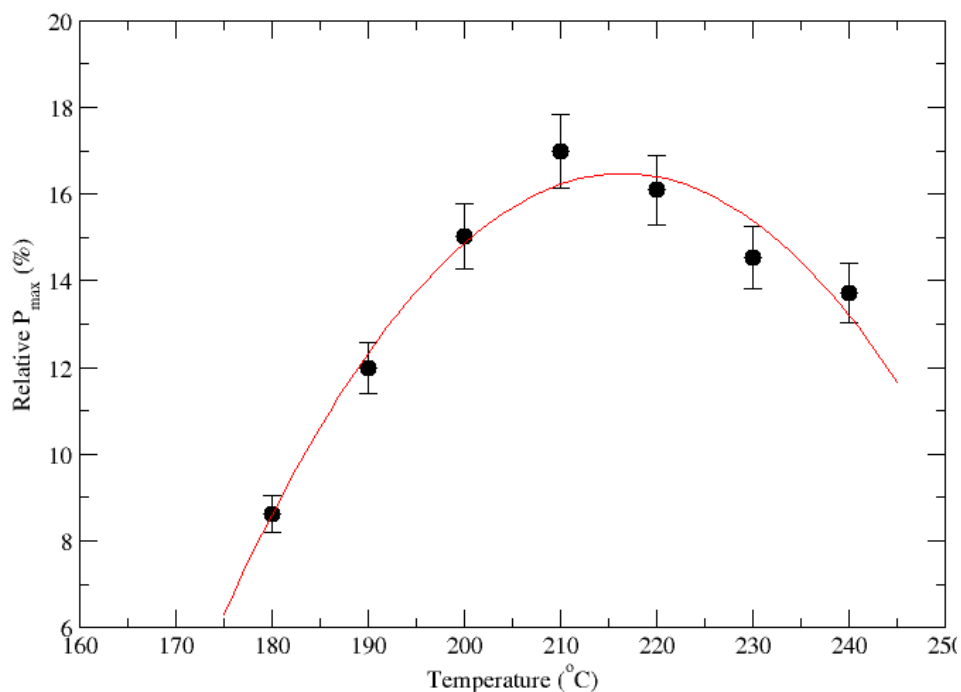
Frequencies measured by photo-diode. Before spin flip the frequency due to the ³He is augmenting the frequency due to holding field. After flip it is detracting from it.

Figure 8.

polarization vs time 210 redux
$$P = 10.8735 * (1 - EXP(-T / 11.5628))$$
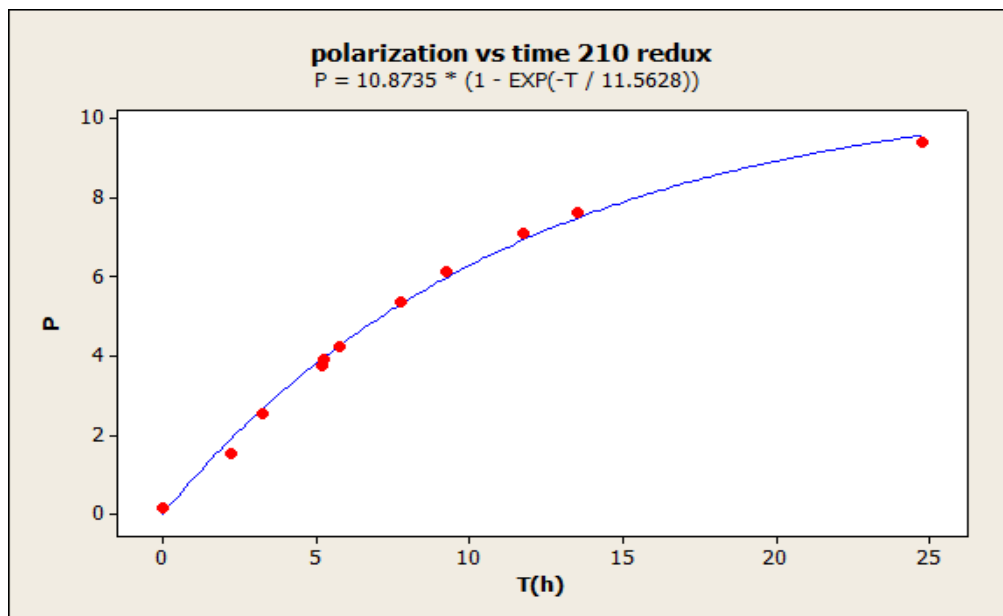
Spin-up of the Boris cell. Oven temperature 210°C, laser power 69.2W.

Figure 9.

Relative Polarization as a function of oven temperature

8. Tables

Table 1

Laser	Current (A)	Photo diode temp (°C)	Power (W)	Polarization
Coherent #3	30	17.6	22.2	99.00%
Coherent #4	30	17.4	23.3	98.00%
Coherent #5	30	19.7	23.7	99.00%

Current to the diode, Temperatuer the photo diode is operating at,power of the photons as measured by a power meter, percent of light rotating circularly in the same direction.

Table 2
Time NMR was taken, polarization NMR gave, corrected polarization using the EPR, Temperature, and AFP corrections. Final data for each temperature after performing linear regression.