

Optical Pumping

Eric D. Black

September 27, 2004

Sometimes you want to magnetize a gas, to align all of the little magnetic moments of the gas atoms in the same direction. You might plan to have somebody inhale the polarized gas, then look at the insides of their lungs using magnetic-resonance imaging, or you might want to investigate the electronic level structure of the gas atoms, and polarizing them is the first step in a spectroscopy program. There are lots of times, in a modern research lab, where you want a polarized gas, and the usual way to produce it is by optical pumping.

In this lab you will learn basic optical pumping, and you will practice it on a gas of Rubidium atoms. Rubidium is a fairly simple atom. Its electronic structure closely approximates that of a Hydrogen atom, so we can get a pretty good theoretical understanding. However, even in Hydrogen-like atoms there are interesting effects that can be investigated spectroscopically using optical pumping, and we will do that in this lab.

1 Prelab I

1.1 Rubidium's atomic energy level structure

Rubidium in its atomic state has just one valence electron and can be well approximated by a one-electron-atom model. Its nuclear properties are different from Hydrogen, however, and this will give it a different energy-level structure. There are two commonly-occurring isotopes of Rubidium in nature, ^{85}Rb , with a natural abundance of 72%, and ^{87}Rb , with an abundance of 28%. In this section you will learn all about ^{87}Rb , but the structure of ^{85}Rb follows the same general principles.

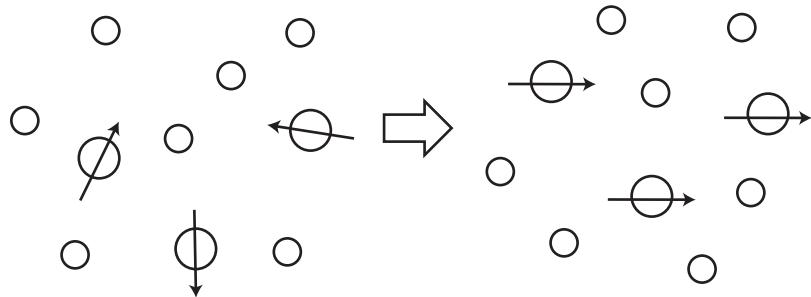


Figure 1: Optical pumping can be used to polarize a gas of atoms that have magnetic dipole moments. In practice, these atoms are often mixed with a nonpolar *buffer gas*, which helps keep the polarized atoms from touching with the walls of the container and losing their polarization.

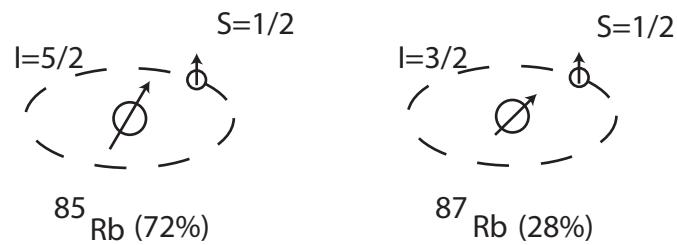


Figure 2: There are two commonly-occurring isotopes of Rubidium found in nature, ^{85}Rb and ^{87}Rb . Both have only one valence electron and can be approximated as one-electron atoms. The major difference between the isotopes is in the nuclear spin I .

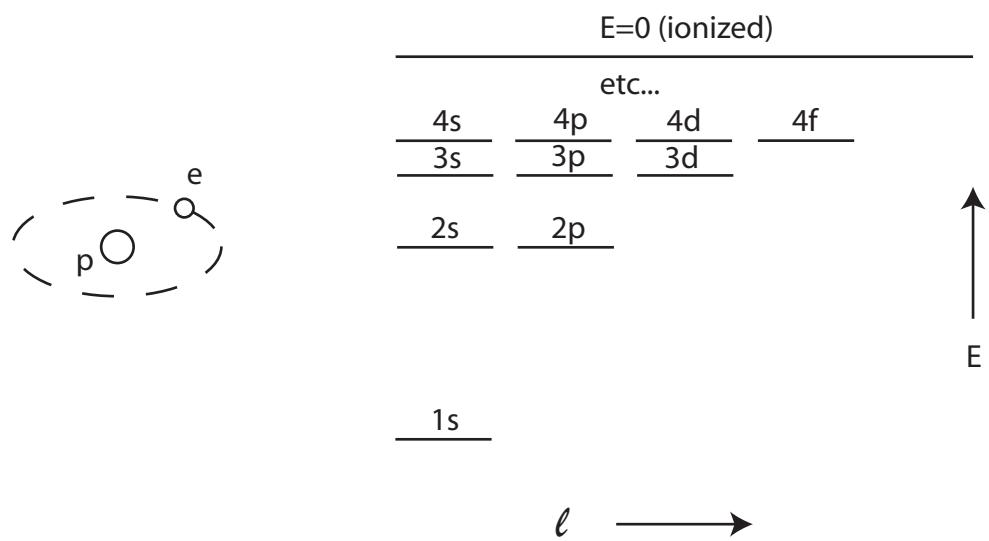


Figure 3: The basic energy-level structure of a one electron atom. Bound states have negative energy and are quantized, with different allowed amounts of angular momentum for each energy level. To first approximation, all of the different angular momentum states have the same energy for a given energy number n .

A crude model of a one-electron atom that neglects both the spin of the electron and the nucleus gives a level structure like that shown in Figure 3. Here, the levels are labeled by their energy quantum number n and the orbital angular momentum ℓ , and the notation is $n\ell$, where $n = 1, 2, \dots$ and $\ell = s, p, d, f, \dots$. The use of letters to represent ℓ is left over from the old days of spectroscopy, before people knew what was going on inside atoms, and the letters stand for “sharp,” “principal,” “diffuse,” and “fundamental.” (After f they go as the alphabet: g, h, i, etc.) This notation is known, appropriately, as *spectroscopic notation*.

The spinless model does a pretty good job, considering how simple it is. A low-resolution spectrometer will see an emission or absorption spectrum of a one-electron atom (any alkali atom) that really does agree with the predictions of this model. However, if we use a higher-resolution instrument, like the optical-pumping apparatus in this lab, additional structure will show up that the spinless model cannot account for. There are many corrections to the basic, spinless model of the atom, but we will only be concerned with three: taking into account the spin of the electron and of the nucleus, and the addition of an external magnetic field.

The spin of the electron is important because it couples with the orbital angular momentum. Essentially, the electron feels an effective magnetic field as it moves through the electrostatic field of the nucleus. Since the electron has an intrinsic magnetic moment, due to its spin, its energy level will be higher if it is aligned opposite this effective magnetic field than if it is aligned with it. This leads to an additional term in the Hamiltonian of the form $\mathbf{L} \cdot \mathbf{S}$, which changes the eigenstates ever so slightly. Only the states with non-zero orbital angular momentum are affected, and the net result is that they are split into multiple levels, depending on the orientation of the spin. This is known as *fine-structure* splitting and is illustrated in Figure 4.

Once we take electron spin into account, we have to modify our spectroscopic notation. The new notation is called, appropriately enough, the *modified spectroscopic notation* or *Russel-Saunders notation*, and it goes like this. Each state is labeled by $^{2S+1}L_J$, where S is the electron spin, L is the orbital angular momentum, and $J = L + S$ is the total angular momentum. The energy quantum number n is dropped. For our single-electron atom, S is always $1/2$, but the notation allows treatment of multi-electron atoms with their spins added in proper quantum fashion. The orbital angular momentum is again labeled with the old spectroscopic letters (S,P,D,etc.), but this time they are capitalized. The quantum number J ranges from $|L - S|$

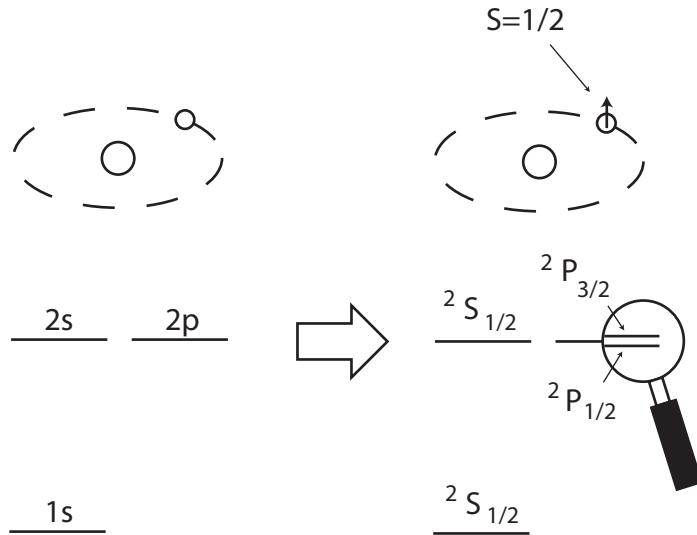


Figure 4: *Fine structure*—Taking the spin of the electron into account leads to a small splitting of the levels with orbital angular momentum. Now that there are two angular momenta to consider, orbital and spin, we must modify our notation. The new notation is $^{2S+1}L_J$, where S is the electron spin ($1/2$ for one electron), L is the orbital angular momentum (S,P,D,etc.), and $J = L + S$ is the total angular momentum. The energy quantum number n is dropped.

to $|L + S|$, according to the rules of addition of angular momentum.

Adding the spin of the nucleus I introduces three additional terms to the Hamiltonian, each of which are related to the nuclear magnetic moment associated with I . You can look up a detailed treatment of this in your favorite quantum mechanics text, but the end result is that the electron's orbital angular momentum, the electron's spin, and the nucleus' spin all add up to a total angular momentum $F = I + J$, with eigenvalues $|I - J| < F < |I + J|$. Each of these eigenstates of F has a slightly different energy, due to the coupling between the electron and the nucleus' intrinsic magnetic field, as illustrated in Figure 5.

Finally, applying an external magnetic field further splits each of the F levels according to their projection number M . (See Figure 6.) All this says is that the energy of the system (atom plus magnetic field) is different for

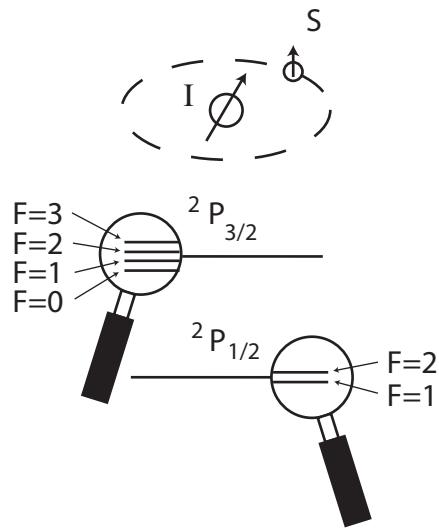


Figure 5: *Hyperfine structure*— Adding the spin of the nucleus further splits the levels. Here we have a closeup of the 2p levels, split into $^2\text{P}_{1/2}$ and $^2\text{P}_{3/2}$ by the fine-structure correction. These fine-structure levels are split according to the total angular momentum $F = I + J$. This example illustrates the splitting for ^{87}Rb , which has $I = 3/2$. The $^2\text{S}_{1/2}$ level also gets split, in the same way as $^2\text{P}_{1/2}$.

different orientations of the atom, which is a perfectly reasonable assertion. This splitting is known as the *Zeeman effect*. The fundamental physics of the Zeeman effect is essentially the same as that of the fine and hyperfine splittings in that all three are due to coupling between magnetic fields and magnetic moments. The difference is that the Zeeman effect is due to a magnetic field we apply in the lab, whereas the fine and hyperfine splittings are due to intrinsic magnetic fields that occur inside the atom itself.

If the magnetic field is relatively weak, the Zeeman splitting is given by a simple expression.

$$E_Z = g_F \mu_B B M$$

Here, E_Z is the Zeeman energy, the difference in energy between the state with projection number M and the unperturbed hyperfine level. The magnetic field strength is B , μ_B is the Bohr magneton, and the coupling constant g_F is known as the Landé g-factor. This g-factor is not the same as the bare electron's g-factor, and it is different for each hyperfine level.

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)},$$

and

$$g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}.$$

A word about magnitudes is in order here. The fine-structure splitting is very, very small compared with the spacing between the excitation levels 1s, 2s, etc., and the hyperfine splitting is much, much smaller still. The Zeeman splitting, however, can conceivably be as big as the hyperfine splitting, or even bigger, if a strong enough magnetic field is applied. In practice, it is difficult to generate high enough fields to get well into the strong-field regime, but even in this lab we will begin to see deviations from the weak-field limit at our highest field values.

1. Evaluate the Landé g-factors for the highest- F hyperfine levels of the $^2S_{1/2}$ state, in both isotopes of Rubidium.
2. Evaluate the Zeeman splitting for the highest- F hyperfine levels of the $^2S_{1/2}$ state, for both isotopes of Rubidium. What frequencies of electromagnetic radiation would you need to induce transitions between these Zeeman levels in a 1 Gauss magnetic field?

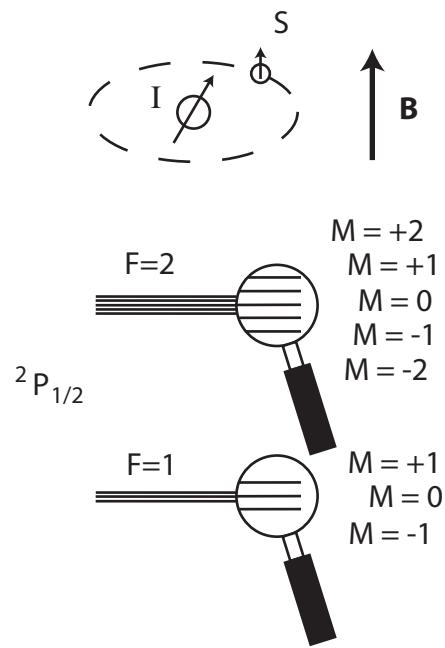


Figure 6: *Zeeman splitting*— If we add an external magnetic field, each of the individual F levels splits according to its angular-momentum projection number M , *i.e.* the orientation of its magnetic moment. Of all the splittings we have considered so far, this is the only one whose magnitude can be comparable to the next larger correction, in this case the hyperfine splitting.

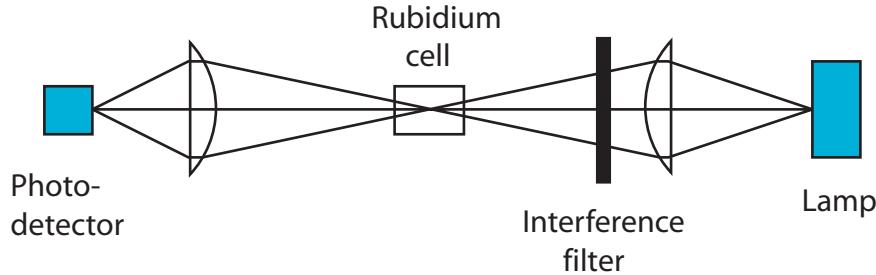


Figure 7: Setup for measuring absorption vs. temperature. Use unpolarized light for this part of the lab, with only the interference filter to block everything but the 795 nm line.

3. Sketch the energy-level structure of ^{85}Rb , including both fine and hyperfine corrections.

1.2 Absorption and cross sections

4. Show that the intensity of light falls off exponentially as the light travels through an absorbing medium. Specifically, show that

$$I = I_0 e^{-\sigma \rho x},$$

where I is the intensity of the light after it travels a distance x in the medium, I_0 is the initial intensity, σ is the effective cross section, and ρ is the density of atoms in the medium. *Hint:* Treat the whole problem classically, pretending that both the photons and the atoms are spheres. Consider any photon that touches an atom to be taken out of the incident beam, either by scattering or absorption, and calculate the probability that a photon will make it through a distance x without touching an atom.

2 Lab I

2.1 Absorption and cross section

Set the apparatus up as shown in Figure 9. Locate the controls for the cell heater, set the temperature to 300 K, and cover the apparatus with the black

cloth provided to block out any stray room light. After the temperature has stabilized (about thirty minutes), measure the transmitted intensity of the light with the photodetector. Measure the transmission as a function of temperature from 300 K to 400 K, then fit the theoretical absorption you calculated in the prelab to your data. You will need to know the density of the Rubidium gas as a function of temperature, which is given approximately in Table I.

Table I: Density of Rubidium gas as a function of temperature [3].

T (K)	ρ (m ⁻³)
290	3.3×10^{15}
300	1.1×10^{16}
310	2.9×10^{16}
320	7.5×10^{16}
330	1.8×10^{17}
340	4.3×10^{17}
350	8.3×10^{17}
360	1.5×10^{18}
370	3.7×10^{18}
380	6.3×10^{18}
390	1.2×10^{19}
400	2.4×10^{19}

(When performing the fit, don't forget to add a constant intensity to your theoretical formula to account for nonresonant light that leaks into the system.) From your fit, calculate a value for the effective cross section σ . How does this compare with the geometric cross section of Rubidium or the photons?

3 Prelab II

3.1 Circularly-polarized light

5. Read Section 2.4 of Fowles [1], and do Exercises 2.5 and 2.6 (p. 54). Pay particular attention to the discussion on how to generate circularly-polarized light using a quarter-wave plate and linear polarizer. You will have to do this for the lab.

6. (Optional) Read Section 2.5 in Fowles, on the Jones Calculus, and do Exercises 2.7 through 2.10.
7. (Optional) Show that the electric-dipole selection rules for circularly polarized light require either $m_f = m_i + 1$ or $m_f = m_i - 1$, *but not both*, for a given handedness of the light. Here, m_f and m_i are the final and initial angular-momentum-projection numbers along the direction of propagation of the light. *Hint:* Don't evaluate the matrix elements! Express the Hamiltonian for the circularly-polarized light as a spherical tensor operator, then use the Wigner-Eckart theorem [4] to decide which transition probabilities are zero and which are not.

3.2 Optical pumping

Now consider what happens when a photon is introduced that has the right energy to lift an electron out of the 1s, $^2S_{1/2}$ state and put it into the 2p, $^2P_{1/2}$ state. The hyperfine levels of the 1s, $^2S_{1/2}$ state are close enough together that they are more or less equally populated. That is, their spacing is much less than $k_B T$, and the valence electron in any given atom has about an equal probability of being in any F state with any M . An electron in any one of these states will get excited by the photon into one of the (F, M) states in $^2P_{1/2}$, but there are only certain states it can go to. For example, the electron's final M value, after it lands in the excited state, can't differ from its initial M value by more than one. A photon is just not capable of changing M by more than one.

Whether M changes by $+1$, -1 , or 0 depends on the nature of the photon. If the applied magnetic field is parallel to the direction of propagation of the photon, then a right-circularly-polarized photon will always induce transitions that have $\Delta M = +1$. Left-circularly-polarized light produces $\Delta M = -1$. The same thing is true for emission. An electron can fall from the 2p, $^2P_{1/2}$ level into the 1s, $^2S_{1/2}$ level and emit a photon with right or left circular polarization, depending on whether ΔM is $+1$ or -1 . For emission, both cases are equally likely. For absorption, we control which kind of photon is incident, so we can control ΔM .

Now we are in a position to polarize atoms. If we shine right-circularly-polarized light and apply a magnetic field, along the same direction, to a gas of atoms, each absorption will force $\Delta M = +1$, whereas each emission event will have, *on average*, $\Delta M = 0$. Repeated absorption and reemission will

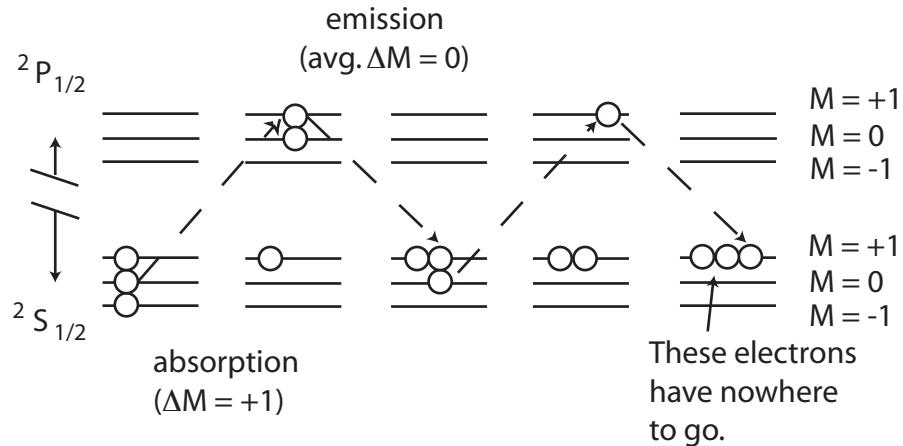


Figure 8: The electrons in a population of atoms can be pumped into the highest M level of the ground state by repeated absorption and reemission of photons. If we only provide right-circularly-polarized photons, then $\Delta M = +1$ for every absorption. The atoms can reemit any kind of photon, so on average $\Delta M = 0$ for reemission. A few iterations of this, and all the electrons end up in the highest M level of the ground state. This diagram illustrates optical pumping between $F = 1$ sublevels in Hydrogen, where $I = 1/2$. Rubidium works the same, except that there are more levels.

“walk” the electrons into the 1s state with the highest value of M , as shown in Figure 8.

Once polarized, the gas will have a total magnetization which we could, in principle, measure. However, it is much more useful to look at how well the gas absorbs the photons. As long as absorption can occur, the gas will be partially opaque to our circularly-polarized light. Once all of the electrons are pumped into the highest M state and the gas is polarized, absorption can no longer occur. The pumped electrons have nowhere to go that would satisfy the $\Delta M = +1$ requirement. When it is in this state, the gas is transparent to our photons.

Our gas will remain transparent as long as the polarization is maintained. If we switch off the magnetic field, the polarization will be lost, and the gas will become opaque. Similarly, pumping, and hence absorption, can occur if we scramble the electrons in the ground state by applying an RF signal that is resonant with the Zeeman splitting. In either case, the light falling on the

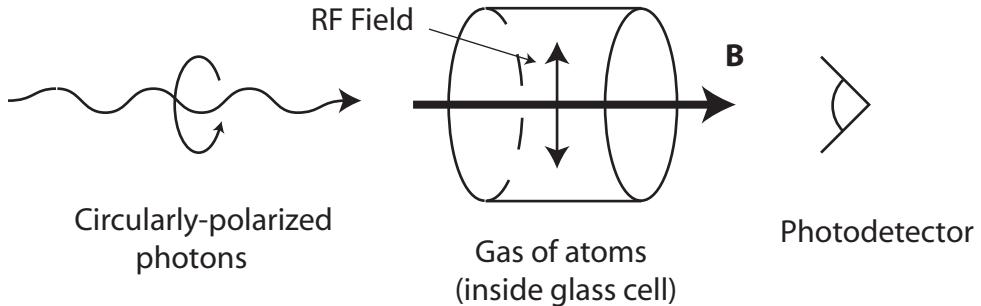


Figure 9: The basic setup for observing absorption changes due to optical pumping. The gas being pumped is inside a cell, usually a cylinder made of glass and sealed, along with a buffer gas. A magnetic field and circularly-polarized light are both applied along the same axis, and the transmission of the cell is measured with a photodiode. An additional radiofrequency (RF) magnetic field may be applied to drive transitions within the Zeeman levels and depolarize the gas.

photodetector drops when pumping is occurring, and is at a maximum when the gas is polarized.

8. The absorption diagram in Figure 10 is representative of a single gas species. The cell you will be working with contains two different isotopes of Rubidium. What absorption curve do you expect for it? Sketch your expected signal, and explain your reasoning.

3.3 The quadratic Zeeman effect

Our simple model of the Zeeman splitting predicts equal splitting between the M levels of a given state. This simple model is only an approximation, valid in the limit of very weak magnetic fields. In the weak-field limit, the Hamiltonian is diagonal in the (F, M_F) basis. However, in the strong-field limit, it is the (J, M_J, I, M_I) basis that diagonalizes the Hamiltonian, and therefore the energy level structure is different. In the transition region between these two regimes, the spacing between the levels is not equal.

9. Read Section E of Chapter XII in Cohen-Tannoudji, volume two [2], on the Zeeman effect. Find a formula for the spacing between the Zeeman

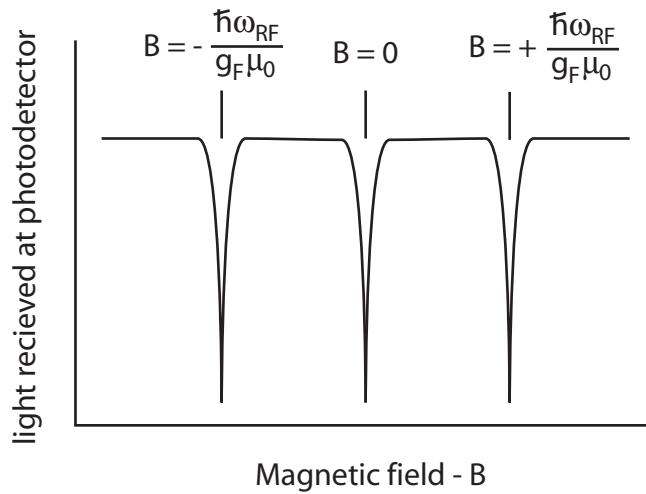


Figure 10: A typical absorption diagram for an apparatus like that shown in Figure 9. Light falling on the photodiode drops to zero whenever pumping occurs and is maximized when the gas is polarized. Depolarization occurs both when the field is switched off and when the RF field becomes resonant with the Zeeman splitting of the ground state.

levels in the Hydrogen 1s, $F = 1$ state as a function of magnetic field ($\hbar\omega_0$). Plot the frequency necessary to induce transitions between these levels as a function of magnetic field from $\hbar\omega_0 = 0$ to $\hbar\omega_0 = A\hbar^2$.

10. Sketch the absorption-vs.-magnetic-field curve you expect in an optical pumping experiment using Hydrogen in the intermediate-field regime. Explain your reasoning.
11. Now consider Rubidium. In this case, $I = 3/2$ or $I = 5/2$, depending on the isotope. Sketch the Zeeman diagram (for arbitrary field) for both isotopes. Identify the energy eigenstates in each field regime. *Hint:* Think about how angular momenta add. If you understand this, you shouldn't have to do any calculations for this exercise, other than adding some fractions.
12. Sketch what you expect the absorption-vs.-field curve to look like for Rubidium in the intermediate-field regime. Explain your reasoning.

3.4 Rabi oscillations

13. Read Section C.3 of Chapter IV in Cohen-Tannoudji, volume one, on induced oscillations between coupled states in a two-state system. You may want to look over all of Section C, just to get used to the notation. If an RF electromagnetic field is used to drive transitions between levels, what relationship do you expect there to be between the frequency of the Rabi oscillations and the strength of the applied field?
14. Read Compliment H_{IV} of Chapter IV in Cohen-Tannoudji. What is one observable signature of the presence of an unstable state? What can a quantitative measurement of this signature tell you about the unstable state?

4 Lab II

4.1 Weak-field resonances

Start with the apparatus set up as you left it at the end of the last lab session, where you measured absorption as a function of temperature for unpolarized

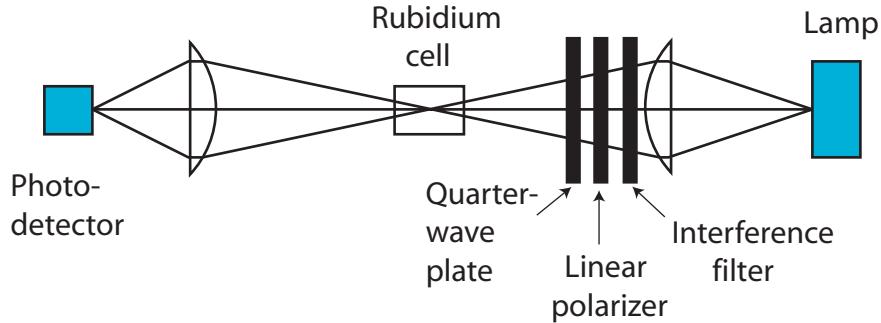


Figure 11: Setup for measuring resonant absorption with circularly polarized light. Orient the apparatus to minimize the effect of the Earth's magnetic field.

light. Now add the linear polarizer and quarter waveplate to produce circular polarization, making sure the orientations are correct. (Remember the description in Fowles of this arrangement, and do this “downstream” of the interference filter.) You will apply a horizontal magnetic field for this experiment, and you will need to make sure that all other magnetic fields are reduced as far as possible. Remove any magnetic objects (anything iron or steel, along with any permanent magnets) from around the apparatus, and turn the apparatus so that its axis is North-South. You will fine-tune this alignment and use the vertical coils to compensate for the Earth’s magnetic field in a little bit.

Use a second linear polarizer after the quarter wave plate to check the circular polarization. If it is properly circular, the light intensity falling on the photodetector should not depend on the angle of the second polarizer. Why is this, and what would it mean if the intensity did depend on the polarizer’s angle? Use an oscilloscope to measure the output of the photodetector, and once you have verified circular polarization, don’t forget to remove the second polarizer!

Now null all the magnetic fields in the Rubidium cell except those along the optical axis. Set the cell temperature to 320 K, and allow it to come to equilibrium. There are two controls for the horizontal-field, one for a static field (large), and one for a slow sweep field (small). Leave the static part at zero, cover the apparatus with the black cloth, and set up a slow sweep. Send both the sweep voltage and the output of the photodetector

to an oscilloscope, and display the photodetector output versus the sweep field in the scope's XY mode. You should see a dip in the output of the photodetector when the total field (applied plus Earth's) on the cell is zero along the optical axis. This dip is the same one as the central absorption line, at $B=0$, in Figure 10, but it will probably not be as well defined. Adjust the rotation of the apparatus and the current through the vertical compensating coils to achieve the minimum width for this absorption line, and experiment with different sweep rates to get a clean-looking line. Take a screen shot of your best zero-field absorption line, print it out, and tape it in your lab book. This line will probably not occur at zero current, since there may be some residual Earth's magnetic field along the optical axis. Record the current, so that you can later determine this residual field.

Now apply a signal to the RF coils at a frequency of about 150kHz. Right now its amplitude is not important. You will optimize it after you have found the Zeeman resonances. Hunt around until you have found the Zeeman resonances, then adjust the RF amplitude to get the cleanest lines. Take a screenshot of the absorption curve, including the Zeeman transitions, and put it in your lab book. How does this compare with the prediction you made in the prelab? The relative amounts of ^{85}Rb and ^{87}Rb inside the cell are not necessarily the same as those found in nature. Assuming that both isotopes have the same cross sections, what are the isotopic abundances of each species inside the cell?

Measure the current in the sweep field at each Zeeman resonance, calculate the magnetic field, and compare with your expectations from the prelab. The relationship between current in the coils and magnetic field at the cell is the usual Helmholtz one,

$$B = 9 \times 10^{-3} \text{Gauss} \frac{NI}{R}, \quad (1)$$

where I is the current in amps, $N = 11$ is the number of turns in the coil, and $R = 0.16\text{m}$ is the average radius of the coils. Do this for several RF frequencies and both isotopes. Plot the Zeeman resonance frequencies of both isotopes as a function of magnetic field. (Don't forget to take the residual Earth's field into account!) Do the slopes of these lines agree with your expectations?

(Optional) Use the Zeeman resonances to calibrate both the sweep-field and static-field coils, obtaining formulas analogous to Equation 1 but more accurate. You will need this if you choose to do study the intermediate-field

Zeeman effect quantitatively using the Breit-Rabi equations, which will be an optional exercise later in the lab.

4.2 Intermediate-field resonances: Quadratic Zeeman effect

Apply a large, horizontal magnetic field using the static-field coils, and sweep around the Zeeman resonances. Does the structure of these resonances agree with your predictions from the prelab? Do you see the expected number of lines? Do this for several different amplitudes of the RF excitation, take screenshots of your results, and comment on what you see. *Hint:* You will have to determine what the best values of the static field, RF frequency and amplitude, etc. should be to do this experiment. Describe, in your lab book, why you chose the values you decided to use.

(Optional) Look up, or derive, a quantitative formula for the energies of the Zeeman transitions in the intermediate-field limit for ^{87}Rb . The formulas for the Zeeman levels in an arbitrary field are known as the *Breit-Rabi* equations in the literature. The Breit-Rabi equations for Hydrogen are given at the end of Chapter XII in Cohen-Tannoudji, which you read as part of your prelab exercises. Make a quantitative prediction for the spacing between the RF transitions for ^{87}Rb , and compare it with your observations. You may want to take data at more than one field value. Also recall that, in order to do this experiment properly, you need to have done the optional field-calibration exercise earlier.

4.3 Rabi oscillations

For this experiment you will want to start with an already-polarized gas, then suddenly turn on an RF field that is resonant with the Zeeman splitting and see what happens. Pick a relatively low RF frequency that will be resonant with the Zeeman splitting in the weak-field limit. Turn off the sweep field, and adjust the static field until you are on resonance for one of the Zeeman transitions. Attach a square-wave signal with an amplitude of 0 to +5 volts and a frequency of a few hertz to the modulation input on the front panel, to chop the RF signal on and off. Look at the transmitted light intensity versus time on an oscilloscope, take a screenshot for your notebook, and explain what you see.

In the prelab exercises, you made two predictions about Rabi oscillations. The first was a quantitative relationship between the frequency of the oscillations and the RF amplitude. Do this experiment for several values of the RF amplitude, and check your prediction. The second prediction was more qualitative. Look for the signature you predicted, and discuss what you see.

References

- [1] Grant R. Fowles, *Introduction to Modern Optics*, Dover Publications, Inc., New York (1989).
- [2] Claude Cohen-Tannoudji, Bernard Diu, and Frank Laloë, *Quantum Mechanics*, John Wiley and Sons, new York (1977).
- [3] *Optical pumping of Rubidium: Guide to the Experiment INSTRUC-TOR'S MANUAL*, TeachSpin, Inc. (2002).
- [4] Rammamurti Shankar, *Principles of Quantum Mechanics*, Plenum Press, New York (1980).