

DOPPLER-FREE SATURATED ABSORPTION SPECTROSCOPY: LASER SPECTROSCOPY

1 Introduction

In this experiment you will use a diode laser to carry out laser spectroscopy of rubidium atoms. You will study the Doppler broadened optical absorption lines, and will then use the technique of saturated absorption spectroscopy to study the lines with resolution beyond the Doppler limit. This will enable you to measure the hyperfine splittings of one of the excited states of rubidium. You will use a Michelson interferometer to calibrate the frequency scale for this measurement. This experiment was developed by Carl Wieman and uses techniques currently in use in his and other research laboratories at CU. It is being duplicated at a number of other schools. To facilitate the widespread use of this experiment, which is new to undergraduate labs, Daryl Preston in collaboration with Wieman wrote a very lengthy writeup. The purpose of this writeup was to allow schools to duplicate the experiment, even if the lab instructors were not familiar with laser spectroscopy. As a result, it is somewhat different in style and far more detailed than your other writeups may have been. Although it is long, it does explain in detail everything you will need to know to carry out and analyze this experiment so you will have little need of other references. References to other chapters or experiments refer to items in Preston's book "The Art of Experimental Physics".

Apparatus

Vortex™ 780-nm diode laser system

Rubidium vapor cell

Photodiode detector circuit (see reference 1)

Triangle waveform generator

optical breadboard

3/8"-thick transparent plastic or glass (beam splitter)

4 flat mirrors

4 mirror mounts

9 or more posts (for mounting vapor cell, photodiode detectors, mirrors, and beam splitter)

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 Fine structure

Saturated absorption spectroscopy

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.

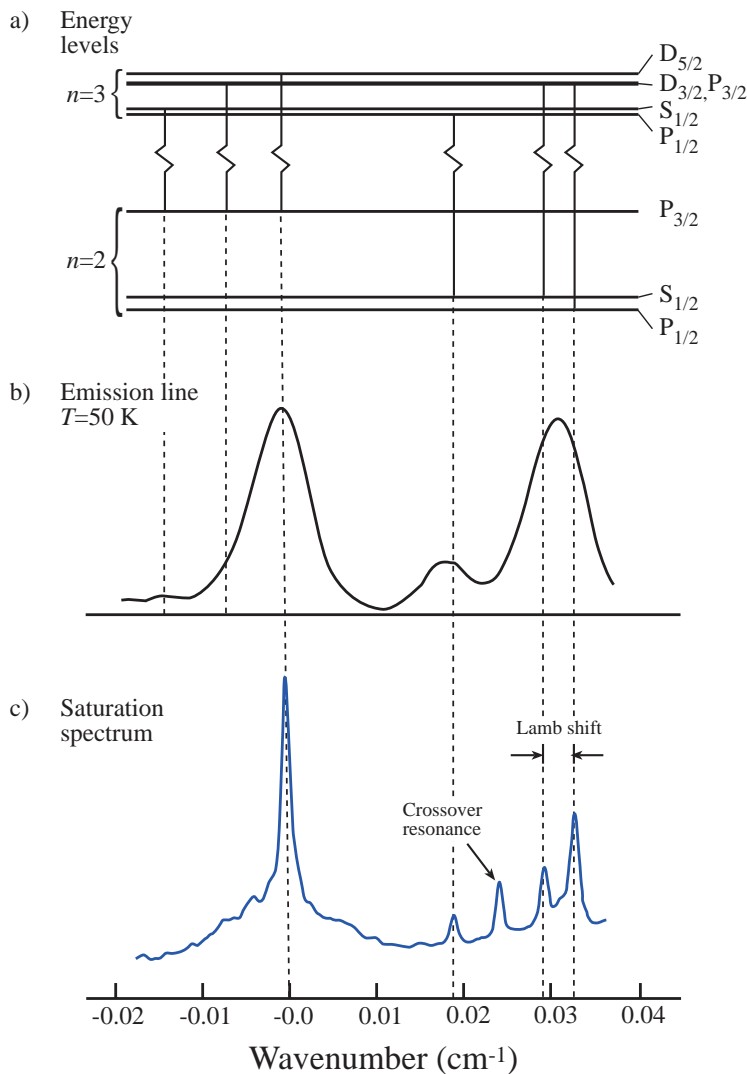


Figure 1. The Balmer α line of atomic deuterium. a) Energy levels and allowed transitions, b) the emission spectrum, c) an early saturated absorption spectrum

beam propagating to the left, the "probe" beam, causes the transition indicated with a solid line. 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, 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 $3.85 \times 10^{14} \text{ Hz} \pm 10 \times 10^6 \text{ Hz}$. Reference 1 points out a method of reducing the spectral linewidth to less than 1 MHz.

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

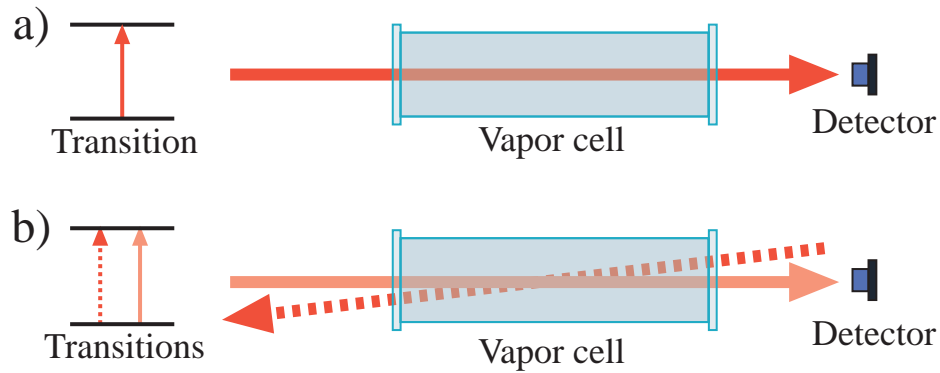


Figure 2. In linear spectroscopy (a) the radiation reaching the detector is proportional to the radiation incident on the sample. In nonlinear spectroscopy (b) the radiation reaching the detector is dependent on both beams.

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,

$$\mathbf{J} = \mathbf{L} + \mathbf{S}, \quad (1)$$

where their magnitudes are

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

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:

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

We label the 5 terms in this equation, in order, as K , V , H_{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

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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 does depend on the particular atom. Ignoring relativistic terms in H , it is H_{SO} that produces the fine structure splitting of rubidium.

Using equation (4) and forming the dot product of $\mathbf{J} \cdot \mathbf{J}$, we solve for $\mathbf{L} \cdot \mathbf{S}$ and obtain

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

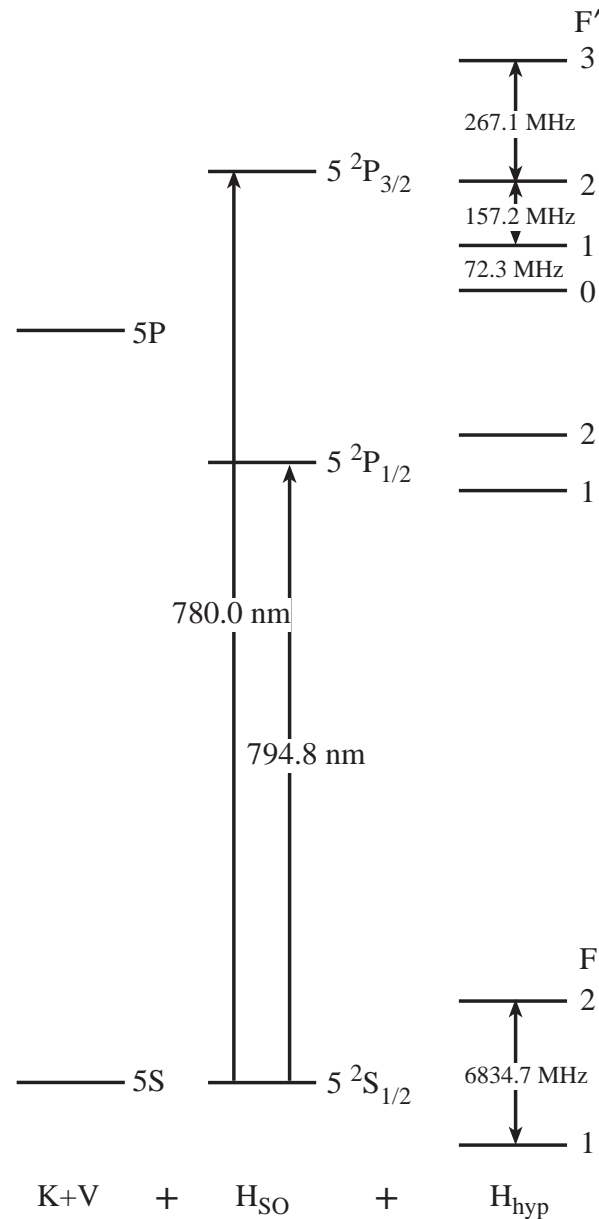


Figure 3. Each interaction in Eq. (3) and its effect on the energy levels of the 5s and 5p electron is shown. The energy level spacings are not to scale. The hyperfine levels are for ^{87}Rb .

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

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

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 $|J - I|, |J - I| + 1, \dots, J + I - 1, J + I$. (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 energy levels in Figure 3b are split by the hyperfine interaction into the levels shown in Figure 3c 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 9 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 $I = 3/2, J = 3/2,$ 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 :

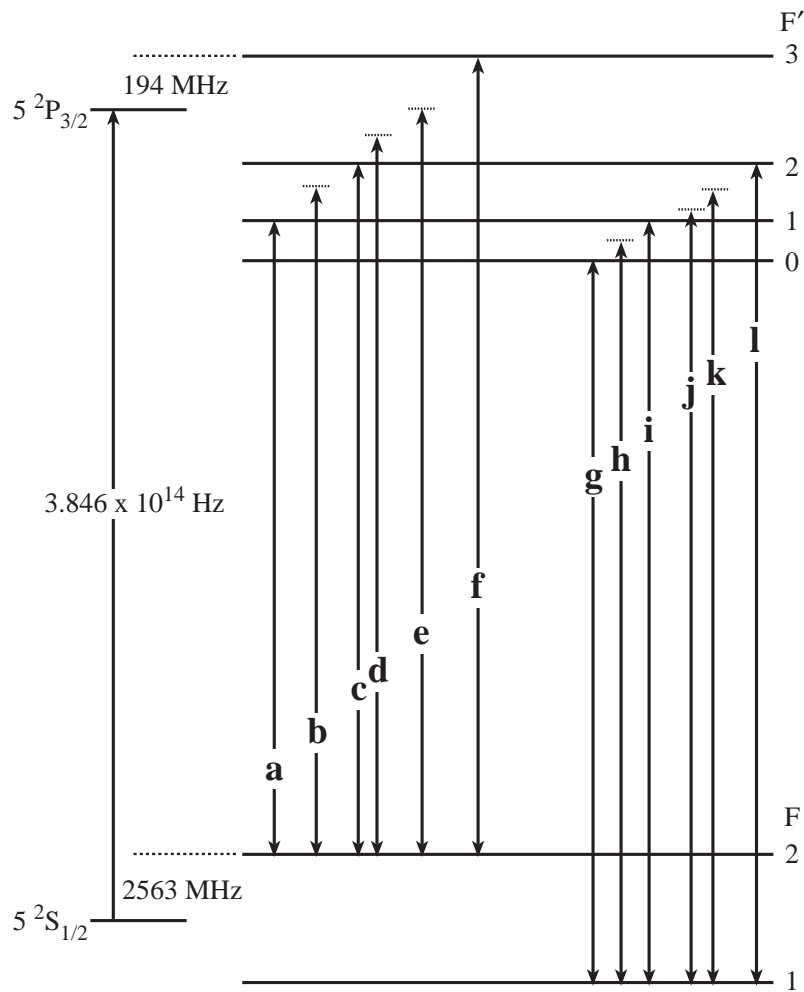


Figure 4. Saturated absorption transitions for ^{87}Rb . The spectral line separation will be derived in the exercises or can be figured out from Figure 3

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

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

For the $5^2\text{S}_{1/2}$ 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 $5^2\text{P}_{3/2}$ state of ^{87}Rb , the accepted values of A and B are 84.85 MHz and 12.52 MHz, respectively. For the $5^2\text{S}_{1/2}$ of ^{85}Rb the accepted value of A is 1011 91 MHz, and for the $5^2\text{P}_{3/2}$ 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 $5^2\text{P}_{3/2}$ 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. What determines the linewidth of a Doppler broadened line? To answer this question we do some theoretical physics.

We first consider the Doppler effect qualitatively. If an atom is moving toward or away from a laser light source, then it "sees" radiation that is blue or red shifted, respectively. If an atom 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 observes 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 observes 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):

$$v_z = (\nu_L - \nu_0)c / \nu_0; \quad dv_z = c d\nu_L / \nu_0 \tag{13}$$

Substituting (16) into (15), 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

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

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

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

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

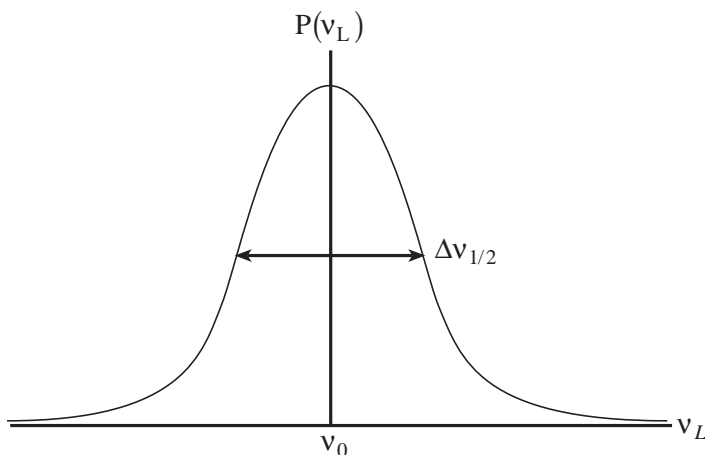


Figure 5. Doppler-broadened spectral line, where $\Delta\nu_{1/2}$ is the FWHM and ν_0 is the absorbed frequency when the atom is at rest in the frame of the laser.

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

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. 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.

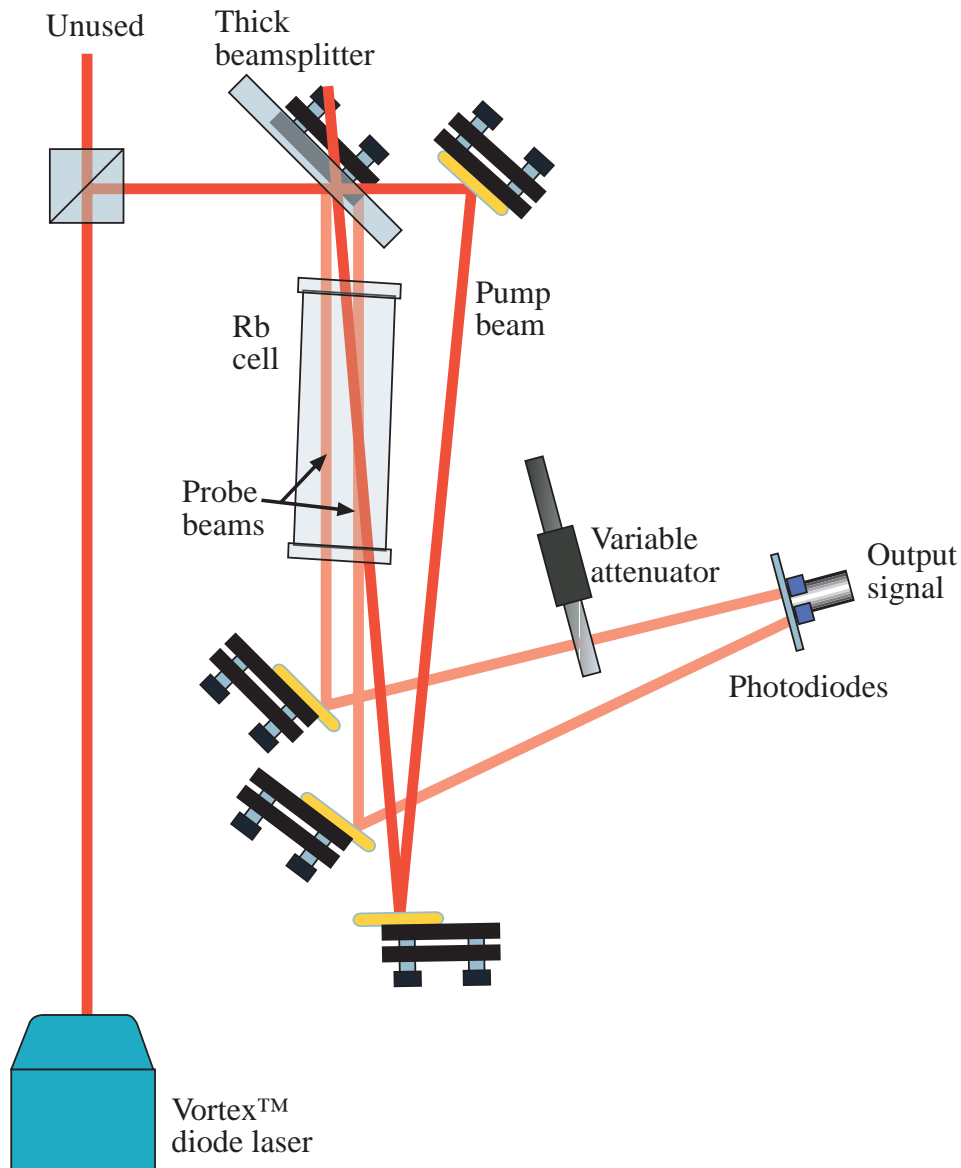


Figure 6. Apparatus for Doppler-free saturated absorption spectroscopy of rubidium.

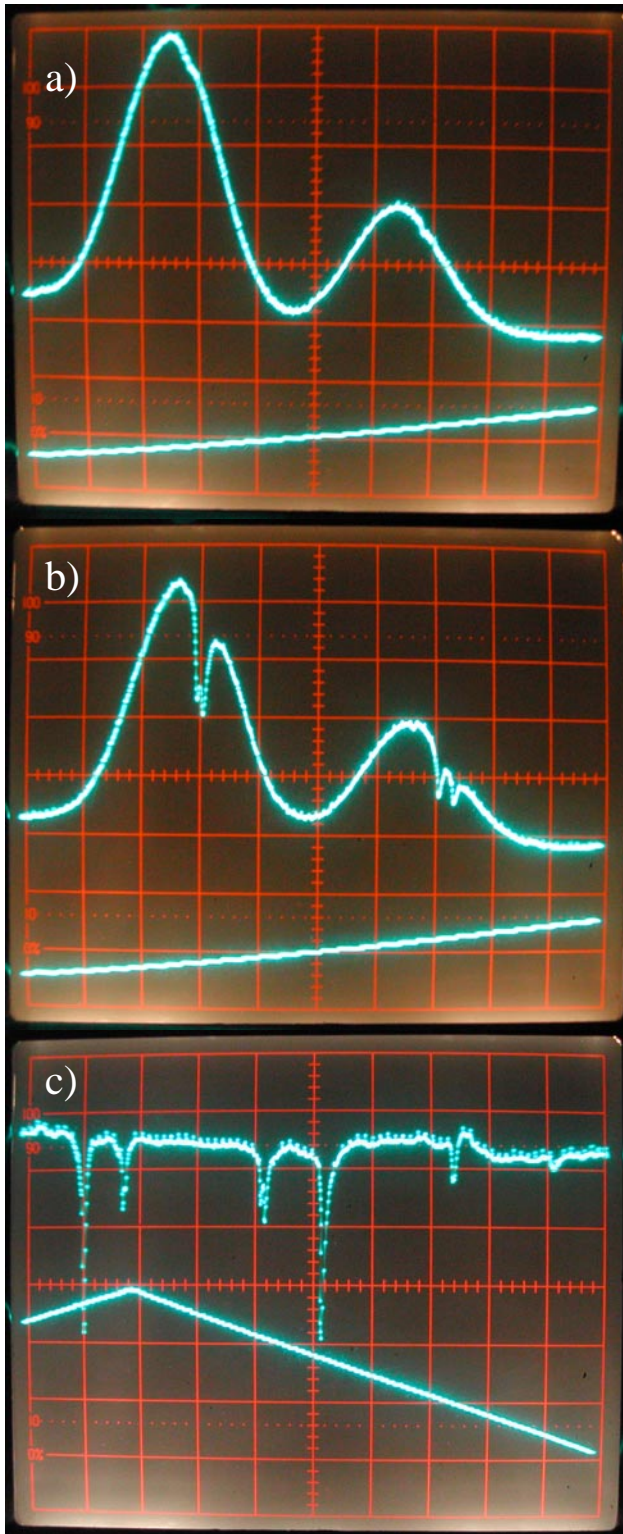


Figure 7. Doppler-free spectroscopy probe signals. In each case the ramp (lower curve) is the piezo voltage. The laser frequency increases with increasing piezo voltage. a) Doppler-broadened spectral lines. b) Doppler-broadened spectral lines with hyperfine structure. c) Doppler-free saturated absorption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

The signal from the second probe beam will be a linear, absorption spectroscopy signal, where the spectral lines will be 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 first probe beams. There are two Doppler-broadened lines shown in the Figure 7a, and a portion of the triangular waveform that drives 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 only is blocked 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 shown in Figure 6 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 . In Figure 7c. 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