Measurement of $7P_{1/2}$ hyperfine splitting and isotope shift in $^{205}\text{Ti}$ and $^{203}\text{Ti}$

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Abstract

We report a preliminary measurement of the $^{205}$Tl $7P_{1/2}$ hyperfine splitting using a two-step excitation in a hot atomic vapor. We find a significant (20 MHz) discrepancy from a previous published value for this splitting. This result is proof that in the main the experimental apparatus works as expected and measurements of the $^{203}$Tl $7P_{1/2}$ hyperfine splitting and isotope shift should follow soon. Two laser beams, 378 nm and 1301 nm, are overlapped in a heated vapor cell containing both isotopes of thallium. The 378 nm laser frequency is locked with a controlled feedback loop, and can excite $^{203}$Tl, $^{205}$Tl, or both isotopes into the intermediate $7S_{1/2}$ state. The 1301 nm laser frequency is swept across the $7P_{1/2}$ hyperfine levels, and this Doppler-free spectrum is detected with a photodiode and lock-in amplifier. We calibrate the frequency scale with a 500 MHz Fabry-Pérot interferometer and a pair of electro-optic modulators.
Thank you

I would like to thank my advisor, Tiku Majumder, for his great help at every stage of the production of this thesis, as well as his patient indulgence, guidance, and education throughout. I would also like to thank Gambhir Ranjit for his tireless work in the lab, ready assistance, and good spirits; Nathan Schine, for his being such a good role model, his company, and his considerable experimental help along the way; Ward Lopes, for his helpful comments and patience; Kirk Swanson, for the summertime experimental assistance and general enthusiasm; Cole Meisenhelder, for the cheerful help over Winter Study; Michael Taylor, for his unparalleled professionalism; and my family, for everything.
Executive summary

This thesis describes the precise measurement of the $7P_{1/2}$ hyperfine anomaly and isotope shift in a hot thallium vapor. By measuring the hyperfine splitting (HFS) of the state in both isotopes, we can determine the different splitting energy between the isotopes (hyperfine anomaly), as well as the gross shift of energy levels (isotope shift); see Figure 1. Historically, the study of atomic thallium has been motivated by measurements of atomic parity non-conservation and the electric dipole moment, effects that scale rapidly with atomic number [1] [2] [3]. Thallium’s high atomic number ($Z = 81$) and tractable, quasi-hydrogenic atomic theory have made it a prime object of study. The $7P_{1/2}$ HFS has already been measured [5], but there is reason to doubt that measurement’s accuracy, and our preliminary results confirm this suspicion.

The ground state for thallium’s lone valence electron is $6P_{1/2}$. Because of selection rules, we must access the excited $7P_{1/2}$ state in two steps. We use two lasers to excite the $6P_{1/2} \rightarrow 7S_{1/2}$ transition (378 nm) and the $7S_{1/2} \rightarrow 7P_{1/2}$ transition (1301 nm) respectively. The Majumder group has already measured the HFS of the intermediate $7S_{1/2}$ state [8], and successfully implemented the two-step excitation method in a similar indium system [4] [9].

Inside of a heated vapor cell containing $^{205}$Tl and $^{203}$Tl in their natural abundances, we overlap a UV (378 nm) laser locked to the $6P_{1/2}(F = 1) \rightarrow 7S_{1/2}(F' = 1)$ transition with an IR (1301 nm) laser whose frequency is swept across the $7P_{1/2}$ hyperfine lines (see the schematic optical setup shown in Figure 2). In principle, we then detect the absorption of the IR laser with a photodiode, and match the absorption dips to the IR laser frequency. In
Figure 1: Thallium energy levels. Drawing is not to scale. $6P_{1/2}$ and $7P_{1/2}$ isotope shifts are rounded from [5], and $7S_{1/2}$ isotope shift is from [8]. $7P_{1/2}$ HFS are from [5]. I have suppressed the (two) hyperfine levels in the $6P_{1/2}$ and $7S_{1/2}$ states. In the experiment we excite the $(F = 1) \rightarrow (F' = 1)$ transition.

In practice, we only have access to the voltage across a nonlinear device that drives the laser frequency. As a tool to linearize the frequency scale, we split the IR beam, sending the majority of the light to the vapor cell and the rest to a Fabry-Pérot interferometer with 500 MHz free spectral range (FSR). The Fabry-Pérot interferometer produces peaks at evenly spaced frequencies, which gives us enough information to linearize our frequency scale. In principle we could calibrate our frequency scale with the known FSR of the Fabry-Pérot interferometer, but in practice the free spectral range is known to insufficient accuracy at the precision we require. We instead calibrate the frequency scale with an electro-optic modulator (EOM), which effectively produces copies of our signal separated by a well-known frequency.

Furthermore, the locked UV laser only excites a few percent of the atoms in its path to the $7S_{1/2}$ state, and, as a result, the IR absorption signal is overwhelmed by background. Instead of detecting the IR absorption directly, then, we modulate the UV beam with a chopper wheel and feed the output from the photodiode to a lock-in amplifier with the chop frequency as the reference. The lock-in amplifier effectively eliminates the background from our signal, and turns the absorption dips into absorption peaks.
The final complication is that, to measure the isotope shift, we need to observe the spectra of both isotopes simultaneously. From Figure 1 it is clear that the $6P_{1/2}(F = 1) \rightarrow 7S_{1/2}(F' = 1)$ transition energy differs between the isotopes due to the isotope shift and hyperfine anomaly, which makes the simultaneous excitation of zero-velocity atoms of both isotopes impossible. To excite both isotopes, we excite Doppler-shifted atoms of both isotopes into the $7S_{1/2}$ state by locking the UV laser in between the two resonances. By co- and counter-propagating the UV beam with the IR beam, we record spectra Doppler-shifted in opposite directions (see Figure 3), and average these observed frequency splittings to remove the Doppler shift and calculate the isotope shift.

By analyzing our first set of spectra, we make a preliminary measurement for the $^{205}$Tl HFS of $2177 \pm 1$ MHz, which is in significant disagreement with the value $2155.5 \pm 0.6$ MHz from [5], as we suspected. The immediate task is to measure the $^{203}$Tl HFS and the isotope shift and to make an exhaustive account of possible systematic error to confirm the results of this measurement.
Figure 3: Sample dual isotope spectrum in co-and counter-propagating configuration. I have labeled the transitions (isotope, final $7P_{1/2} F$ state, responsible beam). Recall that all transitions are from the $7S_{1/2}(F = 1)$. The $x$-axis ramp voltage can be understood as a frequency axis, though the relationship between the two is somewhat nonlinear. In our analysis we linearize the $x$-axis.
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Chapter 1

Introduction

1.1 Motivation

Precise measurements of fundamental physical quantities drive theoretical physics and prove useful for experimental physics as well. Theoretical descriptions of physical phenomena can only be shown to be consistent at levels of precision achieved by experiment, and so more precise measurements of fundamental atomic constants are tests of the assumptions and approximations that go into building atomic theory.

Precise measurements of atomic constants have also been useful in testing the Standard Model. Since the size of atomic parity non-conservation (PNC) and electric dipole moment (EDM) effects scale rapidly with atomic number ($\sim Z^3$), the measurement of the hyperfine structure of thallium’s two isotopes has been historically motivated by interest in probing PNC and EDM because of thallium’s large atomic number [1][2][3], but also its tractable atomic theory. Though thallium has three valence electrons, two are paired in the 6s orbital, and the atom can be treated, at least to lowest order, as "hydrogenic," with one valence electron (whose ground state is 6p). Furthermore, the sizes of the PNC and EDM effects are proportional to both a fundamental particle physics quantity and an atomic scaling factor, which can be derived from atomic theory. In other words, $\epsilon = Q \cdot C(Z)$, where $\epsilon$ is the size of the effect, $Q$ is the fundamental particle physics quantity and $C(Z)$ is deter-
1.2 Previous work

The $7P_{1/2}$ hyperfine splitting (HFS) is the difference in energy between the $F = 0$ and $F = 1$ sublevels of the $n = 7, L = 1, J = 1/2$ state and the focus of this thesis. It has been measured previously using an atomic beam apparatus [5], though there is reason to suspect that the measurement was subject to systematic error, as the same group’s later corrections of other measurements from the same paper suggest [6][7]. For instance, the HFS of the $7S_{1/2}$ state, which is the intermediate state in our two-step excitation, was measured in [6], but later corrected [7]. It was measured subsequently by the Majumder group [8], and was recently measured with good agreement by Chen et al. using an optical frequency comb [2]. See Table 1.1. Preliminary results from this thesis indicate that the value of the $7P_{1/2}$ HFS in [5] requires similar correction.

Our two-step excitation technique was used successfully in measurement of the $6P_{3/2}$ HFS in indium, a system very much analogous to thallium (two steps in indium:
Section 1.3

410 nm $5P_{1/2} \rightarrow 6S_{1/2}$ and 1291 nm $6S_{1/2} \rightarrow 6P_{3/2}$; in thallium: 378 nm $6P_{1/2} \rightarrow 7S_{1/2}$ and 1301 nm $7S_{1/2} \rightarrow 7P_{1/2}$) [9]. Indium, however, has a nuclear spin of $9/2$, whereas thallium’s nuclear spin is $1/2$, resulting in different numbers of hyperfine levels (see below).

1.3 Hyperfine interaction

Thallium’s ground state electron configuration is [Xe] $4f^{14}5d^{10}6s^{2}6p^{1}$. Thus, while there is not a single valence electron as in an alkali metal, two of the three valence electrons (i.e. $6s^{2}6p^{1}$) are paired in the $6s$ orbital. The $6p$ electron can thus be treated as a single valence electron much like as in hydrogen or any of the other alkali metals. The hyperfine splitting is the result of the interaction of this electron’s spin with the nuclear spin, a so-called spin-spin interaction. Let $I$ be the net nuclear spin, and $J$ be the total electron angular momentum. The dominant term in the Hamiltonian describing the hyperfine interaction is the following perturbation,

$$H_{hf} = a I \cdot J,$$

where $a$ is the hyperfine dipole coupling constant, which depends on details of nuclear shape (i.e. the mean charge radius) and the short-range electron wavefunction ($|\psi(0)|^2$). (For $I > 1/2$, it is possible to have higher order quadrupole, octupole, etc. effects. In indium, for example, $I = 9/2$, and the hyperfine structure is considerably more complex. See [10].)

Since thallium’s net nuclear spin $I$ is $1/2$, and we are not sensitive to higher order moments, we can understand the hyperfine splitting in thallium as we would the hyperfine splitting in hydrogen.

As in hydrogen’s case, we are ultimately interested in the first-order perturbation to the energy of the states, which is given by

$$E_{n}^{1} = \langle \psi_{n}^{0} | H_{hf} | \psi_{n}^{0} \rangle,$$
where $E_n^1$ is the first-order correction to the nth energy level and $\psi_n^0$ is the unperturbed nth-level wavefunction. The result will follow trivially if we can calculate $(I \cdot J) |\psi_n^0\rangle$. Proceeding thus, it is helpful to define the total atomic angular momentum $F \equiv I + J$. Then we can write

$$
(I \cdot J) |\psi_n^0\rangle = \frac{1}{2} (F^2 - I^2 - J^2) |\psi_n^0\rangle.
$$

(1.3)

Since we know $I^2 |\psi_n^0\rangle = J^2 |\psi_n^0\rangle = \frac{3}{4} \hbar^2 |\psi_n^0\rangle$ and $F^2 = 0$ or $2\hbar^2 |\psi_n^0\rangle$,

$$
(I \cdot J) |\psi_n^0\rangle = \hbar^2 |\psi_n^0\rangle \begin{cases} +1/4 \\ -3/4 \end{cases}
$$

(1.4)

Then the energy is perturbed by an amount proportional to the hyperfine coupling constant $a$. The two states are known as the triplet and singlet states.

$$
E_n^1 = a \hbar^2 \begin{cases} +1/4 & \text{triplet;} \\ -3/4 & \text{singlet.} \end{cases}
$$

(1.5)

The result is that the degeneracy of the $J$-levels is broken. The $6P_{1/2}$, $7S_{1/2}$, and $7P_{1/2}$ levels are each split into sublevels with $F = 0$ and $F = 1$. For simplicity, I have drawn in Figure 1.1 below only the splitting of the $7P_{1/2}$ state. (Besides simplicity, depicting the first transition with single levels reflects the experimental procedure. In the experiment, the first step is locked to the $6P_{1/2}(F = 1) \rightarrow 7S_{1/2}(F = 1)$ transition because it is the strongest. In principle, however, we could lock to any of the three transitions, as long as it’s always the same one.) By measuring the HFS, then, we can directly calculate the value of the hyperfine coupling constant $a$. 
1.4 Hyperfine anomaly, isotope shift

The spectra of the two isotopes differ in (1) the size of the HFS and (2) a gross shift of the energy levels. The first effect is known as the hyperfine anomaly; the second is the isotope shift. By measuring the hyperfine splittings of both isotopes we are able to directly measure the hyperfine anomaly and calculate the isotope shift, which is calculated in the absence of the HFS (i.e. once the HFS levels’ energy has been averaged). To calculate the isotope shift, we must measure the HFS of both isotopes simultaneously because the two effects (isotope shift, hyperfine anomaly) both contribute to the observed shift of the energy levels.

Unlike in hydrogen and other light elements, in thallium the isotope shift is due not to the difference in the electron’s reduced mass (less than one part in $10^6$), but to the "field shift" caused by the different distributions of nuclear charge brought about by the presence of the two extra neutrons. In that sense, a precise measurement of the HFS in thallium serves as a subtle probe of thallium’s nuclear structure and the short-range electron wavefunction.

The hyperfine anomaly is also related to the difference in the nuclear charge distributions of the two isotopes (the Breit-Rosenthal-Crawford-Schawlow correction), as well as the
change in the nuclear magnetization (the Bohr-Weisskopf effect). (For an overview, see [11].)
Chapter 2

Experiment

2.1 Method and general apparatus

2.1.1 Laser spectroscopy

The $7P_{1/2}$ state is thallium’s third excited state. Selection rules forbid the direct excitation of the ground state $6P_{1/2}$ electron into the $7P_{1/2}$ state — the angular momentum $L$ does not change. We effect the excitation instead by two steps. First we excite the $6P_{1/2} \rightarrow 7S_{1/2}$ transition with one laser; the next transition ($7S_{1/2} \rightarrow 7P_{1/2}$) we excite with a second laser. We sweep the frequency of the laser exciting the second transition and record the transmitted intensity; as the frequency sweeps across the hyperfine levels, the atoms absorb the light and the transmitted intensity drops. To know the laser frequencies at which these drops in transmitted intensity occur is to know the energy of the HFS.

There are three essential challenges in making an accurate measurement, common to all two-step spectroscopy of hot atoms:

1. Line broadening (Section 2.1.1),

2. Frequency measurement/calibration (Section 2.1.3), and

3. The small signal inherent in the two-step excitation (Section 2.1.3).
As for the first, the absorption lines—the frequencies corresponding to the transition energies—are subject to a variety of broadening mechanisms. Line broadening mechanisms are typically separated into "homogeneous" and "inhomogeneous" varieties, i.e. those that affect each atom in the same way and those that affect each atom differently. I will focus on the two most important: lifetime broadening and Doppler broadening. But there are other broadening effects worth some brief mention: power broadening, transit-time broadening, and pressure broadening. Power broadening arises through excess laser intensity stimulating emission out of the excited state, which effectively shortens the excited state’s lifetime. We ignore this effect because it does not change the line’s shape and the broadening is not large enough to substantially affect our measurement. Transit-time broadening is due to the finite time of the interaction of the laser with the atom. If the atom interacted with the beam indefinitely, it would see the laser’s linewidth; for any finite amount of time, it sees a broader distribution of frequencies. In other words, the Fourier transform of the observed electric field is a delta function only in the case where the atom interacts with the laser forever; if the interaction time is finite, the Fourier transform of the field that interacts with the atom has a nonzero width. At 400 °C, the RMS velocity of a thallium atom is nearly 250 m/s and the beam is only a few millimeters wide; a typical time, then, for a thallium atom to interact with the laser is on the order of 10 microseconds. Since optical frequencies we are concerned with are around $10^{14}$ Hz, the 10 microseconds is a long time, and so we can largely ignore the contribution of transit-time broadening to the width and shape of the profile. Perhaps the most significant of these less important broadening mechanisms, however, is pressure broadening. In heated vapor cell experiments, it is possible to achieve high enough pressures to observe pressure broadening. At sufficiently high pressure, the atoms can become close enough to interact with each other (and/or collide), which perturbs their energy levels. But this broadening results in a Lorentzian profile, so it does not effect the shape of the spectrum. Furthermore, the pressure in our cell is not high enough to see an appreciable amount of pressure broadening; at 700 K, thallium’s vapor pressure is less than 1 Pa. Of these three, power broadening and pressure broadening are homoge-
neous, affecting each atom in the same way; transit-time broadening, however, depends on the speed of each atom, and is thus inhomogeneous.

We focus now on the two broadening mechanisms that play the dominant part in determine the shape of our signal. The first is lifetime broadening, due to the finite lifetime of the atom’s excited states. Every atomic transition is subject to "lifetime-limited" broadening, which sets the so-called natural linewidth of the transition. This is a fundamental limit (hence "natural") on the width of the transition, as fundamental as the Heisenberg Uncertainty Principle. In the time-energy form, it reads,

\[ \Delta E \Delta t \geq \frac{\hbar}{2}, \]  

or, since \( E = \hbar \omega \),

\[ \Delta \omega \Delta t \geq \frac{1}{2}. \]  

The uncertainty in time \( \Delta t \) is related to the lifetime of the state, \( \tau \) (often on the order of nano- or microseconds); the smallest width \( \gamma \sim \Delta \omega \) we can expect for the transition turns out to be \( \frac{1}{2\pi \tau} \). For the thallium \( 7P_{1/2} \) state, the lifetime is 60 ns [3], which translates to a natural linewidth of 2.5 MHz. But the width we observe is limited by the natural width of the \( 7S_{1/2} \) line (\( \tau = 7 \) ns), since any width in frequency there turns into width in frequency for the second transition. The limiting width for us is then \( \tau = \frac{1}{\frac{1}{\tau_f} + \frac{1}{\tau_s}} = 23.8 \) MHz.

In some sense, the natural linewidth is the most important because it is fundamental and homogeneous. The broadening mechanism, however, which is the largest, is Doppler broadening. Doppler broadening is inhomogeneous and derived from the Doppler shift,

\[ f_{\text{obs}} = f_{\text{source}} \left(1 + \frac{v_{\text{atom}}}{c}\right), \]  

where \( v_{\text{atom}} \) is the component of the atom’s velocity parallel to the beam axis, measured positive if moving towards the source (and \( c \) is the speed of light). At any nonzero temperature, the distribution of the atoms’ velocity (in any direction) is given by the Maxwell-
Boltzmann distribution:

\[
P(v) = \sqrt{\frac{m}{2\pi kT}}^3 4\pi v^2 \exp\left(-\frac{mv^2}{2kT}\right) \tag{2.4}
\]

Thus the full width at half maximum (FWHM) is given by

\[
2\sqrt{2\log(2)} \sqrt{\frac{kT}{mc^2} f_0}, \tag{2.5}
\]

where \(k\) is the Boltzmann constant, \(T\) is the temperature, and \(m\) is the mass of the atom. As we sweep the laser frequency, then, the line is broadened by the different Doppler shifts observed by the moving atoms. For thallium atoms at 400 °C, the approximate temperature of our ovens, the FWHM of the first transition (UV) is about 1 GHz; for the second transition, due to its lower frequency, it is about 300 MHz. (Recall that the Doppler shift scales with the frequency of the source \(f_0\).)

Ultimately, then, the shape of the absorption lines we observe is a convolution of a Lorentzian and a Gaussian, known as a Voigt profile. That means it is defined by the following integral,

\[
V(x) = \frac{1}{\sigma \pi \sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left(-x'^2/(2\sigma^2)\right) \frac{\gamma}{(x - x')^2 + \gamma^2} dx' \tag{2.6}
\]

where I have chosen the FWHM of the Lorentzian to be \(\gamma\) and the first integral of the Gaussian to be \(\sigma\) and both to be centered at \(x = 0\). One should imagine this integral as a Lorentzian shape weighted by the appropriate (Gaussian) distribution of Doppler shifts; note that the integration variable \(x'\) determines the Gaussian weight \(\exp(-x'^2/(2\sigma^2))\) corresponding to the center frequency \(x'\) of the Lorentzian piece \(\frac{\gamma}{(x - x')^2 + \gamma^2}\). While the resulting shape has some mixture of the features of the Gaussian and Lorentzian distributions, the Gaussian character dominates at large temperature. If we eliminate the Doppler broadening, then the Lorentzian character dominates. So to a good approximation it is possible for us to deal with the (computationally less demanding) Lorentzian profile, once we have
eliminated the Doppler broadening; and before we eliminate the Doppler-broadening, it is possible to consider the lineshapes as essentially Gaussian. For the sake of comparison, see Figure 2.1.

![Figure 2.1: Comparison of Voigt profile with Gaussian and Lorentzian profiles. I have plotted a Voigt profile with comparable Gaussian and Lorentzian components. For comparison, I have also plotted Lorentzian (on the left, dots) and Gaussian (on the right, dashes) distributions. The Gaussian and Lorentzian have equal FWHM and height. The Voigt profile is scaled to have the same height and comparable FWHM. The Voigt profile has much of the shape of the Gaussian, with the wings more characteristic of the Lorentzian. By changing the relative widths of the Lorentzian and Gaussian components, it is of course possible to produce a Voigt profile of almost entirely Gaussian or Lorentzian character.](image)

We can achieve Doppler-free spectroscopy by using the two-step process (for reference, see Figure 2.4). We excite atoms into the $7P_{1/2}$ state by overlapping both laser beams inside a vapor cell. The vapor cell is heated in an oven to approximately 400 °C, because thallium’s vapor pressure at room temperature is negligible and insufficient to perform the measurement. We detect the transmitted intensity from the second step laser. Only those atoms that interact with both beams will absorb light from the second step laser; we over-
lap the two beams. The tuning of the first step laser picks out a velocity class that will be excited into the $7S_{1/2}$ state; if the first step laser is precisely tuned to the appropriate transition, then it is the zero-velocity class that is excited. In any event, the second step laser will only interact with one velocity class of atoms, and so the only Doppler broadening that remains is an artifact of the beams’ nonzero divergence, and the remaining finite width is due largely to the homogeneous natural linewidth.

The two-step excitation conveniently eliminates the Doppler broadening and circumvents the forbidden direct transition from $6P_{1/2} \rightarrow 7P_{1/2}$. But it creates a problem as well: the signal size is small (compared to noise). By picking out the zero-velocity class of atoms, the first step can only excite a maximum of approximately

$$\frac{\Gamma_{\text{Natural}}}{\Gamma_{\text{Doppler}}} \approx \frac{20 \text{ MHz}}{400 \text{ MHz}} = 5\% \quad (2.7)$$

of the available atoms. Thus the signal from the second step laser, i.e. the signal we detect, is small enough that we cannot detect it directly; instead, we modulate the signal with a chopper wheel and detect it with a lock-in amplifier. For more on the detection scheme, see Section 2.1.3.

### 2.1.2 Diode laser, principle and operation

The two step excitation is achieved by two diode lasers (378 nm and 1301 nm). The ultraviolet (UV) 378 nm laser excites the first transition ($6P_{1/2} \rightarrow 7S_{1/2}$), and the infrared (IR) 1301 nm laser excites the second ($7S_{1/2} \rightarrow 7P_{1/2}$). Both lasers are diode lasers stabilized by an external cavity in the so-called Littrow configuration.

The gain medium in the laser is a semiconductor (IR: InGaAs, a well-known laser diode material; UV: GaN, a byproduct of the recent emergence of Blu-Ray™) diode whose bandgap corresponds to an optical frequency close to the desired wavelength of the laser. The semiconductor has two quasi-continuous bands of energy levels, the valence band and the conduction band, separated by the "bandgap." The diode is supplied with power
Figure 2.2: Laser schematic. Spontaneously emitted light from the diode is highly divergent, and collimated with a short-focal-length lens. The diffraction grating is aligned so that the first-order diffraction is directed back into the diode, whose back wall is reflective. The frequency of this light stimulating emission in the diode is determined by the angle of the diffraction grating, which can be finely adjusted by applying a voltage to the PZT mounted behind the grating.

(current), which brings charge carriers from the lower-energy valence band into the conduction band. Electron-hole recombination produces photons; the result is spontaneous emission over a broad range of frequencies, including the desired laser frequency.

The spontaneous emission diffracts off a grating, which is aligned so that the first order diffraction is directed back onto the diode (see Figure 2.2). The other end of the optical cavity is the reflective back face of the diode. In the IR laser, the front face has an anti-reflective coating; in the UV laser, the external cavity modes compete with the modes of the cavity set by the diode. This means that the UV laser is not as good at sweeping its frequency stably, but since we lock the UV laser to one frequency, the poor tuning behavior is not problematic. Different frequencies diffract at different angles, and so, by controlling the angle of the grating, one can control the frequency of light that returns to the cavity, stimulates emission, and is amplified. We achieve this control with a small piezoelectric transducer (PZT) mounted behind the grating, which expands and contracts with a voltage
across it. The laser’s behavior is ultimately determined by the angle of the grating, the length of the cavity, and the gain response of the diode. The first is controlled by the voltage supplied to the PZT; the other two are determined by the temperature of the apparatus and the current supplied to the diode. Changing the current and temperature amounts to (in part) changing the index of refraction of the diode, and so we are able to tune the laser frequency that way. But all fine tuning of the laser frequency is done by tuning the PZT voltage; to sweep the frequency of the IR laser, we simply sweep the voltage across the PZT.

The largest problem the laser can experience while scanning is a modehop. As we sweep the PZT voltage one way, the "lasing" cavity mode passes across the background of the diode’s gain profile. This lasing mode is the mode that produces the best response in the diode. As we tune the lasing mode, we tune the other cavity modes as well. And if one of the other cavity modes can produce a comparable response from the diode, the two modes compete and the laser’s output is no longer in one mode. When the laser jumps to this other competing mode, we say that the laser has experienced a modehop. The IR laser frequency is broadly tunable over approximately 1280 nm - 1330 nm, and we are able to achieve modehop-free fine tuning over more than 8 GHz. As I mentioned, the UV laser does not tune as well: though the diode is temperature-controlled, fixing its length, competition from the two cavities results in a scanning range of about 2 GHz.

Finally, there is one further complication in tuning the laser frequency: the response of the PZT to voltage is nonlinear, which is one of the central issues of our data analysis (see Section 3.2).

2.1.3 Detection scheme

In order to perform the measurement, there are three essential challenges: first, to eliminate the Doppler broadening so that the measured linewidth is comparable to the natural linewidth; second, to identify the line’s center frequency; third, to detect the weak signal. The previous sections have described the general methods and tools with which we
Section 2.1

approach the first challenge. This section describes the primary tools for the second and third.

**Fabry-Pérot interferometer**

Before the IR beam is directed into the cell, a small portion of the beam is directed into a Fabry-Pérot interferometer (see Figure 2.3a). Two spherical mirrors are arranged in a confocal configuration. If the wavelength $\lambda$ of the incident light is resonant with the cavity (if $4r/\lambda$ is an integer), then there is constructive interference and the photodiode records a peak in transmission. Finding the expression for the transmitted intensity is simply a

![Confocal Fabry-Pérot interferometer](image)

(a) Confocal Fabry-Pérot interferometer. Off-axis light, incident from the left, travels a distance that is an integer multiple of $4r$, $r$ the focal length of the spherical mirrors, before being detected at the photodiode on the right. In other words, the free spectral range of the cavity, the difference in frequency between two adjacent peaks as the frequency of the incident beam is swept (see Figure 2.3b), is $4r$. The radius of curvature of the mirrors we use is 15 cm, and so the FSR of our cavity is 500 MHz.

![Simulated plot of normalized transmitted intensity](image)

(b) Simulated plot of normalized transmitted intensity for a Fabry-Pérot interferometer with FSR 500 MHz. I have plotted spectra for three different $R$, $R = 0.90$ (solid), $R = 0.40$ (dashed), and $R = 0.10$ (dotted). Starting at some arbitrary zero-frequency, we sweep the incident beam’s frequency. There are clearly peaks centered on integer multiples of 500 MHz (where all the intensity is transmitted), with troughs out of phase by 250 MHz. Manipulating $R$ affects the peaks’ width and depth.

Figure 2.3: Fabry-Pérot (a) cavity and (b) spectrum.

question of summing the phases accumulated through reflection and distance traveled for
each trip through the interferometer. The result is

\[
\text{transmitted intensity} = \frac{1}{1 + 4R\sin^2(\delta/2)\frac{c^2}{(1-R)c^2}}
\]  

(2.8)

where \(\delta\) is the phase gained during one round trip through the interferometer and \(R\) is the reflectivity of the mirrors, which is assumed to be constant over the frequency range in question (a valid assumption as long as we do not change the frequency too drastically; over the range which we scan the IR laser–5 GHz or so–it is entirely valid). By inspection, it is clearly must be the case that \(\delta \sim r/\lambda\), where \(r\) is the focal length of the mirrors (= the length of the cavity) and \(\lambda\) is the wavelength of the incident light. Thus sweeping the frequency and sweeping the length of the cavity produce a similar effect. Figure 2.3b demonstrates the signal one observes when sweeping the frequency of the incident light in Figure 2.3a and reveals the Fabry-Pérot interferometer’s most important property: the peaks in transmission one observes if one sweeps the frequency of the incident light are evenly spaced at a reasonably well-known interval \((c/4r)\). In a literal sense, then, the Fabry-Pérot spectrum we record is a ruler for measuring the HFS.

EOM

The canonical example of systematic error is the scientist who measures all his lengths wrong because his yardstick is not a yard long. The measurements are reproducible (in the same lab) but inaccurate. This scientist might improve the experiment by comparing to a different yardstick. We find ourselves in precisely this situation. We are using the Fabry-Pérot interferometer as a ruler—but, used alone, we have no way of knowing whether the ruler is accurate at the level of precision we require. Since FSR (the ticks on the ruler) is set by the geometry, namely the focal length of the mirror / separation of the mirrors, it is beholden to the precision with which the mirrors were ground and the interferometer aligned. Furthermore, the geometry is unknown to us; we cannot physically measure
the cavity length with sufficient precision. Like the scientist with the faulty yardstick, we introduce another ruler: the electro-optic modulator (EOM).

The EOM contains a crystal whose index of refraction depends on the voltage supplied to it. (It is analogous to the acousto-optic modulator, which contains a material whose index of refraction depends on the pressure applied to it.) We apply a voltage to the crystal that changes sinusoidally with frequency $\omega$. This adds a sinusoidally varying phase to our laser, which transforms our laser output

$$A e^{i\omega t} \rightarrow A e^{i(\omega t + \beta \sin \omega_m t)},$$

where $\omega_m$ is the modulation frequency and $\beta$ is a measure of how much power is being sent into the modulation. This expression can be expanded in terms of Bessel functions in $\beta$:

$$A e^{i(\omega t + \beta \sin \omega_m t)} = A e^{i\omega t} (J_0(\beta) + \sum_{k=1}^{\infty} J_k(\beta) e^{ik\omega_m t} + \sum_{k=1}^{\infty} (-1)^k J_k(\beta) e^{-ik\omega_m t}),$$

which gives an infinite chain of sidebands. For sufficiently low $\beta$, however, we can approximate the result as a carrier with one set of added sidebands separated in frequency $\pm \omega_m$.

The service the EOM provides is that we can supply a very precisely known frequency to it (known to better than one part in $10^6$), and then compare the frequency-separation of the sidebands in the atomic spectrum (precisely known) to the frequency-separation we calculate from counting the peaks on the Fabry-Pérot signal.

**Lock-in amplifier**

Our signal is easily overwhelmed by noise because we address such a small group of atoms (the zero-velocity class) with the two-step excitation. Only a few percent of the atoms that interact with the first beam are available to be excited into the second state, so we expect a small second step absorption. To find a signal and improve the signal-noise ratio, we chop the UV beam and use a lock-in amplifier (with the chop frequency as the reference).
A more complete description of its use can be found in [14]. Here suffice it to say that it is a piece of electronics that performs the integral

\[ A_{\text{out}}(t) = \frac{1}{\tau} \int_{t-\tau}^{t} \sin(2\pi f_{\text{ref}} t' + \phi) \, A_{\text{in}}(t') \, dt', \]  

(2.11)

where \( A_{\text{out}} \) is the output from the lock-in, \( \tau \) is the time constant (variable), \( f_{\text{ref}} \) is the reference frequency (set by the chopper), \( \phi \) is the phase (variable), and \( A_{\text{in}}(t) \) is the input signal (with noise), a function of time. The principle is that any part of \( A_{\text{in}}(t) \) that is not oscillating at \( f_{\text{ref}} \) will average to zero as it integrates over time; the upshot is that all the background is eliminated (no appreciable amount is oscillating at the reference frequency) and almost all noise as well. The lock-in produces, in short, a signal with zero background; instead of recording mostly background with (minuscule) dips in intensity due to the absorption lines we are interested in, we record peaks on a zero-background signal.

### 2.2 Experimental challenges and apparatus details

The most significant experimental requirement is the stability of the frequency of the UV laser. Since the energy of the transition we are probing is determined by the sum of the UV and IR lasers, any uncertainty due to instability of the UV laser is directly related to uncertainty in the frequency of the IR laser and the measurement.

#### 2.2.1 Locking system

The frequency of the UV laser must remain constant, i.e. "locked". To lock the frequency of the laser, we want a signal that is odd with respect to the desired frequency—at the very least, the sign of this "lock signal" must change as the frequency passes through the lock point. Furthermore, it is desirable that the slope of the lock signal be as linear (and large) as possible. Then, with the appropriate electronics, all we need is to amplify the lock signal, adjust the gain, possibly invert the sign, and feed it back to the PZT controlling the frequency of the locked laser. Luckily the signal out of the photodiode is a DC voltage.
Figure 2.4: Schematic optical setup. Light from two lasers is overlapped in the experiment oven, which contains a quartz vapor cell containing a mixture of $^{203}\text{Tl}$ and $^{205}\text{Tl}$ in their natural abundances. As we sweep the frequency of the IR beam, it is detected at a photodiode, whose output is sent to a lock-in amplifier, whose reference is the chopper wheel modulating the UV beam. The AOM produces two sidebands on the UV beam at $\pm \omega_{\text{AOM}}$, which can be spatially resolved. They are sent to the locking system (see details of the locking system below). The main beam (zero-order out of the AOM) proceeds to the experiment, modulated by the chopper wheel. A 50/50 beamsplitter (marked as 50/50 BS above) and shutters (not pictured) determine whether UV light co- or counter-propagating with the IR beam (or both) will reach the experiment cell. The IR beam is split into two parts. One part is sent into a Fabry-Pérot interferometer (discussed above); the other is sent to the experiment cell via the EOM. (The $\lambda/2$-plates are to ensure that the light incident on the EOM is s-polarized and, with the polarizing beamsplitter—PBS above—control how much light is sent to the Fabry-Pérot.) After the IR beam passes through the experiment cell (collinear with the UV beam), the IR beam is separated from the UV beam with a dichroic mirror and detected at the photodiode, whose output is sent to a lock-in amplifier. The output from the Fabry-Pérot photodiode and lock-in amplifier are sent to the LabVIEW virtual instrument (VI) that we use to record data.
But since we want to lock the UV laser to the $6P_{1/2}(F = 1) \rightarrow 7S_{1/2}(F' = 1)$ transition in each isotope separately (and both at the same time—see below), the signal from the photodiode is (essentially) a Gaussian centered on the frequency of the transition, and thus even about the lock point. If the laser drifts from the lock point, the even function provides no information about the direction (since we are not directly sensitive to the slope of the signal), and so it is impossible to use that signal to correct the frequency. Instead we, in some sense, take the derivative of that even function, which produces an odd function. We use an acousto-optic modulator (AOM) to produce two sidebands at $\pm 250$ MHz. These two beams’ polarizations are made perpendicular by $\lambda/2$-plates and a polarizing beamsplitter (PBS), and then they propagate through a supplementary heated thallium vapor cell. On the other side, the two beams are separated with another PBS, and sent to the two ports of
Figure 2.6: The production of the difference signal. I have used $T = 800$ kelvin. The dashed absorption lines are the detuned spectra that are subtracted to produce the solid difference signal below. The vertical dashed line ($\sim \text{Frequency} = 0$) marks the 203 lock point, where zero-velocity $^{203}\text{Tl}$ atoms are excited to the $7S_{1/2}$ state; the dashed vertical line marks the 205 lock point; and the solid vertical line marks the dual isotope lock point.

a difference photodiode. This is similar to taking a derivative of the signal with respect to frequency (in the case where $\Delta f_{\text{AOM}} < \text{absorption linewidth}$). Thus we produce a lock signal (see Figure 2.6) that is odd about our desired lock points, where the slope is sufficiently constant and large.

We send the lock signal to a PID controller (proportional-integral-differential). The PID box is designed for this kind of closed-loop feedback. It is takes the signal $\epsilon$ and returns

$$P \times (\epsilon + I \int_{t-\tau}^{t} \epsilon \, dt + D \frac{d\epsilon}{dt}) + \text{Offset}, \quad (2.12)$$

where $P$, $I$, and $D$ are adjustable gains, Offset is an adjustable voltage, and $\tau$ is some characteristic time constant. The motivation for this particular machine is that a simple proportional control (essentially, an amplifier) could in principle work to turn a lock signal into a correction signal. But the correction signal and thus frequency would have a tendency to oscillate—there is nothing to damp the feedback. Heuristically, the $I$ term damps out...
the oscillations. When the signal $\epsilon$ is not large, the integral is smaller and the PID pushes back less. (We set $D = 0$. In other systems, the case may arise where one would want a stronger response to a bigger push. But for us, the $D$ term only adds instability. We expect our signal to drift slowly (due to thermal changes or the operating drift of the laser diode); quick changes are often transients or mechanical impulses that die away quickly, and there is no reason to amplify them.) To find near-optimal $P, I, and D$ values, we use the Ziegler-Nichols tuning method, an established heuristic method for tuning such feedback loops, which is convenient if not optimal [12].

### 2.2.2 Lock points and optimization

The difference signal (see Figure 2.6) is odd about the two lock points corresponding to the $^{203}$Tl and $^{205}$Tl lines. But there is another possible lock point in between the other two. The atoms excited by a laser locked at that frequency will be Doppler shifted by precisely the frequency by which the laser is detuned from either isotope’s individual lock point. Now, the shape of the lock signal (= the difference signal $\propto$ the error signal) is determined by the temperature of the oven. Qualitatively, a cold temperature produces an insufficient density of atom vapor, and so the optical depth is low. A hot temperature produces saturation, which washes out the locking feature’s shape. The explanation of how this happens is in Appendix B. The real qualitative upshot of the calculation is that the temperature is optimal for the different lock points at different temperatures, ranging over about 30 degrees near 400 °C; see Figure 2.7.

### 2.2.3 Lock performance

If the laser tunes smoothly and there are no large disturbances, the lock should work indefinitely. If the laser drifts into a modehop, however, the PZT hysteresis results in the error signal getting amplified enough that recovery is impossible. Generally, if the PID controller produces a voltage larger than the peak on the difference signal, the laser becomes unlocked. A large disturbance (e.g. shaking the table) can produce this effect. In
Figure 2.7: Optimization of lock temperature. Zero-crossing slope is plotted as a function of temperature for the three different lock points (203 lock point is dotted; 205 is dashed; dual isotope is solid). The three lock points are optimized at different temperatures, ranging from about 690 K to 715 K. (The sign of the dual isotope plot has been switched for comparison’s sake.)

The long run, modehops are more common than such disturbances. Recall that, as the laser runs, the frequency slowly changes. To a great extent this can be mitigated by slowly ramping the PZT voltage (which is what the lock does); but if the laser drifts into a modehop, the lock breaks. The lock is essentially immune to long-term drift, however, so the laser can stay locked for hours at a time. Figure 2.8 shows the behavior of the laser (via the error signal), locked and unlocked, on a relatively short time scale; the lock data spans about a minute, and the unlocked data spans about ten minutes.

2.2.4 Dual isotope excitation and averaging out the Doppler shift

As is evident from the energy level diagram (Figure 1.1), we cannot excite both isotopes simultaneously with one laser unless we excite Doppler-shifted atoms. The
(a) The laser is left locked for a minute and the error signal (the difference signal, see Figure 2.6) is recorded.

(b) Histogram of data from Figure 2.8a.

(c) The free-running laser. Over time, the laser’s frequency changes without external perturbation, though fast changes in the frequency are likely to be due to mechanical coupling to the environment. Though the vertical scale is the same, note that the time scale here is about 10 times larger than the scale in Figure 2.8a.

(d) Histogram of data from Figure 2.8c. The horizontal scale and bin size are the same as in Figure 2.8b.

Figure 2.8: Characterization of lock and laser drift. Note that the time scales are different between the unlocked plot and the locked plot. While locked, the laser "error signal," measured in volts, is essentially 0. As the laser drifts, it drifts away from the zero point (see the difference signal in Figure 2.6). In terms of frequency, the locked laser is locked to a width of 1 MHz, whereas the unlocked laser drifts around on the order of 100 MHz.
differential locking signal (Figure 2.6) gives us this ability. We can use the relatively unsophisticated plot in Figure 2.6 to find that there are three lock points to choose from, as indicated on Figure 2.6, exciting either

1. Zero-velocity $^{203}$Tl atoms (with trace amounts of highly Doppler-shifted $^{205}$Tl atoms,

2. Zero-velocity $^{205}$Tl atoms (with trace amounts of highly Doppler-shifted $^{203}$Tl atoms, or

3. Atoms of both isotopes, each Doppler-shifted.

At the zero-crossing between the two isotopes’ resonances, all $^{203}$Tl atoms excited by the beam need to be moving parallel to the laser, since they need to see a red-shifted beam to be in resonance by $\sim 755$ MHz. Similarly, all excited $^{205}$Tl atoms see to see blue-shifted light (by $\sim 880$ MHz) to be in resonance, so they will all be moving toward the laser. (Since the Doppler shift is proportional to the source frequency $f_0$, it is simple to calculate these Doppler shifts in the IR spectrum by using the ratio of the two laser frequencies, or the inverse ratio of the two laser wavelengths. These velocities correspond to Doppler shifts of $(\frac{378 \text{ nm}}{1301 \text{ nm}}) 755 \text{ MHz} = 219 \text{ MHz}$ and $(\frac{378 \text{ nm}}{1301 \text{ nm}}) 880 \text{ MHz} = 255 \text{ MHz}$ in the IR spectrum.) But now we co-propagate the IR beam with the UV beam. Clearly all atoms who saw a red-shifted beam will still see a red-shifted beam (to an excellent approximation the absorption of the two photons is simultaneous—the lifetime of the $7S_{1/2}$ state is less than 10 ns); and the same for atoms resonant with a blue-shifted beam. Then we counter-propagate the two beams. Now all atoms that see a red-shift from the UV beam see a blue-shift from the IR beam, and vice versa (see Figure 2.9. What’s more, the switch of red- to blue-shift is a switch of sign; the magnitude of the shift does not change (see Figure 2.9 and Figure 2.12). Then it is simply a matter of averaging to eliminate the Doppler shift.

**The need for accurate alignment**

In averaging out the Doppler shift we depend on the beams’ precise alignment. To understand why, consider the case where the first beam is exactly resonant with the $^{205}$Tl
transition. The first beam selects an axis along which the atoms’ velocity is zero. We may freely call this the $x$-axis. If the other beam is exactly collinear with the $x$-axis (Figure 2.10a), then the atoms are still motionless with respect to the beam and see no Doppler shift (neglecting the second-order relativistic Doppler shift since the atoms’ speed is around 250 m/s). But if the beams are misaligned by an angle $\theta$ (Figure 2.10b), this does not hold. The two beams determine a plane, call it the $xy$-plane. The first laser beam guarantees that the $x$-component of the atoms’ velocity, $v_x$, is zero, but (as long as we only care about the first-order Doppler shift), it is possible that $v_y$ is quite large. The projection of $v_y$ onto the misaligned beam’s axis is obviously $v_y \sin(\theta)$. Thus the atom will experience a Doppler shift of

$$
\Delta f = (1 - \frac{v \sin(\theta)}{c})f_0, \quad (2.13)
$$
(a) The beams are perfectly overlapped. If we let the $x$-axis be the beam axis, then we can say that only the class of atoms with velocity component $v_x = 0$ interacts with both beams.

(b) The beams are overlapped, but misaligned by an angle $\theta$. Now it is not just the $v_x = 0$ velocity class that interacts with both beams.

Figure 2.10: Beam alignment.

where $f_0$ is the laser frequency. Our beams have a maximum possible angle between them of about

$$\theta = \frac{r_{arc}}{R_{oven}} \approx \frac{4 \text{ mm}}{75 \text{ cm}} \approx 0.005 \text{ radians}. \quad (2.14)$$

Using an RMS-velocity atom (250 m/s) and the frequency of the IR laser ($\sim 200,000$ GHz), we get a negligible Doppler shift (one part in 10 million $\sim 10$ kHz).

2.3 Simulated spectra

2.3.1 The signal’s functional form

The shape of our signal is a convolution of a Gaussian and a Lorentzian. However, since we almost entirely eliminate the Doppler broadening with the two-step excitation, the character of each absorption line is a Lorentzian $L(f)$. Suppose we are looking for just this one Lorentzian. The photodiode records the transmitted intensity, $I(f) = I_0 e^{-\alpha L(f)}$, where $\alpha$ is the optical depth. Since such a small number of the atoms can be excited by the second step laser (see Section 2.1.1 and Equation 2.7), the signal is very small. To first order, then, the photodiode records

$$I(f) \approx I_0(1 - \alpha L(f)). \quad (2.15)$$
But we detect this signal with the lock-in amplifier (see Section 2.1.3), which looks for the
signal that is modulated at the reference frequency, removing the large background term;
to a good approximation, we have

\[ \text{Signal}(f) = I_0(1 - \alpha L(f)) - I_0, = -\alpha I_0 L(f), \]  \hspace{1cm} (2.16)

and so our signal is just a Lorentzian. Since the feature we are studying is made up of
several absorption lines, we find our signal to be a sum of Lorentzians, and fit our data
accordingly.

### 2.3.2 Spectra

With the functional form of the absorption features in hand, it is easy to simulate the spec-
tra we expect to record when we lock the UV laser and sweep the frequency of the IR laser.
The eventual goal is to produce a spectrum from which we can rewrite the energy level
diagram in Figure 1.1. Let us turn to a similar diagram, and define the following frequency
intervals, starting from the $7S_{1/2}$ state.

![Diagram showing $7S_{1/2} \rightarrow 7P_{1/2}$ transitions in thallium.]

Referring to Figure 2.11, we have $H_{203}$ as the $^{203}\text{Tl} 7P_{1/2}$ HFS, $H_{205}$ the $^{205}\text{Tl} 7P_{1/2}$
HFS. $G_{203}$ is the $^{203}\text{Tl} 7S_{1/2}$ HFS, $G_{205}$ the $^{205}\text{Tl} 7S_{1/2}$ HFS. The frequency $f_0$ is the frequency
of the $^{203}\text{Tl} 7S_{1/2} \rightarrow 7P_{1/2}$ in the absence of isotope shift; $f'_0$ is the same transition in $^{205}\text{Tl}$. 
Clearly, then, we have

$$I_0 = f_0 - f'_0,$$  
(2.17)

where $I_0$ is the transition isotope shift (TIS). Since we know the $7S_{1/2}$ level isotope shift (LIS) from reference [8], we can calculate the $7P_{1/2}$ LIS trivially.

In the experiment, we excite four transitions from the $7S_{1/2}(F = 1)$ state, corresponding to frequencies $a, b, A,$ and $B$ as marked in Figure 2.11. These correspond to the following transitions, organized from red to blue:

\begin{align*}
A & \implies 7S_{1/2}(F = 1) \rightarrow 7P_{1/2}(F' = 0) \quad ^{205}\text{Tl} \\
a & \implies 7S_{1/2}(F = 1) \rightarrow 7P_{1/2}(F' = 0) \quad ^{203}\text{Tl} \\
B & \implies 7S_{1/2}(F = 1) \rightarrow 7P_{1/2}(F' = 1) \quad ^{205}\text{Tl} \\
b & \implies 7S_{1/2}(F = 1) \rightarrow 7P_{1/2}(F' = 1) \quad ^{203}\text{Tl}.
\end{align*}

It is a question of arithmetic to write the frequencies in terms of these frequencies we have defined.

\begin{align*}
A &= (f_0 - I_0) - \frac{1}{4} G_{205} - \frac{3}{4} H_{205} \quad (2.18) \\
a &= f_0 - \frac{1}{4} G_{203} - \frac{3}{4} H_{205} \quad (2.19) \\
B &= (f_0 - I_0) - \frac{1}{4} G_{205} + \frac{1}{4} H_{205} \quad (2.20) \\
b &= f_0 - \frac{1}{4} G_{203} + \frac{1}{4} H_{203} \quad (2.21)
\end{align*}

To plot, however, it is convenient to add an offset. Since we only care about differences, it does not matter if I add the prudently chosen quantity $\left(\frac{1}{4} G_{205} + I_0 - f_0\right)$ to all the terms,
simplifying the preceding quantities to

\[ A = -\frac{3}{4} H_{205} \quad (2.22) \]
\[ a = I_0 + \frac{1}{4} (G_{205} - G_{203}) - \frac{3}{4} H_{203} \quad (2.23) \]
\[ B = \frac{1}{4} H_{205} \quad (2.24) \]
\[ b = I_0 + \frac{1}{4} (G_{205} - G_{203}) + \frac{1}{4} H_{203}. \quad (2.25) \]

Then it is clear that we can extract the hyperfine anomaly from the measurement of the hyperfine splittings \( H_{203} \) and \( H_{205} \). Unfortunately, however, we cannot observe this spectrum directly. If this were a ground state transition, and there were no need for the two step excitation, we would be able to deal with zero-velocity class atoms of both isotopes, and then we would be able to directly transcribe the information in Figure 2.11 into a sum of Lorentzian absorption lines. We would then observe a spectra such as the one in Figure 2.12a. In reality, we have three lock points available to us, enabling us to either excite (1) zero-velocity \(^{203}\text{Tl}\) atoms (the solid absorption peaks in Figure 2.12a), (2) zero-velocity \(^{205}\text{Tl}\) atoms (the dashed absorption peaks), or (3) Doppler-shifted atoms of both isotopes (see for instance Figure 2.12 and the discussion in Section 3.3). Thus Figure 2.12a could be said to be the \(^{203}\text{Tl}\) spectrum (as if we were locked to lock point 1) overlaid with the \(^{205}\text{Tl}\) spectrum (as if we were locked to lock point 2). If we lock to lock point 3, we can record either the spectrum in Figure 2.12b or Figure 2.12c, depending on whether the two beams are co-propagating or counter-propagating. From these spectra, we can then extract the hyperfine anomaly and the isotope shift. In other words, we record spectra where we see
the following:

\[
A = -\frac{3}{4}H_{205} \pm \Delta_{205} \quad (2.26)
\]

\[
a = I_0 + \frac{1}{4}(G_{205} - G_{203}) - \frac{3}{4}H_{203} \pm \Delta_{203} \quad (2.27)
\]

\[
B = \frac{1}{4}H_{205} \pm \Delta_{205} \quad (2.28)
\]

\[
b = I_0 + \frac{1}{4}(G_{205} - G_{203}) + \frac{1}{4}H_{203} \pm \Delta_{203}, \quad (2.29)
\]

where the top case corresponds to the co-propagating configuration and the bottom case corresponds to the counter-propagating configuration. On our treatment of the Doppler-shifted spectra, see Figure 2.12b and 2.12c and Section 2.2.4.
(a) Simulated spectra overlaid. The $^{203}\text{Tl}$ spectrum is solid and the $^{205}\text{Tl}$ is dashed. We will never acquire this spectrum directly.

(b) Dual isotope excitation in co-propagating configuration. The UV laser and IR laser are co-propagating, exciting atoms Doppler-shifted, say, in the positive direction along the beam axis. As above, the $^{203}\text{Tl}$ spectrum is solid and the $^{205}\text{Tl}$ spectrum is dashed.

(c) Dual isotope excitation in counter-propagating configuration. The UV laser and IR laser are counter-propagating, exciting atoms Doppler-shifted in the negative direction along the beam axis.

Figure 2.12: Simulated $^{203}\text{Tl}$ and $^{205}\text{Tl}$ spectra. In (a) the two zero-velocity spectra are overlaid, which would be ideal, but is in fact impossible. Instead, we record Doppler-shifted spectra, in (b) the co-propagating configuration and (c) the counter-propagating configuration. (N.B. The relative heights of the peaks are qualitatively correct, but no more, based on an estimation from the relative isotopic abundances and different quantum mechanical line strengths for the $F = 1 \rightarrow F' = 0$ and $F = 1 \rightarrow F' = 1$ transitions. The experimental heights of the peaks depends on details of the beams’ polarizations and the UV beam’s detuning from resonance. The center frequencies and widths reflect, however, values for lifetimes and level and transition energies given in references [2] [3] [5]. The features are simulated as sums of Lorentzians, as motivated in Section 2.3.1.)
Chapter 3

Data and analysis

3.1 Data collection procedure

For a run of the experiment, the UV laser is locked to one of the three lock points, so that (practically) only zero-velocity $^{203}$Tl atoms are excited (“the 203 lock point”), only zero-velocity $^{205}$Tl atoms are excited (“the 205 lock point”), or Doppler-shifted velocity classes of both isotopes are excited (“the dual isotope lock point”); see Section 2.2.2 for details. Once that is done, all experimental input and output is driven and collected by a LabVIEW Virtual Instrument (VI). For input, the VI creates a voltage ramp that is sent to the PZT in the IR laser (to sweep its frequency). The voltage at two photodiodes is recorded, one downstream of the experiment cell to detect the absorbed IR beam and the other split off upstream of the EOM and downstream of the Fabry-Pérot interferometer that detects the peaks in transmitted intensity as the frequency sweep progresses (see Figure 2.4 for the optical setup; Section 2.1.3 for the Fabry-Pérot interferometer).

The LabVIEW VI steps the IR PZT voltage, waits, reads the photodiode voltages, and then repeats. (The wait is included because the arm on which the diffraction grating is mounted has some amount of inertia. When the PZT pushes the grating, it does not stop immediately. The wait gives the diffraction grating time to stop.) Each sweep (up or down) takes a few seconds. The resulting output is a list of PZT voltages, a list of photodiode volt-
ages from the Fabry-Pérot interferometer, and a list of voltages from the lock-in amplifier. We typically collect data sets of a few hundred complete frequency sweeps (~ 2 hours of data collection) before changing an experimental parameter (e.g. EOM modulation frequency, oven temperature).

There can be systematic errors associated with hysteresis in the laser diode, particularly if the laser is not operating in a single longitudinal mode [13]. To be sensitive to that kind of systematic error, we separate up-sweeps from down-sweeps and analyze those results separately. The results should be identical if the laser is operating ideally. Similarly, we collect data with the UV beam alternately co- and counter-propagating with the IR beam. The choice of direction is controlled by shutters, which are driven by logic pulses from the VI. For single isotope excitation, when we are locked to the 203 lock point or the 205 lock point, the atoms excited are at zero velocity (along the beam axis), and so analysis of the co- and counter-propagating data should produce identical results. The comparison of these results is another method we have of detecting systematic errors with our methods of data collection.

3.2 Calibration and linearization of frequency sweep

The first step of our data analysis is the linearization and calibration of our frequency scale. It is obvious that any error we accumulate in these two steps is carried forward directly into our measurements. For that reason, we have several methods of calibration that we can compare to each other.

Linearization

Due to nonlinearities inherent in the PZT, the Fabry-Pérot spectra we collect, where transmission is a function of PZT voltage (as opposed to frequency), do not have evenly spaced peaks as in the simulation in Figure 2.3b. Instead, they get bunched up toward one end of the sweep, as in Figure 3.1a. While the peaks are spaced unevenly in PZT voltage, however,
we know they are spaced evenly in laser frequency. So the first part of our data analysis is to transform our Fabry-Pérot signal from voltage-space to frequency-space.

To linearize, we fit the Fabry-Pérot peak centers (voltages) to a fourth-order polynomial. The peak centers are evenly spaced by a reasonably well-known frequency. It is trivial to then establish the function \( \text{frequency} = f(\text{PZT voltage}) \) and transform the list of PZT voltages into a list of frequencies. The Fabry-Pérot transmission peaks should be evenly spaced along this new frequency axis.

![Graph](image)

(a) Typical Fabry-Pérot spectrum. The \( x \)-axis is scaled PZT voltage. The \( y \)-axis is transmitted intensity. Close examination reveals that the peak centers are spaced unevenly. (The data points are fitted by a sum of Lorentzians.)

(b) Least-squares fit of Fabry-Pérot spectrum peak centers to a fourth-order polynomial. Each point is a peak location; the line joining them is the fitted polynomial. The \( x \)-axis is scaled PZT voltage; the \( y \)-axis is frequency. I have also plotted a line joining the first two points to demonstrate the deviation from linearity.

Figure 3.1: Linearization of the frequency scale. To scale the \( x \)-axis, we start with the raw PZT voltage, which is essentially a list of point numbers \( j \). Then we scale \( x_j \) so that it runs from \(-1\) to \(1\). In other words, we take bin numbers \( j \) and transform \( x_j = \frac{j - N/2}{N/2} \), where \( N \) is the total number of points. This rescaling is convenient for our polynomial fits, which are typically centered at \( x_j = 0 \). We fit to a fourth-order polynomial.

**Calibration**

As I mentioned earlier, we know the FSR of the interferometer (the peaks’ spacing in frequency), but not precisely enough (see Section 2.1.3). Before we calibrate, we work with the nominal 500 MHz FSR. Once we make a more accurate and precise measurement of
the FSR, we can simply rescale our frequency axis by the factor \( c_f = \frac{\text{Precise}}{\text{Nominal}} \). We have three current calibration techniques, each sensitive to different possible errors.

**1000 MHz EOM** The first is a 1000 MHz EOM. When we modulate the signal, the sidebands produce copies of our absorption signal separated by precisely (to seven or eight significant figures) \( \pm 1000 \text{ MHz} \). Since we drive the EOM at low modulation depth, we produce no substantial higher-order sidebands. We then correct our FSR-calibrated frequency axis so that the separation of the sidebands and carrier in our spectra is "exactly" (i.e. to a high, known precision) 1000 MHz.

By virtue of the fact that the \( 7P_{1/2} \) hyperfine splitting is close to 2.15 GHz, the 1000 MHz modulation provides a further check on the success of our frequency-scale linearization. If the HFS were exactly 2 GHz, then the sidebands from each hyperfine line would overlap regardless of the linearization (since \( \text{HFS} = 2 \times f_{\text{EOM}} \)). Since the hyperfine splitting is in fact somewhat larger than 2 GHz, but still close, we can calculate the difference between the two adjacent sidebands (\( \sim 150 \text{ MHz} \)) and that number is less sensitive to the accuracy of our linearization than the measurement of the \( \sim 2.15 \text{ GHz} \) separation of the peaks produced by the carrier. This gives us an independent way of measuring the success of our linearization of the frequency scale.

**600 MHz EOM** We also (secondly) have a 600 MHz EOM. In principle the two EOMs work the same way. So the difference is not in method. If it produces identical results—i.e. agrees with the 1000 MHz EOM—the 600 MHz EOM confirms the results of the 1000 MHz EOM; there is nothing peculiar to the 1000 MHz EOM or the RF generator at 1000 MHz. If they disagree, we know there is either something wrong with the RF generator we use to drive the to EOMs or something inconsistent with our frequency linearization procedure.

**Direct wavemeter calibration** Our last calibration method is to use a wavemeter to measure the laser frequency on several (distant) Fabry-Pérot peaks. We split off part of the beam and send it to a wavemeter. The wavemeter takes the input laser and compares it to
Section 3.3

an internal reference laser, whose frequency is known to high precision, higher even than its linewidth. The wavemeter then reads out the frequency with roughly 30 MHz precision. We then manually sweep the frequency of the laser across a transmission peak in the Fabry-Pérot spectrum and record the reading on the wavemeter. Then we tune the laser frequency to a different frequency, maybe even 100 GHz away (it is not necessary for the laser to tune in a single mode), and repeat the process. Then we have two very distantly spaced peaks (in frequency) and we know the distance must be an integer multiple of the FSR. Since we know the FSR to reasonably good precision (\(\sim 500\) MHz), we can make an initial guess at the integer. Then we adjust our integer guess and FSR appropriately, which gives a value for the FSR. We can repeat this process, which has a possibility for error due to the wavemeter’s operating precision, the challenge of tuning the unlocked laser onto a narrow transmission peak, and the beams’ changing alignment (as the frequency is tuned over a wide range) into the Fabry-Pérot and wavemeter. Misalignment into the Fabry-Pérot can affect the apparent FSR, and misalignment into the wavemeter can affect the measured frequency and even hinder measurement entirely (if the power is low).

A recent calibration by this method showed the FSR to be 501.1(5) MHz. This implies a correction factor of \(c_f = \frac{501.1}{500} = 1.002(1)\).

3.3 Summary of data

We take data in a variety of different ways. It is easiest to break it down by lock point. Locking to the 205 lock point, we take data with the UV beam co- and counter-propagating with the IR beam. In principle the spectra should be identical, because the UV beam is tuned to excite zero-velocity atoms. We separate up-sweeps and down-sweeps to be sensitive to hysteresis in the laser diode. We take data with the 1000 MHz EOM and the 600 MHz EOM so that we are sensitive to issues with either one EOM or one range of frequencies from the RF generator. We do the same for the 203 lock point. From these two sets of data (203 data,
205 data) we can have a number for the $^{203}\text{Tl}$ and $^{205}\text{Tl}$ HFS, which gives us a number for the hyperfine anomaly (HFA).

To measure the isotope shift, however, we must excite both isotopes simultaneously. We lock to the dual isotope lock point, and sweep the IR laser frequency. Once again, we record data in either the co- and counter-propagating configurations or both (see Figure 3.2; this time, we expect to see different spectra. We do not collect data with the modulation from the EOM because the spectrum becomes hard to interpret (and fit). Insofar as we use the EOM to calibrate our frequency scale, which depends on the radius of curvature of the mirrors (= their separation) in the Fabry-Pérot interferometer, the EOM only need be used once—the radius of curvature should not change. Furthermore, if we have values for the HFS for both isotopes, that can serve to calibrate our frequency scale.

### 3.4 Single-isotope analysis and preliminary results

In general, we expect to find data qualitatively similar to our simulations in Section 2.3.2, and so far our data have met this expectation. Figure 3.3a displays a typical single isotope (here, $^{205}\text{Tl}$) spectrum, with modulation at 1000 MHz from the EOM. We fit such a spectrum to a sum of six independent Lorentzians using the linearized nominal (FSR = 500 MHz) Fabry-Pérot scale. From the fitted peak centers, we can extract the HFS and the location of the sidebands. The agreement of the sideband separations gives us a handle on systematic error associated with problems in the linearization of the frequency scale. We can then take the average value of these sideband separations to calibrate the frequency scale with the well-known EOM modulation frequency. It is straightforward to calculate the hyperfine splitting. We find the center frequencies of the absorption peaks (the ones due to the carrier frequency, rather than the sidebands), which are parameters that we can extract easily from the fitted function, and find the difference between the two frequencies. (We can also check that our frequency calibration and linearization are consistent: we should get the same answer if we find the separation of the middle two peaks and add 2000 MHz.) If we collect
Figure 3.2: Sample dual isotope spectrum in co-and counter-propagating configuration. I have labeled the transitions (isotope, final $7P_{1/2}$ $F$ state, responsible beam). Recall that all transitions are from the $7S_{1/2}(F = 1)$. The $x$-axis ramp voltage can be understood as a frequency axis, though the relationship between the two is somewhat nonlinear. In our analysis we linearize the $x$-axis.
Section 3.5

We can separate our systematic error into various categories by looking at different data sets separately.

1. **Frequency calibration.** From our fit parameters we can extract the centers of the various peaks with different EOM modulation frequencies. Within one spectrum, discrepan-
Figure 3.3: Histogram of hyperfine splitting calculation results for $^{205}$Tl, with the frequency scale calibrated by 600 MHz EOM. The Gaussian distribution is centered at 2.177 GHz. Compare the older value of 2.15 GHz from [5]. The standard error of this measurement is roughly $\pm 0.6$ MHz.

Discrepancies between sideband separations lead to error associated with the linearization of our frequency scale. There is further error if the HFS measurements using the 1000 MHz calibration differ from those using the 600 MHz calibration, and if these differ from the calibration from the direct calibration method described in Section 3.2. We can separate frequency calibration error into the following categories.

(a) 1000 MHz calibration. Error due to sideband separation discrepancies.

(b) 600 MHz calibration. Error due to sideband separation discrepancies.

(c) Conflicting calibrations. Error due to conflicting frequency calibrations, visible by comparing the 1000 MHz and 600 MHz EOM calibrations with the direct calibration method described above (see Section 3.2).
Figure 3.4: Histograms of uncalibrated sideband separation measurements for 600 MHz EOM modulation for $^{205}$Tl down-sweep spectrum in counter-propagating configuration. For each sideband, the frequency separation from the main peak is measured using the linearized nominal Fabry-Pérot frequency scale. The center of each histogram is recorded above. Taking the average gives an apparent splitting of 597.7(2) MHz, or a frequency correction factor $c_f = \frac{600.00}{597.7} = 1.0038(3)$. 
2. **Sweep direction.** Discrepancy between HFS measurements for up- and down-sweeps of PZT voltage amounts to error in the final measurement. This error is a result of hysteresis in the laser diode.

3. **Beam configuration.** Discrepancy between HFS measurements in the co- and counter-propagating configurations results in error in the final measurement due to alignment and divergence differences between the co- and counter-propagating UV beams.

4. **Fit methods.** Error can be introduced in the separate methods we use to linearize the frequency scale and fit to the absorption spectrum.

   (a) **Linearization.** The success of the linearization depends on the order of the polynomial we use to fit to the PZT voltage vs. frequency curve (see Figure 3.1).

   (b) **Lineshape.** As discussed, the true shape of the absorption line is the Voigt profile (see Equation 2.6), but in the limit with little to no Doppler broadening, the Voigt profile can be approximated well as a Lorentzian (see Figure 2.1). We prefer to fit to a sum of Lorentzians because it is substantially less computationally intensive, though there can be error associated with fitting to the different functions.

Consider now the histograms in Figure 3.4, which show the uncalibrated (i.e. calibrated with the nominal Fabry-Pérot FSR) frequency separation of the sidebands from the center peaks in the atomic spectra for one particular beam configuration and sweep direction. By averaging the center frequencies of the Gaussians, we can calculate that the true FSR of the Fabry-Pérot is given by

\[
\text{FSR} = \frac{600.0 \text{ MHz}}{597.7 \text{ MHz}} \times 500 \text{ MHz} = 501.9 \text{ MHz}. \quad (3.1)
\]

We can estimate the error crudely, based on the different values for the different sidebands, to be ±0.5 MHz. (Compare the value from the direct calibration, 501.1 ± 0.5 MHz.) In this way we can understand where our sources of error are.
As an example of how we would do this on a larger scale, consider all the runs in Figure 3.3 that are calibrated by the 600 MHz EOM. By subdividing that chart, we can begin to study some of the systematic error in our measurement. I tabulate it in Table 3.1 and find a total error at the level of 1.5 MHz.
Chapter 4

Future work

The first and most obvious task is to take more data. At the risk of hubris, the apparatus is working in its essentials. Both lasers are mostly cooperative; it is still best to leave the UV laser on as much as possible, but the IR laser is working well. The locking system seems to be working well enough. The main outstanding issue there is the recurring difficulty with the hiccup with the lock point for the $^{203}$Tl single isotope excitation. (The lock seems to prefer to be locked to the $^{205}$Tl lock point, and willing to jump away from the $^{203}$-thallium lock point to get there.)

But, besides the immediately essential injunction to take more data, there are a variety of desirable tasks that, while not immediately threatening to the wellbeing of the experiment, are nevertheless likely to prevent future headaches. Review/redo the temperature controllers for the ovens. One of them broke in the past (April 2012), which was a big mess for the student working on it at the time. The one that did not break has the same design flaw as the one that did—namely, that the failure mode for the solid state relay (switch) is closed/on, rather than open/off. All things considered, it is an issue of when, not if, that controller will break. A simple voltage limiter circuit should be enough to win some peace of mind. While this experiment is in all likelihood finishing up this summer, it is likely that the oven will remain a key part of whatever experiment is performed next.
Appendix A

Data breakdown

As of 8 May 2013, we have collected the following data. The date, configuration, and isotope are clear enough from the file name, with the exception of the files marked as March 15 $^{203}$Tl data, which are really $^{205}$Tl data.
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Appendix B

Optimization of locking temperature

While the following derivation may seem long-winded (considering that we could in principle find the answer experimentally), it is representative of the way in which we can simulate our results.

The goal is to find the dependence of the lineshape—in particular, the slope at the zero-crossings—on temperature. The line we are examining is the $7S_{1/2}$ line. As mentioned in Section 1.3, this line is split into two hyperfine states (so if we drive the $6P_{1/2} \rightarrow 7S_{1/2}$ transition, we expect to see three sets of lines because of the two isotopes). But we are only interested in the $(F = 1) \rightarrow (F' = 1)$ transition. From Table 1.1, we know that the HFS is about 12 GHz, so the Doppler broadening (FWHM $\approx$ 1 GHz, from Eqn. 2.5), is not enough to make us consider more than this one pair of lines. We also know that the isotope shift is around 1.6 GHz [2], so we know we can resolve both lines.

Because the Doppler broadening is the dominant effect, we can approximate the lineshape as a Gaussian:

$$A(f) = \text{Absorption}(f) = A_{205} \exp\left(-\frac{(f - f_0)^2}{2\sigma_{205}^2}\right) + A_{203} \exp\left(-\frac{f^2}{2\sigma_{203}^2}\right), \quad (B.1)$$

where $A_{205}$ and $A_{203}$ are the abundances of $^{205}$Tl and $^{203}$Tl respectively and $\sigma = \sigma(m, T)$, related to Eqn. 2.5 by some factors of 2 and $\log(2)$. I have put the center frequency for the
Figure B.1: Absorption($f$) for $T = 800$ K. The smaller peak corresponds to the $^{203}$Tl $7S_{1/2}(F'=1)$ line.

$^{203}$Tl line at $f = 0$, and so $f_0 = \Delta f_{IS}$, the isotope shift frequency. Figure B.1 is a plot of absorption as a function of frequency at $T = 800$ K.

We do not, however, directly observe the absorption. Instead we measure the transmitted intensity. The transmitted intensity is given by

$$I_{\text{transmitted}} = I_0 \exp(-\alpha A(f)),$$  \hspace{0.5cm} (B.2)

where $I_0$ is a scaling factor for the laser intensity and $\alpha$ is the so-called optical depth. The optical depth is, heuristically, a measure of opacity. It directly depends on the probability that a transition can be excited, the length of the cell, and the density of the atomic vapor. The density in turn depends on temperature, via the so-called Antoine equation, which
relates vapor pressure $P$ to the temperature of the solid or liquid $T$:

$$\log P = A - \frac{B}{T},$$  \quad (B.3)$$

where $A$ and $B$ are adjustable constants that depend on the material’s properties.

![Log(Pressure(Pa)) vs. Temperature (K)](image)

Figure B.2: $\log_{10} P$ (measured in Pa) vs. $T$ in kelvin. Least-squares fit.

It is easy enough to find the values of $A$ and $B$ in the case of thallium, given some information about the vapor pressure at various temperatures. Using values from the AIP Handbook [16], I optimized my choice of $A$ and $B$ using Kaleidagraph. See Figure B.2; I have
\( A = 10.053, \ B = 8849.2 \) kelvin, so

\[
P (\text{in Pa}) = 10^{10.053-8849.2/T}, \tag{B.4}
\]

where \( T \) is measured in kelvin. Now, since density is a function of pressure (from the ideal gas law, \( PV = nkT \)), I have density \( \rho(T) = n/V = P(T)/kT \). Thus I can write, combining all the parameters related to the cell and the atoms into one constant \( C \),

\[
\alpha(T) = C \frac{10^{10.053-8849.2/T}}{T} \tag{B.5}
\]

We can now find \( C \) and so \( \alpha(T) \) experimentally, by simply looking at our spectrum at any single temperature. A quick measurement shows

\[
\alpha(643) = C \frac{10^{10.053-8849.2/643}}{643} = 3.04 \times 10^{-7} = -\ln\left(\frac{I_{\text{Max absorption}}}{I_0}\right) = 0.226, \tag{B.6}
\]

so \( C = 7.44 \times 10^5 \). Now we can plot the zero-crossing slope as a function of frequency (see Figure B.3 and find the optimal temperature. More important is the qualitative observation that different temperatures optimize different lock points.
(a) Zero-crossing at $^{203}$Tl lock point as a function of temperature.

(b) Zero-crossing at $^{205}$Tl lock point as a function of temperature.

(c) Zero-crossing at dual isotope lock point as a function of temperature.

(d) Overlay of (a), (b), and (c), with the sign of (c) switched for the sake of comparison. The three lock points are optimized at different temperatures, ranging from about 690 K to 715 K.

Figure B.3: Optimization of lock temperature. Zero-crossing slope plotted as a function of temperature for the three lock points.
Bibliography


