saturated absorption spectroscopy: A report

G. A. Kavuri

Abstract

In this project, we explore a well known spectroscopy technique that allows us to overcome the limitations posed by the effect of the Maxwell-Boltzmann velocities (Doppler Broadening) of the sample atoms under investigation . saturated absorption spectroscopy is a well-known experimental technique that allows us to counter Doppler Broadening at room temperatures itself.

1 Introduction

This project was carried out by Niranjan S and the author during the period beginning May 10, 2015 and ending July 10, 2015 at Prof. Vasant Natarajan's lab at IISc, Bangalore, in their capacity as summer interns.

1.1 Spectroscopy

Fundamentally, spectroscopy is the study of the interaction of light with matter. The kind of spectroscopy we explore here involves passing light through a sample of interest (usually gaseous atoms), and observing the variation of either the fluorescence in the sample or the extent of absorption of the incident light, with change in incident light frequency. The variation is usually quantified with the help of a photo-diode that converts the incoming light intensity into a proportional electrical signal. The signal can then be viewed on a device such as an oscilloscope to yield a graph of intensity vs frequency. A study of this graph can yield details about the electronic energy levels of atoms under study.

1.2 Doppler Broadening

Gaseous atoms at room temperature have non-zero total energies. These energies manifest in the different kinds of motions that are executed by these atoms. Along with various oscillatory modes, the atoms also execute translational motion at various velocities. These velocities are known to follow the Boltzmann distribution around a mean velocity that varies with gas temperature and the mass of the gaseous atom.

When a wave of frequency ω_0 is viewed from a certain reference frame, the frequency may appear to be either higher (Blue shifted) or lower (Red

shifted) than at the source reference frame, depending upon the relative motions of the source and viewer. Considering that atoms are in continual motion at room temperature, one can easily see that any light passing through a cell of gaseous atoms will appear to have a frequency different from that measured by a stationary observer. Further, various atoms will experience dissimilar "Doppler shifts" owing to their largely unordered motion. This brings us to the problem with traditional spectroscopy conducted at room temperature. Not all atoms respond to a single incident frequency. Due to the thermal motion, atoms either respond to red or blue shifted light. The degree of shift will further depend on the magnitude of the velocity of the atom, and what results is a broad response to the laser frequency, thus "blurring out" many details.

1.3 Saturated Absorption Spectroscopy[1]

The origins of the technique known as Saturated Absorption spectroscopy are intimately connected with the invention of the laser.

In 1958, Arthur Leonard Schawlow and Charles Townes of Bell labs together published a paper [2] detailing the principles and techniques that might be of use in the construction of a narrow line-width, coherent light source which emitted light in the infrared-optical region of the electromagnetic spectrum. Wallis E. Lamb then predicted what is now known as a "Lamb dip", a drop in the intensity of emitted laser light at the resonance frequency of the gain medium, based on a theoretical treatment.[3] Working on top of this, it was possible to resolve the fine details of atomic spectra, i.e., a "Doppler-free" spectrum of the gas inside a laser could be produced.

Further advancement came in 1970, when Theodor W. Hänsch and Christian Bordè found a way to obtain a Doppler free spectrum of a sample of gas placed outside the laser cavity as well. This technique, almost unaltered, is what we have used in our experiment.

2 Theory

2.1 Spectroscopy [4]

Spectroscopic techniques of some form or the other have been known for more than 150 years now.

Consider a two energy level atomic system, with the electron in the lower or ground state. When this system is impinged by an oscillating electric field of the correct frequency, the electron may get excited to the higher level. Further, the agitated electron may also drop back down to the ground state though the processes of either simulated or spontaneous emission. Thus, when we pass a beam of laser light through a cell containing atoms of a material with an "ex-

citable" electron and a two state system, we observe a so called "fluorescence" in the cell when we image it with the help of a CCD (Charge-Coupled Device) Camera. The fluorescence is due to the spontaneous emission of the electrons in systems excited by the laser beam, and is a good indicator that the laser is operating at or around the resonance frequency.

To quantify the interaction between the incident light and the atoms in question, we can either choose the light that is spontaneously emitted or the light that is transmitted through the gas to convert to a proportional electric signal. These choices will yield signals that differ only in the polarity and relative magnitude of the electric signal produced. The shape of the spectrum remains the same. In this experiment, all signals are absorption signals.

The conversion of light intensity to a proportional electric signal is achieved through the use of a semiconductor based "photo diode", which, roughly speaking, takes advantage of some excitation process inside the semiconductor to produce a proportional voltage difference, which is then run through some amplification before it is fed to the output. The level of this output signal can then be easily visualized with the means of an external oscilloscope. If we vary a parameter (say, the temperature of the gas) with time, and "trigger" the oscilloscope with the right signal, (the trigger dictates how quickly the oscilloscope refreshes the graph on the screen) we will obtain the variation of absorption with change in the parameter.

In this experiment, we vary the frequency of the light source (the laser) and thus obtain graphs that indicate the change in absorption with frequency.

2.2 Doppler Broadening

Consider a sample of gas. At room temperature, the molecules (atoms, if it is an elemental gas, like in this case) will be executing translational motion in various directions, at different velocities. The velocities, as an ideal approximation can be assumed to be following the Maxwell-Boltzmann distribution.

$$f(v) = \sqrt{\left(\frac{m}{2\pi kT}\right)^3} 4\pi v^2 e^{-\frac{mv^2}{2kT}}$$
 (1)

Here, f(v) is the probability density function for finding a particle at a speed v. The mass of the individual atoms is given by m, the temperature of the gas by T, and k is the Boltzmann constant.

Now consider the set of atoms that lie along the line of the laser beam. These are the atoms upon which the beam of light is incident. Assume (although it is not strictly true) that the laser emits light of a *single* frequency λ_0 .

When this light strikes the moving atoms, they experience a frequency that is different from the laser's by a velocity dependent amount, as predicted by the Doppler shift equation (non-relativistic approximation).

$$f_a = f_o \left(1 \pm \frac{v}{c} \right). \tag{2}$$

Here, f_a is the frequency from the reference frame of the atom moving at a speed v relative to the source. f_o is the frequency of the laser light at the source.

The equation for the number of atoms at a velocity v can be derived from equation 1,

$$f(v)dv = \sqrt{\left(\frac{m}{2\pi kT}\right)}e^{-\frac{mv^2}{2kT}}dv.$$
 (3)

Then, the expression for the number of atoms "seeing" light at resonance frequency is given by

$$N(f_a)dv = \sqrt{\left(\frac{m}{2\pi kT}\right)}e^{-\frac{mc^2\left(\frac{f_a}{f_o}-1\right)^2}{2kT}}dv.$$
 (4)

Finally, because

$$\frac{dv}{df_a} = \frac{c}{f_o} \tag{5}$$

(From eq. 2), we get

$$N(f_a)df_a = \sqrt{\left(\frac{mc^2}{2\pi kT f_o^2}\right)}e^{-\frac{mc^2\left(\frac{f_a}{f_o}-1\right)^2}{2kT}}df_a.$$
 (6)

The graph of $N(f_a)$ vs $(f_a - f_o)$ (the frequency offset) will be a Gaussian profile. Thus, the Doppler broadened spectrum will be a Gaussian, as shown

The full width half maximum, a measure of the width of the spectrum will then be given by

$$\delta_F W H M = \sqrt{\frac{8kT \ln(2)}{mc^2}} f_o \tag{7}$$

None of the parameters involved in the above equation are under the control of the experimentalist. We are powerless, therefore, to reduce, or even change, this broadening by a simple variation of a parameter. The broadening, and its magnitude, come married with the technique that we are using to probe the spectrum. For a finer resolution, we must adopt a better technique.

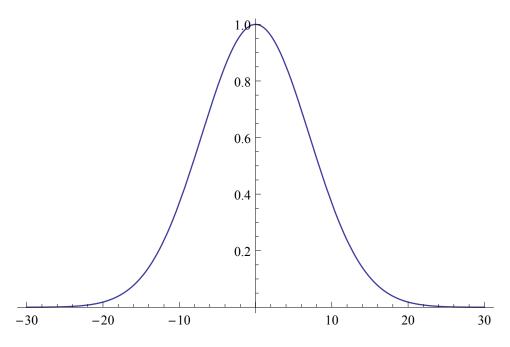


Figure 1: A Gaussian profile

2.3 Saturated Absorption Spectroscopy

Figure 2 shows the schematic of the saturated absorption spectroscopy setup.

The oscilloscope signal is taken from a weak, so called "probe" beam that passes through the vapor. This weak beam is crossed by a relatively strong, so called "pump" beam. In the figure, there are two probe beams, but we can ignore the one that is not crossed by the pump in the cell for the moment.

Consider the pump beam. Say that the "detuning" from the resonance frequency is given by $\delta = f_r - f_o$. Then, atoms of a particular velocity, say v_{δ} in the appropriate direction will respond to the light. These atoms reach an equilibrium, moving between the excited and ground states. Approximately half the population is in the excited state. This is called "saturation" of the sample.

The probe beam is exactly the same frequency as the pump beam, but it is counter-propagating i.e., the wave is propagating in the opposite direction. Because of this, a different set of atoms, moving in the opposite direction respond to the signal, even though the detuning $\delta_p robe$ is the same.

When the detuning δ is zero, both the probe and the pump excite the same set of atoms viz., those that have a zero velocity in the direction of either beam.

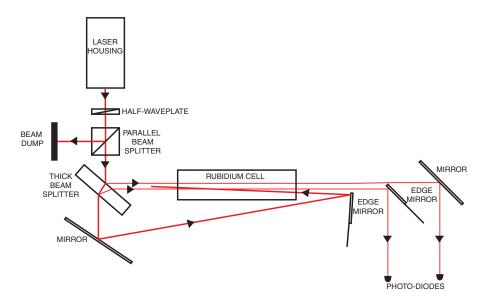


Figure 2: A saturated absorption spectroscopy schematic

Thus, the probe beam will not be absorbed by these atoms, which have been saturated by the pump, and we will see a spike on the Doppler absorption signal corresponding to the location of the transition. In other words, better resolutions are obtainable with the use of this technique.

2.4 Rubidium electronic levels[5]

A complete, thorough analysis and characterization of the hyperfine splitting of the $^{87}{\rm Rb}$ and the $^{85}{\rm Rb}$ is beyond the scope of the author.

The hyperfine splitting is due to the effects of magnetic and electric multiploles of the nucleus. i.e., there would be no splitting if the nucleus were a perfect sphere of positive charge. Due to these effects, the 5s and 5p levels are further split as shown in fig. 3.

The transitions shown in green are the ones that are allowed.

3 Experimental Setup

The experimental setup schematic is as indicated in fig.2.

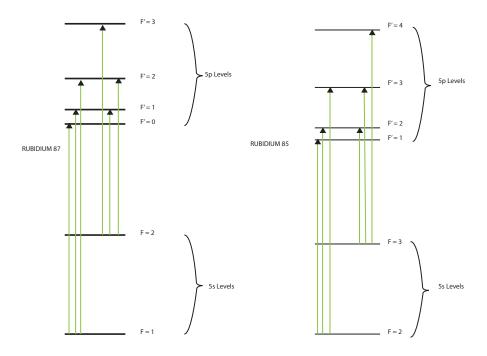


Figure 3: The Rubidium hyperfine splitting

3.1 External Cavity Diode Laser

The source of narrow line width tunable frequency light for use in this experiment was an external cavity diode laser (ECDL). An ECDL setup consists of a semiconductor diode laser, which emits relatively high line width light at around 780nm, a collimating lens, and a fine diffraction grating to achieve frequency selective feedback. The setup, known as the Littrow configuration, is as shown in fig.4.

The diode laser produces divergent, high line width light that is collimated by the collimating lens as shown. The collimated light is then incident on a diffraction grating which has very fine markings on it. These serve to create a diffraction pattern in which different frequencies of light are reflected off at various angles. Thus, by varying the angle of the diffraction grating relative to the incident light, we can control the output light frequency, within a certain range. If we consider a ray of light incident on the grating at an angle θ_i relative to the normal, then the angle (θ_r) at which there is a maximum for reflected light off the grating is given by

$$\theta_r = \arcsin\left(\frac{m\lambda_r}{d} - \sin\left(\theta_i\right)\right) \tag{8}$$

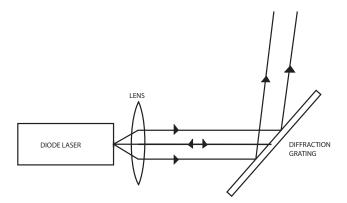


Figure 4: The ECDL Littrow configuration

Here, θ_r is the angle, relative to the normal, at which there is a maximum for reflected light of wavelength λ_r , when light is incident on a grating with grating width d. m is an non-negative integer that indicates the order of the maximum, with the zeroth order fringe being the one with the strongest intensity.

As we can see in the figure, there is also "feedback" from the grating, i.e., some light travels back down the path of the incident ray and impinges on the laser diode. This is an essential part of the lasing process. Because of the feedback from the grating, a longitudinal mode, or a standing wave is set up between the grating and the source. Because of this, only a single frequency is selected, and other light frequencies die out.

One must also note that there is usually a small piezo electric crystal attached behind the diffraction grating in such a manner such that its angle with respect to the incident beam can be varied by varying the voltage applied to the crystal. Changing the angle in this manner affects the feedback given to the diode, and alters the frequency of the finally emitted light. Thus, the piezo voltage can be profitably employed to alter the frequency of the light from the laser.

3.2 Other Devices Used

3.2.1 PBS and $\frac{\lambda}{2}$ assembly

After the beam exits the laser, it first meets the parallel beam splitter (PBS), and the half-wave plate $(\frac{\lambda}{2})$. The half-wave plate is basically a thin slice of crystal with precise thickness. The crystal behaves differently for different polarizations of a light beam. Utilizing this property, the thickness of the half wave plate is carefully engineered to ensure a phase shift of π for one perpendicular component of light relative to the other. This serves to flip the sign on the

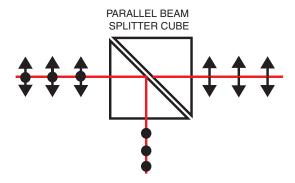


Figure 5: The parallel beam splitter cube

polarization of one component, and thus alters the overall polarization of the linearly polarized light from the laser. This light is then fed to the PBS.

The PBS is constructed of two prism-shaped blocks of material which are placed with together to form a cube. The PBS splits incoming light into two component perpendicular polarizations, as shown in the fig.5.

Thus, as we can alter the orientation of the linearly polarized light with the help of the half-wave plate, and isolate a single perpendicular component with the help of the PBS, the combination of these two can be used to alter the power of the laser beam emerging from the pair.

3.2.2 The thick beam splitter

The thick beam splitter was used to generate the two weak probe beams, from the main laser beam. The beam splitter partially reflected the incident laser light off its front and back faces, and these were used for the purposes of the weak probe beams.

The edge mirrors were simply normal ones mounted so that they would be able to pick up and reflect a beam from very close to another beam without disturbing the original one.

3.2.3 Rubidium cell

The rubidium cell contains rubidium vapor at low pressure. It is the "source" of the atoms that are irradiated as part of the experiment. The cell contains both the $^{87}{\rm Rb}$ and the $^{85}{\rm Rb}$ isotopes, in their naturally occurring ratio of $\approx 72:28$ ($^{85}{\rm Rb}:^{87}{\rm Rb}$). It is basically a borosilicate glass container with the gas inside it. The laser beam is passed through two specially treated sides.

4 RESULTS 10

4 Results

The figures below illustrate two of the waveforms observed on the oscilloscope.

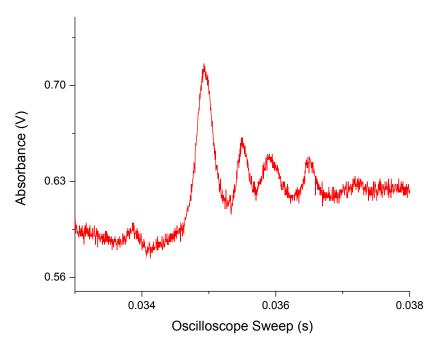


Figure 6: A transition Spectrum

Fig.6 is the absorption vs frequency graph that corresponds to the transitions from the F=1 of the $^{87}{\rm Rb}$ isotope.

Fig.7 is a similar graph, but for the transitions from F=2 for the $^{85}{\rm Rb}$ isotope.

One feature of note is clear appearance of not three, but six peaks in the absorption plot in Fig. 6. This phenomenon has already been studied, and the peaks are called "crossover" peaks. These are due to a spike in absorption when the incident light frequency is exactly the average of the frequencies corresponding to two hyperfine transitions.

5 Conclusion

There is hardly anything to say, in conclusion to this experiment, that has not already been well documented. This is not only because saturated absorption spectroscopy, as a technique, has been studied for almost fifty years now, but

REFERENCES 11

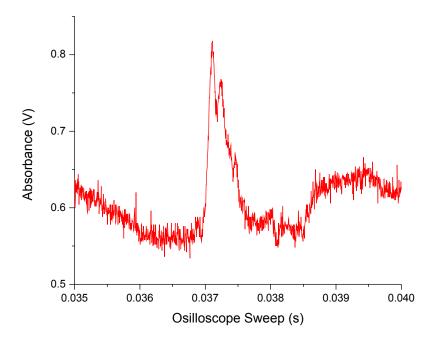


Figure 7: Another Spectrum

also because the experiment done during the period of the project was not designed and begun with the intention of doing a careful enough study of the technique, so as to yield new insights. It was in fact, the useful tool that it is, supposed to be used as part of a bigger experiment viz., the observation of the Coherent Population Trapping phenomenon.

References

- [1] A L Schawlow. Spectroscopy in a new light. Nobel Lecture, 1982.
- [2] A. L. Schawlow and C. H. Townes. Infrared and Optical Masers, 1958.
- [3] Willis E. Lamb. Theory of an optical maser. *Physical Review*, 134(6A), 1964.
- [4] Preston Daryl W. Doppler-free saturated absorption: Laser spectroscopy. *American Journal of Physics*, 64:1432–1436, 1996.
- [5] Daniel Adam Steck. Rubidium 87 D Line Data. Unpublished.