

## OPTICAL PUMPING OF RUBIDIUM VAPOR

### Introduction

The process of optical pumping is a beautiful example of the interaction between light and matter. In the Advanced Lab experiment, you use circularly polarized light to pump a particular level in rubidium vapor. Then, using magnetic fields and radio-frequency excitations, you manipulate the population of the pumped state in a manner similar to that used in the Spin Echo experiment. You will determine the energy separation between the magnetic substates (Zeeman levels) in rubidium as well as determine the Bohr magneton and observe two-photon transitions.

Although the experiment is relatively simple to perform, you will need to understand a fair amount of atomic physics and experimental technique to appreciate the signals you witness.

#### *A simple example of optical pumping*

Let's imagine a nearly trivial atom: no nuclear spin and only one electron. For concreteness, you can think of the  ${}^4\text{He}^+$  ion, which is similar to a Hydrogen atom, but without the nuclear spin of the proton. Its ground state is  $1S_{1/2}$  ( $n = 1, S = 1/2, L = 0, J = 1/2$ ). Photon absorption can excite it to the  $2P_{1/2}$  ( $n = 2, S = 1/2, L = 1, J = 1/2$ ) state. If you place it in a magnetic field, the energy levels become split as indicated in Figure 1. In effect, each original level really consists of two levels with the same energy; when you apply a field, the "spin up" state becomes higher in energy, the "spin down" lower. The spin energy splitting is exaggerated on the figure. At low fields, the amount of splitting is very nearly proportional to the field strength.

If we shine light of appropriate frequency (and sufficient frequency spread) onto the atoms, we can induce transitions from either of the lower levels to either of the upper ones. If the light is circularly polarized with the appropriate "handedness" along the axis of the  $B$  field, the transitions will be subject to the requirement  $\Delta m = +1$ , as indicated by the arrow in Figure 1. Such behavior illustrates a selection rule: in this case, the rule amounts to nothing more than conservation of angular momentum along the field axis, since circularly polarized photons carry angular momentum of  $\hbar$  along their direction of propagation.

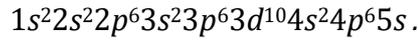
The excited atoms can decay to one of the split levels of the ground state through spontaneous emission, as illustrated in Figure 1. For this process the selection rule is  $\Delta m = 0$  or  $\pm 1$ , because the emitted photon can have any polarization.

Although the atoms can relax to either of the split ground-state levels, only the  $m = -1/2$  level can absorb the incident polarized photons—so there is a net tendency to "pump" atoms into the  $m = +1/2$  level of the ground state. When a significant fraction of the atoms has been pumped, the transparency of the sample to this particular frequency and polarization increases, because the number of possible absorber's has decreased.

#### *The rubidium atom level scheme*

Alkali-metal atoms are often used for optical pumping because they have a single valence electron and thus resemble a hydrogen atom. The photon wavelength needed for optical excitation is in a convenient range of the spectrum, where diode lasers are available. However, alkali-metal atoms have a nuclear spin, which complicates their level structure. We will use rubidium atoms in this experiment.

The occupied orbitals of rubidium atom are



The first 36 electrons are in closed sub-shells and so their total angular momentum is zero. The 5s electron acts like hydrogen's electron. In particular, we will look at transitions between the sub-levels of the 5s–5p

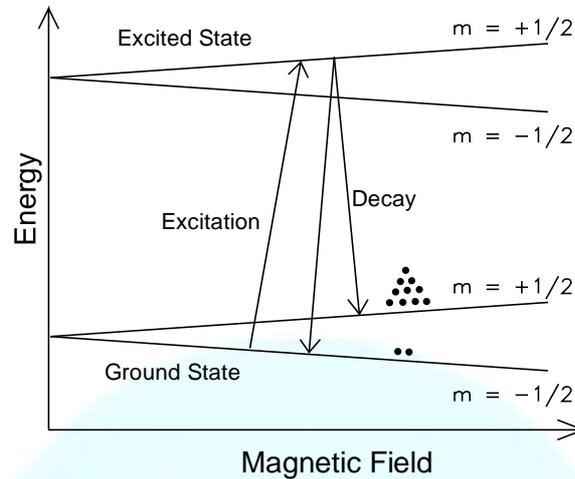


Figure 1: Level diagram of a simplified atom. The energy level splitting, which is not drawn to scale, increases with applied magnetic field. There is only one transition for absorption of a positive helicity photon. There are two paths for the decay of the atom. The atoms pile up in the  $m = +1/2$  state.

transition. To understand the energy levels of rubidium, we need to consider the interaction between the electron and the nucleus and between both and an external magnetic field. The angular momentum of the valence electron is given by

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

where  $\mathbf{L}$  is the orbital angular momentum and  $\mathbf{S}$  is the spin angular momentum. To get the *total* angular momentum of the atom, we must also include the nuclear component which is denoted as  $\mathbf{I}$ . The atom angular momentum is then given by

$$\mathbf{F} = \mathbf{J} + \mathbf{I} \quad (2.)$$

There are four principle mechanisms that determine the rubidium energy levels. They are, in order of decreasing strength, the coulomb interaction, the spin-orbit interaction, the hyperfine interaction, and the Zeeman splitting. These are shown in Figures 2a and 2b. The Hamiltonian for the Coulomb interaction is given by

$$H_o = \frac{\mathbf{p}^2}{2m} - \frac{Z'e^2}{r}. \quad (3.)$$

It is characterized by the principal quantum number  $n$  and the orbital angular quantum number  $l$ . The typical energy scale for  $H_o$  is 1 eV.

The first perturbation to  $H_o$ , called spin-orbit coupling, is due to the interaction of the spin magnetic moment of the electron and the magnetic moment produced by the orbit of the electron around the nucleus. It is given by,

$$H_{so} = \frac{g_e Z e^2}{4m^2 c^2 r^3} \mathbf{L} \cdot \mathbf{S} \quad (4.)$$

where  $g_e$  is the electronic “g factor.” The resulting level splittings are called the fine structure and are typically  $10^{-4}$  eV or  $\alpha^2 H_0$  where  $\alpha$  is the fine structure constant. The levels are split according to their value of  $J$ , where  $J(J+1)\hbar^2$  is the eigenvalue of the operator  $\mathbf{J}^2$ . Of course, there is no energy shift to the 5s state because  $l = 0$ . The fine structure states are labeled with the spectroscopic notation  $^{2S+1}L_J$ , where

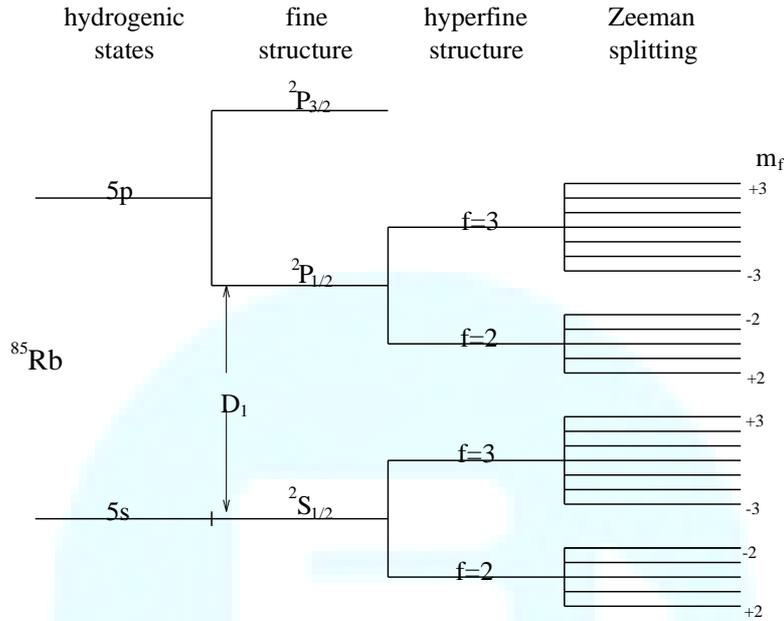


Figure 2: Level diagram of  $^{85}\text{Rb}$ .  $I = 5/2$ . The splittings are not to scale.

$S(S+1)\hbar^2$  is the eigenvalue of  $\mathbf{S}^2$ , and  $L(L+1)\hbar^2$  the eigenvalue of  $\mathbf{L}^2$ . The values of  $L$  are denoted by letters: ‘S’ for  $L = 0$  and ‘P’ for  $L = 1$ . Radiation corresponding to the energy difference between the  $^2S_{1/2}$  and  $^2P_{1/2}$  levels of rubidium is termed the  $D_1$  line; its wavelength is roughly 794.8 nm. The  $^2P_{3/2}$  state is separated from the ground state by an energy corresponding to 780 nm wavelength, it is called the  $D_2$  line.

The next splitting of the energy levels arises from an interaction of the magnetic moment of the electron with the nuclear magnetic moment. It is called the hyperfine interaction, with the Hamiltonian give by,

$$H_{hf} = \frac{Ze^2 g_N}{2mM_N c^2 4\pi} \mathbf{S} \cdot \left[ -\mathbf{I} \nabla^2 \frac{1}{r} + \nabla(\mathbf{I} \cdot \nabla) \frac{1}{r} \right] = A \mathbf{S} \cdot \mathbf{I} \quad (5.)$$

where  $g_N$  is the nuclear “g-value.” Here the levels are split according to their value of  $F$ , the eigenvalue of the operator  $\mathbf{F}^2$ . Rubidium metal has two isotopes,  $^{85}\text{Rb}$  (72% abundance) and  $^{87}\text{Rb}$  (28% abundance). For the  $^2S_{1/2}$  and  $^2P_{1/2}$  states of  $^{85}\text{Rb}$ , which has nuclear angular momentum  $I = 5/2$ , the allowed values of  $F$  are  $5/2 - 1/2 = 2$  and  $5/2 + 1/2 = 3$ . The angular momentum  $I$  of the  $^{87}\text{Rb}$  nucleus is  $3/2$ , and so the allowed values of  $F$  for the  $^2S_{1/2}$  and  $^2P_{1/2}$  states of this isotope are  $3/2 - 1/2 = 1$  and  $3/2 + 1/2 = 2$ . The magnitude of the energy splitting due to the hyperfine interaction is on the order of  $m/M_N \approx 1/2000$  times the size of the splitting due to the spin-orbit interaction or roughly  $10^{-6}$  eV.

The final level of splitting occurs in the presence of a weak external magnetic field  $\mathbf{B}$ . Classically, the magnetic moment  $\mu$  of a particle of charge  $q$  and angular momentum  $\mathbf{L}$  is given by  $\mu = (q/2mc)\mathbf{L}$ . Extending this result quantum mechanically, one obtains

$$\mu_J = -g_J \frac{e}{2mc} \mathbf{J} \quad (6.)$$

for the magnetic moment  $\mu_J$  due to the total electronic angular momentum, and

$$\mu_F = -g_F \frac{e}{2mc} \mathbf{F} \quad (7.)$$

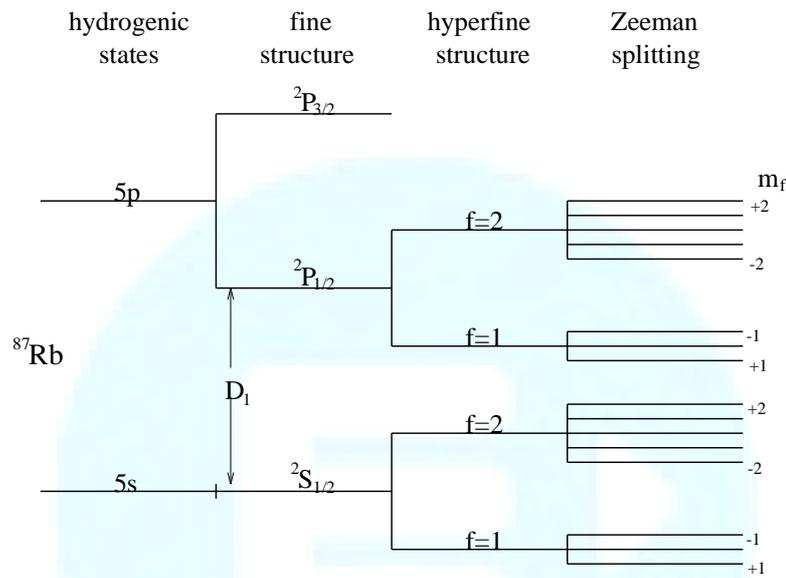


Figure 3: Level diagram of  $^{87}\text{Rb}$ .  $l = 3/2$ . The splittings are not to scale.

for the total magnetic moment  $\mu_F$  of the atom, where  $g_J$  and  $g_F$  are the Lande g-factors which arise from a quantum mechanical treatment of the addition of the angular momentum operators. The g-factors are

$$g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} \quad (9.)$$

and

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} \quad (9.)$$

The energy of the magnetic moment  $\mu_F$  in a field  $\mathbf{B}$  is given by  $E = -\mu \cdot \mathbf{B}$ ; this leads to the term  $H_B$  in the Hamiltonian for the Zeeman splitting in a magnetic field of magnitude  $B_z$  in the z-direction:

$$H_B = g_F \mu_B m_F B_z. \quad (10.)$$

Here,  $\mu_B = eh/2m_e c$  is the Bohr magneton and  $m_F \hbar$  is the eigenvalue of the operator  $F_z$  (i.e., the projection of  $\mathbf{F}$  onto the z-axis). From Eq. 10., we find that the energy difference between adjacent

Zeeman levels of the  $^2S_{1/2}$ ,  $F = 3$  state of  $^{85}\text{Rb}$  is 0.466 MHz/Gauss, and for the  $^2S_{1/2}$ ,  $F = 2$  state of  $^{87}\text{Rb}$ , 0.701 MHz/Gauss (verify this).

The linear relationship between energy levels and magnetic field only holds for small magnetic fields. When the Zeeman splitting grows relative to the hyperfine energy difference one has to take into account the quantum mixing of the states.

To do that we find the eigenvalues of the Hamiltonian

$$H = H_{hf} + H_B = A\mathbf{J} \cdot \mathbf{I} + g_J\mu_B\mathbf{J} \cdot \mathbf{B}$$

The result is known as the Breit-Rabi formula:

$$E(F = I \pm 1/2, m_F) = -\frac{E_{hf}}{2(2I + 1)} \pm \frac{1}{2} \sqrt{E_{hf}^2 + \frac{4m_F}{2I + 1} g_J\mu_B B E_{hf} + (g_J\mu_B B)^2}$$

where  $E_{hf} = A(I + 1/2)$  is the zero-magnetic field hyperfine splitting and we have ignored a small effect due to nuclear magnetic moment, see Ref. 2. The hyperfine energy splitting at zero magnetic field is  $E_{hf} = 3036$  MHz for  $^{85}\text{Rb}$  and  $E_{hf} = 6835$  MHz for  $^{87}\text{Rb}$ .

### Optical pumping

The transition rules for the 5s Rb electron are similar to those for the simplified atom discussed above. For concreteness, we focus on  $^{87}\text{Rb}$  though both isotopes are pumped in our apparatus. As always, both spin and orbital angular momentum must be conserved. When the Rb vapor is illuminated with circularly polarized  $D_1$  radiation of positive helicity, the condition that must be satisfied is  $\Delta m_F = +1$ . Because there are no  $m_F = +3$  magnetic sub-states in the  $^2P_{1/2}$  state, any electrons in the  $m_F = +2$  sub-state of the  $^2S_{1/2}$  level are trapped there. Meanwhile, the spontaneous and stimulated emission of the  $^2P_{1/2}$  states (decaying back down to the  $^2S_{1/2}$  level) need only obey the magnetic quantum number selection rule  $\Delta m_F = 0, \pm 1$ . Since collisions provide the only possible means for depopulation of the highest magnetic sublevel, most all of the atoms end up in the sub-state ( $m_F = +2$ ) of the  $^2S_{1/2}$  ground state. We say that this level has been pumped.

To detect optical pumping, you could, for instance, monitor the transparency of the sample by shutting off the light beam, allowing the atoms to “relax”, and then observing the transient in absorption when the beam is turned back on. A more convenient way of equalizing the populations of the Zeeman states is to resonantly relax them with a strong radio frequency (RF) field,  $\Delta E = \hbar\omega_0$ , where

$$\omega_0 = \frac{g_F e}{2mc} B_z.$$

You could imagine sweeping the frequency of the RF generator over some range around the expected resonance, while observing the transparency with a photodiode. This is very nearly the path we will take, except instead of varying the frequency, it is simpler and cleaner to vary the energy gap by sweeping the magnetic field slightly around a central value so that  $B_z = B_0 + B^0(t)$ .

### Outline of the experimental apparatus

A block diagram of the experimental set-up is shown in Figure 4.

**Rb Laser.** The light at the Rb  $D_1$  line is generated by a VCSEL (vertical cavity surface emitting diode laser) source. The laser wavelength depends on laser current and temperature and needs to be tuned to

the Rb resonance. Turn on the temperature controller and press “Output” to enable it. The temperature setting is 20 °C. The laser current is controlled by adjusting the voltage knob on the power supply, it should be around 18 V. The fine-control voltage knob can be used to fine-tune the wavelength of the laser.

*Sample Cell.* The sample cell contains in natural abundance rubidium and a nitrogen buffer gas. The purpose of the buffer gas is two-fold. First, it slows the diffusion of atoms, so they spend more time interacting with the laser beam. Second, nitrogen in particular can “quench” atoms in the excited state, so they decay to the ground state without emitting a photon. This is important because spontaneous emission photons are unpolarized. They can be absorbed by other atoms and lead to depolarization. A coil around the cell delivers the level-equalizing RF magnetic field and is connected to a RF generator. The sample cell is heated by air that has been passed over heater coils carrying an AC current. The raised temperature melts the metallic rubidium from the side of the bulb and increases its vapor pressure in the gas.

The air flow to the Rb cell is crucial for maintaining its temperature. Open the main (chrome-plated) air valve 2 turns. Open the small valve leading to the cell by one quarter of a turn. You should hear air flow noise. Turn on the Variac used to control the current in the heater and set it to 3 A. The temperature of

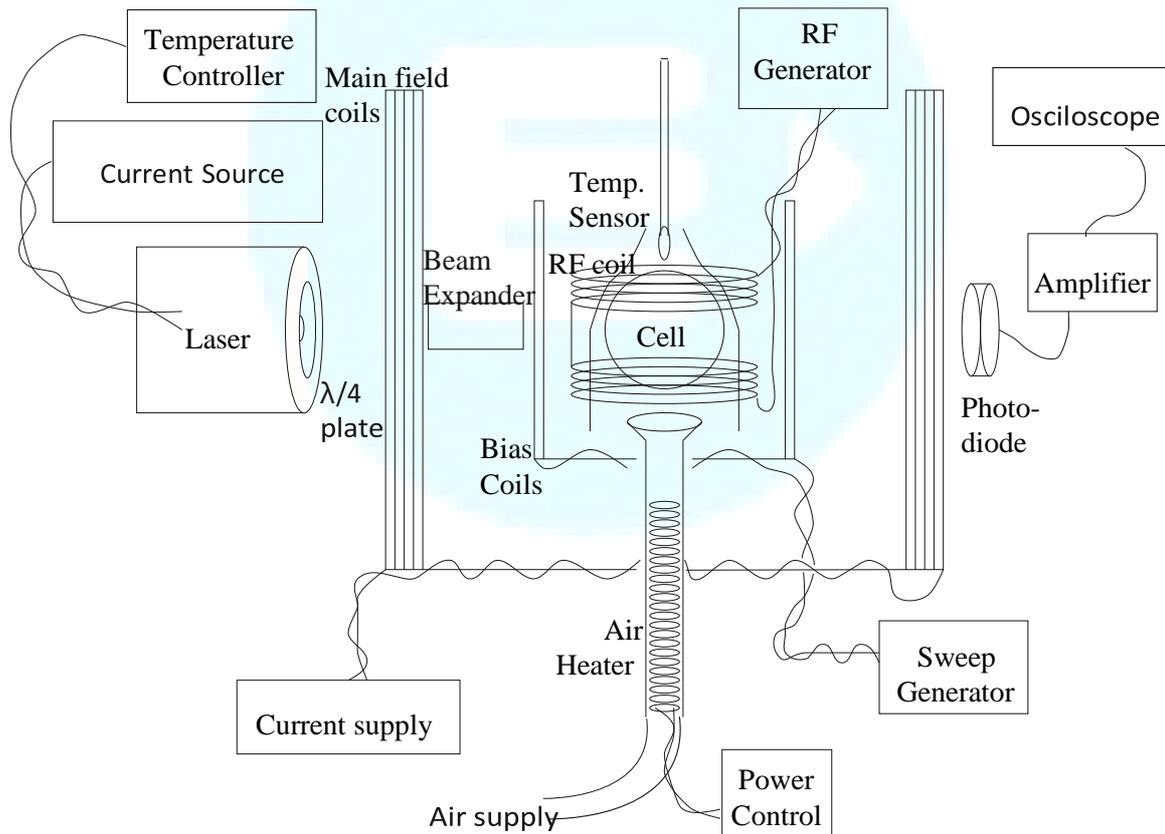


Figure 4: Block diagram of experiment. the cell is measured with a thermocouple and a “speaking” meter. The temperature should be somewhere between 60 °C and 75 °C, it takes about 10 minutes to reach equilibrium.

*Optics.* The light coming from the laser is naturally linearly polarized. A quarter-wave plate mounted directly on the laser converts the light to circular polarization (learn how it works). Light from the laser passes through a beam expander, the cell and is measured on the photodiode. The wavelength (789 nm) is just on the edge of being visible, it is hard to see. One can use a yellow “IR” card that fluoresces in the visible to trace the path of the beam. The components get bumped sometimes, so you should check that the laser beam passes cleanly through all elements and hits the center of the photodiode. The output current of the photodiode is converted to voltage using an operational amplifier. The circuit is powered by 9V batteries, don’t forget to turn it off after use.

*Magnetic fields.* There are two sets of Helmholtz coils. The larger main field coils have 225 turns each and are fed from a current-regulated DC supply and provide the nominal magnetic field,  $B_0$ . The smaller coils are driven by a sweep generator and provide a small triangular sweep  $B^0$ . Note that because of the inductive effects in the smaller coils, the voltage across the coils is not necessarily in phase with the electric current.

*Field Measurement.* In addition to measuring the frequencies of the resonance peaks, you will need to determine the magnetic field strength  $B_0$ . This can be done with the Gauss meter. The zero of the probe can be determined by inserting it into a small cylindrical magnetic shield. You can also check the magnetic field calibration by calculating the magnetic fields from the parameters of the coils and taking into account the Earth’s field.

### Operating the experiment

The goal of this experiment is to explore optical pumping. Using this technique, you should determine the separation between the Zeeman levels of the ground states of both rubidium isotopes and measure the Bohr magneton. You should also observe the non-linear Breit-Rabi splitting and study the dependence of the magnetic field transitions on the strength of the RF field.

1. Turn on the power supply and temperature controller for the laser, air supply and heater for the cell. Wait 10 min until the cell heats up above 60 °C.

2. Using “IR” card, verify that the laser beam passes cleanly through the cell and hits the photodiode. Turn on the photodiode amplifier (remember to turn it off later lest you drain the batteries) and measure DC output voltage. You should get a signal of 1-2 V due to laser light.

3. After the cell becomes hot, look at the DC photodiode signal as a function of the diode laser supply voltage. You should see an absorption dip when the laser wavelength is in resonance with the atomic transition. Note that the overall power of the laser increases linearly with current.

4. Turn on the rest of the electronics. Compute the combination of magnetic fields and RF frequency at which you expect to see a signal. A starting point might be  $B_0 = 3$  Gauss and a RF frequency of 1.4 MHz. Which isotope is this for? The small coil should sweep with a frequency between 5 Hz and 100 Hz. The digital RF generator is used to set the frequency and amplitude of the RF field. Start with the maximum amplitude both for the RF field and the sweep field to make it easy to find the resonance.

5. Turn on the Gaussmeter, zero the probe and verify that it gives reasonable readings by measurement the Earth’s magnetic field (away from the coils). Calibrate the magnetic field by making several measurements of the field in the center of the coils as a function of the current.

6. Observe the Zeeman resonances on the scope. This will require setting proper triggering of the scope relative to the small coil sweep.

7. When you obtain a signal, convince yourself that it is real by detuning the RF field and watching the signal go away, or change the magnetic field of the Helmholtz coils. You can also improve signal-to-noise ratio by using the averaging function of the scope.

### Exploration

Now you can study many aspects of the signal:

- Find two resonances corresponding to the two isotopes of Rb.
- How does the shape of the resonance depends on the magnitude of the main field? You can go up to fields of 10-20 G.
- How does the shape of the resonance depends on the direction of the field? (Remember to think about the vector addition of the main coil field, sweep field and Earth's field).
- How does the shape of the resonance depends on the amplitude of the RF field?
- How does the shape of the resonance depends on the frequency and the amplitude of the sweep field?

### Measurements

1. With a small RF amplitude and slow sweep rate, perform detailed measurements of the resonance frequencies as a function of the current in the main coil. You should observe a linear relationship but at higher field the resonance for each isotope will split into several in accordance with the Breit-Rabi equation. What is the effect of the transverse components of Earth's field at low magnetic field?

2. At high magnetic field measure the width of the resonance as a function of the RF amplitude. This is known as a power broadening of the magnetic resonance. At some point you will also see new, narrow peaks appear in the spectrum. Measure how their amplitude depends on the RF field. Convince yourself that they are due to two-photon RF transitions.

For extra credit: Try to see Rabi Oscillations, where the atoms are coherently cycled between two Zeeman states. To see them, hold the external magnetic field fixed exactly at the resonance and suddenly turn the RF on for a short interval of time. You can do that by programming the digital function generator or using an amplitude modulation input on the back.