Doppler-Free Saturated Absorption Laser Spectroscopy

Lisa M. Larrimore

The fundamentals of Doppler-free saturated absorption spectroscopy were explored for a rubidium sample. Two probe laser beams from a 780-nm diode laser were directed through the Rb cell, and one of them was crossed by a pump beam in the opposite direction. Because of the changes in the ground-state population caused by the pump beam, subtraction of the intensity signals from the two probe beams resulted in a non-Doppler-broadened signal. The three Doppler-free peaks corresponded to three of the ground state levels of Rb: the $F = 2$ energy level of ^{87}Rb, the $F = 3$ energy level of ^{85}Rb, and the $F = 2$ energy level of ^{85}Rb. These three peaks were separated by 0.0575 ms and 0.145 ms in our data. The theoretical separations of 1.2 GHz and 3.1 GHz both give the same calibration: 21 GHz/ms. This means there is a 24% error in our rough estimate of the calibration at 16 GHz/ms. Using the 21 GHz/ms calibration, the FWHM of the peaks were 525 MHz, 630 MHz, and 735 MHz, which are all above the Doppler limit of 500 MHz. Errors in this experiment included mistakes during the calibration, the improper use of AC coupling, and the wide bandwidth of the diode laser.

Introduction

The technique of Doppler-free saturated absorption spectroscopy was developed by the research group of Arthur L. Schawlow, who was one of the recipients of the 1981 Nobel prize in physics for this work [1]. Before the development of this technique, Doppler widths of about 500 MHz, caused by the random thermal motion of the atoms being studied, limited the resolution of optical spectroscopy [2]. Doppler-free saturated absorption spectroscopy is insensitive to this effect, and thus allows the splitting between energy levels of a sample to be measured with much greater precision. This has allowed previously unobservable atomic and molecular interactions to be measured. For this experiment, the hyperfine splitting of rubidium was first examined using traditional spectroscopy, and then measured beyond the Doppler limit using saturated absorption spectroscopy.

Our rubidium sample can be theoretically treated as a hydrogen-like atom because of its single valence electron. It thus has predictable fine and hyperfine corrections to the Hamiltonian, resulting in a splitting of its energy levels. Rubidium can absorb radiation when the frequency corresponds to a difference in energy levels, exciting the valence electron to a higher energy. This decreases the intensity of radiation that passes through the sample, an effect that is measured with an oscilloscope. By determining the frequencies at which the sample absorbs radiation, an absorption spectrum can be obtained. This technique is known as spectroscopy, and it allows the different energy levels to be measured.

The absorption spectra in traditional linear spectroscopy are characterized by broad signals around each absorbed frequency, despite the fact that the atom can only absorb at specific frequencies. This is a result of the Doppler effect; the frequency of the radiation in the frame of the atom is different from the frequency in the laboratory frame. By sending a pump beam through the sample in the opposite direction as the probe beams, the resonant frequency can be precisely determined. This is known as saturated absorption spectroscopy because tuning the laser above or below the resonant frequency will cause saturation of atoms moving at a certain speed, in either the positive or negative direction. When the laser is tuned to the resonant frequency, the difference between the probe beam that passes through the pump beam (which is unable to be absorbed by the already-excited atoms at rest) and the probe beam that is less affected by the pump beam gives the Doppler-free emission spectrum.

Theory

Atomic Structure of Rubidium

The Hamiltonian for an atom with a single valence electron can be written as

$$\hat{H} = \hat{K} + \hat{V} + \hat{H}_{SO} + \hat{H}_{1,hyp} + \hat{H}_{2,hyp}. \tag{1}$$

$\hat{K}$ represents the kinetic energy, $\hat{p}^2/2m$, and $\hat{V}$ represents the potential energy, $Z_{\text{eff}}e^2/4\pi\epsilon_0\hat{r}$. The fine structure comes from the spin-orbit coupling term, $\hat{H}_{SO} = \zeta(r)\hat{\boldsymbol{L}} \cdot \hat{\boldsymbol{S}}$, and the final two terms are hyperfine corrections due to magnetic and quadrupole interactions [1].

In its ground state, Rubidium has a single valence electron in the $5^2S_{1/2}$ state, with quantum numbers $n = 5$, $l = 0$, and $j = 1/2$. In Rubidium's first excited state, this electron jumps to the $5^2P_{1/2}$ or $5^2P_{3/2}$, where $n = 5$, $l = 1$, and $j = 1/2$ or $j = 3/2$. Since the spin-orbit interaction can be expanded as

$$\hat{H}_{SO} = \varsigma(r)\frac{\hbar}{2}[j(j+1) - l(l+1) - s(s+1)], \tag{2}$$

the fine splitting between the $5^2P_{1/2}$ and $5^2P_{3/2}$ levels is given by $\varsigma(r)3\hbar^2/2$. This splitting is illustrated in the left side of Figure (1). As shown, the wavelength of radiation corresponding to the energy difference between the $5^2S_{1/2}$ and $5^2P_{3/2}$ energy levels is 780 nm [1]. Since a 780-nm diode laser was used for this experiment, this transition was measured; transitions involving the $5^2P_{1/2}$ state were not examined.

The splitting of these fine structure energy levels into hyperfine structure is related to the angular momentum, $\boldsymbol{F}$, which is the sum of the electron angular momentum $\boldsymbol{J}$ and nuclear angular momentum $\boldsymbol{I}$. The rules of addition of angular momentum dictate that the possible values of the quantum number F are $|J - I|$, $|J - I + 1|$, $\ldots$ $J + I$. Because rubidium nuclei are not spinless, natural Rb consists of two isotopes: 28% abundant ^{87}Rb, with nuclear spin quantum number $I = 3/2$, and 72% abundant ^{85}Rb, with nuclear spin quantum number $I = 5/2$ [2]. We thus see that the for the $5^2S_{1/2}$ and $5^2P_{1/2}$ states of ^{87}Rb, $F = 1, 2$, and for ^{85}Rb, $F = 2, 3$. Similarly, for the $5^2P_{3/2}$ state, $F = 0, 1, 2, 3$ for ^{87}Rb, and $F = 1, 2, 3, 4$ for ^{85}Rb. From now on, F will be used to describe splittings within the $5^2S_{1/2}$ state, and F' will be used to describe splittings within the $5^2P_{3/2}$ state. The energy levels corresponding to these values of F are illustrated on the right side of Figure (1). During an electric dipole transition, F cannot change by more than one; for instance, the transition $F = 3$ to $F' = 1$ is not allowed [1].

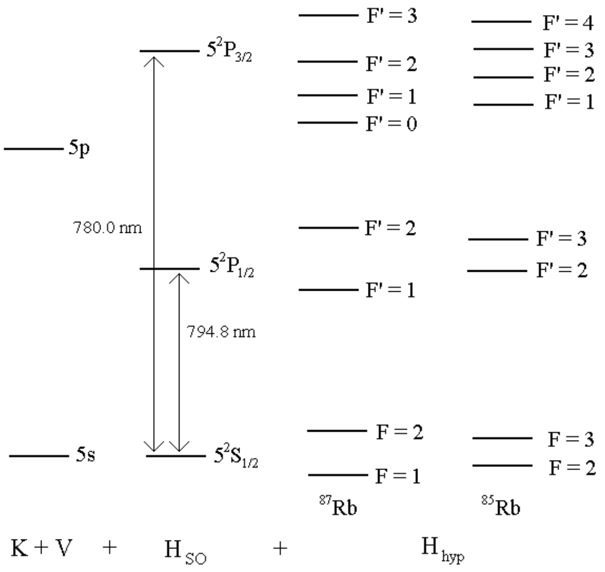

Figure 1: The interactions from Eq. (1) and their effect on the energy levels of rubidium, shown for both ^{87}Rb and ^{85}Rb. For this experiment, the transition between $5^2S_{1/2}$ and $5^2P_{3/2}$ were measured using a 780-nm laser. (Note: these energy level splittings are not drawn to scale.)

Doppler Broadening and Absorption Spectroscopy

If the radiation from the laser, oriented along the z axis, has a frequency ν_L in the lab frame, atoms moving at velocity v_z with respect to the lab frame will see a frequency ν_A such that

$$\nu_L = \nu_A \left(1 + \frac{v_z}{c}\right). \tag{3}$$

When ν_A equals one of the resonant frequencies of the atom, ν_o, the atom will absorb the radiation to jump to an excited state. This means that atoms moving towards the laser will absorb at a frequency that, when measured in the lab frame, is lower than their resonant frequency, and atoms moving away from the laser will absorb at higher than their resonant frequency. The measured absorption spectrum will thus be characterized by broad signals around each absorbed frequency. If the atom velocities are given by a Maxwell distribution, the full-width at half maximum amplitude (FWHM) of the Doppler-broadened signal is given by [1]

$$\nu_{FWHM} = 2\frac{\nu_o}{c}\left(\frac{2kT}{M}\ln 2\right)^{1/2}, \tag{4}$$

which is about 500 MHz for rubidium [2].

In Doppler-free saturated absorption spectroscopy, the absorption spectra of two probe beams are measured, as described above. One of the probe beams, however, is interacting with a different group of atoms, since it is crossed by a pump beam of identical frequency in the opposite direction. This pump beam changes the ground state population of the rubidium atoms in two ways: hyperfine pumping and saturation [1].

Hyperfine pumping occurs when excited atoms can decay to a different ground state than they started in. Whether transitions are allowed is determined by the selection rule $\Delta F = 0, \pm 1$. For instance, ^{85}Rb atoms that start in the $F = 3$ state and are excited to the $F' = 2$ excited state can decay back to the $F = 3$ state or the $F = 2$ ground state. If a group of atoms are subjected to a beam of the frequency corresponding to the energy difference between the $F = 3$ and $F' = 2$ states, a larger than normal percentage will soon end up in the $F = 2$ ground state.

The other method by which the pump beam changes the ground state population levels is saturation, which means that a smaller than normal percentage of atoms will be in the ground state because more will have recently been excited. In the above example of the $F = 3$ to $F' = 2$ transition, the pump beam will further lower the population of $F = 3$ ground state because more atoms will be in the $F' = 2$ state. This is a weaker effect than hyperfine pumping because the latter occurs at a much lower intensity [1].

The Doppler-free signal is given by subtracting the two probe beams, which gets rid of their common Doppler-broadened absorption and gives the difference caused by the probe beam. It is important that the probe beam is going in the opposite direction of the pump beam, which means that it generally interacts with different atoms than the pump beam due to the Doppler effect. For instance, an atom moving towards the source of the probe beam will see it as having a much higher frequency than the pump beam. If the frequency of the two beams is higher or lower than a resonant frequency of the atoms, then the pump beam will not affect the probe beam signal. When the laser is tuned to a resonant frequency, however, the pump beam will lower the percentage of atoms that the probe beam can interact with, so fewer atoms will absorb the probe beam and the measured intensity will be higher than that of the unaffected probe beam. Subtracting the crossed probe beam signal from the unaffected probe beam signal will result in sharp peaks at the resonant frequencies.

There is, however, an additional effect that results in unwanted peaks in an absorption spectrum. We have already seen that probe beam is affected when the laser is tuned to a resonant frequency of the atoms. The probe beam can also be affected when the laser is tuned to the frequency midway between two resonant frequencies; the resulting peak is known as a crossover peak, and it is often stronger than the peaks at the resonant frequencies [3]. Crossover peaks occur because the Doppler shift allows certain moving atoms to be in resonance with both the pump beam and the probe beam. For instance, if the laser is tuned halfway between the transitions from $F = 3$ to $F' = 3$ and from $F = 3$ to $F' = 4$, atoms moving towards the probe beam could see the pump beam as the lower frequency resonance, causing them to depopulate the $F = 3$ ground state, and they could see the probe beam as the higher frequency resonance, resulting in a signal where none ought exist.

The line-width of each peak in the Doppler-free signal is theoretically limited by the Heisenberg uncertainty principle [1]. For this experiment, however, the limiting factor was the bandwidth of the diode laser.

Procedure

Using mirrors and beamsplitters, the beam from a diode laser was split into three parts. Two parallel and closely-spaced beams, referred to as probe beams, were sent through a cell containing rubidium atoms. After decreasing their intensity so as not to saturate the detectors, they were measured by two photodiode detectors connected to an oscilloscope. The third beam was directed into the Rb cell from the opposite direction so that it overlapped with one of the probe beams without touching the other.

The laser frequency was modulated so that its wavelength varied around 780 nm, and the output of the detectors was measured over this range. The two probe beam signals on the digital oscilloscope were subtracted to form the Doppler-free signal.

In order to accurately determine what transitions were measured, the timescale of the oscilloscope had to be related to a frequency. Unfortunately, this was not done correctly, as will be explained in the Discussion section.

Results and Discussion

The absorption spectra from the two probe beams is seen in Figure (2). In Figure (3), the two probe beams were subtracted to give the Doppler-free signal, which showed three peaks at -0.130 ms, -0.0725 ms and 0.0725 ms on the oscilloscope.

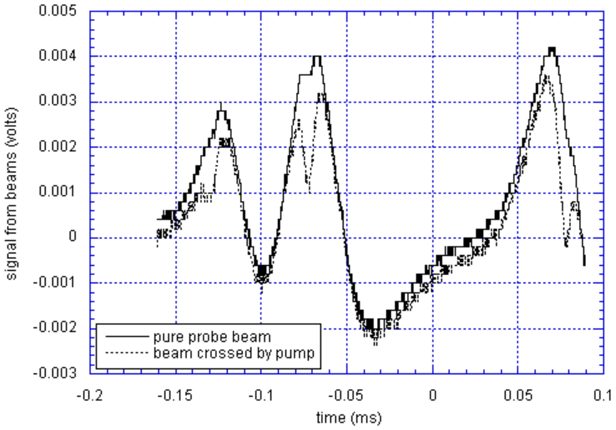

Figure 2: Measured signal from the two probe beams, one of which is crossed by the pump beam in the opposite direction.

To relate the timescale of the oscilloscope to a frequency, first a spectrum analyzer was attached to the non-digital oscilloscope to relate frequency to boxes on the screen, and then the Doppler-free signal was compared on the two oscilloscopes to relate boxes to time on the digital oscilloscope. Unfortunately, this is not a correct conversion between frequency and time. A spectrum analyzer contains a cavity with two highly reflective internal mirrors,

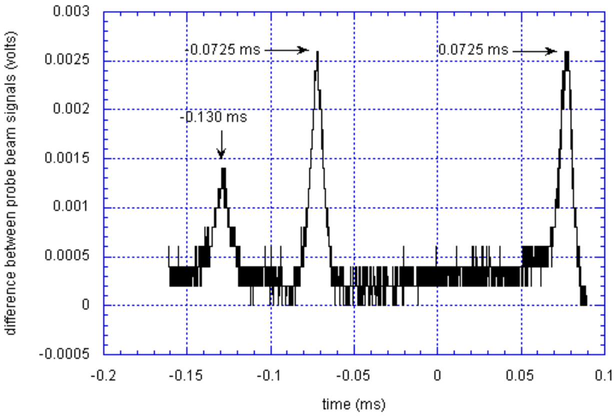

Figure 3: The difference between the two probe beam signals seen in Figure (2) gives the Doppler-free saturated absorption spectral lines. Note that the three peaks correspond to -0.130 ms, -0.0725 ms, and 0.0725 ms.

so that an electric field sent in one side is only transmitted if it corresponds to a mode of the cavity. The output is thus zero except at certain frequencies at which a spike occurs. Because the separation between these spikes is 7.5 GHz for our instrument, by determining the change in current necessary to move one spike by this distance we could determine that 7.5 GHz corresponded to about 2 mA. The instrument controlling the current gave us the current to voltage conversion, 20 mA to 1 V. To relate a change in voltage to a change in time, we should have measured the amplitude and period of the triangle wave with which we were modulating the laser while we were performing the experiment. The amplitude was measured several weeks later to be 53 mV, and it would have been unlikely to have been changed. From the timescale of our scan in Figure (2), we can estimate that this 53 mV corresponds to 0.25 ms, resulting in a calibration of 16 GHz/ms.

Because all of the calibration measurements were performed several weeks after the rest of the experiment, we cannot be sure of their accuracy. The spectrum in Figure (2), however, can be identified as the ground-state splitting of Rb, with the first peak corresponding to the $F = 2$ energy level of ^{87}Rb, the second corresponding to the $F = 3$ energy level of ^{85}Rb, and the third corresponding to the $F = 2$ energy level of ^{85}Rb. The complete spectrum of the ground-state would have a fourth peak further to the right corresponding to the $F = 1$ energy level of ^{87}Rb; this was apparently out of the range of our scan. Given this identification, the separation between the first two peaks should be 1.2 GHz, and the separation between the second two should be 3.1 GHz [4]. Indeed, these are the correct proportions for our data; both of these separations give a calibration of 21 GHz/ms. This means that our rough estimate of the calibration was off by 24%, but the fact that it is of the right order of magnitude gives us some confidance in our results.

As a further check, estimations of the FWHM for the three peaks on the Doppler-broadened spectrum in Figure (2) are 0.025 ms, 0.03 ms, and 0.035 ms, respectively. These correspond to frequencies of 525 MHz, 630 MHz, and 735 MHz, which are all above the Doppler-limited FWHM of 500 MHz. They may be slightly wider than this limit because the measurements were taken using AC coupling, which can smooth out the data.

While the peaks shown in Figure (3) are Doppler-free, they do not show the hyperfine splitting of the $5^2P_{3/2}$ state. While this may be partly due to the use of AC coupling, the main cause is probably the bandwidth of the laser. A rough estimation of 33 MHz was made for the bandwidth while examining the spikes output by the spectrum analyzer, which is of the same order of magnitude as the excited state hyperfine splittings.

In future investigations, the calibration should be done at the same time the data is taken to allow for more accurate measurements. Setting the oscilloscope to DC coupling might allow for better resolution, as would using a laser with a narrower bandwidth. These improvements might allow the hyperfine splitting of the excited state to be measured.

Conclusion

To see how Doppler-free spectroscopy can be a useful technique for measuring the hyperfine splitting of an atom, a simple experiment was performed to measure these transitions in a rubidium sample. Two probe beams were sent through the Rb cell and into photodiode detectors connected to an oscilloscope. When their frequencies were modulated, a Doppler-broadened signal was seen on the oscilloscope because the random thermal motion of the atoms caused a distribution of atoms to see the laser at a resonance frequency. Then a pump beam was crossed with one of the probe beams, and through hyperfine pumping and saturation it decreased the population of atoms in the ground state. Only when the laser was tuned to a resonance frequency would the pump beam and the probe beam be interacting with the same atoms, resulting in a dip in the probe beam signal. Subtraction the two probe beams resulted in a Doppler-free signal, which was identified as the hyperfine splitting of the ground state.

Three of the ground state levels were shown: the $F = 2$ energy level of ^{87}Rb, the $F = 3$ energy level of ^{85}Rb, and the $F = 2$ energy level of ^{85}Rb. These three peaks were separated by 0.0575 ms and 0.145 ms in our data. The theoretical separations of 1.2 GHz and 3.1 GHz both give the same calibration: 21 GHz/ms. This means there is a 24% error in our rough estimate of the calibration at 16 GHz/ms. Using the 21 GHz/ms calibration, the FWHM of the peaks were 525 MHz, 630 MHz, and 735 MHz, which are all above the Doppler limit of 500 MHz.

The errors in this experiment came from mistakes during the calibration, the improper use of AC coupling, and the wide bandwidth of the diode laser; the first two could easily be corrected in a future investigation.

References

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