

Towards Feshbach Resonances in the S-WAVE Channel

by

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Author's Declaration

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

I understand that my thesis may be made electronically available to the public.

Abstract

Ultracold molecules offer unique opportunities for studying quantum phenomena. This thesis presents work undertaken to repair and optimize an older experimental setup which last studied Feshbach resonances in the p -wave channel for ultracold [Sodium-Lithium \(NaLi\)](#) molecules. The primary focus of this work was on the process of repairing and reconfiguring the molecule machine back to a working state. This involved the repair of the optical systems which provide the laser light at the required frequencies. The characterization of the dual species atomic beam, leading to the replacement of the [Sodium \(Na\)](#) and [Lithium \(Li\)](#) sources. The re-optimization of the Zeeman slower current and light alignment. The alignment, configuration and optimization of the magneto optical trap, along with configuration of the transfer processes to the magnetic trap. Significant effort was dedicated to evaporative cooling, where we reached near quantum degeneracy temperatures for both species. With both ultracold gases, we optimized the transfer into an optical dipole trap where we could sweep a magnetic field to produce Feshbach molecules. Progress was made towards [Stimulated Raman Adiabatic Passage \(STIRAP\)](#) for efficient transfer into the triplet ground state. Although the final step in the [STIRAP](#) process was not completed, the successful detection of Feshbach molecules was achieved. This work provides a solid foundation for future experiments, including the completion of the triplet ground-state molecule formation and enhanced stability which will allow for the exploration of s -wave Feshbach resonances in [NaLi + NaLi](#) collision complexes. This thesis contributes to the understanding of ultracold molecular interactions and offers insight into the problems encountered in the process of restoring and optimizing a complex experimental apparatus.

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Dedication

This thesis isn't just a product of my efforts but a testament to all the support and guidance I've received by everybody mentioned above. I'm deeply grateful for all the help I've received and I feel privileged.

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List of Abbreviations

AO Acousto-Optical Modulator [28](#), [39](#), [40](#)

BEC Bose-Einstein Condensation [x](#), [2](#), [7](#), [32](#), [34](#)

BEC-BCS Bose-Einstein Condensate to Bardeen-Cooper-Schrieffer [2](#)

BEC3 Bose-Einstein Condensation Lab 3 [x](#), [4](#), [6](#), [15](#), [25](#), [40](#), [50](#), [56](#), [58](#), [59](#), [63](#), [65](#), [66](#)

CCD Charge-Coupled Device [ix](#), [11](#)

Cs Cesium [3](#)

ECDL External Cavity Diode Laser [56](#)

EOM Electro-Optical Modulator [56](#)

IQC Institute for Quantum Computing [iv](#)

KRb Potassium Rubidium Molecule [3](#)

Li Lithium [iii](#), [ix](#), [xi](#), [4](#), [15](#), [16](#), [35](#), [50](#)

M.I.T Massachusetts Institute of Technology [2](#), [4](#), [6](#)

MOT Magneto Optical Trap [ix](#), [2](#), [6](#), [7](#), [9](#), [11](#), [13](#), [18–22](#), [24](#), [27](#), [28](#), [35](#), [40](#), [67](#)

Na Sodium [iii](#), [ix](#), [5](#), [15](#), [17](#), [18](#), [35](#), [43](#)

NaLi Sodium-Lithium [iii](#), [xi](#), [4–6](#), [35](#), [36](#), [47](#), [53](#), [56](#), [59](#), [62](#), [67](#)

ODT Optical Dipole Trap [x](#), [6](#), [36](#), [38–42](#), [45](#), [65](#), [67](#)

PD Photodiode [22](#), [40](#)

PDH Pound-Drever-Hall [56](#)

PID Proportional-Integral-Derivative Controller [40](#)

RF Radio Frequency [x](#), [xi](#), [29–35](#), [43](#), [44](#), [62–64](#)

RMS Root Mean Square [15](#)

STIRAP Stimulated Raman Adiabatic Passage [iii](#), [3](#), [5](#), [6](#), [36](#), [52](#), [56](#), [66](#), [67](#)

ToF Time of Flight [41](#)

UHV Ultra High Vacuum [6](#)

UW University of Waterloo [iv](#)

VCO Voltage Controlled Oscillator [56](#)

Chapter 1

Introduction

1.1 History of Ultracold Chemistry

In the following introduction, we're going to briefly discuss the history of ultracold atoms, the making of molecules, and the beginnings of ultracold chemistry. There is a long history of theoretical and experimental developments which led up to the modern achievements that are currently being pushed forward in labs around the world.

For this introduction, we will jump into the story in the 1980s where most of the techniques we use to this day were developed. I cannot do complete justice to all those who have contributed along the way. We will focus on specific developments and seminal papers that highlight key aspects of our current understanding¹.

1.1.1 Ultracold Atoms

The quest for ultracold atoms began much earlier, but it wasn't until the 1980s that practical techniques were developed, opening up the possibility of achieving ultracold states of matter. In 1985, Chu et al. [5] demonstrated the first experimental results for cooling atoms with optical molasses. Chu et al. demonstrated a cooling method that allowed for temperatures as low as $240\mu\text{k}$. This method opened up the possibility of reaching the Doppler cooling limit.

¹For those interested, a detailed historical account up to Bose-Einstein condensation can be found in William Phillips's Nobel Lecture: 'Laser cooling and trapping of neutral atoms,' which offers fascinating details of contributors to this field.

By 1988, Phillips et al. [37] showed that you could achieve temperatures lower than the Doppler cooling limit. However, in the mid 1990s it became clear that laser cooling of neutral alkali atoms had reached a maximum phase-space density. This technique could not be used to achieve [Bose-Einstein Condensation \(BEC\)](#), but, it did lay the foundation in preparation for what was to come next.

In parallel with the advancements in cooling, David Pritchard developed the first [Magnetic Optical Trap \(MOT\)](#), in 1987 which uses magnetic fields in combination with laser cooling to trap and cool atoms. [38]. Through steady improvements of these techniques and experimental equipment, researchers began to achieve an ever increasing density of trapped atoms at ever decreasing temperatures.

Another major development that came at about the same time was the first demonstrations of magnetic trapping and eventually evaporative cooling of dilute gases of neutral atoms. Although evaporation occurs in nature, it wasn't applied to atom cooling until 1988 [25]. Researchers could get to low temperatures with magnetic traps but the phase-space density remained too low.

Now, in the early 1990s, many of these techniques began to coalesce and open up the possibility of reaching ultra low temperatures and achieving [BEC](#). This led to a now infamous scientific race between competing labs, which culminated in 1995 when Cornell, Wieman et al. reported, in a now seminal paper, the first ever evidence of Bose-Einstein condensation [1].

Soon after, the team at [Massachusetts Institute of Technology \(M.I.T\)](#), led by Wolfgang Ketterle, also achieved Bose-Einstein condensation, however in a more robust system. The machine at [M.I.T](#) allowed for a greater number of atoms to be condensed and easier experimental access to make, probe, and understand this new state of matter. This led to an explosion of research into the behavior of Bose-Einstein condensates [22] and ultimately won Wolfgang, Eric, and Carl a Nobel Prize.

1.1.2 Making Molecules

In 1999, the first quantum degenerate Fermi gas had been achieved [7]. This sparked a flurry of experiments measuring the interaction properties between bosonic and fermionic atoms. Researchers investigated the [Bose-Einstein Condensate to Bardeen-Cooper-Schrieffer \(BEC-BCS\)](#) crossover, which is a crossover in the nature of the bound pairs that Bose condense to give a Fermi superfluid. They uncovered new quantum phases and many-body phenomena. And as the field matured, the specter of possibly forming molecules arose.

In the early 2000s, researchers began binding two ultracold atoms of the same species into molecules. The two main processes that were developed are photoassociation and magnetoassociation. The first uses laser light to bind two atoms into a long-range molecule [19]. The second uses magnetic fields to induce a Feshbach resonance that binds atoms into a highly excited state molecule [23]. Each offered their own advantages and disadvantages but both methods are still used today.

By the mid 2000s, the first experimental techniques for the conversion of Feshbach molecules into stable ground-state molecules appeared. The group Grimm and Chin created used Feshbach resonances to form **Cesium (Cs)** dimers [14]. Then Ospelkaus et al. created the first ground-state **Potassium Rubidium Molecule (KRb)** by first combining them into high energy state via Feshbach resonance, and then used **STIRAP** to transfer down into the ground state [29].

Not long after, researchers began working on bringing molecules down to the ground state at ever decreasing temperatures and increasing phase-space densities, just as the previous generation did for ultracold atoms. This opened up the avenue for studying interaction properties and dynamics of molecules. For example, Quemener et al. studied the collisional properties of polar molecules in the quantum regime [39]. With these improved techniques, the possibility of using ultracold molecules as a platform for studying chemistry became a real possibility.

1.1.3 Ultracold Chemistry

With high phase-space density at ultracold temperatures being achieved, along with the precision measurement of collisional interactions, researchers began to demonstrate that we could control chemical reactions.

Notably in 2010, Ospelkaus et al.’s first observation of the reaction dynamics at ultracold temperatures of **KRb + KRb** reactions [32]. The researchers found that preparing fermionic molecules in a single quantum state, they could observe *p*-wave dominated collisions. And by preparing them in different states, the reaction rates could be enhanced by two orders of magnitude. In this way they show that a clear understanding of molecular reactivity could be found using quantum statistics and threshold laws to understand how chemical reactions proceed at ultra low temperatures.

In 2013, a group of researchers led by Deborah Jin and Jun Ye, used dipolar interactions of molecules in 3D lattices to realize a lattice spin model [50]. The spin information is coded in the rotational states of molecules. The resonant exchange of angular momentum between them gives us a spin exchange interaction. They showed that, in this system,

they could suppress the loss rates by continuously probing the state, in what is called the continuous quantum Zeno effect. These studies opened up control of direct long-range spin interactions.

Researchers also began to investigate the energetics of reactions. For some molecules, exchange reactions forming homonuclear dimers are energetically allowed but some trimer formation reactions are energetically forbidden. These results show that the stability of quantum gases is highly dependent on the allowed interactions. It's not just that any two molecules can collide; but some collisions have little to no effect [17].

Also the study of transient intermediate complexes, a temporary bound state formed when two molecules interact during a collision, in ultracold molecule collisions began. Gas phase molecular reactions complexes last on the order of picoseconds before dissociating so they're not particularly externally influenced. However, with ultracold molecules, one can show a significant photo excitation in the presence of continuous wave lasers as well as increasing the lifetime of the complex by nearly nine orders of magnitude [24]. This opened up the possibility of probing the structure spectroscopically and untangling reaction dynamics.

1.1.4 BEC3

In recent years, as the field has become more accessible, it has also broadened out. The scope of research covers so much fertile ground that it's not feasible to cover everything in this brief overview. So, from here on I will focus on the contributions and direction that [Bose-Einstein Condensation Lab 3 \(BEC3\)](#), the [NaLi](#) lab at [M.I.T](#), has directly led or contributed to.

The exploration of Feshbach resonances and their role in forming ultracold molecules at [BEC3](#) started in 2013. Collisions between [Li₂](#) and free [Li](#) atoms were shown to deviate significantly from the universal rate² predictions near Feshbach resonances [49]. Feshbach resonances occur when a bound state of the system's potential energy curve is tuned to coincide with the energy of the colliding particles, leading to a strong enhancement of interaction strength. This results in deviations from the universal rate predictions because the interactions become highly dependent on the specific details of the molecular potentials involved.

The group performed photoassociation spectroscopy to study the excited triplet molecular potential. They observed 50 vibrational states and their substructure [41]. This was

²The universal loss rate refers to the rate at which pairs of atoms or molecules are lost due to inelastic collisions.

the first observation of the $c^3\Sigma^+$ potential as well as the first experimental achievement of photoassociation with [NaLi](#).

Continuing on this success, this was followed by two photon spectroscopy of the [NaLi](#) triplet ground-state [40]. The team took an ultracold mixture of both sodium and lithium and photo associated them into an excited triplet molecular state. The team then measured the vibrational state binding energies, the potential fitting curves, line strengths, and the rotational splitting of the lowest vibrational state.

By 2017, the group had achieved the creation of fermionic dipolar [NaLi](#) molecules in their triplet ground-state. They used magnetoassociation across a narrow Feshbach resonance to form a highly excited Feshbach molecule; and then, using [STIRAP](#), they brought the molecule down to the triplet ground-state[42]. Using electron spin resonance spectroscopy, the team measured the hyperfine structure of this molecular state.

In 2020, the lab successfully sympathetically cooled [NaLi](#) molecules down to nanokelvin temperatures through collisions with [Na](#) atoms. They discovered that the ratio of elastic to inelastic collisions is sufficiently large to support collisional cooling [45]. This opened up another method for cooling molecules and increased the phase-space density achievable, which ultimately resulted in a method to drive temperatures into the nanokelvin regime. An important idea came out of this work, that it may be possible to create quantum degenerate dipolar molecules and possibly use them to cool other molecules.

The next advancement to come out of the lab was the magnetic control of the reactive scattering of [Na](#) with [NaLi](#) molecules [46]. By controlling the phase of the scattering wave function through a Feshbach resonance, the group modified the loss rate by more than two orders of magnitude. This work constituted direct control over reaction rates via quantum interference.

In a productive series of experiments, the group showed that there is a very narrow Feshbach resonance in collisions between two triplet ground-state molecules. This resonance enhances the collisional loss rate by two orders of magnitude above the background loss rate. These observations proved the existence of coherent intermediate complexes that are long-lived, further opening up the possibility of controlling chemical reactions [34].

Further developments in increasing phase-space density were made at this time. Using a magnetic trap, along with sympathetically cooling [NaLi](#) molecules, extends the lab’s capability to explore ultracold chemical reactions [33].

In the most recent results, the spectrum of Feshbach resonances in [NaLi](#) + [Na](#) collisions were mapped out [35]. The team discovered 25 resonances, which is in agreement with the theoretical calculations, showing that the process at work are spin/rotation and spin/spin coupling, along with the anisotropic Coulomb interaction with [Na](#).

1.2 Thesis Structure

In Jamison’s Lab, I developed an injection lock with a stability control system. I constructed a lithium spectroscopy cell and configured the surrounding optics and electronics to lock the main laser to the crossover feature. Additionally, I assembled and tested a new crucible design for the mixing machine, built and prepared the [MOT](#) and main chamber [Ultra High Vacuum \(UHV\)](#) assembly, wrote code, repaired and tested equipment, and set up various optical systems. However, I will not discuss these tasks due to space constraints. I will instead focus on my work at [M.I.T](#) under Wolfgang Ketterle in his ultracold [NaLi](#) molecule experiment lab ([BEC3](#)).

The structure of the thesis is as follows: each topic begins with general theory, followed by a discussion of the experimental setup (where fruitful). Then we’ll cover some of the issues we faced and the solutions we implemented including tests, repairs, troubleshooting, key insights, calibrations, and benchmarks. The goal is to provide new graduate students with some initial insights into the underlying physics, an understanding of the experimental setup, and the process of troubleshooting inevitable problems.

In chapter two, we will cover the trapping and cooling of atoms. Starting from how we generate and characterize a dual species atomic beam. Then we’ll discuss the theory and optimization of a Zeeman Slower. Next, we will cover the theory and optimization of a [MOT](#) which leads us to the theory of how we magnetically trap and evaporatively cool atoms.

In chapter three, we begin discussing how we make triplet ground-state sodium lithium molecules. First, we cover the theory for an [Optical Dipole Trap \(ODT\)](#). Secondly, we show how we transfer to the high field by performing a Landau-Zener sweep. Thirdly, we cover the general theory of Feshbach molecule formation along with how to measure and optimize the process. And lastly, we briefly review [STIRAP](#), the technique we use to bring the molecules into the triplet ground-state.

In Chapter 4, we outline the future directions of our experimental setup. Building on the groundwork established over the past decade, we will search for *s*-wave Feshbach resonances between molecules, measure the collisional loss rate as a function of rotational energy, and assess the coherence of mixed rotational states in the magnetic trap. Additionally, we will attempt to photo-associate molecules into an excited four-body state.

Chapter 2

Trapping and Cooling Atoms

This chapter will cover how to trap and cool atoms down to near quantum degeneracy temperatures. We will discuss the creation of a two species atomic beam, how we slow the atoms down with a Zeeman slower, the capture of atoms into a [MOT](#), how we then transfer them into a magnetic trap, and the process of evaporative cooling down to [BEC](#).

I have provided an updated diagram of the apparatus for reference [A.1](#). The optics and electronics surrounding the main setup have changed over the years, but the core apparatus has remained.

2.1 Atomic Beam

In the following section, we will discuss a few details about our source of atoms: We will discuss how we create a two-species oven, the velocity distribution of the beam, and the two tests we conducted to diagnose the need for an oven change.

2.1.1 Theory

Atomic beam production in a dual species oven begins with a vapor of the two elements, sodium and lithium, within separate high-temperature chambers [A.1](#). The sodium beam first passes through an orifice into the lithium chamber; and because the lithium is kept at a higher temperature, the sodium tends to pass straight through. (As we will discover, over long periods of time this mixing does occur and has consequences for the lifetime of the

lithium.) In the mixing chamber, we now have a vapor that is both lithium and sodium. This vapor then effuses through two orifices which results in a collimated atomic beam.

The first orifice acts as a point source for the atomic vapor. However, due to the alignment of the second orifice, the setup preferentially allows atoms moving parallel to the axis to pass through. The degree of collimation is determined by the aspect ratio of the orifices, specifically the ratio of their separation to their diameter, as well as their relative spatial positioning. Well aligned, small orifices with large separation significantly improve collimation and reduce angular spreading.

The velocity distribution of atoms in the atomic beam is described by a distribution similar to Maxwell-Boltzmann [8]. It characterizes the statistical distribution of speeds in a gas of non-interacting particles at thermal equilibrium. However, the process of effusion through the orifices introduces a selection bias towards higher longitudinal component of velocity [30]. This is due to the fact that atoms with higher velocities are more likely to pass through the orifices given their higher kinetic energy; and consequently, their increased likelihood of moving towards and through these narrow openings.

The Maxwell-Boltzmann distribution for the speed of particles is

$$f(v) = 4\pi \left(\frac{m}{2\pi kT} \right)^{\frac{3}{2}} v^2 e^{-\frac{mv^2}{2kT}}, \quad (2.1)$$

where m is the mass of an atom, k is the Boltzmann constant, T is the temperature of the gas, and v is the speed of the particles [8].

When considering the effusion through an orifice, the probability of an atom making it through the hole is proportional to its longitudinal velocity towards the orifice. This leads to an additional factor of v [30] in the distribution

$$f_{\text{eff}}(v) = 4\pi \left(\frac{m}{2\pi kT} \right)^{\frac{3}{2}} v^3 e^{-\frac{mv^2}{2kT}}. \quad (2.2)$$

There is a higher probability of faster atoms to effuse through. This results in a beam with a slightly shifted velocity distribution that favors higher velocities than would be predicted by the Maxwell-Boltzmann distribution alone.

A final important detail for our discussion is the growth of the beam diameter as a function of length. Although the beam is collimated, it still has some transverse velocity. We can model the beam's diameter D at a distance L from the orifice as being related to the transverse velocity $v_{\perp}$ [30]. Making a small angle approximation we have

$$D(L) = 2L \tan(\theta) \approx 2L\theta, \quad (2.3)$$

where θ is the angle of divergence. This is related to the average transverse velocity $v_{\perp}$ and the average longitudinal velocity $v_{\parallel}$. So the relationship between beam diameter and distance becomes

$$D(L) \approx 2L \cdot \frac{v_{\perp}}{v_{\parallel}}. \quad (2.4)$$

This shows us that the beam diameter grows linearly with distance from the orifice. The growing beam diameter impacts the flux we can slow down and catch in the [MOT](#).

2.1.2 Absorption Spectroscopy

As we repaired the system, we struggled to make a lithium [MOT](#). After some time, we began to suspect an issue with the lithium source itself. So we setup to measure the flux. We used a straightforward absorption spectroscopy scheme with our light intersecting with the beam just after the differential pumping tube.

Alignment is difficult due to limited access, however we used the differential tube itself as a guide [2.1](#). By shining the probe light onto the end of the tube we could then adjust the light down until the beam was just clipping the end of the tube where we would get the highest flux.

With the light aligned, we could be certain that we were intersecting the atomic beam. After processing an average of 1024 measurements, with and without the atomic beam present, the results were inconclusive [2.2](#). Because lithium's absorption is weak¹, along with the difficulties of detecting the signal given the location of our setup and the time required to break vacuum and change the lithium, we decided to confirm our results with fluorescence.

¹There are two main reasons for this: 1. The strength of the lithium transition is lower than that of sodium. And 2. lithium atoms are lighter than sodium atoms so there is greater Doppler broadening at the same temperature.

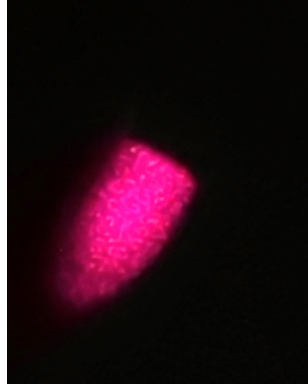


Figure 2.1: Using the differential pumping tube (between the oven and differential chamber just before the start of the slower) as a guide to align the probe light perpendicular to the atomic beam.

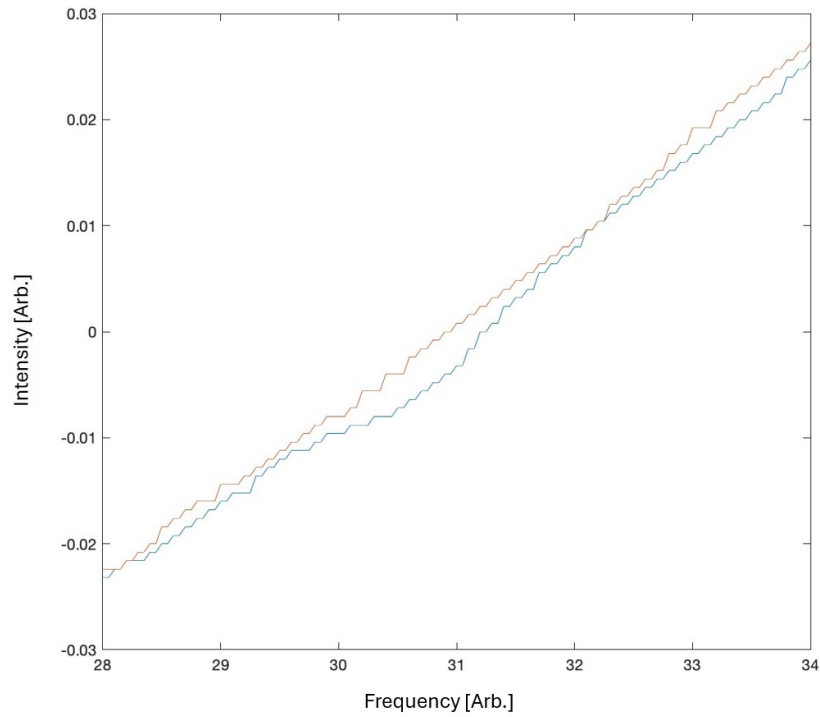


Figure 2.2: An average of 1024 measurements of absorption spectroscopy for lithium in blue and without the atomic beam present in red.



Figure 2.3: The [CCD](#) camera side of the fluorescence setup. There is limited access since this is at the top of the machine surrounded by the rest of the experimental setup.

2.1.3 Fluorescence Detection

The setup for fluorescence detection is straight forward. We installed a [Charge-Coupled Device \(CCD\)](#) camera after a lens that is placed so the image of the beam (considered as a point source) is collimated and falls on the sensor [2.3](#).

The resulting fluorescence image, after turning the oven up to 390C and turning the slower laser up as high as possible, was 60mW. After some post processing and averaging, this resulted in the following [2.4](#).

With this data confirming that the flux of the lithium beam was low, we decided to break vacuum and change the lithium cup. We followed the standard process; and as a result of the oven change, we successfully established a lithium [MOT](#).

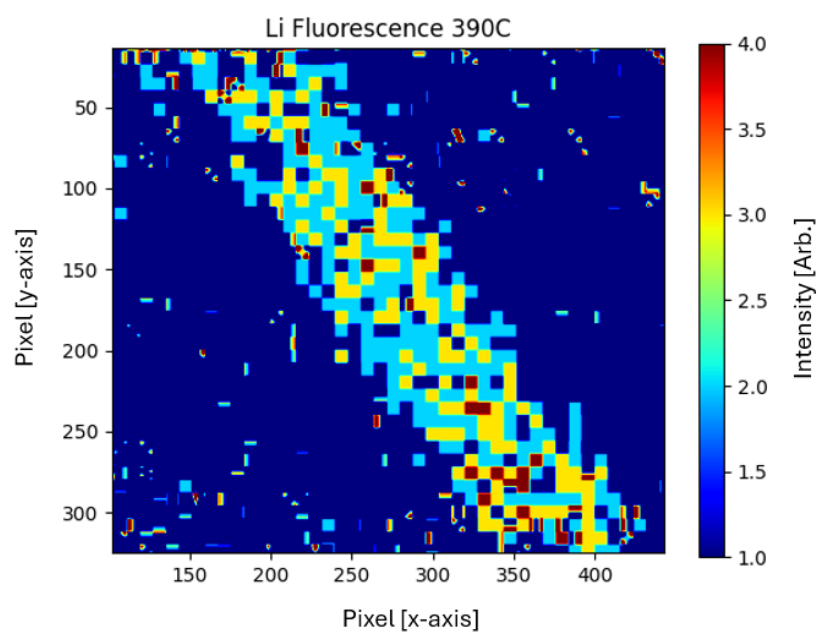


Figure 2.4: Lithium fluorescence image post processing. 2D array of pixel intensity values that range from 1 to 256. This definitively showed that the lithium flux was low and needed to be replaced.

2.2 Zeeman Slower

After the creation of an atomic beam, we need to slow the atoms down to a trappable velocity. In our experiment, we use a Zeeman Slower. In the following section, we'll discuss the general theory of Zeeman slowing, the main considerations to maximize the number of atoms slowed and ultimately trapped in our MOT, as well as the experimental setup and a technique for slower laser light alignment.

2.2.1 Theory

A Zeeman slower takes advantage of the Zeeman shift, where an atom in a static magnetic field experiences a change in the energy difference between the ground and excited states in proportion to the magnitude of a magnetic field. In essence, we can lift the degeneracies of the hyperfine levels (F) in a magnetic field, separating them into sublevels (m_F), each with different interaction strengths [8]. And if we consider the case where the magnetic field is weaker than the hyperfine interaction $A\vec{I} \cdot \vec{J}$, then the magnitude of the change is directly proportional to the magnetic field strength as

$$E = g_F \mu_B B m_F, \quad (2.5)$$

where g_F and m_F depend on the chosen substates [8]. This gives us a way to change the energy required for the atom to interact with light.

For example, let's choose the D_2 cycling transition for sodium. Here the stretched ground and excited states ($|F, m_F\rangle = |2, 2\rangle$ and $|3', 3'\rangle$) are connected via σ^+ polarized light [47]. So the difference in Zeeman shifts for this transition is

$$\begin{aligned} E &= \mu_B B (g'_F m'_F - g_F m_F) \\ &= \mu_B B \left(\frac{2}{3} 3 - \frac{1}{2} 2 \right) \\ &= \mu_B B, \end{aligned} \quad (2.6)$$

where the values of g_F are found in [8]. We see that the energy required for the transition is growing linearly with the magnitude of the magnetic field.

Now that we have a way to change the energy splitting between transition levels, we can discuss the force applied by classical laser light on an atom. In a two level system, in a steady state, the force applied is

$$F = \frac{\hbar k \Gamma}{2} \frac{s_0}{1 + s_0 + (2\delta/\Gamma)^2}, \quad (2.7)$$

where k is the wave vector for the given light, Γ is the line width, and s_0 is the fraction of the saturation intensity for the given transition with $\left(s_0 = \frac{I_l}{I_s}\right)$ and $I_s = \frac{\hbar c k^3 \Gamma}{12\pi}$, and δ is the Doppler and Zeeman detuning from resonance [28]. The force experienced by an atom depends on the lifetime of the interaction (since in an excited state the light cannot interact with the atom), as well as depending on the intensity of the light (more photons gives more ‘pushes’), and how far detuned that light is from the resonance frequency (how probable it is the photon interacts with the atom).

Focusing on δ , we see that the detuning consists of three parts,

$$\delta = \delta_0 - kv + \frac{\mu_B}{\hbar} B. \quad (2.8)$$

The first term is the frequency of the laser, the second frequency component accounts for the Doppler shift, and the final term accounts for the frequency change required for the Zeeman shift. From this it directly follows that the minimum and maximum magnetic fields are

$$\begin{aligned} B_{\min} &= \frac{\hbar k}{\mu_B} v_f \\ B_{\max} &= \frac{\hbar k}{\mu_B} v_i. \end{aligned} \quad (2.9)$$

We want the highest possible v_i in order to slow the largest fraction of the atomic beam velocity distribution. Since there is a maximum acceleration

$$a_{\max} = \frac{\hbar k \Gamma}{2m} \frac{s_0}{1 + s_0}, \quad (2.10)$$

where we solved (2.4) for v given that $\delta = 0$ and substituted into (2.3). This suggests that we want a long Zeeman slower to have plenty of time to slow the high velocity atoms down.

However, there is another more subtle issue: as atoms absorb photons, they spontaneously re-emit isotropically. Over time, this produces a random walk in velocity space

[44]. As a result, even though the longitudinal velocity of the atoms is being reduced, their transverse velocity acquires a non-zero [Root Mean Square \(RMS\)](#) value. Along with the geometric spreading of a beam, the atoms experience ‘transverse heating’ and expand further [31]. With these complexities in mind, we want to make the Zeeman slower as short as possible.

By imposing that the acceleration must be less than the maximum, we get a constraint on our magnetic field,

$$\alpha^2 \frac{a_{\max}}{B} \geq -\frac{\partial B(z)}{\partial z}, \quad (2.11)$$

where $\alpha = \frac{\hbar k}{\mu_B}$. This is the so-called ‘adiabatic following’ condition, the maximum change in magnetic field the atoms can follow without dropping out of the deceleration cycle. You can just set the RHS and LHS of (2.7) equal and solve for the longitudinal magnetic field profile,

$$B(z) = \frac{\hbar k}{\mu_B} \left(\sqrt{v_i^2 - 2a_{\max}z} \right) \quad (2.12)$$

but this wouldn’t allow for any fluctuations in the laser intensity or magnetic fields. So it’s common practice to stretch the magnetic field out by including a ‘noise’ factor, η , on the acceleration. If we assume that the noise is independent of position, which is good to first order with a short Zeeman slower, then we simply have

$$B(z) = \frac{\hbar k}{\mu_B} \left(\sqrt{v_i^2 - 2\eta a_{\max}z} \right). \quad (2.13)$$

For an initial velocity of 450 m/s and assuming full saturation, we can plot the optimal magnetic field profile for a [Li](#) Zeeman slower [2.5](#).

2.2.2 Experimental Setup

In [BEC3](#), we use a long ‘spin flip’ Zeeman slower [A.1](#). Originally designed for [Na](#), the slower’s magnetic field profile also works for [Li](#). Despite the inability to tune the field for specific velocity classes, the slower captures enough of both velocity distributions to be effective for the ratio of [Na](#) to [Li](#) we need for our experiments.

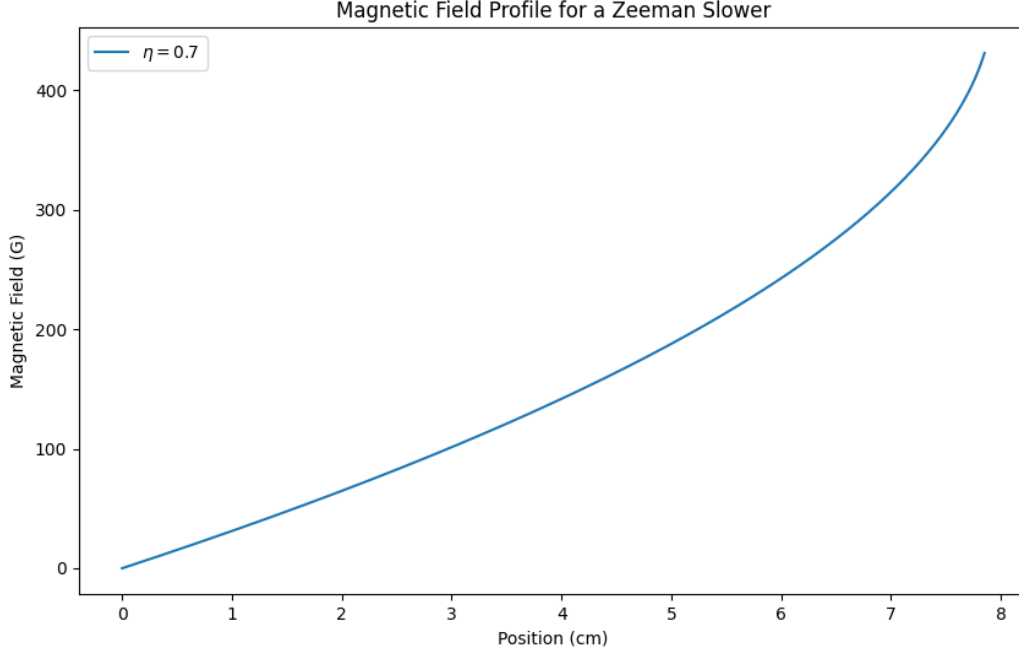


Figure 2.5: Optimal magnetic field profile for a Zeeman slower with noise parameter of 0.7 for [Li](#) with 450 m/s initial velocity and at maximum saturation intensity. Note: for physical insight this is plotted as $B_{\max} - B(z)$.

One major limitation of our slower setup is the increased divergence and transverse heating of the atomic beam from the long length of our slower. Even though we are slowing more of the velocity distribution, the greater length essentially cancels out the majority of the benefits. Most modern experimental setups now use slower measured in centimeters instead of meters².

2.2.3 Fluorescence Guided Alignment

A major issue with the design, which only becomes obvious after many years, is the difficulty in realigning the slower beam. All the slower optics are deep in the machine setup, which is now buried under all the other optics that have grown up around the chamber.

²Including the apparatus we are constructing in Jamison's Lab.

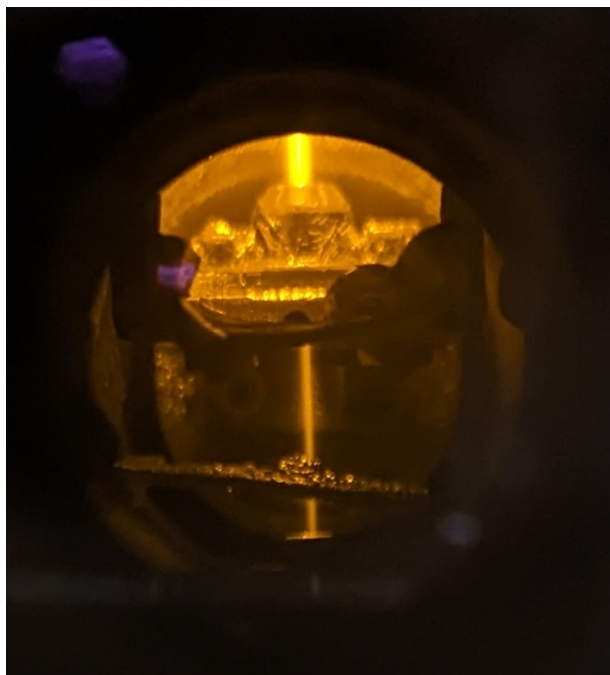


Figure 2.6: Fluorescence of the Na slower beam before the differential pumping tube directly after the oven.

Even when you gain access, there is no simple way to align the beam. It becomes a major project to fix the alignment and divergence if, and more likely when, the beam needs to be re-positioned. Ensuring the slower properly converges near the nozzle of the oven and is parallel to the slower inner walls (not reflecting and making it out the other side for example) is challenging.

One location where you have line of sight is after the differential pumping tube just before the oven nozzle 2.6. Although this gives you valuable information, it does not resolve the internal bouncing issue, nor does it tell you if the beam is clipping on openings of the differential pumping tube.

The way we resolved this problem was two fold. First, we setup a camera and lens system, perpendicular to the Zeeman slower path (see figure A.1 ‘window’) to image the fluorescence of the Na light passing through the atomic beam 2.7. By imaging the beam passing through the atomic beam you can measure the angle at which the light travels through, along with the fluorescence before the differential pumping tube, allows you to resolve the ‘reflecting’ problem. The clipping problem is similarly resolved via the fluores-

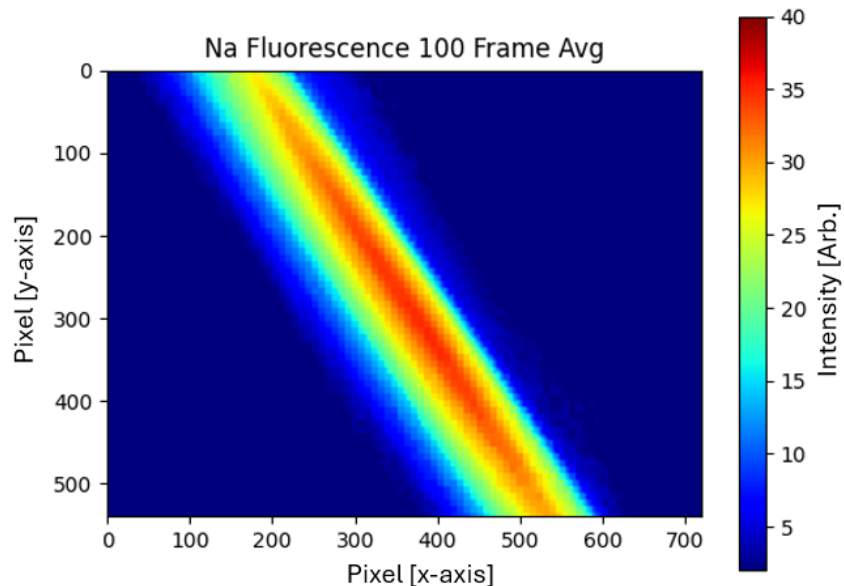


Figure 2.7: Fluorescence image of the [Na](#) light passing through the atomic beam used to align the slower light. 2D array of camera pixel intensity from 1 to 256. As can be seen by the asymmetry of the fluorescence, the slower light is slightly misaligned to the atomic beam.

cence. You can see how centered and large your light is relative to the atomic beam (this is not a perfect one to one mapping, but enough to move forward with minimal loss).

Once the slower was aligned, we could switch to absorption imaging of the [MOT](#). We setup a quick sequence in our hardware control software, Cicero, to repeatedly load and image, so we could adjust the slower beam to maximize the number of atoms captured.

2.3 Magneto Optical Trapping

Once the atoms have been Zeeman slowed to within the capture velocity, we can catch them in a MOT. In the following section, we will discuss how we use laser light and a magnetic field to trap atoms, along with a discussion about how we can model the trapping force. Then we'll briefly cover how we can increase the density using a 'Dark Spot'. And finally we'll talk about our experimental setup and discuss how we maximized the total number of atoms.

2.3.1 Theory

The main components of a MOT are a quadrupole magnetic field and 3 pairs of counter propagating laser beams from all axial directions. Both of these together create the region that traps atoms. For our discussion, we will simplify the model by reducing down to a single dimension, however, the model can be extended to three dimensions readily.

There are two important features of a quadrupole magnetic field, generated by two counter circulating, current carrying loops set up in the anti-Helmholtz configuration as shown in figure 2.8. First, for the cancellation of the B field at the center; and secondly, so the magnitude of the field is approximately linear in the region near zero. So as an atom moves away from the centre of our trap, it will experience an approximately linearly increasing B field, which along with the Zeeman shift, provides a spatially varying energy level.

The Zeeman shift, in the linear region of the trap, is

$$\beta z = \frac{g\mu_B}{\hbar} \frac{dB}{dz} z, \quad (2.14)$$

where β is a constant of proportionality, which is taken to be a constant in the region of interest.

Next, the force applied to an atom comes from the scattering of resonant light which is proportional to the photon momentum and the scattering rate. We can model this interaction as

$$F_{\text{scatt}} = \hbar k R_{\text{scatt}}, \quad (2.15)$$

where $\hbar k$ is the momentum and R_{scatt} is the rate at which the light scatters off the atom. The rate of scattering is related to the intensity of the light, the lifetime of the excited

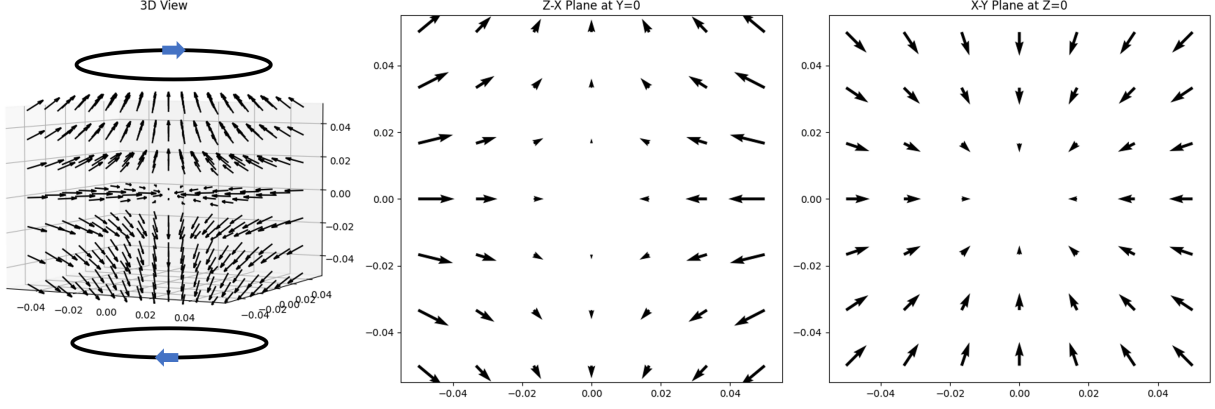


Figure 2.8: The quadrupole magnetic field used in a MOT, generated by two counter propagating current carrying loops. a) The 3D model. b) The field as seen from the side. c) The field as viewed from the top.

state and the detuning from the transition resonance [8] as

$$R_{\text{scatt}} = \frac{\Gamma}{2} \frac{I/I_{\text{sat}}}{1 + I/I_{\text{sat}} + 4\delta^2/\Gamma^2}, \quad (2.16)$$

which for a single beam gives

$$F_{\text{scatt}} = \hbar k \frac{\Gamma}{2} \frac{I/I_{\text{sat}}}{1 + I/I_{\text{sat}} + 4\delta^2/\Gamma^2}. \quad (2.17)$$

The atom is moving and so there is a Doppler shift we have to take into account. The shift in frequency seen by the atom relative to the resonant frequency of the transition is simply given by the light's wave vector times the velocity, $\vec{k} \cdot \vec{v}$. Simplifying the notation, we have that

$$F_{\text{scatt}}^{\pm}(\omega - \omega_0 \pm kv) \quad (2.18)$$

where the function F is to be evaluated at the point given by $(\omega - \omega_0 \pm kv)$, with the laser frequency ω , the resonant frequency ω_0 , and the Doppler shift kv .

In a MOT, we have counter propagating beams along each axis. To model this situation, we take the difference between the forces from opposite directions (e.g. the left and the right)

$$F_{\text{N}} = F_{\text{L}}(\omega - \omega_0 - kv) - F_{\text{R}}(\omega - \omega_0 + kv). \quad (2.19)$$

Since $kv \ll \Gamma$, we can Taylor expand

$$F_N = F_L(\omega - \omega_0) - kv \frac{\partial F}{\partial \omega} - F_R(\omega - \omega_0) - kv \frac{\partial F}{\partial \omega}, \quad (2.20)$$

because the two beams are the same the constant terms cancel and we're left with

$$F_N = -2kv \frac{\partial F}{\partial \omega}. \quad (2.21)$$

If we now make the reasonable assumption that the $\frac{dF}{d\omega}$ is constant over the low velocities we're interested in, we see that the force is simply a damping force $F_N = -\alpha v$. It's important to keep in mind that this kind of force does not restore the atoms to a specific position; instead, it slows them down, effectively providing a viscous drag that damps their motion³.

We want to trap atoms in a specific volume not just slow them down. So we need to introduce a spatially varying component. This is where the Zeeman shift term comes in

$$F_{\text{scatt}}^{\pm}(\omega - \omega_0 \pm kv \pm \beta z). \quad (2.22)$$

Following the same procedure as before, and using the fact that $\frac{\partial F}{\partial \omega} = -\frac{\partial F}{\partial \omega_0}$, we have

$$F_N = \alpha v - \alpha \beta z, \quad (2.23)$$

where the first term is a velocity-proportional damping force that provides viscous resistance slowing the atoms down and the second term is a position-dependent force that acts to restore atoms towards the centre of the trap. Together these forces result in a damped simple harmonic oscillator where the atoms experience a steady deceleration as they approach the zero magnetic field region at the centre of the trap.

The final idea we will discuss is a 'Dark Spot' [MOT](#). In a standard [MOT](#), atoms are continuously excited by resonant laser light and may occasionally transition into a dark state that does not interact with the trapping light. These atoms are usually recycled back into the cycling transition using a repumping laser. However, at the centre of the trap, high atomic density leads to an increased rate of light-induced collisions where some atoms gain enough energy to escape. By introducing a dark spot overlapping the centre of the magnetic field where the repumping light is blocked, the atoms stop scattering photons

³This is called 'Optical Molasses' since the interaction is as if an object was falling through a thick fluid like molasses, see [\[8\]](#) for details.

for some time [22]. This reduces the rate of collisional losses. When these atoms drift out of the dark spot, they will reenter the repumping light and be restored to the cycling transition and be pushed back towards the center.

2.3.2 Optimizing a MOT

The process of optimizing a MOT is straightforward. There are five factors that need to be optimized. First, the alignment of each beam to spatially align the point of overlap with the zero centre of the magnetic field. Using the fluorescence of the MOT as a guide, you can maximize the signal on a Photodiode (PD) for each axis direction.

Second, the intensity of each beam, at the final viewport, should be the same so as not to introduce an imbalanced scattering force along any direction. One needs to be careful here as an imbalance initially only moves the centre of the MOT relative to the magnetic field. This not only shows up as a displacement but as an increase in the temperature. You begin with just a power meter at the viewports, but eventually, the MOT is optimized relative to how much is transferred into the magnetic trap, where we can measure the total number of atoms at a given temperature most accurately.

Thirdly, the polarization of each beam needs to be maximally circularly polarized. The ellipticity of your polarization has a similar effect as a power imbalance since the scattering probability is directly related to the polarization. With one significant difference, the component that is oppositely polarized will push the atoms on the other side of the zero crossing outward.

And the fourth is the frequency of the light. The detuning required for sodium is approximately 25 MHz. However, this number can be fine tuned. As you increase the detuning you increase the size of the trap (since the atoms need to pick up greater spatial shift before becoming on resonance.) However by doing so, you also allow faster atoms (which need a greater Doppler shift) and thereby increase the temperature of the MOT. There is a trade off between these effects. Here we do the same optimization as before where we maximize the number of atoms in the magnetic trap at a given temperature by adjusting the detuning.

Finally, the fifth factor is the alignment of the 'Dark Spot'. The Dark Spot size, alignment, and focal point are all critical. Thankfully, the size of the spot is fixed. This leaves only the angular alignment and the divergence angle of the light hitting the spot to be aligned so that we are casting the shadow on the centre of the trapped atoms. The ideal dark spot creates a conical cylinder with the waist at the centre of the MOT. Here we can optimize how many atoms are transferred into the magnetic trap at a given temperature

by adjusting the distance between the two beam shaping lenses and the angular alignment of the final two mirrors⁴.

⁴As you may have noticed, the best measure for each of these is going into the magnetic trap, evaporatively cooling to a set temperature, and then imaging the atoms. However, the initial fluorescence measurement is indispensable for quickly getting some signal, which you can then optimize.

2.4 Magnetic Trapping

The general principle of magnetically trapping atoms is straightforward. In the following section, we will discuss where the trapping force comes from, how the magnetic trap confines atoms in both the radial and axial directions, and how we can model the oscillation frequencies of the trap. Then, we will discuss how we can trap atoms of different species in the same magnetic field. And finally, the main experimental difficulty is transferring atoms into the magnetic trap from a MOT, so we will finish this section by discussing how we tackled these issues and optimized the number of atoms captured.

2.4.1 Theory

Atoms are trapped in magnetic fields due to their magnetic dipole moments associated with their electron spin [8]. The mechanism for trapping is the interaction of the magnetic dipole moment with the magnetic field, which is described by the force due to magnetic field gradients [22]. The force experienced by a magnetic dipole in a magnetic field is given by

$$\vec{F} = -\nabla V = -\nabla(\vec{\mu} \cdot \vec{B}). \quad (2.24)$$

The force depends on the gradient of the magnetic field. It acts to move the atom towards regions of higher magnetic field strength if the dipole is aligned with the field. Also, a torque is exerted on the dipole that aligns it with the field

$$\vec{\tau} = \vec{\mu} \times \vec{B}. \quad (2.25)$$

The potential energy $V = \vec{\mu} \cdot \vec{B}$ associated with the Zeeman effect is

$$V = g_F \mu_B m_F B, \quad (2.26)$$

where g_F is the Landé g-factor, μ_B is the Bohr magneton, and m_F is the magnetic quantum number. Assuming adiabatic following, defined as when the atom's magnetic moment maintains its orientation relative to the slowly varying magnetic field, the resulting force in the z-direction is

$$F = -g_F \mu_B m_F \frac{dB}{dz}, \quad (2.27)$$

which shows that the force is proportional to the gradient of the magnetic field.

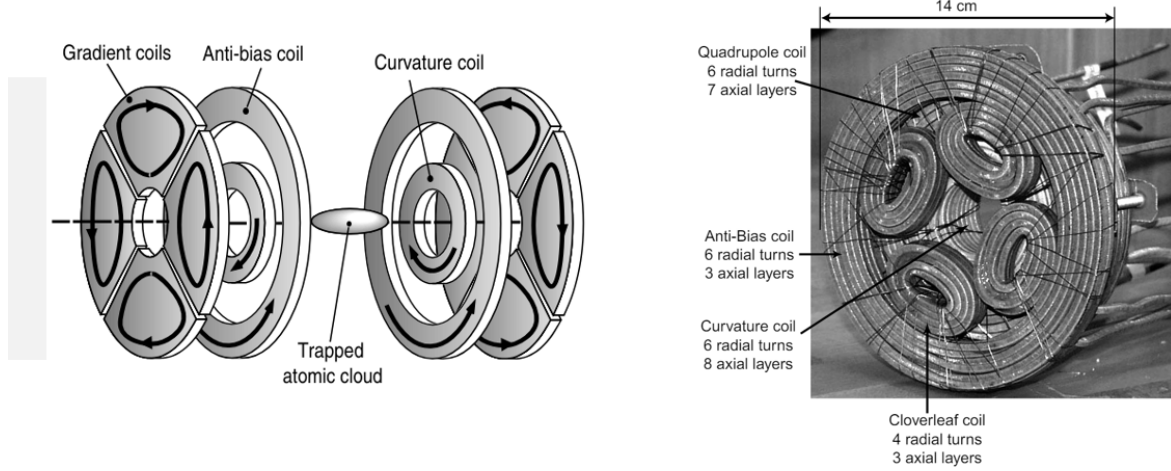


Figure 2.9: The magnetic coils layout including the cloverleaf gradient coils, the curvature 'pinch' coils, and anti-bias coils. Also a picture of the actual coils used in the experiment [3],[11].

If the temperature of the atoms is sufficiently low, their kinetic energy is not enough to overcome the potential barriers created by the magnetic field gradients. Therefore, the atom trajectories can be confined within closed loops, effectively trapping them in the magnetic field [22].

The main consideration then is the setup of the magnetic field such that we can create a sufficiently localized trapping potential. In BEC3 we have a cloverleaf magnetic field trap with additional curvature coils 2.9.

The B field for a cloverleaf configuration is

$$\mathbf{B} = g(z\hat{i} - y\hat{j}) + g(y\hat{i} + z\hat{j}) + g_x x^2 \hat{k}, \quad (2.28)$$

where g and g_z are the gradient strengths of the horizontal and vertical components of the field respectively. As shown in 2.10, this results in a radially increasing gradient in the YZ plane and linear gradients in the ZX and XY planes. The trap provides radial, but not coaxial confinement because the atoms are free to move along the x-axis.

To resolve this problem, we introduce another set of two coils that provide a curvature along the coaxial direction 2.11. This setup creates a trapping region that is shaped as an ellipsoid. The magnetic field gradient is different along the semi-major and semi-minor

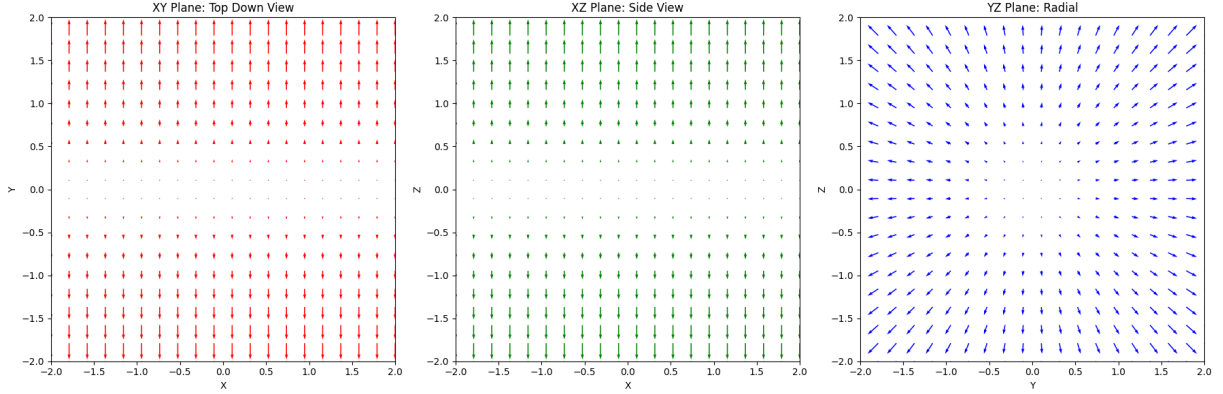


Figure 2.10: From left to right we have the magnetic field in the XY plane (as viewed from the top down), the XZ plane (side view), and the YZ plane (coaxial view).

axes which results in two different oscillation frequencies.

In our configuration, if we consider a small region at the centre of the trap (where $b'r \ll B_0$) the atoms will experience a harmonic potential. This approximation allows us to find the oscillation frequency in both directions. In the radial direction we have

$$w_{\text{radial}} = \sqrt{\frac{g_F \mu_B m_F}{m B_0}} \times b', \quad (2.29)$$

where b' is the magnetic field gradient. The same procedure can be done for the coaxial direction [8].

A final note: we are dealing with two different species of atom. The magnetic dipole moments for two alkali atoms in the ground state, when we're restricted to the stretched state, are the same. However, their masses are significantly different. The mass of sodium is roughly four times that of lithium. Even though both atoms are subject to the same trapping potentials, they experience different accelerations and this results in different oscillation frequencies. Nevertheless, these frequencies are close enough that both atomic species can be trapped by the same magnetic field [43]. There are other practical implications of these differences beyond oscillation frequencies, such as collisional dynamics or state-dependent trapping efficiencies, but we will not discuss these other than to mention their existence.

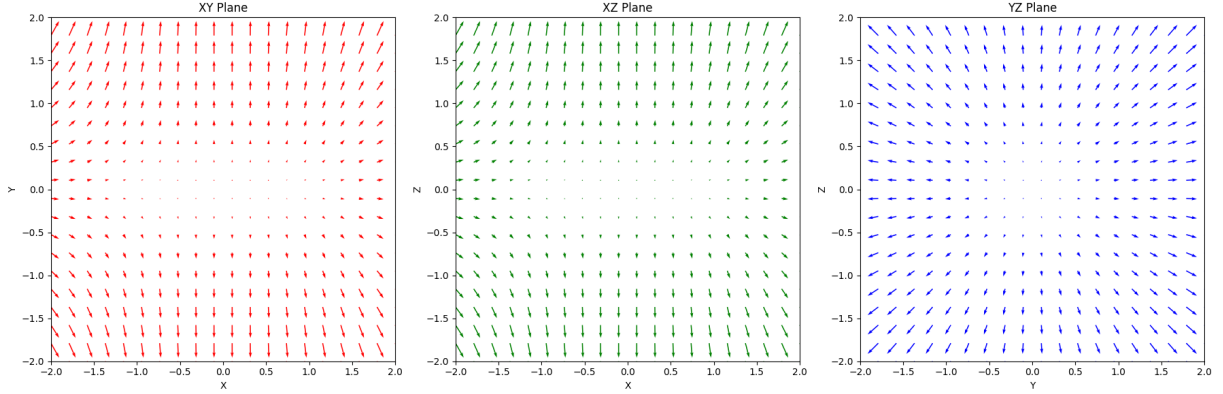


Figure 2.11: The magnetic field from a cloverleaf setup along with curvature coils providing a gradient in the coaxial (x -axis) direction.

2.4.2 Aligning The MOT to The Magnetic Trap

The successful transfer of atoms to a magnetic trap is important for our experiment. We therefore try to achieve optimal spatial overlap between the MOT and the magnetic trap with minimal loss of atoms and at as low a temperature as possible.

We have additional bias fields along the x , y , and z axes. By modifying the relative strengths along each axis, we can alter the balance of forces acting on the atoms. This effectively changes the spatial location of the cloud. It's effective for moving the MOT; however, it is important to note that altering the fields can also modify the trapping potential, potentially affecting the temperature.

Another control knob is the intensity of the trapping light. Precise measurement of light intensity is challenging due to focusing effects at the atomic location. Generally, we utilize a power meter placed at the viewport to gauge the intensity, further refining it by observing the shape of the MOT. Adjusting the power, and therefore the intensity, can inadvertently increase the potential energy of the atoms by pushing them further away from the centre of the magnetic field. When the atoms are transferred to the magnetic trap, this adds energy to the atoms increasing the atoms initial temperature in the trap.

The purity of circular polarization from the counter-propagating beams also plays a crucial role. Imbalances in polarization can reduce the scattering force along an axis, similarly leading to the centre of the MOT being dislocated from the magnetic trap center. Before each viewport, we have quarter wave plates for this reason. The quarter wave plates effect 671nm and 598nm light slightly differently, so the optimal setting is a trade

off between the two.

Lastly, we can adjust the frequency detuning of the trapping light. More detuned light enlarges the trap, which can improve overlap but at the cost of elevating the potential energy once atoms are transferred to the magnetic trap.

With these in mind, the procedure started by loading the trap for less than one second with as small a cloud as possible to clearly measure the centre of the MOT. We then utilized a long time of flight to catch some atoms in the magnetic trap. This let us determine the centre of the magnetic trap as viewed from the top down. This initial step provided crucial information that guided the subsequent short time of flight adjustments to move the centre of the MOT over to the centre of the magnetic field.

Once some overlap was achieved, we proceeded to sweep all parameters, optimizing for the number of atoms retained in the trap after evaporatively cooling to a fixed temperature. We then did this procedure again for lithium. Lithium takes longer to optimize as the time one has before the atoms fly out of the trappable region is significantly less.

2.4.3 F1 Pumping and Catch

Once we deactivate the MOT lights, we apply light tuned to $F=1$ ground state to pump the atoms from a dark state to the upper stretched state. The timing of this light is crucial. It must be long enough to adequately pump atoms into a trappable state, yet short enough to prevent atoms in free flight from escaping the trap region. If atoms drift too far from the trap's center, they gain significant potential energy and can 'slosh' out of the trap. Thus, the precision in timing and intensity of the light is essential for optimizing both the number and temperature of the trapped atoms.

Lithium atoms are lighter, so they present a bigger challenge. They have a shorter time of flight before leaving the trap or having too much potential energy and 'slosh' out. We typically have on the order of a few milliseconds to pump and catch. Proper synchronization of the shutters and Acousto-Optical Modulator (AO)s is critical. Adjusting this was the main key to transitioning from catching nothing to successful lithium trapping.

Additionally, the task of trapping two different atomic species complicates the timing optimization as each species requires different conditions. Optimal timing becomes a trade-off between the amount of sodium versus the amount of lithium trapped. We use sodium to sympathetically cool the lithium and this process results in significant sodium loss. Allowing for longer pumping times for sodium despite trapping fewer lithium atoms works in our favor because we need more sodium than lithium to effectively cool the atoms.

2.5 Evaporative Cooling

Now that we have atoms magnetically trapped, we can start to evaporatively cool them down. In this section, we will discuss the general theory behind how evaporative cooling works. We'll talk about how to prepare the atoms by transferring them into a single state and cleaning out the remaining atoms. We'll discuss how we tested and optimized this process. Then we'll cover how to measure the bottom of the magnetic trap to set the final [Radio Frequency \(RF\)](#) knife frequency. We'll talk about how to determine that we've reached Bose-Einstein condensation; and finally, what happens when you introduce lithium into the system.

2.5.1 Theory

Evaporative cooling involves two main mechanisms. First, the depth of the magnetic trap can hold atoms up to a specific kinetic energy. Atoms with kinetic energy greater than the threshold escape the trap. The process is very similar to how the hottest molecules in a cup of coffee evaporate. Once the high energy atoms evaporate, the remaining atoms redistribute their kinetic energies through collisions. Thermalization results in a lower overall temperature of the atomic cloud.

The second involves using [RF](#) light to couple trapped and anti-trapped states. Atoms with sufficient kinetic energy penetrate far enough into the magnetic field for the Zeeman shift between these states to bring the transition into resonance with the [RF](#) light. The atoms transition from the trapped state to the anti-trapped state and are ejected from the trap. This effectively removes the high energy atoms. Again, the atoms thermalize, and this leads to a lower temperature of the cloud.

In a model for evaporative cooling [\[9\]](#), we consider a sequence of discrete steps of energy cuts and re-thermalization. Initially, we have a gas of atoms trapped in a harmonic potential, as shown in the magnetic trapping section, which is in thermal equilibrium. The energy distribution of these atoms is

$$f_0(E) = N_0 \frac{E^2}{2(k_B T_0)^3} e^{-E/k_B T_0}, \quad (2.30)$$

where N_0 is the total number of atoms, k_B is the Boltzmann constant, and T_0 is the initial temperature. From this distribution, we can calculate the mean energy per particle using

the equipartition theorem. Since each degree of freedom (3) contributes a $\frac{1}{2}$, when we integrate over the energy distribution we have $3k_{\text{B}}T_0$

To cool the gas, we now apply [RF](#) light to cut the tail end of the distribution which removes atoms with energies exceeding a chosen threshold. This threshold is defined by a truncation parameter η , such that the cut is applied at energy $E = \eta k_{\text{B}}T$. This selective removal of high energy atoms results in a non-thermal distribution. We then allow the gas to re-thermalize through elastic collisions, restoring a thermal distribution at a lower temperature.

Integrating over the truncated energy distribution, we calculate a new number of atoms N_1 and the new total energy E_1 . These can be expressed as

$$N_1 = N_0 \int_0^{\eta k_{\text{B}}T_0} f_0(E) dE, \quad (2.31)$$

$$E_1 = \int_0^{\eta k_{\text{B}}T_0} E f_0(E) dE, \quad (2.32)$$

After numerical integration, the remaining fraction of atoms can be approximated as

$$\frac{N_1}{N_0} \approx 1 - \frac{1}{2}\eta^2 e^{-\eta}, \quad (2.33)$$

And likewise the resulting total energy as

$$\frac{E_1}{E_0} \approx 1 - \frac{1}{6}(\eta^3 + 3\eta^2 + 6\eta)e^{-\eta}, \quad (2.34)$$

The new temperature T_1 after re-thermalization is found using the relation

$$T_1 = \frac{E_1}{3N_1 k_{\text{B}}} \quad (2.35)$$

This gives a new temperature of

$$\frac{T_1}{T_0} = \frac{E_1}{E_0} \frac{N_0}{N_1}. \quad (2.36)$$

For each single cut and re-thermalization cycle, we get a significant reduction in temperature. We can then repeat this process until we reach the desired temperature. In real world experiments the process is continuous, but this model captures the essence of evaporative cooling.

As mentioned in the introduction, we also weaken the trap as we cool the cloud. There is an interplay between reducing the trap depth, the [RF](#) knife, and the total phase-space density we can achieve in the final trap. The exact sequence has been optimized for our setup over many years of experimentation.

Continuing with our model, we can analyze the phase-space density, which is a crucial parameter for achieving quantum degeneracy in a dilute atomic gas. The peak phase-space density in a harmonic trap is

$$\rho = \frac{N}{(k_B T)^3}, \quad (2.37)$$

so the relative change in phase-space density after one evaporation step is

$$\frac{\rho_1}{\rho_0} = \frac{N_1}{N_0} \left(\frac{T_1}{T_0} \right)^{-3}. \quad (2.38)$$

It's convenient to introduce a parameter for evaporation efficiency ξ [\[21\]](#). This parameter characterizes the gain in phase-space density relative to the loss of atoms

$$\xi = \frac{\log(\rho_f/\rho_i)}{\log(N_i/N_f)}, \quad (2.39)$$

where ρ_i and ρ_f are the initial and final phase-space densities, and N_i and N_f are the initial and final atom numbers.

In experiments, the additional losses, like background gas collisions and inelastic two-body collisions, limit efficiency. These losses can be accounted for by adjusting the efficiency parameter. We can get more effective evaporation when the energy threshold for evaporation is continuously adapted to the decreasing temperature, keeping the truncation parameter η constant. This will give us the highest phase-space density.

For us to get a better picture of the dynamics of evaporative cooling, we need to consider the collision rates in the trap. The elastic scattering rate per atom is

$$\gamma_{\text{el}} = \bar{n}\sigma\bar{v}_{\text{rel}}, \quad (2.40)$$

where $\bar{n}$ is the mean number density, σ is the elastic scattering cross-section, and $\bar{v}_{\text{rel}}$ is the mean relative velocity of the atoms [13]. The mean number of elastic collisions per atom in one evaporation step can be approximated by

$$N_{\text{el}} = p_{\text{ev}}N_{\text{ev}} \approx 2 + \frac{4}{\eta}, \quad (2.41)$$

where p_{ev} is the probability of an atom being evaporated and N_{ev} is the average number of elastic collisions an atom experiences before leaving the trap [13]. Efficient evaporative cooling requires a high ratio of elastic to inelastic collisions because every inelastic collision causes atoms to be lost from the trap.

2.5.2 Evaporative Cooling Optimization

In this section, we will briefly discuss how we optimized the evaporative cooling stage in our experiment.

Optimizing the RF Knife

Optimizing the RF knife is a major step to reach BEC. We needed to re-optimize the light sequence due to recent changes in magnetic field strengths. It's important to note that while the decay function for the frequency changes does not require adjustment, the final frequency does. This adjustment is necessary because the final frequency is highly dependent on the bottom of the magnetic trap, which in turn depends on the magnetic field strength.

It's important for us to find the bottom of the magnetic trap. This is done by taking absorption images at set intervals of the RF final frequency. We analyze the data by fitting it to a straight line to predict the frequency at which zero atoms remain, effectively determining the trap's bottom 2.12. This process gives us a frequency region around which we can optimize.

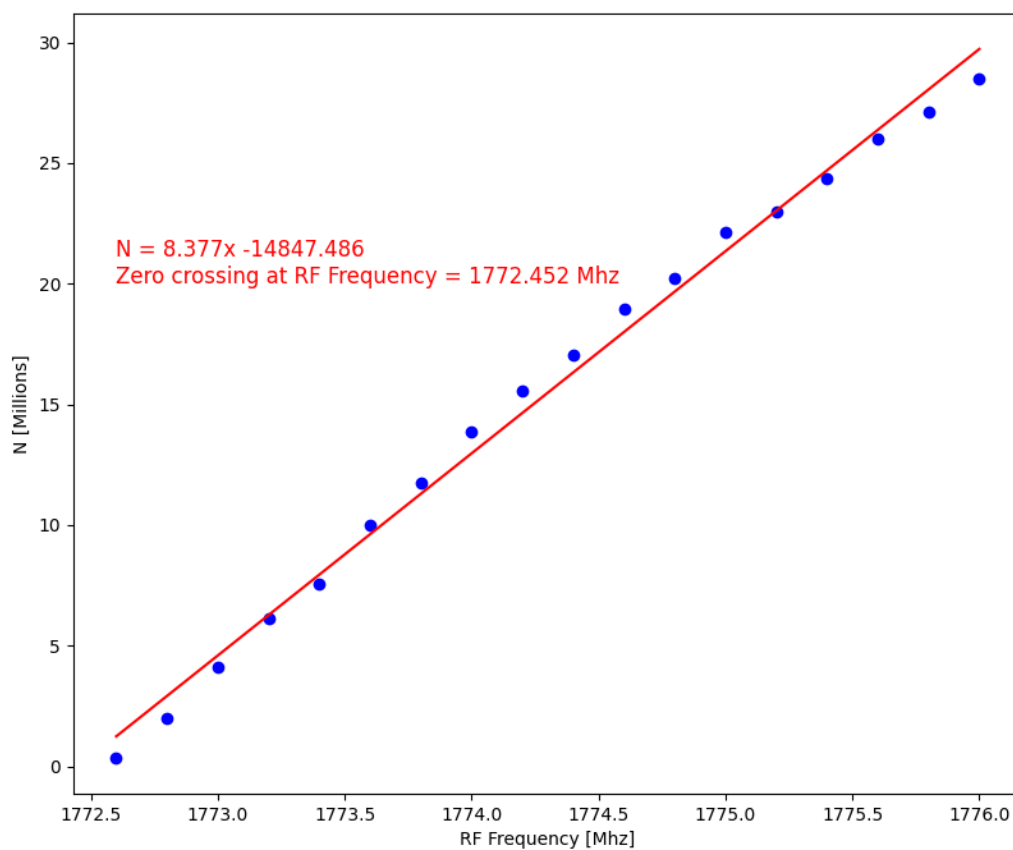


Figure 2.12: Optimization of the RF 'knife' frequency. The RF frequency corresponding to the bottom of the magnetic trap was found to be 1772.4 MHz.

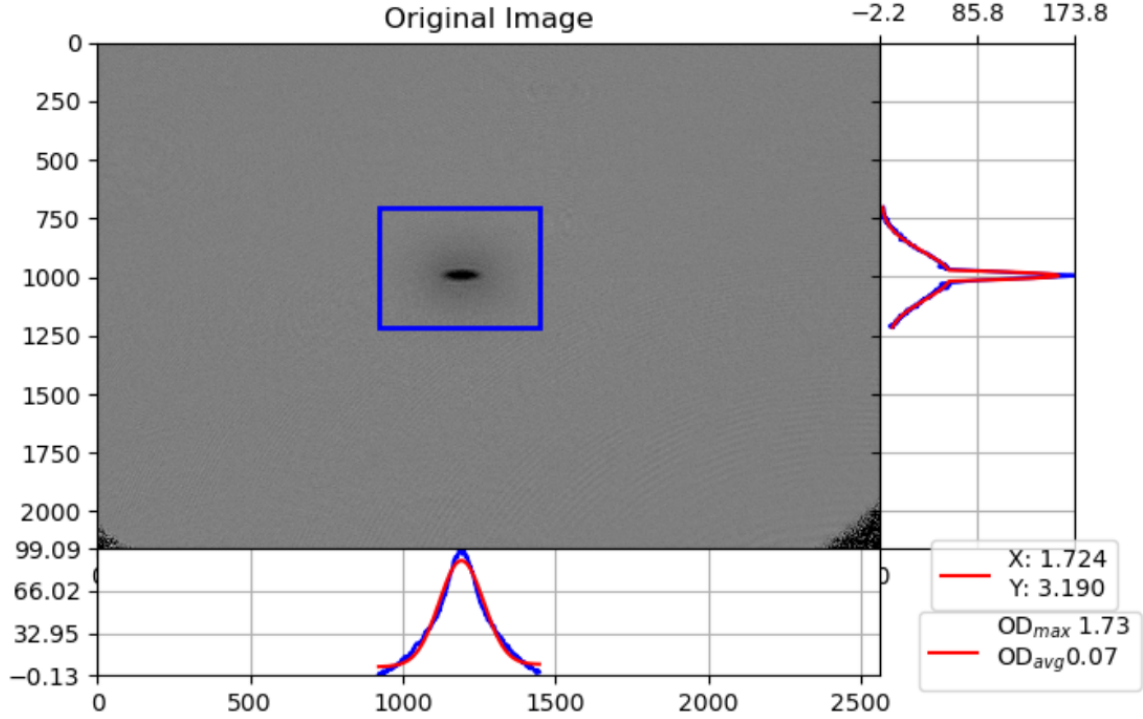


Figure 2.13: In this image, we have both an isotropically expanding thermal cloud and a BEC. The BEC is expanding at a much greater rate along one axis of the trap due to greater initial confinement along that axis than along the others.

Achieving Bose-Einstein Condensation

With the bottom of the trap identified, we proceed to optimize the duration of the evaporative cooling sequence to facilitate Bose-Einstein condensation. As illustrated in figure 2.13, we initially observe condensation by noting the inversion of the widths, unlike a thermal cloud which expands isotropically. We then adjust the timing and final RF knife frequency to maximize the number of atoms in the BEC relative to the thermal cloud.

Sympathetic Cooling Lithium

Although lithium can be evaporatively cooled, it is more technically challenging than sympathetically cooling lithium with sodium directly. Lithium undergoes elastic collisions with sodium. We leverage this property by sympathetically cooling lithium using sodium as a heat sink. The process involves cooling sodium slightly, then allowing time for sodium and lithium to thermalize, and repeating this process to gradually cool the lithium.

Starting with an approximate ratio of 100 Na atoms for every Li atom in the magnetic trap. This ratio works well to get sufficient cooling efficiency and the thermalization dynamics between the two species.

Optimizing the final temperature of the mixture, we initially adjusted the lithium load time until the desired temperature and atom number trade off was reached. Adjustments in the RF final frequency timings are essential once lithium is introduced. We achieved a final ratio of 1 to 1 with about 2 million atoms of each species which prepares the system for Feshbach molecule formation. Achieving quantum degeneracy in the atomic mixture is not necessary for forming NaLi molecules. This simplifies the optimization process as there is headroom for what temperature and final atom count is achieved at this stage.

2.5.3 Conclusion

In this chapter we covered the trapping and cooling of atoms. We discussed how to characterize and create a dual species atomic beam providing the foundation for the experiment. We then covered the theory behind a Zeeman slower, discussing the importance of designing the correct field profile. Next, we covered the physics behind a MOT and a magnetic trap. And importantly, the importance of properly aligning the atoms for transferring between them. And finally, the process of evaporative cooling, where we achieve the ultracold temperatures required to form Feshbach molecules. In the following chapter, we will use our ultracold atoms in the process of making triplet ground-state NaLi molecules.

Chapter 3

Making triplet ground-state NaLi Molecules

This section will cover the formation of [NaLi](#) molecules. We'll begin by discussing the theory of optical dipole traps along with how to align the beam. We will cover how to transfer atoms from a low field seeking to a high field seeking state using a Landau-Zener sweep. Next, we will review Feshbach resonances and how we use this phenomenon to create Feshbach molecules. Lastly, we will discuss the theory behind [STIRAP](#) and the experimental setup that brings our highly excited molecules down to the triplet ground-state.

3.1 Optical Dipole Trap

An [ODT](#) exploits the dipole force generated by the gradient of an intense laser field to confine neutral atoms. This technique works via the interaction between the induced dipole moment of atoms in an electric field and the spatially varying electric field of the laser.

In contrast to magnetic traps, [ODTs](#) offer the advantage of trapping states independent of their magnetic sublevels, opening up the possibility of studying properties of interacting atoms, like Feshbach resonances, that would otherwise be inaccessible. In the following we will discuss the general theory of optical dipole traps, the experimental setup, and two important procedures for successful operation.

3.1.1 Theory

Dipole forces arise due to the interaction between an induced atomic dipole moment and the external electric field. The potential energy V_{dip} of the induced dipole moment d in an oscillating electric field $\mathbf{E}$ is

$$V_{\text{dip}} = - \int_0^E \mathbf{d} \cdot d\mathbf{E} = -\frac{1}{2}\alpha E^2, \quad (3.1)$$

where α is the atomic polarizability and $\mathbf{d} = \alpha\mathbf{E}$ is the dipole. For our purposes, we want to write the expression in terms of intensity so we have

$$V_{\text{dip}} = -\frac{1}{2\epsilon_0 c} \text{Re}[\alpha(\omega)] I(\mathbf{r}), \quad (3.2)$$

where η_0 is the vacuum impedance. Putting this into a form for the polarizability, expressing the slowly varying electric field as $I = |E_0|^2 / 2\eta_0$, and defining the detuning as $\Delta = \omega - \omega_0$, we can write the intensity dependent dipole potential as follows

$$V_{\text{dip}} = \frac{e^2}{2m\epsilon_0 c} \frac{(\omega_0 + \omega) \Delta}{[(\omega_0 + \omega) \Delta]^2 + \Gamma^2 \omega^2} I(\mathbf{r}). \quad (3.3)$$

The sign of V_{dip} is determined by the detuning. Because the corresponding dipole force is gradient-dependent, $\mathbf{F}_{\text{dip}} = -\nabla V_{\text{dip}}$, we get attraction or repulsion from regions of high intensity depending on the sign of Δ .

Using standard techniques [47], we can find the polarizability of a two-level atom to be

$$V_{\text{dip}} = - \sum_j \frac{\hbar \omega_{j0} \Gamma_j^2}{4} \frac{\omega_{j0}^2 - \omega^2}{(\omega_{j0}^2 - \omega^2)^2 + \Gamma_j^2 \omega^2} \frac{I(\mathbf{r})}{I_{\text{sat},j}}, \quad (3.4)$$

where I_{sat} is the saturation intensity for the specific transition level we are addressing. We can treat the alkali atom as a two level system because the D_2 line dominates the polarizability. At large detuning where the laser frequency ω is far away from the resonance ω_0 such that $|\Delta| \gg \Gamma$, we can simplify to

$$V_{\text{dip}} = \frac{\hbar \Gamma^2}{8\Delta} \frac{I(\mathbf{r})}{I_{\text{sat}}}. \quad (3.5)$$

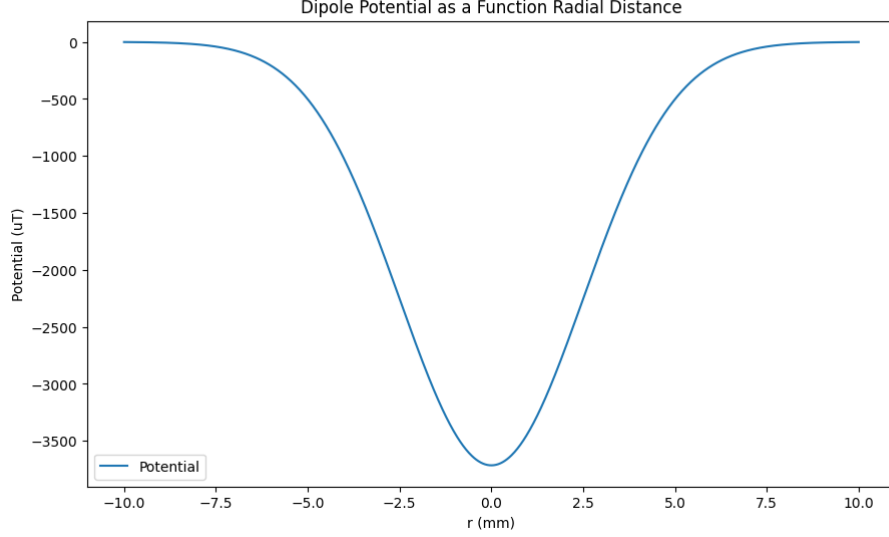


Figure 3.1: Dipole potential as a function of radial distance. Using a Gaussian beam profile, we have an initial trap depth of $3500 \mu K$.

Using our setup as an example, we can now calculate the potential curve at the initial stage of transferring from the magnetic trap to the [ODT](#), where we are using a 1550nm (red detuned) laser with an intensity of 25Wcm^{-2} , we get a trap depth of $3500\mu K$ shown in [Fig. 3.1](#).

From this potential we can approximate the density distribution of our trapped atoms [\[12\]](#) as follows

$$n(r, T) = n_0 \exp\left(-\frac{V_{\text{dip}}(r)}{k_b T}\right), \quad (3.6)$$

where n_0 is the density at the centre of the trap as in [Fig. 3.2](#).

On a final note, we use a retro-reflected beam for the [ODT](#), outside of creating a pancake like structure of atomic clouds, we get a factor of four increase in peak intensity because for a single traveling wave propagating along the z -axis, the intensity is

$$I \propto |E_0|^2, \quad (3.7)$$

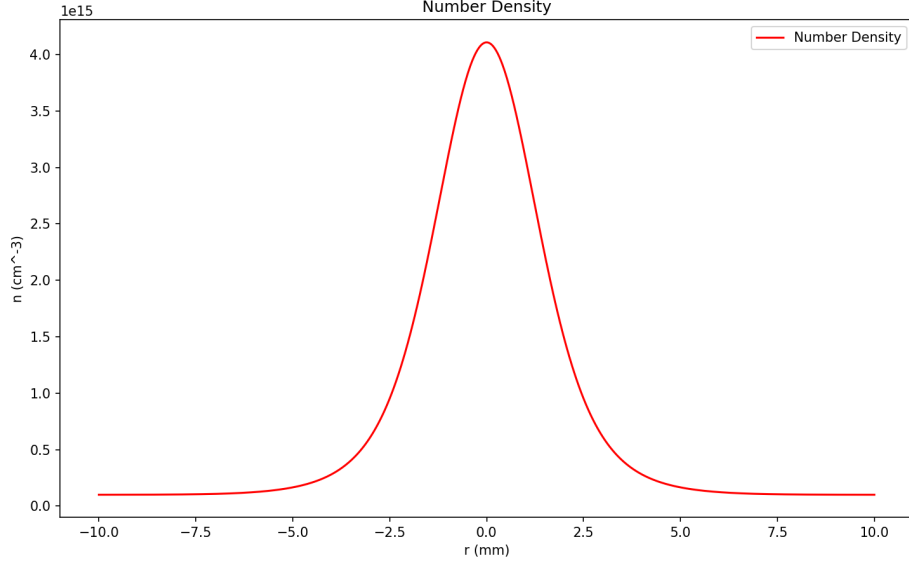


Figure 3.2: Using an arbitrary centre density value, the plot shows that the spatial density profile is determined by the spatial intensity profile of the [ODT](#) beam.

and for a standing wave the intensity is

$$I \propto |2E_0|^2 \cos^2(kz), \quad (3.8)$$

This is because the standing wave is the superposition of two counter-propagating waves [\[10\]](#). Comparing, we see that the maximum intensity of a standing wave is four times that of a traveling wave $4|E_0|^2$ vs. $|E_0|^2$.

3.1.2 Experimental Setup

The experimental setup for an [ODT](#) is fairly straightforward. As shown in the following figure [3.3](#), there is a high power (25W) 1550nm laser that is aligned to pass through the cloud of atoms and then retro-reflected via a “cats eye” configuration to create a standing wave and increase the trapping potential. There is a locking servo setup that monitors the laser power and adjusts an [AO](#)’s input power to change the fraction of optical power pushed into the first order.

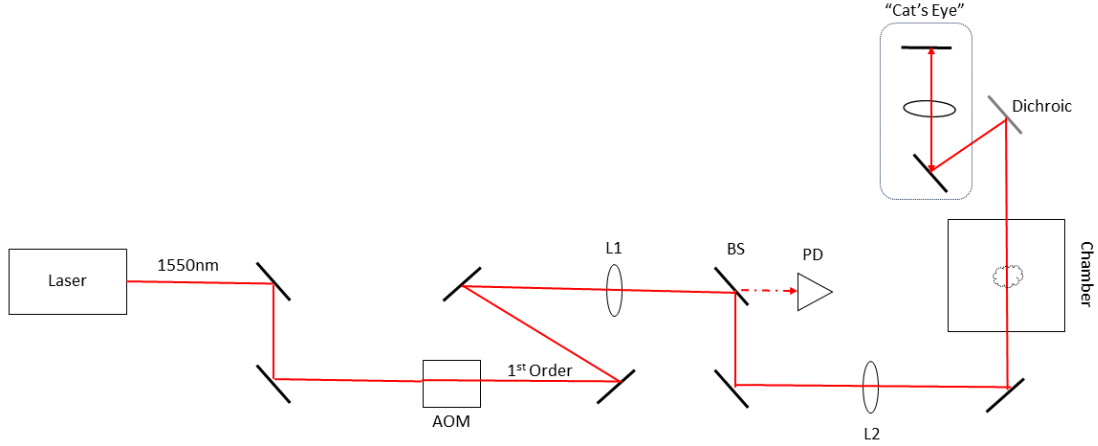


Figure 3.3: Schematic Layout of the ODT Setup in BEC3.

Optical Layout

At the experimental table, the ODT begins with the laser which is in the infrared spectrum and a typical choice for trapping because of its deep potential wells and the ability to trap a variety of atoms without causing significant spontaneous emission. From there the light passes through an AO. It is driven at 80 MHz, which is an insignificant frequency shift, this allows for the servo to control the optical power. From here the light enters the first of a two lens beam expansion setup, where it encounters a beam sampler. The light that transmits though goes to a PD which is used as the feedback signal for the Proportional-Integral-Derivative Controller (PID). The light is then collimated at the second lens and sent through the main chamber. The beam then reflects off a dichroic lens (allowing the MOT and imaging light to pass unobstructed) and is then retro-reflected in a ‘cat’s eye’ configuration.

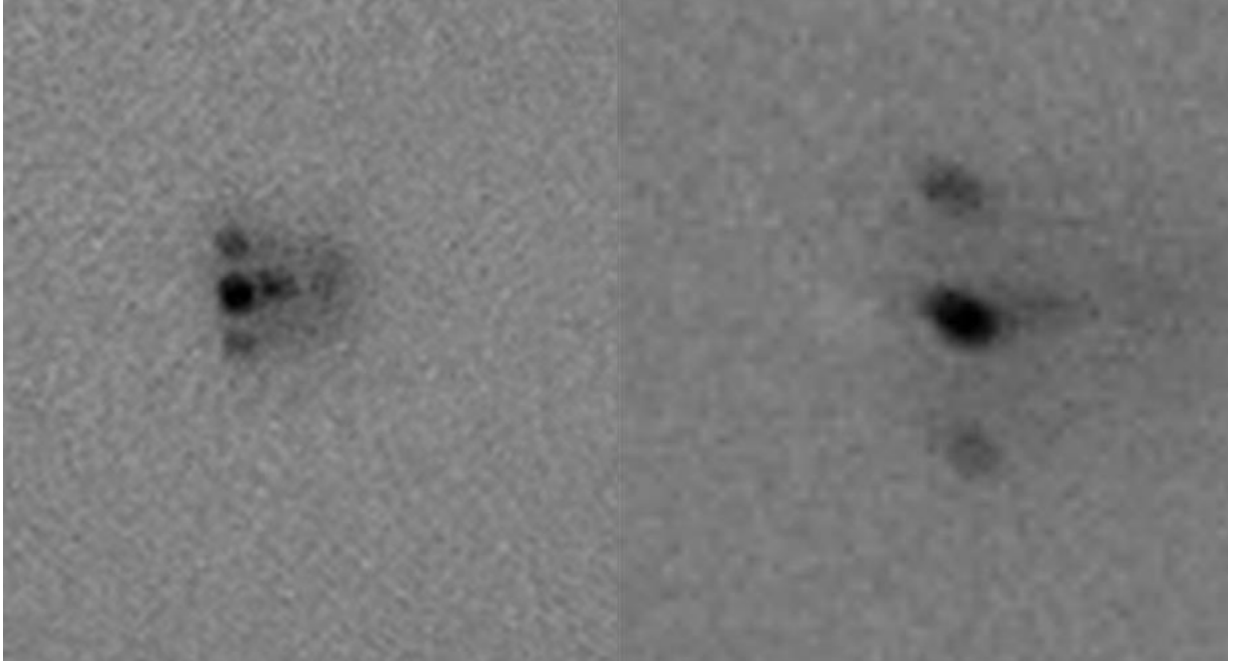


Figure 3.4: Using the Kapitza-Dirac effect to align the ODT. On the left, we see that the cloud is asymmetrically divided by the standing wave. On the right, the ODT is well aligned as indicated by the single central feature along with the two lobes traveling in opposite directions.

3.1.3 ODT Alignment

Here we discuss using the Kapitza-Dirac effect to align the ODT. A standing wave of light diffracts the cloud of cold atoms since the light's periodic potential acts as a grating, scattering the atoms into discrete momentum states. The duration of the light is tuned to be brief enough to cause only minimal photon scattering, which allows for a clear resolution of these states. After the light, the trap is turned off and the atoms freely expand where their different momentum states cause them to spatially separate. Observing the Time of Flight (ToF) expansion reveals the diffraction pattern, which is contingent on the alignment of the atomic cloud with the standing wave. A misaligned trap will produce an asymmetric pattern in the images 3.4. A well-aligned trap will produce a symmetric diffraction pattern, indicating that the trap is centered with the atomic cloud. We can use this for precise adjustments to the trap's alignment.

3.2 Transferring to the High Field

In the following section, we'll discuss how we go from the [ODT](#) with no magnetic field, to the high magnetic field regime. The high field is where we can use a Feshbach resonance to create a highly excited bound molecular state. We'll discuss Landau-Zener Theory for a two level system to better understand the Landau-Zener sweeping technique which coherently transfers population from one sublevel to another. Doing this sweep will allow us to go to the high field while maintaining a large population of atoms in the trap. Then we'll briefly cover the RF transfer frequency program and how it calculates the correct centre frequency required. And we'll finish by covering how we optimized the sweep to get over 90% transfer efficiency.

3.2.1 Theory

Landau-Zener theory gives a way for us to calculate the probabilities of non-adiabatic transitions between quantum states when an external control parameter, such as a magnetic field, is varied linearly across an avoided crossing.

We begin our discussion by considering a two-level system described by the Hamiltonian

$$H(t) = \frac{1}{2} \begin{pmatrix} \epsilon & \Delta \\ \Delta & -\epsilon \end{pmatrix} \quad (3.9)$$

where Δ is the minimal energy spacing and ϵ is the energy bias. For our two-level system, we will assume a linear bias where $\epsilon = \nu t$, where ν is the controllable parameter (in our case the B field strength), giving us

$$H(t) = \frac{1}{2} \begin{pmatrix} \nu t & \Delta \\ \Delta & -\nu t \end{pmatrix}. \quad (3.10)$$

The diabatic states are when $\Delta = 0$ and the energy levels are simply $\epsilon/2$. For the other case, we use the Schrodinger equation and obtain the adiabatic energy levels

$$E = \frac{1}{2} \sqrt{\Delta^2 + \epsilon(t)^2}. \quad (3.11)$$

The adiabatic energy levels approach the diabatic as ϵ gets much larger than Δ . The energy spacing at the avoided crossing grows and the coupling between the two states approaches zero.

Landau and Zener developed a time-dependent Schrodinger equation solution taking the two asymptotic states as boundary conditions in solving for the probability of transferring between states [18]. This results in the well known formula

$$P = \exp\left(-\frac{2\pi\Delta^2}{\hbar|\nu|}\right). \quad (3.12)$$

If we sweep quickly through the avoided crossing, i.e change ν quickly, we follow the non-adiabatic transitions and the system will switch states with high probability. Otherwise, the system will stay in the same state and follow the avoided crossing.

RF Transfer Frequency

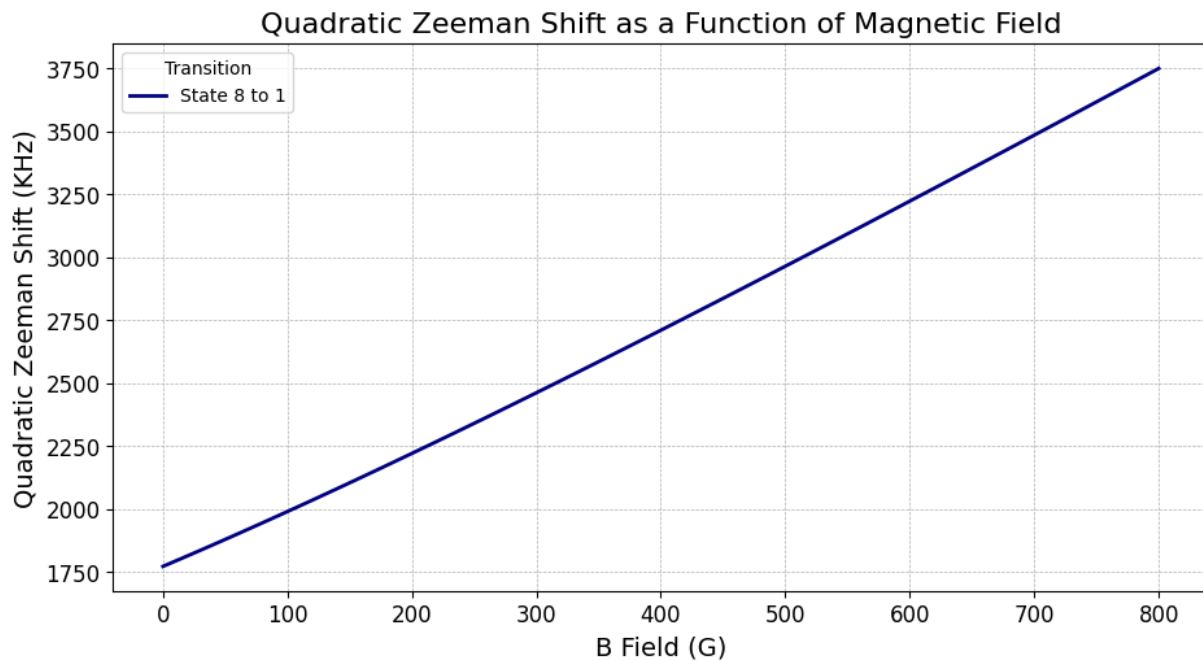
It's important to calculate the RF transfer centre frequency required for the sweep. In appendix A, there is a python program that calculates the transition frequency between two states by using the quantum numbers associated with the atoms nuclear (I) and electron (J) spins and calculating the hyper-fine Hamiltonian which combines the scalar product of these spins and accounting for magnetic dipole and electric quadrupole interactions. The total magnetic moment of the atom in an external magnetic field is determined by the weighted contributions of the electron and nuclear magnetic moments using respective g-factors. The eigenvalues and eigenvectors of the total Hamiltonian are computed to find the energy levels and hence the transition frequencies between these levels for a given B field 3.5. For example, for Na state 8 to 1 (where we label the m_F levels of the ground and excited states 1 through 8), the centre frequency is 1777.66 MHz at 2.87 Gauss B field¹.

3.2.2 Landau Zener Sweep Optimization

In this section, we will quickly discuss how we optimized the Landau-Zener sweep sequence and achieved 95% population transfer.

We begin by setting the start and finish frequencies, approximately 5 MHz wide centered on the calculated RF transfer centre frequency. The sequence progresses to the optical

¹We always keep a magnetic field present to define the quantization axis. In our experiment, this is approximately 2.87 Gauss.



For B-Field: 2.87 G
RF Transfer Frequency: 1777.66 Hz

Figure 3.5: Plot of the RF transfer frequency between state 8 and state 1 as a function of B field. For our setup, with a bias field of approximately 3G, the transfer frequency is 1777.66 MHz.

dipole trap where we perform the Landau-Zener sweep. Then we increase the magnetic field to a consistent value (in our case 10 Gauss) and then absorption image the atoms.

Some careful consideration needs to be made with the imaging frequency given that we are imaging at two different field strengths. So we first optimized the imaging frequency at the new field before we moved on to optimizing the sweep [3.6](#). By comparing the atom count in the [ODT](#) before the Landau-Zener sweep to the count after the magnetic field increase, we can determine the efficiency of the sweep. Next, we narrow the width of the sweep (effectively decreasing the speed) and then look to maximize the atom count at the higher magnetic field.

As we narrow in on the optimal conditions for maximum population transfer, we enhance the precision of our measurements by averaging the results of three shots. This increases our sensitivity, although practical considerations limit further optimization past 95% transfer efficiency because of system fluctuations and time constraints.

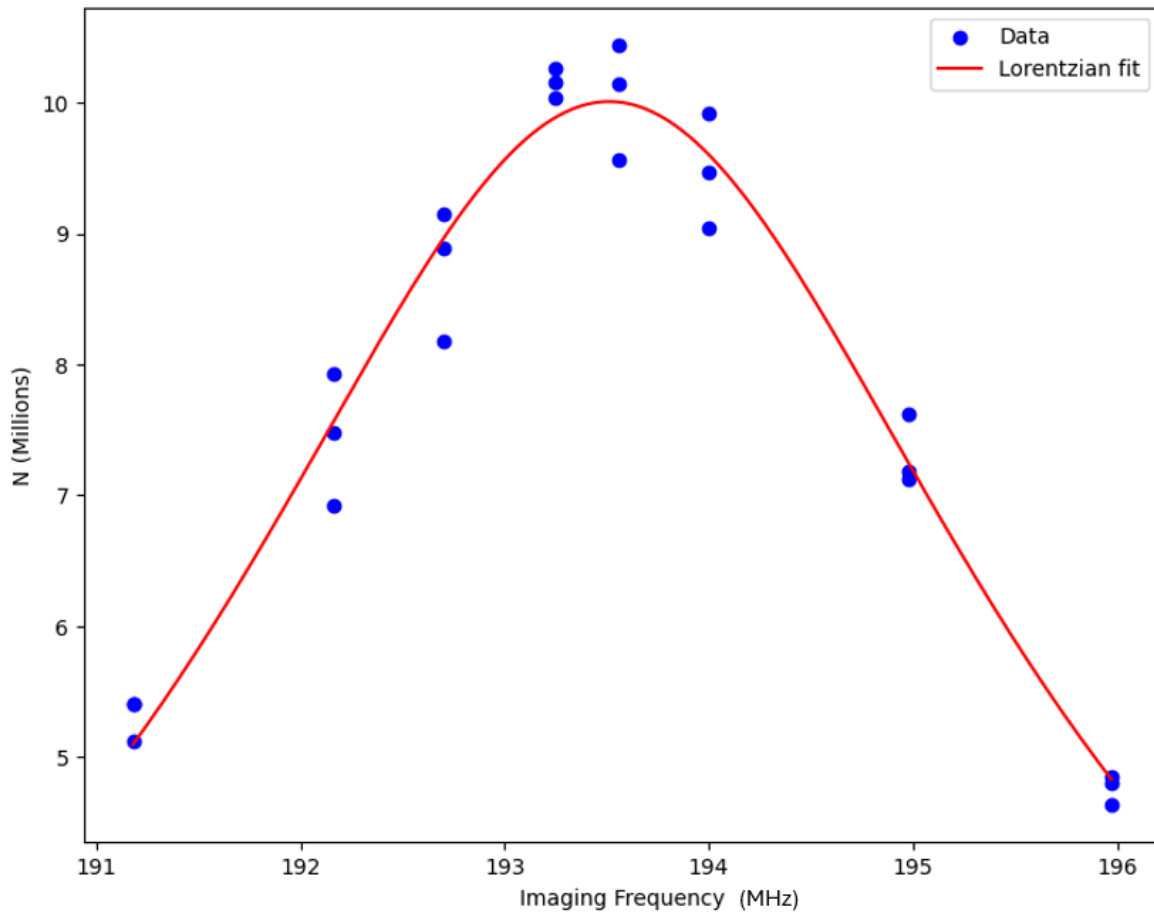


Figure 3.6: Imaging frequency optimization. We increased the number of shots taken near the maximum value to improve sensitivity. Here the centre of the Lorentzian is at 193.5 MHz.

3.3 Feshbach Molecule Formation

In the following section, we will discuss the formation of Feshbach molecules. First, we will discuss what a Feshbach resonance is in the context of a magnetically tunable interaction. We will also present a model for predicting the variation of the scattering length as a function of magnetic field. With this background, we will review the formation of Feshbach molecules. Next we'll cover the experimental procedure which implements these ideas. And finally, we'll talk about how we measured and optimized the total number of Feshbach molecules we could produce.

3.3.1 Theory

In ultracold collisions, a Feshbach resonance occurs when the bound state energy of a closed channel overlaps with the energy of an open channel [4]. Through the Zeeman effect, we can tune a closed channel energy level towards an open channel, then even a weak coupling between the two states leads to strong mixing. Because in this situation the scattering cross-section, which is a measure of the probability of colliding, for the two atoms is modified and is a function of the tunable parameter.

For a magnetically induced resonance [27], the scattering length varies according to

$$a(B) = a_{\text{bg}} \left(1 - \frac{\Delta}{B - B_0} \right), \quad (3.13)$$

where a_{bg} is the background scattering length (the scattering you measure with no magnetic field), B_0 is the resonance position (where the scattering length diverges), and Δ is the resonance width.

These Feshbach resonances can be broad and strong or narrow and weak. The width can be modeled as

$$\Delta = \frac{m(2\pi\hbar)^3}{4\pi\hbar^2 a_{\text{bg}} \Delta\mu} |M|^2, \quad (3.14)$$

where $\Delta\mu$ is the difference in magnetic moments, and M is the coupling strength. While $\Delta\mu$ is generally on the order of $2\mu_{\text{B}}$ for alkali atoms, the coupling strength $|M|^2$ can vary over a wide range of values. The widths often range from μG , as is the case for the Feshbach resonance we use for [NaLi](#), up to hundreds of Gauss.

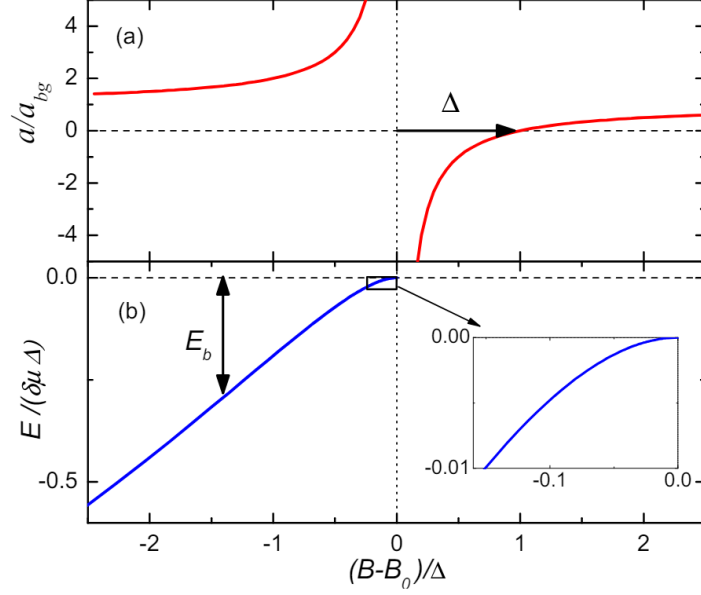


Figure 3.7: a) Scattering length and b) molecular state energy near a magnetically tuned Feshbach resonance. The inset shows the ‘universal regime’ near the point of resonance where a is very large and positive [4].

The coupling of the two channels via a Feshbach resonance also modifies the energy of the bound molecular state. This means that the dressed molecular state has an energy

$$E_b = -\frac{\hbar^2}{2ma^2}, \quad (3.15)$$

which shows that the binding energy around the pole of the resonance depends quadratically on the magnetic field

$$E_b \propto (B - B_0)^2, \quad (3.16)$$

as can be seen by substituting in how the scattering length depends on the B field. So the further away you get from the resonance, the more bound the molecular state.

An important detail is that if the bound state of a closed channel approaches the resonance from below, the scattering length increases, while it decreases for bound states located above the resonance energy [4]. We take advantage of this in our setup by ramping

the magnetic field up to a higher level initially and then sweeping the field strength down through the resonance.

Another important consideration is that a Feshbach resonance also increases inelastic losses. This leads to an experimental difficulty where you have to find a compromise between how long you spend in the resonance region such that you get Feshbach molecules, but not so long as to also lose molecules through enhanced inelastic collisions.

In general, these treatments simplify using s -wave collisional channels only. This assumption can be justifiable because in ultracold atomic gases, energies are so low that most of the time they cannot overcome the centrifugal barrier which suppresses p -wave and above collisions.

For Feshbach molecule formation, we use the magnetically tunable Feshbach resonance to resonantly enhance atom to molecule coupling. By sweeping the magnetic field across the narrow width resonance, we can transfer atoms into a bound Feshbach molecule state.

If we assume a two-level system, we can use the Landau-Zener theory, described above, to model the probability of transitioning into a bound state. We find the probability using

$$P = 1 - e^{\epsilon}, \quad (3.17)$$

and for our two level system model we have that

$$\epsilon = \frac{\alpha n \hbar}{m} \frac{\Delta a_{\text{bg}}}{\dot{B}}, \quad (3.18)$$

where $\dot{B}$ is the rate of change of the magnetic field [23]. This shows that the maximum conversion efficiency is dependent on phase-space density of the atomic cloud at ultracold temperatures, which in turn relies on the resonance parameters.

Unlike the Landau-Zener sweep between two states, where you can get near 100% efficiency, the typical efficiency for molecule formation we can achieve is a mere 5%. This is largely due to inelastic atom-molecule collisions. As mentioned earlier, the Feshbach resonance also enhances these collisions which limits us. Also, another source of loss is molecule-molecule collisions. As we form molecules, they begin to collide with each other which releases energy and hence they're lost from the trap.

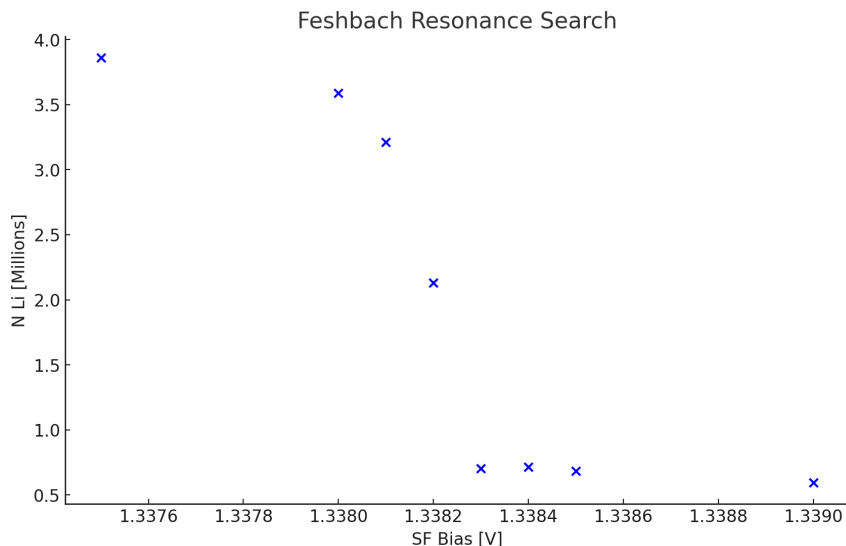


Figure 3.8: Feshbach resonance molecule formation search. The number of [Li](#) atoms remaining after blasting and dissociation for a given SF Bias analog voltage control setting.

3.3.2 Measurement and Optimization

With the atoms now transferred to the high field seeking state, we can begin the process of optimizing Feshbach molecule formation.

Because we have changed the magnetic fields of the experimental setup, the first thing we needed to do was find the power supply current settings to give us the correct magnetic field for the Feshbach resonance. [BEC3](#) has three power supplies, a large 500A Lambda, a medium power supply (named SF Bias), and a small but quick power supply (named MOL). This gives us the flexibility to ramp up to a large field, then trim the field to the correct field strength, and finally use the small power supply to sweep the Feshbach resonance.

Finding the right values is straightforward. We ramped up to a fixed value with the large power supply and then use the medium power supply to ramp slowly up to ever increasing values until we see the signature of a Feshbach resonance. Using the medium power supply is not ideal for molecule formation, however, as shown in [3.8](#), you can see a point at which the atom number drops significantly.

We set the medium power supply to just above this value, and as mentioned earlier, we use the small but quick power supply to sweep the field down through the Feshbach

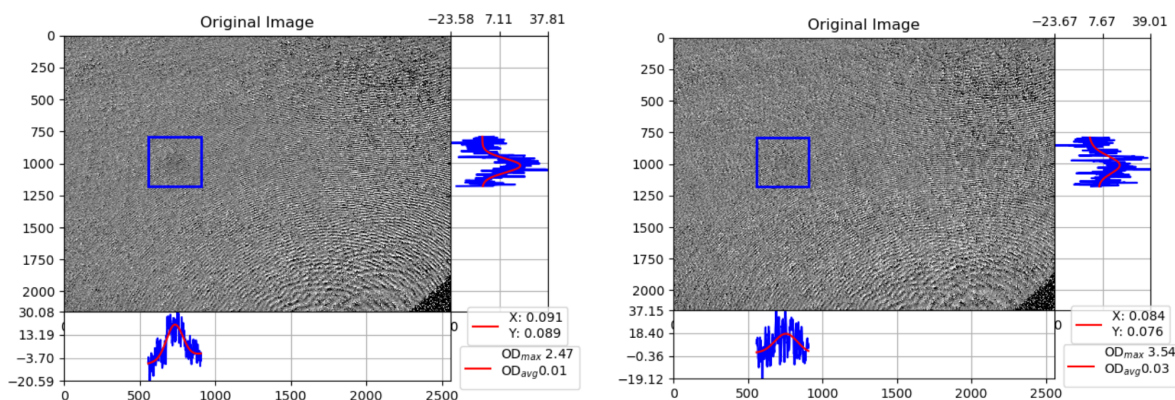


Figure 3.9: On the left, we have an absorption image of the remaining atoms after dissociation with the sweep, and on the right without the sweep. It can be seen that the blast step was not fully optimized since there are remaining atoms with no sweep.

resonance. To optimize this final step, we start with a wide sweep and narrow it until we find the maximum.

A quick note on the imaging process. Since we cannot image the molecules directly, we have to follow a three-step procedure. We sweep to form the molecules, we use high field resonant light to blast away any remaining atoms, then we dissociate the molecules and take an absorption image of them. In this way, we can measure how many atoms must have been in bound in a molecule. To optimize the imaging process, we avoided forming the molecules and adjusted the blast time and strength until no atoms were left. This ensures that there will be no atoms surviving unless bound in a molecule.

3.4 STIRAP

In this section, we will first discuss the basic theory behind the [STIRAP](#) process. We will cover a model for the dynamics. After which, we'll discuss the five stages to gain a better understanding of what's happening. Then we'll give a brief description of the laser setup. And finally, we'll go over how we found the correct upleg frequency.

3.4.1 Theory

The [STIRAP](#) process uses two coherent lasers to achieve a population transfer between molecular states. It involves a three-level quantum system that consists of an initial state $|i\rangle$, an intermediate/excited state $|e\rangle$, and a target/ground state $|g\rangle$.

The direct population transfer from $|i\rangle$ to $|g\rangle$ is forbidden due to selection rules; however, we can use the third state to modify and couple the states together. The intermediate state $|e\rangle$ is typically a radiatively decaying state, but because the state is kept dark, we generally don't get much loss.

The dynamics of the [STIRAP](#) process are governed by the time-dependent Schrodinger equation $i\hbar \frac{d}{dt}\Psi(t) = H(t)\Psi(t)$. By using the rotating-wave approximation [\[43\]](#) we have the Hamiltonian

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega_1(t) & 0 \\ \Omega_1(t) & 2\Delta & \Omega_2(t) \\ 0 & \Omega_2(t) & 2\delta \end{pmatrix}, \quad (3.19)$$

where the Rabi frequencies $\Omega_1(t)$ and $\Omega_2(t)$ are the interaction strengths of the laser fields with the respective quantum states. We have that

$$\Omega_n = \frac{-d_{i,j}\epsilon(t)}{\hbar} \quad (3.20)$$

where $d_{i,j}$ is the dipole transition moment between the initial and excited state, and $\epsilon(t)$ is the slowly varying amplitude.

We can break down the process into five stages and describe what is happening [\[48\]](#). The initial phase involves turning on the Stokes laser adiabatically. This causes an Autler-Townes splitting of the intermediate state but the initial state $|i\rangle$ remains unaffected. The dark state, which is derived from the Hamiltonian's eigenstates is

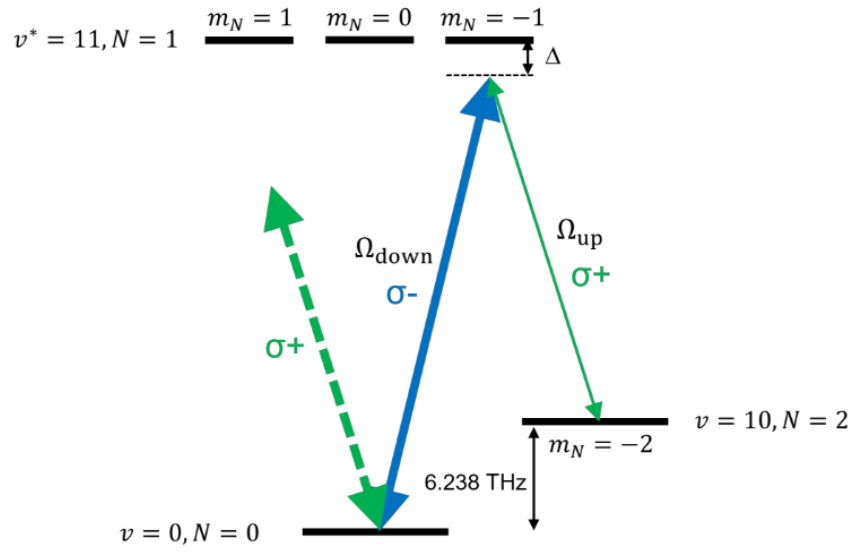


Figure 3.10: A schematic representation of the chosen three-level system for forming NaLi triplet ground-state molecules [33].

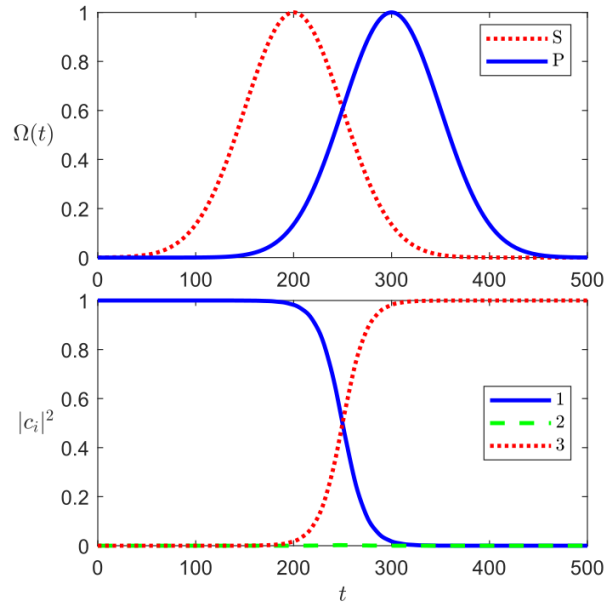


Figure 3.11: Top - The Rabi frequency for the Stokes and probe beams over time during the sequence. Bottom - The probability of being in the initial or ground state as a function of time [48]

$$|a_0\rangle = \cos \Theta(t)|i\rangle - \sin \Theta(t)|g\rangle, \quad (3.21)$$

where $\Theta(t)$ is the mixing angle defined as

$$\tan \Theta(t) = \frac{\Omega_1(t)}{\Omega_2(t)} = \frac{\sin \Theta(t)}{\cos \Theta(t)}. \quad (3.22)$$

The mixing angle is a time-dependent parameter that represents the ratio of the Rabi frequencies of the pump and Stokes fields. It describes the evolution of the system during the population transfer. Also, the Rabi frequencies must be significantly greater than the rate of change of the mixing angle. This eigenstate completely avoids the intermediate state. Even though the Stokes field splits the energy levels of $|e\rangle$, the population is still in $|i\rangle$ at this point.

In the next phase, the Stokes field reaches its peak intensity. The pump field is then turned on and this leads to coherent population trapping, where the atoms are ‘trapped’ in a superposition of states. The system remains in the dark state avoiding radiative decay as mentioned above. For this to happen, the local adiabatic condition must be met where

$$\Omega_{\text{eff}}(t) \gg |\dot{\Theta}(t)|, \quad (3.23)$$

where the effective Rabi frequency is simply the quadrature sum of the two Rabi frequencies. This ensures adiabatic evolution of the system. The strong Stokes field also prevents population transfer to $|e\rangle$, so most of the molecules remain in $|i\rangle$.

In the following phase, we get adiabatic passage between states. Both fields are now strong and the population of molecules transition from $|i\rangle$ to $|g\rangle$ through the dark state. Here the mixing angle $\Theta(t)$ evolves from 0 to $\pi/2$ during this stage.

In the fourth phase, the pump field becomes stronger than the Stokes field. The strong pump field couples $|i\rangle$ to $|e\rangle$ and also splits $|e\rangle$. The transition from $|g\rangle$ to $|e\rangle$ is suppressed and the population remains in $|g\rangle$.

The final phase involves turning off the Stokes laser. The pump laser must remain at maximum intensity because this will isolate the population in the ground state $|g\rangle$.

3.4.2 Experimental Setup

The formation of [NaLi](#) molecules in the triplet ro-vibrational ground state $v = 0, N = 0$ from the Feshbach state $v = 10, N = 2, m_N = -2$ is achieved using 833nm upleg and 819nm downleg laser frequencies to couple the Feshbach and ground states via the $v^* = 11$ excited vibrational state in the $c^3\Sigma^+$ potential.

The strong downleg transition requires only a few 100 μ W power. [BEC3](#) uses a custom built 818nm [External Cavity Diode Laser \(ECDL\)](#) for this purpose. The weak upleg transition requires higher power, so we use an injection lock setup with a high power laser diode seeded by another custom 833nm [ECDL](#).

Both [STIRAP](#) lasers are locked to a commercial optical cavity made of ultra low expansion glass with a linewidth of 15 kHz. Using this cavity, we can lock lasers between 810-880nm to relatively narrow linewidths of $\leq 1kHz$. [\[33\]](#).

The scanning of the frequencies is achieved via a fiber [Electro-Optical Modulator \(EOM\)](#), which generates a secondary feature on the light (in a [Pound-Drever-Hall \(PDH\)](#) locking scheme) using a [Voltage Controlled Oscillator \(VCO\)](#). This signal is then amplified to 15 dBm for the upleg and 17 dBm for the downleg. The use of an analog [VCO](#), rather than a digital signal generator, is essential because it maintains the [PDH](#) lock during small and continuous frequency detunings, whereas a digital signal generator would cause the lasers to unlock because of a short disconnection when changing frequencies.

3.4.3 Finding The Pump Frequency

The [STIRAP](#) process requires a narrow linewidth frequency light. Although in theory this is a simple matter of selecting the correct frequency, in practice there are many practical factors that influence the exact configuration required.

We measured the cavity offset and compared this to previous measurements and estimated that the cavity drifts by 2 MHz per year. From this we could estimate that the correct frequency for the downleg would be 525 MHz. This gives us the starting point for the following optimization process. While the downleg is active at a given frequency, we scan the upleg (probe) during the photoassociation process. During this sweep, we observe a pronounced gain feature. This typically appears near the centre of the initial loss feature associated with the photoassociation.

Once we have this gain feature, if it is not in the centre of the loss curve, then we know that the downleg frequency is offset. We then update the frequency and repeat the process

until we have the gain peak in the centre of the loss feature. The gain feature needs to be centered in the loss feature to ensure that we have resonant coupling between the laser frequencies and the energy levels involved. Resonance is crucial for efficient population transfer and centering reduces off resonant transitions which can lower the efficiency of the process.

3.4.4 Conclusion

In this chapter, we covered how we make triplet ground-state sodium-lithium molecules. We discussed the theory of optical dipole traps, which allows us to trap and cool atoms efficiently. We talked about how we transfer to the high field state by performing a Landau-Zener sweep, which is a critical step to prepare the atoms for Feshbach molecule formation. We covered the theory and experimental setup used to generate Feshbach molecules. Finally, we reviewed the technique for transferring to the triplet ground-state. In the next chapter, we'll discuss four possible experiments we can do with our triplet ground-state molecules.

Chapter 4

The Future of Ultracold Chemistry in BEC3

In this chapter we will explore four experimental goals for our setup in [BEC3](#). We will discuss the theoretical foundations that underpin these experiments and we'll cover why they are interesting areas of research.

4.1 Introduction

First, we will talk about searching for Feshbach resonances in the s -wave channel. Feshbach resonances are important for controlling interactions between atoms in ultracold gas mixtures. In the most recent work done in [BEC3](#), Julie Park observed a universal loss rate below the expected threshold. This suggests that a s -wave resonance might be resolvable. Investigating this could provide insights into atomic interactions. It may also improve our ability to control quantum systems.

We'll also discuss measuring the collision loss rate in $N = 1$ rotational states. While the $N = 0$ stretched hyperfine states are known to be collisionally stable, it remains an open question whether rotational stretching preserves this stability for $N > 0$. By examining Feshbach resonances in these states, we may gain a better understanding of collision dynamics and stability.

Next, we will test the coherence of $N = 1$ and $N = 0$ states in magnetic trap. These states possess coupled electric dipole moments. This interaction could lead to long coherence times. By choosing states with similar magnetic moments, we are trying to optimize

coherence. If we achieve long coherence times, we open up the possibility of using ultracold molecules for quantum computing.

Finally, we are going to attempt photoassociation of molecules in the magnetic trap. Instead of forming molecular states directly in the magnetic trap, we aim to induce photoassociative loss on collision complexes. This experiment will help us measure the dynamics during molecule/molecule collisions, hopefully giving us new insights into these interactions.

4.2 Searching for s -wave Feshbach Resonances

The last experiment in BEC3 was done by Julie Park [46]. She studied the collisional properties of fermionic ultracold sodium lithium molecules in the same hyperfine, triplet ground-state. s -wave collisions are forbidden since they're in the same state. It was an open question whether Feshbach resonances in p -wave collisions could be experimentally measured due to the high density of states. Julie's work showed that you can indeed observe Feshbach resonances in p -wave collisions 4.1.

The team also studied the collision properties of NaLi in different hyperfine states. They showed that the s -wave collisional loss rate was significantly below the universal loss rate. According to the theory for molecule/molecule s -wave collisions, we expect many overlapping Feshbach resonances 4.2. This overlap usually results in the universal loss rate. However, the observed loss rate was below the universal rate. This suggests that the resonances might be spaced out enough to observe them.

An important idea for this experiment is the Universal Loss Rate. The full calculation for the universal loss rate is complex. For molecules, it is generally analytically intractable.

The group presented a simplifying model [46]. They showed that you can model the collision between two atoms like a Fabry-Perot cavity. Atoms in our trap come close to each other and first reflect off the long-range potential. Those atoms don't collide and stay in the trap. Atoms that make it into the short range have a chance of colliding, releasing energy, and being lost from the trap. The model shows that in the short range there is a build up of probability over time just like a resonant cavity building up amplitude.

The "Universal Rate" assumes that every atom that makes it to the short range is lost. For our work, it is important to know that with Feshbach resonances, you can control the reflection/transmission off the first "mirror." This allows us to manipulate the loss rate in the cavity.

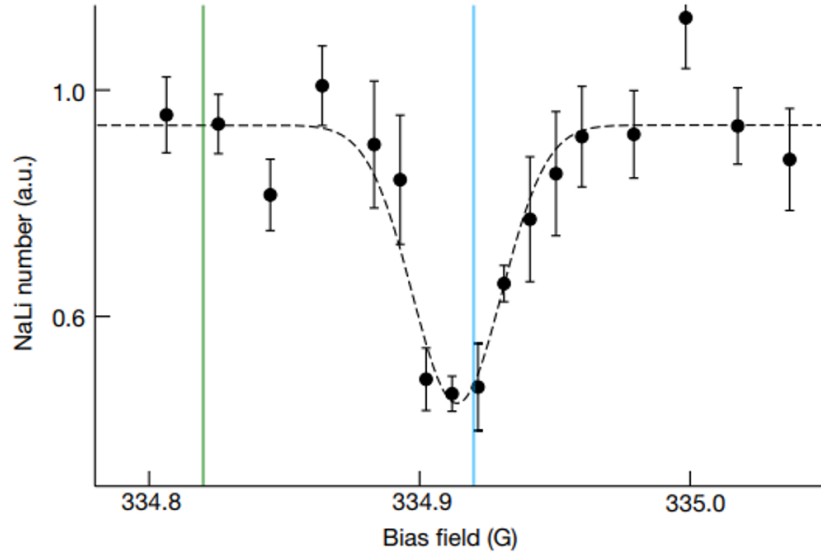


Figure 4.1: The average number of molecules remaining after a fixed hold time for a given magnetic field. At 334.9 Gauss, the collisional losses go dramatically up, the unique signature of a Feshbach resonance [34].

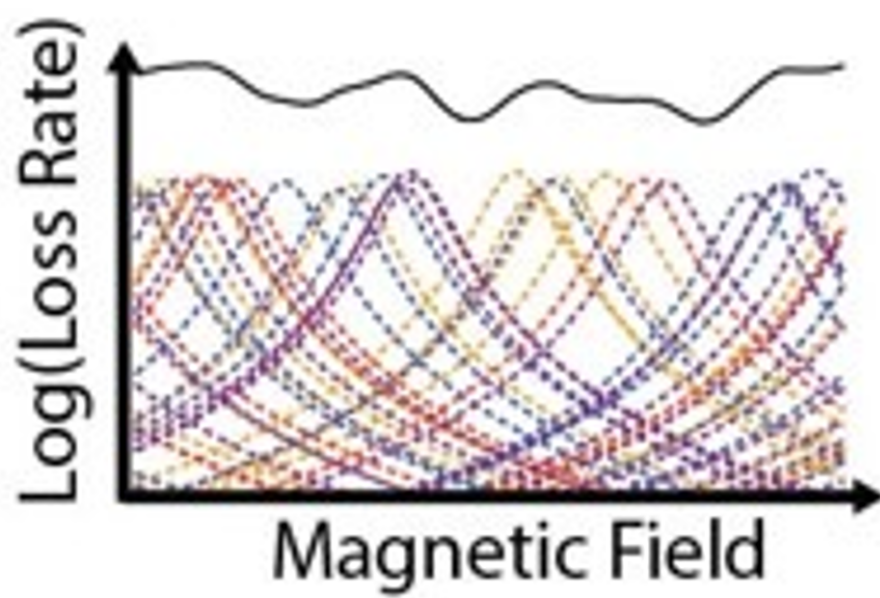


Figure 4.2: Illustration of how the universal loss rate was believed to be a result of multiple overlapping resonances.

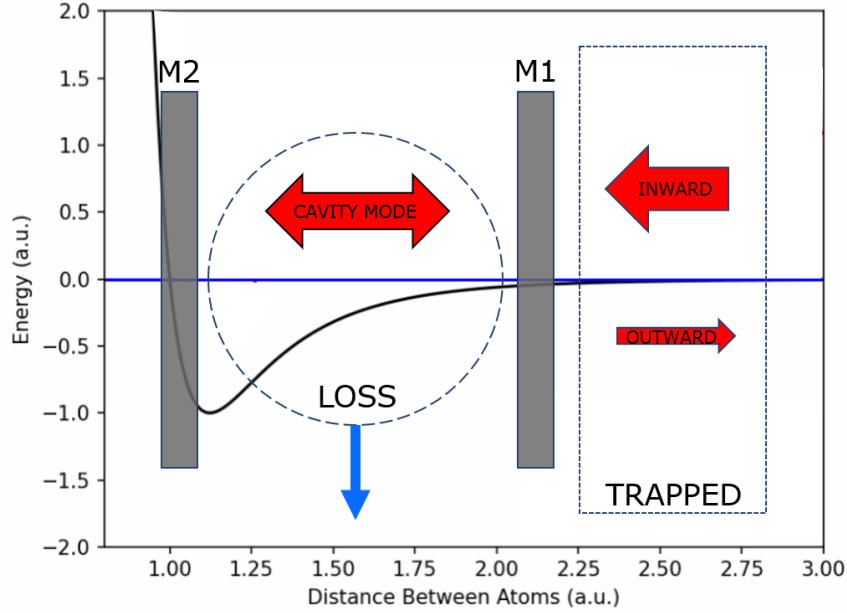


Figure 4.3: Universal Rate Loss Modelled as a Fabry-Perot Cavity [41].

4.2.1 triplet ground-state Molecule Mixture

In the proposed experiment, we will prepare a triplet ground-state NaLi molecule in a mixture of two states by applying a RF light. The technique induces a superposition of states, which then rapidly decoheres into a mixture. To find the RF frequency we need to use, we construct the Hamiltonian of a molecule in a magnetic field. We then diagonalize the matrix to determine the eigenvalues and find the energy levels. Through this model, we can then calculate the energy separation between levels at a specific magnetic field giving us the required RF light frequency.

The Hamiltonian includes a hyperfine coupling term and Zeeman energy term for triplet ground-state

$$H = H_{HF} + H_{\text{Zeeman}}. \quad (4.1)$$

The hyperfine interaction Hamiltonian for two electron spins interacting with nuclear spins is

$$H_{HF} = a_1 \vec{S} \cdot \vec{I}_{\text{Na}} + a_2 \vec{S} \cdot \vec{I}_{\text{Li}}, \quad (4.2)$$

where $a_1 = 433.2$ MHz and $a_2 = 74.6$ MHz [34] are the hyperfine coupling constants. The

Zeeman Hamiltonian accounts for the interaction of the magnetic moment with the B field

$$H_{\text{Zeeman}} = -\mu_B g_S \vec{B} \cdot \vec{S} - \mu_N g_{I1} \vec{B} \cdot \vec{I}_{Na} - \mu_N g_{I2} \vec{B} \cdot \vec{I}_{Li}, \quad (4.3)$$

The nuclear magneton μ_N is significantly smaller than the Bohr magneton μ_B because it is proportional to the inverse of the proton mass which is approximately 1836 times the mass of an electron. Because of this the electron magnetic moment dominates the interaction with the magnetic field.

Combining these two, the total Hamiltonian is

$$H = a_1 \vec{S} \cdot \vec{I}_{Na} + a_2 \vec{S} \cdot \vec{I}_{Li} + \frac{\mu_B}{\hbar} \left(g_S \vec{S} + g_{I_{Na}} \vec{I}_{Na} + g_{I_{Li}} \vec{I}_{Li} \right) \cdot \vec{B}. \quad (4.4)$$

Using python to solve for the eigenvalues, we can plot them at each value of B. From the solution, we see that the Zeeman effect breaks the degeneracies between our 36 states allowing us to distinguish the 8 energy levels into their respective Zeeman levels, figure 4.4. We can calculate the separation between two states and determine the required RF transfer light¹.

4.3 Collisions Loss Rate in N = 1

During the previous study in BEC3, the team saw new interactions between internal spin/spin and spin/rotational dynamics (the discovery of the unexpected *p*-wave Feshbach resonance.) Here we'd like to investigate whether these interactions can also be observed when you add a unit of rotational energy.

Previously, the stretched hyperfine state reduced the loss rate. However, if we go to N=1 state, it's an open question to whether we lose this protective effect or not. It's even possible that the protective effective is enhanced.

Also, the reduced lost rate in the N=0 state allowed us to resolve a Feshbach resonance. If going to the higher rotational state, and there is still some protection, we may be able to resolve a possibly new spectrum of resonances.

With the interaction of rotational energy, this could lead to new interactions. Conversely, if the protection effect is enhanced, this could lead us to a new set of stable

¹For example, the RF Transfer Frequency between states 36 and 27 at 10 Gauss is 112.052543 MHz. For the details on how the program performs the calculation, see the code comments in the provided code A.

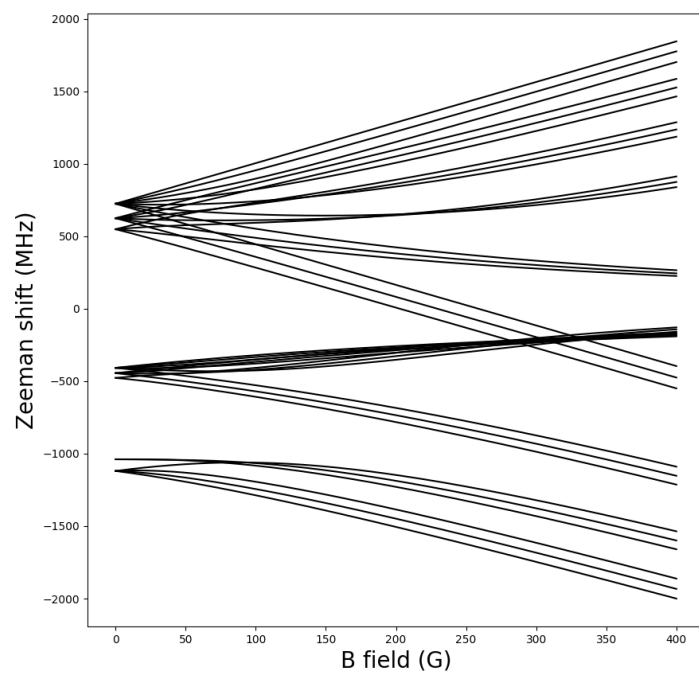


Figure 4.4: Solving the Hamiltonian of the triplet ground-state molecule, we see that there are total of 36 states, which become degenerate and result in 8 energy levels at low field. From these results, the separation between two states can be calculated. For example, the RF transfer frequency between states 36 and 27 at 10 Gauss is 112.052543 MHz.

interaction dynamics. Either way, by measuring the loss rate and the Feshbach resonance spectrum, we may be able to adjust or update current models where these effects are less well understood. This will help us to understand the role of rotation in molecular collisions.

4.4 Coherence of $N=1$ and $N=0$ in a Magnetic Trap

Another avenue that we will pursue in [BEC3](#) is the measurement of the coherence of mixed rotational states in the magnetic trap. Because these states have coupled electric dipole moments, this could lead to long coherence times.

By selecting states with similar magnetic moments, we may be able to optimize coherence times. Long times would increase the stability of the molecular states, which would demonstrate an advantage for ultracold molecules in quantum computing.

Because of the electric dipole coupling, these interactions can more readily be controlled. This means that ultracold molecules might be a good platform for quantum computing with a long enough coherence time to implement error correction schemes.

Exploring the coherence properties of the system could have implications for precision measurement (for example, through precise interference measurements after interacting with external forces) and opening up a route to quantum simulations of many body physics (by setting up large lattice arrays of ultracold molecules with long coherence times and precisely controlling the interaction dynamics between them).

4.5 Photoassociation In The Magnetic Trap

The final experiment we will discuss in this chapter is an attempt to photo-associate molecules into a four body excited bound state. Given the current setup, there's very little modification to the experimental apparatus necessary.

After we have formed the Feshbach molecules and transferred them down to the triplet ground-state. This state is magnetically trappable, so it should be a relatively straightforward sequence to transfer from the [ODT](#) to the magnetic trap.

With molecules magnetically trapped, we will attempt to induce photoassociative losses on the collisional complex. If it is possible to photo-associate the molecules, we can then search for a down leg frequency to possibly transfer into a new four-body ground state in a

similar atom-atom [STIRAP](#) process. This work would enable us to gain new insights into the collision interactions between molecules and resulting bound states.

In this chapter, we explored four experimental goals for our setup in [BEC3](#). We began by discussing the search for *s*-wave Feshbach resonances, emphasizing their potential to provide deeper insights into atomic interactions and enhance our ability to control quantum systems. We then examined the collision loss rate in $N = 1$ rotational states, probing whether the stability observed in $N = 0$ states extends to higher rotational states. Next, we discussed the coherence properties of $N = 1$ and $N = 0$ states in a magnetic trap. We focused on optimizing coherence times for potential quantum computing applications. Finally, we outlined an experiment to photo-associate molecules in a magnetic trap where we might gain new insights into molecule/molecule collision dynamics.

Chapter 5

Conclusion

Throughout this thesis, we have explored the theory and practical implementation of making ultracold molecules. In Chapter 1, after a brief overview of the history of ultracold physics focused on advancements used in the lab, we discussed the three main stages of making ultracold molecules: catch, cool, and make molecules. In chapter 2, we covered how to establish a source of atoms and how we slowed them down enough to catch them in a MOT. Then we covered how we took slowed, trapped atoms and transferred them into a magnetic trap so we could perform evaporative cooling. In chapter 3, we reviewed optical dipole traps and how we transferred the ultracold atoms to the ODT to perform a Landau Zener sweep, preparing our sample for high field Feshbach molecule formation. After obtaining excited triplet state NaLi molecules, we covered the process of transitioning to the triplet ground-state using STIRAP. Finally, we gave an introductory-level overview of four possible experiments that we believe would lead to interesting physics: searching for s -wave Feshbach resonances, collisional loss rate measurement in $N = 1$, coherence times of $N = 1$ and $N = 0$ in a magnetic trap, and photoassociation in the magnetic trap.

It is my hope that this thesis can serve as a starting place for any new graduate students working on ultracold molecules looking to garner an introductory understanding of the overall process, as well as learn some techniques for solving the inevitable problems that arise during these complex experiments. Although we are not operating at the high energy frontier or the high precision frontier, we're at the forefront of the complexity frontier. These are some of the most complicated tabletop experiments in the world. This presents its own unique problems, but also its own unique rewards. Working with ultracold molecules unlocks a vast landscape of new physics ready for those willing to take up the challenge.

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APPENDICES

Appendix A

A.1 Triplet Ground State Molecule Python Program

```
import numpy as np
import matplotlib.pyplot as plt

#####
# VALUES FOR NA AND LI #
#####
S = 1.                                #electron spin
I1 = 1.5                             #1.5 Sodium nucleus
I2 = 1.                               #1 Lithium nucleus
gS = 2.002319                         #electron spin g factor
gI1 = -0.00080461                     #sodium nuclear g factor
gI2 = -0.000447654                    #lithium nuclear g factor
a1 = 433.2                            #Hyperfine Constant aNa = 433.2MHz
a2 = 74.6                             #Hyperfine Constant aLi = 74.6MHz
muB = 1.3996245                       #Bohr magneton in frequency units

#####
# FUNCTIONS #
#####

# Find the number of states and check J
def _getN(J):
    if not(2*J == int(2*J)):
        raise ValueError("J must be half of an integer")
    return int(2*J+1)

# Finds the index of m in the standard basis for spin J
def AngIndex(J,m):
    ind_f = m+J #formula to go from m to index, but potentially gives floats
    ind_i = int(ind_f) #make it an int
    assert ind_f == ind_i #make sure we were given valid inputs
    return ind_i

# Returns the state vector for m in the standard basis for spin J
def AngState(J,m):
    N = _getN(J)
    state = np.zeros(N, dtype=np.complex)
    state[AngIndex(J,m)] = 1
    return state

# Angular momentum operators
def AngLadder(J,sign):
    """
    Finds the ladder operator for spin J
    sign = 1: raising
    sign = -1: lowering
    assumes the basis |J m_J> order starting with m_J = -J
    """
    if sign != 1 and sign != -1:
```

```

        raise ValueError("sign must be +- 1")

    N = _getN(J)
    Mat = np.zeros((N,N),dtype=np.complex128)
    mRange = np.arange(-J,J) if sign == 1 else np.arange(-J+1,J+1)
    for m in mRange:
        Mat[AngIndex(J,m+sign),AngIndex(J,m)] = np.sqrt(J*(J+1)-m*(m+sign))
    return Mat

# Operator matrix for z component of angular momentum in standard basis
def AngZ(J):
    N = _getN(J)
    Mat = np.zeros((N,N),dtype=np.complex128)
    for m in np.arange(-J,J+1):
        Mat[AngIndex(J,m),AngIndex(J,m)] = m
    return Mat

def AngX(J):
    return 0.5*(AngLadder(J,1) + AngLadder(J,-1))

def AngY(J):
    return -1j*0.5*(AngLadder(J,1) - AngLadder(J,-1))

def sorted_eigs(H):
    vals, vecs = np.linalg.eig(H)
    valsr = [v.real for v in vals]
    vecs = np.transpose(vecs)
    pairs = sorted(zip(valsr,vecs), key=lambda x: x[0])
    vals2 = [p[0] for p in pairs]
    N=len(vals)
    vecs2 = [p[1].reshape(N,1) for p in pairs]
    return vals2, vecs2

#####
# MOLECULE CLASS #

class Molecule(object):
    def __init__(self,S,I1,I2,gS,gI1,gI2,a1,a2):
        self.S=S
        self.I1=I1
        self.I2=I2
        self.gS=gS
        self.gI1=gI1
        self.gI2=gI2
        self.a1=a1
        self.a2=a2

        # construct the Hamiltonian
        eyeI1 = np.eye(_getN(I1))
        eyeI2 = np.eye(_getN(I2))
        eyeI = np.kron(eyeI1, eyeI2)

        I1x = np.kron(AngX(I1), eyeI2)
        I1y = np.kron(AngY(I1), eyeI2)
        I1z = np.kron(AngZ(I1), eyeI2)

        I2x = np.kron(eyeI1, AngX(I2))
        I2y = np.kron(eyeI1, AngY(I2))
        I2z = np.kron(eyeI1, AngZ(I2))

        # S-I
        SdotI1 = np.kron(I1x, AngX(S)) + np.kron(I1y, AngY(S)) + np.kron(I1z, AngZ(S))
        SdotI2 = np.kron(I2x, AngX(S)) + np.kron(I2y, AngY(S)) + np.kron(I2z, AngZ(S))

        # RF operators
        eyeS = np.eye(_getN(S))
        self.Sx = np.kron(eyeI, AngX(S))
        self.Sy = np.kron(eyeI, AngY(S))
        self.Sz = Sz = np.kron(eyeI, AngZ(S))
        self.F1z = Sz + np.kron(I1z, eyeS)
        self.N = _getN(S)*_getN(I1)*_getN(I2)
        self.H_HF = self.a1*SdotI1 + self.a2*SdotI2
        self.m1 = m1 = np.kron(AngZ(I1), np.kron(eyeI2, eyeS))
        self.m2 = m2 = np.kron(eyeI1, np.kron(AngZ(I2), eyeS))
        self.mu = muB*gS* Sz
        self.mu = muB*(gS* Sz + gI1*m1 + gI2*m2)

```

```

def eig(self, B):
    H = self.H_HF + B*self.mu
    vals2, vecs2=sorted_eigs(H)
    return vals2, vecs2

#####
# CALCULATIONS #

NaLi = Molecule(S,I1,I2,gS,gI1,gI2,a1,a2)

# PLOT ZEEMAN VS BFIELD
# Eigenvalues for each B value.
B = 400 #in Gauss
N = 1000
B_span = np.linspace(0, B, N)
vals = []
vecs = []
for i in range(N):
    a, b= NaLi.eig(B_span[i])
    vals.append(a)
    vecs.append(b)

fig = plt.figure(figsize=(10, 10))
axs = fig.add_subplot(111)
axs.plot(B_span, vals, '-o', c="k", markersize=0.5, label = '0')
axs.set_ylabel('Zeeman shift (MHz)', fontsize=20)
axs.set_xlabel('B field (G)', fontsize=20)
#plt.savefig('GS mol shift 3.png')
plt.show()

#RF TRANSFER BETWEEN TWO STATES (36 DIFFERENT STATES) AT BFIELD
State_A = 36
State_B = 27
B_RF = 10
n, m = NaLi.eig(B_RF)
frequency_difference = round(n[State_A - 1] - n[State_B - 1], 6)
print(f"The RF Transfer Frequency between {State_A} and {State_B} at {B_RF} Gauss is {frequency_difference} MHz.")

```

A.2 RF Transfer Python Program

```
#####
#####

"Refactored Code. Paul Del Franco 12/15/2023"

#####
#####

#IMPORTS
import numpy as np
import matplotlib.pyplot as plt

#####
#####

# CLASSES
class AtomIJ(object):

    """
    Models an atom as a nuclear spin I and electron spin J
    Assumes we have chosen n,L
    """
    def __init__(self,name,L_label,I,J,gI, gJ, A, B=0, muBref=1.399624604):
        self.name = name
        self.L_label = L_label
        self.I = I
        self.J = J
        self.gJ = gJ          #electron Lande g factor
        self.gI = gI          #nuclear g factor
        self.A = A            #hyperfine constant A
        self.B = B            #hyperfine constant B
        self.muB = muBref     #Bohr magneton
        self._init()

    def _init(self):
        #initialize the Hamiltonian
        I = self.I; J=self.J
        NI = _getN(I)
        NJ = _getN(J)
        eyeI = np.eye(NI); eyeJ = np.eye(NJ)
        eye = np.kron(eyeI, eyeJ)

        IdotJ = np.kron(AngX(I), AngX(J)) + np.kron(AngY(I), AngY(J))+np.kron(AngZ(I), AngZ(J))
        if J != .5 and I != .5 and self.B != 0:
            Bterm = self.B*(3*np.dot(IdotJ,IdotJ)+ 1.5*IdotJ - eye*I*(I+1)*J*(J+1))/(2*I*(2*I-1)*J*(2*J-1))
        else:
            Bterm = 0*IdotJ

        self.HF = self.A*IdotJ + Bterm #hyperfine Hamiltonian / h

        mJ = np.kron(eyeI, AngZ(J))
        mI = np.kron(AngZ(I), eyeJ)
        self.mu = self.muB*(self.gJ*mJ + self.gI*mI) #total magnetic moment operator / h

        if J == int(J):
            Jlabel = "$_{%d}$" % int(J)
        else:
            Jlabel = "$_{%d/%d}$" % (int(2*J), 2)
        self.term = self.L_label + Jlabel

    def eig(self,Bz):
        """
        find the energies at field Bz
        returns vals, vecs
        """
        H = self.HF + self.mu*Bz
        vals, vecs = np.linalg.eig(H)
        #sort:
        vecs = np.array([x for (y,x) in sorted(zip(vals,np.transpose(vecs)), key=lambda pair: pair[0].real)])
        vals = np.array(sorted(vals))

        return vals.real, vecs

#####
```

```

#####

#FUNCTIONS
def _getN(J):
    """
    #Find the number of states and check J
    """
    if not(2*J == int(2*J)):
        raise ValueError("J must be half of an integer")
    return int(2*J+1)

def AngIndex(J,m):
    """
    Finds the index of m in the standard basis for spin J
    """
    ind_f = m+J #formula to go from m to index, but potentially gives floats
    ind_i = int(ind_f) #make it an int
    assert ind_f == ind_i #make sure we were given valid inputs
    return ind_i

def AngLadder(J,sign):
    """
    Finds the ladder operator for spin J
    sign = 1: raising
    sign = -1: lowering

    assumes the basis |J m_J> order starting with m_J = -J
    """
    if sign != 1 and sign != -1:
        raise ValueError("sign must be +- 1")

    N = _getN(J)
    Mat = np.zeros((N,N))
    mRange = np.arange(-J,J) if sign == 1 else np.arange(-J+1,J+1)
    for m in mRange:
        Mat[AngIndex(J,m+sign),AngIndex(J,m)] = np.sqrt(J*(J+1)-m*(m+sign))
    return Mat

def AngZ(J):
    """
    Operator matrix for z component of angular momentum in standard basis
    """
    N = _getN(J)
    Mat = np.zeros((N,N))
    for m in np.arange(-J,J+1):
        Mat[AngIndex(J,m),AngIndex(J,m)] = m
    return Mat

def AngX(J):
    """
    Operator matrix for x component of angular momentum in standard basis
    """
    return 0.5*(AngLadder(J,1) + AngLadder(J,-1))

def AngY(J):
    """
    Operator matrix for y component of angular momentum in standard basis
    """
    return -1j*0.5*(AngLadder(J,1) - AngLadder(J,-1))

def landeG(L,L1,L2,g1,g2):
    def term(g,a,b):
        return g*(L*(L+1.)-b*(b+1.)+a*(a+1.))/(2.*L*(L+1.))
    return term(g1,L1,L2) + term(g2,L2,L1)

def findDiffFreq(atom_gnd, atom_exc, ind_g, ind_e, Bz):
    """
    Find the transition frequency between ground and excited states
    Gives the answer relative to the difference between the lines' centers of mass
    i.e. don't make any correction for overall energy offset
    """
    vals_g, vecs_g = atom_gnd.eig(Bz)
    vals_g = sorted(vals_g)

    vals_e, vecs_e = atom_exc.eig(Bz)
    vals_e = sorted(vals_e)
    return vals_e[ind_e] - vals_g[ind_g]

```

```

#####
#####

# Sodium Values
I = 1.5                #Nuclear Spin
S = 0.5                #Electron Spin
gL = 0.99997613        #Electron Orbital g-factor
gS = 2.0023193043622   #Electron Spin g-factor
gI = -0.00080461080    #Nuclear g-factor

#Ground State S_1/2
L_g = 0; J_g = 0.5
gJ_g = landeG(J_g, L_g, S, gL, gS)
A_gnd = 885.81306440    #Magnetic Dipole Constant
B_gnd = 0               #Electric Quadrupole Constant
Na_gnd = AtomIJ("$~{23}$Na", "S", I, J_g, gI, gJ_g, A_gnd, B_gnd)

#Excited State P_1/2
L_e1 = 1; J_e1 = 0.5
gJ_e1 = landeG(J_e1, L_e1, S, gL, gS)
A_D1 = 94.44
B_D1 = 0
Na_exc1 = AtomIJ("$~{23}$Na", "P", I, J_e1, gI, gJ_e1, A_D1, B_D1)

#Excited State P_3/2
L_e2 = 1; J_e2 = 1.5
gJ_e2 = landeG(J_e2, L_e2, S, gL, gS)
A_D2 = 18.534
B_D2 = 2.724
Na_exc2 = AtomIJ("$~{23}$Na", "P", I, J_e2, gI, gJ_e2, A_D2, B_D2)

# Lithium Values
I = 1
S = 0.5
gL = 0.99999587
gS = 2.0023193043737
gI = -0.00080461080

#ground state
L_g = 0; J_g = 0.5
gJ_g = landeG(J_g, L_g, S, gL, gS)
A_gnd = 152.1368407
B_gnd = 0
Li_gnd = AtomIJ("$~{6}$Li", "S", I, J_g, gI, gJ_g, A_gnd, B_gnd)

#excited state P_1/2
L_e1 = 1; J_e1 = 0.5
gJ_e1 = landeG(J_e1, L_e1, S, gL, gS)
A_D1 = 17.386
Li_exc1 = AtomIJ("$~{6}$Li", "P", I, J_e1, gI, gJ_e1, A_D1, B=0)

#excited state P_3/2
L_e2 = 1; J_e2 = 1.5
gJ_e2 = landeG(J_e2, L_e2, S, gL, gS)
A_D2 = -1.155
B_D2 = -0.10
Li_exc2 = AtomIJ("$~{6}$Li", "P", I, J_e2, gI, gJ_e2, A_D2, B_D2)

#####
#####

#FIND and PLOT THE RF TRANSFER FREQUENCY

#Choose Levels and States (Defined Above)
Level_1 = Na_gnd      # From Ground, First Excited, Second Excited
Level_2 = Na_gnd      # To Ground, First Excited, Second Excited
State_1 = 0           # From m_F level
State_2 = 7           # To m_F level

#Choose Bz Value [Gauss] (BEC3 Experimental Value of 2.87G)
Bz = 2.87

#Select States (Defined Above) and m_F levels
nu_rf = findDiffFreq(Level_1, Level_2, State_2, State_1, Bz)
rf_info = f"For B-Field: {Bz} G\nRF Transfer Frequency: {np.round(-nu_rf, 2)} Hz"

```

```

#Display result and Plot RF Transfer Freq vs Gauss
diff_81 = []
Bfield = np.linspace(0, 800, 100)
for B in Bfield:
    diff_81.append(-findDiffFreq(Level_1, Level_2, State_2, State_1, B))

fig, ax = plt.subplots(figsize=(10, 6))

ax.plot(Bfield, diff_81, label=f"State {State_2} to {State_1}", color='navy', linewidth=2)
ax.legend(title='Transition', loc='upper left')
ax.set_ylabel('Quadratic Zeeman Shift (KHz)', fontsize=14)
ax.set_xlabel('B Field (G)', fontsize=14)
ax.set_title('Quadratic Zeeman Shift as a Function of Magnetic Field', fontsize=16)
ax.tick_params(axis='both', which='major', labelsize=12)
ax.grid(True, which='both', linestyle='--', linewidth=0.5)

# Adjusting the position and style of the additional information
ax.text(0.5, -0.35, rf_info, transform=ax.transAxes, ha='center', va='center', fontsize=16,
        bbox=dict(facecolor='white', alpha=0.5, boxstyle='round,pad=1'))

plt.tight_layout()
plt.show()

```

A.3 BEC3 Apparatus

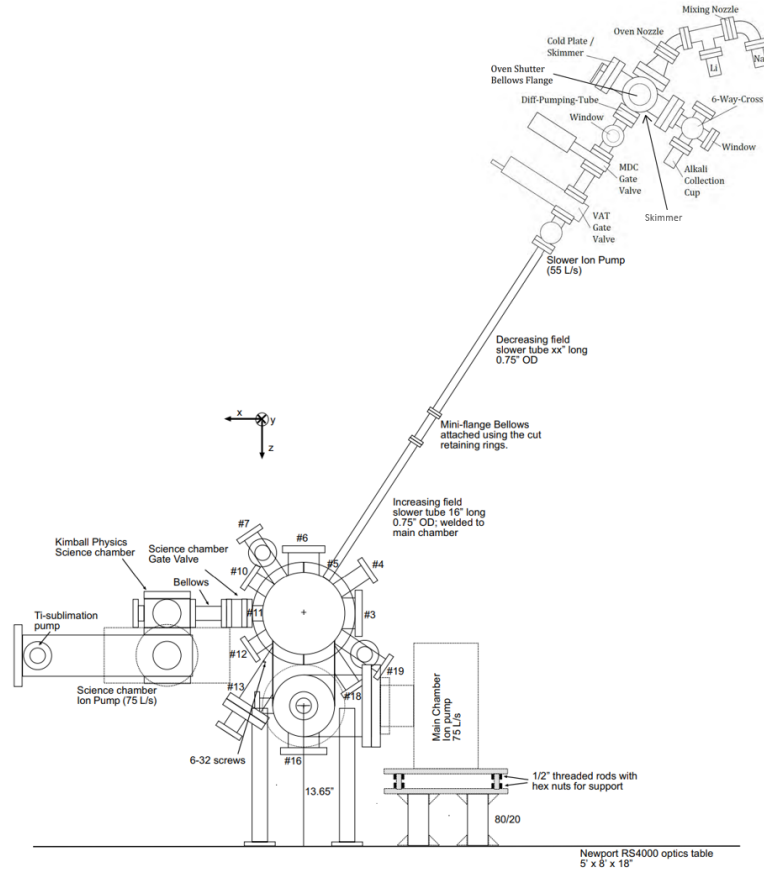


Figure A.1: BEC3 Apparatus Diagram with updated two species oven design [3].