Progress Towards Cooling of an Atomic Beam of Lithium

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Executive Summary

This experiment aims to measure the $D_1$ and $D_2$ lines, from the ground state to the first excited state, in lithium 6 and 7 to high precision, and by extension, the fine and hyperfine splittings, and the isotope shift. This is achieved by probing an atomic beam of lithium with a diode laser. This probe laser is referenced to an optical frequency comb, which translates optical frequencies to measurable radio frequencies, and is in turn referenced to a GPS steered atomic clock. Thus when the probe laser is locked, we can calibrate its frequency to a few kHz. High precision measurements of these state will help resolve discrepancies in previous measurements, inform the addition of Quantum Electro Dynamic (QED) corrections to calculations of these energy levels, and provide a window into the nuclear structure of lithium.

This experiment has made preliminary measurements of the $D_1$ lines, but various splittings are not self consistent due to issues with the line shape and unresolved peaks. By counterpropigating laser light, red detuned from a resonance, across the atomic beam, we can alter the velocity distribution and narrow the Doppler width of our beam. This process is called transverse cooling. We have made some progress towards this transverse cooling, which we hope will narrow our linewidths and resolve the internal discrepancies in our data, and in addition allow for measurements of the less resolved $D_2$ lines.
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Chapter 1

Introduction

1.1 Motivation

As one of the lightest atoms and the first atom with a closed shell, lithium has been of significant theoretical interest. As a few electron system, the error in atomic structure calculations due to relativistic corrections is small enough that Quantum Electro Dynamic (QED) corrections are significant. With a closed shell, lithium is also the simplest system where interactions between this shell and the valence electron can be studied. In addition the calculation of the isotope shift between $^6\text{Li}$ and $^7\text{Li}$ is limited by how well the charge radius of the nucleus is known. Thus a measurement of the isotope shift is effectively a measurement of this charge radius. With advances in precision measurement and spectroscopy, namely the development of frequency combs and cold atoms, measurements can be made to probe this theory. More extensive information on the interest in precision measurement of lithium can be found in Ref [1], [2], and [3].

So far there have been large discrepancies in the measurements that have been made of these lines as seen in Fig 1.1. As discussed in Ref [6], the discrepancy in measurements of the $D_2$ lines is partially caused by quantum interference between the unresolved hyperfine lines. Our system should allow us to thoroughly investigate
Figure 1.1: Deviation from the weighted mean for previous measurements of (i) the fine structure splitting of $^6\text{Li}$, (ii) the fine structure splitting of $^7\text{Li}$, and (iii) the isotope shift between $^6\text{Li}$ and $^7\text{Li}$. We can see that there are significant discrepancies between the measurements well outside their error bars. Measurements are from (a) Puchalski PRL 113 [4] (Theory), (b) Brown PRA 87 [5], (c) Sansonetti PRL 107 [6], (d) Das PRA 75 [7], (e) Noble PRA 74 [8], (f) Walls EPJD 22 [9], (g) Orth ZPA 273 [10].
this effect in the future by controlling the polarization of the probe beam, and the polarization detected. Quantum interference, however, does not resolve the discrepancy in measurements of the $D_1$ lines, and thus additional measurements of these lines is also necessary.

1.2 History of the Experiment

Progress towards making these measurements have been a long running project in this lab. In 2009 the frequency comb for the project was stabilized, this work is mostly presented in Ref [11]. In the years since there have been a number of changes to the setup of the comb which has improved its functioning significantly, including a new mount for the crystal that allows for easier cleaning and a slightly improved offset frequency detection scheme. The first iteration of the vacuum system and atomic beam had a very wide peak widths as seen in Ref [12]. Do to the large widths in 2009, saturated fluorescence spectroscopy was attempted which would have given Doppler-free profiles. Unfortunately, the small hyperfine splitting causes crossover resonances which made this technique ineffective, as outlined in Ref [13]. Instead we worked on narrowing the width of the peaks, by trying different nozzles for our ovens and different collimation schemes. The widths where narrowed significantly, and preliminary data was taken in 2015, as presented in Ref [14]. This thesis investigates internal inconsistencies within new data taken with a slightly improved system, and presents progress towards transverse cooling of the atomic beam. So far the cooling system has been set up, and we have locked both lasers necessary to achieve cooling, but have not been able to lock close enough to the resonance to see significant changes in the velocity distribution.
Chapter 2

Atomic Physics Theory

2.1 Hydrogen Energy Levels

To better understand the atomic structure of lithium it is useful to recall the structure of hydrogen, before considering the contributions from more electrons and a larger nucleus. A more detailed treatment of hydrogen can be found in Ref [15]. To find the energy levels we start with the time-independent Schroedinger equation in three dimensions,

$$-\frac{\hbar^2}{2m_e} \nabla^2 \psi + V \psi = E \psi,$$

where, if we treat the nucleus as a large mass at zero, our potential is given by

$$V(r) = -\frac{e^2}{4\pi\epsilon_0 r}.$$

When we solve this equation, we end up with eigenfunctions of the form

$$\psi_{nlm} = \sqrt{\left(\frac{2}{na}\right)^3 \frac{(n - l - 1)!}{2n((n + l)!)^3}} e^{-r/na} \left(\frac{2r}{na}\right)^l L_{n-l-1}^{2l+1}(2r/na)Y_l^{m_1}(\theta, \phi),$$

where the $L_{n-l-1}^{2l+1}$'s are Laguerre polynomials, and the $Y_l^{m_1}$'s are spherical harmonics. We say that $n$ is our principal quantum number, $l$ is the angular momentum quantum number.
number, and $m_l$ is the projection of the angular momentum onto the $z$-axis. These eigenfunctions yield energies of

$$E_n = -\frac{m_e}{2\hbar n^2} \left( \frac{e^2}{4\pi\epsilon_0} \right)^2, \quad n \in \mathbb{N}.$$ 

Note that the energy is only dependent on the principal quantum number. Since $l$ can vary from 0 to $n - 1$, and $m_l$ can vary from $-l$ to $l$, both in integer steps, we end up with a degeneracy of $n^2$ for the $n^{th}$ energy level. This degeneracy will be broken when we consider the fine and hyperfine structure as well as electron-electron interactions in higher $Z$ atoms.

### 2.2 Gross Structure of Alkalis

This section loosely follows a treatment similar to those in Refs [16], and [17]. In general we can’t solve Schroedinger’s equation for multi-electron atoms, as electron-electron interactions preclude considering each electron separately. However, we can attempt to understand how the energy levels would shift. Alkali atoms, like lithium, have a closed inner shell with zero spin and orbital angular momentum, this allows for a simplified analysis. The first simplifying assumption we will make is that this closed shell forms a spherically symmetric potential $V(r)$. This is called the central field approximation. At large $r$ we expect that the electrons in the closed shell screen the charge of the nucleus, and the valence electron should see a potential that looks like the usual coulomb potential (for neutral atoms)

$$V(r) \xrightarrow{r \to \infty} -\frac{e^2}{4\pi\epsilon_0 r}.$$ 

At smaller $r$, there is less shielding of the nuclear charge. Thus we expect that levels with higher $l$, where the electron is farther from the nucleus, should have a higher energy than lower $l$ levels. Since the potential is always lower than the hydrogen potential the energies will all be lower than the energies of the associated hydrogen
levels. We can model by the equation
\[ E_{n,l} = -\frac{Ry}{(n - \delta_l)^2}, \]
where \( Ry = 13.6 \text{ eV} \), and \( \delta_l \) is an \( l \) dependent correction to the principle quantum number called the quantum defect. This correction lifts the degeneracy in \( l \) that is apparent in the gross structure of hydrogen. Fig 2.1 shows some of this gross structure of lithium, listing their energies in \( \text{1/cm} \) and the associated wavelengths between them in nm, with data from Ref [18].

2.3 Fine Structure

Relativistic effects shift the energy levels from the previous section though multiple mechanisms. While these shifts fundamentally come out of the Dirac equation we will use perturbation theory, and an ad hoc addition of electron spin to evaluate the effects within the scope of non-relativistic quantum mechanics. A more in depth explanations of fine structure shifts can be found in Refs [16], and [15].

First we will consider the shifts from a relativistic correction to kinetic energy. When we found the energy levels of hydrogen we used
\[ \hat{H} = -\frac{\hbar^2}{2m_e} \nabla^2 + V. \]
The term \( -\frac{\hbar^2}{2m_e} \) represents the classical kinetic energy \( \frac{\mathbf{p}^2}{2m_e} \), in order to do a more accurate relativistic calculation we must consider the relativistic kinetic energy
\[ T = \sqrt{\mathbf{p}^2 c^2 + m_e c^4} - m_e c^2 \approx \frac{\mathbf{p}^2}{2m_e} - \frac{\mathbf{p}^4}{8m_e^3 c^2}. \]
Writing it in this form, as a Taylor series, we can use perturbation theory and take our perturbing Hamiltonian to be
\[ H' = -\frac{\hat{p}^4}{8m_e^3 c^2}. \]
Figure 2.1: A partial energy level diagram for the gross structure of Li. Energies are from Ref [18] and are quoted in units of 1/cm, while the transitions between low lying states are quoted as wavelengths in nm.
2.3. FINE STRUCTURE

It turns out that $n, l, m_l$ are already good quantum numbers for this Hamiltonian, so we do not need to do full degenerate perturbation theory. When applied to hydrogen, this relativistic correction shifts the energies by

$$\Delta E_{n,rel} = -\frac{E_n^2}{2m_e c^2} \left( \frac{4n}{l + 1/2} - 3 \right).$$

This correction shifts $l$ states different amounts, but in multi-electron atoms, does not add any additional splittings.

Out of the Dirac Equation, electrons attain an intrinsic form of angular momentum called spin which we denote as $S$. Spin takes two quantum numbers: $s$, and $m_s$ which represent the total spin, and projection of the spin along the $z$-axis, just as with orbital angular momentum. This spin couples with the angular momentum and adds another fine structure shift.

The spin of an electron is related to its magnetic moment through

$$\mu_e = -\frac{g_e e}{2m_e} S,$$

where $g_e \approx 2$ is the $g$-factor for an electron. In the electron’s frame we have a positive current loop, from the nucleus orbiting around the electron, with a magnetic field of

$$B = \frac{1}{4\pi\varepsilon_0} \frac{e}{m_e c^2 r^3} L.$$

From the interaction between this magnetic field and the magnetic moment of the electron we can add a perturbation to our Hamiltonian

$$H' = -\frac{1}{2} \mu \cdot B = \left( \frac{e^2}{8\pi\varepsilon_0} \right) \frac{1}{m_e^2 c^2 r^3} S \cdot L,$$

where the factor of $1/2$ is a correction from working in a non-inertial reference frame. However, $m_l$ and $m_s$ are no longer good quantum numbers for this Hamiltonian, so we will now consider $J = L + S$ the total angular momentum, and our quantum numbers will be $n, s, l, j, m_j$. Using these quantum numbers for $l \neq 0$ we get energy levels, shifted by the spin orbit coupling, of

$$\Delta E_{n,so} = \frac{E_n^2}{m_e c^2} \left( \frac{n(j(j+1) - l(l+1) - 3/4)}{l(l+1/2)(l+1)} \right).$$
Then adding the terms together we get that the fine structure shifts energy levels by

$$\Delta E_{n,fs} = \frac{E_n^2}{2m_ec^2} \left( 3 - \frac{4n}{j+1/2} \right).$$

Now the energies have split depending on the value of $j$. For a given $l$ we have two options for $j$, either $l + 1/2$ or $l - 1/2$. Now the splitting between these two levels is

$$\Delta E = \frac{E_n^2}{m_ec^2} \frac{4n}{l(l+1)} = \frac{Ry^2}{n^3m_ec^2} \frac{4}{l(l+1)}.$$

When we want to consider the fine structure of the neutral alkali atoms we can modify this equation. First we replace $n$ with $n - \delta_l$ and then multiply by a factor $Z^2$ to account for the increase in nuclear charge. Making these changes we end up with [17]

$$\Delta E = \frac{Z^2Ry^2}{(n-\delta_L)^3m_ec^2} \frac{4}{L(L+1)},$$

where we are now using the total orbital angular momentum $L$, which for alkali atoms makes little difference. For $L = 0$ we have that $J = S$ so there is no splitting, instead there is a third correction called the Darwin Term. This term only affects $s$ orbitals and requires a full relativistic analysis, that I will not go through here but can be found in Ref [16]. In this experiment we consider the $D_1$ and $D_2$ lines, the $D_1$ being the transition from the ground state, to the excited state with $L = 1$, $J = 1/2$, and the $D_2$ being the transition from the ground state to the state with $L = 1$, $J = 3/2$.

2.4 Hyperfine Structure

This section follows the treatment of hyperfine structure found in [16]. Beyond fine structure there are additional splittings in energy levels due to small scale effects from higher order electromagnetic moments of the nucleus. The most significant moments are the magnetic dipole, and electric quadrupole moments.
2.4. HYPERFINE STRUCTURE

Just as the electron has spin, the nucleus can have a spin $I$ which, as with the electron, has a magnetic moment associated with it of

$$\mu_I = \frac{g_I e}{2m_p} I,$$

where $m_p$ is the mass of a proton, and $g_I$ is the nuclear g factor. We will consider the contribution of this magnetic moment to the energy of the system as a perturbation $H'_I$ to the Hamiltonian. Evaluating $H'_I$ we see that it splits into two contributions. The first comes from the coupling between the spin of the nucleus $I$ and the orbital angular momentum $L$ of the electron. We will denote this term $H'_1$ and it comes out to

$$H'_1 = \mu_0 g_I \mu_B \frac{e \hbar}{2m_p r^3} L \cdot I,$$

where $\mu_B = \frac{e \hbar}{2m_e}$ is the Bohr magneton. This term is reminiscent of spin orbit coupling of the fine structure. The second term comes from dipole-dipole interaction between the nucleus and electron and can be written as

$$H'_2 = \mu_0 g_I \mu_B \frac{e \hbar}{2m_p r^3} \left( S \cdot I - 3 \left( \frac{S \cdot r}{r^2} \right) \right).$$

In order to diagonalize $H'_I$ we need to again change our quantum numbers. As with the fine structure, we care about the total angular momentum $F = I + J$, and our good quantum numbers will be $n, L, S, J, I, F, m_F$, (However $I$ will be constant). In the end we get an energy shift of

$$\Delta E_I = \frac{1}{2} \frac{e \alpha}{m_p} \mu^2 \frac{\alpha^2}{m^3} \frac{F(F+1) - I(I+1) - J(J+1)}{J(J+1)(2L+1)},$$

where $\alpha$ is the fine structure constant, and $\mu$ is the reduced mass. Note that the interaction with the nuclear spin further splits the energy levels.

The second significant contribution to hyperfine structure is from the electric quadrupole moment of the nucleus $Q$. The Hamiltonian from this interaction is

$$H'_Q = Q \left( \frac{\partial^2 V_e}{\partial z^2} \right) \left( \frac{3I \cdot J(2I \cdot J + 1) - I^2 J^2}{2I(2I-1)J(2J-1)} \right).$$
and gives an energy shift of

\[
\Delta E_Q = \frac{Q}{4} \left( \frac{\partial^2 V_e}{\partial z^2} \right) \frac{\frac{3}{2}K(K + 1) - 2I(I + 1)J(J + 1)}{I(2I - 1)J(2J - 1)}
\]

where

\[ K = F(F + 1) - I(I + 1) - J(J + 1). \]

Fig 2.2 shows the fine and hyperfine structure of both isotopes of lithium. We see that the hyperfine splittings of the \( D_2 \) lines are less than the natural linewidth, so these peaks are not resolved, this causes the quantum interference issues mentioned in Sec 1.1.

### 2.5 Isotope Shift

Refs [17] and [16] both have similar treatments for isotope shifts. In our derivation of the gross and fine structure we have made the assumption that the mass of the nucleus is infinite, so we have used \( m_e \) instead of the reduced mass

\[
\mu = \frac{m_e A m_p}{m_e + A m_p},
\]

where \( A \) is the mass number and \( m_p \) is the mass of a proton. Using this reduced mass instead of the mass of an electron will result in a shift of a state with energy \( E \) to a new value of

\[
E' = E \frac{A m_p}{m_e + A m_p}.
\]

Now suppose we have two nuclei, one with \( A \) nucleons and the other with \( A + 1 \) nucleons, and consider the same state with base energy \( E \), then energy difference between the isotopes is

\[
\Delta E = E \frac{(A + 1)m_p}{m_e + (A + 1)m_p} - E \frac{A m_p}{m_e + A m_p} = E \left( \frac{(A + 1)m_p(m_e + A m_p) - A m_p(m_e + (A + 1)m_p)}{(m_e + (A + 1)m_p)(m_e + A)} \right)
\]
\[ E = \frac{m_p m_e}{m_e^2 + (4A + 1)m_p m_e + A(A + 1)m_p^2}. \]

This shift is called the normal mass shift, however, this only holds for single-electron atoms. For multi-electron atoms there is an additional shift, called the specific mass shift, since we are no longer in a two body picture which is more complicated and we will not go into here.

In addition to the shift from the mass of the nucleus, there is also a shift from the size of the nucleus, called the volume shift. For small \( r \) we need to consider what the potential is within the nucleus. For a nucleus with radius \( R \), if we assume that the charge density is uniform within the nucleus, the potential looks like

\[
V(r) = \begin{cases} 
\frac{Ze^2}{4\pi\epsilon_0 (2R)} \left( \frac{r^2}{R^2} - 3 \right) & r \leq R \\
-\frac{Ze^2}{4\pi\epsilon_0 r} & r \geq R
\end{cases}
\]

Using this potential we get a shift of

\[
\Delta E = \frac{Z^5 e^2}{\pi\epsilon_0 a_\mu n^3} \frac{R}{5} \Delta R,
\]

where \( \Delta R \) is the difference in radius between the two isotopes and \( a_\mu = a_0 \frac{m_e}{\mu} \). In this experiment we have two isotopes \(^7\)Li and \(^6\)Li. Fig 2.2 also shows the Isotope shift between these two isotopes for the \( D_1 \) and \( D_2 \) transitions, in addition to the fine and hyperfine structure. In particular we see that the isotope shift is on the order of the fine structure splitting for the excited state, this causes the \( D_2 \) line of \(^6\)Li to be overlapped with the \( D_1 \) line of \(^7\)Li which complicates measurement, this overlap can be seen in Fig 4.1.

### 2.6 Zeeman Effect

We follow the treatment found in Ref [19] for the weak field Zeeman effect. The fine and hyperfine structure give us splittings and shifts for isolated atoms, but external fields also shift the energy levels. The first shift we will consider is that from a small
Figure 2.2: The fine, hyperfine, and isotope structure of the $D_1$ and $D_2$ lines of $^6\text{Li}$ and $^7\text{Li}$. Note that the splittings are not to scale. We see that the isotope shift causes the $D_2$ line of $^6\text{Li}$ to overlap with the $D_1$ line of $^7\text{Li}$, and that the hyperfine structure of the $D_2$ line for both isotopes is unresolved. Data from Ref [18].
constant magnetic field. Small means that the shifts are below the level of the fine structure. For this effect we can add a perturbing Hamiltonian

$$H_1 = -\mu \cdot B = -\mu_z B,$$

where we have assumed $B$ is along the $z$-axis. The difficulty comes from finding $\mu$. We will consider the Zeeman effect on the fine structure, so the magnetic moment of our atom will be pointed along $J$. since we know the $g$-factors for $L$ and $S$ are 1 and 2 respectively we can relate the $g$-factor for $J$ to $L$ and $S$, and we end up with

$$g_J = \frac{3J(J + 1) + S(S + 1) - L(L + 1)}{2J(J + 1)}.$$

This means that our energy shift is

$$\Delta E = g_J \mu_B B m_J$$

where the $m_J$ is due to projecting onto the $z$-axis. This shift breaks the degeneracy in $m_J$, and states with different $J$ projections spread apart. These levels are aptly called the Zeeman sublevels.

A similar analysis can be done to find the shift to the hyperfine structure where we take

$$g_F = g_J \left( \frac{F(F + 1) + J(J + 1) - I(I + 1)}{2F(F + 1)} \right).$$

Then we get

$$\Delta E = g_F \mu_B B m_F.$$

### 2.7 Semi-Classical Interaction of Atoms with Light

In this section we will consider a general two level system with states $|1\rangle$ and $|2\rangle$, interacting with light that has an electric field of the form

$$\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t).$$
We follow the treatment found in Ref [17]. Let the resonance of the system be
\( \omega_0 = (E_2 - E_1)/\hbar \). We will assume that the frequency of the light is near resonance
\( |\omega - \omega_0| \ll \omega_0 \), and that the wavelength is much larger than the size of the atom so
that \( |E| \) is constant across the atom, this is called the electric dipole approximation.
To analyze this system we will use time dependent perturbation theory with
\[
H = H_0 + H_1(t)
\]
where \( H_0 \) is the unperturbed Hamiltonian and
\[
H_1(t) = -d \cdot E = e r \cdot E_0 \cos(\omega t)
\]
is the perturbation from the light with \( d = -e r \) being the dipole operator. Now we
look for solutions to the Schroedinger equation
\[
i\hbar \frac{\partial \Psi}{\partial t} = H \Psi.
\]
We will let \( \Psi \) be a linear combination of the two stationary states of \( H_0 \),
\[
\Psi(r, t) = c_1(t) |1\rangle e^{-i\omega_1 t} + c_2(t) |2\rangle e^{-i\omega_2 t}
\]
where \( \omega_1 = E_1/\hbar \), and \( \omega_2 = E_2/\hbar \). Solving for the coefficients we get
\[
i c_1(t) = \Omega \cos(\omega t)e^{-i\omega_0 t} c_2(t)
\]
\[
i c_2(t) = \Omega \cos(\omega t)e^{i\omega_0 t} c_1(t),
\]
where
\[
\Omega = \frac{e \langle 2 | r \cdot E_0 | 1 \rangle}{\hbar}.
\]
Since we are only considering bound states of the electron, \( \Omega \) is real, so we can ignore
complex conjugation of \( \Omega \). We can rewrite the cos above to get
\[
i c_1(t) = \frac{\Omega}{2} \left( e^{i(\omega - \omega_0)t} + e^{-i(\omega_+\omega_0)t} \right)
\]
\[
i c_2(t) = \frac{\Omega}{2} \left( e^{-i(\omega - \omega_0)t} + e^{+i(\omega+\omega_0)t} \right).
\]
Since the laser is near resonance the term $e^{-i(\omega + \omega_0)}$ averages to 0 quickly compared with the other term, so we will neglect it; this is the rotating wave approximation. Then our coefficients become

$$
i\dot{c}_1(t) = \frac{\Omega c_2(t)}{2} e^{i\delta t}$$

$$
i\dot{c}_2(t) = \frac{\Omega c_1(t)}{2} e^{-i\delta t},$$

where $\delta := \omega - \omega_0$ as the detuning of the light from the resonance. If initially an atom is in $|1\rangle$, solving these differential equations allows us to find the probability that at a time $t$ the atom is in $|2\rangle$,

$$|c_2(t)|^2 = \frac{\Omega^2}{\Omega^2 + (\omega - \omega_0)^2} \sin^2 \left( \frac{t \sqrt{\Omega^2 + (\omega - \omega_0)^2}}{2} \right).$$

Here we see that the atoms are oscillating between the two states at a frequency $\Omega$. Note that we are using a semi-classical description where we are treating the light classically, and the atom quantum-mechanically. In addition we have ignored spontaneous emission, which complicates the picture.

### 2.8 AC Stark Shift

This section combines the discussion of AC Stark shifts from Refs [20], [17], and [21]. We would like to be able to look at the energies of our stationary states in the time varying perturbation presented in Sec 2.7. Our semi-classical picture gives us a Hamiltonian which is not diagonal and is time varying. To get rid of this time variation, we must transform into a rotating frame, by defining

$$\tilde{c}_1 = c_1 e^{-i\delta t/2}$$

$$\tilde{c}_2 = c_2 e^{i\delta t/2}.$$

Then we can write our differential equations, in matrix form as

$$i\hbar \frac{d}{dt} \begin{pmatrix} \tilde{c}_1 \\ \tilde{c}_2 \end{pmatrix} = \hbar \begin{pmatrix} \delta/2 & \Omega/2 \\ \Omega/2 & -\delta/2 \end{pmatrix} \begin{pmatrix} \tilde{c}_1 \\ \tilde{c}_2 \end{pmatrix}.$$
CHAPTER 2. ATOMIC PHYSICS THEORY

The matrix here is the Hamiltonian for our rotated picture, but it also represents two states separated by $\hbar(\omega - \omega_0)$, under a perturbation from a static electric field with magnitude $E_0/2$. These two states are the excited state, and the ground state with the added photon energy $\hbar \omega$. We can now use second order perturbation theory to show that our states are shifted by

$$\Delta E \approx \pm \frac{\Omega^2}{4\delta} = \pm \frac{\Omega^2}{4(\omega - \omega_0)},$$

where the “+” is for the higher state and the “−” is for the lower state. If we don’t make the rotating wave approximation, we get an additional term with $(\omega + \omega_0)$, and end up with

$$\Delta E \approx \pm \frac{\Omega^2 \omega_0}{2(\omega_0^2 - \omega^2)}.$$

Unfortunately this doesn’t hold near the resonance as we have assumed a linewidth of zero. Near resonance the shift will go to zero as in Fig 2.3. These shifts are known as both as AC Stark shifts or light shifts.

2.9 Steady State Population

This section combines the treatments found in Refs [21], and [17]. When we consider a collection of $N$ atoms, rather than a single atom it is useful to consider the density matrix, defined as

$$\rho_{nm} = \frac{1}{N} \sum_{i=1}^{N} c_{n}^{(i)} c_{m}^{(i)*},$$

where $c_{n}^{(i)}$ is the coefficient in front of $|n\rangle$ in the $i$th atom’s wavefunction. Using density matrices allows for easier evaluation of observables for ensembles of atoms. In particular, the diagonal elements $\rho_{nn}$ give the expectation value for the fraction of atoms in $|n\rangle$. More information on density matrices can be found in Ref [22]. If we rotate our frame and use $\tilde{c}_1, \tilde{c}_2$, we can define

$$\tilde{\rho}_{nm} = \frac{1}{N} \sum_{i=1}^{N} \tilde{c}_{n}^{(i)} \tilde{c}_{m}^{(i)*}. $$
Figure 2.3: The AC Stark shift for the excited state plotted as the frequency of the shift vs. the detuning of the laser light. We see that it has the expected relationship away from the resonance, and near the resonance the shift goes to zero. Here we have set the natural linewidth to 6 MHz, the natural linewidth of lithium, and have set $\Omega$ to 3 MHz.
This rotation does not change the populations, but will allow us to simplify our expressions. We will model spontaneous emission at a rate $\Gamma \tilde{\rho}_{22}$, where $\Gamma$ is the decay rate or natural linewidth related to the lifetime, $\tau$, of the state through $1/\Gamma = \tau$.

Then we will consider the variables $\tilde{\rho}_{21}$ and $w = \tilde{\rho}_{11} - \tilde{\rho}_{22}$, the difference in population between the ground and excited states. Using these variables, and including the spontaneous decay, our differential equations become

$$\dot{\tilde{\rho}}_{21} = -\left(\frac{\Gamma}{2} - i(\omega - \omega_0)\right) \tilde{\rho}_{21} + \frac{iw\Omega}{2}$$

$$\dot{w} = -\Gamma w - i(\Omega \tilde{\rho}_{21}^* - \Omega \tilde{\rho}_{21}) + \Gamma.$$

If we set the derivatives to 0, we get a steady state population in the upper state of

$$\tilde{\rho}_{22} = \frac{1 - w}{2} = \frac{\Omega^2/4}{(\omega - \omega_0)^2 + \Omega^2/2 + \Gamma^2/4}.$$

In the previous three sections we have assumed an isolated two state system. In reality the picture becomes more complicated when we consider multiple states, and allow for excitation and relaxation between them. In particular this steady state population will be different for different transitions. The next three sections will go through some simple ways we can extend the two state picture to compare the strengths of different possible transitions.

2.10 Transition Rules

Before talking about transition strengths we can look at which transitions are allowed. Since lithium is an alkali atom, the total spin and angular momentum are carried by the valence electron, thus we will simplify the picture by only considering a single electron transitioning. In addition we will only consider transitions induced by dipole radiation. We can rule out some transitions by invoking angular momentum conservation. Consider a transition from $|n, S, L, J, I, F, m_F\rangle$ to $|n', S, L', J', I, F', m'_F\rangle$ where we leave $S$ and $I$ are unchanged since they are not affected by dipole radiation. Since photons carry an angular momentum of 1 we can
use the triangle rule of angular momentum addition to say that
\[ F' - F = 0, \pm 1, \quad \text{and} \quad \text{if } F = 0 \Rightarrow F' \neq 0. \]

Since dipole radiation only changes the orbital angular momentum of the electron we have similar rules for \( L \) and \( J \):
\[
J' - J = 0, \pm 1, \quad \text{and} \quad \text{if } J = 0 \Rightarrow J' \neq 0
\]
\[
L' - L = 0, \pm 1, \quad \text{and} \quad \text{if } L = 0 \Rightarrow L' \neq 0.
\]

In fact, we can make the rules for \( L \) even stricter by considering the parity of states. The dipole operator is odd, so it will only couple states of opposite parity. Otherwise when we take the integral corresponding to the matrix element, we will have an odd integrand which will go to zero; this puts additional restrictions on \( L' \). In the case of lithium, this manifests in \( L' - L \neq 0 \).

In addition to these transition rules, the polarization with respect to the quantization axis of the photon which excites the atoms dictates the change in \( m_F \). First we must decompose the photon’s polarization \( \hat{\epsilon} \) in the spherical basis \( \{ \hat{\epsilon}_{-1}, \hat{\epsilon}_0, \hat{\epsilon}_1 \} = \{ (1/\sqrt{2})(\hat{x} - i\hat{y}), z, -(1/\sqrt{2})(\hat{x} + i\hat{y}) \} \) with respect to the quantization axis, \( z \), then we have.
\[
m'_{F} - m_F = \begin{cases} 
-1 & \text{If } \hat{\epsilon} = \epsilon_{-1} \\
0 & \text{If } \hat{\epsilon} = \epsilon_0 \\
+1 & \text{If } \hat{\epsilon} = \epsilon_1 
\end{cases}.
\]

Later we will find how to evaluate how the electron will transition where \( \hat{\epsilon} \) is not a pure spherical basis element.

These rules are somewhat over-restrictive, as some of these transitions are weakly allowed if we relax some of our assumptions. More explanation of these transition rules, in addition to a discussion of their exceptions can be found in Ref [19]. While these rules help determine which transitions we expect atoms to be able to make, they don’t give the relative amplitudes of allowed transitions. To get relative transition amplitudes we need to manipulate the matrix element, which requires the Wigner-Eckart Theorem.
2.11 Wigner-Eckart Theorem

The Wigner-Eckart Theorem is useful for calculating matrix elements of tensor operators, a proof and additional explanation can be found in Ref [23]. Consider $T^{(k)}_q$, a spherical tensor of rank $k$. We will see that the spherical components $q$ are related to the projection of the angular momentum $m_F$ and $k$ is related to the total angular momentum $F$. Before we state the theorem we must go through some notation related to adding angular momenta. First we recall that the decomposition of angular momenta into possible constituents invokes the Clebsch-Gordan coefficients. For example decomposing an $F$ state into its $J$ and $I$ components yields [19].

\[ |Fm_F\rangle = \sum_{m_J,m_I} \langle Jm_Jm_I|Fm_F\rangle |Jm_J\rangle |Im_I\rangle \]

where $\langle Jm_Jm_I|Fm_F\rangle$ are the Clebsch-Gordan coefficients. Without proof the Wigner-Eckart Theorem says that the matrix element $\langle \alpha', F', m'_{F}|T^{(k)}_q|\alpha, F, m_F\rangle$ can be written as

\[ \langle \alpha', F', m'_{F}|T^{(k)}_q|\alpha, F, m_F\rangle = \langle Fm_Fkq|F'm'_{F}\rangle \frac{\langle \alpha'F'||T^{(k)}||\alphaF\rangle}{\sqrt{2F+1}} \]

where $\langle \alpha'F'||T^{(k)}||\alphaF\rangle$ is called the reduced matrix element. This is significant since $\langle \alpha'F'||T^{(k)}||\alphaF\rangle$ does not depend on $m_F$ or $m'_{F}$. This allows us to look at ratios of matrix elements between Zeeman sublevels.

2.12 Relative Transition Amplitudes

A more in depth discussion of transition amplitudes, as well as the quantum angular momentum theory used in this section can be found in Refs [20], [19], and [24]. From the Wigner-Eckart theorem we have the tools necessary to look at the relative strengths of transitions between Zeeman sublevels. If we consider each transition in our two state picture, the only difference between transitions is the matrix element

\[ | \langle 2|er \cdot E_0|1 \rangle |^2. \]
We can simplify this further if we consider \( E_0 = E_0 \hat{e} \) where \( \hat{e} \) is the direction of the polarization, and we can reintroduce the dipole operator \( d = -e \mathbf{r} \), then pulling \( E_0 \) out, so the matrix element we care about becomes

\[
| \langle 2 | d \cdot \hat{e} | 1 \rangle |^2.
\]

Now, first working in the fine structure, we will let \( |1\rangle = |\alpha, J, m_J\rangle \) and \( |2\rangle = |\alpha', J', m'_J\rangle \), where \( \alpha = \{n, S, L\} \), \( \alpha' = \{n', S', L'\} \), since lithium has a filled shell the total angular momentum and spin are the same as the electron’s angular momentum and spin. To apply the Wigner-Eckart Theorem we need to decompose our operator into the spherical basis

\[
\langle \alpha', J', m'_J | d \cdot \hat{e} | \alpha, J, m_J \rangle = \sum_q (-1)^q \langle \alpha', J', m'_J | d_q \epsilon_{-q} | \alpha, J, m_J \rangle.
\]

Then applying the theorem to \( d_q \) recalling that that our rank is \( k = 1 \) we get

\[
\langle \alpha', J', m'_J | d \cdot \hat{e} | \alpha, J, m_J \rangle = \frac{\langle \alpha', J' || d || \alpha, J \rangle}{\sqrt{2J' + 1}} \sum_q (-1)^q \epsilon_{-q} \langle J, m_J, 1, q | J', m'_J \rangle.
\]

If we only consider transitions with the same \( \alpha, \alpha', J, \) and \( J' \), only the Clebsch-Gordan coefficients will vary, this allows us to find the relative transition strengths of the Zeeman sublevels by decomposing the polarization of the excitation light.

If we want to look beyond the Zeeman sublevels we have find some way to evaluate the reduced matrix element \( \langle \alpha', J' || d || \alpha, J \rangle \). It can be shown that

\[
\langle n', S', L', J' || d || n, S, L, J \rangle = (-1)^{L' + S + J + 1} \sqrt{(2J + 1)(2J' + 1)} \begin{pmatrix} L' & J' & S' \\ J & L & 1 \end{pmatrix} \langle n', L' || d || n, L \rangle.
\]

The term with the curly braces is called a 6j symbol and is related to adding three angular momenta; they can be written as

\[
\begin{pmatrix} J_3 & J & J_a \\ J_1 & J_2 & J_b \end{pmatrix} = \frac{(-1)^{J_1 + J_2 + J_a + J}}{\sqrt{(2J_a + 1)(2J_b + 1)}} \begin{pmatrix} L' & J' & S' \\ J & L & 1 \end{pmatrix} \langle J_a, J_3, J | J_b, J_1, J \rangle.
\]
where the last term is a coefficient relating two different bases, similar to the Clebsch-Gordan coefficients. This allows us to find the relative strengths between fine structure levels.

For hyperfine structure we can write

\[
\langle \alpha', J', F', m_F' \mid \hat{d} \cdot \hat{\epsilon} \mid \alpha, J, F, m_F \rangle = \frac{\langle \alpha', J', F' \mid |d\rangle |\alpha, J, F \rangle}{\sqrt{2J' + 1}} \sum_q (-1)^q \epsilon_{-q} \langle J, m_J, 1, q \mid J', m_J' \rangle
\]

\[
\langle n', S', L', I, J', F' \mid |d\rangle |n, S, L, I, J, F \rangle = (-1)^{J' + I + F + 1} \sqrt{(2F + 1)(2F' + 1)} \begin{bmatrix} J' & F' & I \\ F & J & 1 \end{bmatrix} \langle n', S', L', J' \mid |d\rangle |n, S, L, J \rangle.
\]

Using these relations allows us to find the relative line strengths between transitions with the same \( L \) and \( L' \). For example, using this analysis we find that the \( D_2 \) line is twice as strong as the \( D_1 \) line in lithium, if we excite with unpolarized light.

### 2.13 Radiative Cooling

The basics for this section can be found in both Ref [17], and Ref [19]. For an in depth look at laser cooling see Ref [21]. We know that photons carry momentum, thus we expect that, when they are absorbed, they should impart that momentum to the atom. Similarly when atoms emit photons, momentum should be conserved. The force should be equal to the rate at which momentum is transfered to the atoms. Consider a two state system; we let \( R_{abs} \) be the rate that atoms are excited to the upper state and \( R_{stim} \) be that rate that atoms decay to the lower state via stimulated emission. Since we are considering the steady state situation we must balance the rate equations so that

\[
(R_{stim} + \Gamma) \rho_{22} = R_{abs} \rho_{11},
\]

where \( \Gamma \) is the rate of spontaneous emission as before. When an atom absorbs a photon the photon imparts a momentum \( \hbar k \) to the atom so the average force from
2.13. RADIATIVE COOLING

absorption becomes

\[ F_{\text{abs}} = \hbar k R_{\text{abs}} \rho_{11}, \]

where a positive force is in the direction of the laser light. In stimulated emission
the atom emits a photon in the same direction, and with the same frequency as the
laser light so there is an average force in the opposite direction of the laser light of

\[ F_{\text{stim}} = -\hbar k R_{\text{stim}} \rho_{22}. \]

Finally, in spontaneous emission, we will assume the light is emitted isotropically, so
that on average the force is 0. This gives an average net force of

\[ F = F_{\text{abs}} + F_{\text{stim}} = \hbar k (R_{\text{abs}} \rho_{11} - R_{\text{stim}} \rho_{22}). \]

In reality light is not excited completely isotropically, but the assumption is good
enough for our purposes. We can then plug in our rate equation to get

\[ F = \hbar k ((R_{\text{stim}} + \Gamma) \rho_{22} - R_{\text{stim}} \rho_{22}) = \hbar k \Gamma \rho_{22}. \]

Now we can plug in our steady state population to find

\[ F = \hbar k \Gamma \frac{\Omega^2/4}{(\omega - \omega_0)^2 + \Omega^2/2 + \Gamma^2/4}. \]

If we include a Doppler shift of \( \omega_D = \omega_{c} \), and rewrite our population in terms of
an intensity dependent saturation parameter \( s_0 = 2 \Omega^2/\Gamma^2 = I/I_0 \) where \( I \) is our
intensity and \( I_0 \) is some saturation intensity, we end up with

\[ F = \hbar k \frac{\Gamma}{2} \left( \frac{s_0}{1 + s_0 + (2(\omega - \omega_D - \omega_0)/\Gamma)^2} \right). \]

Now suppose we add another counter-propigating beam of the same laser light. This
beam will impart a force in the other direction, but the Doppler shift will change
sign as well. Then we get

\[ F = \hbar k \frac{\Gamma}{2} \left( \frac{s_0}{1 + s_0 + (2(\omega - \omega_D - \omega_0)/\Gamma)^2} - \frac{s_0}{1 + s_0 + (2(\omega + \omega_D - \omega_0)/\Gamma)^2} \right). \]
Figure 2.4: The force on an atom as a function of that atom's Doppler shift from resonance. If the atom has a negative velocity it will be pushed in the positive direction, and if it has a positive velocity it will be pushed in the negative direction. This force thus acts to push all atoms towards the velocity class with zero Doppler shift.

For lithium $\Gamma = 6 \text{ MHz}$, $I_0 = 2.6 \text{ mW/cm}^2$, and in our experiment $s_0 \approx 2$, Fig 2.4 shows how this force depends on the Doppler shift for this saturation parameter and a detuning of $-3 \text{ MHz}$.

If the Doppler shift is positive, or the atoms are moving in the positive direction, then the force is negative slowing them down. If the Doppler shift is negative, or the atoms are moving in the negative direction, the force is positive, again slowing them. In this way all atoms are pushed toward some zero velocity class, where their velocity has no component parallel to the laser beams. Since the widths of our peaks are determined by Doppler shift given by the velocity distribution of the atoms, this cooling will narrow our peak widths. Since we have an expression for the force and the mass of lithium we can set up a differential equation to get the change in velocity as a function of time

$$m \frac{dv}{dt} = \hbar k \frac{\Gamma}{2} \left( \frac{s_0}{1 + s_0 + (2(\omega - \omega_c - \omega_0)/\Gamma)^2} - \frac{s_0}{1 + s_0 + (2(\omega + \omega_c - \omega_0)/\Gamma)^2} \right).$$

The atoms in our beam are moving at about 1500 m/s, given the size of our
2.13. RADIATIVE COOLING

Figure 2.5: This plot shows the distribution of Doppler shift for no cooling light, and detunings of -1, -3, -10, and -20 MHz. These are simply the distributions of atoms and do not translate directly to fluorescence peaks, in addition we have assumed a zero linewidth laser.

laser beam this corresponds to interaction with the light for about 3 $\mu$s. This allows us to solve for the change in velocity and change in Doppler shift that an atom will experience when moving through the beam. With this information we can see how our velocity distribution will change after cooling. We start with an uncooled Gaussian distribution with a Doppler width of 16 MHz, then Fig 2.5 shows how our distribution will change for a detuning of -3, -10, and -20 MHz. Each detuning is mostly effecting a certain velocity class of the distribution causing non-Gaussian final distributions. In addition the width of the distributions narrow as the detuning decreases in magnitude only until some point at which they widen again, we can see this widening in the distribution with a -1 MHz detuning. The optimal detuning is $-\Gamma/2 = -3$ MHz [21]. So far in the experiment we have not been able to get closer than 20 MHz from resonance, for which we see little change in the distribution. Note
that these are just based on the velocity distributions, and do not include optical effects such as the natural linewidth, or optical pumping which will be relevant for the fluorescence peaks, in addition they assume a zero linewidth laser. The actual system is further complicated since we are cooling on a wider distribution with a width of around 200 MHz, before it is reduced to 16 MHz by a collimating plate.

This treatment so far assumes an isolated two state system. The $F = 2 \rightarrow 3$ transition of the $D_2$ line of lithium is almost such a system. Unfortunately, since the hyperfine structure is unresolved, some atoms can be excited to the $F' = 2$ state, from there they can decay to the $F = 1$ hyperfine level of the ground state which will be dark to our laser light. This will cause many atoms to be optically pumped into this state before they have been cooled sufficiently. To counteract this we need to introduce re-pump light resonant with the transition from $F = 1 \rightarrow 2$ where they can decay to the proper ground state and be cooled again. The cooling transitions used can be seen in Fig 2.6.
Figure 2.6: The transitions on which we cool and re-pump our atoms. Atoms can be excited by the cooling light to the $F' = 2$ excited state, where they can decay to the $F = 1$ ground state. We have the re-pump light so that these atoms can be moved back onto resonance with the cooling light.
Chapter 3

Experimental Setup

3.1 Overview

In this experiment the $D_1$ and $D_2$ lines of $^6\text{Li}$ and $^7\text{Li}$ are being measured. The measurement is made by scanning the frequency of a probe laser, orthogonal to an atomic beam of Li, over the resonances. When the probe laser is on resonance with a transition, fluorescence of the atoms is detected. The probe beam is also retro-reflected back through the chamber, this symmetrizes the peaks. The frequency of the probe laser is determined by locking it to the stabilized frequency comb, and the probe frequency is scanned by scanning the comb’s repetition rate. A overview of the frequency locks for this system can be seen in Fig 3.1

In addition to the main system, an additional system is in the process of being developed to cool the transverse components of the atomic beam. This involves two additional diode lasers resonant with the $D_2$ line, one to supply the main force of transverse cooling, and the other to pump atoms out of a dark state. Both of these processes happen in a preliminary interaction region before the atoms interact with the main probe beam. The cooling laser’s frequency is locked using the fluorescence from its interaction with the atoms, and the re-pump laser is locked to the cooling laser. To simplify the system when looking for cooling, the probe laser is locked to
Figure 3.1: A rough block diagram for the locks involved in the main experiment. The probe beam, which interacts with the atoms is locked to the comb using the mixed down signal of the beat note between the probe and the nearest mode of the comb. The repetition rate is locked using the mixed down signal of the beat note between adjacent modes of the comb. The offset frequency detected by doubling low frequency light beating it with the undoubled comb, it is then mixed down and locked. The offset lock has a few more steps not shown in this diagram but discussed in Sec 3.3.
Figure 3.2: A rough block diagram for the locks necessary for transverse cooling. Note that both the cooling laser and re-pump laser have two possible locks which are represented here. The cooling laser is locked to the fluorescence peaks through a level lock described in Sec 3.11, or through a dither lock described in Sec 3.12. The re-pump is locked to the cooling laser either with a delay line lock described in Sec 3.9 or the same phase lock as the offset frequency as described in Sec 3.10. The probe laser is locked to a Fabry-Perot through a Pound-Drever-Hall lock as described in Sec 3.13.

a Fabry-Perot cavity, which allows us to scan over the resonances without using the comb. An overview of the frequency locks of this cooling system can be seen in Fig 3.2. Note that we are trying out multiple types of locks for the re-pump and cooling lasers.

3.2 Optical Frequency Comb

This section will go through a brief overview of optical frequency combs, and the particular comb in this lab. Other overviews can be found in Refs [25], and [26]. For
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Figure 3.3: A diagram of the cavity of the comb. The pump laser hits the Ti:Sapphire crystal and generates pulses which propagate through the bowtie cavity. There’s a piezo ceramic on M3 which allows us to change the path length to control the repetition rate.

A more detailed description of our comb see Ref [11]. The output spectrum of the frequency comb consists of discrete frequencies evenly spaced by a repetition rate $f_r$, and offset from zero by an offset frequency $f_0$. This allows us to write the frequencies as

$$\nu_n = n f_r + f_0,$$

where $n$ is a natural number. This gives us a frequency spectrum resembling the representation of the comb in Fig 3.1. The repetition rate and offset frequency are typically in the radio frequency regime. These frequencies can be measured to arbitrary precision with the aid of a GPS steered atomic clock. By measuring these two frequencies we can determine the optical frequencies in the comb spectrum. In this experiment, the purpose of the optical frequency comb is to provide an absolute frequency reference. Normally optical frequencies are too fast to measure accurately, by stabilizing our probe laser to a known frequency in the comb, we translate this problem into the measurable radio frequency regime.

The characteristic comb spectrum is obtained by creating a femtosecond pulse which propagates through a bowtie cavity as seen in 3.3, some of this light escapes through the output coupler. Then the output consists of a short pulse pulses every
3.2. OPTICAL FREQUENCY COMB

$T = L/c$ seconds where $L$ is the effective path length of the cavity and $c$ is the speed of light. Our cavity has a length of about 30 cm and generates pulses that are about 40 fs long. From Fourier analysis we know that to create such a short pulse we need a large range of frequencies. The gain medium for the comb is a titanium doped sapphire crystal (Ti:Sapphire) which is pumped by a 5 mW laser. This crystal has a bandwidth from 770-800 nm which is wide enough to create these short pulses. Just because the bandwidth is available, does not ensure that we will get a pulsed output. In our comb, the pulsed output is ensured though Kerr lensing in the crystal. This effect relies on a third order correction to the index of refraction that is proportional to the intensity of light. When the output is a continuous wave (CW), the peak intensity is the same as the average intensity and this correction is small. In pulsed operation the peak intensity is very high and the change in the index of refraction of the crystal acts as a lens to focus the light. When the beam is focused there is more overlap with the gain medium, so pulsed operation is preferred. This is sufficient if all frequencies see the same path length, but in general this is not the case. The index of refraction in the crystal will be frequency dependent, an effect called dispersion, then the path length is a function of frequency. To account for this, and to avoid the pulses spreading out, special chirped mirrors are used in the cavity, that allow low frequencies to penetrate deeper, increasing the path length for low frequencies, and offsetting the dispersion of the crystal. In pulsed operation, when the comb is mode locked, there is a fixed phase difference $\Delta \phi$ between the carrier wave in consecutive pulses. When we consider this mode locked spectrum, the train of pulses separated in time by $T$ gives effectively discrete frequencies separated by

$$f_r = 1/T.$$  

This is our repetition rate, which for this comb is about 900 MHz. The offset frequency is related to the phase offset $\Delta \phi$ through

$$f_0 = \frac{1}{2\pi} f_r \Delta \phi.$$  

This frequency is usually between 100 and 500 MHz.
CHAPTER 3. EXPERIMENTAL SETUP

When our comb is mode locked its output does not span an optical octave, which is necessary as we will see in Sec 3.3. To achieve an octave spanning laser, the output is sent through microstructure fiber. The fiber is single mode and has negative dispersion for optical wavelengths, this compresses the pulses, and in turn expands the frequency range of our comb. More information on this fiber can be found in Ref [27], or Ref [14].

3.3 Repetition Rate Stabilization

For the comb to be stable, and provide an absolute frequency measurement, we need the offset frequency and repetition rate to be locked to known radio frequencies, which are directly linked to a GPS steered atomic frequency standard. To achieve a lock, we must first detect the repetition rate $f_r$. Luckily any two adjacent modes of the comb, when heterodyned on a detector, will yield a beat note at the repetition rate. Once it is detected the signal is filtered and mixed down with a synthesizer, set to the desired locking frequency. The mixer outputs a signal at the difference frequency between the actual repetition rate and the frequency of the synthesizer, we can use this as an error signal. This difference signal is sent to a loop filter which allows us to shape the signal for better feedback, and is then amplified by a high voltage amplifier. The high voltage error signal is sent to a piezo-ceramic in the comb cavity which changes the length of the cavity as voltage is applied. This change in length changes the period of pulses, which in turn changes the repetition rate keeping it at the desired frequency. A more detailed description of this lock can be found in Refs [11], and [13].

3.4 Offset Frequency Stabilization

The offset frequency $f_0$ is stabilized in a similar way. It is detected by frequency doubling the comb light with wavelengths around 1060 nm and heterodyning it with
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Undoubled comb light with wavelengths around 530 nm. In order to double the light, the comb beam is sent through a frequency doubling crystal, however this adds a time delay between the doubled and undoubled light. To account for this, comb light is split such that the high frequencies and low frequencies are sent along different paths before being recombined. By changing the length of these paths we can correct for the time delay and get a heterodyned signal. For a given $n$ the doubled light with mode number $n$ and the undoubled light with mode number $2n$ then yield a beat note at the offset frequency, since

$$|2(n f_r + f_0) - (2n f_r + f_0)| = |f_0|.$$ 

The sign of the offset frequency can be determined by looking at its response while locking, or by finding the sign which matches the fluorescence peaks with their expected frequencies. The detected signal typically has frequency noise which moves it around by about 15 kHz, to account for this it is filtered and mixed up to about 1240 MHz, then divided by 8 which decreases the noise on a frequency of around 155 MHz. A digital phase detector then mixes this signal down with a signal at 155 MHz from a synthesizer which gives us an error signal. Just as with $f_r$, this error signal is then shaped with a loop filer. The signal from the loop filter is sent to the driver of an Acousto-Optic Modulator (AOM) in the comb's pump beam. The AOM scatters some of the light off of lattice vibrations from the modulation of the driver, decreasing the pump power. Decreasing the power alters the phase relationship $\Delta \phi$ between consecutive pulses, and thus changes the offset frequency. A block diagram is shown in Fig 3.4, and a more detailed description of this lock can also be found in Refs [11], and [13].

3.5 Diode Lasers

We use Extended Cavity Diode Lasers (ECDLs) at 671 nm to probe the transitions and for cooling. There are three nearly identical ECDLs in this lab, one to probe the transitions of interest and two lasers used for cooling. We can get about 25
mW of power out of these lasers when they are operating at their best, but this often drops to around 15 mW or even lower over time. These diodes have an anti-reflective coating on their output so that there is no internal cavity, instead the cavity is created outside the diode using the first order reflection off of a grating. Our cavities are in the Littrow configuration as seen in Fig 3.5, the first order off of the grating is sent back to the diode, and the zeroth order is sent out to the table. Using a grating allows for a narrower line width laser, since only part of the output spectrum is sent back to the diode. By adjusting the angle of the grating, we can change which frequencies get fed back and make large adjustments to the frequency of our laser. Smaller scale adjustments can be made by changing the temperature and the current of the diode. In addition, the frequency can be tuned by applying a voltage to a piezo-ceramic on the grating which changes the cavity length; this is our finest frequency control and we feedback to this piezo in order to lock the frequency. In addition, we can supply a small amount of feedback to the current of the diodes.
Figure 3.5: The configuration of our diode lasers. The first order off of a grating is sent back to the laser diode, while the zeroth order is sent to the table. The grating includes a piezo ceramic which allows us to tune the frequency.

which also affects the frequency and improves our locks. More details of our specific diode lasers and other possible configurations can be found in Ref [14].

3.6 Probe Beat Note Stabilization

The absolute frequency reference of the comb is not useful unless it can be related to the frequency of the probe laser. By stabilizing the beat note $f_b$ between the probe and the comb we can find the frequency of the probe through

$$\nu = nf_r + f_0 + f_b$$

where the mode number $n$ is determined by referencing fluorescence peaks. The beat note is detected by heterodyning the probe laser with filtered light from the comb, giving a beat note between the probe frequency and the closest mode of the comb. In order to reduce the noise, this signal is filtered and mixed up to 1240 MHz, before being divided by 16. It is then mixed down with the digital phase detector and sent through a loop filter and high voltage amplifier. This lock feeds back to
CHAPTER 3. EXPERIMENTAL SETUP

Figure 3.6: A block diagram for the beat note frequency lock. Comb light is filtered and heterodyned with the probe laser. The detected beat note is mixed up to 1240 MHz, divided by 16, and mixed down with a digital phase detector, before being sent to the grating and current of the probe laser.

the piezo-ceramic in the diode laser cavity which changes the cavity length and thus the frequency. There is additional feedback with proportional gain to the current of the laser diode which provides higher frequency feedback. Typically we lock the beat note to 200 MHz. Fig 3.6 gives a block diagram for this lock, and more details can be found in Ref [13].

3.7 Power Stabilization

While stabilizing the diode laser frequency eliminates most of the frequency noise, power fluctuations will impact our signal to noise, and can introduce systematic shifts when the power is frequency dependent. To compensate for this, the power is measured and locked. Before the beam is sent through the interaction region, a beam splitter sends some light to a photodiode which measures the power level. A DC voltage at the desired locking level is subtracted from the photodiode signal.
which is then sent to a loop filter and amplified by a high voltage amplifier. This signal is sent to an Electro-Optic Modulator (EOM) in the beam path. When the high voltage signal is applied to the crystal in the EOM, the index of refraction along one axis changes, effectively rotating the polarization of the light passing through it. A polarizer on the output of the EOM then changes the power, allowing for stabilization. More details can be found in Ref [13]. This is the last lock in the main experiment, after discussing the vacuum system used for both the main experiment and the transverse cooling, we will discuss the cooling system locks.

3.8 Vacuum System and Atomic Beam

Our lithium atomic beam is created by heating solid Li to around 420° c in an oven. The oven has a nozzle consisting of a single hole through which the Li escapes creating a wide beam. This wide beam then passes through the cooling interaction region, where it can interact with the cooling and re-pump lasers. These lasers pass through the interaction region and are retro reflected back through the other direction with a mirror. This ensures cooling of atoms moving in either transverse direction. Mounted on top of this region is a Photomultiplier tube (PMT) to detect the fluorescence signals. To narrow the atomic beam before the main interaction region there is a plate with a small hole, which the atoms must pass through. Before this collimation plate, the signals are about 200 MHz wide, after the plate they are reduced to about 16 MHz wide. However, the velocity distribution after the plate is not entirely symmetric. After making its way through the plate, the atomic beam interacts with the probe beam. The probe beam is counter propagated through the chamber with a reto-reflecting corner cube. The retro-reflected beam excites the velocity class of atoms mirror to the velocity class excited by the main beam. This symmetrizes our peaks, even with an asymmetric velocity distribution. In addition this retro reflection allows us to align our probe beam perpendicular to the atomic beam, as outlined in Ref [14]. Two large lenses hang in vacuum above the main
interaction region to collect light from the fluorescence of the atoms, which is then detected by a larger PMT mounted atop the system. After interacting with the probe beam, the atoms hit the back wall of the chamber. The system is pumped with an ion pump which ionizes stray gas molecules in the system and pulls the remaining charged particles out. The ion pump gets the whole system down to around $10^{-8}$ Torr when the ovens are off. Fig 3.7 shows the vacuum system.

3.9 Re-Pump Laser Delay Line Lock

As mentioned, we need two lasers to achieve cooling, a main cooling laser, and a re-pump laser. From Sec 2.13 we know that the re-pump laser must be detuned by about 813 MHz above the cooling laser. We get about 803.5 MHz from the ground state splitting and about 9.3 MHz from the hyperfine splitting of the excited state. To achieve the fixed frequency difference between the cooling and re-pump lasers, we lock the beat note between them to 813 MHz. Our first attempt at this lock involved
a variable delay line, a device which allows us to finely adjust the path length taken by a radio frequency signal. To lock the frequency, we heterodyne the cooling and re-pump lasers onto a detector, and then split the resulting beat note signal and send it though two paths, one of which includes the variable delay line. Just as with any interferometer we get complete destructive interference if the difference in path lengths is equal to a half integer number of wavelengths

\[ \Delta L = \left( n + \frac{1}{2} \right) \lambda = \left( n + \frac{1}{2} \right) \frac{c'}{f}, \]

where \( \Delta L \) is the difference in path length, \( \lambda \) is the wavelength, \( f \) is the frequency and \( c' \) is the effective speed of the signal in the cables or other elements of the system. We use the output of the interferometer as an error signal, where we can lock to any frequency satisfying

\[ f = \left( n + \frac{1}{2} \right) \frac{c'}{\Delta L}. \]

The delay line allows us to change \( \Delta L \) so that we get a good error signal at 813 MHz. We then send the signal through a loop filter and high voltage amplifier which sends feedback to the grating of the re-pump laser. With this setup however, the lock is not very stable since the locking frequency, set by the delay line, fluctuates over a variety of time scales. While a narrow laser linewidth is not necessary to pump atoms out of the dark state, the frequency noise of this lock means the delay line must be adjusted frequently, and it makes the fluorescence signal harder to lock the cooling laser to, as described in Sec 3.11.

### 3.10 Re-Pump Laser Phase Lock

In order to maintain a tighter lock, the beat note between the re-pump and cooling lasers can be sent through the same locking procedure as the offset frequency lock discussed in Sec 3.3. This lock allows for a tighter lock than the delay line lock, as the synthesizer has a much higher resolution, and doesn’t drift like the mechanical delay line. In addition, we have been able to achieve more feedback to the current of


Figure 3.8: Signals from the cooling, and re-pump, as well as their combined signal for two different detunings between them. We see a large peak when both the cooling and re-pump lasers are on resonance, corresponding to pumping into the cooling state.

the re-pump laser with this lock. This gain in the frequency stability of the re-pump gives a more stable fluorescence signal to lock our cooling laser to.

3.11 Cooling Laser Level Lock

Our first attempt to frequency stabilize the cooling laser involved locking the level of the fluorescence signal from the preliminary interaction region. In the process of cooling and re-pumping, the atoms within the preliminary interaction region fluoresce. This fluorescence is then detected by a PMT. While these signals are independent of the probe beam, they include contributions from both the cooling and re-pump lasers. A typical signal from scanning both the probe and re-pump can be seen in Fig 3.8.

Here there are a few preliminary things to note. The peaks are much wider here than in main interaction region, the width seen in Fig 3.8 is closer to 200 MHz, compared to about 16 MHz in the main interaction region. This is because the preliminary interaction region is before the collimation plate, and thus we are seeing
the full Doppler width out of the oven. Also there are saturation dips in the peaks at the zero velocity class where the counter-propagating beams excite the same atoms. Lastly, since we also see the atoms’ interaction with the re-pump, the signal includes two sets of the resonances, each offset about 813 MHz from the other. When both the re-pump and cooling lasers are on resonance with the atoms, there is a sharp peak in the signal, as atoms are pumped out of the $F = 1$ ground state. The two plots show the signal for two different detunings between the re-pump and probe laser, and we see that there is a significant shift in the character of the peak.

When we want to lock the frequency of the cooling laser, a fixed DC voltage at the desired locking level is subtracted from the PMT signal and put through a loop filter and high voltage amplifier, sending feedback to the piezo-ceramic in the cooling laser cavity. As shown in Sec 2.13 we want the cooling laser to be slightly red-shifted from the resonance. Unfortunately, the level lock is not stable enough to lock close enough to the resonance, as any change in the re-pump frequency significantly changes the character of the sharp peak we see in the signal. Since the lock only stabilizes the level of the signal, and not the frequency of the cooling laser directly, noise in the re-pump lock will write noise onto the cooling laser lock. In addition when we try to lock very close to the resonance, a small change in the re-pump laser can make the desired level unattainable, and we quickly lose our lock.

### 3.12 Cooling Laser Dither Lock

To lock the cooling laser closer to the resonance, we have attempted a dither lock. To achieve this lock, an approximately 10 kHz signal from a lock-in amplifier is used to modulate either the grating or current, and thus the frequency, of the cooling laser. The fluorescence signal is then sent back to the lock-in amplifier, which multiplies the signal by the driving signal. This process outputs a signal that is effectively the derivative of the fluorescence signal. Since the derivative crosses zero when the cooling laser is at the top of the resonance, and has opposite sign on either side, the
CHAPTER 3. EXPERIMENTAL SETUP

output of the lock-in amplifier can be used as an error signal. The phase between the multiplied signals can be changed by the lock-in amplifier to shift where on the peak the error signal is zero. This allows us to lock our laser at small detunings. As with most of the other locks this signal is sent through a loop filter and a high voltage amplifier which controls the grating in the cooling laser cavity.

This technique has the advantage that we can lock anywhere on the top of the peak. In addition, since we only modulate the cooling laser, this lock is mostly independent of the fluorescence signal from the re-pump laser. This ensures we don’t write noise onto the cooling laser as the re-pump frequency varies. Unfortunately, the modulation of the cooling laser makes its beat note with the re-pump modulate as well, and this makes the re-pump laser lock very unstable. It’s possible that if we prestabilize the re-pump laser to a cavity this could be improved.

3.13 Probe Pound-Drever-Hall Lock

Ref [28] is the original paper describing this type of lock, and Ref [29] provides a detailed explanation for how it works. Even if the re-pump and cooling lasers are locked to their desired positions, we still need to scan the probe laser over the peaks to investigate their widths, and determine whether we are cooling the atoms. We can scan the probe laser, unlocked, by scanning the voltage applied to the piezo-ceramic, but due to frequency jitter and drift, the resolution is not good enough to determine if we are cooling. We can also lock the probe laser to the comb, as in the main experimental setup, but this is time and effort intensive. Our solution is to lock our probe laser to a confocal Fabry-Perot cavity. The Fabry-Perot is a highly reflective confocal cavity as seen in Fig 3.9. Since the light has a path length of $4L$, the frequency difference between adjacent resonant modes is given by

$$\Delta \nu = \frac{nc}{4L}.$$ 

These modes can build up in the cavity, while all others will destructively interfere. When light builds up in the cavity, some of it leaks out and can be detected at the
3.13. PROBE POUND-DREVER-HALL LOCK

Figure 3.9: A confocal Fabry-Perot cavity

output. As the laser frequency is changed the output of the Fabry-Perot has peaks at frequencies separated by $c/4L$.

Initially some amount of light is reflected off the input of the Fabry-Perot. When the laser is resonant with the cavity, the light that leaks out of the cavity, back towards the laser, is out of phase with the reflected light from the input, and we get a minimum intensity reflected. This signal can be detected by splitting the light with a polarizing cube, and then sending it through a quarter wave plate. When the light passes through the waveplate on its way to and back from the Fabry-Perot, its polarization will be rotated so that it is transmitted, instead of reflected, by the cube on its way back, and a signal can be detected. This setup is shown in Fig 3.10.

In order to get an error signal the beam is sent through an EOM. The EOM is driven with high voltage, though a resonant tank circuit, at 35 MHz. The tank circuit consists of an inductor, tuned with the capacitance of the crystal in the EOM, to be resonant with the driving frequency. When driven with an AC signal the EOM phase modulates the laser light and adds sidebands at the driving frequency. Thus, when the back reflection is detected there will be a beat note at the driving frequency from interference between the fundamental and the sidebands. This signal is then mixed with the driving frequency to get our error signal. When the laser is on resonance the fundamental is not reflected, so there is no signal off the detector at 35 MHz, and the error signal is zero, as we would like. The phase between the signal off the detector and the driver can be altered such that the error signal looks like Fig 3.11.
CHAPTER 3. EXPERIMENTAL SETUP

Figure 3.10: A block diagram for the Pound-Drever-Hall lock. The EOM is driven to add sidebands to the light. The signal from the back reflection off of the Fabry-Perot cavity is then mixed with the driving frequency to get an error signal.

Figure 3.11: The error signal for a Pound-Dever-Hall Lock for a driving frequency of 35 MHz. The capture range is determined by the location of the sidebands, this allows for a larger capture range than other locks.
The error signal then feeds back to the grating and current of the probe laser. This setup allows for a large capture range and stable locks. To scan the frequency a voltage can be applied to a piezo-ceramic on one of the mirrors of the Fabry-Perot which changes the length and thus the resonant frequencies. While this lock gives us a stable laser on short time scales, there is drift in the Fabry-Perot and hysteresis on the piezo-ceramic which cause non-linearities in the scan. These are accounted for by recording the output of another Fabry-Perot which gives us relative frequency markers.
Chapter 4

Results

4.1 Improvements to main setup

A few improvements have been made to the system since the last time data was presented in Ref [14]. The first improvement is new windows on the chamber which give a negligible amount of loss. The previous data were much more sensitive to misalignments between the retro-reflected beam and the main beam since reflections off of the windows caused the former to have lower power and contribute less to the peak. In addition this caused asymmetry in the peak, which made our fits less accurate. While this effect is not as bad now, there is still loss from the corner cube. In order to match the lineshapes between the two beams we want the retro-reflected beam to fall back along the main beam, however in this configuration it hits the corner cube right at the center, where there is more loss due to the cube’s structure; this introduces some asymmetry to our peaks. In addition to the new windows we can now continuously count either the beat note or offset frequency. This allows us to cut scans where the locks drift or are not sufficiently tight, and to cut individual points where the locks vary from the desired value. Additional cuts can be made by looking at the standard deviation of the error signals for these locks, which detects places where the locks oscillate, but maintain a good average value. These oscillations can
lead to variations in the laser linewidth over a scan.

4.2 Data Collection

Typically when data are taken for the main experiment, we record the repetition rate, the PMT signal, the power in the probe beam, the error signals for the offset and beat note locks, the signal through a Fabry-Perot, and the count of the beat note. All of these signals are recorded by a 16 bit analog to digital converter, which, for most signals, averages between 500 – 2000 samples for each point recorded in the scan, at a rate of 50000 samples per second. This allows for the standard deviation of each signal to be recorded as well. The computer controls and scans the repetition rate by setting the synthesizer of the lock to the desired value, and records the value it is set to. The count is measured over the time it takes to record a point in all the other signals, and is recorded before the next point is taken.

4.3 Peak Fitting

The peaks have contributions from the velocity distribution of the atoms which should be close to Gaussian and the natural lineshape of the peaks which is a Lorentzian. Peaks were thus fit to a combination of a Lorentzian and Gaussian lineshape specified by

\[ f(\nu) = A \left[ FL \left( \frac{W_L^2/4}{W_L^2/4 + (\nu - \nu_0)^2} \right) + e^{-\frac{(\nu - \nu_0)^2}{2W_G^2}} \right] + B, \]

where \( FL \), the fractional Lorentzian, scales the relative Lorentzian contribution to the peaks, and \( A \) is a total scaling factor, making the absolute amplitude of the peaks \( A(1 + FL) \). The center frequency of the peaks is given by \( \nu_0 \), and \( B \) gives the vertical offset from zero, due to an offset of the PMT and scattered light. \( W_L \) is the full width at half maximum (FWHM) for the Lorentzian component, and the
4.3. **PEAK FITTING**

FWHM of the Gaussian component is $2\sqrt{2\ln2} W_G$. Combining these we can find the FWHM for our combined lineshape by solving

$$FL \left( \frac{W_L^2/4}{W_L^2/4 + (\text{FWHM}/2)^2} \right) + e^{-\frac{(\text{FWHM}/2)^2}{2W_G^2}} = \frac{1}{2} (1 + FL).$$

We find our absolute frequency $\nu$ by solving the comb equation, combined with the beat note between the comb and the probe beam

$$\nu = nf_r \pm |f_0| - |f_b|.$$

The mode number $n$ and the sign of the offset frequency can be found by comparing the possible values with the expected values of our center frequencies, which are known well enough for this process. Typically the mode number is around 488 000. Currently the sign of the beat note is defined to be negative. These fits are weighted using the standard deviation of the PMT signal and dividing by the square root of the number of samples per point. This weighting underestimates the error, so it is scaled to get a reduced $\chi^2$ of 1.

Before fitting, scans are excluded where too many points were cut due to bad locks, or where the power in the probe beam was too high such that AC stark shifts, or saturation would be significant. After the fits were completed, scans were cut where fit parameters, other than the center frequency, were outside of a normal range, due to noise in the signal affecting the fits, or where there was significant structure in the residuals. This structure either due to asymmetry in the peak, or the probe power being high enough that the top of the peak flattens off as the atoms start to saturate. In addition there were some scans where either the Lorentzian width or Gaussian width became large, while decreasing the corresponding amplitude, to fit noise in the background rather than the fluorescence peak. This could be accommodated for by cutting out the Lorentzian component, but even so, these fits were cut on $^7\text{Li}$. For the $^6\text{Li}$ peaks, this width effect happened too often to cut these scans, instead, the widths where fixed so that all peaks in the scan had the same width. Examples of good fits can be seen in Fig 4.1.
Figure 4.1: Examples of fits for the D1 lines of $^6$Li and $^7$Li. The peaks were fit to a combination of Lorentzian and Gaussian lineshapes. Residuals can be seen at the top of the plots.
4.3. PEAK FITTING

Figure 4.2: An fit to a lineshape modeled off of data. This is a fit of the $^7\text{Li} D_1 F = 1 \rightarrow F'$ lines. The residuals do not have structure, but due to interpolation, the fit function has its own noise.

The fits described above gave self inconsistencies in the splittings between states as discussed in Sec 4.4. We think this could be due to an issue with an asymmetric lineshape. In an attempt to resolve these lineshape issues, we tried another fit method by creating an interpolation function out of data taken without the retro beam, to mimic our base lineshape. While we used the most isolated peak to create this function, we still subtracted off the contribution from its closest neighbor. This function was then be reflected around the center frequency to model the contribution from the retro-reflection. Horizontal and vertical scaling factors were added to the function and the amplitude ratio between the main contribution and the retro contribution was allowed to vary, so as to model loss in the retro-reflected beam. An example of one of these fits can be found in Fig 4.2. The results of these fits are discussed in Sec 4.4.
<table>
<thead>
<tr>
<th>Transition</th>
<th>$F$</th>
<th>$F'$</th>
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<tbody>
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<td>$1/2$</td>
<td>446 789 502.750(37)</td>
</tr>
<tr>
<td>$^6$Li $D_1$</td>
<td>$3/2$</td>
<td>$3/2$</td>
<td>446 789 528.739(34)</td>
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<td>$3/2$</td>
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<td></td>
<td>446 789 596.27(10)</td>
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<td>$1$</td>
<td>446 799 771.173(18)</td>
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<td></td>
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<td>446 800 129.868(44)</td>
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Table 4.1: Absolute frequencies of $D_1$ hyperfine structure, and center of gravities

### 4.4 Results

Tab 4.1 gives the absolute frequency measurements for the $^6$Li and $^7$Li $D_1$ lines, with systematic effects from Zeeman shifts, AC stark shifts, frequency stabilization, and misalignment taken into account. Information on the evaluation of these systematics can be found in Ref [14], with the only change being the contribution from beam alignment. The process of beam alignment described in Ref [14] assumes that the corner cube is a perfect retro-reflector. Looking at the specifications for angle misalignment with our corner cube, we can estimate the uncertainty this would cause in our center frequency by estimating the doppler shift of atoms perpendicular to the misaligned beam. This gives us an uncertainty in the center frequency from misalignment of 15 kHz. In this analysis we use this 15 kHz uncertainty from misalignment instead of the 9 kHz uncertainty presented in Ref [14]. We should be able to decrease this misalignment uncertainty through better evaluation of the corner cube, but this was not done for these data.
4.4. RESULTS

The center of gravity (cog) of a fine structure state is defined as the average of the hyperfine energies, weighted by the degeneracy of the hyperfine state. Then the center of gravity of a transition is the difference between the center of gravity of the two states. For $^7\text{Li} \ D_1$, this gives

$$\text{cog} = \frac{3E_1' + 5E_2'}{8} - \frac{3E_1 + 5E_2}{8} = \frac{3(E_1' - E_1) + 5(E_2' - E_2)}{8},$$

where $E_1', E_2'$ are the energies of the $F = 1$ and $F = 2$ hyperfine levels of the excited state respectively, and $E_1, E_2$ are the energies of the $F = 1, F = 2$ hyperfine levels of the ground state respectively. Since we measured the frequency of the $F = 1 \rightarrow 1$ and $F = 2 \rightarrow 2$ transitions, we can calculate the center of gravity frequency of the $D_1$ line of $^7\text{Li}$, and similarly for $^6\text{Li}$. These numbers can also be found in Tab 4.1.

The preliminary data presented in Ref [14], was only taken over one or two days, and did not investigate effects that vary day to day, like the quality of the locks, changes in the character of the probe laser, or other aspects of the system. In contrast, these data were taken over more days, and variation of the system day to day increased the scatter in the data accounting for the increase in uncertainty. Since we get center frequency measurements for every $D_1$ line, we have multiple ways to calculate the hyperfine splitting between states. For example to calculate the ground state splitting of $^7\text{Li}$ we can subtract the frequency of the $F = 1 \rightarrow 1$ transition from the frequency of the $F = 2 \rightarrow 1$ transition, or we can subtract the frequency of the $F = 1 \rightarrow 2$ transition from the frequency of the $F = 2 \rightarrow 2$ transition. Doing this allows us to check the internal consistency of our data, as these two calculations should be consistent. The data presented in Ref [14] was not self consistent splittings, and even with the improvements and the increase in uncertainty these new data are still not self consistent, as presented in Tab 4.2.

This lack of self consistency could be coming from a variety of sources. The most probable is issues with the lineshape of our peaks, as most other effects would not give such a dramatic systematic shift. As mentioned earlier, there is still power loss in the retro-reflected beam, which would make our peaks not perfectly symmetric. In addition we see that the inconsistencies are worse for the less resolved peaks of
CHAPTER 4. RESULTS

<table>
<thead>
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<th>Splitting</th>
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<th>F'</th>
<th>Frequency (MHz)</th>
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<td>1,2</td>
<td>2</td>
<td>803.482(16)</td>
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<td>10533.60(11)</td>
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</table>

Table 4.2: Hyperfine splittings and the isotope shift. Each hyperfine splitting is measured twice using transitions between different states. There are large discrepancies in the splittings.

$^6\text{Li}$, and this does not change if we allow the widths of the peaks to be different.

As mentioned in Sec 4.3, we tried to model our asymmetric distribution and retro reflection better, using an interpolation function as a fit. Using this fitting scheme gives frequencies shown in Tab 4.3, and Tab 4.4.

These interpolation fits did not resolve the discrepancies in the measurements. This could be because we used the lineshape of a single scan of a particular peak, and it’s possible that the lineshape will shift between scans in ways we cannot model with the interpolation function. For example, in the previous fitting scheme the fractional Lorentzian is allowed to vary, but there is no equivalent parameter for these fits. Note that an uncertainty analysis was not done for these fits, since the discrepancies were not resolved.

We hope that transverse cooling of the beam should allow us to further narrow the Doppler width, and better resolve the peaks. Since we expect the discrepancies are related to the overlap of the Doppler broadened peaks, with increased resolution from transverse cooling we should be able to get better fits, and investigate this effect.
### 4.4. RESULTS

<table>
<thead>
<tr>
<th>Transition</th>
<th>$F$</th>
<th>$F'$</th>
<th>Frequency (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^6\text{Li } D_1$</td>
<td>$3/2$</td>
<td>$1/2$</td>
<td>446 789 596.24</td>
</tr>
<tr>
<td></td>
<td>$3/2$</td>
<td>$3/2$</td>
<td>446 789 528.686</td>
</tr>
<tr>
<td></td>
<td>$1/2$</td>
<td>$1/2$</td>
<td>446 789 731.35</td>
</tr>
<tr>
<td></td>
<td>$1/2$</td>
<td>$3/2$</td>
<td>446 789 756.908</td>
</tr>
<tr>
<td>$^6\text{Li } D_1$ cog</td>
<td></td>
<td></td>
<td>446 789 596.24</td>
</tr>
<tr>
<td>$^7\text{Li } D_1$</td>
<td>2</td>
<td>1</td>
<td>446 799 771.153</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>2</td>
<td>446 799 863.049</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>1</td>
<td>446 800 574.687</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>2</td>
<td>446 800 666.515</td>
</tr>
<tr>
<td>$^7\text{Li } D_1$ cog</td>
<td></td>
<td></td>
<td>446 800 129.913</td>
</tr>
</tbody>
</table>

**Table 4.3:** Absolute Frequencies of $D_1$ lines (Interpolation Fits)

<table>
<thead>
<tr>
<th>Splitting</th>
<th>$F$</th>
<th>$F'$</th>
<th>Frequency (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^6\text{Li } 2s\ ^2S_{1/2}$ hfs</td>
<td>$1/2, 3/2$</td>
<td>$1/2$</td>
<td>228.668</td>
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<tr>
<td></td>
<td>$1/2, 3/2$</td>
<td>$3/2$</td>
<td>228.222</td>
</tr>
<tr>
<td>$^7\text{Li } 2s\ ^2S_{1/2}$ hfs</td>
<td>1,2</td>
<td>1</td>
<td>803.534</td>
</tr>
<tr>
<td></td>
<td>1,2</td>
<td>2</td>
<td>803.466</td>
</tr>
<tr>
<td>$^6\text{Li } 2p\ ^2P_{1/2}$ hfs</td>
<td>$3/2$</td>
<td>$1/2, 3/2$</td>
<td>26.004</td>
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<tr>
<td></td>
<td>$1/2$</td>
<td>$1/2, 3/2$</td>
<td>25.558</td>
</tr>
<tr>
<td>$^7\text{Li } 2p\ ^2P_{1/2}$ hfs</td>
<td>2</td>
<td>1, 2</td>
<td>91.896</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>1, 2</td>
<td>91.828</td>
</tr>
</tbody>
</table>

**Table 4.4:** Hyperfine and Isotope Splittings (Interpolation Fits)
more fully. In addition the transverse cooling should lower our uncertainties, and allow for investigation of the $D_2$ lines.

### 4.5 Progress Towards Cooling

When we take data to look for cooling, the computer controls the probe laser frequency by sending a voltage to the Fabry-Perot of the Pound-Drever-Hall lock, varying its length, and scanning the probe laser over the peaks. We then record the fluorescence peaks off of the PMT in the main interaction region, the transmission through the Fabry-Perot we are locking the probe laser to, the signal from another Fabry-Perot which gives us relative frequency markers, and the fluorescence from the cooling laser, detected by the PMT at the first interaction region. The second Fabry-Perot signal gives us frequency markers every 250 MHz, these markers allow us to correct for non-linearities in our scan and calibrate the relative frequency of the probe laser.

Without the re-pump laser we see optical pumping into the $F = 1$ ground state as expected. When we add in the re-pump and are close to resonance, so that we are locking on the sharp peak seen in Fig 3.8, we see optical pumping into the $F = 2$ ground state that we are attempting to cool on. Examples of this optical pumping can be seen in Fig 4.3, and a plot showing how the relative amplitudes of the two states shift with the level of the cooling level lock can be seen in Fig 4.4.

While we see optical pumping, we see no measurable change in the width. This is shown in Fig 4.5 which scales the $F = 2 \rightarrow F'$ peaks of the two scans in Fig 4.3. The peak where the cooling laser is on resonance lies slightly within the peak where the cooling laser is off resonance. This could signify slight cooling, but the effect is small enough that it could be caused by our frequency calibration. We can roughly determine the detuning by scanning the cooling and re-pump lasers and calibrating the frequency based on the splitting between peaks. Then by looking at the voltage level of the level lock we can find the detuning from the center frequency. For the
4.5. PROGRESS TOWARDS COOLING

(a) Cooling system off resonance

Figure 4.3: Examples of the $D_2$ lines of lithium interacting with the probe laser. We can see in (b) that when the cooling laser is locked near resonance, the amplitude of the $F = 2 \rightarrow F'$ increases, as we would expect for our re-pumped system.
CHAPTER 4. RESULTS

Figure 4.4: Here we see how the amplitudes vary when we lock closer to the resonance. The higher the level of the PMT signal, the closer to resonance we are locking, and the more optical pumping we see.

optically pumped scan in Fig 4.5 the cooling laser is detuned about -20 MHz from the resonance, and the difference in width is consistent with what we expect as seen in Fig 2.5.

There are a few things we need to check to make sure we expect to see cooling. Firstly, since there is a collimation plate after the initial interaction region, we need to make sure the atoms we think we are cooling are getting to the main interaction region, rather than just hitting this plate. Since we see the correct optical pumping, we know that the cooling and re-pump lasers are aligned well enough so that the atoms we are exciting in the first interaction region are getting through the collimation plate to the main interaction region. In particular this means our vertical alignment is good enough, however, the horizontal alignment could still be a little off. The velocity class we are cooling around is dependent on the angle of the cooling and re-pump lasers relative to the atomic beam, since the zero velocity class is defined as the one perpendicular to these laser beams. Thus it’s possible that, even though
Figure 4.5: Were we have overlapped the peak without the cooling lasers on resonance, with the optically pumped peak. We see only a slight change in the width, which could be due to cooling, or our frequency calibration.
we are exciting atoms that make it through the plate, we are cooling to a horizontal velocity class that is dieing on the plate. We can correct for this by sending the probe laser through both interaction regions. Then when we scan the probe laser can see where the zero velocity classes of each region are relative to each other. Then we can change the horizontal alignment of the beam in the first interaction region such that the zero velocity classes line up. By aligning the cooling and re-pump lasers to this angle we should be cooling around the appropriate velocity class. This leaves the detuning of the cooling laser as the most likely reason we do not see cooling, and this is consistent with what we expect from our simulations. Hopefully, with a few improvements, we should be able to lock closer to resonance and see cooling.
Chapter 5

Conclusions and Future Improvements

5.1 Transverse Cooling

With the system as it is now we should be close to achieving transverse cooling; we only need a reliable method to lock the cooling laser closer to the resonance, or make the re-pump lock more robust to the modulation of the dither lock. Once we can reliably cool the atoms, we can try to run the experiment with the comb, and resolve the inconsistencies in our fine structure splitting measurements on the $D_1$ lines, while decreasing our uncertainties. Then, with our narrower line-width we should be able to make preliminary measurements of the less resolved $D_2$ lines and investigate quantum interference effects.

5.2 Future Improvements

Our data is sensitive to the linewidth of the probe laser. If the linewidth changes over the peaks, this would shift the peaks, and is likely part of the larger scatter we get when looking at data over multiple days. Utilizing the Pound-Drever-Hall
lock used to scan the probe laser while looking for cooling, we could pre-stabilize the probe laser to the Fabry-Perot cavity when return to running with the comb, which will hopefully improve the lock and narrow the linewidth.

Recently we came up to air and put the oven and the first interaction region on a bellows before the chamber. This allows for some flexibility in adjusting the angle of the atomic beam. By tuning this angle we will hopefully be able to adjust which velocity classes get through the collimation plate, and symmetrize the distribution in the main interaction region. In addition we will also be able to adjust the angle to ensure that the velocity class we are cooling around makes it through the collimation plate.

As a longer term goal we can work towards trapping the atoms in a Magneto-Optical trap (MOT) this would further narrow the peaks, and allow for longer interrogation times and better signal to noise. Building this MOT would require a Zeeman slower to initially cool the atoms along the direction of their motion, and a tapered amplifier to supply enough power to trap the atoms. With a MOT we could also investigate higher lying states with direct frequency comb spectroscopy.
Bibliography


