Laser Locking with Doppler-free Saturated Absorption Spectroscopy

Paul L. Stubbs, Advisor: Irina Novikova
W&M Quantum Optics Group

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Abstract

The goal of this project was to lock the frequency of a 795 nm diode laser using a saturated absorption spectroscopy method. Laser locking in AMO physics is done to stabilize the frequency of lasers used in the laboratory in order to make results more reliable and reproducible. Locking the laser frequency to a particular absorption resonance requires creating a feedback loop, in which an error signal, proportional to the laser detuning from the desired frequency, is fed back to the laser controller to adjust accordingly. The laser was first locked to a rubidium resonance generated from basic linear absorption. In order to enhance results, a lock was acquired in our Doppler-free saturated absorption spectroscopy experiment with rubidium, and then refined with the use of Zeeman modulation. Well-defined atomic transitions of the outer valence electron of ^{87}Rb and ^{85}Rb between the $5^2S_{1/2}$ and the $5^2P_{1/2}$ states corresponds to a separation of roughly 795 nm (D1 line), the wavelength of the diode laser used for this experiment. Utilizing the hyperfine structure of rubidium made visible with this method, allowed for a precise and stable lock of the laser frequency.

Theory overview

Laser locking is essentially the stabilization of the laser's frequency using some stable frequency reference. In Atomic, Molecular and Optical (AMO) physics, the stabilization of the lasers used in the laboratory through locking is an important step in attaining more reliable and reproducible results. While there are many ways to stabilize the frequency of a laser, atomic absorption lines are particularly accurate because of the well-defined nature of optical frequencies of atomic transitions [1].

Laser Spectroscopy

The energy associated with transitions for a valence electron of an atom is given by difference between the excited state energy and the ground state energy, which used with Planck's law, gives the frequency v_0 of transitions between these two states:

$$\Delta E = \hbar v_0 \tag{1}$$

When an electron in an excited state decays down to the ground state of the atom, it emits a photon of frequency v_0 [2].

An atom can undergo three basic transition processes: stimulated absorption, stimulated emission, and spontaneous emission. Stimulated emission and absorption are related to interactions with a laser or other forms of electromagnetic fields. Spontaneous emission is a transition that occurs regardless of any sort of laser field and the frequency, direction, and polarization of emitted photons does not correspond to the external field.

The method for saturated absorption spectroscopy of rubidium allows for very precise measurements of the hyperfine structure of these atoms. From basic laser spectroscopy, one is able to observe Doppler

broadened absorption. As adjustments are made, it becomes possible to resolve the saturated absorption lines that correspond to specific atomic transitions and crossover resonances.

An idealized two-level atom consists of a ground state and an excited state. Passing a single laser beam through a vapor cell of a two-level gas results in basic laser absorption spectroscopy. This affects the number of atoms in the ground state and excited state, but the random thermal motion of absorbing atoms causes Doppler broadening. The horizontal velocity parallel to direction of laser propagation, v_z, of these atoms causes a Doppler-shift of the resonance frequency ν_0, as described by the equation below.

$$\nu_0' = \nu_0 \left(1 + \frac{v_z}{c} \right) \tag{2}$$

In order to counteract this Doppler broadening and obtain higher resolution, a second counter-propagating "pump" beam is passed through the cell. The Doppler shifted resonance frequency for atoms moving with velocity v_z is opposite that of the probe beam. That is, the pump beam interacts with the group of atoms with velocities $-v_z$, exactly opposite the group the probe beam interacts with. The large absorption dips are only seen when the laser frequency is on resonance and both beams interact with the same group of atoms, which effectively picks out atoms with zero velocity and eliminates Doppler broadening. A strong enough pump beam at a resonant frequency causes rapid transitions that evenly distribute the population of atoms between the ground and excited state. When this occurs, the laser is said to saturate the transition, hence the name saturated absorption [2]. Figure 1 shows the density of atoms in the ground state versus their velocity when the frequency of the pump beam is greater than resonance, on resonance and less than resonance. The largest saturated absorption dip is seen when the pump is on resonance, a process called "hole burning."
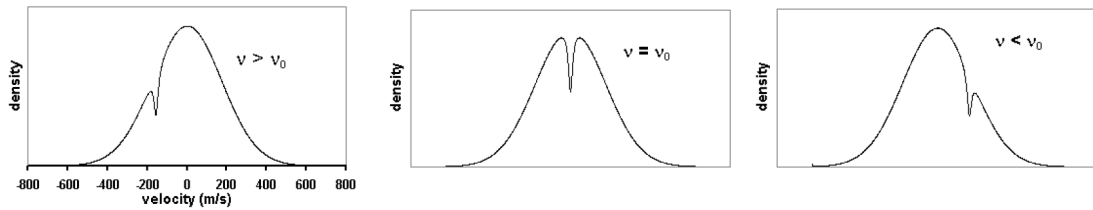

Figure 1: Atom density in the ground state plotted versus velocity for the frequency of the pump beam. Resonance frequencies are seen Doppler-shifted in the left and right picture [2].

Rubidium atoms and multilevel effects

Actual atomic systems are more complex than the two-level system considered before. The basic energy levels of hydrogen and hydrogen-like atoms, such as rubidium, are further corrected by looking at the effects of both the fine and hyperfine structures. Rubidium is an alkali atom with a single free electron outside of closed shells and has the following electron configuration: $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 5s$. The two isotopes of rubidium, 28% abundant ^{87}Rb and 72% abundant ^{85}Rb, have a nuclear spin quantum number of $I = 3/2$ and $5/2$, respectively. The fine structure splitting in rubidium is described by spin-orbit coupling and accounts for the difference in separation for the excited $5^2P_{1/2}$ and $5^2P_{3/2}$ states. The hyperfine structure is due to the dipole moment of the proton in the nucleus and its interaction with the orbiting electron [3]. This results in a number of hyperfine states within the ground and two excited states. Transitions of the valence electron in both isotopes of rubidium will be examined using a 795 nm tunable diode laser. The separation between the $5^2S_{1/2}$ and the $5^2P_{1/2}$ state of rubidium is 794.764 nm, often called the D_1 transition line [3].
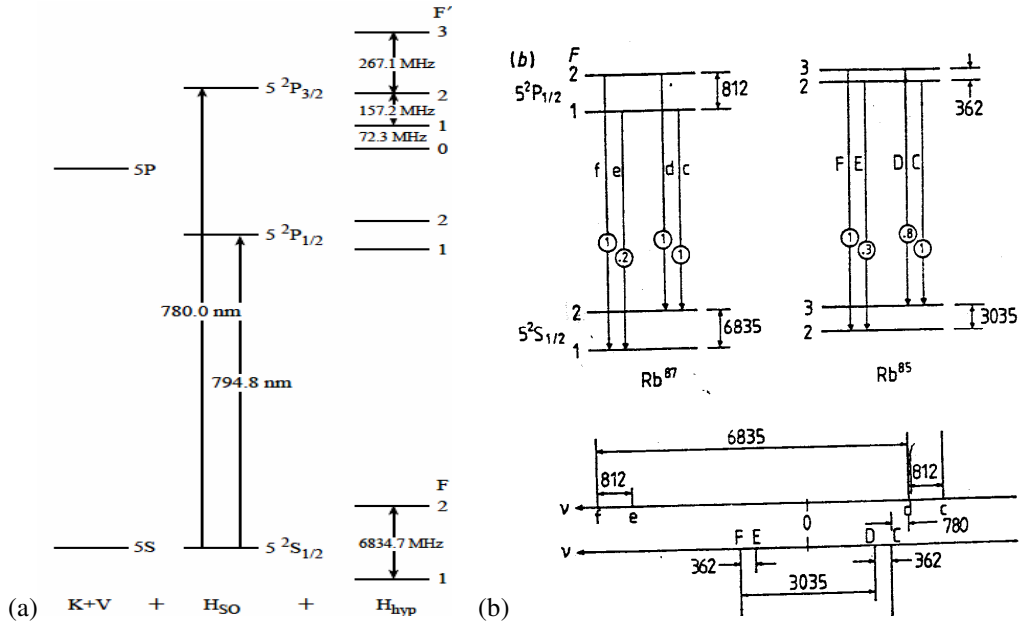
Figure 2: (a)Energy level diagram for rubidium showing splitting due to the basic Hamiltonian, the spin-orbit interaction, and the hyperfine splitting (Rb[87] shown) [3]. (b) Fine and hyperfine splitting for rubidium between the $5^2P_{1/2}$ and $5^2P_{1/2}$ states (795 nm D_1 line). The lower part gives the relative position of the hyperfine lines in MHz [4].

With these multi-level atoms, additional variables such as optical pumping and crossover resonances must be considered. Crossover resonances are additional absorption dips that are seen due to the overlapping of Doppler-broadened signals from the extremely close splitting of energy levels [2]. The pump beam then interacts with two different groups of atoms at the same time, as does the probe beam. As long at these groups of atoms are within one Doppler width of each other, when the laser frequency is increased there will be a point where the same group of atoms is resonant with different probe and pump resonance frequencies, and vice versa. Optical pumping happens when an excited atom has the ability to spontaneously decay to more than one lower energy level that may or may not interact with the incident laser beam. When atoms are excited and then decay to a non-resonant lower level, the population of atoms in the resonant ground state becomes depleted and causes an increase in the size of the saturated absorption signal [2].

Saturation absorption spectroscopy provides the narrow transmission peaks within much broader Doppler-broadened absorption lines in atomic vapor. Since these atomic transitions are so well-defined, it makes this technique particularly useful for laser locking.

Lock-in Amplifier

The lock-in amplifier will be used within the laser-saturated absorption spectroscopy experiment. Locking the laser frequency to a particular absorption resonance requires creating a feedback loop, in which an error signal, proportional to the laser detuning from the desired frequency, is fed back to the laser controller to produce the required adjustment. The use of a lock-in amplifier takes the signal from the photodiode and multiplies it by a sine wave at the reference frequency. This gives a proportional DC output signal corresponding to the proportion of the signal from the diode that was locked to the reference already [5]. Integrating that provides the required error signal which is sent to the laser controller to make the correct adjustment.

Fourier's theorem states that if some parameter of the original system is modulated at frequency ν, then the output signal can be represented by the sum of numerous sinusoidal functions. This signal can be represented mathematically as:

$$signal = noise + s_1 \sin(vt) + s_2 \sin(2vt) + ... \qquad (3)$$

As stated before, the lock-in then multiplies this signal by a sinusoid at the reference frequency, v, which upon integrating yields $(1/2)s_1$, where s_1 is the DC error signal proportional to the detuning that is sent to the laser control box to make any small correction and stabilize the laser frequency.

$$\lim_{T \to \infty} \frac{1}{T} \int_0^T (signal \cdot \sin vt) dt = \frac{1}{2} s_1 \qquad (4)$$

The lock-in operation is based on this phase-sensitive detection, mathematically described above. When multiplying the input signal by a sine function at the reference frequency and integrating, the part of the signal that is not at the reference frequency or the correct phase is discarded. For this reason, lock-in amplifiers are also referred to as phase-sensitive detectors. The part of the signal at a specific reference frequency and phase is picked out and used as feedback to stabilize the system [6].

Experimental setup and procedures

There are two major parts of the research that were completed and within each part the laser was locked to resonances of rubidium. The two parts were the standard Doppler-free saturated absorption and then the use of a solenoid to lock the laser with Zeeman modulation. The first dithers the laser frequency and utilizing that error signal to lock the laser, whereas Zeeman modulation dithers the actual resonances. Each of these setups utilized the same lock-in amplifier but required a few different components to conduct the experiment.

There is little extra equipment when working with a lock-in amplifier, but there are a number of components used for the saturation spectroscopy setup. The extended cavity diode laser ECDL-7960R was already in place, but the lock-in amplifier LIHV200 was incorporated, shown in Figure 3.

Figure 3: Lock-in amplifier LIHV200 without photodiode INPUT connection

As a preliminary test, we locked the laser to an atomic resonance using a simpler single-beam configuration. The output of photodetector was hooked up to the input of the lock-in amplifier. The modulation of the LI was connected to the control box of the laser, with the monitor output sent to an oscilloscope for observation and another output signal with the PID-control to the laser input. We tuned the laser frequency to find a reference resonance of the rubidium and used the lock-in with that resonance. By adjusting the SWEEP knob we were able to achieve an effective lock.

For the saturated absorption spectroscopy part of the experiment, the diode laser (ECDL-7960), a beam splitter and adjustable mount, two flat mirrors with mounts, a Rb cell, and a differential photodiode setup were needed. As the diode laser and the Rb cell already existed from the previous setup, a minimal amount of rearranging was required to achieve the setup pictured in the schematic below. The specific setup used for this experiment eliminates Doppler broadening in measurements made and will allow for Doppler-free saturated absorption. Three beams of light are derived from the same laser using a series of mirrors and a beam splitter, which pass the beams through a cell containing a low-density vapor of rubidium atoms [7].
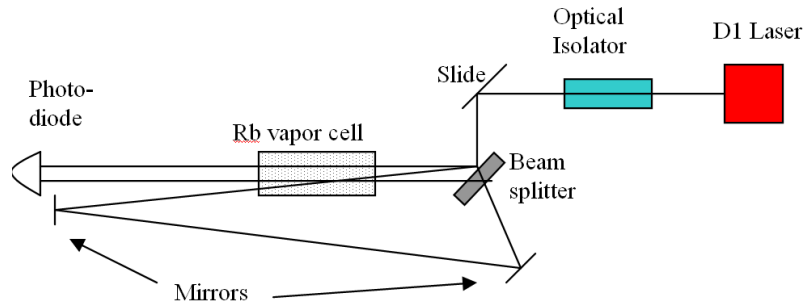
Figure 4: Schematic of diode laser-saturated absorption spectrometer.

The schematic of the experimental setup for Doppler-free saturated absorption spectroscopy is shown in Figure 4. The frequency of the laser can be varied by changing to voltage across the piezoelectric transducer (PZT) inside laser assembly in order to sweep through the atomic absorption lines [7]. The laser then passed through the optical isolator, reflected off the slide and was sent to the beam splitter. The beam splitter splits the incident beam into three beams, the more intense counter-propagating "pump" beam, and two weaker "probe" beams, arranged such that they all pass through the Rb cell. Mirrors were placed, one behind the beam splitter and the other at the far end of the cell, to reflect the pump beam which was made to overlap one probe beam within the sample. The two probe beams were denoted the "overlap" beam and "reference" beam. The subtraction of the overlap and reference beam circumvents Doppler broadening, leading to Doppler-free laser-saturated absorption spectroscopy.
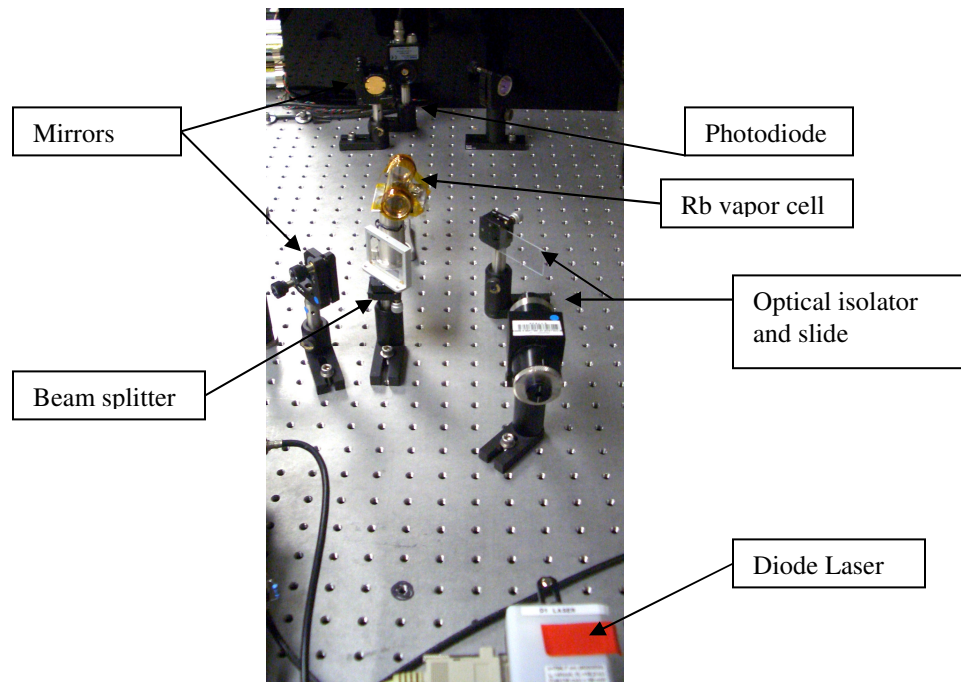

Figure 5: Constructed saturation spectroscopy setup on optical table.

The setup for the Doppler-free saturated absorption spectroscopy is shown in Figure 5 above with each component labeled. The optical isolator only allows the transmission of laser light in one direction and prevents the light from reflecting back into the optical cavity of the laser. The single photodiode at the top of the picture needed to be replaced with a differential photodiode, in order to subtract the signal from the overlap and reference probe beams, which will be discussed shortly. The laser beam traveled to the slide, where 90% was transmitted toward the far mirror and another laboratory experiment, and the other 10%

was reflected to the beam splitter and this experiment. This setup incorporated the beam splitter which created two probe beams and the additional mirrors which were needed to reflect the pump beam.

The three beams the laser is broken into, other than the one sent to another experiment are shown below in Figure 6. The more intense counter-propagating pump beam comes from the laser passing through the beam splitter. The two weaker probe beams, formed by reflections from the beam splitter, pass through the Rb cell where the pump beam overlaps one probe beam. When the overlap beam is subtracted from the reference beam using a differential photodiode, Doppler broadening in these measurements is eliminated.
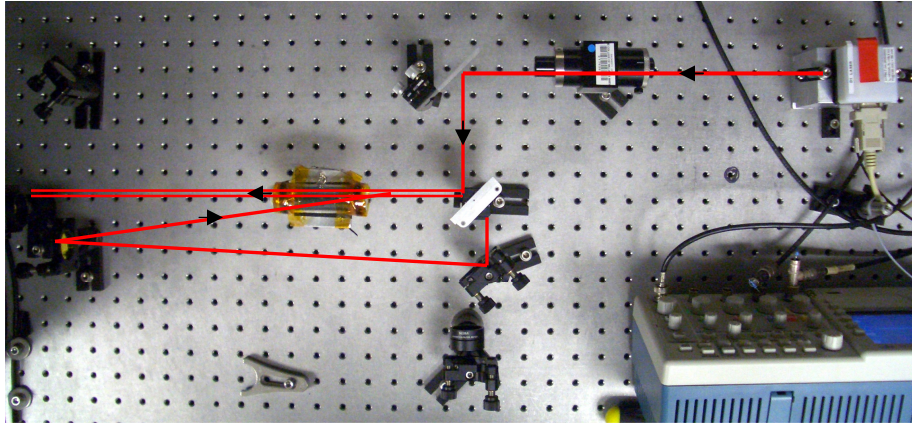

Figure 6: Laser path for Doppler-free saturation spectroscopy.

The preliminary single laser-beam configuration lock was successful as mentioned previously. A lock was achieved with this new setup while utilizing the differential photodiode circuit. Shown below in Figure 7 are the two photodiodes that detected the overlap and reference probe beam signals. Since the two beams come in parallel to the detector, it was necessary to remove the top PD in the picture and place it next to the other one so that each PD could read their respective probe beam simultaneously. Once this was accomplished, the differential PD was connected to the lock-in amplifier and allowed for the observation of some absorption lines, specifically the Doppler-free saturated absorption lines when everything was correctly aligned.
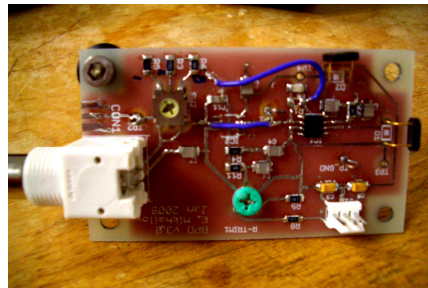

Figure 7: Differential Photodiode circuitry with BNC output.

The adjustment for the saturation spectroscopy with Zeeman modulation first required making a solenoid. This was done by wrapping .75mm wire around a plastic spool which was constructed specifically to fit the Rb cell. The solenoid consisted of 7 x 120 turns, made with .75mm copper wire, which in theory should give a potential magnetic field of roughly 117.29 G/A. Sending an alternating current through the solenoid will dither the resonances themselves and allow for an even more precise lock of the laser.
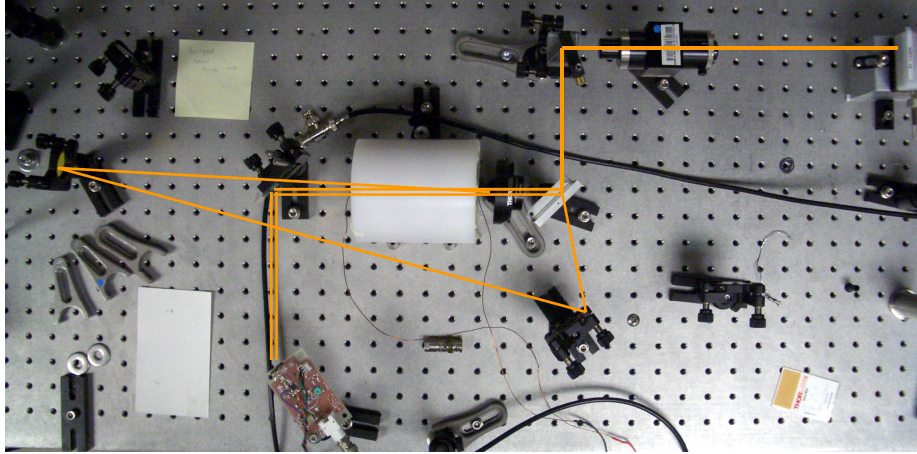
Figure 8: Saturation Absorption Spectroscopy setup with solenoid.

The saturation absorption spectroscopy with Zeeman modulation is shown in Figure 8 below. A beam cube was placed after the scope which the laser first passed through. A quarter-wave plate, which makes the laser light circularly polarized, was placed after the beam splitter and acted to attenuate the probe beams. The large white cylinder in the middle of the image is the solenoid mentioned before, which is attached to a function generator that provided the modulation for the Zeeman splitting of the rubidium atoms. The function generator was set at a frequency of 10 kHz. The differential photodiode is seen at the bottom of the image and subtracts the Doppler-broadened profile from the absorption lines to acquire the Doppler-free atomic absorption lines desired.

Results

Figure 9(a) shows the Doppler-broadened spectrum lines when the overlap beam is blocked. This is equivalent to ordinary laser absorption spectroscopy, where a single laser beam passes through a vapor cell to a photodiode. As the laser passes through the cell, the stimulated emission and absorption processes change the intensity of the laser measured by the photodetector and Doppler-shifts of the rubidium atoms cause the signal to be Doppler-broadened. The two signals of larger amplitude are of ^{85}Rb; whereas the outer two dips correspond to absorption in ^{87}Rb. The reason that the signal from ^{85}Rb is considerably larger than ^{87}Rb is because it is roughly three times more abundant. The larger of the two signals on the right, about 55 mV, corresponds to the F = 3 to F' = 2, 3, transitions whereas the smaller dip of about 35 mV is for the F = 2 to F' = 2, 3, both for the more abundant ^{85}Rb.
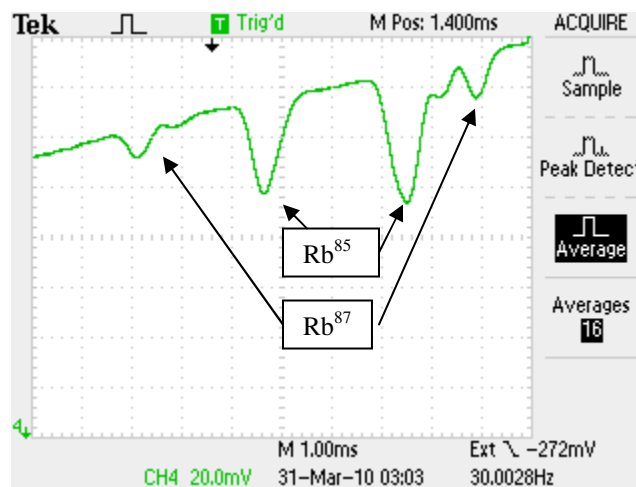

Figure 9(a): Doppler-broadened absorption lines

When the reference beam is blocked so that only the overlap beam is detected, the resulting picture is that of saturated absorption spectroscopy. As shown in Figure 9(b), the saturated absorption lines are seen on top of the two Doppler-broadened signals seen in the previous image. These small absorption peaks correspond to the hyperfine splitting of excited levels in rubidium, in contrast to the smooth signal from basic absorption spectroscopy. This begins to exhibit the refined precision allowed by saturated absorption spectroscopy. The hyperfine energy level transitions due to the D1 (795 nm) laser are shown.
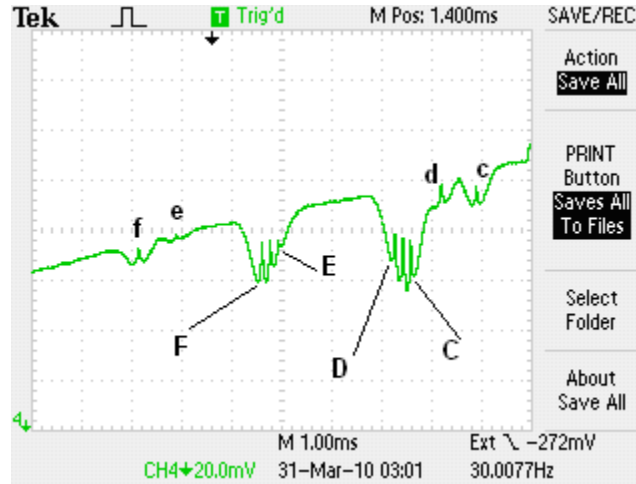

Figure 9(b): Doppler-broadened absorption lines with hyperfine structure. Labels on the resonances correspond to the notation in Figure 2.

Figure 9(c) shows the subtraction of the signals in the previous two graphs, (a) and (b), which results in showing the Doppler-free saturated absorption lines. This is because the Doppler-broadened absorption of the first graph is subtracted from the same graph with hyperfine structure. Subtracting off the Doppler provides the Doppler-free nature desired of the data. The differential photodetector circuit shown earlier subtracts the signals that arise from both the overlap and reference beam hitting each photodiode. Each absorption peak shown below corresponds to a specific resonance or crossover resonance for rubidium. Looking at the figure below, from right to left are the resonances and crossover resonances of ^{87}Rb, ^{85}Rb for both large signals, and ^{87}Rb for the left most peaks.
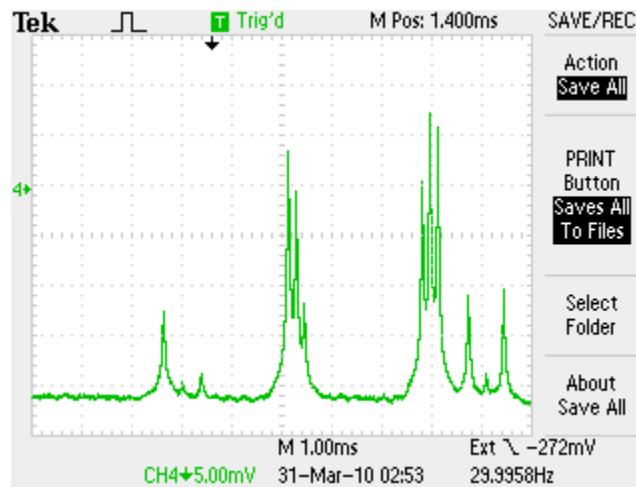

Figure 9(c): Doppler-free saturated absorption lines.

Using a function generator, an AC current was applied to the solenoid in order to shake the resonance. Each resonance was seen to shift slightly when the modulating current was applied. This dithering of the

resonances through Zeeman modulation provided the error signal to lock the laser. The resonance that shifted greatest when the magnetic field was on was most likely to generate a stable frequency lock. This turned out to be the F = 1 to F' = 2 (f) transition of ^{87}Rb.
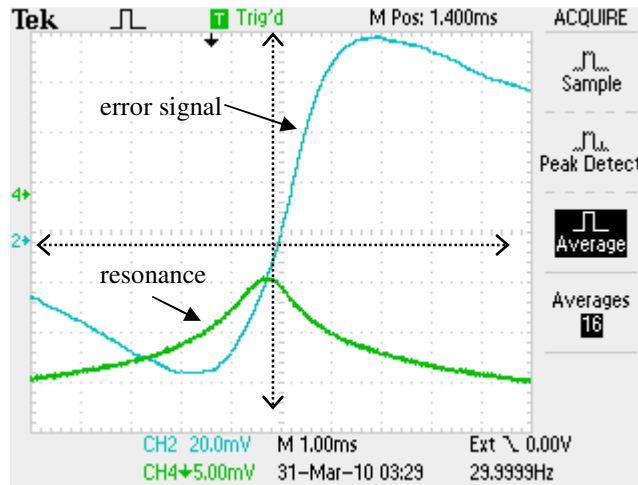

Figure 10: CH2: Error signal CH4: Rubidium resonance peak.

The resonance above in Figure 10 was used along with the sinusoidal error signal shown to lock the laser. The error signal crosses the x-axis at the peak of the resonance as expected since it is the derivative signal of the resonance. This error signal was used as the external modulation for the laser controller, essentially fed back to the controller to make the necessary correction of laser frequency. Once switching the laser control box over to the EXT MOD mode, which used the signal from the lock-in amplifier to adjust the laser frequency and locked it to the top of the resonance. This is shown below in Figure 11, where the error signal is hovering right around zero volts as expected and the laser lock is complete.
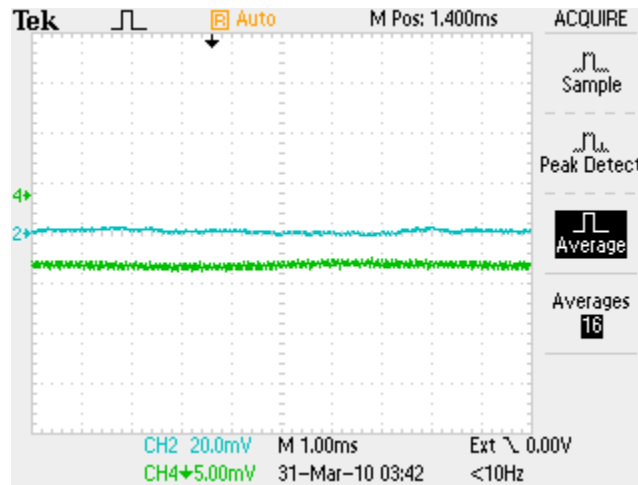

Figure 11: Locked laser signal with roughly zero error signal.

Conclusion

The use of laser locking in AMO physics to stabilize lasers in the laboratory makes results more reliable and reproducible. Locking the laser frequency to a particular absorption resonance requires creating a feedback loop, in which the lock-in amplifier provides an error signal proportional to the laser detuning from the desired frequency, and produces the required adjustment when fed back to the laser controller. The Doppler-free saturated absorption spectroscopy experiment utilizes this technique and allows for a much more precise laser lock. The transition of the outer electron of ^{87}Rb between the $5^2S_{1/2}$ and the $5^2P_{1/2}$ states corresponds to a separation of roughly 795 nm, the wavelength of the diode laser used for this experiment.

For the Doppler-free saturated absorption spectroscopy part of the experiment, the diode laser was broken into three beams, a more intense pump beam and two less intense probe beams. The counter-propagating pump beam overlapping one probe beam within the Rb cell, when incident on the differential photodiode, eliminated the Doppler broadening found in measurements done with the simpler single-laser configuration. This was from the subtraction of the Doppler-broadened absorption signal seen without the overlap beam from the signal of just the overlap beam.

The Rb cell was then placed inside the solenoid, to which an AC current was applied by a function generator in order to shake the resonance. Each resonance shifted slightly when the modulating current was applied and this dithering of resonances through Zeeman modulation provided the error signal to lock the laser. The resonance that shifted greatest when the magnetic field was on determined the one to which we chose to lock the laser, which happened to be the F = 1 to F' = 2 transition of ^{87}Rb. The benefit of utilizing the Zeeman modulation over a dithering of the laser frequency to provide the error signal is that finer splitting can be observed when an external magnetic field is applied than can be obtained through a modulation of the laser frequency. This results in a more precise error signal and a similarly precise laser lock without disturbing the exact instrument which is trying to be stabilized.

Quantifying just how well the lock-in amplifier is doing to stabilize the laser frequency would be a potential next step when looking for ways to improve the overall performance of the lock as well as the longevity of the stabilization after switching the laser control box to external modulation. However, the laser was successfully locked to a resonance of basic absorption spectroscopy, and then for Doppler-free saturated absorption spectroscopy without and then with Zeeman modulation.

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