

DOPPLER-FREE SATURATED ABSORPTION SPECTROSCOPY: LASER SPECTROSCOPY

1 Introduction

In this experiment you will use an external cavity diode laser to carry out laser spectroscopy of rubidium atoms. You will study the Doppler broadened optical absorption lines (linear spectroscopy), and will then use the technique of saturated absorption spectroscopy to study the lines with resolution beyond the Doppler limit (nonlinear spectroscopy). This will enable you to measure the hyperfine splittings of one of the excited states of rubidium. You will use a balanced photodetector to perform background suppression and noise cancellation in the photo-detection process. You will use a Michelson interferometer to calibrate the frequency scale for this measurement. This writeup is based on an earlier version written by Carl Wieman.

Apparatus

Vortex™ 780-nm diode laser system
Rubidium vapor cell
Balanced photo detector
Triangle waveform generator
optical breadboard
3/8"-thick transparent plastic or glass (beam splitter)
Mirrors and mirror mounts
Oscilloscope (Optional: storage scope with a plotter)
Digital camera
Kodak IR detection card
Hand held IR viewer or a CCD surveillance camera

Purpose

- 1) To appreciate the distinction between linear and nonlinear spectroscopy.
- 2) To understand term states, fine structure, and hyperfine structure of rubidium.
- 3) To record and analyze the Doppler-broadened 780-nm rubidium spectral line (linear optics).
- 4) To record and analyze the Doppler-free saturated absorption lines of rubidium (nonlinear optics), and thereby determine the hyperfine splitting of the $5P(3/2)$ state.

Key Concepts

Linear optics	Electric dipole selection rules
Nonlinear optics	Doppler broadening
Absorption spectroscopy	Saturated absorption spectroscopy
Fine structure	Hyperfine structure

2 Background

Figure 1 compares an older method with a modern method of doing optical spectroscopy. Figure 1a shows the energy levels of atomic deuterium and the allowed transitions for $n = 2$ and 3, 1b shows the Balmer α emission line of deuterium recorded at 50 K with a spectrograph, where the vertical lines in 1b are the theoretical frequencies, and 1c shows an early high resolution laser measurement, where one spectral line is arbitrarily assigned 0 cm^{-1} . The "crossover" resonance line shown in 1c will be discussed later. Even at 50 K the emission lines are Doppler-broadened by the random thermal motion of the emitting atoms, while the laser method using a technique known as Doppler-free, saturated absorption spectroscopy eliminates Doppler-broadening.

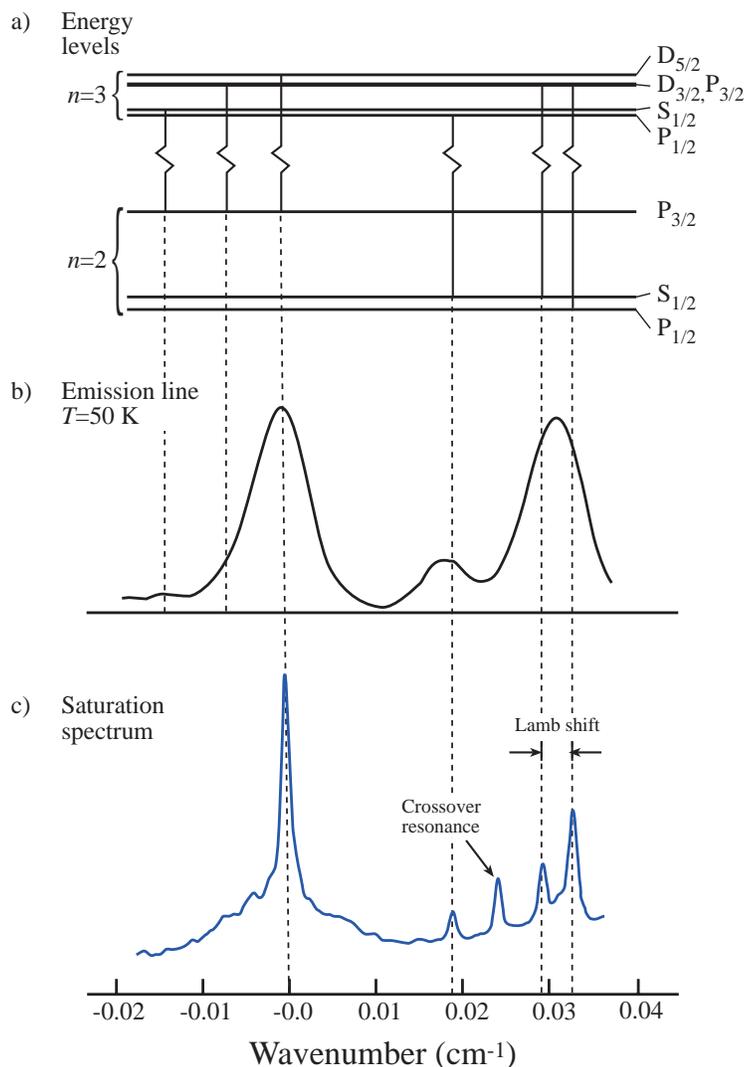


Figure 1. The Balmer α line of atomic deuterium. a) Energy levels and allowed transitions, b) the emission spectrum, c) an early saturated absorption spectrum

In this case the field reaching the detector is a function of both fields, hence, nonlinear spectroscopy. Prior to the development of the laser, the interaction between optical frequency fields and matter were weak enough that linear theories were adequate.

What feature of the laser gives rise to high resolution spectroscopy? Well, it is the narrow spectral linewidth, which is about 20 MHz for the diode laser, ($\sim 1 \text{ MHz}$ and below for an extended cavity laser diode) and the tunability of lasers that have revolutionized optical spectroscopy. Note that when the 780-nm diode laser is operating at its center frequency then most of its power output is in the frequency range of $3.85 \times 10^{14} \text{ Hz} \pm 1 \times 10^6 \text{ Hz}$.

The topics to be discussed in the Introduction are linear and nonlinear optics, atomic structure of rubidium, absorption spectroscopy and Doppler broadening, and Doppler-free saturated absorption spectroscopy.

A simplified diagram of linear spectroscopy is shown in figure 2a, where a single propagating wave is incident on the sample, some photons are absorbed, as shown in the two-energy diagram, and some fraction of the wave reaches the detector. Nonlinear spectroscopy is illustrated in figure 2b, where there are two counter-propagating waves that interact with the same atoms in the region where they intersect. The beam propagating to the right, the *pump* beam, causes the transition indicated with a dashed line in the energy level diagram, and the beam propagating to the left, the "probe" beam, causes the transition indicated with a solid line. In this

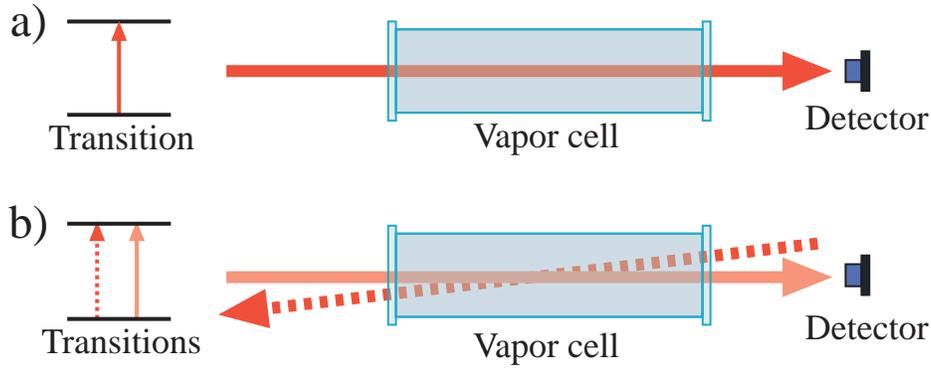


Figure 2. In linear spectroscopy (a) the radiation reaching the detector is proportional to the radiation incident on the sample. In nonlinear spectroscopy (b) the radiation reaching the detector is dependent on both beams.

3 Atomic Structure of Rubidium

The ground electron configuration of rubidium (Rb) is: $1s^2; 2s^2, 2p^6; 3s^2, 3p^6, 3d^{10}; 4s^2, 4p^6; 5s^1$, and with its single $5s^1$ electron outside of closed shells it has an energy-level structure that resembles hydrogen. For Rb in its first excited state the single electron becomes a $5p^1$ electron. Also natural rubidium has two isotopes, the 28% abundant ^{87}Rb , where the nuclear spin quantum number $I = 3/2$, and the 72% ^{85}Rb , where $I = 5/2$.

3.1 TERM STATES

A term state is a state specified by the angular momentum quantum numbers s, ℓ , and j (or S, L , and J), and the notation for such a state is $^{2s+1}\ell_j$ (or $^{2s+1}L_j$). The spectroscopic notation for ℓ values is $\ell = 0(S), 1(P), 2(D), 3(F), 4(G), 5(H)$, and so on.

The total angular momentum \mathbf{J} is defined by,

$$\mathbf{J} = \mathbf{L} + \mathbf{S}, \tag{1}$$

where their magnitudes are

$$J = \hbar\sqrt{j(j+1)}; L = \hbar\sqrt{\ell(\ell+1)}; S = \hbar\sqrt{s(s+1)}, \tag{2}$$

and the possible values of the total angular momentum quantum number j are $|\ell - s|, |\ell - s| + 1, \dots, \ell + s - 1, \ell + s$; where for a single electron $s = 1/2$.

The $5s^1$ electron gives rise to a $5^2S_{1/2}$ ground term state. The first excited term state corresponds to the single electron becoming a $5p^1$ electron, and there are two term states, the $5^2P_{1/2}$ and the $5^2P_{3/2}$.

3.2 HAMILTONIAN

Assuming an infinitely massive nucleus, the nonrelativistic Hamiltonian for an atom having a single electron is given by:

$$H = \frac{p^2}{2m} - \frac{Z_{\text{eff}}e^2}{4\pi\epsilon_0 r} + \zeta(r)\mathbf{L} \cdot \mathbf{S} + \alpha\mathbf{J} \cdot \mathbf{I} + \frac{\beta}{2I(2I-1)J(2J-1)} \left[3(\mathbf{I} \cdot \mathbf{J})^2 + \frac{3}{2}(\mathbf{I} \cdot \mathbf{J}) - I(I+1)J(J+1) \right] \tag{3}$$

We label the 5 terms in this equation, in order, as $K, V, H_{\text{SO}}, H_{1,\text{hyp}}$ and $H_{2,\text{hyp}}$ respectively. K is the kinetic energy of the single electron; where $\mathbf{p} = -i\hbar\nabla$, classically \mathbf{p} is the mechanical momentum of the electron of mass m . V is the Coulomb interaction of the single electron with the nucleus and the core

electrons (this assumes the nucleus and core electrons form a spherical symmetric potential with charge $Z_{\text{eff}}e$ where Z_{eff} is an effective atomic number).

H_{SO} is the spin orbit interaction, where \mathbf{L} and \mathbf{S} are the orbital and spin angular momenta of the single electron. $H_{1,\text{hyp}}$ is the magnetic hyperfine interaction, where \mathbf{J} and \mathbf{I} are the total electron and nuclear angular momenta, respectively. This interaction is $-\mu_n \cdot \mathbf{B}_e$ where μ_n , the nuclear magnetic dipole moment, is proportional to \mathbf{I} , and \mathbf{B}_e , the magnetic field produced at the nucleus by the single electron, is proportional to \mathbf{J} . Hence the interaction is expressed as $\alpha \mathbf{I} \cdot \mathbf{J}$. α is called the magnetic hyperfine structure constant, and it has units of energy, that is, the angular momenta \mathbf{I} and \mathbf{J} are dimensionless.

$H_{2,\text{hyp}}$ is the electric quadrupole hyperfine interaction, where β is the electric quadrupole interaction constant, and non-bold I and J are angular momenta quantum numbers. The major electric pole of the rubidium nucleus is the spherically symmetric electric monopole, which gives rise to the Coulomb interaction; however, it also has an electric quadrupole moment (but not an electric dipole moment). The electrostatic interaction of the single electron with the nuclear electric quadrupole moment is $-eV_q$, that is, it is the product of the electron's charge and the electrostatic quadrupole potential. Although it is not at all obvious, this interaction can be expressed in terms of \mathbf{I} and \mathbf{J} . In both hyperfine interactions \mathbf{I} and \mathbf{J} are dimensionless, that is, the constants α and β have units of joules.

We will not use the above Hamiltonian in the time independent Schrödinger equation and solve for the eigenvalues or quantum states of rubidium, but rather we present a qualitative discussion of how each interaction effects such states.

3.2.1 $K + V$

The $K + V$ interactions separate the 5s ground configuration and the 5p excited configuration. This is shown in Figure 3a. Qualitatively, if the potential energy is not a strictly Coulomb potential energy then for a given value of n , electrons with higher ℓ have a higher orbital angular momentum (a more positive kinetic energy) and on the average are farther from the nucleus (a less negative Coulomb potential energy), hence higher ℓ value means a higher (more positive) energy. This scenario does not occur in hydrogen because the potential energy is coulombic.

3.2.2 *Fine Structure, H_{SO}*

The spin-orbit interaction and its physical basis are discussed in standard quantum mechanics text books. Fine structure, the splitting of spectral lines into several distinct components, is found in all atoms. The interactions that give rise to fine structure do depend on the particular atom. Ignoring relativistic terms in H , it is H_{SO} that produces the fine structure splitting of rubidium.

Using equations (1) and (2) and forming the dot product of $\mathbf{J} \cdot \mathbf{J}$, we solve for $\mathbf{L} \cdot \mathbf{S}$ and obtain

$$\begin{aligned} \mathbf{L} \cdot \mathbf{S} &= (J^2 - L^2 - S^2) / 2 \\ &= \frac{\hbar^2}{2} [j(j+1) - \ell(\ell+1) - s(s+1)] \end{aligned} \tag{4}$$

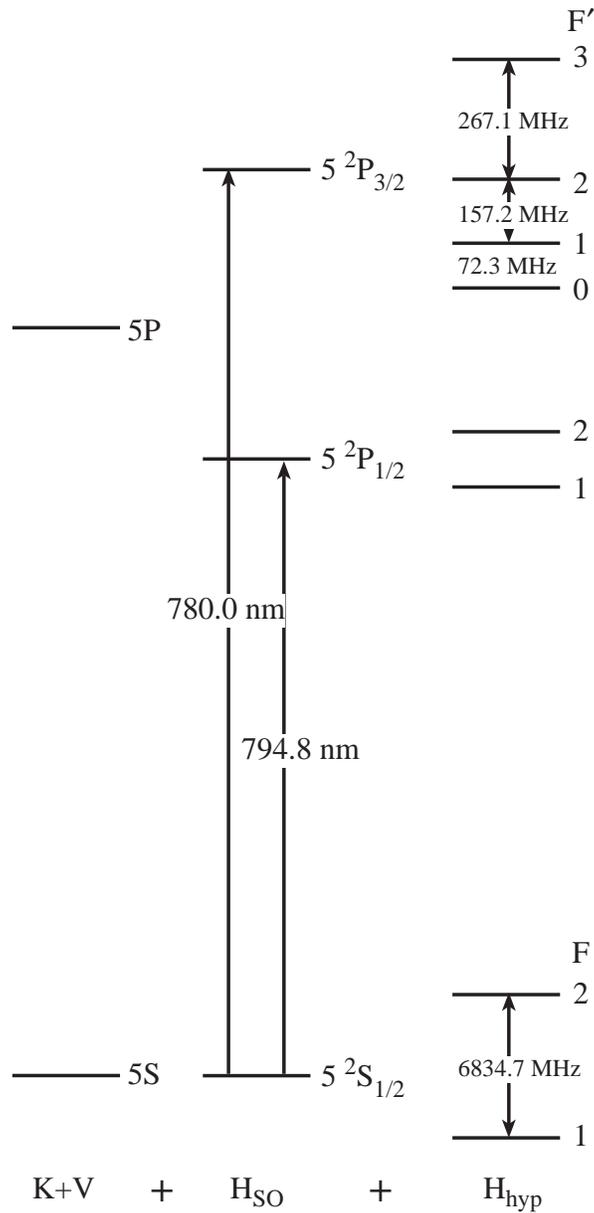


Figure 3. Each interaction in Eq. (3) and its effect on the energy levels of the 5s and 5p electron is shown. The energy level spacings are not to scale. The hyperfine levels are for ^{87}Rb .

where the magnitudes of the vectors were used in the last equality. Using Eq (4) H_{SO} can be written

$$H_{SO} = \zeta(r) \frac{\hbar^2}{2} [j(j+1) - \ell(\ell+1) - s(s+1)] \quad (5)$$

Figure 3b shows the effect of H_{SO} on the quantum states. The separation of the $5^2S_{1/2}$ and the $5^2P_{3/2}$ states, in units of wavelength, is 780.023 nm , and the separation of the $5^2S_{1/2}$ and the $5^2P_{1/2}$ states is 794.764 nm . It is the transition between the $5^2S_{1/2}$ and the $5^2P_{3/2}$ states that will be studied using the 780-nm laser.

3.2.3 Hyperfine Structure, H_{hyp}

For either hyperfine interaction, the interaction couples the electron angular momentum \mathbf{J} and the nuclear angular momentum \mathbf{I} to form the total angular momentum, which we label as \mathbf{F} , where

$$\mathbf{F} = \mathbf{J} + \mathbf{I}, \quad (6)$$

and the possible quantum numbers F are $|J - I|, |J - 1| + 1, \dots, J + I - 1, J + I$. (In this case the non-bold capital letters are being used for quantum numbers, which, for the hyperfine interaction, is more standard practice than using f, j and i as the quantum numbers.)

The hyperfine structure of both ^{85}Rb and ^{87}Rb will be observed in this experiment; however, it is the hyperfine structure of ^{87}Rb that will be studied. The split of energy levels by the hyperfine interaction is shown in the right column of Figure 3 for ^{87}Rb . These levels are known as *hyperfine levels*, where the total angular momentum quantum numbers are labeled as F' and F for the $5^2P_{3/2}$ and the $5^2S_{1/2}$ states, respectively. The selection rules for electric dipole transitions are given by

$$\begin{aligned} \Delta F &= 0 \text{ or } \pm 1 \text{ (but not } 0 \rightarrow 0) \\ \Delta J &= 0 \text{ or } \pm 1 \\ \Delta s &= 0 \end{aligned} \quad (7)$$

In addition to the normal resonance lines, there are "crossover" resonances peculiar to saturated absorption spectroscopy, which occur at frequencies $(\nu_1 + \nu_2)/2$ for each pair of true or normal transitions at frequency ν_1 and ν_2 . A crossover resonance is indicated in Figure 1c. The crossover transitions are often more intense than the normal transitions. In Figure 4 six crossover transitions, $b, d, e, h, j,$ and k , and six normal transitions, $a, c, f, g, i,$ and l , are shown, where for the normal transitions $\Delta F = 0, \pm 1$. The frequency of the emitted radiation increases from a to l .

What are the expected frequencies of the normal transitions $a, c, f, g, i,$ and l ? To answer this question we first determine the energies of the hyperfine levels. Using Eq. (6) and forming the dot product of $\mathbf{F} \cdot \mathbf{F}$, we solve for $\mathbf{J} \cdot \mathbf{I}$ and obtain

$$\begin{aligned} \mathbf{J} \cdot \mathbf{I} &= (\mathbf{F}^2 - \mathbf{J}^2 - \mathbf{I}^2) / 2 \\ &= [F(F+1) - J(J+1) - I(I+1)] / 2, \\ &= C / 2 \end{aligned} \quad (8)$$

where dimensionless magnitudes were used in the second equality and the last equality defines C . Replacing $\mathbf{J} \cdot \mathbf{I}$ in the hyperfine interactions of Eq. (3) with Eq. (8), the magnitude of the interactions or the energy $E_{J,F}$ is given by

$$\begin{aligned} E_{J,F} &= E_J + E_{hyp} \\ &= E_J + \alpha \frac{C}{2} + \beta \frac{3C^2 / 4 + 3C / 4 - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)} \end{aligned} \quad (9)$$

where E_J is the energy of the $n^{2S+1}L_J$ state, that is, the $5^2P_{3/2}$ or the $5^2S_{1/2}$ state shown in Figure 4. From the figure note that in Eq. (9) for the $5^2P_{3/2}$ state of ^{87}Rb , $I = 3/2$, $J = 3/2$, and $F = 0, 1, 2, 3$; and for the $5^2S_{1/2}$ state, $I = 3/2$, $J = 1/2$, and $F = 1, 2$.

The frequencies $\nu_{J,F}$ (energy/h) of the various hyperfine levels are obtained by dividing Eq. (9) by Planck's constant h :

$$\nu_{J,F} = \nu_J + A \frac{C}{2} + B \frac{[\frac{3}{4}C(C+1) - I(I+1)J(J+1)]}{2I(2I-1)J(2J-1)} \quad (10)$$

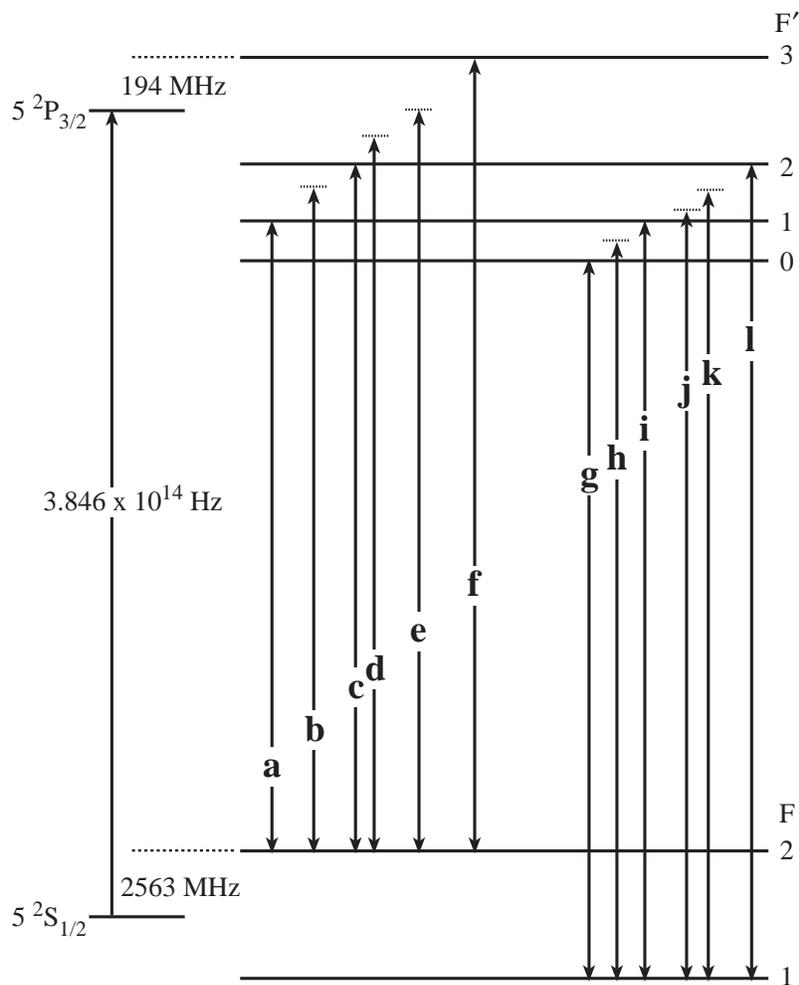


Figure 4. Saturated absorption transitions for ^{87}Rb . The spectral line separation will be derived in the exercises or can be figured out from Figure 3

where $A = \alpha / \hbar$ and $B = \beta / \hbar$ have units of hertz.

For the $5^2S_{1/2}$ state of ^{87}Rb , the term that multiplies B in Eq. (10) reduces to zero and the accepted value of A is 3417.34 MHz. For the $5^2P_{3/2}$ state of ^{87}Rb , the accepted values of A and B are 84.72 MHz and 12.50 MHz, respectively (See Ref. 12). For the $5^2S_{1/2}$ of ^{85}Rb the accepted value of A is 1011 91 MHz, and for the $5^2P_{3/2}$ the accepted values of A and B are 25.01 MHz and 25.9 MHz, respectively. One goal of this experiment is to experimentally determine A and B for the $5^2P_{3/2}$ state of ^{87}Rb .

4 Doppler Broadening and Absorption Spectroscopy

Random thermal motion of atoms or molecules creates a Doppler shift in the emitted or absorbed radiation. The spectral lines of such atoms or molecules are said to be Doppler broadened since the frequency of the radiation emitted or absorbed depends on the atomic velocities. Individual spectral lines may not be resolved due to Doppler broadening, and, hence, subtle details in the atomic or molecular structure are not revealed. Below we will answer the question on what determines the linewidth of a Doppler broadened line.

We first consider the Doppler effect qualitatively. If an atom is moving toward or away from a laser light source, then it receives radiation that is blue or red shifted, respectively. If an atom is at rest,

relative to the laser, absorbs radiation of frequency ν_0 , then when the atom is approaching the laser it will see blue-shifted radiation, hence for absorption to occur the frequency of the laser must be less than ν_0 in order for it to be blue-shifted to the resonance value of ν_0 . Similarly, if the atom is receding from the laser, the laser frequency must be greater than ν_0 for absorption to occur.

We now offer a more quantitative argument of the Doppler effect and atomic resonance, where, as before, ν_0 is the atomic resonance frequency when the atom is at rest in the frame of the laser. If the atom is moving along the z axis, say, relative to the laser with $V_z \ll c$, then the frequency of the absorbed radiation in the rest frame of the laser will be ν_L , where

$$\nu_L = \nu_0 \left(1 + \frac{V_z}{c} \right). \tag{11}$$

If V_z is negative (motion toward the laser), then $\nu_L < \nu_0$, that is, the atom moving toward the laser will absorb radiation that is blue-shifted from ν_L up to ν_0 . If V_z is positive (motion away from the laser) then $\nu_L > \nu_0$, that is, the atom absorbs radiation that is red-shifted from ν_L down to ν_0 . Therefore, an ensemble of atoms having a distribution of speeds will absorb light over a range of frequencies.

The probability that an atom has a velocity between V_z and $V_z + dV_z$ is given by the Maxwell distribution

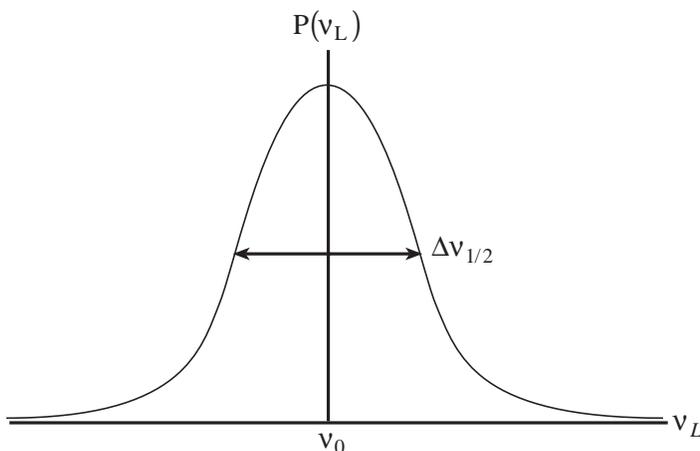
$$P(V_z)dV_z = \left(\frac{M}{2\pi kT} \right)^{1/2} \exp\left(-\frac{MV_z^2}{2kT} \right) dV_z \tag{12}$$

where M is the mass of the atom, k is the Boltzmann constant, and T is the absolute temperature. From Eq. (11):

$$V_z = (\nu_L - \nu_0) \frac{c}{\nu_0}; \quad dV_z = \frac{c}{\nu_0} d\nu_L \tag{13}$$

Substituting (13) into (12), the probability of absorbing a wave with a frequency between ν_L and $\nu_L + d\nu_L$ is given in terms of the so-called linewidth parameter $\delta \equiv 2(\nu_0 / c)(2kT / M)^{1/2}$ by

$$P(\nu_L) d\nu_L = \frac{2}{\delta\pi^{1/2}} \exp\left[-4(\nu_L - \nu_0)^2 / \delta^2 \right] d\nu_L \tag{14}$$



The half width, which is the full-width at half maximum amplitude (FWHM), of the Doppler broadened line is given by

$$\Delta\nu_{1/2} = \delta(\ln 2)^{1/2} = 2\frac{\nu_0}{c} \left(\frac{2kT}{M} \ln 2 \right)^{1/2} \tag{15}$$

The profile of a Doppler-broadened spectral line is shown in Figure 5. Substituting numerical values for the constants, (15) becomes

$$\Delta\nu_{1/2} = 2.92 \cdot 10^{-20} \nu_0 \left(\frac{T}{M} \right)^{1/2} \tag{16}$$

Figure 5. Doppler-broadened spectral line, where $\Delta\nu_{1/2}$ is the FWHM and ν_0 is the absorbed frequency when the atom is at rest in the frame of the laser.

where M is the mass of the absorbing atom in kilograms and T is the absolute temperature in

Kelvin. So from Eq. (15) the FWHM of a Doppler broadened line is a function of ν_0 , M , and T .

5 Doppler-Free Saturated Absorption Spectroscopy

The apparatus for the Doppler-free saturated absorption spectroscopy of rubidium is shown in Figure 6. The output beam from the laser is split into three beams, two less intense probe beams and a more intense pump beam, at the thick beamsplitter. The two probe beams pass through the rubidium cell from top to bottom, and they are separately detected by two photodiodes. The two photodiodes form a balanced photodetector. After being reflected twice by mirrors, the more intense pump beam passes through the rubidium cell from bottom to top. Inside the rubidium cell there is a region of space where the pump and a probe beam overlap and, hence, interact with the same atoms. This overlapping probe beam will be referred to as the first probe beam and the other one the second probe beam.

The signal from the second probe beam will be a linear absorption signal, where the spectral lines are Doppler-broadened. The signal is shown in Figure 7a, and it was photographed from the screen of an oscilloscope. This signal was obtained by blocking the pump and the first probe beams. There are two Doppler-broadened lines shown in the Figure 7a, and a portion of the triangular waveform that drives

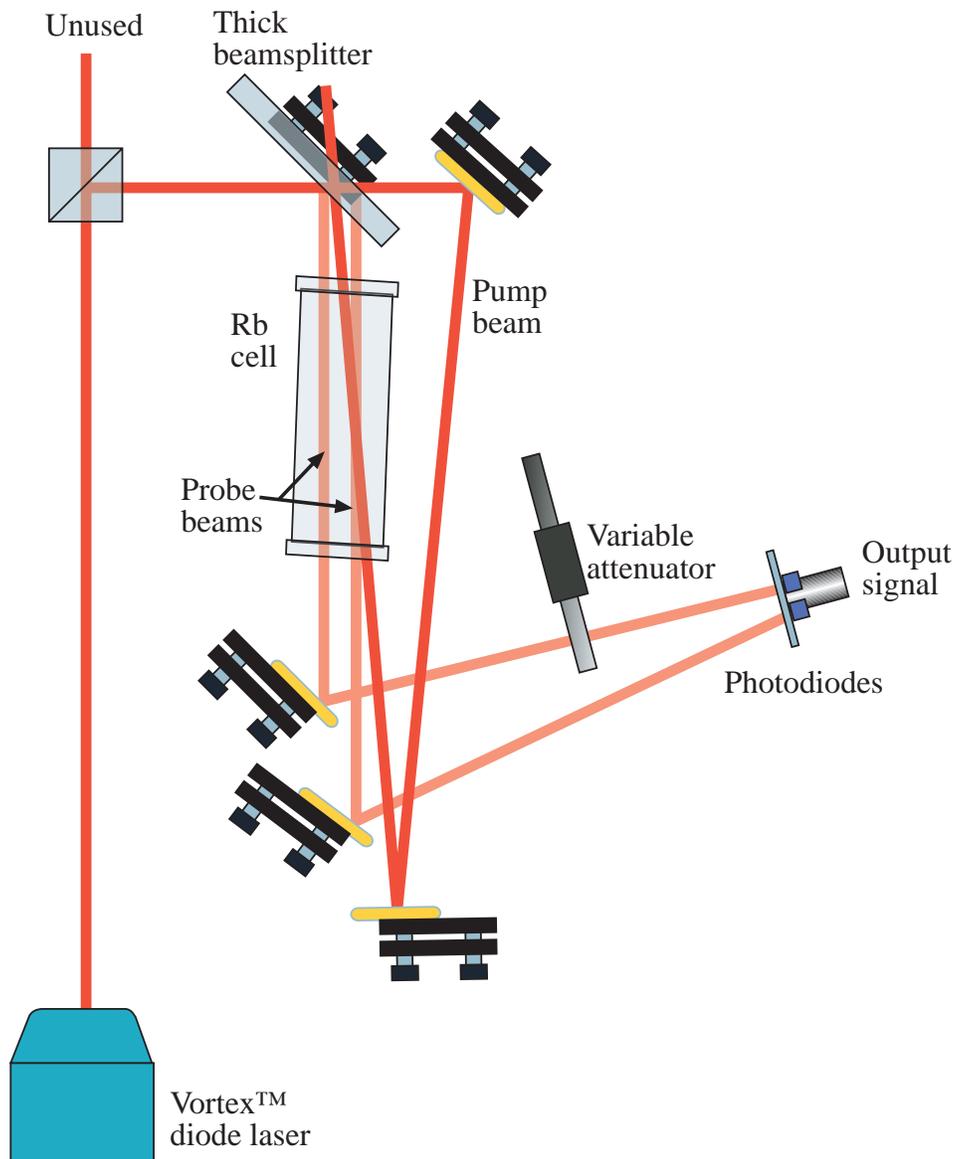


Figure 6. Apparatus for Doppler-free saturated absorption spectroscopy of rubidium.

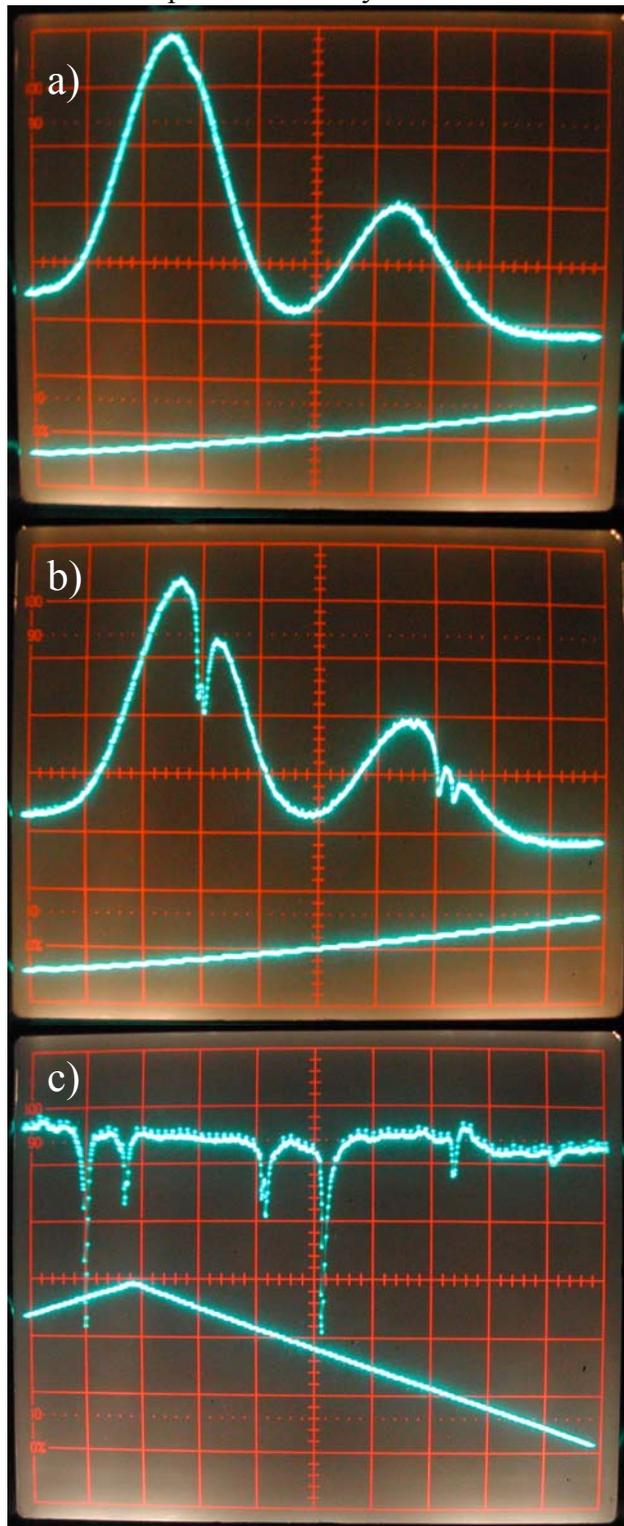


Figure 7. Doppler-free spectroscopy probe signals. In each case the ramp (lower curve) is the piezo voltage. The laser frequency increases with increasing piezo voltage. a) Doppler-broadened spectral lines. b) Doppler-broadened spectral lines with hyperfine structure. c) Doppler-free saturated absorption spectral lines.

the piezo, and, hence sweeps the laser frequency, is also shown. The larger amplitude signal is that of the 72% abundant ^{85}Rb and the smaller amplitude signal is that of the 28% abundant ^{87}Rb . The ^{87}Rb transition is the $F = 2$ to $F' = 1, 2$ and 3 transition, and the ^{85}Rb transition is $F=3$ to $F'= 2, 3$, and 4 .

If the second probe beam is blocked instead, then the signal from the first probe beam will be a nonlinear, saturated absorption spectroscopy signal "riding on" the Doppler-broadened line. This signal is shown in Figure 7b, where the two Doppler-broadened lines are the same transitions as in Figure 7a, but note the hyperfine structure riding on these lines.

If the two signals in Figure 7a and Figure 7b are subtracted from each other, then the Doppler-broadened line cancels and the hyperfine structure remains. The two photodiodes are wired such that their signals subtract, and the signal obtained when none of the beams are blocked is shown in Figure 7c for ^{87}Rb . Note that the frequency of the laser of Figure 7c is ramping first up and then down, so one obtains a partial mirror image of the spectrum. The signal shown in Figure 7c is the Doppler-free saturated absorption signal. We now consider in detail the physics behind Figure 7.

We start by focusing on the first probe and pump beams. The pump beam changes the populations of the atomic states and the probe detects these changes. Let us first consider how the pump beam changes the populations, and then we will discuss how these changes effect the first probe signal. As discussed above, because of the Doppler shift only atoms with a particular velocity V_z will be in resonance with the pump beam, and thereby be excited. This velocity dependent excitation process changes the populations in two ways, one way is known as "hyperfine pumping" and the other as "saturation". Hyperfine pumping is the larger of the two effects, and it will be discussed first.

Hyperfine pumping is optical pumping of the atoms between the hyperfine levels of the $5^2\text{S}_{1/2}$ state. This happens in the following manner. Suppose the laser frequency is such that an atom in the $F = 1$ ground state is excited to the $F' = 1$ excited state. The ΔF selection rule indicates that this state can then

decay back to either the $F = 1$ or $F = 2$ ground states, with roughly equal probabilities. When the atom

decays back to the $F = 1$ state it will be re-excited by the laser light and the process repeated. Thus after a very short time interval, most of the atoms will be in the $F = 2$ state, and only a small fraction will remain in the $F = 1$ state. If the atoms never left the pump laser beam, even a very weak laser would quickly pump all the atoms into the $F = 2$ state. However, the laser beam diameter is small and the atoms are moving rapidly so that in a few microseconds the optically pumped atoms leave the beam and are replaced by un-pumped atoms whose populations are equally distributed between the $F = 1$ and $F = 2$ levels. (You are asked to show in an Exercise, that the populations of these two levels are essentially equal at room temperature with the laser turned off.) The average populations are determined by the balance between the rate at which the atoms are being excited and hence optically pumped, and the rate they are leaving the beam to be replaced by fresh ones. Without solving the problem in detail, one can see that if the laser intensity is sufficient to excite an atom in something like 1 microsecond it will cause a significant change in the populations of the $F = 1$ and $F = 2$ levels, that is, more atoms will be in the $F = 2$ level than the $F = 1$ level. This mechanism is called hyperfine pumping since the net effect is pumping the atomic population from the $F = 1$ to the $F = 2$ ground hyperfine level. Although the example used was for the $F = 1$ to $F' = 1$ transition, similar hyperfine pumping will occur for any excitation where the excited state can decay back into a ground state which is different from the initial ground state.

The other process by which the laser excitation to an excited F' level changes the ground state population is known as saturation. It is pointed out in an exercise that when an atom is excited to an F' level it spends 28 ns in this level before it decays back to the ground state. If the pump beam intensity is low, it will stay in the ground state for much more than 28 ns before it is re-excited, and thus on the average almost all the atoms are in the ground state. However, if the pump intensity is high enough, it can re-excite the atom very rapidly. One might expect that if it were very high it would excite the atom in less than 28 ns. In this case most of the population would be in the excited state and very little would be left in the ground state. In fact, because the pump laser can "excite" atoms down just as well as up, this does not happen. For very high intensity a limit is reached where half the population is in the excited state and half is in the ground state. For realistic intensities the population of the excited state will be less than 0.5, something like 2 to 20 % is more typical. The effect of using high power to rapidly pump the atoms to an excited state is known as "saturating the transition" or just saturation. (In many references hyperfine pumping is also referred to as saturation.)

This saturation effect will be present on the transitions that have hyperfine pumping as well as the transitions that do not. However, it is generally a smaller effect than the hyperfine pumping. This can be understood by considering the intensity dependence of these two effects on the population. We previously indicated that the hyperfine pumping starts to become important when the excitation rate is about once per microsecond. From the discussion above, it can be seen that the saturation of the transition will become important when the excitation rate is comparable to the excited state lifetime, or once every 28 ns. Thus hyperfine pumping will occur at much lower intensities than saturation. The intensities you will be using are low enough that the hyperfine pumping will be substantially larger than saturation. This is also why the $F = 2$ to $F' = 3$ and $F = 1$ to $F' = 0$ signals are much smaller than the other transitions in the saturated absorption signals. These two transitions are called "cycling" transitions, in the sense that by selection rules, the atoms decay from the excited state back to the same ground state. Therefore the hyperfine pumping is a much weaker effect. As one increases the intensity these peaks will become larger relative to the other peaks when the saturation becomes effective.

Summarizing, in the absence of laser light the $F = 2$ and $F = 1$ levels have nearly equal populations, and when the pump beam is on and tuned to either the $F = 1$ to $F' = 1$, $F = 1$ to $F' = 2$, $F = 2$ to $F' = 1$, or the $F = 2$ to $F' = 2$ transition, then hyperfine pumping produces a larger population in either the $F = 1$ or the $F = 2$ level.

So how does the hyperfine pumping by the pump beam affect the first probe beam? Well, in the arrangement of Figure 6 imagine that all of the atoms in the rubidium cell are at rest and consider what happens when the laser frequency ν_L is tuned to ν_0 , the frequency of an atomic absorption line of the Rb atoms, for example, the $F = 1$ to $F' = 1$ transition. The hyperfine pumping by the more intense pump beam produces a smaller population in the $F = 1$ level than that in the $F = 2$ level. This means there are fewer atoms in the $F = 1$ level that will absorb power from the first probe beam, hence the number of photons in the first probe beam that reach the photodiode detector will increase. Now the second probe beam is interacting with a different group of atoms in the vapor cell, hence it is not influenced by the pump beam; therefore, the second probe beam's intensity at its photodiode detector will be less than that of the first probe beam. Thus after subtracting the two signals in the current-to-voltage converter, the resulting signal will show the difference between the two probe beams due to the effects of hyperfine pumping by the pump beam. Also both probe beams have the same Doppler broadened absorption (neglecting the effect of the pump beam), and the subtraction cancels this common absorption as shown in Figure 7c, leaving only the pump beam induced difference.

The atoms in the vapor cell will, of course, not all be at rest; instead they will have a distribution of velocities given by Eq. (12), the Maxwell-Boltzman distribution. An atom that absorbs light at frequency ν_0 when at rest, will absorb laser light of frequency ν_L , where ν_L is given by Eq. (11), when the atom moves with velocity $\pm V_z$ along the axis of the vapor cell. Consider the Maxwell distribution of atomic velocities shown in Figure 8, where the number of ground state atoms $N_{gs}(V_z)$ is plotted against the atom velocity V_z .

The positive z axis is arbitrarily chosen in the direction of the probe beams. We will consider the three cases of Figure 8 in order.

(a) $\nu_L < \nu_0$. Atoms moving toward the right are moving toward the pump beam and they will see its light blue-shifted. At appropriate positive V_z (Eq. (11)), the light will be shifted to ν_0 in the rest frame of the atoms, where ν_0 is the frequency of a transition from an F level to an F' level. Atoms moving at this velocity will absorb the pump laser light. The probe beam is pointed to the right, hence atoms moving to the left with the same velocity magnitude will absorb the probe beams. It is important to recognize that the three beams are interacting with three different groups of atom. The two probe beams are interacting with atoms in different regions of the vapor cell moving to the left with velocity V_z , while the pump beam is interacting with atoms moving to the right with velocity V_z . Also the more intense pump beam causes a greater reduction in the number of atoms in the ground state than the less intense probe beams. Subtraction of the two probe beams gives a null signal.

(b) $\nu_L = \nu_0$. Atoms with speed $V_z = 0$ in the region of overlapping first probe and pump beams can absorb light from both the first probe and pump beams. For the $F = 1$ to $F' = 1$ transition, for example, the pump beam depletes the population of the $F = 1$ level, and then the first probe beam passes through with reduced absorption. Reduced absorption of the second probe beam does not occur, hence subtraction of the two signals gives an absorption signal without Doppler broadening. The interaction is nonlinear in that the first probe signal depends upon the fields of both the first probe and pump beams.

(c) $\nu_L > \nu_0$. This is like (a) with directions reverse, and subtraction of the two probe beam signals gives a null result.

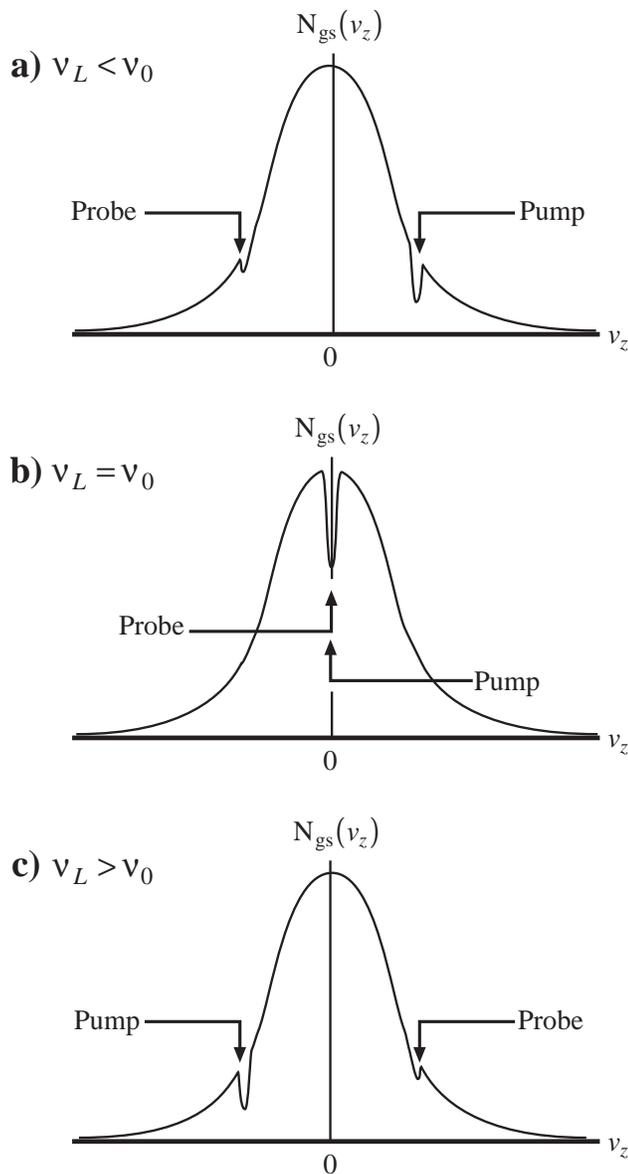


Figure 8. Absorption of pump and probe beams by ground-state atoms, assuming a Maxwell-Boltzmann velocity distribution, for the cases when the laser frequency is a) below ($v_L < v_0$), b) equal to ($v_L = v_0$) and c) above ($v_L > v_0$) the transition resonance frequency.

Methods of tuning and typical tuning rates are: (1) Vortex laser current: (about 200 MHz/mA), and (2) piezoelectric tuning: (about 1 GHz/V). When the laser is tuned to either transition the vapor cell will fluoresce along each beam path. The fluorescence may be observed with either a hand held IR viewer or a CCD surveillance camera. If neither device is available, then place the IR detection card near the cell such that you can observe IR radiation reflected from the card. To search for fluorescence:

1. Start with the laser at a modest lasing current and the piezo voltage set to mid range.
2. Turn on the triangle wave generator with the gain so that the laser piezo is driven through its maximum range (refer to the operating manual).

A crossover peak appears midway between any two transitions that have the same lower level and two different excited levels. This occurs because, when the laser is tuned to the frequency midway between two such transitions, atoms with a particular nonzero velocity can simultaneously be in resonance with both the saturating beam and the probe beam and thus have nonlinear absorption. In other words, the two beams excite resonances to different transitions. For example, those atoms which are moving toward the probe beam with a velocity which gives a Doppler shift exactly equals to half the frequency difference between the two transitions will be shifted into resonance with the higher frequency transition. These atoms will see the pump beam with exactly the opposite Doppler shift. This shift will make the pump beam frequency just right to excite these same atoms to the lower frequency transition. As a result, the absorption is saturated not in stationary atoms but rather in two complementary classes of moving ones.

6 Experiments

The Vortex™ tunable diode laser manufactured by New Focus Inc used in this experiment is a research-quality instrument, and it should be handled with considerable care. Please read the operating instructions before using the laser.

6.1 DOPPLER-BROADENED SPECTRAL LINE

In this part of the experiment you will observe the Doppler-broadened line without hyperfine structure riding on it, that is, you will observe a signal similar to that shown in Figure 7a

The first step is to tune the laser frequency to either the $F = 1$ to $F' = 0, 1, 2$ transition or the $F = 2$ to $F' = 1, 2, 3$ transition. Remember the hyperfine structure will not be resolved in this Doppler-broadened case. Methods of tuning and typical tuning

- Look for fluorescence as you tune the laser current up to its maximum value, but do not exceed the maximum value (the instrument should prevent you from doing so). If you do not see the fluorescence, then see your instructor.

Once fluorescence is observed, block the second probe beam such that it does not reach the photodiode as shown in Figure 6. (It does not matter if the beam is blocked before or after passing through the vapor cell.) Block the pump beam before it reaches the vapor cell. Trigger the scope from the sync out pulse of the triangle wave generator. Adjust the offset of the triangle wave generator and, if necessary, make slight adjustments in the laser current until the spectral line of ^{87}Rb is centered on the triangle waveform as shown in Figure 7. Record the signal with the data acquisition system at your station, for example, a digital camera.

6.2 DOPPLER-BROADENED LINE WITH HYPERFINE STRUCTURE

Remove the block from the pump beam. You should observe a Doppler-broadened line with hyperfine structure riding on it, somewhat like that shown in Figure 7b. Record the observed signal BE SURE AND RECORD THE piezo VOLTAGE SCALE FOR ALL SPECTRA. YOU WILL LATER USE THE MICHELSON TO CONVERT THIS VOLTAGE INTO FREQUENCY.

6.3 DOPPLER-FREE SATURATED ABSORPTION SPECTRAL LINES

Remove the block from the second probe beam, and, hence, observe Doppler-free saturated absorption spectral lines. Reduce the gain of the triangle waveform generator and observe the effect on spectral line resolution. Insert the variable optical attenuator in the beam path to observe the signal using reduced laser power. Record the signal.

Now tune the laser, as described above, to the other transitions so that you observe transitions from both hyperfine ground states of ^{87}Rb ($F=2$ to F' , and $F=1$ to F'), and from both ground states of ^{85}Rb ($F=2$ to F' , and $F=3$ to F') and record similar data. Figure 7c shows the $F = 2$ to F' transitions for ^{87}Rb .

6.3.1 Calibration: Michelson Interferometer

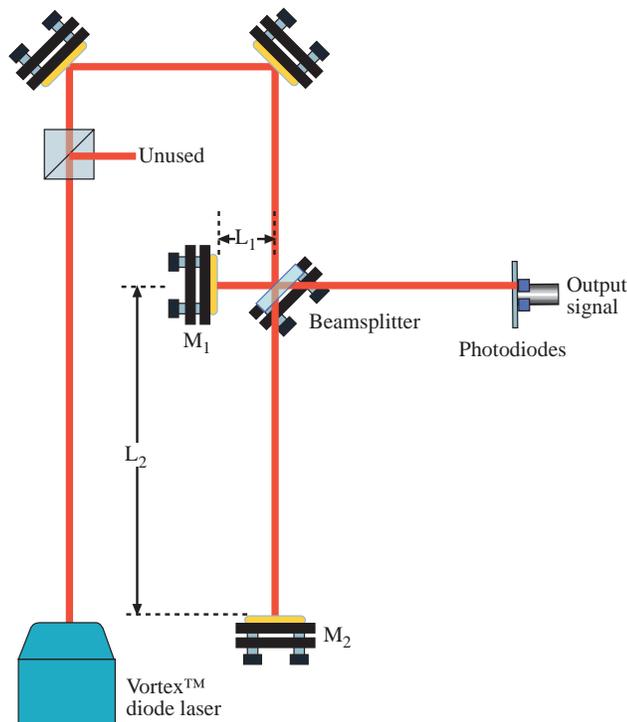


Figure 9. Michelson interferometer.

A Michelson interferometer will be used to calibrate the horizontal axis of the oscilloscope in frequency units. Set up a Michelson interferometer similar to the one shown in Figure 9. Your setup does not have to look like the one in the figure —only the critical elements are shown.

The laser beam is divided by a beam splitter (a microscope slide is an adequate beam splitter), and the two beams are recombined at the beam splitter after they have traveled distances of $2L_1$, and $2L_2$. After returning to the beam splitter, and thereafter, the two beams will have a phase difference due to their path difference given by:

$$\begin{aligned} \Phi_1 - \Phi_2 &= \frac{(2L_1 - 2L_2)}{\lambda} 2\pi \\ &= \frac{2\nu}{c} (L_1 - L_2) 2\pi \end{aligned} \quad (17)$$

where ν is the laser frequency. The frequency is not fixed, rather it is being swept by the triangle

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waveform applied to the piezo of the Vortex™ laser, hence as the frequency changes the phase difference will change. The changes in frequency and phase are expressed by writing Eq. (17) as

$$\Delta(\Phi_1 - \Phi_2) = 2 \frac{\Delta\nu}{c} (L_1 - L_2) 2\pi. \quad (18)$$

The intensity of the two superimposed waves, aside from a constant of proportionality, is given by,

$$\begin{aligned} I &= EE^* \\ &= \left(E_1 e^{i\left(\frac{2L_1}{\lambda} 2\pi - 2\pi\nu t\right)} + E_2 e^{i\left(\frac{2L_2}{\lambda} 2\pi - 2\pi\nu t\right)} \right) \cdot \left(E_1 e^{i\left(\frac{2L_1}{\lambda} 2\pi - 2\pi\nu t\right)} + E_2 e^{i\left(\frac{2L_2}{\lambda} 2\pi - 2\pi\nu t\right)} \right)^* \\ &= |E_1|^2 + |E_2|^2 + 2E_1 E_2 \cos[\Delta(\Phi_1 - \Phi_2)] \end{aligned} \quad (19)$$

where Δ was inserted to indicate the change in the phase from sweeping the frequency. The interference is a maximum whenever

$$\Delta(\Phi_1 - \Phi_2) = 2\pi \quad (20)$$

Substituting Eq. (20) into (18) yields the frequency spacing of the interference maxima

$$\Delta\nu = \frac{c}{2(L_1 - L_2)}. \quad (21)$$

Figure 10 is a photo of the scope screen showing the triangle waveform and the interference pattern, where the frequency spacing between maxima is given by Eq. (21).

There are a few things it is helpful to know in order to get good fringes from the interferometer. First, you should not be misled by the weak fringes you will get from the interference of the beams reflecting off the two sides of the beam splitter. They will not have the correct spacing and they will not require both beams returning off M3 and M4. Second, you need to realize that in order to give good fringes the two beams must not only overlap at the photodiode, but they must also be parallel. It is possible to have them come back from M3 and M4 and go through different parts of the beam splitter but still overlap at the photodiode. By drawing some simple triangles you should be able to convince yourself that in this case the path length difference $L_1 - L_2$ will be different at different places where the beams overlap. This will result in a series of bright and dark fringes across the photodiode, which you can see if you look

closely with the viewer. If the phase difference across the face of the photodiode is a full 2π there will always be a bright and dark fringe present and changing the laser frequency will give no change in the total power on the photodiode, and hence no signal. The larger the angle between the beams the closer will be the spacing of the fringes. As you adjust the beams to be parallel (but still overlapping) the spacing between the dark fringes will become larger until it is as large as the photodiode, and you will see a large modulation in the photodiode output as you change the frequency. Often the easiest way to get good fringes is to first make the beams as parallel and overlapping as possible and then do the final adjustments by looking at the photodiode output and align the beams to get the largest fringes as you ramp

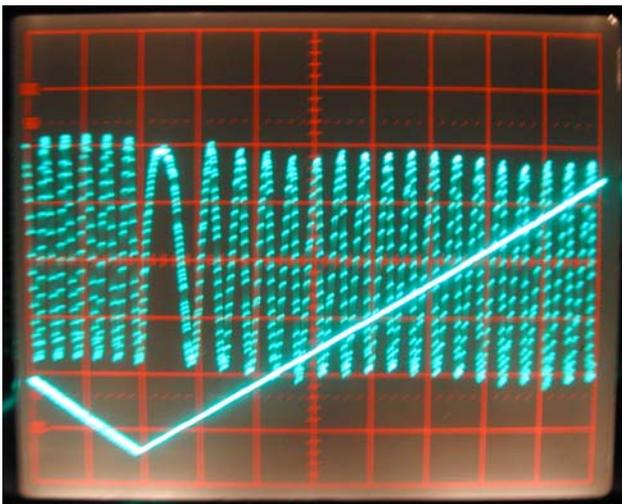


Figure 10. Interference fringes and the triangle waveform. The dotted appearance is caused by the digital oscilloscope used in this photo.

the laser frequency. The final thing you need to realize is that you want the beams to go nearly but not exactly back along the incident laser beam. If they are going exactly back they will go back into the laser and cause the frequency to jump around (optical feedback). This usually shows up as a lot of noise on the fringe pattern signal you see on the oscilloscope. If you keep these things in mind you should not have trouble aligning and using the Michelson interferometer with the following procedure:

4. Choose L_1 and L_2 such that $\Delta\nu$ is approximately 0.5 GHz.
5. Remove the photodiode and place an IR detection card near the output beam splitter. Adjust the tilts of the mirrors M_1 and M_2 such that the two beams are superimposed.
6. Place the IR detection card far away from the beam splitter. Check the superposition of the two spots and adjust the beam splitter mount to overlap the spots. Move the card back near the beamsplitter and re-align M_2 if necessary. Go back and forth with the card far away and near and adjust mounts as indicated above. Once alignment is achieved the scope trace should resemble Figure 10.

Carefully calibrate the horizontal axis of the oscilloscope for the gain setting of the triangle wave generator you used in measuring the spectra. If you determine how large a frequency change is produced for each volt that is applied to the piezo drive, you can then convert the horizontal scale of the spectra from ramp voltage to frequency, and then determine splittings and linewidths. You might check that different ranges on the ramp generator give the same calibration factor.

As the laser frequency is changed you will observe a cosine modulation on the oscilloscope. However, if you make a large sweep you will see sudden jumps in the signal as if the phase has abruptly changed. What has actually happened is that the laser has jumped to a different frequency (a so called "mode hop" as was discussed in the Diode Laser writeup). The frequency where the laser jumps will move if you change the laser current. Under ideal conditions with this laser setup you can get a scan of about 8 GHz without a mode-hop. More typically you will get continuous scans (no mode hops) of 3 or 4 GHz. If the frequency range over which you get a continuous scan is shorter than this it probably means that the vertical alignment is off. The length of continuous scan can also be affected by the laser temperature.

7 Data Analysis

1. Measure the FWHM for each of the two Doppler-broadened lines of ^{87}Rb . Compare your values with your calculated value from Exercise 9.
2. Measure the separation of the hyperfine lines for the $F=2$ to F' transitions, use Eq. (10) to obtain a theoretical expression for the frequency separation of the hyperfine lines, and then solve for the constants A and B for the $5^2P_{3/2}$ state of ^{87}Rb . Compare your results with the accepted values given earlier. If you have time, carry out the same analysis for the other transitions.

8 References

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9 Prelab Exercises

The parentheses indicate the relevant section of this lab manual.

1. (Section 3.1)

Show for a $5s^1$ electron that the term state is a $5^2S_{1/2}$. For a $5p^1$ electron show that the term states are $5^2P_{1/2}$ and $5^2P_{3/2}$.

2. (Section 3.2.2)

Show that the splitting of the $5^2P_{1/2}$ and the $5^2P_{3/2}$ term states due to H_{SO} is $\zeta(r)3\hbar^2 / 2$.

3. (Section 3.2.3)

For ^{87}Rb show: (1) for the $5^2S_{1/2}$ state that $F = 1, 2$; (2) for the $5^2P_{1/2}$ state that $F = 1, 2$; (3) for the $5^2P_{3/2}$ state that $F = 0, 1, 2, 3$.

4. (Section 3.2.3).

Assuming all of the spectral lines are resolved for transitions from the $5^2P_{3/2}$ excited state to the $5^2S_{1/2}$ ground state, how many spectral lines do you expect to observe for ^{87}Rb ?

5. Section 3.2.3)

For the $5^2S_{1/2}$ state of ^{87}Rb substitute numerical values for I , J , and F into the third term of Eq.(10) and show for both $F = 1$ and 2 that this formula only makes sense if $B = 0$ for these two cases.

6. Section 3.2.3)

For ^{87}Rb use Eq. (10) to show that: (1) for the $5^2S_{1/2}$ state the splitting of the $F = 1$ and $F = 2$ levels, $\nu_{1/2,2} - \nu_{1/2,1}$ is 6834.7 MHz, as shown in Figure 3, (2) for the $5^2P_{3/2}$ state the splitting of the $F = 3$ and $F = 2$ levels, $\nu_{3/2,3} - \nu_{3/2,2}$ is 267.1 MHz, as shown in Figure 3, (3) the frequency spacing $\Delta\nu = \nu_{3/2,2} - \nu_{1/2,2}$ is 194.0 MHz as shown in Figure 4. (4) Now that you know how to do the calculations using Eq. (10), just use the energy level spacings given in Figure 3 and Figure 4 to show the frequency of transitions a and b are $3.846 \times 10^{14} - 2.7933 \times 10^9$ Hz and $3.846 \times 10^{14} - 2.7147 \times 10^9$ Hz, respectively, hence the separation of these two spectral lines is 79 MHz. (5) Show that the frequency separation of spectral lines a and l is 6.993 GHz and then show that their wavelength separation is 0.0142 nm.

7. (Section 4)

Show that the FWHM of the 780 nm spectral line of Rb at 297 K due to Doppler-broadening is 513 MHz. The spectral absorption lines you will observe using absorption spectroscopy will be Doppler-broadened. However, the absorption lines you will observe using saturated absorption spectroscopy will not be Doppler-broadened, in fact, they will approach the minimum or natural linewidth determined by the Heisenberg uncertainty principle.

8. (Section 4)

For an atom with no Doppler shift, the linewidth is determined by the Heisenberg uncertainty principle, $\Delta E \approx \hbar / \tau$, where τ is the excited state lifetime, and the range of frequencies absorbed is given by $\Delta \nu \approx 1 / 2\pi\tau$. From the expression for $\Delta \nu$ make a reasonable estimation of an expression for the FWHM, $\Delta \nu_{1/2}$, and using the 28-ns lifetime of the $^2P_{3/2}$ excited state of ^{87}Rb calculate the FWHM. Compare your answer to the 513 MHz FWHM in Exercise 7.

9. (Section 4)

When you perform absorption spectroscopy on ^{87}Rb how many spectral lines do you predict will be resolved? To answer this question use the 513 MHz FWHM from Exercise 7 and use the spectral line separations that are given in Figure 4.

10. (Section 5)

With the laser off, the ratio of the number of atoms in the energy level $E_{1/2,2}$ ($J = 1/2, F = 2$), $N(E_{1/2,2})$, to the number of atoms in the energy level $E_{1/2,1}$ ($J = 1/2, F = 1$), $N(E_{1/2,1})$, is determined by Maxwell-Boltzmann statistics:

$$\frac{N(E_{1/2,2})}{N(E_{1/2,1})} = \exp\left[-\left(\frac{E_{1/2,2} - E_{1/2,1}}{kT}\right)\right] \quad (22)$$

Assuming a temperature of 295 K, show that the above population ratio is 0.999, hence the two levels are essentially equal populated with the laser off. You may want to refer to Exercise 2 and the discussion that precedes it in the Introduction to Magnetic Resonance.

11. (Section 5)

Show that laser excitations from $F = 1$ to $F' = 2$, $F = 2$ to $F' = 1$, and $F = 2$ to $F' = 2$ will also produce hyperfine pumping of the $5^2S_{1/2}$ ground state, where the first excitation produces a larger population in the $F = 2$ than in the $F = 1$ level, and the other two excitations produce the larger population in the $F = 1$ level. Also show that laser excitations from $F = 2$ to $F' = 3$ and $F = 1$ to $F' = 0$ will not produce hyperfine pumping of the $5^2S_{1/2}$ state. One way to show the production, or non-production, of hyperfine pumping for each case is to show the upward transition and all of the allowed downward transitions on an energy level diagram like that in Figure 3, where the hyperfine levels of the $5^2P_{1/2}$ state do not need to be shown since we are not interested in transitions to these levels.