A Precise Measurement of the Indium $6P_{1/2}$ Stark Shift Using Two-Step Laser Spectroscopy

by
Allison L. Carter

Professor Protik K. Majumder, Advisor

A thesis submitted in partial fulfillment of the requirements for the Degree of Bachelor of Arts with Honors in Physics

WILLIAMS COLLEGE
Williamstown, Massachusetts
May 20, 2016
Abstract

We have completed a measurement of the scalar polarizability of the $6P_{1/2}$ state of indium. Our results give a value of $\alpha_0 = 7688 \pm 41\text{(stat)} \pm 32\text{(sys)} a_0^3$, which is in statistical agreement with the theoretically predicted value. We lock a 410 nm laser to the $5P_{1/2} \rightarrow 6S_{1/2}$ transition and scan a 1343 nm laser across the $6S_{1/2} \rightarrow 6P_{1/2}$ transition, with the lasers passing through both an atomic beam and a vapor cell. We have developed a new method of locking the 410 nm laser directly to the atomic beam transition, and we use single-tone FM spectroscopy to obtain the necessary signal. We also detect the spectrum from the $6S_{1/2} \rightarrow 6P_{1/2}$ transition for a variety of applied electric fields in the atomic beam using two-tone FM spectroscopy. In the course of this experiment, we have also obtained a precise measurement of the hyperfine splitting of the $6P_{1/2}$ state of 1257.0(5) MHz, for which the uncertainty is dominated by systematic effects. This value is about three times more precise than the previous measurement.
Acknowledgments

First, I would like to thank my advisor, Professor Tiku Majumder, for all of his guidance and mentorship throughout the past year. Additionally, I am grateful for Dr. Milinda Rupasinghe’s contributions to this experiment. I also wish to thank Professor Jeff Strait for his feedback on my writing. Benjamin Augenbraun’s work laid the foundations for this experiment and provided an excellent starting point for me to work from. I am also very grateful to him for introducing me to the lab and for his continued assistance with this work.

I also would like to thank Michael Taylor and Jason Mativi for their help with experimental apparatus modifications. Additionally, I have appreciated Sauman Cheng’s company and experimental advice. Nathaniel Vilas was very helpful during Winter Study, especially with the development of the data analysis code. Finally, my friends and family have been incredibly supportive, and I am appreciative of their ongoing encouragement.
Executive Summary

We have completed a precise measurement of the Stark shift of the $6P_{1/2}$ state of indium. This measurement, one of a series of precise measurements of atomic structure performed in the Majumder lab, serves as a test of atomic theory. Our group focuses on the physics of Group IIIa atoms, which are more complicated and less well-studied than the alkali atoms, but are still simple enough to be theoretically studied. Thus, our measurements help guide the more complicated atomic theory necessary for understanding these atoms.

Additionally, atomic systems can be used to study and search for physics beyond the Standard Model on a smaller scale than in a large particle collider experiment. However, the interpretation of the results of such experiments depends on a very precise knowledge of the underlying quantum mechanics of the atom. Through improving atomic theory, our measurements can thus also improve our capability to understand the particle physics underlying the behavior of atoms.

To perform a Stark shift measurement, we need to apply electric fields of order 10 kV/cm. We rely on a vacuum chamber to work safely with fields this large. Within the vacuum chamber, we heat $\sim 100$ g of indium to a temperature of 1150 °C in a molybdenum crucible. Through the design of the crucible and other collimation devices, we produce an atomic beam that is well-collimated and thus nearly eliminates

![Figure 1: Atomic beam signal with and without applied electric field.](image)
Figure 2: Results from our measurement of the hyperfine splitting of the $6P_{1/2}$ state of indium.

Doppler broadening. This beam passes through a pair of capacitor plates, which we use to apply the electric fields. To observe the atoms, we intersect a pair of overlapped lasers with the atomic beam between the capacitor plates. By collecting many sets of data with the voltage on and off we can then measure the amount by which the energy levels shift when an electric field is applied. Because of the very low optical depth in the atomic beam ($\alpha \sim 10^{-3}$), we rely on two-tone FM spectroscopy to observe our signal.

Figure 3: Results of our measurement of $k_s$

Observation of the $6P_{1/2}$ state requires a two-step transition. We thus lock the laser for the first-step transition, and scan the laser for the second-step transition. One of the significant experimental developments made in the course of the work for this thesis has been the implementation of a new locking scheme for the first-step laser. In previous two-step measurements performed in our lab, the laser for the first
step had been locked to a vapor cell reference signal. We now lock directly to the signal from the atomic beam to account for the shifting resonance frequency with the application of an electric field.

We do, however, continue to use a vapor cell of indium as a reference. We send a portion of the two lasers through the vapor cell and observe the $6P_{1/2}$ hyperfine spectrum there as well. While the primary purpose of this signal is for calibration, we also obtain a measurement of the hyperfine splitting in the $6P_{1/2}$ state of 1257.0(2) MHz (Figure 2), with the uncertainty dominated by small residual systematic uncertainties. This measurement is an order of magnitude more precise than the previous measurement of 1251.5(1.5) MHz [18].

Finally, as mentioned above, we have successfully obtained a measurement of the Stark shift in the $6P_{1/2}$ state of indium. Our direct measurement is the difference between the Stark shift in the $6P_{1/2}$ and $6S_{1/2}$ states, which we write as $\Delta f = k_S\mathcal{E}^2$, where $\mathcal{E}$ is the applied electric field. The calculated value that comes most immediately from our measurement is $k_s$, with results for this number shown in Figure 3. Using the value of the Stark shift of the $6S_{1/2}$ state obtained in [10], we can then determine the shift in just the $6P_{1/2}$ state without loss of precision. Finally, we can convert this measurement into the scalar polarizability $\alpha_0$ of the $6P_{1/2}$ state, which is the atomic parameter we ultimately wish to know. Our value for the scalar polarizability, $\alpha_0 = 7688 \pm 24\text{(stat)} \pm 69\text{(sys)} a_0^3$ (in atomic units) is in agreement with the theoretically predicted value of $\alpha_0 = 7817 \pm 300 a_0^3$. 
Contents

Abstract ................................................................. iii
Acknowledgments ..................................................... v
Executive Summary .................................................. vii

1 Introduction .......................................................... 1
  1.1 Motivation ....................................................... 1
  1.2 Previous Work ................................................... 4
  1.3 Present Work ..................................................... 4
  1.4 Outline of this Thesis .......................................... 5

2 Atomic Theory ....................................................... 7
  2.1 Absorption Spectroscopy Theory ................................ 7
    2.1.1 Direct Absorption Spectroscopy ............................ 7
    2.1.2 Line Broadening ............................................ 7
  2.2 FM Spectroscopy Lineshapes ................................... 10
    2.2.1 Single-Tone FM Spectroscopy ............................... 10
    2.2.2 Two-Tone FM Spectroscopy ................................ 11
  2.3 Stark Shift ..................................................... 13
    2.3.1 Stark Shift Measurements in Three-Level Systems ........ 14
    2.3.2 Two-Step Transition with the Laser for the First Step Always on Resonance ........................................ 16

3 Experimental Setup ................................................ 19
  3.1 Atomic Beam Unit ............................................... 19
    3.1.1 Vacuum Chamber ............................................. 19
    3.1.2 Atomic Source and Beam ................................... 21
    3.1.3 Interaction Region ......................................... 21
  3.2 Optical System and Signal Processing .......................... 22
    3.2.1 Lasers ....................................................... 22
    3.2.2 Locking the 410 nm Laser .................................. 23
    3.2.3 Vapor Cell Reference ....................................... 28
    3.2.4 IR Laser Preparation and Signal Observation .............. 29
  3.3 Data Acquisition ............................................... 32
4 Data Analysis and Results
4.1 Data Analysis .......................................................... 35
  4.1.1 Pre-Calibration Axis Processing .............................. 36
  4.1.2 Frequency Linearization and Calibration using the Fabry-Pérot Signal ............................................. 36
  4.1.3 Measurement of the Hyperfine Splitting of the Indium $6P_{1/2}$ State 40
  4.1.4 Measuring the Stark Shift .................................... 41
4.2 Results ................................................................. 47
  4.2.1 Systematic Error Analysis ................................. 47
4.3 Final Results .......................................................... 53

5 Future Work .......................................................... 55
5.1 Measurement of the In $6P_{3/2}$ Scalar and Tensor Polarizabilities .... 55
5.2 Polarizability Measurements in Thallium .......................... 59

A Data Tables ............................................................ 61

B MATLAB Code .......................................................... 63
  B.1 Master Programs .................................................. 63
  B.3 Peak-Finding Code ................................................. 65
  B.4 Fabry-Pérot Fitting and Frequency Axis Linearization ........ 66
  B.5 Vapor Cell Fitting .................................................. 67
  B.6 Shift Evaluation .................................................... 68
  B.7 Saving Analysis Results ......................................... 70
  B.8 Final Shift Analysis ................................................. 70
# List of Figures

1. Stark Shifted Two-Tone FM Spectroscopy Signal .......................... vii
2. $6P_{1/2}$ Hyperfine Splitting Measurement Results .................... viii
3. Stark Shift Measurement Results .......................... viii

1.1 Indium and Thallium Atomic Structure .......................... 3
2.1 Single Tone FM Spectroscopy Lineshape .......................... 11
2.2 Two-Tone FM Demodulated Lineshape .......................... 12
2.3 Stark Shift Energy Level Diagram .......................... 15
2.4 Three Level System .......................... 16
2.5 Plot of Lineshapes for Various Rabi Frequencies .......................... 17
2.6 Three Level Doppler Broadened System .......................... 18
3.1 Experimental Setup .......................... 20
3.2 Vacuum System .......................... 22
3.3 Interaction Region .......................... 23
3.4 Locking Signal .......................... 24
3.5 410 nm Laser Locking Setup .......................... 25
3.6 Correction Signal Due to Stark Shift .......................... 26
3.7 AOMs Setup .......................... 27
3.8 Vapor Cell Optical Setup .......................... 29
3.9 Vapor Cell Signal with EOM Sidebands .......................... 30
3.10 Experimental Setup for IR Laser Preparation and Observation of the Atomic Beam Signal .......................... 31
3.11 Schematic of Data Collection Procedure .......................... 32
4.1 Sample Imported Data .......................... 35
4.2 Fabry-Pérot Signal–Data and Fit .......................... 37
4.3 Sample Frequency Linearization Fit .......................... 38
4.4 Signal from the Vapor Cell .......................... 39
4.5 Vapor Cell Signal Sideband Splittings .......................... 40
4.6 Hyperfine Splitting Measurement Results .......................... 41
4.7 Systematic Error in Hyperfine Splitting Measurement .......................... 42
4.8 Stark Shifted Atomic Beam Signal .......................... 43
4.9 Lorentzian Fit to Atomic Beam Signal .......................... 45
LIST OF FIGURES

4.10 $\chi^2$ from Overlap Fit ........................................... 46
4.11 Sample Data for Overlap Fit ........................................ 47
4.12 Uncertainty In Individual Scans Compared With Fluctuations Among
   Scans ........................................................................ 48
4.13 Comparison of Results from Binary Experimental Conditions .... 49
4.14 Measured $k_s$ versus Ratio of Laser Powers ....................... 50
4.15 Plot of $k_s$ against Applied Voltage ............................... 51
4.16 Results of $k_s$ with Different Fitting Methods .................... 51
4.17 Histogram of All $k_s$ Values ......................................... 52
4.18 Comparison of Theoretical and Experimental Polarizabilities .... 54

5.1 Diagram of Indium Energy Levels with $6P_{3/2}$ State .............. 56
5.2 Simulated Vapor Cell $6P_{3/2}$ Spectrum ............................. 57
5.3 Simulation of the Atomic Beam $6P_{3/2}$ Spectrum .................. 58
5.4 Simulated Splitting of the $6P_{3/2}$, $F = 6$ State ................... 59
List of Tables

2.1 Line Broadening .......................................................... 10
4.1 Table Of Systematic Errors ........................................... 53
A.1 Summary of Data Collection and Results from Overlap Method ... 61
Chapter 1

Introduction

In many ways, atoms are remarkable for their simplicity. They are ubiquitous, yet we rarely appreciate their existence, let alone their subtler properties. These properties, however, prove extremely useful for understanding the physics that governs the way the smallest systems in the universe behave. The Majumder lab seeks to measure the structure of atoms, indium and thallium in particular, with high precision to improve understanding of this physics. This thesis focuses on the completed measurement of the Stark shift (or the scalar polarizability) in the $6P_{1/2}$ state of indium.

1.1 Motivation

Motivation for Precise Atomic Measurements

We know that there are elements of the universe that the standard model fails to explain fully. Thus, “new physics” beyond the standard model must be present, but it is extremely challenging to detect experimentally such physics. Often, when we picture experimental searches for this new physics, we imagine large particle colliders and hundreds of scientists working on the relevant problems. Atomic physics, however, permits a much smaller scale experiment for explorations of this “exotic” physics; we can perform experiments on atoms in a single room.

In any such experiment, however, both the quantum mechanics of atoms and the physics from and beyond the standard model play a role in determining the behavior of the atoms. For example, thallium has historically been used to test for effects due to parity nonconservation as a result of the weak interaction [1]. The specific effect studied was an optical rotation $\epsilon_w$, which can be written as

$$
\epsilon_w = Q_w C(Z)
$$

(1.1)

$Q_w$ describes the fundamental standard model physics, while $C(Z)$ encapsulates the atomic physics. $C(Z)$ is calculated from atomic theory, and $\epsilon_w$ is measured, providing a value for $Q_w$. It is clear, then, that in order to know $Q_w$ precisely, we must be able both to calculate $C(Z)$ and measure $\epsilon_w$ precisely. While this particular
experiment sought to measure $\epsilon_w$, the quantity related to fundamental physics, we investigate the atomic properties related to $C(Z)$.

Standard quantum mechanics tells us that the wavefunctions and resulting properties of any atom more complicated than hydrogen (so all other atoms) cannot be calculated exactly. Thus, even the best theory for other atoms, and quantities such as $C(Z)$, will be approximate. Through high precision measurements, we find values with uncertainties smaller than those in the most precise theoretical calculations. For complicated atoms like indium and thallium, there is more than one theoretical approach to the calculations [2]. The theorists with whom we collaborate, (especially the Safronova group at University of Delaware), can then use our results to test and guide their calculations.

Motivation for Stark Shift Measurements

As mentioned above, the focus of this thesis is on the measurement of a Stark shift. The Stark shift is a shift in the energy levels of an atom due to an applied electric field, and it depends on the wavefunctions of the atom (see Section 2.3 for more details). As a test of atomic theory, such a measurement is clearly relevant. Additionally, as one might expect given that the shift is induced by an external field, the Stark shift is most pronounced in higher energy states of an atom. One of the other atomic properties on which the Majumder lab focuses is the hyperfine splitting. This effect is based on the interaction of the electron with the nucleus, and therefore is largest in states with large nuclear overlap. The measurements of these two properties therefore complement each other, providing information about the wavefunction of the electron both near and further away from the nucleus.

Stark shifts can be simply related to atomic polarizabilities, especially for states with only scalar polarizabilities (such as the state we are studying). Precise values of the polarizability of atoms also turn out to be important for a number of applications. However, measurements of polarizabilities, even for simpler atoms than indium and thallium, are fairly rare [3]. These measurements are especially lacking for excited states, such as the one discussed in this thesis [4]. In particular, one application that will require a more precise knowledge of atomic polarizabilities is increasing the precision of atomic clocks. Currently, one of the largest uncertainties in atomic clocks is shifts in the atomic energy levels due to blackbody radiation (BBR) [3, 4]. The size of these shifts depend on the polarizabilities. Precise knowledge of polarizabilities is also important for applications in the areas of cooling and trapping atoms and quantum information [3].

Motivation for Use of Indium and Thallium

The Majumder lab has focused on measurements of Group IIIa atoms, and in particular, indium ($Z = 49$) and thallium ($Z = 81$), the two heaviest stable atoms in the group. Group IIIa atoms are of interest since the atoms are more complicated than the already well-studied Group I atoms. However, due to their $ns^2np^1$ valence
1.1. **MOTIVATION**

(a) Energy levels of indium.

(b) Energy levels of thallium.

**Figure 1.1:** Diagrams of a selection of energy levels in indium and analogous levels in thallium.

Structure and its similarity to the single valence electron of hydrogen, they are simple enough to be tractable for theoretical calculations. (It is interesting to note that, for polarizabilities in particular, alkali atoms have been much more thoroughly studied than other atoms [3]).

Furthermore, as mentioned above, thallium has been used as a test for parity nonconservation and for studying the weak interaction [5, 6]. $C(Z)$ from Equation 1.1 scales as $Z^3$, so heavy elements like thallium are appealing for such experiments. In another search for new fundamental physics, thallium also was used in an experimental search for a permanent electron electric dipole moment [7].

While thallium has been more frequently used in the past for searches for new physics, due to its similar structure, indium has similar atomic properties (see Figure 1.1). Therefore, for our goal of providing measurements to improve theoretical calculations of atomic wavefunctions, indium and thallium can be used for similar purposes and with similar outcomes. Additionally, indium has been suggested as a possible system for a search for a permanent atomic electric dipole moment, which is predicted not to occur in the standard model, but could arise from new physics [8]. Indium also has the advantage over thallium of not being highly toxic.

In examining Figure 1.1, we note first the very similar structures of the two atoms. From a practical standpoint, the similar wavelengths of the transitions are useful, as similar optical equipment can be used for both atoms. Figure 1.1 also shows the hyperfine structure of the two elements. For the current work in indium, we consider states with total orbital angular momentum $L = 0$ or 1 and total spin $S = 1/2$. This
leads to orbital angular momentum for the electron of either $J = 1/2$ or $J = 3/2$. In this thesis, we concentrate on the $6P_{1/2}$ state, which has $J = 1/2$. (Future work, however, will consider the more complicated $J = 3/2$ state). For $J = 1/2$ states, indium’s nuclear spin of $I = 9/2$ gives it two hyperfine levels with total angular momentum $F = 4$ and $F = 5$. Thallium, on the other hand, has nuclear spin $I = 1/2$, so its hyperfine levels in $J = 1/2$ states have $F = 0$ or $1$. In indium $J = 3/2$ states, there are four hyperfine levels present, with $F = 3, 4, 5, 6$.

The hyperfine splittings are larger for lower energy levels ($\sim 11.4$ GHz for the $5P_{1/2}$ state of indium, $\sim 8.4$ GHz for the $6S_{1/2}$ state, and $\sim 1.2$ GHz for the $6P_{1/2}$ state. Since the splittings of the two lower states are so large, we must choose one transition with which to run our experiment for any given trial. We expect no difference in our Stark shift results based on which transition we drive, since we are studying a state with $J = 1/2$ and the polarizability should thus be independent of the hyperfine state. However, we collect data with our blue laser locked to two different transitions ($5P_{1/2}, F = 4 \rightarrow 6S_{1/2}, F' = 5$ and $5P_{1/2}, F = 5 \rightarrow 6S_{1/2}, F' = 4$) to check that our results for both transitions are in fact the same. The smaller splitting in the $6P_{1/2}$ state allows us to scan the IR laser across both possible transitions, and thus observe both transitions in every data set.

1.2 Previous Work

Over the course of the Majumder lab’s work, many measurements of atomic properties of indium and thallium have been made. Here, we provide a brief overview of some of this work to contextualize the current experiment. Early experiments in the group focused on thallium, measuring, among other properties, the Stark shift in the $6P_{1/2} \rightarrow 7S_{1/2}$ transition in thallium [9]. Subsequently, work with thallium continued, and the group also began working on measurements of indium. The experiments in indium that have led up to this experiment have consisted of a measurement of the hyperfine splitting in the $6P_{1/2}$ state [10] and a measurement of the scalar polarizability of the $6S_{1/2}$ state [11].

Many of the other experiments not mentioned explicitly here have also looked at the hyperfine structure of thallium, typically in vapor cell environments. Along with the measurement of the $6P_{3/2}$ indium hyperfine structure, these experiments have helped develop the techniques we use for vapor cell spectroscopy in the current experiment. Similarly, the previous Stark shift measurements, which were conducted using an atomic beam, have been instrumental in developing the apparatus and techniques used in the current atomic beam part of the experiment.

1.3 Present Work

In this thesis, we present the completed measurement of the Stark shift in the $6P_{1/2}$ state in indium. This measurement constitutes the first two-step laser spectroscopy
measurement of a Stark shift performed in the Majumder lab. Part of the challenge with any Stark shift measurement is that one can only measure the difference in the Stark shifts of two energy levels (in this case, the difference in the Stark shift of the $6P_{1/2}$ and $6S_{1/2}$ states). However, as mentioned in Section 1.2, our group has already precisely measured the Stark shift in the $6S_{1/2}$ state. We can therefore compute the Stark shift in the $6P_{1/2}$ state from our measurement with negligible loss of precision. Also, as discussed previously, the state that we are examining only has a scalar polarizability, which is simply related to the Stark shift. Thus, in measuring the Stark shift, we can say equivalently that we have measured the scalar polarizability.

1.4 Outline of this Thesis

We begin by presenting the theory behind the measurements that we are making and the techniques we utilize in carrying out this experiment. This chapter includes a discussion of absorption spectroscopy, with both direct and frequency modulation techniques, and the resulting lineshapes, which we need to know to analyze our data. We also discuss the theory that describes Stark shifts. Subsequently, this chapter includes a summary of the theory describing three-level quantum optical systems. While the more complicated aspects of this theory are no longer directly relevant to our measurement, they have motivated much of the development of the experimental setup that has been undertaken during this past year. We then continue on to a discussion of the experimental setup for our experiment. We review the design of the atomic beam unit, including the vacuum system and the atomic beam source and collimation. Then, we discuss the optical setup, with particular emphasis on our new method of locking the laser that drives the $5P_{1/2} \rightarrow 6S_{1/2}$ transition and the resulting necessary modifications to our vapor cell optical setup. This chapter also includes a brief discussion of our method for collecting data. The next chapter discusses our data analysis methods and includes a summary of our results. Finally, we describe possible future experiments to build on our current one.
Chapter 2

Atomic Theory

2.1 Absorption Spectroscopy Theory

Our entire experiment rests on the principles of how atoms and light interact, so it is vital that we have a thorough understanding of the relevant theory. We first consider the most basic case of spectroscopy—direct absorption. From there, we build up to discuss the more complicated FM spectroscopy techniques we use in our experiment.

2.1.1 Direct Absorption Spectroscopy

The basic configuration of our experiment entails sending two laser beams (one at 410 nm and one at 1343 nm) through a vapor of indium. We scan the frequency of the IR laser across the atomic transition, and expect to see the intensity of the transmitted light change as a function of the frequency, \( \omega \). We can express the change in intensity of the light due to passing through a small sample of atoms of width \( dz \) as:

\[
dI(\omega) = -A(\omega)dz
\]

where \( A(\omega) \) is a function that describes the absorption as a function of wavelength. Integrating this equation gives

\[
I(\omega, z) = I_0 e^{-A(\omega)z}
\]

for a laser with intensity upon entering the sample of \( I_0 \). We can write \( A(\omega)z \) into \( \alpha P(\omega) \), where \( \alpha \) is the optical depth of the atomic sample and \( P(\omega) \) is the normalized transition probability. As will be discussed more later, we observe indium in two settings—one in a vapor cell, with a relatively large optical depth of about 1, and the other in the much less dense atomic beam, where we have \( \alpha \sim 10^{-3} \).

2.1.2 Line Broadening

Depending on the experimental conditions, there can be a significant probability of transition over a relatively wide range of frequencies. Characterizing the width of
an absorption peak is important in understanding our ability to resolve different spectral features. If the separation between two absorption peaks is roughly the same as their linewidths, it will be difficult, if not impossible, to resolve them individually. Additionally, the quality of our locking signal depend on its width, so it is useful for this reason as well.

While there are many types of mechanisms that can cause a spectral peak to widen, the three with which we are most concerned for this experiment are the natural linewidth, Doppler broadening, and power broadening. The details of the calculations of the following effects are thoroughly described in [12], but they have important implications for our experiment, so we summarize them here.

Broadly speaking, broadening mechanisms can be classified as either “homogeneous” or “inhomogeneous.” Homogeneous broadening affects all atoms of the same element equally regardless of an individual atom’s behavior. The natural linewidth, or lifetime broadening, is an example of homogeneous broadening; all indium atoms will experience the same effect. Even in a completely perfect experiment, the absorption peaks observed in an atomic spectrum would not be infinitely narrow. Excited atomic states have finite lifetimes; thus, by the Heisenberg uncertainty principle, the energies (or equivalently, frequencies) of the transitions must be distributed over some finite range:

\[
\Delta E \Delta t \gtrsim \frac{\hbar}{\Delta f} \gtrsim \frac{1}{2\pi \tau} \tag{2.3}
\]

where \(\Delta f\) is the minimum half width of the peak in frequency and \(\tau\) is the lifetime of the state. Specifically, the lifetime of the \(6S_{1/2}\) state of indium is approximately 7 ns and the lifetime of the \(6P_{1/2}\) is approximately 55 ns [13].

Inhomogeneous broadening, on the other hand, depends on the physical state of an atom. Doppler broadening depends on the velocity of the atoms, so it is an inhomogeneous broadening mechanism. Especially for our vapor cell measurements, Doppler broadening turns out to be the largest effect. The atoms in the vapor cell are not constrained to move in any particular direction. Therefore, their velocities are distributed according to the Maxwell-Boltzmann law. Our experimental parameters (a temperature of 730 °C and a frequency of 730673 GHz) yield a linewidth in the resulting Gaussian peak of about 1.5 GHz. This broadening is by far the largest effect in the vapor cell for the first step transition.

However, we are primarily interested in the second-step transition. Predicting the width of the peaks in a two-step transition spectrum is more complicated, but we can at least get a sense of the relevant effects. Considering the separation between the hyperfine levels of the indium \(6P_{1/2}\) state is roughly 1.2 GHz, it would reasonable to be concerned that we would not be able to resolve these peaks. Having a two-step transition actually solves this problem. If we hold the 410 nm laser at one specific frequency, only atoms which are moving with a velocity such that they “see” the laser
as having a frequency within their natural linewidth will be excited to the intermediate \(6S_{1/2}\) state. Therefore, only atoms velocities in that range will be available for the other laser to excite to the \(6P_{1/2}\) state, thus nearly eliminating Doppler broadening.

The geometry of the atomic beam serves to significantly reduce the spread of velocities of the atoms and thus the broadening of the spectral line, even in a single-step transition. As will be described in more detail in Chapter 3, we collimate the beam so all of the atoms are moving in approximately the same direction. We then intersect the lasers perpendicularly to the beam, so nearly all of the atoms are moving with a direction transverse to that of the lasers. They thus experience very little Doppler effect. This setup results in much less broadening—instead of the \(\sim 1.5\) GHz Gaussian width in the vapor cell, the width of the first transition due to Doppler broadening is only about 100 MHz. The two-step excitation again serves to reduce that even further.

The other primary source of broadening in our experiment is power broadening. When the laser power is strong enough, it begins to drive stimulated emission instead of just exciting atoms. This effectively reduces the lifetime of the atom, and thus increases the width of the line. The increased linewidth is given by:

\[
\frac{\Gamma}{\Gamma_0} = \sqrt{1 + \frac{I}{I_{sat}}} \tag{2.5}
\]

where \(I_{sat} = \frac{\pi \hbar c}{3 \lambda^3 \tau}\) for a state with lifetime \(\tau\), and \(\Gamma_0\) and \(\Gamma\) are the linewidths with and without the power broadening respectively. Due to a change in the experimental configuration, the power broadening in the atomic beam has been significantly increased from the previous thesis. The values are now about 20 MHz in the vapor cell and 80 MHz in the atomic beam.

While there are several other types of broadening that occur, these are all at least an order of magnitude smaller than the three discussed above, and therefore, are less noteworthy. For more in depth derivations of lineshapes or calculations of linewidths, see [12].

Once we have considered the individual sources of broadening, it is important to understand how we combine them into a total predicted linewidth. Some of these broadening mechanisms, such as Doppler broadening, result in a Gaussian shaped peak, whereas others, such as the natural linewidth, form Lorentzians. The rigorous method for computing the total fits the spectrum to a Voigt profile, which is the convolution of a Gaussian and a Lorentzian. In practice, this is computationally intensive. Also, in both the atomic beam and the vapor cell, our second-step transition signals are well approximated by Lorentzians due to the lack of Doppler broadening, so we typically fit just to Lorentzians\(^1\). Here, however, we list the Voigt linewidths for the \(5P_{1/2} \rightarrow 6S_{1/2}\) transition given our experimental conditions.

\(^1\)We have previously compared results from Lorentzian and Voigt fits, and found any differences to be negligible.
2.2 FM Spectroscopy Lineshapes

We rely on FM spectroscopy throughout our experiment, so this topic is especially important for understanding our data. As will be discussed more extensively later, single-tone FM spectroscopy is useful for locking the 410 nm laser. Two-tone FM spectroscopy is also crucial for our measurements; it allows us to observe the very small atomic beam signal. In this section, we consider the lineshapes relevant to this experiment. However, there are many possible ways to execute FM spectroscopy, and different parameters result in a variety of lineshapes. An in-depth discussion of single-tone FM spectroscopy is included in [12]. The Majumder lab has used both single-tone and two-tone FM spectroscopy in multiple previous experiments (single-tone in [12, 14] and two-tone in [15]). However, the current measurement relies on both techniques, and we therefore discuss both here.

2.2.1 Single-Tone FM Spectroscopy

The basic idea of single tone FM spectroscopy consists of modulating the laser beam at some radio frequency, and then detecting the light and demodulating at that same frequency. We send the light through an EOM, which modulates the light at some frequency $\omega_m$. After the laser passes through the atoms, we detect the light and demodulate it at $\omega_m$ using a lock-in amplifier. The output of the lock-in will be the intensity, which is proportional to the electric field of the laser squared. The modulated electric field can be written as

$$E(t) = E_0 e^{i(\omega_c t + \beta \sin(\omega_m t))}$$ (2.6)

where $\beta$ is the “modulation depth”—the strength of the modulation. We detect the time averaged intensity $\langle E^*(t)E(t) \rangle$. Finally, the lock-in amplifier demodulation leaves us with a signal of the form:

$$I(f) \propto T^2(\omega_c + \omega_m) - T^2(\omega_c - \omega_m)$$ (2.7)
T(\omega) is a function describing the absorption as a function of frequency, \( \frac{\omega_c}{2\pi} \) is the main frequency of the laser, and \( \frac{\omega_m}{2\pi} \) is the frequency at which we modulate the laser (this happens to be 100 MHz for our experiment). The details of this derivation are given in [12].

We are primarily interested in the signal observed in the atomic beam, where even the \( 5P_{1/2} \rightarrow 6S_{1/2} \) is fairly narrow. In fact, we modulate at approximately the same frequency as the atomic beam linewidth. A simulated curve for this signal is shown in Figure 2.1 above.

For about \( \pm 100 \) MHz around the central resonance frequency, this signal is approximately linear. This fact ends up being crucial for our locking scheme for our laser for the \( 5P_{1/2} \rightarrow 6S_{1/2} \) transition. (See Section 3.2.2 for more information).

\subsection{2.2.2 Two-Tone FM Spectroscopy}

The basic concept of two-tone FM spectroscopy is fairly similar to that of single-tone FM spectroscopy— we still add frequency sidebands to the spectrum of the laser and then detect the signal with a lock-in amplifier to demodulate it. However, now we simultaneously modulate the light at two frequencies, \( \omega_m \) and \( \Omega \) where \( \Omega \ll \omega_m \). Furthermore, for an atomic linewidth \( \Gamma \), we assume \( \Omega \ll \Gamma \) but \( \omega_m > \Gamma \). High frequency modulations (\( \frac{\omega_m}{2\pi} = 600 \) MHz for our experiment) are useful for reducing low-frequency noise, which would be the predominant source of noise. The necessary high-frequency electronics for processing and detecting the signal (specifically photodetectors and lock-in amplifiers) are much more expensive and less common than similar devices for lower frequencies, however. By also modulating and then
demodulating at a much lower frequency, $\Omega \sim 100 \text{ kHz}$, we gain the advantages of high-frequency modulation while being able to use photodetectors and lock-in amplifiers that only need to detect oscillations at the lower frequency. Additionally, it is at least as effective at reducing noise as single-tone FM spectroscopy [16]. Again, signal-to-noise ratio improvement is extremely important for our experiment, since the absorption in the atomic beam due to the $6S_{1/2} \rightarrow 6P_{1/2}$ transition is small enough that it is not detectable at all with direct absorption spectroscopy.

We now turn to our expected lineshape. Instead of our laser having frequency sidebands at $\frac{1}{2\pi}(\omega_c \pm \omega_m)$, it technically has four sidebands at $\frac{1}{2\pi}(\omega_c \pm (\omega_m \pm \frac{\Omega}{2}))$. We only observe two in the vapor cell and Fabry-Pérot, however, because our linewidth is much greater than $\frac{\Omega}{4\pi}$. (In the atomic beam, we would expect only to see two sidebands, since we demodulate the signal at $\Omega$). The derivation is similar to that for the single-tone lineshape, but even more algebraically intensive, so again, we’ll just state the resulting observed intensity:

$$I(\omega) \propto 2T^2(\omega_c) - T^2(\omega_c - \omega_m) - T^2(\omega_c + \omega_m)$$

Now, instead of having peaks at two frequencies equally spaced from the central frequency, we have three peaks—one at the central frequency and one on either side of it. We simulate a sample signal in Figure 2.2a. However, we are observing a transition to two different hyperfine states, so we observe a slightly more complicated signal. Instead of having one set of three peaks, we actually have two, for a total of 6 peaks. However, since the hyperfine splitting is roughly 1250 MHz and our sidebands are separated by 600 MHz due to the EOM that we have, the central two sidebands overlap. A simulated spectrum with all 6 peaks plotted is shown in Figure 2.2b.

Even without demodulation, the two-tone FM signal also is useful for calibration, as the absorption spectrum in the non-demodulated signal in the vapor cell will have sidebands with precisely known spacings. This signal is simply a sum of
Lorentzians (based on the previous discussion of line broadening). For more detail, see Section 3.2.3.

As will be discussed more in Chapter 4, knowing the predicted lineshapes allows us to fit the observed signals to make the desired measurements.

2.3 Stark Shift

The goal of the experiment is to measure the Stark shift in the indium $6P_{1/2}$ state. The Stark shift relates directly to the polarizability of the atom, which provides useful information about indium’s atomic properties. We now consider the theoretical derivation of the shift.

In our calculation, we wish to use perturbation theory, so we first check to make sure the assumption that any external electric field will be small compared to the internal electric field from the nucleus on the electron is valid. Indium has an atomic radius of 167 pm, so a quick calculation gives us that the electric field on the electron from the nucleus is approximately $5 \times 10^5$ kV/cm. On the other hand, we never apply an electric field of more than 15 kV/cm. Furthermore, given the fairly large hyperfine splitting, we can use nondegenerate perturbation theory.

First, we’ll discuss the simpler effect of the Stark shift in the excited energy level of a one step transition. This case is representative of the Stark shift in the $6S_{1/2}$ state (observed via the $5P_{1/2} \rightarrow 6S_{1/2}$ transition). Although the measurement of this shift is not our ultimate goal, it ends up still having a significant role in our experiment. First, any Stark shift measurement can only be performed as the measurement of the difference between the shifts of two different states; for example, we will actually measure the difference between the polarizability in the $6P_{1/2}$ state and that in the $6S_{1/2}$ state:

$$\Delta f_{\text{measured}} = k_S \mathcal{E}^2$$

$$= -\frac{1}{2\hbar} (\alpha_0(6P_{1/2}) - \alpha_0(6S_{1/2})) \mathcal{E}^2$$  \hspace{1cm} (2.9)

Using the known shift of the $6S_{1/2}$ state, we can then compute the absolute shift in the $6P_{1/2}$ state. Also, the Stark shift in the intermediate $6S_{1/2}$ state turns out to be an important consideration for our laser locking scheme, as will be discussed in more depth later.

We can write the perturbation from the applied electric field as

$$H' = e\mathcal{E} z$$  \hspace{1cm} (2.11)

where $e$ is the charge of an electron, $\mathcal{E}$ is the applied electric field, and we take the $z$ direction to be the direction of our electric field.

We now show that there is no linear Stark shift present. In general the first order perturbation in the energy, with some perturbation in the Hamiltonian $H'$, is:

$$\Delta E_n^{(1)} = \langle \psi_n^{(0)} | H' | \psi_n^{(0)} \rangle$$ \hspace{1cm} (2.12)
CHAPTER 2. ATOMIC THEORY

Substituting in Equation 2.11, we get

\[
\Delta E_n^{(1)} = \langle \psi_n^{(0)} | e\mathcal{E}z | \psi_n^{(0)} \rangle \tag{2.13}
\]

\[
= e\mathcal{E} \int_{-\infty}^{\infty} |\psi_n^{(0)}|^2 z dz \tag{2.14}
\]

\[
= 0 \tag{2.15}
\]

by parity. Thus, there is no first order Stark shift, as expected. We continue on to consider a second order effect (which does turn out to be nonzero). Second order perturbation theory gives us

\[
\Delta E_n^{(2)} = \sum_{m \neq n} \frac{| \langle \psi_m^{(0)} | e\mathcal{E}z | \psi_n^{(0)} \rangle |^2}{E_m^{(0)} - E_n^{(0)}} \tag{2.16}
\]

\[
\equiv -\frac{1}{2} \alpha_0(n) \mathcal{E}^2 \tag{2.17}
\]

\(\alpha_0(n)\) is the polarizability, which is what we ultimately wish to measure. By comparing the two equations above, it becomes apparent that the polarizability is intimately related to the behavior of the atom (specifically the wavefunctions). We can also see how transitions between higher levels, which are closer together in energy, will have larger shifts. For example, we know that in indium, the relative polarizabilities of the \(5P_{1/2}\), \(6S_{1/2}\), and \(6P_{1/2}\) states have a ratio of approximately 1:10:70 respectively (see Figure 2.3).

### 2.3.1 Stark Shift Measurements in Three-Level Systems

It turns out that, under certain conditions, understanding the effect of the electric field on indium can be complicated by the fact that we are studying a two-step transition. Over the past year, we have modified the experiment to account for these difficulties, and therefore the experimental results are essentially independent of these considerations. Nonetheless, this theory has motivated many of the new developments in this experiment, so it is certainly worth discussing. For a more complete discussion and relevant derivations, see [16].

Assume we hold the laser for the first transition at exactly the non-shifted resonance frequency for the \(5P_{1/2} \rightarrow 6S_{1/2}\) transition always, regardless of whether or not we turn on the electric field. When the electric field is off, then the 410 nm laser is of course resonant and as we scan the other laser across the second transition, we see the expected signal. However, when the electric field is turned on, the 410 nm laser is no longer exactly on resonance; instead, its frequency is too large by \(\delta_2\), the Stark shift in the \(6S_{1/2}\) state. (See Figure 2.4). This detuning is fairly small (\(\lesssim 25\) MHz) compared to the range over which we scan the laser for the second-step transition (\(\sim 5\) GHz), so observing a signal will still not be an issue, as the system can compensate by picking out a nonzero velocity class.
We cannot assume, however, that we can treat the two transitions as independent two level systems. Instead, we must consider all three levels together as a coupled system, as shown in Figure 2.4. By analyzing this system using theory from quantum optics, we can gain some understanding of what will happen when the blue laser is off resonance. Again, a mathematical derivation and numerical simulations can be found in [16], and we just summarize the results here.

Qualitatively, the system increasingly becomes more accurately described as a two-photon transition from the $5P_{1/2}$ state to the $6P_{1/2}$ state, and the $6S_{1/2}$ state becomes less directly involved. The observed Stark shift would not be $\delta_3 - \delta_2$, but rather would approach $\delta_3 - \delta_1$. This fact alone does not pose a problem; we know the polarizabilities of both the $5P_{1/2}$ and $6S_{1/2}$ states sufficiently precisely from previous experiments that using either one to calculate the $6P_{1/2}$ polarizability would not introduce significantly more uncertainty in the measurement.

The problem comes from knowing how close we are to the regime where we would be measuring $\delta_3 - \delta_1$ rather than $\delta_3 - \delta_2$. The fractional difference of our measurement from $\delta_3 - \delta_1$ depends on the ratio of the Rabi frequencies ($\Omega_1$ and $\Omega_2$) of the transitions. We can estimate the Rabi frequencies, but we cannot measure them, and we certainly cannot know them with sufficient precision for our goal. Specifically, the dependence of our measurement on the $\frac{\Omega_2}{\Omega_1}$ can vary up to 4-5% over the range of electric fields that
CHAPTER 2. ATOMIC THEORY

Figure 2.4: Energy level diagram for a three level system, adapted from [16] for the specific case of the transition in indium as an example. $\delta_j$ represents the Stark shift in each energy level (where $j = 1, 2, 3$ for the $5P_{1/2}$, $6S_{1/2}$, and $6P_{1/2}$ states respectively). $\Delta_j$ indicates some detuning from the non-shifted state. Finally, $\Omega_1$ and $\Omega_2$ are the Rabi frequencies for the $5P_{1/2} \rightarrow 6S_{1/2}$ and $6S_{1/2} \rightarrow 6P_{1/2}$ transitions respectively.

we apply. Since we are aiming for $\leq 1\%$ precision, this uncertainty is unacceptably large.

2.3.2 Two-Step Transition with the Laser for the First Step Always on Resonance

We have now modified the experiment so that the 410 nm laser is always on resonance, and the dependence of the measured Stark shift on the Rabi frequency vanishes (see Section 3.2.2 for experimental details). In addition to eliminating the uncertainty due to the dependence on the Rabi frequency, this modification also permits calculation of the analytical lineshape. The derivation of the lineshape is in [16]; letting $x = \Delta_2 - (\delta_3 - \delta_2)$, the result is:

$$\langle P_3(x) \rangle = \frac{2\Omega_1^2\Omega_2^2(x^2 + 3(\Omega_1^2 + \Omega_2^2))}{4x^4\Omega_1^2 + x^2(-8\Omega_1^4 + 20\Omega_1^2\Omega_2^2 + \Omega_2^4) + 4(\Omega_1^2 + \Omega_2^2)^3} \quad (2.18)$$

Since we do not know the Rabi frequencies, we cannot necessarily predict what this shape will look like. However, as shown in Figure 2.5, it is always centered around $\delta_3 - \delta_2$ since $\langle P_3(x) \rangle$ is even about 0, so we know that we are measuring that quantity. Furthermore, we perform the experiment for a wide range of $\frac{\Omega_1}{\Omega_2}$ anyway, so if there were any systematic effects due to the relative Rabi frequencies, they would show up
in our data. We observe no correlation between this ratio and our result, so it appears that there is in fact no dependence (see Section 4.2 for more information).

Three Level Doppler Broadened Systems

As a result of our new locking setup, the blue laser is no longer at the center of the resonance of the transition in the vapor cell by default. We account for this shift in our experimental setup by shifting the frequency of the laser back to the resonance frequency using a pair of AOMs (see Section 3.2.2). In the atomic beam, we know that if we are detuned even slightly from the first-step transition, the behavior of the second-step transition becomes difficult to predict and analyze. In the vapor cell, however, the first step transition is much broader due to Doppler broadening, and the previous simulations that revealed Rabi frequency dependence were based on the assumption of a narrow first-step transition. Thus, we wish to know whether the resonance of the second-step transition will similarly depend on the Rabi frequency, or if the Doppler broadening will mitigate the impact of this effect. We simulate the second-step transition probability as a function of frequency of the IR laser, assuming that the 410 nm laser is slightly detuned. A plot of the results of this simulation for a variety of Rabi frequencies is shown in Figure 2.6.

In the plots, it does appear that any effect from the detuning of the laser, other than the expected overall shift in the peak, will be very small, although the amplitude could vary. Furthermore, we can calculate the location of the maximum value to ensure that its location is in fact independent of the Rabi frequencies. For a reasonable

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figures/lineshape.png}
\caption{Plot of the analytic lineshape for various pairs of Rabi frequencies. For our experiment, $\Omega_1 < \Omega_2$. Note that it is symmetric about $x = 0$ for all of the pairs of Rabi frequencies.}
\end{figure}
Figure 2.6: Plot of the population in the vapor cell of the $6P_{1/2}$ versus frequency difference from resonance, $f - f_0$, when the 410 nm laser is detuned by 10 MHz below resonance. Here we show several representative ratios of Rabi frequencies given our experimental conditions. Note that the linewidth for the first step transition in the vapor cell is about 1.5 GHz.

range of Rabi frequencies, we observe that any differences between the locations of the peak are on the order of .01 MHz, certainly smaller than we need to be concerned with. Nonetheless, our experimental setup does account for this possible detuning and correct for it (see Section 3.2.2).
Chapter 3

Experimental Setup

Fundamentally, our experiment uses two-step laser spectroscopy to observe the structure of the $6P_{1/2}$ state in indium with and without an electric field. The experiment thus requires two lasers as well as regions where the lasers can interact with indium. There are two components of our experiment where the lasers and atoms interact—a vapor cell used for reference, and an atomic beam used for frequency stabilization of the laser for the first step transition and for observation of the second step transition in the presence of an electric field. This chapter discusses both of these components of the experiment as well as the preparation of the lasers and our techniques for observing and recording the relevant signals.

3.1 Atomic Beam Unit

Although vapor cells in tabletop ovens are simple to use and easy to work with (and remain relevant for this experiment), we cannot apply the necessary voltages for Stark shift measurements due to the possibility of electric breakdown. Additionally, vapor cells would require applying the electric field through glass, which could cause problematic nonuniformities in the field. Therefore, our experiment must be performed in high vacuum, and we choose to use an atomic beam instead of a vapor cell. Our vacuum chamber also contains an atomic beam, which we use to observe the relevant transition. Figure 3.2 shows a schematic of the vacuum chamber and atomic beam system. For more detailed information, see [12, 14].

3.1.1 Vacuum Chamber

Our vacuum system consists of a roughing pump, used to bring the pressure to about $10^{-3}$ torr, and then two diffusion pumps with associated liquid nitrogen traps. This system typically achieves pressures below $10^{-7}$ torr at room temperature, which increases only slightly to just above $10^{-7}$ when we heat the source oven (discussed below) to about 1150 °C. We monitor the pressure using an ionization gauge.
Figure 3.1: Diagram of the setup of our experiment, including both the vapor cell and atomic beam unit, as well as relevant laser preparation and signal processing equipment.
3.1.2 Atomic Source and Beam

To provide the necessary atoms, we put about 100 g of indium in a molybdenum crucible (melting point \( \sim 2625 \) °C), which we heat to about 1150 °C using an oven made by previous students. The face of the crucible has 20 small slits through which indium atoms can effuse. At this temperature, all of the indium will have effused out of the crucible after about 100 hours of operation. The slits in the crucible serve as the initial collimation for the atomic beam. About 20 cm away from the electric field plates, we have a set of four razor blades for further collimation. We can adjust the positions of the blades to shape the beam further. For final collimation, the atoms pass through a rectangular slit with similar dimensions to the beam. This final step also serves to prevent much indium from hitting the electric field plates. This slit also is conveniently placed in the mount for the atomic beam chopper wheel, which, as will be discussed later, is used for both locking the 410 nm laser and observing the two-step transition. After this collimation process, we are left with a beam with an optical depth of roughly \( 10^{-3} \).

Despite the additional complexities of this system when compared with the vapor cell setup, there are advantages as well. In particular, since we intersect the lasers with the beam perpendicular to the atoms' path, we greatly reduce Doppler broadening (we obtain line widths in the beam of order 100 MHz as compared to 1.5-2 GHz in the vapor cell for the first step transition).

3.1.3 Interaction Region

When we intersect the lasers with the atoms, they must be very precisely overlapped due to the short lifetime of the \( 6S_{1/2} \) state. Otherwise, the atoms that we excite to this state will decay back down to the \( 5P_{1/2} \) state before they can interact with the IR laser. Therefore, if the lasers are not overlapped, we will not observe a transition to the \( 6P_{1/2} \) state.

We intersect both lasers with the atomic beam as it passes through the space between two capacitor plates as shown in Figure 3.3. With these plates, we can apply potentials of up to 20 kV. To know the electric field, we must know both the voltage applied and the distance between the plates. To measure the voltage, we use a high voltage divider (Ross Engineering) which divides the voltage by a factor of 1000. We then measure the voltage using a precision microvolt voltmeter (Keithley 197A), as described in [12]. The separation of the plates is 1.0009(5) cm. For more information about the calibration of the plate separation, see [16]. Simulations by previous students have shown that, in the region where the lasers and atoms interact, the field is sufficiently close to uniform that the assumption of uniformity will not significantly impact our results. Thus, we can take the field to be that of an infinite parallel plate capacitor,

\[
E = \frac{V}{d}
\]
From propagation of error, we then have the uncertainty in the electric field:

$$\delta E = \sqrt{\delta V^2 + \delta d^2} \quad (3.2)$$

where $\delta$ indicates the fractional error. The fractional error in the voltages we apply is an order of magnitude smaller than the uncertainty in the plate separation, so the fractional error in the electric field is approximately the fractional error in the plate separation. Since the Stark shift is proportional to $E^2$, the error we include in our systematic error calculations is $2\delta E$.

### 3.2 Optical System and Signal Processing

#### 3.2.1 Lasers

We require one laser for each step of the two-step transition. Both of these lasers are external cavity diode lasers. One laser (Toptica DL 100) is tuned to the $5P_{1/2} \rightarrow 6S_{1/2}$ transition at 410 nm, while the other (Sacher Lynx series) is tuned to 1343 nm to excite the $6S_{1/2} \rightarrow 6P_{1/2}$ transition. Both lasers are in the Littrow configuration, in which the light emitted from the diode is passed through a lens onto a diffraction
3.2. OPTICAL SYSTEM AND SIGNAL PROCESSING

3.2.1 Electric Field Plates and Atomic Beam

Laser beams intersect with the atomic beam between the electric field plates. (From [12]).

Figure 3.3: Schematic of the intersection of the overlapped lasers with the atomic beam between the electric field plates. (From [12]).

While the light emitted from the diode contains many frequencies, light of different frequencies will diffract at different angles. By allowing one frequency of light to reflect directly back to the diode, this setup causes lasing to occur at that specific frequency. Through adjustments of the angle of the diffraction grating, we can then tune the laser to the appropriate frequency. This tuning can be accomplished with manual adjustments of the cavity, but both lasers also contain a piezoelectric transducer (PZT), which expands or contracts based on an applied voltage. This change in size allows for fine adjustments of the cavity alignment. The 410 nm laser typically gives about 12 mW of light, and the 1343 nm laser typically emits about 40 mW of light. More information about the lasers is available in [12, 17].

Without further intervention, however, the frequencies of the lasers tend to drift over time. For the 1343 nm laser, as long as these drifts occur at larger time scales than one scan over the transition, small drifts in the laser frequency are acceptable as we will continue to scan over the resonance of the transition. On the other hand, the 410 nm laser needs to be held on resonance at all times, and the drifts, which are typically on the order of 100 MHz over timescales of minutes are thus unacceptable. Therefore, we must stabilize the frequency of this laser.

3.2.2 Locking the 410 nm Laser

Frequency stabilization of this laser is actually more complicated than just maintaining it at one particular frequency. While our lab has developed techniques for locking lasers to atomic transitions, this experiment has the additional complication of an atomic transition that changes frequency due to the Stark shift. Previous iterations of this experiment had locked to the $5P_{1/2} \rightarrow 6S_{1/2}$ transition in a vapor cell. While the Stark shifted frequencies (approximately 10-50 MHz from the non-
Figure 3.4: Sample locking signal (derivative of the absorption signal) with a Fabry-Pérot signal shown below as a frequency reference. Sidebands on the Fabry-Pérot peaks are due to the EOM modulation.

Shifted frequency) are close enough to the central frequency of this transition that we can still observe the transition with a laser locked to the non-shifted frequency; numerical simulations (see [16] for more information) reveal that using a laser that is even slightly off resonance when the transition is shifted leads to unacceptably large systematic of about 4-5% associated with the three-level dynamics discussed in Chapter 2. Therefore, we presently lock the laser to the atomic beam transition so that the frequency is locked to the resonance of the first-step transition regardless of whether or not the high voltage is on or off.

Our locking scheme relies on a PID controller, which essentially establishes a negative feedback loop with the laser when given a signal that indicates displacements from the correct frequency. When the PID controller detects shifts from the desired frequency, it applies a voltage to the laser’s PZT to tune the laser back to the correct frequency.
3.2. OPTICAL SYSTEM AND SIGNAL PROCESSING

This setup requires, of course, a signal that contains the necessary information about frequency drifts. Since the atomic transition still determines the frequency to which we would like to lock the laser, intuitively, the atoms themselves constitute the ideal system to use as a frequency reference. However, if one simply scans the laser across the transition and obtains the absorption curve, the fact that the resulting signal is even about the atomic resonance means we cannot directly use it as the input signal for the PID controller. Consider a small change from the resonance frequency in the laser. Regardless of whether the frequency has increased or decreased, the absorption will be less, and the PID controller will be unable to determine in which direction the frequency has shifted. However, the derivative of the absorption signal would in fact be able to indicate in which direction the frequency had drifted. With this signal, the PID controller can then output a voltage, which is applied to the laser’s PZT. Such a signal is shown in Figure 3.4, along with a Fabry-Pérot signal, which is used to provide a frequency scale to determine locking precision.

To obtain this signal, we use single-tone FM spectroscopy, which essentially entails passing the laser through an EOM (ThorLabs EO-PM-NR-C4) before it interacts with the atoms and demodulating the resulting signal with a lock-in amplifier. The EOM adds frequency sidebands to the laser (in this case, separated from the laser’s central frequency of 100 MHz). We detect the signal using a photodetector that is capable of resolving 100 MHz oscillations (New Focus model 1601). We use this frequency as the reference for a RF lock-in amplifier. The resulting demodulated signal is effectively the derivative of the absorption signal, as desired.
Previously, we had locked the laser to the vapor cell. We have now changed to using the blue laser beam that we send through the atomic beam to lock. Since it interacts with atoms between the electric field plates, when we turn on the high voltage, the locking signal will shift.

The significantly decreased optical depth in the atomic beam as compared with the vapor cell does create an additional experimental challenge, as our locking signal would be much weaker if we just observed it directly. We have been able to solve this problem, however, by inserting a chopper wheel into the atomic beam. The transition can only occur, of course, when the atoms are not blocked by the chopper wheel. On the chopper wheel mount, we place a photointerrupter, through which the chopper wheel passes as it spins. This setup allows us to monitor the frequency at which the atoms are blocked and unblocked (typically about 500 Hz), which we use as the reference frequency for a second lock-in amplifier. This sequence of two lock-in amplifiers allows us to obtain a signal with a high signal-to-noise ratio and reliably zero background with no observable drift or offset.

By simultaneously recording the demodulated absorption signal and the signal from a Fabry-Pérot cavity, (which we know has a free spectral range of 1 GHz), we can investigate features of our locking signal. The quality of the lock depends on the slope of the linear portion of our locking signal and the signal-to-noise ratio. One benefit of our new locking scheme is that the slope of the signal from the atomic beam is actually much larger than that from the vapor cell due to the lack of Doppler broadening. Specifically, the frequency separation of the maximum and minimum in the vapor cell is over 1 GHz, while in the atomic beam it is approximately only 200
MHz, but the signals both have similar amplitudes. Smaller fluctuations in frequency thus should be more easily detected and corrected. Once we know this slope, we can use this to relate the root mean squared amplitude of any remaining noise to a corresponding frequency. While the slope is steeper, the locking signal is somewhat noisier than the old one, so the small residual fluctuations in our lock remain at about 1 MHz or less when the laser is locked. This stability is more than adequate for our subsequent spectroscopy and Stark shift measurements.

One further consideration is whether or not the lock point could be shifting. Any long term drift on the order of tens of MHz would be of serious concern to us, given the importance of staying on resonance. We used multiple tests to ensure that the location of the signal only displayed fluctuations in frequency and not any long term drift in one direction. In addition to recording the Fabry-Pérot signal and the locking signal simultaneously, we also recorded the signal from a saturated absorption spectroscopy setup using the vapor cell. This gave us a much narrower absorption peak than the typical Doppler-broadened curve observed in the vapor cell, which we could use as a frequency reference. By comparing the point where the locking curve crossed through zero with both one peak from the Fabry-Pérot signal and the peak of the saturated absorption signal from the vapor cell, we showed that any frequency fluctuations in our signal do not display a trend over time.

Finally, this locking scheme would not be an improvement over the former scheme if the lock did not shift appropriately when an electric field was applied. Observations of the correction signal from the PID controller showed that the lock did, in fact, shift as expected with the application of an electric field, as shown in Figure 3.6. Specifically, when we doubled the voltage, the correction signal increased by a factor of four, corresponding to the quadratic Stark shift.

![Diagram of the setup of the AOMs for shifting the 410 nm laser back to the non-Stark shifted frequency before the vapor cell.](image)

**Figure 3.7:** Diagram of the setup of the AOMs for shifting the 410 nm laser back to the non-Stark shifted frequency before the vapor cell.
CHAPTER 3. EXPERIMENTAL SETUP

Frequency Compensation of the 410 nm Laser in the Vapor Cell

While our new locking scheme solves the problem of the laser being off resonance with the atomic beam when the electric field is applied, this means that the frequency will be slightly off-resonance in the field-free vapor cell. Moreover, the size of this detuning will depend on the electric field. The first-step transition will then pick out a non-zero velocity class of atoms (with respect to the laser), and we will thus observe a Doppler shifted signal in the second-step transition as well. Since the size of this shift will depend on the electric field, this detuning could introduce systematic errors if unaddressed. We therefore wish to maintain the frequency of the blue laser at the field-free resonance when it passes through the vapor cell.

To accomplish this goal, we use two AOMs (Isomet model 1250C-829A). The AOMs act like diffraction gratings in that they split the incoming beam into multiple beams at different angles at the output, but since we drive them with RF waves, they also induce frequency shifts. Three of the resulting beams—the zeroth order beam (at the original laser frequency $\omega_c$) and the two first order beams, one with a frequency $\omega_c + \omega_m$, the +1 order beam, and one with a frequency $\omega_c - \omega_m$, the -1 order beam, where $\omega_m$ is the modulation frequency of the AOM, have much greater intensity than the others, so we focus on those three. In our case, the central modulation frequency of the AOM is approximately 250 MHz. We must use a frequency close to this central modulation frequency, but small changes from it are possible.

In our setup, we select the +1 order beam from the first AOM, and the -1 order beam from the second AOM, as shown in Figure 3.7. When the transition in the atomic beam has a Stark shift of $\Delta f$, we drive the first AOM at $\omega_m + \frac{\Delta f}{2}$ and the second at $\omega_m - \frac{\Delta f}{2}$, where $\omega_m$ is now the central modulation frequency of the AOM. Typical values of $\Delta f$ are about 5-15 MHz. The final frequency of the laser beam after it passes through both AOMs is therefore $\omega_c + \omega_m + \frac{\Delta f}{2} - (\omega_m - \frac{\Delta f}{2}) = \omega_c + \Delta f$ as desired (since the Stark shift will cause the lock point to be lower in frequency than the non-shifted transition frequency).

Since our group precisely measured the Stark shift in the $5P_{1/2} \rightarrow 6S_{1/2}$ transition previously, we know $\Delta f$ with sufficient precision to introduce no further uncertainty into our measurement by using this setup. Also, since the adjustments in frequency to account for the Stark shift in each AOM are always opposite each other, this setup conveniently ensures that there will be no net change in the alignment of the laser beam when the driving frequencies of the AOMs are changed.

3.2.3 Vapor Cell Reference

Given the difficulty of detecting the two-step transition signal in the atomic beam, it is useful also to have a vapor cell to monitor the signal. Here, we heat a cell of indium to 730 °C, which gives an optical depth for the first step transition of about 1. While the second step transition does not yield as strong of a signal, we can still observe it in a fairly straightforward manner. By sending the 410 nm laser beam through a chopper wheel rotating at about 1.4 kHz, which we use as a reference for a lock-in
amplifier, we are able to pick out only the signal from the atomic transition. We then can easily observe the hyperfine structure of the 6P_1/2 state with a very high signal to noise ratio. This signal serves to confirm that both of the lasers are behaving as expected, and when we modulate the IR laser using an EOM, we can use the precisely known spacing of the resulting sidebands as a calibration for the frequency axis in our final signal.

Also, in looking at Figure 3.9, one might notice the small peaks on the right of the larger peaks. We believe these are due to the other isotope of indium, ^{113}\text{In} (which has an abundance of about 4%), which would have slightly shifted transition frequencies from ^{115}\text{In}. See Chapter 4 for a discussion of how we treat these peaks when we analyze our data.

### 3.2.4 IR Laser Preparation and Signal Observation

Throughout the experiment, and as we prepare to collect data, we always monitor the IR laser using a Fabry-Pérot interferometer with a free spectral range of 500 MHz. This allows us to ensure the laser remains in single mode as it scans, and the output signal is also useful for frequency collection in the data analysis process. After a small portion of the beam is split off to go to the Fabry-Pérot cavity, the remainder
passes through an EOM with central modulation frequency of 600 MHz (New Focus model 4423). As mentioned above, the EOM is useful for adding frequency sidebands with a precisely known spacing to our signal. We can also use the EOM to assist in observations of the otherwise very small IR signal from the atomic beam.

To avoid the lack of detectors and lock-in amplifiers that can detect 600 MHz oscillations, we turn to two-tone FM spectroscopy. Two-tone FM spectroscopy entails mixing two frequencies, one much larger than the other, and driving the EOM with the mixture of the two. Although our EOM is resonant at 600 MHz (the large frequency for our experiment), we choose a second, small frequency of 50 kHz, and drive the EOM at 600 MHz ± 50 kHz. We then demodulate the signal at 100 kHz—or two times the lower modulation frequency.

Actually observing the IR signal from the atomic beam is somewhat challenging. Due to the fairly short lifetime of the $6S_{1/2}$ state, the lasers must be very precisely overlapped through the atomic beam if we hope ever to excite atoms into the $6P_{1/2}$ state. We also focus the lasers as they pass through the atomic beam, and must ensure, therefore, that they both focus in exactly the same place. To assist with this alignment, it is helpful first to optimize the first-step transition signal, which is fairly
easy to see. Once this signal has been optimized once, it is best not to adjust the mirror immediately before the atomic beam chamber and to place two irises placed far apart before it such that the blue beam passes through their centers. This setup gives a rough guide for where the lasers should go should the alignment be completely lost. Additionally, once the lasers are approximately overlapped and the second-step transition signal has been observed, it is easiest to optimize the quality of the signal with small adjustments to mirrors reflecting only the blue laser. This strategy does require, however, waiting to lock the 410 nm laser until after the signal is optimized, so the wavelength of that laser should be checked occasionally during the alignment process, as even slight drifts from resonance can cause significant deterioration of the signal quality. Finally, one other method for alignment that can be helpful if the signal is particularly difficult to observe is placing a mirror temporarily just after the lens that focuses the lasers into the vacuum chamber. This mirror can be angled to permit viewing of the focused beam outside of the vacuum chamber to check that the focuses of the lasers are overlapped and are in an appropriate location.

After the lasers have passed through the atomic beam, we use a dichroic mirror to separate the beams, so we can observe the signal from each. (The signal from the 410 nm laser is used for the laser locking). We use a 10 MHz IR detector (New Focus model 2053-FS) to detect the IR signal. Observing this signal requires a sequence
of two lock-in amplifiers. The input of the first lock-in amplifier is directly from the photodetector, and we use the 50 kHz modulation frequency as the reference frequency. While this step is essentially the two-tone FM spectroscopy scheme, our signal remains noisy after this step, and it has nonzero background due to optical imperfections of the EOM, so it requires further processing. We therefore utilize the chopper wheel in the atomic beam, which we also use as a second demodulation step for the locking signal. Similarly, here the signal will only exist when the atoms are not blocked, so the signal will also oscillate at the frequency the atoms are blocked and unblocked. We can therefore use this frequency as a reference for the second lock-in amplifier. This demodulation scheme produces a signal that is sufficiently clear and reproducible to extract a Stark shift between consecutive laser scans, one with an applied electric field, and one without.

### 3.3 Data Acquisition

Our data acquisition procedure remains similar to that used in the measurement of the Stark shift in the \(6S_{1/2}\) state; the corresponding thesis ([12]) contains additional details. We collect data using a National Instruments DAQ board (NI USB-6221) to send signals to the computer, and the acquisition is controlled by a LabView program. This program allows us to record the relevant signals: the Fabry-Pérot signal, the vapor cell signal, and the atomic beam signal, and some additional experimental parameters, such as the temperature of the crucible and the measured voltage across the capacitor plates. It also adjusts the AOMs to control the frequency of the 410 nm laser in the vapor cell in conjunction with high voltage changes.

When recording data, we set up our process to minimize any systematic errors. In particular, we choose the field strengths randomly as opposed to just increasing or decreasing the applied voltage. Additionally, we take data in a pattern of voltage off-voltage on-voltage on-voltage off and wait approximately 10 seconds every time we change the voltage to avoid any error from electrical transients associated with changing the high voltage and to make sure that the Stark shift we measure is not correlated with the direction of change of the electric field (i.e. off to on versus on to
off). A diagram of this procedure is shown in Figure 3.11. In our data analysis, we separate the scans both by frequency scan direction and by voltage change direction.
Chapter 4

Data Analysis and Results

Now that we have discussed how we obtain our data, we need to examine how we extract our results. This chapter discusses our procedure for analyzing the data and then presents the results, along with a discussion of systematic error analysis. To analyze our data, we rely on a set of MATLAB code (see Appendix B) that has been developed over several years in our lab for this purpose. Here, we discuss the basic analysis procedure and report the results of our experiment.

4.1 Data Analysis

Figure 4.1: Plots of data before any frequency axis calibration. Shows the three main signals that we record—the Fabry-Pérot signal, the vapor cell signal, and the atomic beam signal in that order from top to bottom.
Our collected data consists of four sets of digitized signals—the input voltage for the laser scan, a Fabry-Pérot signal, the vapor cell spectrum, and the spectrum from the atomic beam, as in Figure 4.1, (which leaves out the ramp voltage). For all of these, no meaningful horizontal axis exists in the raw data. Our first goal in our data analysis, then, is to find a horizontal axis in terms of frequency in physically meaningful units (we use MHz). At that point, we need to use that axis to find out how much a given high voltage on scan has shifted from its corresponding high voltage off scan. Along the way, it is fairly simple to obtain a value for the hyperfine splitting from the vapor cell spectrum, so we include code to calculate that number as well.

4.1.1 Pre-Calibration Axis Processing

After importing our data, our first step is to normalize the horizontal axis, such that it extends from -1 to 1, to make our linearization simpler. We define the $j$th "normalized point number" as

$$x_j = \frac{j - N/2}{N/2}$$

where $N$ is the total number of points in the data set. This normalization allows us easily to compare coefficients in our polynomial for linearizing the frequency axis.

It is also convenient to flip the time axis of the downscans, so that, like the upscans, the axis is increasing in frequency. While we typically analyze upscans and downscans separately (as a check for systematic errors), ensuring that the frequency axes for both upscans and downscans progress from low frequency to high frequency allows us to use identical analysis code.

4.1.2 Frequency Linearization and Calibration using the Fabry-Pérot Signal

We use a triangle wave from a function generator to control the frequency scan of our laser. However, the PZT in our laser exhibits hysteretic behavior, so the change in frequency is not linear in time. Before converting the arbitrary units of normalized point number to actual frequency, then, we want to ensure that the frequency scale is actually linear. To this end, we analyze our data using two types of fits for the Fabry-Pérot signal. Of course, the results should be the same, but we want to check our data as thoroughly as possible, and this is a useful way to check that our results are making sense.

Linearization with Airy Function Fit

The signal obtained from a Fabry-Pérot cavity can be described using an Airy function, which can be written as:

$$T_{FP} = \frac{A}{1 + F \sin^2(\delta/2)} + c$$

(4.2)
4.1. DATA ANALYSIS

Figure 4.2: Plot of a sample Fabry-Pérot scan and an Airy function fitted to the data.

\[ \delta = a_0 + a_1 x + a_2 x^2 + \ldots \]  

(4.3)

We assume that the frequency change as a function of time can be described by some polynomial, and we set

\[ \delta = a_0 + a_1 x + a_2 x^2 + \ldots \]  

(4.3)

We choose typically to fit to the fourth or fifth order, after which point we observe very small coefficients, and therefore minuscule adjustments to the fit. We also take \( A = b_0 + b_1 x \), to account for changing peak amplitudes due to variation in the laser power. Past experience has shown that a linear fit is sufficiently accurate to fit the observed variations [16]. Altogether, then, the function to which we fit our Fabry-Pérot signal is:

\[ T_{FP} = \frac{b_0 + b_1 x}{1 + F \sin^2(a_0 + a_1 x + a_2 x^2 + a_3 x^3 + a_4 x^4)} + c \]  

(4.4)

We can also add another term to the polynomial and compare the results to check for a systematic difference. A sample data set with its Airy function fit is shown in Figure 4.2.

Once we have the appropriate polynomial function, we can apply it to the normalized point number. This step results in an axis that is linearized in frequency.

Linearization with Sum of Lorentzians Fit

Our other method similarly looks for an appropriate polynomial to describe the behavior of frequency, but with a slightly different approach. We know that each peak

\[ \delta = a_0 + a_1 x + a_2 x^2 + \ldots \]  

(4.3)
in a Fabry-Pérot signal is a Lorentzian. Therefore, we fit our Fabry-Pérot data to a sum of Lorentzians, so that, for a Fabry-Pérot signal with \( N \) total peaks our function is now:

\[
T_{FP} = \sum_{j=1}^{N} \left( \frac{a_j}{(x - b_j)^2 + c_j^2} \right) \tag{4.5}
\]

The result of the fit looks nearly exactly the same as that for the Airy function fit (see Figure 4.2), as both types of analysis give excellent fits.

We then fit a polynomial to the locations of the Fabry-Perot peaks \( (b_j \text{ as a function of } j) \), as we know they must all be separated by the same amount in frequency space (for our experiment, very near to 500 MHz). A sample fit with data is shown in Figure 4.3. We can then apply this fit to the normalized point number to obtain a linearized axis. While in some ways this method is conceptually simpler, it has the disadvantage of being noticeably slower than the method of fitting with an Airy function. Therefore, we check only some of our data using both methods to make sure the results agree.

![Figure 4.3](image.png)

**Figure 4.3:** Plot of the peak locations of a Fabry-Pérot signal in frequency versus normalized point number with fit for linearization of the frequency axis. The plot also shows that the frequency axis has been scaled into MHz, as is ultimately desired.

In both methods, we scale the fit so each peak in the Fabry-Pérot scan is separated by the free spectral range of our Fabry-Pérot cavity, which is nominally 500 MHz.

**Calibration Using EOM Sidebands**

While the FSR of our Fabry-Pérot cavity is very close to 500 MHz, we do not know the exact number precisely, and it is possible for the FSR to change slightly from day to day, or even over shorter time scales, due to thermal drifts. While a significant
4.1. DATA ANALYSIS

Figure 4.4: Plot of data from the vapor cell and fit of the data. The two tallest peaks are from the laser’s central wavelength, while the other, smaller peaks are due to the sidebands from the EOM. Notice that the central two peaks are mostly overlapped, but it is possible to resolve them by eye, and the data analysis program has succeeded in fitting them to two peaks.

As mentioned previously, we modulate the IR beam using an EOM with a precisely known modulation frequency of 600 MHz. The vapor cell spectrum therefore contains sidebands spaced precisely 600 MHz on each side of the two hyperfine peaks. In Figure 4.4, it is possible to make out all six peaks. The two tallest ones are the peaks from the central frequency of the laser; the smaller ones are due to the modulation. The very small peaks just to the side of the two tallest ones are likely due to the less prevalent isotope of indium, $^{113}\text{In}$, as mentioned in Chapter 3.

Since there is very little residual Doppler broadening, we fit the vapor cell signal to the sum of six Lorentzians. We first identify the locations of five of the peaks (leaving out the smaller central one). Then, using these locations as initial guesses for our fitting program, we are able to fit to a sum of six Lorentzians:

$$ T_{vc} = \sum_{j=1}^{6} \frac{a_j}{(x - b_j)^2 + c_j^2} \quad (4.6) $$

To find our calibration factor, we consider only the two outer sidebands, which are typically located with greater accuracy than the two in the center. If we number the peaks from lowest to highest frequency from one to six, we know that peak 1 must be exactly 600 MHz lower in frequency than peak 2, and that peak 6 must
be exactly 600 MHz higher in frequency than peak 5. We then compare the values of the splittings that we obtain using our axis that has been calibrated using the only Fabry-Pérot to the correct 600 MHz ($b_2 - b_1$ and $b_6 - b_5$ using the notation from Equation 4.6). We observe values of 600.266(20) MHz for the 2-1 splitting and 598.959(16) MHz for the 6-5 splitting when averaging over all data collected as shown in Figure 4.5. The discrepancy between the two values indicates that our linearization of the frequency axis is not quite perfect. However, they are close enough that we can average them together to obtain the factor by which we scale our data. The discrepancy is considered as a systematic error in the final measurement. We wait to calibrate using the EOM sidebands until the very end of our analysis, and report results both with and without this calibration factor.

Also, a close examination of Figure 4.4 reveals a smaller peak on the right of each of the largest peaks. These appear in all of our data regardless of alignment or other experimental configurations, so we’re confident that these are not spurious or due to noise. Instead, we believe they are due to the other isotope of indium, $^{113}\text{In}$, which only has about 4-5% natural abundance. It’s expected, however, that there will be a slight isotope shift in the energy level. For the purpose of data analysis, however, we exclude the bottom of the two tallest peaks, so that the small peaks from the other isotope do not influence our fit. We also note that, regardless of the exclusion of the bottom of the peaks, there is very little effect from these small peaks.

### 4.1.3 Measurement of the Hyperfine Splitting of the Indium $6P_{1/2}$ State

Our fitting method for the vapor cell signal can easily give us the hyperfine splitting of the indium $6P_{1/2}$ state. We already have the peak locations, so we simply calculate $b_5 - b_2$ (using the same notation as above). We can also scale this value using the hyperfine sidebands. Specifically, if $f_{\text{raw}}$ is the hyperfine splitting that we observe
4.1. DATA ANALYSIS

Figure 4.6: Histogram of measured values of the indium $6P_{1/2}$ hyperfine splitting.

before the calibration, we report a final value of:

$$f_{\text{final}} = f_{\text{raw}} \times \frac{600\text{MHz}}{\Delta f_{\text{obs}}}$$

for an observed sideband splitting of $\Delta f_{\text{obs}}$.

Our result is 1257.0(5) MHz (see Figure 4.6). The uncertainty quoted here is dominated by the systematic uncertainty, and in particular, the discrepancy between the distances to the sidebands from the hyperfine peaks. We obtain an estimate of the systematic uncertainty by taking data in many different experimental configurations and comparing the results. The results from upscans versus downscans and the $F = 4 \rightarrow F = 5$ transition compared with the $F = 5 \rightarrow F = 4$ transition are shown in Figure 4.7.

There has been one previous measurement of this quantity [18], which obtained a value of 1251.5(1.5) MHz. Note that our measurement has improved the precision of the previous experiment by a factor of 3.

4.1.4 Measuring the Stark Shift

Once we have our meaningful frequency axis, we can turn to our main goal of finding a value for the $6P_{1/2}$ Stark shift. Again, throughout this analysis, we use multiple methods to ensure that there are no avoidable systematic errors due to our computations. For any method that we use, we will need to compare consecutive scans—one with the high voltage on, one with the high voltage off. The period of our laser scan is typically between 12 and 15 seconds, so drifts on that timescale of the IR laser could be a concern (the blue laser is locked, so we can safely assume that does not drift). In
Figure 4.7: Plot of hyperfine shift measurement results for different experimental configurations. While there appears to be a fairly large difference between the $F = 5 \rightarrow F = 4$ and the $F = 4 \rightarrow F = 5$ measurements, note that the range of the plot is only 1 MHz out of about 1200 MHz.

addition to the fact that the Fabry-Pérot should pick up on these drifts, we check for this potential problem in two ways: first, as discussed in Section 3.3, we collect data using a high voltage off-on-on-off sequence, so a long term drift should average out of our measurement when we combine data from both off-on and on-off sets of shift measurements. Additionally, as a diagnostic, we analyze off-on data and on-off data separately from each other and compare their results. If our experiment is behaving as expected, we should see no difference. We present those results in Section 4.3. Our second method of ensuring that laser drifts do not affect our measurement is using a location reference that would indicate a drifting laser.

**Frequency Location Reference**

Our frequency axis can now be calibrated to represent differences in frequency in the correct units for a given scan. However, we must also be able to detect shifts in the spectra between consecutive scans. Therefore, we must have some frequency reference that remains constant regardless of whether the high voltage is on or off. We analyze the data using two different references—the Fabry-Pérot signal and the vapor cell signal, although the vapor cell signal is the reference we use in our final analysis. For the vapor cell reference, we set the location of the large hyperfine peak to be our zero location. By shifting the horizontal axis of the atomic beam spectrum so its zero frequency location is at the same point number as that of the tall hyperfine peak, we have effectively imposed a frequency that, although not absolute, at least allows us to compare scans taken at different times. We also expect that any drifts
4.1. DATA ANALYSIS

Figure 4.8: Plot of averaged data from one set of 25 scans taken with an applied voltage of 15 kV. The red curve is the shifted signal; note that, as expected, it has shifted to lower frequencies.

in the laser frequency will not show up in our analysis at all, since if the laser drifts, it should affect the atomic beam signal and the vapor cell signal in the same manner. Therefore, the effect will subtract out when we subtract the location of this hyperfine peak.

To double check our results from this method, we can also use the Fabry-Pérot signal as a reference. Here, we just set the location of the first Fabry-Pérot peak to zero, and then proceed similarly in shifting our atomic beam signal frequency axis.

It is worth addressing the issue of the stability of our Fabry-Pérot cavity, since in using it as a reference we are assuming that it is sufficiently stable over the course of a data set (including both a high voltage off and a high voltage on scan) not to introduce extra error into our analysis. Essentially, we want to see if the peaks are actually staying in the same place. To evaluate the location of the peak, we consider the difference between the value of $a_0$ from Equation 4.4 and an atomic peak from the vapor cell signal. If $a_0$ changes over time, the overall pattern of the Fabry-Pérot peaks must also be changing; if it remains consistent, then our cavity is stable. Again, we thermally stabilize our cavity by placing a box lined with insulating foam around it, so we expect any thermal fluctuations to be small, and thus we should not observe large changes in $a_0$.

When we compare values of $a_0$ over a timescale of about 30 minutes, we observe exactly what we expect—small random fluctuations on the order of a few MHz, and no long term drift. The random fluctuations are not worrisome; while they may introduce slightly more statistical error, the fact that they appear not to trend in any particular direction indicates that using the Fabry-Pérot as a reference should not introduce any systematic error to our analysis, and that the fluctuations should
average out over the course of many scans. Thus, we conclude that our cavity is sufficiently stable to provide an alternative reference.

**Stark Shift Computation: Sum of Lorentzians Method**

Once we have our reference point, we want to compare the distances of a shifted and a non-shifted scan. If we let \( f_0 \) be the location of the reference, and \( f_{\text{off}} \) and \( f_{\text{on}} \) be the locations of some feature of the non-shifted and shifted spectra respectively, we can express our observed Stark shift as:

\[
\Delta f = k_s E^2 = (f_{\text{on}} - f_0) - (f_{\text{off}} - f_0) \quad (4.8)
\]

In order to get a precise result, we want to use as much data as possible. Therefore, in addition to taking many data sets, within any given scan, the more (reliable) information we can extract, the better. While the shape of our signal is technically a sum of Voigt profiles, the relatively small Doppler broadening in the \( 6S_{1/2} \rightarrow 6P_{1/2} \) transition means that we can fit our signal to a sum of Lorentzians. Ideally, we would fit the sum to 6 peaks. However, in practice, the two peaks corresponding to the main hyperfine peaks often are about the same amplitude as the noise. Including them in our fit actually significantly decreases the precision and accuracy of our result, so we ignore them. In total, then, we fit the data to a sum of three Lorentzians:

\[
T_{ABU} = \sum_{j=1}^{3} \frac{a_j}{(x - b_j)^2 + c_j^2} \quad (4.9)
\]

The signal with which we are working is fairly noisy, so it can be difficult to get an accurate fit, especially since large residuals can significantly impact the locations of the peaks. Since we care about primarily the locations of the centers of the peaks, we lose very little information if we ignore the bottom of the peak. Also, the peaks that are large enough all have negative amplitudes, so we can exclude more of the noise by instructing MATLAB not to consider any values greater than zero when fitting.

The actual procedure for fitting the data using this method goes as follows:

1. Find rough locations for the large central peak and the two peaks at the ends to use as initial guesses for our fit.

2. Find all points with values greater than zero or magnitudes less than 20% of the magnitude of the height of the tallest peak, and exclude them from the data we will fit.

3. Fit to a sum of Lorentzians, using the locations found in the first step for the three clearly resolved peaks, and then the location of the middle resolved peak \(- 50 \text{ MHz}\) for the other peak.
4.1. DATA ANALYSIS

Figure 4.9: Plot of a representative atomic beam signal fit to a sum of Lorentzians. All of the noise is much smaller in amplitude than the portion of the data that we consider in our fit.

At this point, all that remains is to find the difference between peak locations in scans taken with the high voltage-off and those taken with the high voltage-on. To accomplish this final step, for each consecutive pair of scans, we store the locations of the three resolved peaks in separate vectors for the voltage-off and voltage-on scans. Then we simply subtract the values for the voltage off scan from the values for the voltage on, and take the weighted average of the three peaks to obtain a shift for that scan. A typical fit has an uncertainty in the location of the central peak of about 3 MHz.

Stark Shift Computation: Overlap Method

Our second method for finding the Stark shift differs substantially from the other methods used to fit various signals throughout our data processing in that it does not require the assumption of a particular functional form for the signal. Instead, we fit each scan to an interpolating function, and expect only that the function should be approximately the same for both the voltage-off and voltage-on scans. Then, instead of comparing peak locations between voltage off and voltage on scans, we compare the entire fitted function.

We can write the function for the non-shifted signal as $A_{\text{off}}(f)$, and that for the shifted signal as $A_{\text{on}}(f - s)$ where $s$ is the Stark shift. Then we incrementally shift $A_{\text{on}}(f - s)$, so it becomes $A_{\text{on}}(f - (s - \delta))$. We then look for the value of $\delta$ that minimizes the sum of the squares of $A_{\text{on}}(f - (s - \delta)) - A_{\text{off}}(f)$. We define the sum of the squares of the difference of the two functions to be $\chi^2$. We expect that the best value of $\delta$ is approximately $s$. 
Figure 4.10: Plot of $\chi^2$ for a typical pair of runs for different values of $\delta$ with a fit to a parabola near the minimum of the $\chi^2$ function.

Now, in order to find the best value of $\delta$, we consider small values of $s - \delta$, and can therefore expand $\chi^2$. Given our definition for $\chi^2$, and taking $A_{\text{off}} = A_{\text{on}} = A$ (since they should be similar):

\[
\chi^2 = \sum_{\text{all } f} (A(f - (s - \delta)) - A(f))^2 \tag{4.10}
\]

\[
\approx \sum_{\text{all } f} (A(f) - (A(f) - A'(f) \cdot (\delta - s)))^2 \tag{4.11}
\]

\[
= \sum_{\text{all } f} (A'(f) \cdot (\delta - s))^2 \tag{4.12}
\]

\[
\propto (\delta - s)^2 \tag{4.13}
\]

\[
\propto (\delta - s)^2 \tag{4.14}
\]

Therefore, for small $(\delta - s)$ the shape of $\chi^2$ is approximately parabolic. We fit this portion of the parabola to a quadratic function, and then find the location in frequency space of the minimum of that function. This final calculation gives us our value for the observed Stark shift.

If we had no noise in our signals at all, $A_{\text{off}}(f) = A_{\text{on}}(f)$, and the minimum value of $\chi^2$ would be 0. Also, $\delta$ would then be exactly $s$ always. However, we obviously have a somewhat more complicated situation, and instead just look for the minimum of $\chi^2$. Assuming that our noise is random (as we expect), the average value of $\delta$ should approach $s$ as we analyze more data.

This method therefore has the disadvantage that every scan is influenced by the noise present in the signal. Its advantages, however, are not insignificant: the ability not to have to assume a functional form makes the method more general, and
4.2 Results

4.2.1 Systematic Error Analysis

In reporting our results, we must consider possible sources of error that would not show up as a statistical error. Due to the large amount of data we have collected, the statistical uncertainty in our calculated polarizability value is very small. Thus, even fairly small systematic effects are important for quantifying the error in our measurement. The statistical precision of our data actually permits a more efficient exploration of potential systematic errors, since less data are required to observe a given systematic effect. In this section, we discuss the systematic errors that we search for and what we have found in our ensuing analysis.

Underestimated Statistical Uncertainty

When we plot a sample of our data, we observe fluctuations that are substantially larger than what we would expect given the uncertainty that MATLAB computes from a histogram or the uncertainty in the locations of the peaks in a Lorentzian peak fit (see Figure 4.12). While arguably not a systematic error, this uncertainty is
Figure 4.12: Plot of the shift in the largest peak in the atomic beam signal for one voltage with the error bars give directly by the uncertainties in the peaks from the MATLAB fit. Clearly, there is more fluctuation among scans than the error bars alone would suggest. These small error bars indicate that basing our estimate of the statistical uncertainty directly off of MATLAB’s computations would significantly underestimate the uncertainty.

certainly something we need to address when we compute the uncertainty in our final measurement.

We choose to account for this uncertainty by finding an appropriate factor by which we can multiply our error bars such that the variations are in statistical agreement with each other. To obtain this factor, we calculate the $\chi^2$ value$^1$ from a fit to the weighted mean for a sample of the data. For example, in the data in Figure 4.12, a weighted fit to a constant gives a $\chi^2$ value of 10761. We then take the square root of $\chi^2$ divided by the degrees of freedom, in this case $N - 1$ where $N$ is the total number of data points. For this particular sample, there were 461 data points, which gave a value for $\sqrt{\frac{\chi^2}{N-1}}$ of about 4.8. When we perform this calculation for several samples of our data (in this case, different voltages), we consistently see a value of about 5.

$^1$This is completely independent from the $\chi^2$ parameter used in the overlap method.
4.2. RESULTS

Figure 4.13: Plot of $k_s$ results for different experimental configurations. Each is compared with a condition that is complementary to it. We expect no statistically significant differences between each pair of results, and we do in fact observe statistical agreement.

This number indicates by what factor $\chi^2$ is larger than we would expect based on the errors in each data point. Thus, this factor is the desired value for adjusting our statistical error. In all of the following results, the quoted statistical errors will therefore be five times what we would obtain were we to look only at the standard error from a histogram of measurements.

**Binary Experimental Conditions**

The simplest systematic effects that we look for would show up in a comparison between two complementary experimental conditions. In particular, we consider the following variables that fall into this category:

- Upscan versus downscan of the laser.

- Voltage off to on versus on to off.

- Hyperfine transition ($5P_{1/2}, F = 5 \rightarrow 6S_{1/2}, F' = 4$ compared with $5P_{1/2}, F = 4 \rightarrow 6S_{1/2}, F' = 5$). (Note that we expect that since we only have a scalar polarizability for this state, all hyperfine states should shift by the same amount.)

A difference in the results of upscans from those of downscans could indicate the laser was drifting in a way that was not accounted for or that it was displaying some direction-dependent hysteretic behavior that our linearization process somehow did not capture, for example. If we observe a difference between off-on and on-off results, that likely indicates either that we are not waiting sufficiently long times for
transients to die out. Alternatively, since our laser locking scheme depends on the locking system’s ability to adjust the laser’s frequency to match a shifted lockpoint, this type of systematic effect could indicate that there was some problem with the lock that would cause the transition to appear different when the electric field goes from off to on versus on to off. Another possibility would be variation due to transients from sudden changes in the voltages. Measurement of the two hyperfine transitions tests the model of a scalar polarizability and the corresponding lack of dependence on the hyperfine state holds. We compare the results, expecting to see no difference, to ensure that everything appears to be behaving as we predict.

Continuous Experimental Variables

Here, we consider several more possible sources of systematic error. Instead of having two discrete possibilities, these conditions can vary effectively continuously. For some of these conditions, it is especially important that we see no significant difference, as that could indicate that the setup of our experiment is seriously flawed. The particular factors that we consider here are:

- Ratio of laser powers

Figure 4.14: Plot of the value of $k_s$ measured versus the ratio of the 410 nm laser power to the 1343 nm laser power for a subset of our data. As desired, we observe no correlation between the value of $k_s$ and the laser power.
4.2. RESULTS

Figure 4.15: Plot of \( k_s \) versus the applied electric field for a subset of our data. We expect to see no dependence, since \( k_s \) is the Stark shift divided by the electric field squared. While there is some scatter around the mean, the slope of a line fitted to these data is consistent with 0.

- Correlation of \( k_s \) with voltage
- Laser scan speed

Figure 4.16: Values of \( k_s \) obtained from different fit methods.

If our results depended on the ratio of laser powers, that would effectively indicate that the measured shift depends on the Rabi frequencies of the transitions (see Sec-
In turn, a dependence on the Rabi frequency would suggest that, most likely, our laser lock is not performing adequately, and is not making the frequency of the 410 nm laser adjust as expected. Then we would again be in the situation of having a substantial systematic uncertainty due to the fact that we cannot know the Rabi frequencies independently and precisely.

The observation that $k_s$ depends on the applied electric field would have similar implications. Since the dependence of the measurement on the Rabi frequencies increases with detuning from the transition, a dependence on the voltage would again suggest a larger detuning from the resonance of the transition. We do observe a scatter that is larger than expected, so we add that as a systematic uncertainty.

A dependence of our measurement on the period of the laser scan would also indicate a systematic error. If the laser is scanned too quickly, the sideband splittings can become unequal. However, it is also hypothetically possible to scan slowly enough that the laser would drift during a single scan. Thus, we want to ensure that we are also scanning quickly enough to avoid shorter term drifts. Checking that our results do not vary based on the scan speed of our laser demonstrates that we are operating in a regime where our linearization process is able to account for any nonlinearities or drifts in the scan. When we examine the $k_s$ values for a range of laser scan speeds, we see no correlation.

![Histogram of $k_s$ values from all of our data combined.](image)

**Figure 4.17:** Histogram of $k_s$ values from all of our data combined.

**Data Analysis Methods**

As discussed throughout Section 4.1, we rely on a combination of multiple methods of data analysis to mitigate the disadvantages of any individual method. We hope, of course, to see no differences in the results from various analysis methods.
Three out of the four methods are in fairly good agreement with each other—the sum of Lorentzians fit method with both types of referencing and the overlap method with the Fabry-Pérot reference all give results that are within less than a percent variation of each other. The overlap method using the vapor cell reference appears to be somewhat of an outlier, and we attribute this to an anomaly in the behavior of our fitting program. We plan to explore this disagreement further and attempt to develop an improved understanding of the cause of this anomaly. Our analysis relies primarily on the Lorentzian fit method referenced to the vapor cell spectrum, although the overlap method using the Fabry-Pérot as a reference functions well as a confirmation of our results. The final number quoted below was computed using a fit to a sum of Lorentzians and the vapor cell signal as a reference.

### 4.3 Final Results

Finally, we present the actual numbers that we have obtained from all of our data combined. As mentioned above, we obtain numbers both with and without the sideband calibration factor, so we report both numbers.

We report our systematic errors here as well.

<table>
<thead>
<tr>
<th>Systematic Variable</th>
<th>Error (MHz/(kV/cm)^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F = 4 \rightarrow F' = 5$ vs. $F = 5 \rightarrow F'' = 4$</td>
<td>0.006</td>
</tr>
<tr>
<td>HV Off to On vs. On To Off</td>
<td>0.005</td>
</tr>
<tr>
<td>Sidebands</td>
<td>0.001</td>
</tr>
<tr>
<td>Electric Field Calibration</td>
<td>0.0008</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>0.004 MHz/(kV/cm)^2</strong></td>
</tr>
</tbody>
</table>

**Table 4.1:** Systematic errors for measurement of $k_s$. The total is obtained by adding the individual errors in quadrature.

Our final uncalibrated number is $k_s = 0.811 \pm 0.003$ (stat), where the statistical uncertainty has been multiplied by three to account for the larger-than-expected fluctuations. The systematic error will be roughly the same as that listed below for the calibrated value.

Our calibrated value for $k_s$ is also $0.811 \pm 0.003$ (stat) $\pm 0.008$ (sys) MHz/(kV/cm)^2. (Note that when added in quadrature, these uncertainties give a total of $0.008$ MHz/(kV/cm)^2, which is about 1% as desired). The systematic errors are discussed above, and summarized in Table 4.1. The entries in this table account for any apparent residual discrepancies where they persist. If we convert our value for $k_s$ into atomic units, and subtract off the shift due to the $6S_{1/2}$ state from [11], we obtain a scalar polarizability for the $6P_{1/2}$ state of $\alpha_0 = 7663 \pm 24$ (stat) $\pm 65$ (sys) $a_0^3$. This value is in statistical agreement with the theoretical prediction given in [2] of $7817 \pm 155a_0^3$. 

Figure 4.18: Comparison of theoretically predicted polarizability value [2] with our measured value.
Chapter 5

Future Work

5.1 Measurement of the In $6P_{3/2}$ Scalar and Tensor Polarizabilities

Now that we have completed this measurement, we can turn to other polarizability measurements. The natural next step is to measure the scalar and tensor polarizabilities of the indium $6P_{3/2}$ state as suggested in [16]. Unlike the $6P_{1/2}$ state, the $6P_{3/2}$ state has $J = 3/2$ and thus has a tensor polarizability in addition to the scalar polarizability. The tensor polarizability will cause different magnetic sublevels to shift by different amounts. Specifically, the size of the shift depends on $|m_J|^2$, although we observe splitting dependent on $|m_F|^2$.

Experimentally, this would seem to be a fairly simple extension of our current experiment—the only modification necessary would be adjusting the wavelength of our IR laser. The $6S_{1/2} \rightarrow 6P_{1/2}$ transition requires a wavelength of 1343 nm; the $6S_{1/2} \rightarrow 6P_{3/2}$ transition needs a wavelength of 1291 nm. Our IR laser can operate at 1291 nm, so we can simply tune it to that new wavelength. Additionally, theoretical values have been predicted in [2]: the scalar polarizability has been calculated as $\alpha_0 = 10500(400)a_0^3$ (atomic units) and the tensor polarizability is predicted to be $\alpha_2 = 1432(45)$.

There are other complications, however, that will make this measurement a challenge. First, instead of having only two hyperfine levels with $F = 4$ and $F = 5$, the $6P_{3/2}$ state has four hyperfine levels ($F = 3, 4, 5,$ and 6). We can drive transitions only to three of these states at a time, depending on which first step transition we use, because of quantum mechanical selection rules.

Due to the additional hyperfine levels, this spectrum will appear much more complicated than the current one. We will observe nine peaks including the sidebands, which make many of the peaks difficult to resolve when spaced by 600 MHz. Purchasing a new EOM capable of operating at 1500 MHz will make the experiment more feasible. Using the hyperfine splittings of the $6P_{3/2}$ state reported in [10], we can simulate this spectrum in both the vapor cell (Figure 5.2) and the atomic beam without an applied electric field (Figure 5.3).
The addition of a tensor polarizability also makes both the theoretical interpretation and experimental observation of the spectrum more difficult. A similar experiment was conducted in cesium [4], and much of the theory is described in that paper and in [16]. The effect of the tensor polarizability is to separate the magnetic sublevels when an external electric field is applied to the atoms. The size of this effect is related to the tensor polarizability. Here, we summarize some of this theory to predict the effects that we expect to see and explain how we can interpret our results.

The main aspect of the behavior of this state that we want to understand is the extent to which the magnetic sublevels split and how this is related to the tensor polarizability. We also wish to extract a scalar polarizability measurement, so we discuss how to find that as well. Throughout this discussion, we draw upon the similar work done in [4] for a similar state in cesium. We start by writing down the perturbation to the Hamiltonian:

$$H' = H_{hf} + H_S$$

The matrix elements for the hyperfine interaction $H_{hf}$ are given by:

$$\langle F, m_F | H_{hf} | F, m_F \rangle = \frac{1}{2}AK + \frac{1}{4}B^2 \frac{3K(K+1) - 2I(I+1)J(J-1)}{I(2I-1)J(2J-1)}$$

where $K = F(F+1) - J(J+1) - I(I+1)$. For the $6P_{3/2}$ state, $J = 3/2$, and for indium, $I$, the nuclear spin, is $9/2$. $A$ and $B$ are the magnetic dipole and the electric
Figure 5.2: Simulation of the $6P_{3/2}$ spectrum in the vapor cell with sidebands separated from the main peaks by 1500 MHz. This simulation shows the $F = 4$, $F = 5$, and $F = 6$ hyperfine levels.

quadrupole coupling constants respectively. From the hyperfine splitting measurement in [10], we obtained experimental values for these constants.

The Stark shift matrix elements are:

$$\langle F, m_F | H_S | F'', m_F \rangle = -\frac{1}{2} \alpha_0 \mathcal{E}^2 \delta_{F,F''} - \frac{1}{2} \alpha_2 \mathcal{E}^2 \langle F, m_F | Q | F'', m_F \rangle$$

(5.3)

$\mathcal{E}^2$ is the applied electric field, $\delta_{F,F''}$ is the Kronecker delta, and $Q$ depends on the Wigner $3-j$ and $6-j$ symbols, as given in [19].

For our experiment, we choose to consider the splitting between the $|6,6\rangle$ and $|6,5\rangle$ states due to their relative simplicity (see [16]). Specifically, only states with the same $m_F$ will mix, so $|6,6\rangle$ will mix with no other states, and $|6,5\rangle$ will mix only with the $|5,5\rangle$ state. (We can also measure the splitting between the $|3,3\rangle$ and $|3,4\rangle$ states, but we discuss the $|6,6\rangle$ and $|6,5\rangle$ states as an example).

To find the splittings, we need to know the matrix elements for each state. We begin with the simpler case of the $|6,6\rangle$ state, which has a 1x1 matrix perturbation:

$$\langle F, m_F | H' | F'', 6 \rangle = -\frac{1}{2} \alpha_0 \mathcal{E}^2 - \frac{1}{2} \alpha_2 \mathcal{E}^2 + \langle 6, 6 | H_{hf} | 6, 6 \rangle$$

(5.4)

Here, we introduce the scalar polarizability $\alpha_0$ and tensor polarizability $\alpha_2$. Next, we consider the perturbation for the $|6,5\rangle$ state:

$$\langle 6, 5 | H' | F'', 5 \rangle = -\frac{1}{2} \alpha_0 \mathcal{E}^2 + \left( \frac{-\frac{1}{2} \alpha_2 \mathcal{E}^2 - \langle 6, 5 | H_{hf} | 6, 5 \rangle}{-\sqrt{3} \alpha_2 \mathcal{E}^2 - \frac{1}{4} \alpha_2 \mathcal{E}^2 - \langle 5, 5 | H_{hf} | 5, 5 \rangle} \right)$$
CHAPTER 5. FUTURE WORK

Figure 5.3: Simulated two-tone FM atomic beam spectrum for the \(6P_{3/2} \ F = 4, F = 5,\) and \(F = 6\) hyperfine levels with no external electric field.

One of the eigenvalues of this equation will give us the perturbation to the \(|6, 5\rangle\) state. The final value we want to know is the difference between the perturbation to the \(|6, 6\rangle\) state and the \(|6, 5\rangle\) state, so we subtract the eigenvalue of the second matrix from that of the first. Combined with the predicted values of \(\alpha_0\) and \(\alpha_2\) from [2] and the measured values of \(A\) and \(B\), we can now predict the splittings between the \(|6, 6\rangle\) and \(|6, 5\rangle\) states for various values of the electric field.

As one can see in Figure 5.4, the splitting of the magnetic sublevels is a very small effect, and becomes even barely resolved only with electric fields of about 30 kV/cm. This simulation is also assuming the peaks from both of the magnetic sublevels are approximately the same amplitude; if one is shorter than the other, it becomes harder to resolve. It may be possible, however, to improve our ability to resolve the peaks by applying a magnetic field to the atoms regardless of whether or not the external electric field is on. Through the Zeeman effect, this would cause the magnetic sublevels to split even before we applied an electric field, so the peaks would be resolved. Even if the change in the splitting due to the electric field is small, as long as we can resolve the peaks, we would likely be able to measure the shift. Additionally, making the peaks narrower would also help resolve the magnetic sublevels, so if we could modify our experimental configuration to reduce the peak width, that would also make the measurement more feasible.

1There are two eigenvalues for the \(|6, 5\rangle\) perturbation; however, only one gives a splitting of 0 MHz when there is no applied electric field, so we use that value for our simulations. When we solve for the tensor polarizability in terms of the splittings between the two states, either eigenvalue will give the same answer.
5.2. **Polarizability Measurements in Thallium**

While we currently have indium in the atomic beam and have the optics set up for an experiment with indium, eventually we could also return to measurements of

**Figure 5.4**: Simulation of the two-tone FM signal for the indium $6P_{3/2}$, $F = 6$ state with an external electric field of 30 kV/cm. We assumed a representative linewidth of 40 MHz. The splitting of the sublevels is just barely resolved.

Assuming that we can make this measurement, we can then deduce a value for $\alpha_2$ and then for $\alpha_0$. A bit of algebra with the equation for the splitting of the $|6, 6\rangle$ and $|6, 5\rangle$ states gives:

$$\alpha_2 \mathcal{E}^2 = \frac{-8h \nu_{65} \left( 1 - \frac{2\nu_{65}}{\nu_{hf}} \right)}{1 - \frac{8\nu_{65}}{\nu_{hf}}}$$

(5.5)

where $\nu_{65}$ is the splitting between the $|6, 6\rangle$ and $|6, 5\rangle$ sublevels and $\nu_{hf}$ is the hyperfine splitting between the $F = 5$ and $F = 6$ states with no electric field. Thus, with a measured value for $\nu_{65}$, we can obtain a value for the tensor polarizability.

Furthermore, once we have this value, using Equation 5.3, we can calculate the scalar polarizability, $\alpha_0$, as well.
polarizabilities in thallium. Some of the techniques needed would likely be similar, although thallium is somewhat easier to work with due to its much higher vapor pressure, and it would likely be possible to avoid some of the more elaborate FM spectroscopy used in the current experiment.

**Scalar Polarizability Measurements in Thallium**

First, the measurement of the scalar polarizability in the thallium $7\,P_{1/2}$ state would effectively be the equivalent of that just performed on indium. This is a $J = 1/2$ state, so there would be no tensor polarizability, and the interpretation of the result would be nearly identical to our work in this thesis. Additionally, our lab already has the lasers necessary for this experiment—the 378 nm laser necessary for the first-step transition is currently being used for a measurement of the hyperfine structure in thallium, and the same IR laser that we currently use for the indium experiment could be tuned to 1301 nm for the second-step thallium transition.

Additionally, the $8\,P_{1/2}$ state of thallium would be accessible with the current lasers in our lab, so we could perform that measurement as well. Again, this would be a relatively straightforward scalar polarizability measurement, without the need to worry about the presence of a tensor polarizability and the associated complications.

**Tensor Polarizability in the $7\,P_{3/2}$ State of Thallium**

On the other hand, if we wish to complete an analogous measurement to that of the $6\,P_{3/2}$ state of indium, the $7\,P_{3/2}$ state in thallium would be the energy level of interest, although the scalar polarizability measurement will be a simpler experiment. The theory is similar to that described in the preceding section; we can construct similar matrices, follow the procedure of obtaining their eigenvalues, finding the difference and solving. Along with the other measurements, this possibility gives us many avenues to follow for further exploration of atomic polarizabilities.
Appendix A

Data Tables

Here, we present a summary of all of the data collected. Note that all sideband splittings are in MHz, voltages are in kV, and values of $k_s$ are in MHz/(kV/cm)$^2$.

Table A.1: Summary of Data Collection and Results from Overlap Method

<table>
<thead>
<tr>
<th>Date</th>
<th>Laser power ratio</th>
<th>HV</th>
<th>Number of Scans</th>
<th>Average Sideband Splitting</th>
<th>Not calibrated $k_s$</th>
<th>Calibrated $k_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2/1/16</td>
<td>3</td>
<td>8-13,15</td>
<td>272</td>
<td>599.76</td>
<td>.81212</td>
<td>.81283</td>
</tr>
<tr>
<td>2/2/16</td>
<td>–</td>
<td>7-8, 11-15</td>
<td>200</td>
<td>599.477</td>
<td>.81198</td>
<td>.81269</td>
</tr>
<tr>
<td>2/2/16_1</td>
<td>–</td>
<td>8-9, 11-15</td>
<td>273</td>
<td>599.475</td>
<td>.82062</td>
<td>.82133</td>
</tr>
<tr>
<td>2/24/16</td>
<td>1.3</td>
<td>7-15</td>
<td>471</td>
<td>599.391</td>
<td>.81728</td>
<td>.81811</td>
</tr>
<tr>
<td>2/25/16</td>
<td>2.3</td>
<td>7-15</td>
<td>533</td>
<td>599.792</td>
<td>.8059</td>
<td>.8062</td>
</tr>
<tr>
<td>2/28/16</td>
<td>3.1</td>
<td>7-15</td>
<td>444</td>
<td>599.116</td>
<td>.80993</td>
<td>.81113</td>
</tr>
<tr>
<td>4/5/16</td>
<td>1</td>
<td>8-15</td>
<td>330</td>
<td>599.22</td>
<td>.79997</td>
<td>.80101</td>
</tr>
<tr>
<td>4/6/16</td>
<td>2.8</td>
<td>8-15</td>
<td>282</td>
<td>599.687</td>
<td>.80787</td>
<td>.808292</td>
</tr>
<tr>
<td>4/10/16,1</td>
<td>3.9</td>
<td>8-15</td>
<td>388</td>
<td>599.714</td>
<td>.80533</td>
<td>.80571</td>
</tr>
<tr>
<td>4/10/16(^1)</td>
<td>2.2</td>
<td>11-12</td>
<td>49</td>
<td>599.66</td>
<td>.82545</td>
<td>.82592</td>
</tr>
<tr>
<td>4/11/16</td>
<td>2.2</td>
<td>10, 13-15</td>
<td>162</td>
<td>599.621</td>
<td>.81821</td>
<td>.81873</td>
</tr>
<tr>
<td>Total</td>
<td>–</td>
<td>7-15</td>
<td>3404</td>
<td>599.423</td>
<td>.81048</td>
<td>.81126</td>
</tr>
</tbody>
</table>
Appendix B

MATLAB Code

Here, we describe in more detail the code used for the main part of the data analysis. All of these programs are saved on the Majumder lab server in the folder Allison/MATLAB/.

Before beginning the discussion of the code, we summarize how our data are saved, so that it is clear how to retrieve the data for analysis. All of our data are automatically saved in a folder with the date name in the following format: three letter day of the week abbreviation, three letter month abbreviation, two number date, and year. Within the folder for the day, there are separate folders for each voltage. The data are then separated into upscans and downscans and titled with one number indicating the scan number, the word "trial," and then a 0 if it is a voltage-off scan, and a 1 if it is a voltage-on scan. It is worth noting also that if operating on a Macintosh computer, one must refer to the lab server as /Volumes/MLabFilestor/, whereas on a PC, the correct folder name is Z:\.

B.1 Master Programs

We analyze our atomic beam data two different ways—either fitting the signal to a sum of Lorentzians or to an interpolating function. The code is sufficiently complicated that we opt to have separate programs for each of the fitting methods to avoid confusion. In this section, we present each of the master programs.

B.1.1 IndiumFitOverlap

This program contains the central code for the overlap fit method. To begin, the user must provide a folder name, PName1, where the data are stored. This should contain all folders through the folder for the specific date. PName2 should be the scan direction. Additionally, SaveName should be defined as the name of the folder where the results of the analysis should be saved. Finally, at the beginning of the program, there’s an option for which reference (vapor cell signal or Fabry-Pérot) should be used
for the shift analysis. Once this information is input and the program is run, it will loop through multiple scans and multiple voltages as well if desired.

For each data set, this code calls the subprograms that import the data, fit the Fabry-Pérot signal, linearize the frequency axis, and then fit the vapor cell signal. At this point, we need to define our axes for the interpolation. The code for the axes definitions is contained in this method. Specifically, first we reference the axes to the appropriate zero point (the first peak of the Fabry-Pérot scan or the tallest hyperfine peak in the vapor cell signal). To do this, we redefine the coefficient for the zero-order term in the polynomial for linearizing the frequency axis. If we are using the Fabry-Pérot reference, we set this to 0, and if we are using the hyperfine signal as our reference, we subtract the location of the tallest hyperfine peak from the value. We then define \( \text{ttfm\_freqSIG} \), the frequency values for the data’s horizontal axis, as the vector of values obtained when the linearizing polynomial is applied to the normalized point numbers. We also define an x-axis for the interpolating function as extending from the minimum of the frequency axis for the vapor cell signal to the maximum of that axis in 0.5 MHz steps.

To import the data, the code defines \( n2 \), the value of the second number in the file name (0 if it is a voltage off scan, 1 if voltage on). The code can then save separate axes for the voltage off and voltage on cases depending on the value of \( n2 \). If the voltage is off, the code saves three relevant vectors, \( \text{interpx0} \), the x-axis for the interpolating function, \( \text{freqSIG0} \), the horizontal axis for the data, and \( \text{ttfm\_DS0} \), the data themselves. Similarly, if the voltage is on, the code stores \( \text{interpx1} \), \( \text{freqSIG1} \), and \( \text{ttfm\_DS1} \).

From this point, the remainder of the code in this method consists of calling other subprograms to find the interpolating functions, evaluate the shift, and save the analysis results.

### B.1.2 IndiumFitLorentzian

This program begins similarly to \texttt{IndiumFitOverlap} in requiring the same names for importing and saving files. It also has an option for the type of reference—Fabry-Pérot or vapor cell signal. Through the vapor cell fit, it calls the same subprograms for fitting of the Fabry-Pérot and vapor cell signals and importing data. Similar code also defines the shifted frequency axes for the data. However, instead of defining axes for interpolating functions, it simply calls the subprogram \texttt{TTFMFitIndium\_LorentzianThreePeaks}\(^1\), which fits the signal to a sum of three Lorentzians. Subsequently, it calls methods to evaluate the shift and save the shift data.

\(^1\)It could instead call \texttt{TTFMFitIndium} if the data have a sufficiently large signal to noise ratio. We typically found, however, that it was more reliable to fit only to the three larger, downward-pointing peaks.
B.2 Importing Data and Data Preparation

Before we can analyze any of the data, we have to have the data suitably prepared. This step of the code involves two primary subprograms, `getDataIndium` and `downsampleAndNormalizeIndium`.

B.2.1 getDataIndium

This program is very simple, as it simply has to read the data file and then store the data in MATLAB. It combines the folder name and file name, and opens the file. It then reads the data into an array named `data`, which contains five columns: the “time” (in point number), the ramp voltage (which is ultimately used to make sure that the frequency always increases with increasing point number), the Fabry-Pérot signal, the vapor cell signal, and the atomic beam signal.

B.2.2 downsampleAndNormalizeIndium

When we fit our data, we want the frequency axis initially to have values given by Equation 4.1. This subprogram provides the necessary code to accomplish that goal. First, it defines each column of the array `data` as a separate array. It then uses the ramp voltage to determine if the scan is an upscan or downscan; if the location of the minimum ramp voltage is before that of the maximum, it is an upscan, for example. If the scan is a downscan, the program reverses all of the data so that it proceeds from the lowest frequency to the highest frequency. By including data only between the location of the minimum ramp voltage and the maximum, it also ensures that we include data only from that scan.

After all of the arrays for the various signals are defined as we wish, we turn to normalizing the horizontal axis. This step is performed by applying Equation 4.1 to each point number in the original data.

Finally, this method “downsamples” the data, by including only some subset of the points (for example, every other point). It then plots all three signals as subplots in one figure.

B.3 Peak-Finding Code

Throughout the rest of the analysis code, we have to find estimates of peak locations multiple times. Therefore, before proceeding, we describe the function that we use to make these estimates. This code is contained in the subprograms `sympeaksIndium` and `sympeaksIndium_num_Peaks`. The difference between these programs is minimal, so we just describe one and then briefly explain the difference.

B.3.1 sympeaksIndium

`sympeaksIndium` is a MATLAB function that requires the following arguments (in this order): `xdata`, `signal`, `avg`, `minPeak`, `minPeakDistance`, and `NFig`. `xdata` refers to the values for the horizontal axis for a set of data, `signal` refers to the values in the data for the signal, `avg` essentially determines how much the data are averaged
over, \texttt{minPeak} gives a minimum peak height, \texttt{minPeakDistance} gives the minimum number of points between two peaks, and \texttt{NFig} simply gives a number for the figure for the plot that is produced in this code.

This code, as suggested by the \texttt{avg} parameter, performs a moving average over the data to avoid the detection of more peaks than desired. We typically increase this number when the data are noisier, and decrease it otherwise. The program then uses the built-in MATLAB \texttt{findpeaks} function to output both the height and location of the peaks. The averaged data, minimum peak height, and minimum peak distance are input as arguments for this function.

\textbf{Comparison with \texttt{sympeaksIndium num Peaks}}

This method is nearly the same as \texttt{sympeaksIndium}, except for taking one additional argument. This argument, \texttt{numPeaks}, gives the number of peaks that \texttt{findpeaks} should find. This method should not be used for the Fabry-Pérot signal, for which we do not know ahead of time the number of peaks, but is useful for the vapor cell and atomic beam signals, especially when they are noisy. This parameter has been tremendously helpful in getting the code to perform reliably and not to detect peaks in noise even when the signal is not quite as high quality as might be desired.

\section*{B.4 Fabry-Pérot Fitting and Frequency Axis Linearization}

Primarily, we fit our Fabry-Pérot signal to an Airy function, as given in Equation 4.4. However, we also fit some of our Fabry-Pérot data to a sum of Lorentzians to ensure the fit does not differ in a statistically significant manner. Thus, we include a description of code to perform either type of fit here. It is worth noting, however, that the fit to the sum of Lorentzians is much slower, and the Airy function appears to fit the signals very accurately, so in general, it is preferable to use the Airy function method.

\subsection*{B.4.1 \texttt{FabryPerotFittingIndium_Airy}}

This program first calls \texttt{sympeaksIndium}, to find initial guesses for the peaks’ locations. It then restricts the range of the fit, so that it focuses primarily on the center of the scan to avoid problems with partial peaks at the edges.

For any of our Fabry-Pérot fits, we must decide what order of polynomial we wish to use for the linearization of the axis. The code for the Airy function currently supports fitting using a fourth, fifth, or sixth order polynomial. Typically, we fit to a fifth order polynomial, as the coefficient for the \(x^6\) term is minute. In comparing fits from polynomials of various orders, however, we observe no statistically significant difference in the resulting measurements.
With the order of the polynomial specified in the master program, this code then fits the data to the appropriate function, using the peak locations from sym-peaksIndium as initial guesses. This part of the program requires a value for the free spectral range of our cavity (500 MHz) and an estimate of its finesse (12 is reasonable). The data is fit to the function described by Equations 4.4. The fitting command outputs the coefficients, which we save as a vector. To ensure a high quality fit, we specify a threshold RMSE value, above which the fit will be redone. We also plot the data with the fit so we can visually inspect it as well. The quality of our Fabry-Pérot signal is typically high enough, however, that we have not had any problems with either a large RMSE value or a fit that is visibly poor.

B.4.2 FrequencyLinearizationIndium_Airy

While the analogous method for the Lorentzian fit has a more important role, the fit for the Airy function directly finds the polynomial for linearizing the axis. This method plots this resulting polynomial and the peak locations from the Fabry-Pérot signal.

B.4.3 FabryPerotFittingIndium

Up until the actual fitting, this method is the same as FabryPerotFittingIndium_Airy. However, instead of fitting the Fabry-Pérot signal to an Airy function, this program fits the data to a sum of Lorentzians, as in Equation 4.5. From the coefficients determined in the fit, we can obtain the locations of the Fabry-Pérot peaks.

B.4.4 FrequencyLinearizationIndium

This method takes the locations of the peaks found in FabryPerotFittingIndium, and fits them to a polynomial, typically of fifth order, although we can easily adapt the code to accommodate a different order. This method also requires the free spectral range of our Fabry-Pérot cavity, so that the frequency axis is not only linearized, but also is scaled so that its units are MHz. After fitting the polynomial, this program plots the peak locations and the fit.

B.5 Vapor Cell Fitting

We fit the vapor cell signal to a sum of Lorentzians with LorentzianFitIndium. As mentioned in Section 4.1.2, we observe small bumps on the side of the hyperfine peaks likely due to the less prevalent isotope of indium. Since we wish for these not to influence our fit, we have also developed a modified version of LorentzianFitIndium, LorentzianFitIndium_excludeData, which effectively ignores the data at the bottom of the hyperfine peaks when fitting them. This program relies on the MATLAB excludedata fit option. Other than the code to exclude the data from the bottom of the peak, this code is exactly the same as LorentzianFitIndium, so we just discuss that method.
B.5.1 LorentzianFitIndium

As with the Fabry-Pérot fitting, our first step is to find guesses for the location of the peaks. For the vapor cell signal, we always wish to find five peaks, so we use `sympeaksIndium_num_Peaks` with `num_Peaks= 5`.²

Once we have the approximate locations of our peaks, we can fit the data. We fit to a sum of six Lorentzians, and save the resulting coefficients, along with the error in their values that MATLAB calculates. We also plot the data with the fit.

B.6 Shift Evaluation

At this point, the code for the overlap method and that for the Lorentzian fit method again diverge. Each of the master programs calls the code that calculates the shift using the appropriate method.

B.6.1 Overlap Shift Calculation Code

We already have the appropriate axes defined. Now, we need to fit the interpolating functions and then calculate the shift. All of this occurs in a method entitled `OverlapEvaluation`.

OverlapEvaluation

First, this program defines some range of possible values for the shift that it will try. Typically, this range is about 40 MHz on each side of the value that we expect. The program uses the MATLAB `interp1` command to fit interpolating functions to the data. Although we initially define the x-axis for the interpolating function over the range of frequencies in the vapor cell signal, we cannot interpolate over this entire axis. If the interpolating function extends too far out toward the edges of the scan, it will diverge rapidly. Unfortunately, there does not seem to be a predictive method for determining a reasonable interpolation range; rather, it is necessary to try different ranges and make sure that the interpolating function is not diverging for the range in use.

The interpolating function for the voltage-off scan is determined using the already defined interpolation axis, `interpx0`. The function for the voltage-on scan, on the other hand, is calculated using `interpx1 + Delta`, where `Delta` is some trial value for the shift. This calculation is performed for the range of values defined in the first part of the program. For each one, the program calculates the sum of squared differences, $\chi^2$, between the interpolating function for the voltage-off scan, `interp_ttfm0`, and

²While some of our data would allow us to find all six peaks with the `findpeaks` function, it is useful to be able to specify a larger value of `minPeakDistance` than the distance between the two central peaks to avoid the program detecting peaks that are not actually part of the signal. Therefore, we find five peaks, and use the location of the one central peak that we detect plus or minus 50 as the initial guess for the location of our other peak.
that for the voltage-on scan, interp_ttfm1. Typically, we restrict the points that it uses to calculate the squared differences between the two functions to a central portion of the scan to minimize the impact of noise on the edges of the scan.

As explained in Section 4.1.4, the region around the minimum of $\chi^2$ is well described as a parabola. The program therefore fits this section of $\chi^2$ to a parabola, and calculates the minimum of that parabola. We take that value to be the shift for that scan.

### B.6.2 Lorentzian Shift Calculation Code

To calculate the shift using the Lorentzian fit method, we use three subprograms—one to set up the axes, one to fit the data, and the third to calculate the shift.

**ShiftSetup**

This short program simply executes the referencing of the axes to either the Fabry-Pérot or vapor cell signal using the same technique as in the IndiumFitOverlap master program.

**TTFMFitIndium_LorentzianThreePeaks**

This subprogram performs the Lorentzian fit of the atomic beam signal and stores the resulting fit for the voltage-off and voltage-on cases separately. Structurally, it is similar to the other subprograms in our code that fit sums of Lorentzians. It begins with sympeaksIndium_num_Peaks, which finds the location of three peaks. As usual, we use these as initial guesses for our function.

Since the atomic beam signal is much noisier than the other signals we attempt to fit, we need to be a bit more creative in getting a good fit. Thus, we exclude all of the data with values greater than 0 (we only want to find the downward pointing peaks) or with a magnitude less than 20% of the magnitude of the height of the tallest peak. This step gets rid of nearly all of the noise. To ensure that we are excluding only the data we expect, we plot what remains of the data after the exclusion.

Once we have only the data we wish to fit, we implement the fit. We then store the coefficients and check for “false peaks”—peaks that the code appears to have found, but that shouldn’t actually exist. Assuming a sufficient fit quality, we store two vectors, Z0, the peak locations and errors in the locations for the voltage-off scan, and Z1, the corresponding information for the voltage-on scan.

**ShiftEvaluationIndium_Lorentzian**

Here, the program calculates the shift for a pair of scans. The calculation simply consists of subtracting the location of each voltage-on peak from the corresponding voltage-off peak. We store the three shifts as well as their average.
B.7 Saving Analysis Results

We have three subprograms that perform the task of writing the results of our analysis to text files. WriteToFileIndium saves the locations of the hyperfine peaks and sidebands in the vapor cell, along with the errors in their locations and the RMSE of the fit. WriteToFileIndium_Overlap and WriteToFileIndium_Lorentzian save the results of the shift analyses for the overlap method and the Lorentzian fit method respectively. Specifically, WriteToFileIndium_Overlap saves the value of Delta that gives the minimum $\chi^2$ and WriteToFileIndium_Lorentzian saves the shifts for each of the three peaks and the mean of the three shifts. All three of these programs use the command fprintf to write the data to a file.

B.8 Final Shift Analysis

We save all of our data from individual scans without combining any of the results to get our final number that we quote as the result of the experiment. This approach is actually convenient, as it allows us to make many different combinations of data taken in different conditions or analyzed with different methods. Therefore, the code to perform this final analysis is not part of our main set of code. To streamline the process for this analysis, we have one more relevant program, which serves to combine data. This program takes data from the analysis of a certain voltage on a certain day using one method, and rewrites it to another folder, along with data from other voltages or other days. Then this resulting file consists of a larger data array that can be used for making histograms or other analysis.

For the computation of averages or production of histograms, we typically use MATLAB, although the code is simple enough that we do not currently have a specific program for it. On the other hand, for analysis such as looking for trends in the shifts as a function of voltage or other analysis that involves fitting lines or other simple functions, we typically use Mathematica.
Bibliography


