NONLINEAR SPECTROSCOPY OF RUBIDIUM
AND FREQUENCY STABILIZATION OF
A 1560 nm DIODE LASER

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Abstract

Frequency-stabilized lasers are ubiquitous tools in laser spectroscopy and optical metrology. This thesis describes precision nonlinear spectroscopy of rubidium (Rb) at 780 nm that is achieved using the second harmonic of a diode laser at 1560 nm. The frequency of the laser is then actively stabilized to Rb resonances. It will ultimately serve as a frequency reference for optical time transfer and clock comparisons of frequency standards over a free-space optical link.

A low power 1560 nm external cavity diode laser is frequency-doubled to 780 nm efficiently by sending the laser into a periodically-poled lithium niobate waveguide. We generate the laser’s second harmonic with a conversion efficiency of 1%/mW. With the 200 microwatts of second harmonic power generated, we then perform saturated absorption spectroscopy (SAS) on a Rb vapor cell, so that the laser can access the $D_2$ lines at 780 nm. SAS is utilized to counter the effects of thermal Doppler broadening.

Once we obtain a SAS resonance signal whose width at 7 MHz approaches the natural linewidth of the electronic transition, we are ready to frequency-lock the laser. Applying a modulation-locking technique, we dither the laser frequency about resonance. This generates an error signal proportional to the frequency deviation from resonance. We use the error signal as a feedback mechanism with a novel RF-locking method to lock the diode laser’s frequency to the saturated absorption resonances, ultimately achieving frequency instabilities of 90 kHz in 1 s averaging time.
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Chapter 1

Introduction

The following thesis describes the frequency stabilization of a 1560 nm external cavity diode laser by the traditional method of saturated absorption spectroscopy (SAS) on the rubidium $D_2$ lines. The project has been carried out in support of developing precise methods to transfer time and frequency signals through free-space optical links. Precision timing is a crucial tool for many aspects of fundamental and applied physics, including tests of Einstein’s general relativity, tests of local Lorentz invariance, and enhancement of navigation systems, e.g. Global Navigation Satellite System (GNSS).

We stabilize the 1560 nm diode laser to an absolute frequency reference, provided by a rubidium atomic transition at 780 nm. Chapter 2 describes how the second harmonic of the 1560 nm cw laser is generated using a nonlinear crystal, to access this transition. Then, saturated absorption spectroscopy is performed on a Rb vapor cell to find this transition (c.f. Chapter 3).

Finally, we establish a feedback control system to stabilize (lock) the cw laser, as described in Chapter 4. The purpose of active stabilization is to suppress the low-frequency drift of the laser.
Chapter 2

Accessing the Rb Line using Second Harmonic Generation

This chapter describes how we produce the light to access the external frequency reference for laser stabilization purposes. Atomic transitions provide unchanging reference frequencies. We choose a Rb D\textsubscript{2} line transition near 780 nm as our reference. Naturally, our first step is to access the transition with our 1560 nm laser. To do this, we frequency double our light source with a nonlinear crystal.

We start with a discussion of the theory of second harmonic generation and quasi-phase-matching. Then, we describe the characterization process of the nonlinear crystal. Finally, we discuss the resulting second harmonic conversion efficiencies obtained.

2.1 Background: Second Harmonic Generation

Frequency doubling, or second harmonic generation (SHG), is a consequence of nonlinearity in optical materials. It was realized soon after the invention of the laser, and marked the birth of the field of nonlinear optics. First reported in 1961 by Peter Franken et al in a piece of crystalline quartz, SHG has become an important and...
ubiquitous tool in many atomic physics laboratories [1]. The process consists of two photons of the same frequency interacting to produce a photon of twice the energy (and frequency) of the initial photons. Second harmonic generation is a special case of sum frequency generation, whereby two photons of energies $\hbar \omega_1$ and $\hbar \omega_2$ combine to form a third photon of energy $\hbar \omega_3$.

This section will give an overview of the theory behind SHG, following the derivations of Milonni and Eberly, Ch. 10 [5]. Then, it will address the implementation of SHG in the context of accessing the Rb $D_2$ transition with the lab’s 1560 nm laser.

### 2.1.1 Derivation of second harmonic power expression

We start with the transverse wave equation, derived from Maxwell’s equations in matter.

$$\nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P}{\partial t^2}$$

Equation (2.1)

For simplicity, we consider a single component of the electric field $E$ propagating in the $\hat{z}$ direction. We further approximate the incoming field to be monochromatic upon a non-centrosymmetric medium of polarization $P$ and refractive index $n(\omega)$. The plane wave solutions for the second harmonic field will be

$$E = \frac{1}{2} \mathcal{E}_{2\omega}(z)e^{-i(2\omega t - k_{2\omega} z)} + \frac{1}{2} \mathcal{E}^*_{2\omega}(z)e^{i(2\omega t - k_{2\omega} z)} \quad (2.2)$$

where $k_{2\omega} = n(2\omega) \frac{2\omega}{c}$. Plugging back into the left hand side of Equation (2.1), we obtain

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{2} \frac{2ik_{2\omega}}{c^2} \frac{d\mathcal{E}}{dz} - k_{2\omega}^2 \mathcal{E}_{2\omega} + \frac{d\mathcal{E}^2}{dz} [e^{-i(2\omega t - k_{2\omega} z)}] e^{-i(2\omega t - k_{2\omega} z)}$$

$$- \frac{2\omega^2}{c^2} \mathcal{E}_{2\omega} e^{-i(2\omega t - k_{2\omega} z)} + c.c.$$  

Equation (2.3)
We assume that the envelope function $E_{2\omega}$ will be varying slowly with respect to the phase term $\exp(ikz)$, so we may drop the term proportional to $e^{-2i(2\omega t - k_{2\omega}z)}$ from the wave equation \[5\].

So far we have just assumed the existence of a second harmonic solution, without treating the material properties required for such a solution to exist. Only media without inversion symmetry can generate second harmonic light. Mathematically, this is expressed in terms of the polarization. To the lowest order, a medium’s polarization has a linear dependence on the electric field. The next highest order polarization term originates from a quadratic dependence on $E$. The $i$th component of polarizability is the sum of linear and nonlinear components,

$$P_i/\epsilon_0 = \frac{1}{\epsilon_0}(P_i^L + P_i^N) = \sum_j \chi_{ij}^L E_j + \sum_{jk} \chi_{ijk}^N E_j E_k + \ldots \quad (2.5)$$

where $\chi$ are the susceptibility tensors. It is the nonlinear term proportional to $E^2$ in Equation (2.5) that enables SHG. Only in materials which lack inversion symmetry (i.e., the quantum states of electric dipole transitions have definite parity) is the term $\chi_{ijk}$ nonvanishing \[5\]. In such materials, three-wave mixing can occur, giving rise to two photons of energy $\hbar\omega$ producing one photon of energy $2\hbar\omega$.

The plane-wave contribution to the right-hand side of Equation (2.1) is

$$P = \frac{1}{2}[P_{2\omega}^L + P_{2\omega}^N]e^{-i(2\omega t - k_{2\omega}z)} \quad (2.6)$$

Combining the above equation with Equation (2.1) obtains

$$[ik_{2\omega}\frac{dE_{2\omega}}{dz} - \frac{E_{2\omega}}{2}(k_{2\omega}^2 - 4\omega^2/c^2)]e^{ik_{2\omega}z} = -2\mu_0\omega^2(P^L e^{ik_{2\omega}z} + P^N e^{2ik_{\omega}z}) \quad (2.7)$$
We substitute the relation $P^{L}_{2\omega} = \epsilon_0 \chi^L(2\omega) E_{2\omega}$ into Equation (2.7). We also utilize the relation $P^{N}_{2\omega} = \frac{\omega}{2} \chi^N(2\omega) E_{2\omega}^2$. Then, we match the terms from both sides of Equation (2.7) by their degrees in $E_{2\omega}$.

From terms linear in electric field,

$$k_{2\omega}^2 = \frac{4\omega^2}{c^2} (1 + \chi^L(2\omega))$$  \hfill (2.8)

The remaining nonlinear terms give

$$\frac{dE_{2\omega}}{dz} = i\omega \frac{\sqrt{\mu_0/\epsilon_0} E_{2\omega}^2(z) \exp[iz(2k_\omega - k_{2\omega})]}{dz}$$  \hfill (2.9)

where, for convenience, $d = \frac{\omega}{2} \chi^N(2\omega)$. The argument of the exponential in Equation (2.9) reflects the difference in phase resulting from different refractive indices for the fundamental and second harmonic light. For this reason, we define $\Delta \equiv 2k_\omega - k_{2\omega}$.

To solve Equation (2.9), we take the fundamental field as constant with respect to $z$. We are assuming that the fundamental field is not depleted significantly over a distance $z$.

Finally, we have

$$E_{2\omega} = i\omega \sqrt{\frac{\mu_0}{\epsilon_0}} \int_{0}^{z} \exp(i\Delta k z') dz'$$  \hfill (2.10)

$$= i\omega \sqrt{\frac{\mu_0}{\epsilon_0}} E_{2\omega}^2 (e^{i\Delta k z} - 1) \frac{1}{i\Delta k}$$  \hfill (2.11)

$$= i\omega \sqrt{\frac{\mu_0}{\epsilon_0}} E_{2\omega}^2 ze^{i\Delta k z/2} \frac{\sin(\Delta k z/2)}{\Delta k z/2}$$  \hfill (2.12)

In the lab, the quantity of interest is optical power, not electric field. The time-averaged intensity (power per beam area) for the second harmonic, as a function of distance, is therefore
When there is no phase difference ($\Delta = 0$), the sinc function evaluates to zero, giving a second harmonic efficiency $\eta$ of

$$\eta(z) = 2\sqrt{\frac{\mu_0^3/\epsilon_0^3}{\omega^2 d^2 I_\omega(0) z^2}} \frac{(\Delta k z / 2)^2}{n^3}$$  \hfill (2.16)$$

Having zero phase difference is a special case, of course, and most crystals used in SHG do not satisfy the requirements. In general, the phases between the fundamental and second harmonic will slip due to differences in refractive index. In the next section, we will discuss how phase-matching may be obtained to maximize conversion efficiency, in order to use Equation (2.16).

### 2.1.2 Quasi-Phase-Matching

A phase mismatch ($\Delta k \neq 0$) can severely lower second harmonic intensities. The fundamental and second harmonic light will suffer a relative phase mismatch over a coherence length $L_c = \pi/\Delta$. In contradiction to the simplified expression in Equation...
increasing the length $z$ of the medium past its coherence length $L_c$ does not increase second harmonic efficiency. Rather, the two frequencies of light will continue to periodically interfere constructively and destructively (Figure 2.2). For example, using Equation 2.9 the second harmonic field at $z = L_c$ is

$$E_{2\omega}(L_c) = i\omega d\mathcal{E}_\omega^2 \sqrt{\mu_0/\varepsilon_{2\omega}} \int_0^{L_c} \exp(i\pi z/L_c)dz = -2\omega/\pi \sqrt{\mu_0/\varepsilon_{2\omega}} d\mathcal{E}_\omega^2 L_c \quad (2.17)$$

whereas at $z = 2L_c$, the field is

$$E_{2\omega}(2L_c) \propto \int_0^{2L_c} \exp(i\pi z/L_c)dz = \frac{L_c}{i\pi}(e^{2\pi i} - 1) = 0 \quad (2.18)$$

showing that at twice the coherence length, the second harmonic field vanishes.

One way to enforce phase-matching is to utilize birefringence of the uniaxial crystals used in SHG [5]. Such a doubly refracting material has different indices of refraction depending on incoming light polarization. To achieve phase-matching, the fundamental wave is propagated into the material as an extraordinary wave at some angle $\theta$ to the optic axis, where $\theta$ is chosen to satisfy $n_0(2\omega) = n_e(\omega, \theta)$. Then the second harmonic wave is generated as an ordinary wave in the direction $\theta$. However, few birefringent materials also have a substantially high nonlinear index, which limits the $d$ value in the second harmonic intensity (Equation 2.13).

Another technique, quasi-phase-matching (QPM), enables use of non-birefringent crystals with high nonlinear index such as lithium niobate (LiNbO$_3$). This technique in LiNbO$_3$ can improve conversion efficiency by $(2d_{33}/\pi d_{31})^2 \approx 20$ compared to birefringent phase matching [8]. In QPM, the material is structured such that the crystal axis orientation changes periodically. Thus, the nonlinear coefficient changes periodically with position, with a value of $d$ at $0 \leq z \leq L_c$ to a different value $-d$ at $L_c \leq z \leq 2L_c$. In this case, Equation 2.13 gives
Figure 2.2: A comparison of the second harmonic light generation as a function of crystal length. In the case of no phase-matching, $\Delta \neq 0$ and the efficiency is limited by the coherence length $L_c$. The destructive interference can be overcome with QPM, which causes the electric field to increase monotonically with distance. Image from ThorLabs.

\[ \mathcal{E}'_{2\omega}(2L_c) = \mathcal{E}_{2\omega,d}(L_c) - \mathcal{E}_{2\omega,-d}(L_c) = 2\mathcal{E}_{2\omega,d}(L_c) = -\frac{4\omega d\varepsilon_0^2 L_c}{\pi \varepsilon_{2\omega}} \sqrt{\frac{\mu_0}{\varepsilon_{2\omega}}} \] (2.19)

The period of the variation in $d$ is clearly $2L_c$, so the second harmonic intensity continues to grow, following Equation 2.19. The resultant field increases linearly with distance, $z$, leading to a quadratic growth of second harmonic intensity over the length of the crystal. A representation of QPM can be seen in Figure 2.2.

Periodically poled lithium niobate (PPLN) is often used for QPM. Each unit cell of the crystal LiN0$_3$ has a permanent, small electric dipole moment. Consequently, a strong electric field can change these dipoles’ orientations. A field (typically on the order of 20kV/mm) can be applied for a few milliseconds along the longitudinal length of the crystal, at periodic intervals. If such fields act on equally spaced sections, they can cause a periodic variation on dipole orientation. Therefore, the sign of the nonlinear coefficient also varies.
2.1.3 SHG in waveguides

Additional SH power can be obtained using a waveguide structure in the frequency-doubling medium. A waveguide maintains a smaller mode area over a greater length, and thus a higher intensity, as compared to a bulk frequency doubler. In a bulk medium, diffraction limits the interaction length. Efficient doubling in nonlinear waveguides can be achieved for laser power levels below 100 mW (well within the range of diode lasers), whereas bulk crystals require much higher input power (several watts, for instance) because of the larger input beam size.

A QPM waveguide has efficiency

\[
\eta(z) = \frac{32\pi^2 d^2 \omega^2}{\alpha n^2(\omega)n(2\omega)\epsilon_0 P_\omega z^2} \frac{\kappa}{A_{eff}} \frac{\sin^2(\Delta k L/2)}{\Delta k L/2} \tag{2.20}
\]

The preceding equation is different from Equation 2.16 by the parameters \(\kappa\) and \(A_{eff}\). \(\kappa\) is the coefficient that takes into account the number of modes in the waveguide; typically it will be 1 for a single-mode spectrum. \(A_{eff}\) relates to the effective area of the waveguide; for a derivation of this quantity, see [4].

LiNbO\(_3\) waveguides are typically fabricated by proton exchange [8]. The waveguide PPLN device used in this project was 59 mm long and single mode for the input of 1560 nm. The mode size was 4 \(\mu\)m \(\times\)3\(\mu\)m.

2.2 Generating and optimizing second harmonic power using a PPLN crystal

In our experiment, a 1560 nm diode laser (RIO Orion, Redfern) was injected to a fiber-coupled PPLN waveguide doubler to produce the second harmonic at 780 nm. This light was then used to perform spectroscopy on Rb atoms. Our PPLN crystal was fabricated by Carsten Langrock, in Martin Fejer’s group in the Department of
Applied Physics at Stanford. It is 59 mm long with poling periods $2L_c = 14.5 \mu m$. The device was single mode at the 1560 nm input wavelength.

Our first step was to maximize the SH output of our desired wavelength out of the PPLN crystal. The effectiveness of quasi-phase-matching depends on the coherence length $L_c$, total device length, and refractive indices, all of which are temperature-dependent. Accordingly, we needed a way to reliably and uniformly control the crystal’s temperature.

An aluminum mount for the PPLN crystal was devised, with holes drilled to allow for four 3 W cartridge heaters to be placed. The heaters were connected in series and epoxied into the holes. A thermistor was placed within the mount. It was bonded by a thermal compound to be underneath the waveguide, for diagnostics. Before the PPLN was placed, the entire system was subjected to a 100°C heat gun to allow outgassing of the adhesive components, which may damage the crystal. Figure 2.3 shows the mount, without its lid, and how the PPLN crystal is slotted within the aluminum. The edges of the waveguide were secured to the mount using thin strips of Kapton tape.

The entire ensemble is placed on the optics table, where an aluminum foil-lined box thermally shields the PPLN crystal and its mount from the environment. Temperature stability in the laboratory is to within 1 degree, which is low enough to not necessitate active temperature control of the PPLN crystal.

The incoming light from the polarizing paddles is p-polarized. Light is fiber-coupled to the PPLN waveguide, resulting in a coupling loss of 40%. With the fiber core index of refraction of 1.6 and LiNiO$_3$ refractive index of 2.3, input Fresnel loss is approximately 3%. The PPLN crystal-air interface is polished roughly at 10° to suppress parasitic etaloning. From Fresnel reflection, we expect a 13% loss from the LiNiO$_3$ crystal at 10°. We collimate the beam using a 5 mm focusing lens. See Figure 2.5 for details.
To optimize SH power, we experimentally tune the temperature on the waveguide while injecting 1560 nm light. A silicon photodiode measures the SH power (since 1560 nm is lower than silicon’s conduction band, we are sure to only pick up 780 nm light and higher harmonics). The optimal temperature for phase-matching was found to be 38.0 °C when the laser was tuned to the center of the $^{87}$Rb $F = 2 - F' = 1, 2, 3$ transition. Even with the crystal temperature fixed on the value optimized for one frequency, the phase matching was sufficient for spectroscopy spanning $\approx 10$ GHz and all four fine structure lines.

To measure $\eta \equiv \frac{P_{SH}}{P_F^2}$, we measure the fundamental input $P_F$ into the PPLN as well as the second harmonic output, $P_{SH}$. The silicon photodiode picks up the 780 nm output. Our data are shown in Figure 2.4. The quadratic dependence of 780 nm power is evident at low input powers (1 - 2 mW), as predicted by Equation 2.20. However, the output power seemingly has a linear dependence on input power past $P_{in} \geq 2.5$ mW, due to depletion of the pump source.
Typically, conversion efficiencies are reported in %/W, due to the nonlinearity of second harmonic generation (see Equation 2.16). After performing these measurements and accounting for fiber coupling and Fresnel losses, we obtain $\eta \approx 1%/mW$. We can reliably obtain 200 $\mu W$ of 780 nm light for Rb spectroscopy, which is enough to saturate the relevant transitions (see Chapter 3).

With frequency-doubling capabilities verified, we proceed to perform nonlinear spectroscopy on Rb.
Figure 2.5: The setup for frequency doubling using a periodically poled LiNbO$_3$ crystal (PPLN). The fundamental light at 1560 nm enters the crystal, and a second harmonic signal is also produced. A 5 mm lens collimates the beam.
Chapter 3

Saturated Absorption

Spectroscopy on the Rb D2 Line

This chapter describes the method of saturated absorption spectroscopy (SAS) on rubidium, using the SH light described in the preceding chapter. We begin with an introduction to saturated absorption spectroscopy, and discuss the varying limits on the linewidths of the Rb transitions. Then, we discuss our experimental setup for saturated absorption.

3.1 Widths of spectral lines

Traditional spectroscopy uses a narrowband light source with tunable frequency, sweeping over the sample in question in order to drive an electron transition. In fluorescence spectroscopy, the incident light has intensity sufficient to raise the excited state population so that stimulated emission is detectable. To do so, incident light must be within a specific spectral distribution about the central transition frequency \( \omega_0 = \Delta E/h \) corresponding to an atomic transition of energy \( \Delta E \). This distribution is characterized by the full-width at half-maximum of the line, the FWHM (or the linewidth).
There are various mechanisms that broaden the value of the linewidth. The following section will treat the most important broadening mechanisms: the natural linewidth, Doppler broadening, collisional broadening, and power broadening.

The greatest limit to the achievable resolution on $\omega_0$ is often the Doppler broadening of spectral lines, a result of the particles’ thermal motion. Saturated absorption (or Doppler-free) spectroscopy overcomes this limit without cooling of the particles. Often, this technique can resolve the transition down to its natural linewidth, typically orders of magnitude less than the Doppler width. The following sections will then explore this type of nonlinear spectroscopy applied to rubidium atoms. Our light source is a diode laser at 1560 nm, frequency doubled to 780 nm.

3.1.1 Natural Linewidth

The natural linewidth is a finite value that will always broaden the spectral line of an atom, even when the atom does not interact with its environment. It is due to spontaneous emission from an atomic state with a finite lifetime, represented by Einstein’s A coefficients.

For a semiclassical derivation of the natural linewidth, we use the harmonic oscillator model: an excited electron of mass $m$ and frequency $\omega$ releases excitation energy through spontaneous emission of a photon, described by the damping constant $\gamma$. For real atoms, $\gamma/\omega \ll 1$. Electron motion follows the equation

$$\ddot{x} + \gamma \dot{x} + \frac{k}{m} x = 0 \quad (3.1)$$

where $k$ is some restoring force constant. The central frequency of the transition is $\sqrt{k/m}$. The real solution of 3.1 approximating $\gamma \ll \sqrt{k/m} \equiv \omega_0$ is

$$x(t) = x_0 e^{-\gamma t/2} \cos(\omega_0 t) \quad (3.2)$$
The amplitude of \( x(t) \) falls over time as a decaying exponential, so its Fourier transform pair \( x(\omega) \) must have a Lorentzian lineshape. Finally, the real intensity \( I(\omega) = x(\omega)x^*(\omega) \) has the form

\[
I(\omega - \omega_0) = \frac{N}{(\omega - \omega_0)^2 + (\gamma/2)^2}
\] (3.3)

The constant \( N \) is due to normalization of intensity. According to Equation 3.3, the FWHM is \( \gamma \). The FWHM is related to the mean lifetime of the transition \( \tau \) by \( \tau = 1/\gamma \). For the D\(_2\) lines of \(^{85}\)Rb and \(^{87}\)Rb, the mean lifetime is \( \tau = 26.24 \) ns, giving a natural linewidth of \( 2\pi \times 6.067 \) MHz [7].

### 3.1.2 Doppler Broadened Linewidth

At room temperature, all transitions are broadened far beyond their natural linewidths by Doppler broadening. Different velocities of emitting atoms result in Doppler-shifted frequencies of

\[
\nu = \nu_0 (1 + v/c)
\] (3.4)

where \( \nu, \nu_0 \) are the observed and rest frequencies. In the case of thermal Doppler broadening (the strongest effect), atomic velocity distribution \( P_v(v) \) is given by the Maxwell distribution,

\[
P_v(v)dv = \sqrt{\frac{m}{2\pi kT}} \exp\left(-\frac{mv^2}{2kT}\right)dv
\] (3.5)

where \( m \) is the atomic mass and \( T \) is the temperature. Substituting \( v \) for \( \nu \) using Equation 3.4 and performing the corresponding change of coordinates, we obtain the frequency distribution

\[
P_\nu(\nu)d\nu = \sqrt{\frac{mc^2}{\pi w^4 kT \nu_0^2}} \exp\left(-\frac{mc^2(\nu - \nu_0)^2}{2kT \nu_0^2}\right) d\nu
\] (3.6)
This Gaussian profile has FWHM equal to \( \sqrt{\frac{8kT \log(2)}{mc^2}} \nu_0 \), which can be around 500 MHz for Rb at room temperature. Clearly, thermal Doppler broadening results in a linewidth of two orders of magnitude greater than the natural linewidth, at room temperature.

### 3.1.3 Collisional Broadening

When two atoms interact, their energy levels are shifted because of their interaction. Depending on whether the atoms have attractive or repulsive interaction \( \Delta E \) may be negative or positive. There are two mechanisms for collisional broadening: elastic and inelastic collisions. The former leads to a phase shift in the oscillator, and is also termed a *phase-perturbing collision* \[2\]. It is determined by the number density of atoms and the collision cross section.

Inelastic collisions (also called *quenching collisions*) effectively shorten the lifetime of the upper energy level. A quenching collision between atoms A and B transfers excitation energy \( E_i \) of A into internal energy of B or translational energy of A and B. Therefore such a collision decreases the number of excited atoms in level \( E_i \) and quenches the fluorescence intensity. The effective lifetime of \( E_i \) decreases, causing an increase in linewidth.

Together, these effects cause the collisional self-broadening of Rb to be 56 MHz/torr at \( T = 300K \) \[6\]. At a vapor pressure of \( 10^{-6} \) torr, collisional broadening’s effect is much suppressed beyond the natural linewidth uncertainty in the transition.

### 3.1.4 Power Broadening

The laser introduces some oscillatory electric field \( E = E_0 \cos(\omega t) \), which shifts the energy levels by the AC Stark shift. Following the derivation of Demtroder \[2\], the upper state population has a mean population probability, as a function of laser frequency \( \omega \), of
\[ P_b(\omega) = \frac{1}{2} \frac{\Omega_R^2}{(\omega_0 - \omega)^2 + \gamma^2(1 + S)} \]  

(3.7)

where \( \Omega_R = \vec{d} \cdot \vec{E}/\hbar \) is the Rabi flopping frequency of the two-level system, and \( \vec{E}, \vec{d} \) are the electric field amplitude and transition dipole moment respectively. The saturation parameter \( S = \Omega_R^2/\gamma^2 \). \( P_b(\omega) \) is proportional to the absorption line profile, so power broadening gives the linewidth \( \gamma_P = \gamma\sqrt{1 + S} \).

A summary of the discussed broadening mechanisms and their values are given in Table 3.1.4.

<table>
<thead>
<tr>
<th></th>
<th>Natural Linewidth</th>
<th>Thermal Doppler</th>
<th>Collisonal</th>
<th>Power</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{87}\text{Rb} ) (MHz)</td>
<td>6.08</td>
<td>509</td>
<td>0.00006</td>
<td>7</td>
</tr>
<tr>
<td>(^{85}\text{Rb} ) (MHz)</td>
<td>6.08</td>
<td>515</td>
<td>0.00008</td>
<td>7</td>
</tr>
</tbody>
</table>

Table 3.1: Typical values of FWHM broadening of the Rb \( D_2 \) line at room temperature (300K). Laser intensities range from 1 - 2 mW/cm\(^2\).

### 3.2 Rb \(^{85}, 87\) Energy Levels

Our atomic reference is a vapor cell of rubidium atoms composed of 28\% \(^{87}\text{Rb} \) and 72\% \(^{85}\text{Rb} \) (the natural abundance of the isotopes). Rb is ubiquitous in quantum optics experiments due to the availability of diode laser light at the relevant wavelength. In particular, the hyperfine structure of Rb energy levels make it useful for high-precision timing. The following section explores the structure of \(^{87}\text{Rb} \) and \(^{85}\text{Rb} \).

Rb, as an alkali metal, is the lightest element to populate the \( n = 5 \) level. The first excited state transitions are split from spin-orbit interactions into a fine-structure doublet (quantum number \( J = \mathbf{L} + \mathbf{S} = 1/2 \) or \( 3/2 \)). \( J = 1/2 \) corresponds to the \( D_1 \) line, and \( J = 3/2 \) is the \( D_2 \) line used for spectroscopy. The fine structure doublets can be resolved in linear, Doppler-limited spectroscopy (see Figure 3.2). Each of these doublet lines additionally has hyperfine structure. The hyperfine states in the absence of an external magnetic fields are shown in Figure 3.1 and are denoted with
quantum number $F = J + I$, where $I$ is the nuclear angular momentum ($I = 3/2$ for $^{87}\text{Rb}$ and $5/2$ for $^{85}\text{Rb}$). The $2F+1$ hyperfine magnetic sublevels (not pictured) are degenerate without external magnetic field. To prevent lifting of this degeneracy (and a further shift in transition frequencies), the Rb cell used in the experiment is magnetically shielded by mu-metal.

### 3.3 Theory of Saturated Absorption Spectroscopy

Saturated absorption spectroscopy (SAS) is a setup that enables the determination of a transition frequency to an accuracy limited, ideally, only by the width of the excited state (the natural linewidth). Without cooling to lower temperatures, SAS can theoretically eliminate the uncertainties due to Doppler broadening. It is a common method to lock the laser frequency to the atomic transition.
Figure 3.2: Linear spectroscopy showing the four Rb $D_2$ lines. (A) $^{87}\text{Rb } 5S_{1/2} \ F = 2 - 5P_{3/2} \ F' = 1,2,3$ (B) $^{85}\text{Rb } 5S_{1/2} \ F = 3 - 5P_{3/2} \ F' = 2,3,4$; (C) $^{85}\text{Rb } 5S_{1/2} \ F = 2 - 5P_{3/2} \ F' = 1,2,3$; (D) $^{87}\text{Rb } 5S_{1/2} \ F = 1 - 5P_{3/2} \ F' = 0,1,2$.

Figure 3.3: A schematic of the classic pump-probe scheme for spectroscopy

A classical pump-probe scheme is used to overcome the problem of Doppler broadening. A strong pump beam is incident on the atomic gas, and a weak probe beam is counterpropagated over the pump, as in Figure 3.3.

Both beams are from the same laser and have the same frequency, even as the frequencies are tuned through resonance.

The pump beam is strong enough to affect the populations of the ground and excited states; this will change the probe’s measured absorption. Depending on the pump intensity, this beam may saturate the transition when tuned to the appropriate frequency, as dictated by the Doppler shift (see Equation 3.4). The pump will
only perturb the population of atoms moving at the lab frame velocity corresponding to the Doppler-shifted resonance frequency. Consequently, different portions of the Maxwellian atomic velocity distribution are affected by the laser. The corresponding ground state densities will drop significantly at these precise frequencies, a phenomenon called hole burning.

Meanwhile, the weak probe beam is burning a weaker hole in the Maxwellian velocity distribution. However, when the laser frequency is detuned from the natural resonance (|ν − ν₀| ≫ γ), the probe absorption comes from a completely different set of atoms than the pump absorption: the atoms moving at the opposite velocity. In this case, the pump beam does not affect the population of the relevant atoms for the probe, and the probe absorption follows the standard Doppler-broadened profile.

In the special case of the laser frequency near resonance, ν = ν₀, the pump burns a hole for atoms with almost zero velocity parallel to the beam path. The same group of atoms is also accessed by the probe beam. This results in a saturation absorption (or Lamb) dip in the absorption-versus-frequency plot (Figure 3.4). The dip arises from the following reasoning: when the laser is at resonance, the probe’s photons are likely to encounter atoms already in the excited state, due to the transition being saturated by the pump. Then, the photons may cause the atom to undergo stimulated emission, releasing an equal-energy photon. Thus, small dips in absorption occur at each transition (typically the hyperfine resonances). The equivalent dip can be seen in the fluorescence signal. Under ideal conditions (well-collimated beam, low magnetic field, sufficiently narrow laser linewidth), the dip will approach the natural linewidth of the atomic transition.
Figure 3.4: (a) Saturated absorption dips of two example transitions a, b with overlapping Doppler profiles. (b) The SAS signal with background Doppler profile subtracted.

### 3.3.1 Lamb dip width

To quantify the width of the Lamb dip, we define the saturation parameter

\[
S(\omega) = \frac{B_{12}(\omega)I}{cR_1(\omega)R_2(\omega)}
\]  

(3.8)

where \(B_{12}\) is the Einstein absorption coefficient, \(I\) is laser intensity, and \(R_1, R_2\) the relaxation rates (including spontaneous emission) that depopulate a level. This saturation parameter can be shown to equal the quantity \(S\) in Equation 3.7. Then, it can be shown that the Lamb dip has width (22)

\[
\gamma_s = \gamma \sqrt{1 + S(\omega_0 = 0)}
\]  

(3.9)

For purposes of using SAS to stabilize a laser, it is advantageous to choose a small saturation parameter to minimize the width. However, minimizing \(S(0)\) clearly also decreases overall signal strength.
3.3.2 Cross-over signals

The hyperfine transitions will not be the only structures visible in SAS, in general. When two transitions share a common lower or upper level and overlap within their Doppler width, extra resonances called cross-over signals will be observed.

If a laser operates at the average of two adjacent transitions, $\omega_1$ and $\omega_2$, the incident beam is shifted against $\omega_1$ by $\Delta \omega = \omega - \omega_1 = (\omega_2 - \omega_1)/2$, thus saturating the velocity class of particles $(\omega_2 - \omega_1)/2k \pm \gamma k$. This velocity class of atoms experiences resonance conditions. The reflected beam has the opposite Doppler shift and can cause an analogous velocity class of atoms to be resonant on transition 2. When 1 and 2 share a common lower level, both waves contribute to the decrease of $\Delta N_1$ at laser frequency $\omega = (\omega_1 + \omega_2)/2$. This causes a decrease in observed fluorescence at that frequency. We observe several cross-over signals in both Rb 87 and 85.

3.4 Implementation of SAS on Rb

The experimental realization of SAS is illustrated in Figure 3.5. Our diode laser source at 1560 nm is sent through non-polarization-preserving single-mode fiber to the PPLN waveguide for second harmonic conversion. Polarization is controlled using manual fiber polarization controllers, and set to optimize SHG output. The 780 nm output is collimated and sent through the Rb cell. The cell is heated to approximately 34 °C with adhesive resistors. It is contained in a cylinder of $\mu$-metal to shield against magnetic fields.

The beam is retro-reflected by a dielectric mirror optimized to 780 nm. To prevent instabilities in the laser source, a Faraday isolator blocks the reflected light from reaching the diode laser. A detailed configuration of the light’s path is shown in Figure 3.6.
Figure 3.5: The layout for saturated absorption spectroscopy.

Figure 3.6: The 780 nm light follows the trajectory A-B-C-D before it is reflected by a dielectric mirror and returns by overlapping the incident path. The 80 cm Rb cell is shown in black, with finite window thickness illustrated.
The 1560 nm laser is tuned across multiple Rb resonances by changing the drive current on the diode’s thermoelectric cooler (TEC). The TEC heats or cools the diode chip through the Seebeck-Peltier effect. As we sweep across the relevant transitions (using LabView software), we detect our SAS signals with a photodiode array that surrounds the Rb cell.

3.4.1 Photodetector design

To see a saturated absorption fluorescence signal, we collect the resonance fluorescence signal scattered from the laser beam path. Light travels down the Rb cell, interacts with atoms; emitted photons provide the signal detected by our photodiode array.

**Geometry of detector**

The array consists of 16 photodiodes (Advanced Photonix Solderable Silicon Photo-diodes). Each diode has dimensions 10 mm x 10 mm, and is red-enhanced with a responsivity of 0.55 A/W at 780 nm. The diodes are placed inside a copper tube which encircles the cylindrical Rb cell. On each end of the copper tube, eight diodes are attached in a ring configuration, enclosing the cell. The geometry is shown in Figure 3.7.

**Detector efficiency** To estimate the 780 nm power our detectors should be able to collect, we need to know the solid angle subtended by our photodetectors, as well as the angular radiation distribution of scattered light from the incident beam. For linearly polarized light, the angular distribution for resonance fluorescence is

\[ f(\gamma) = \frac{3}{8\pi} \sin^2(\gamma) \]

which is the classical radiation pattern for an oscillating dipole. \( \gamma \) is the angle between the polarization vector and the emitted photon’s direction.
Figure 3.7: The front and side view of the mount for the silicon photodiodes, used to detect fluorescence. There are eight diodes pictured (in black), each attached to the copper tube that will surround the Rb cell.

Treating every point on the beam line as a radiating source, we can integrate Equation 3.10 over the solid angle that the detectors subtend. Let the polarization be in the $\hat{x}$ direction, and the beam propagation be in $\hat{z}$. Figure 3.8 lays out the geometry of the setup.

We use the law of cosines to find $\gamma$ in terms of $\phi, \theta$.

$$
\cos(\gamma) = \frac{\sin^2(\phi) - \cos^2(\phi) + \cot^2(\theta) - \csc^2(\theta)}{2 \cos(\phi) \csc(\theta)} 
$$

To calculate detection probability $P(z)$ of a photon emitted at $z$, the angular distribution is integrated over the solid angle subtended by the detectors.

$$
P(z) = \frac{3}{4\pi} \int_0^{\phi_1} \int_{\theta_0}^{\theta_1} \sin^2(\gamma) \sin(\theta) d\theta d\phi
$$

where $\theta_0 = \arctan(r/(A + d - z)), \theta_1 = \arctan(r/(A - z))$ label the position of the detector. $\phi_1 = 4\pi/5$ is the azimuthal position of the bottom-most detector on the
Figure 3.8: The incoming light polarization is in the \( \hat{x} \) direction, and light travels in the \( \hat{z} \) direction. The beam within the Rb cell extends from \( z = 0 \) to \( z = 8 \) cm. The dipole radiation pattern is proportional to \( \sin^2(\gamma) \). Limits of integration are dictated by the location and dimensions of the detector. Each detector is at a radius \( r = 1.78 \) cm away from the beam line and has width of 1 cm. The two rings of detectors are located at \( A = z = 2.3 \) cm and 5.7 cm. Each ring is cylindrically symmetric about the \( z \)-axis.

Next, integrating over the entire 8 cm beam,

\[
P = 2 \int_0^8 P(z)dz \quad (3.13)
\]

with the assumption of low pump depletion (so that the first ring of detectors is equivalent to the second ring). We predict a 15% chance that a fluorescent photon will be absorbed by a detector.

**Circuitry**

We wish to obtain the aggregate signal of 16 photodiodes. The diodes are split into groups of four. For each group, the four diodes are connected as parallel current sources. A transimpedance amplifier converts the current signals into voltages, which are aggregated by a summing amplifier.
When evaluating the circuit performance, it can be shown that thermal Johnson noise will dominate any noise level at above roughly 10 Hz. The equation for Johnson noise is

\[ v_{rms} = \sqrt{4k_bTR\Delta f} \]  

at temperature T. The noise is \( \frac{v_{rms}}{\sqrt{Hz}} = 64 \text{nV/}\sqrt{Hz} \) at room temperature and with feedback resistance \( R = 250 \text{k}\Omega \). The four separate transimpedance circuits will contribute noise, added in quadrature, to give expected noise on the level of 100 nV/\( \sqrt{Hz} \).
Figure 3.10: A. View down the length of the Rb cell, which is enclosed by the photodiode array (the copper tube). Signals are carried by the ribbon cables. B. An interior view of the photodiodes.

A complete schematic of the circuit is shown in Figure 3.4.1 along with a picture of the completed photodiode array in Figure 3.10.

3.4.2 Results

For our SAS setup, incident 780 nm power ranged from 150-200 $\mu$W. Several hyperfine and crossover signals were detected on the oscilloscope (see Figure 3.11). For locking, we primarily utilized the $^{87}$Rb crossover signal of $F = 2$ to $F' = 1$ and 3. The FWHM of the transition used, as defined by Equation 3.9, is 7.2 MHz, which approaches the natural linewidth of 6 MHz. Details of the different signals, their strengths, and widths can be found in Table 3.4.2.
Figure 3.11: Saturated absorption dips in the resonance fluorescence signal of $^{87}$Rb and $^{85}$Rb. The top figure pictures data collected by our photodiode array; the bottom pictures data collected by a single photodiode, in an earlier setup. The labeled signals in $^{87}$Rb correspond to the electronic transitions between the initial state of $F = 2$ to final states of (a) $F' = 1$ to 3. (b) $F' = 2$ to 3. (c) $F' = 3$. 
Table 3.2: Table of SAS signals for the largest resonance or crossover (co) signals observed, in both $^{87}$Rb and $^{85}$Rb. The initial states are all from $5S_{1/2}$ and the specified F level. The final states are $5P_{3/2}$ $F'$, or a crossover to multiple $F'$ hyperfine states. The FWHM and signal strength are also listed. Signal strength is the amount of power from the SAS signal, with an incident $\approx 100 \, \mu W$ of 780 nm power on the cell.
Chapter 4

Locking to the atomic resonance

The electronic resonance is used to frequency-stabilize the laser in a modulation lock.
Our aim is to suppress low-frequency drift of the laser.

4.1 Modulation Lock Basics

The principle of modulation locking (or dither locking) is as follows: we modulate the laser signal across a resonance to generate an error signal, which is proportional to the difference between the reference (atomic) frequency and the possibly perturbed laser frequency. The error signal is fed back to the laser to lock its frequency to the reference. Our atomic resonance is an electronic transition between $5S_{1/2}$ and $5P_{3/2}$ involving different hyperfine states, as discussed in Ch. 3. In general, frequency references could also be Fabry-Perot cavity transmission signals. The technique of modulation locking is widely used because of its simplicity, stability against intensity fluctuations, and generally high signal-to-noise ratio.

The laser frequency $\omega$ is modulated (typically with a sinusoidal wave) by changing some parameter to the diode that controls laser frequency. There are many ways to achieve this. Laser frequency can be modulated externally through an acousto-optical or electro-optical modulator, but these methods can have a significant cost.
Figure 4.1: The mechanism to tune the diode laser frequency. A dc signal (VCA voltage) controls the RF power to the laser injection current, which effects a change (frequency excursion) of the central laser frequency. We always operate in the quasi-linear regime, shaded in green. An increase of 1 V corresponds to a 12.2 MHz increase in frequency.

in laser power. For certain external cavity diode lasers, a piezo-electric transducer (PZT) can phase-modulate the laser, or a diffraction grating mounted on the external resonator can selectively filter wavelengths. However, for our diode laser, a novel scheme involving heating of the diode chip is used to tune frequencies. We apply a high frequency RF signal near 800 MHz, which causes the center frequency of the laser to move. The RF signal changes the laser injection current by direct heating of the diode chip. RF power is controlled by a voltage-controlled attenuator (VCA); attenuation is set by the error signal. Figure 4.1 shows the change in laser frequency as a function of the VCA voltage. We always operate in the quasi-linear regime, where a dc heating signal of 1 V corresponds to a frequency increase of approximately 12 MHz.
To find the error signal, we analyze the layout pictured in Figure 4.2. The diode laser signal passes through the atomic gas cell, generating a fluorescence signal whose power is dependent on $\omega$ and has a SAS dip at some resonance, $\omega_0$. At first, we apply a slow-varying ramp voltage to find a hyperfine Rb resonance. We scan over almost the entire FWHM of the Doppler-broadened $^{87}\text{Rb}$ transition, sweeping over 100 MHz laser frequency with a modulation frequency of 0.1 MHz. As we draw nearer to $\omega_0$, we progressively decrease the ramp voltage, until we are confidently centered on resonance. The ramp voltage is taken off.

Meanwhile, a constant dither voltage of frequency $\Omega_d = 20$ kHz has been applied to the diode. This causes fast-varying changes in $\omega$ of $\Delta \omega \approx \pm 1$ Mhz. We can write the photodetector fluorescence signal as

$$V_p(\omega) \approx V_p(\omega_0) + \left. \frac{dV_p}{d\omega} \right|_{\omega_0} \Delta \omega \cos(\Omega_d t)$$  \hspace{1cm} (4.1)
in the case that $\omega_0 \gg \Delta \omega$. A lock-in amplifier can multiply $V_p$ by $\cos(\Omega d t)$ to obtain a signal of three frequency components: 0 (DC), $\Omega d$, and $2\Omega d$:

$$V_p(\omega) \cos(\Omega d t) = V_p(\omega_0) \cos(\Omega d t) + \frac{\Delta \omega}{2} \left( \cos(2\Omega d t) + 1 \right) \frac{dV_p}{d\omega}$$

(4.2)

from which the DC component can be extracted using a low-pass filter. The error signal is

$$\epsilon(\omega) = \frac{dV}{d\omega} \bigg|_{\omega_0}$$

(4.3)

The error signal is antisymmetric about $\omega_0$, which is desired for negative feedback.

$\epsilon(\omega)$ is the switch that sets the temperature-controlling modulation voltage. Through a voltage-controlled attenuator, $\epsilon(\omega)$ allows varying amplitudes of a 800 MHz voltage signal to enter the diode tuning port.

### 4.2 System Performance

#### 4.2.1 Error Signal

The ramp voltage, which broadly tunes the laser frequency with efficiency of 12.2 MHz/V, has a DC offset of 3.1 V and an amplitude of 1.0 V. The lock-in amplifier outputs an error signal that is the derivative of the fluorescence signal. The laser frequency swings about resonance to give an error signal shown in Figure 4.3.

As a demonstration of negative feedback, when the servo begins operation, the error signal is summed with the perturbing ramp voltage. This sum is fed back to the voltage tuning port of the diode, effectively canceling the perturbation. The method of active control keeps the frequency from drifting from the center of resonance. Figure 4.4 shows the error signal while the servo is operating.
Figure 4.3: Ramp voltage and error signal as a function of time, when the servo is off.

Figure 4.4: Ramp voltage and error signal as a function of time, when the servo is on. The sum of the ramp and error signal will set the VCA to control the power of the RF heating signal, which tunes the frequency of the laser.
4.2.2 Frequency Stability

The frequency instability is estimated by calculating the change $\Delta V$ of the error signal per unit modulation frequency, $\Delta \text{MHz}$. This quantity is inverted, then multiplied by the rms noise of the error signal. The latter is obtained observing the power spectral density of noise in the error signal. At 0.5 Hz, the frequency instability is $\approx 200 \text{ kHz}$ over 1 second averaging while unlocked, and $\approx 30 \text{ kHz}$ while locked. Integrating over all frequencies up to 0.5 Hz, low-frequency instability is $\approx 90 \text{ kHz}$ over 1 s averaging. The frequency power spectral density for the laser, both free-running and locked, is shown in 4.5.

With this approach, the laser can be locked to the resonant transition for durations of over 12 hours without problems. Temperature shifts in the laboratory can contribute to long-term instabilities.
Chapter 5

Conclusion

We have been able to lock a diode laser at 1560 nm to an atomic transition in Rb. By generating the second harmonic of the laser and using saturated absorption spectroscopy, we could use frequency modulation locking to actively stabilize the laser to a hyperfine transition in Rb 87. The low-frequency instability of the laser center frequency when locked is approximately 90 kHz with 1 s averaging. The atomic transition in $^{87}$Rb is excellent for an absolute frequency reference. However, a narrower resonance could be obtained by a Fabry-Perot cavity. Locking to such a highly stable optical reference cavity can reduce the spectral linewidth of the laser, and provide better frequency stability on shorter timescales (or frequencies greater than 0.1 Hz). The Rb SAS signals are an excellent frequency reference for longer time scales (frequencies less than 0.1 Hz).
Bibliography


