# **Optical-Pumping Method**

# 1 Purpose

The object of this experiment is to measure the energy splitting of the hyperfine structure of the lowest energy state of rubidium which is subjected to a uniform magnetic field whose magnitude can be determined by other means. From the method called "optical pumping", you will deduce the frequency of the RF-transitions in this magnetic dipole interaction which will then allow you to determine the *g*-factor of the resultant magnetic moment for two isotopes of Rb in a series of measurements with progressively increasing sensitivity:

- A rough measurement performed by sweeping the uniform magnetic field and finding the frequency of an applied transverse RF field at which de-pumping occurs at a particular spot along the sweep.
- A second measurement performed by finding the transverse field frequency at which the amplitude of Rabi oscillations are maximal.
- A third measurement using the dependence of the Rabi oscillation frequency as a function of amplitude of the transverse RF field.

The results of these measurements will be used to also find the ratio of the g-factors for the two isotopes. You will note that this experiment requires the conservation of both energy (in the resonance phenomenon) as well as angular momentum (in the absorption of photon quanta) in order to proceed.

The Nobel Prize in Physics was awarded to Alfred Kastler for his pioneering work on this highly important experimental method. Kastler (with the help of Jean Brossel) was the first to propose a successful optical method to investigate RF resonances in atoms. He believed that selected magnetic sublevels could be excited from higher lying energy levels using polarized light having the correct resonance frequency. In 1952, Kastler performed an experiment that confirmed the practicality of his recommendation and the validity of his theory. Other important developments that followed *as a result* of Kastler's work include such things as lasers, sensitive magnetometers, and atomic clocks.

# 2 Theoretical background

## 2.1 Thermal population distribution

The atoms of some material at a specific temperature, T, are usually distributed amongst a number of different energy states. Under ordinary conditions, the higher the energy level, the smaller the number of atoms in each level. Thus, the lowest energy state is generally populated to the greatest amount. Such a distribution is referred to as a thermal distribution. The relative population of any two (negative) energy levels,  $E_1$  and  $E_2$  with  $E_1 < E_2$ , is given by the Boltzmann factor such that

$$\frac{N_2}{N_1} = e^{-(E_2 - E_1)/kT} \,. \tag{1}$$

This relation yields the fractional difference in population,  $\frac{\Delta N}{N} = 1 - \frac{N_2}{N_1}$  given approximately by

$$\frac{\Delta N}{N} \approx \frac{E_2 - E_1}{kT} \tag{2}$$

when the energy kT is much larger than the energy difference between levels. Of course, in the absence of an external field, all of the *magnetic* substates are equally populated, since they all have the same energy.

**Exercise 1** For fields on the order of a Gauss, and for temperatures typical of those used in this experiment, verify that the fractional excess population is less than 0.1 ppm in level-1. Thus, ground-state rubidium atoms indeed reside with equal populations throughout all of its magnetic substates.

#### 2.2 Example: A simple 1-electron system

Under the right conditions, it is possible to cause the atoms to have a different distribution. When a non-thermal distribution is achieved by using a source of light, the process is then called "optical pumping". In order to see the essence of this scheme, let us first describe a simple energy level system (with no hyperfine structure as described later in this write up) for a sample of gas atoms which has a  ${}^{2}S_{1/2}$  ground state and a  ${}^{2}P_{1/2}$  excited state. Assume in this case that the atoms in the sample are all found in the ground state with its magnetic sublevels equally populated. In a weak magnetic field, the energy level diagram will look something like that shown in Fig. 1.



Figure 1: Hypothetical energy level diagram showing each fine-structure term split into doublets by the magnetic interaction. The optical absorption of a photon polarized in the  $\sigma^+$  state, shown with state-A  $\rightarrow$  state-D, corresponds to +1 change in the magnetic quantum number ( $\Delta m_S = +1$ ). Note, however, that spontaneous emission has equal probability of ending with states A or B. Repeated absorption-emissions would eventually populate state B above the thermal-equilibrium value.

[Note: The theory section of the "Zeeman Effect" write-up reviews the rules for "Russell-Saunders coupling." This first-order energy coupling (or so-called spin-orbit interaction) yields the fine-

structure in the energy level diagram, i.e. the primary energy separation between the  ${}^{2}P_{1/2}$  and the  ${}^{2}S_{1/2}$  states when the magnetic interaction is absent. Also in the "Zeeman Effect" write-up is a good description of the semi-classical vector model for angular momentum addition.]

In the presence of a magnetic field, the degeneracy between the  $m_s$  sublevels is lifted. The sublevel energies acquire an additional term due to the spin's orientation relative to the direction of the magnetic field,  $\vec{H}_0$ :

$$E(\text{magnetic}) = g_s \mu_B H_0 m_s \tag{3}$$

where  $\mu_B = e\hbar/2m_e$  is the Bohr magneton and  $g_s$  is the electron's Lande g-factor whose tiny difference from exactly 2.0 (as predicted by Dirac) is due to fluctuations in the electromagnetic vacuum. The quantity,  $m_s$ , is the magnetic quantum number which can have a value of either  $+\frac{1}{2}$  or  $-\frac{1}{2}$ , depending on the spin's alignment with the direction of the magnetic field.

Now assume that some resonant optical radiation impinges on the atoms in the sample, and the energy of that radiation is absorbed as indicated by the upward pointing broken arrow. The selection rules for the absorption of electric dipole radiation into the excited state from the ground state are  $\Delta \ell = \pm 1$  and  $\Delta m_s = 0, \pm 1$ . The first selection rule is valid because the transition is  ${}^{2}S_{1/2} \rightarrow {}^{2}P_{1/2}$  with  $\Delta \ell = +1$ . As for the second rule, assume that the impinging radiation is circularly polarized. This rule indicates that  $\Delta m_s = +1$  or  $\Delta m_s = -1$ , but not both. Which one is valid depends on the the direction of the magnetic field relative to the direction of light propagation as well as the direction of circular polarization. If the angular momentum of the photon (according to the electric field's rotation by the right-hand rule) is along the direction of propagation of the light (i.e. the direction of the photon's momentum), the light is said to be right-circularly polarized and is given the notation  $\sigma^+$ . This polarization state produces  $\Delta m_s = +1$  transitions. Likewise, left-circularly polarized  $\sigma^-$  light will produce  $\Delta m_s = -1$  transitions.

Figure 1 illustrates the absorption of  $\sigma^+$  light which *only* allows a transition from state-A to state-D. Assuming no disorientation by collisions occurs during the lifetime of about  $10^{-8}$  seconds for this excited state, the atom will fluoresce from state-D to state-A and state-B with equal probability. Thus, some of those atoms leaving state-A do not return to state-A, but end up in state-B. Without any means of transferring atoms from state-B back to state-A, the ensemble of atoms in the sample will *all* eventually end up in the higher energy state-B, thus producing a non-thermal population distribution.

If one is observing the intensity of the light transmitted through the sample, one initially notes the existence of absorption by the reduction of the light intensity, but as the state-B becomes fully populated, the gas becomes transparent and the light intensity through the gas is at a maximum. In a real system, state-B may not become fully populated because the final distribution will depend on such parameters as the intensity of the pumping light (see Sec. 3.1), the relative transition probabilities of multiple allowed states (which might compete for sublevels), and the relaxation rates through various internal-gas or wall collisions.

#### 2.3 Hyperfine structure interaction

The main features of atomic spectra for atoms with one optically-active electron are associated with the interaction of the spin magnetic moment of that electron and its orbital magnetic moment. These features have become known as the "fine structure" of the atomic spectra because they showed up first with improved resolution of optical spectrometers. One can show that the interaction responsible for the fine-structure energy shift is given by

$$\Delta E(\mathrm{fs}) = a\mathbf{L} \cdot \mathbf{S} \tag{4}$$

where  $\mathbf{L}$  and  $\mathbf{S}$  are the orbital and spin angular momentum quanta, respectively, and the finestructure coupling constant a is given by

$$a \propto \frac{1}{r} \frac{dV(r)}{dr} \sim \mu_B^2 \left\langle \frac{1}{r^3} \right\rangle$$
 (5)

The function V(r) is the electrostatic potential of the nuclear charge as it interacts with the electrons orbiting the nucleus. The spatial average (denoted by the angle brackets) is taken using the electronic wave-function  $\psi_e$ .

As spectrometer resolution was further improved, a still finer structure was observed, now referred to as the "hyperfine structure" of the atomic spectra. When the nucleus is given the added property of quantized angular momentum,  $\mathbf{I}$ , we see that a nuclear magnetic moment,  $\vec{\boldsymbol{\mu}}_{I}$ , will exist. Its interaction with the magnetic field produced by the atomic electrons at the position of the nucleus is given by

$$\Delta E(\text{hfs}) = A\mathbf{I} \cdot \mathbf{J} \tag{6}$$

where **J** is the sum of **L** and **S** and the hyperfine-structure coupling constant A depends on the nuclear magneton but is otherwise similarly dependent on r as the fine-structure interaction above:

$$A \propto |\psi_e(0)|^2 \sim \mu_n \mu_B \left\langle \frac{1}{r^3} \right\rangle. \tag{7}$$

In this case, r represents the electron's radial distance from an assumed "point" dipole located at the center of the nucleus. The nuclear magneton  $\mu_n = \mu_B m_e/m_p$  is 1836 times smaller than the Bohr magneton. Thus,  $\Delta E(\text{hfs})$  is typically three orders of magnitude smaller than  $\Delta E(\text{fs})$ . In weak external magnetic fields, the strong coupling of angular momenta, **I** and **J**, yields another quantum mechanical angular momentum **F** which is the vector sum **I** + **J**. Thus, **F** is quantized with values ranging from I + J to |I - J|. It should be noted that transitions between levels of different F, which are referred to as the hyperfine transitions, will involve absorption or emission of magnetic dipole radiation (to change direction of a nuclear magnetic moment) with  $\Delta F = 0, \pm 1$ only. Likewise, transitions between different  $m_F$  levels with F remaining constant comprise the Zeeman transitions (which are also due to magnetic dipole interactions), but now with  $\Delta m_F = 0, \pm 1$ only.

#### 2.4 *g*-Factor calculations

Before proceeding to the rubidium experiment, let us first review the rules for computing the g-factors associated with a magnetic moment that depends on the coupling of the spin angular momentum  $\vec{S} = \mathbf{S}\hbar$  and the orbital angular momentum  $\vec{L} = \mathbf{L}\hbar$ . The associated magnetic moments are

$$\vec{\mu}_{S} = -g_{s} \cdot \frac{\mu_{B}}{\hbar} \vec{S}$$
(8)

$$\vec{\mu}_{L} = -g_{\ell} \cdot \frac{\mu_{B}}{\hbar} \vec{L}$$
(9)

where the quantum mechanical values for the angular momenta are

$$|\vec{S}| = \sqrt{S(S+1)}\hbar \tag{10}$$

$$|\vec{L}| = \sqrt{L(L+1)}\hbar.$$
(11)

The above magnetic moments add vectorally to yield magnetic moment,  $\vec{\mu}$ , which precesses around an axis parallel to the resultant angular momentum,  $\mathbf{J} = \mathbf{L} + \mathbf{S}$  with quantum-mechanical value  $\sqrt{J(J+1)}\hbar$ . However, the magnetic behavior of this system depends only on  $\vec{\mu}_J$ , the component of  $\vec{\mu}$  in the direction of  $\vec{J} = \mathbf{J}\hbar$  where

$$\vec{\boldsymbol{\mu}}_{J} = -g_{J} \cdot \frac{\mu_{B}}{\hbar} \vec{\boldsymbol{J}} \,. \tag{12}$$

Figure 2 illustrates the vector addition of **L** and **S** as well as the associated magnetic moments. The component,  $\mu_J$ , is given by the relation



Figure 2: Origin of magnetic moment  $\vec{\mu}_{J}$ . It should be noted that the direction of the angular momentum vectors is opposite to that of the corresponding magnetic moments.

$$\mu_J = \mu_L \cos \theta_{LJ} + \mu_S \cos \theta_{SJ} \tag{13}$$

where the law of cosines yields

$$L^{2} = S^{2} + J^{2} - 2SJ\cos\theta_{SJ}$$
(14)

$$S^{2} = L^{2} + J^{2} - 2LJ\cos\theta_{LJ}.$$
(15)

Upon substituting for the magnetic moments given in (8), (9) and (12), one obtains

$$g_J J = g_L L \cos \theta_{LJ} + g_S S \cos \theta_{SJ} \,. \tag{16}$$

Finally, upon substituting for the values of the cosines in (14) and (15) into (16), using the fact that  $g_{\ell} = 1$  and  $g_s \approx 2$ , and solving for the g-factor  $g_J$ , one obtains

$$g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} \,. \tag{17}$$

Following the same procedure for the hyperfine structure using

$$\vec{\mu}_{I} = -g_{I} \cdot \frac{\mu_{n}}{\hbar} \vec{I}$$
<sup>(18)</sup>

for the magnetic moment associated with nuclear spin angular momentum  $\vec{I} = \mathbf{I}\hbar$ , you should be able to show that the hyperfine g-factor  $g_F$  is given by

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} - g_I \left(\frac{\mu_n}{\mu_B}\right) \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)}$$
(19)

where the second term in the above is often ignored since  $\mu_n$  is three orders of magnitude smaller than  $\mu_B$ :

$$\frac{\mu_n}{\mu_B} = \frac{m_e}{m_p} \,. \tag{20}$$

#### 2.5 Rabi Oscillations

In this experiment we prepare a fully pumped state and then introduce an oscillatory field that drives transitions to neighboring states. Under the right conditions, the system will undergo *Rabi* Oscillations (named after I. I. Rabi, usually pronounced "robby"), an effect similar to what one sees in pulsed nuclear magnetic resonance, and important for quantum computing among other applications. Rabi oscillations are seen as a periodic variation in the intensity of the light, caused by the forced population and depopulation of the states that can absorb the polarized light.

Consider a spin-J system in a constant magnetic field  $\vec{H}_0 = H_0 \hat{z}$ , with magnetic moment  $\vec{\mu} = g_J \mu_B \vec{S}_J$ , where  $\vec{S}_J$  is the J-dimensional spin operator. The state splits into 2J+1 sub-states with Zeeman splitting  $\Delta E = g_J \mu_B H_0$ . For simplicity, we will consider just two states (e.g. states A and B of Fig. 1, corresponding to a spin 1/2, with  $\vec{S}_J = \vec{\sigma}/2$ , where  $\vec{\sigma}$  are the Pauli spin matrices.) The energies of each state are  $E_A = -\Delta E/2$  and  $E_B = +\Delta E/2$ .

Let the system begin in the fully pumped state, i.e.  $|\psi(0)\rangle = |A\rangle$ . On timescales short compared to the pumping time, in the field  $\vec{H}_0$  the system would remain in state  $|A\rangle$ . However, if at time t = 0, we turn on an oscillating transverse field  $\vec{H}_1 = H_1 \cos \omega t \hat{x}$ ,  $|A\rangle$  is no longer an energy eigenstate, and the system will evolve into some linear combination  $|\psi(t)\rangle = c_A(t)|A\rangle + c_B(t)|B\rangle$ . To compute  $|\psi(t)\rangle$ , we evolve the system with the Hamiltonian:  $|\psi(t)\rangle = e^{i\frac{\mathcal{H}}{\hbar}t}|\psi(0)\rangle$ , where  $\mathcal{H} = -\vec{\mu} \cdot (\vec{H}_0 + \vec{H}_1)$ . We find that the population of state 1 oscillates in time as

$$c_A^2(t) = 1 - \left(\frac{\omega_1}{\Omega}\right)^2 \sin^2\left(\frac{\Omega t}{2}\right),\tag{21}$$

where  $\hbar\omega_0 = g_J\mu_B H_0$ ,  $\hbar\omega_1 = g_J\mu_B H_1$ , and  $\Omega$  is the "Rabi frequency," given by

$$\Omega = \sqrt{\omega_1^2 + (\omega - \omega_0)^2} \tag{22}$$

The oscillation is maximal when the frequency of the transverse field matches the energy splitting  $\Delta E$ , which corresponds to the resonance condition  $\omega = \omega_0$ .

Note that on resonance, the Rabi frequency depends linearly on the strength of the transverse field  $H_1$ . If the  $H_1$  field strength is not spatially homogeneous across the system, the system will oscillate with a distribution of Rabi frequencies. At small t, the oscillations will be in phase, with apparent frequency  $\langle \Omega \rangle$  given approximately by the average field strength  $\langle H_1 \rangle$ . At longer t, the oscillations will begin to decohere, and the oscillation envelope will decay. The contribution to the decay constant from decoherence is given roughly by  $\langle \Omega \rangle / \sigma_{\Omega}^2$ , where the denominator quantifies the variance of the frequency distribution. For higher spin systems, the Hamiltonian  $\mathcal{H}$  drives transitions from the pumped state not just into the neighboring state, but also into all other spin states. Yet qualitatively the system still exhibits a pumped state population that oscillates at frequency  $\Omega$ . Semi-classically, one can imagine that in one Rabi cycle those atoms in the pumped state will mostly transition to the neighboring state, while in the next cycle half will return to the original state, and the other half will go to the next neighboring state, and so on. However, in our setup, pumping continues even while we are generating level mixing with  $H_1$ . In every time interval a fraction of those atoms not in the pumped state are re-pumped back into it, leading to further degradation of the Rabi oscillations. Ultimately a balance is achieved between coherent depopulation of the pumped state via  $H_1$  and incoherent repopulation of the pumped state via optical pumping, and the system levels off to a steady state. With a constant rate of pumping, the population of the pumped state at long times can be made higher or lower by decreasing or increasing  $H_1$ , respectively.

## 3 The rubidium experiment

For this experiment, a special apparatus has been constructed whose purpose is to be compact and self-contained, but its components can not be readily viewed. The apparatus is housed inside of a phenolic tube, which also gives it some thermal isolation from the environment. As shown in Fig. 3, the phenolic tube contains the Rb-spectral lamp, a lens, an interference filter, a circular polarizer, the Rb-absorption cell, and a photo-diode detector. The converging lens is needed in order to collect the diverging rays from the spectral lamp and produce a parallel beam of light that propagates along the axis of the tube, through the absorption cell, and onto the photo-diode detector. A mask in the form of a thin disk with a small hole is placed directly in front of the Rb lamp. It prevents light reflected by the concave mirror in back of the lamp from entering the absorption cell. Since most of this reflected light would not be parallel to the z-axis of the tube (and therefore not be parallel to the  $\vec{H}_0$  field), it would cause a broadening of the resonance which is observed when applying the perpendicular RF magnetic field,  $\vec{H}_1$ . Although the mask cuts down on the intensity of the transmitted light being detected, the linewidth of the resonance is narrowed considerably. In order to eliminate the  $D_2$  spectral component in this light beam, an interference filter which is strongly peaked at 794.8 nm (the  $D_1$  line) follows the lens. Next, a linear polarizer and quarter-wave plate are mounted in the light path, immediately before the absorption cell. This combination serves to circularly polarize the light from the Rb-lamp. The light coming out of the linear polarizer (which is linearly polarized) is set at a  $45^{\circ}$  relative to the slow and fast axes of the quarter-wave plate; then the relative phases of the fast and slow components of the light's electric vector change by 90° in order to produce circularly polarized light. Finally, a 5-cm diameter, 7.5cm long absorption cell follows, containing the tiny amount of purified rubidium metal. In order to increase the degree of polarization of the optical-pumped sample, 50 Torr of neon buffer gas is included inside the absorption cell. In addition, the cell walls are prepared with a thin coating of tetracontane which is a long-chain saturated hydrocarbon. Such films have been found to reduce the wall-relaxation rate by more than a factor of five.

In order to produce the time dependence in the polarization of the sample, a solenoid is wound around the phenolic tube such that the axis of the  $\vec{H}_0$ -modulation field is co-linear with the direction of the pumping light. In order to increase the essential uniformity of this field, the coils are made about twice the length of the absorption cell. It is crucial that the  $\vec{H}_0$ -modulation field be made parallel to the Earth field so that the two can be added like scalars. The other method of treating this problem (which we do not use) is to buck out the Earth field using special Helmholtz



Figure 3: Experimental arrangement of optical components in a phenolic tube that comprise the optical pumping system. The polarizer and quarter-wave plate are mounted together, but the polarizer must be closer to the Rb-vapor lamp.

coils, known for their high-degree of uniformity. To complete the configuration that yields the time dependence of the sample polarization, another set of modulation coils are wound outside of the  $\vec{H}_0$  solenoid (windings not shown in Fig. 3) in order to produce a perpendicular  $\vec{H}_1$  rf-field that will be used to disorient the polarization of the sample.

In this particular experiment, the rubidium metal, which is sealed into the glass cell, should be heated above the metal's 38.5°C melting point. At room temperature, the absorption cell should contain enough rubidium vapor in order to get a weak resonance signal. The signal will improve a lot after allowing the lamp to warm the cell: about an hour of warm-up is sufficient. The large aluminum housing that holds the cell also insulates it thermally. [If this is not the case, the cell can be heated gently by blowing warm air from a hair dryer through the aluminum tubes that shield the phenolic tube assembly. This should only be done with the guidance of one of the TA's in the lab.]

There are two common isotopes that are present in the resulting vapor, <sup>85</sup>Rb with nuclear spin I = 5/2 and <sup>87</sup>Rb with nuclear spin I = 3/2, whose relative abundances are 72% : 28% respectively. When this vapor cell is placed in a weak magnetic field, the Zeeman splitting of the hyperfine structure is created. A schematic energy-level diagram for the single optically-active electron in <sup>87</sup>Rb is shown in Fig. 4. Note the relative energy separations between the fine structure splitting,



Figure 4: A diagram of the energy levels of <sup>87</sup>Rb with I = 3/2.

the hyperfine-structure splitting, and the Zeeman splitting. These are clearly not shown to scale; if they were, they would then be 1,500,000,000:27,000:1 in relative proportion.

For <sup>85</sup>Rb, the nuclear spin of 5/2 generates hyperfine levels F = 2 and F = 3. Using the expected

magnetic interaction in the hyperfine Zeeman structure

$$\Delta E(\text{magnetic}) = h\nu_F = g_F \mu_B H_0 \Delta m_F , \qquad (23)$$

you should be able to show that the energy differences arising from the  $\Delta m_F = 1$  transitions using <sup>85</sup>Rb instead of <sup>87</sup>Rb are 2/3 as large.

#### 3.1 Time dependence of optical pumping

Atoms in the fully-pumped state tend to become disoriented mainly because of collisions with the glass walls. The fully-pumped state is called "fully oriented" because the *m* sublevel is maximized making the magnetic moment greatest along the direction of the magnetic field. The rate of disorientation is called the "relaxation rate." On the other hand, a disoriented atom becomes oriented through optical pumping, and this process leads to a competing "orientation rate." Both processes determine the overall rate at which the gas of atoms reaches its maximum orientation and the steady-state value of maximum orientation that can be sustained with the pumping light on. The time constants of the overall process can be studied by observing the oscilloscope trace, similar to that shown in Fig. 5, as the sample proceeds from the completely *disoriented* state to the maximally-pumped state. If one can control the intensity of the pumping light, it becomes possible



Figure 5: Trace of the transmitted light intensity versus time. The level marked  $I_{\infty}$  represents the fully-pumped state and the level marked  $I_0$  is the initial intensity after the disturbance to the fully-pumped state. The horizontal scale is 2.5 ms per major division.

to extract the disorientation rate alone from such traces, as will be evident from the discussion below. Unfortunately, for our old optical pumping system, the pumping light intensity is fixed.

For the sake of simplicity, the following discussion of the two rates assumes that there is only the <sup>87</sup>Rb isotope in the cell. First we must prepare the sample. Let D<sub>1</sub> radiation at 794.8 nm be incident on the cell, causing absorption from  ${}^{2}S_{1/2} \rightarrow {}^{2}P_{1/2}$ . If the light has  $\sigma^{+}$  polarization, then in the absence of collisions, all the atoms will end up in the F = 2,  $m_{F} = 2$ , sublevel, as described earlier. This represents the fully pumped state which can no longer absorb electric dipole radiation

at the D<sub>1</sub> wavelength. It is important that D<sub>2</sub> light must not be available, or else an escape channel is opened to the  ${}^{2}P_{3/2}$ , F = 3,  $m_{F} = 3$  excited state, and atoms in this state experience a higher relaxation rate by collisions with the background vapor.

Now, we apply an RF magnetic field  $\vec{H}_1$  to the sample at right angles to the weak dc  $\vec{H}_0$  field aligned along the axis of the phenolic tube shown in Fig. 3. Further assume that the frequency of the  $\vec{H}_1$  field is close, but slightly above  $\nu_F$  where  $h\nu_F$  represents the energy difference between adjacent magnetic sublevels in the  ${}^2S_{1/2}$ , F = 2 ground state. By slowly ramping up  $H_0$ , the  $\vec{H}_1$ field can become resonant to this energy spacing and thus induce rapid absorption or stimulated emission by induced magnetic dipole transitions. This process induces a rapid de-orientation of the atoms, much faster than through collisions, since it tends to equalize the population in the Zeeman sublevels and destroy the original magnetic orientation corresponding to the sample being in the fully-pumped ground state.

The trace was made in Fig. 5 using the same ramp to generate the time sweep for the oscilloscope's horizontal scale as used to sweep the  $H_0$  field. The slope of the initial sharp drop in intensity, from the  $I_{\infty}$  level to  $I_0$  (from left to right), depends on the degree of homogeneity of the  $H_0$  field across the sample cell. If the field were completely homogeneous, this edge would be exactly vertical. As it is, the atoms in the cell become disoriented at different locations throughout the sample volume, causing the initial non-vertical slope.

As soon as all parts of the sample have passed through resonance with the  $\vec{H}_1$  field, the pumping process becomes evident as the light transmitted through the sample begins to increase in intensity. In effect, we assume that the pumping process consists essentially of raising atoms indiscriminately from all the other ground-state sublevels to the  $m_F = 2$  ground sublevel. Let dn/dt be the rate of change of the  $m_F = 2$  ground-state population, n, and let dN/dt be the rate of change of population, N, for all other ground-state sublevels ( $m_F \neq 2$ ). Further, define  $W_d dt$  to be the probability for downward transitions from the  $m_F = 2$  level (mostly via collisions), and define  $W_u dt$  to be the probability for upward transitions to the  $m_F = 2$  level (via the pumping light). As for our initial conditions at t = 0 when  $I = I_0$  (the lowest intensity level of the transmitted light), let  $n = n_0$ (the initial population of the  $m_F = 2$  sublevel) and  $N = N_0$  (the initial population of all other ground-state sublevels). The rate equations for the time-dependent values of these populations becomes

$$\frac{dN}{dt} = nW_d - NW_u \tag{24}$$

$$\frac{dn}{dt} = NW_u - nW_d . (25)$$

The solutions to these two rate equations consistent with initial conditions are

$$N = N_0 - C(1 - e^{-t/\tau}) \tag{26}$$

$$n = n_0 + C(1 - e^{-t/\tau}) \tag{27}$$

where  $\tau \equiv (W_u + W_d)^{-1}$  and  $C = (N_0 W_u - n_0 W_d) / (W_u + W_d)$ .

Finally, if one assumes that the transmitted intensity is proportional to the number of excess atoms in the  $m_F = +2$  ground sublevel, according to  $I = \alpha(n - N)$ , it follows that

$$I = I_{\infty} - \alpha C e^{-t/\tau} \tag{28}$$

where  $\alpha C = I_{\infty} - I_0$  is the amount of light lost at full disorientation. The above equation is in qualitative agreement with observed results. At t = 0, when the sample is completely disoriented,

I is a minimum. Thereafter, as the pumping proceeds, I increases exponentially to the limit,  $I_{\infty}$ . The quantity  $\tau$  can thus be estimated from the slope and magnitude of the curve in Fig 5 at any point:

$$\frac{dI}{dt} = \frac{I_{\infty} - I}{\tau} \,. \tag{29}$$

Note also: if the light intensity is reduced,  $W_u$  must reduce (because of fewer photons available to make upward transitions), and thus C would approach zero. However, for pumping to occur, C must be positive which yields

$$W_u \ge \frac{n_0}{N_0} W_d \,. \tag{30}$$

Let  $(1/\tau)_{\min}$  be the rate measured at minimum pumping light. Since  $1/\tau \equiv W_u + W_d$ , it then follows that

$$\left(\frac{1}{\tau}\right)_{\min} \to \left(\frac{n_0}{N_0} + 1\right) W_d \tag{31}$$

where  $W_d^{-1}$  is a measure of the relaxation time by all causes from the fully pumped state.

For <sup>87</sup>Rb,  $n_0/N_0$  is equal to 1/7, assuming equal populations in all eight ground-state sublevels. Thus, in the limit as the pumping light intensity is reduced, the rate back to the pumped state becomes  $\tau_{\min}(87) = (7/8)W_d^{-1}$ . And for <sup>85</sup>Rb,  $n_0/N_0$  is equal to 1/11, so  $\tau_{\min}(85) = (11/12)W_d^{-1}$ . This suggests that the rate from a fully disoriented sample towards maximally pumped one should be slightly quicker for the <sup>87</sup>Rb atoms than for the <sup>85</sup>Rb atoms, assuming that the processes that cause relaxation do not depend on the isotope.

## 4 Procedure

In this experiment you will determine the g-factor for the ground state of the two isotopes of rubidium, <sup>85</sup>Rb and <sup>87</sup>Rb. You will investigate the time constant  $\tau$  for pumping the Rb atoms into the highest Zeeman (magnetic) sublevel of the ground state under two experimental conditions: 1. Equalizing the populations of the ground state sublevels by passing through the resonance condition; 2. completely inverting the ground state sublevels by reversing the magnetic field. You will also estimate the field inhomogeneity across the Rb cell, expressed as so many ppm/cm.

#### 4.1 Preliminary procedure

A diagram of the setup is shown in Fig. 6. Turn on all the electronics. The power supply for the Rb lamp should already be on; the resonance signal does not achieve full intensity until the lamp has been on for an hour or so. The power switch for the ADA400A preamp power supply is on the back of the black box. Other equipment settings follow.

#### Rb lamp power supply

Turn on the Power switch, and then after the current comes up turn on the Standby switch.

Current should read 22–25 mA.



Figure 6: Setup of the optical pumping apparatus for the  $H_0$  sweep experiments.

#### ADA400A preamp

Signal to +input, AC coupled, -input switch set to ground;

Gain = 100;

3 kHz upper bandwidth;

Offset OFF;

Output (from black box/power supply) goes to digital scope, Ch1.

**AFG3022B function generator Ch1** Ch1 provides the sweep of the  $H_0$  field. Select Ch1 by pressing the Ch1/Ch2 button to enable channel 1 (yellow trace). Run Mode: Continuous

Function: Ramp. Then under Ramp Parameter Menu select 100% Symmetry. This makes a positive ramp (shown in Fig. 7).

Frequency = 50.000 Hz;

Amplitude = 2.00 V(olts pk-pk)

To change the amplitude, for example, press the Amplitude/High button, enter the desired voltage with the number keys and then press the softkey for the appropriate unit (V). This is the general scheme for changing the value of any parameter on this generator: press the button for the desired parameter, enter the desired numerical value, then press the button for the desired units.

Offset = 0.00 V;

Ch1 output goes through  $50\Omega$  resistor to apparatus and to scope, Ch1. TTL out goes to AUX input on digital scope to provide external trigger. Press Output On to turn on generator signal.

#### AFG3022B function generator Ch2 Ch2 provides the $H_1$ resonance RF field.

Run Mode: Continuous

 $Function: \ Sine$ 

Frequency  $\approx 220 \text{ kHz}$ 

Amplitude = 2.0 V pk-pk;

Offset = 0.00 V;

Ch2 output goes to back side of Rb Lamp Supply labeled "H1 FN GEN." Press Output On to turn on generator signal.

**Tektronix digital scope** The settings below are approximate. Adjust as needed to make an easy-to-see picture.

Horizontal: 5–10 milliseconds (ms) per division

Vertical: Ch1, 1 V per division, Ch2, 20 mV per division.;

Trigger source: AUX, Use a TTL level: 1.4 V trigger level;

Cursors: OFF;

Averaging feature: OFF.

Before attempting to observe a signal, make sure that aluminum cylinder holding the apparatus is aligned parallel to the Earth field. A compass/tiltmeter is available in the lab to indicate the direction of the Earth field. To use the tiltmeter, first set it horizontal (like a compass) to find magnetic North, and align the long edge of wooden base of the aluminum tube with this direction. Then rotate the tiltmeter in its holder so that it will follow the vertical component of the Earth field. Align the tilt of the tube to be parallel to the tilt-meter needle.

The tiltmeter should not be left close to the apparatus, as the magnetic fields from the needles will distort the field inside the apparatus resulting in a distorted output signal.

Having set the equipment as indicated above, you should see a signal something like that in Fig. 7. If you do not see anything like the dip in the Ch1 signal, adjust the frequency of the RF channel (Ch2) between 200 and 280 kHz. (Press Frequency/Period and use the knob.) This channel provides the  $H_1$  (see Fig. 3) signal which disorients the polarization upon passing through the resonance condition. If you still don't see anything, make sure all the equipment is on, and check all of the above settings. If you still don't see a signal, consult with TA, Professor or lab manager.

To get a more stable pattern on the scope, you can display the average of a number of sweeps. To activate this feature, press the ACQUIRE button, set Average to be on and set the number of sweeps to be averaged with the knob at the upper left of the control panel. Averaging just 2 or 4 sweeps will significantly reduce the noise and stabilize the trace while still allowing for a fairly fast response time.

If you are using the Tektronix MSO2024 digital scope, you can save a screenshot to a USB memory stick. Insert the stick into the USB port at lower left. Then press Menu in the Save/Recall buttons and follow the instructions. (Or, you could take a picture with your phone or tablet.)

If the scope you are using is attached to a printer, push the HARDCOPY button on the scope and the printer underneath the scope will print the screen. To get a less cluttered printout, press the REMOVE MENU button to the lower right of the screen.



Figure 7: Initial signal from the optical pumping experiment. The resonance dip shows the effect of disorienting the polarization of the Rb atoms leading to an increase in the absorption of the light. As optical pumping proceeds, the cell becomes more transparent.

#### 4.2 Measurements to determine $g_F$

The first set of measurements will be to determine the resonant frequency,  $f_{res}$ , for the  $H_1$  signal. Once this frequency is determined and the static  $H_0$  field, i.e., the Earth field, is measured, one can derive  $g_F$  using Eq. (23). The simplest way to proceed would be to measure the frequency of  $H_1$ at the zero crossing of the  $H_0$  sweep field. But this method would provide only one measurement from which to derive  $g_F$ . A better method is to introduce a series of offsets in  $H_0$ , measure  $f_{res}$  at each offset, and then derive the frequency at zero offset from a least squares fit of the data. We shall follow this method to determine  $g_F$  for both isotopes of rubidium. We will also be able to determine the offset necessary to cancel the Earth field altogether, since from Eq. (23) one can see that  $f_{res} = 0$  when  $H_0 = 0$ .

After getting a signal like that in Fig. 7 on the scope, turn on the scope cursors (press CURSORS button) and select the vertical cursors. Move a cursor by rotating the multipurpose knob at the upper left of the control panel. Set the left cursor just on the vertical edge of the ramp waveform, as in Fig. 8. Make sure that the frequency of the  $H_0$  sweep is *exactly* 50 Hz. Set the right cursor at  $\Delta = 10.0$  ms (milliseconds). This is just 1/2 the period of the  $H_0$  sweep waveform and is therefore the zero crossing of this waveform when there is no offset.

Check that the OFFSET of the  $H_0$  sweep channel is set to 0. Adjust the frequency of the  $H_1$  generator until the leading edge of the dip is centered on the right cursor as in Fig. 8. Note the frequency of  $H_1$ ; it should be approximately 220–250 kHz. To see how reproducible this measurement is, you may want to move the dip away from the cursor by changing the frequency and then back to where the leading edge is again centered on the cursor. You can get some idea of the uncertainty by repeating this process several times. You should get at least three significant figures on your frequency measurements; for fine tuning, the frequency increment should be 0.1 kHz (or smaller). Check that the time interval between the cursors is still 10 milliseconds. It should remain at 10 milliseconds for this entire set of measurements.



Figure 8: Scope setup used to find  $H_0$ . Note that cursors are set to span the range from the beginning of the sweep to the exact midpoint (10 ms). The frequency of  $H_1$  is adjusted to position the dip at the midpoint cursor.

Now introduce an offset into  $H_0$  by setting the offset to 0.1 volt (With Ch1 active, press Offset/Low. You figure it out from here.) Adjust the frequency of  $H_1$  until the leading edge of the dip is again centered on the right cursor. Again note the frequency. Repeat this process for a series of offsets between -3.00 volts and +1.00 volts. When you analyze your data, perform a (linear) least squares fit for  $f_{res}$  vs.  $V_{offset}$  to obtain the value of  $f_{res}$  at zero offset. This will be the resonant frequency in the Earth field. You will measure this field using the Hall effect gaussmeter at the end of the experiment.

As you obtain the data, make a plot in order to obtain the offset at  $f_{res} = 0$ . This will be useful later when measuring the relaxation times, as it will give the offset necessary to null the Earth field.

Set the  $H_0$  offset to zero. Adjust the frequency of  $H_1$  to about 330 kHz. You should again see a dip similar to the one you observed at 220 kHz with no offset. The dip at 330 kHz is smaller as it is due to the <sup>87</sup>Rb isotope which has  $\approx 1/3$ rd the abundance of the <sup>85</sup>Rb isotope. You may want to rescale the vertical gain in order to see the resonance more clearly, and take advantage of the averaging feature of the digital scope to clean up the trace. Make sure the time interval between the cursors is still 10 milliseconds and the  $H_0$  frequency is still set at 50 Hz. Now repeat the process of introducing a series of offsets, centering the leading edge of the dip, and noting the frequency, just as for the <sup>85</sup>Rb isotope. Again, as part of your analysis, perform a fit for these data to determine the resonant frequency  $H_1$  in the Earth field and the offset necessary to null the Earth field.

**Exercise 2** It is interesting to see just how sensitive the signal is to any change in the magnetic field at the location of the Rb cell. A small magnet is available for this test. With a resonance dip like that in Fig. 7 displayed on the scope, start far away from the cylinder and slowly bring the magnet close to the aluminum cylinder as you observe the effect on the scope waveform. Then see what happens when you flip the magnet, and bring the opposite end to the cylinder. Can you explain what you see, both in the direction of the shift of the resonance and in how the shape changes?

**Exercise 3** Following the description in section 3.1, estimate the re-pumping time constant  $\tau$  for both Rb isotopes. Do they differ? By how much? Is the difference consistent with the expected behavior predicted in section 3.1?

#### 4.3 Another method of observing optical pumping

There is another way to observe optical pumping without using the perturbation of the  $H_1$  field. The idea is to redistribute the population of the magnetic sublevels of the ground state when they are in the fully pumped condition. Given the configuration of the experiment, changing anything about the light is not possible. But we do have other ways to control the applied field. Note that when taking measurements to determine  $g_F$ , the net magnetic field never changed polarity (how do you justify this claim?). But what if the magnetic field were to reverse polarity? Referring to Eq. (23), if  $H_0$  changes to  $-H_0$ , the energy changes sign. This implies that atoms in the  $m_F = +1$  and +2 states are now in the  $m_F = -1$  and -2 states, respectively. But this is just the goal we were seeking—to change the population distribution of the magnetic sublevels. After flipping the field from  $H_0$  to  $-H_0$ , transitions with  $\Delta m_F = +1$  are again possible, and pumping action can be observed. Question: are both isotopes participating in the pumping action under these conditions?

Set  $H_1 = 0$  by turning off its channel. Select the square wave waveform on the  $H_0$  channel, set the amplitude to 0.6 volts, and the frequency to 7 Hz (this is a frequency that will allow for averaging out the 60 Hz noise; to see that this is important, try a frequency of 20 Hz). From your graph of  $V_{offset}$  vs.  $f_{res}$  which you made during the procedure to determine  $g_F$ , determine the offset corresponding to  $f_{res} = 0$ ; this gives the condition necessary to null the Earth field. Set the offset on the  $H_0$  channel to this value. What do you expect to see when the square wave transitions from one level to the other? Change the offset in the positive direction by several volts. What happens to the signal? Change the offset in the negative direction. What do you see now? Finally, adjust the offset slightly to find the value where the signals on both edges of the square wave are most nearly equal. (Note: you may find that as you move the offset up and down that you hit a point where the generator display shows an error; this means you have reached the limit of the generator output. The generator can produce a maximum voltage of  $\pm 10$  volts into an open circuit, or  $\pm 5$ volts into  $50\Omega$ . You may be able to move the offset to greater values by reducing the amplitude of the square wave, and vice versa.)

**Exercise 4** Estimate the repumping time constant of this signal. How does it compare to the time constants you observed using the  $H_1$  resonance signal in Exercise 3?

#### 4.4 Rabi Oscillations

To observe Rabi Oscillations with this apparatus, we must prepare a constant background magnetic field, for which we will use  $H_{\text{Earth}}$ , and then we must pulse the system with the transverse field  $H_1$ . When the transverse field turns on, the system will undergo Rabi cycles, and the oscillating population in the pumped state (described by Eq. 21) will result in variation of the transparency of the Rb cell, giving an oscillating signal from the photodiode.

To set this up, you should reconfigure the connections to the function generator in order to make an RF burst that has its start coincident with the TTL output trigger signal. Ch1 on the AFG3022B is the only channel that can be used this way. To make the changes:

1. Turn off Ch1 and Ch2.

- 2. Disconnect Ch1 from the 50 $\Omega$  resistor that feeds the  $H_0$  coil. But, leave the connection to the scope in place.
- 3. Connect the output of Ch1 to the  $H_1$  input on the Rb lamp supply.

When you are done, you connections should look like those shown in Fig. 9.



Figure 9: Setup for observing Rabi oscillations. The only change is in the connections from the function generator. Use Ch1 to generate the RF burst for  $H_1$ .

Now, you need to configure an RF burst signal on Ch1. To do this:

- 1. Set Ch1 to be active.
- 2. Select Function: Sine, and set the frequency to be the resonant frequency for the <sup>85</sup>Rb isotope (the stronger one). Set the amplitude for 5 V peak-to-peak.
- 3. Select Run Mode: Burst.
- 4. In the menu that appears, select N-cycles and set the number to 1000.
- 5. At the bottom of the menu where it says "-more-", select the next set of parameters and set the Trigger interval to 200ms. This will give 5 burst per second.

When you turn on Ch1 you should see something similar to the fields are set up correctly, you should see a small "wiggle" at the bottom of your depumping dip, like that shown in Fig 10. Tune the frequency so that the feature is as strong as possible. Use this frequency to derive another value for  $g_F$ . Compare to your value from Section 4.2. You might attempt to optimize the angle of the apparatus relative to the Earth magnetic field, using the Rabi oscillation features as a guide.

Next, develop a procedure for measuring the frequency of your Rabi oscillations using the scope. You should explore the relationship between  $\Omega$  and both the  $H_1$  frequency and  $H_1$  amplitude. What is predicted by Eq. (22)?



Figure 10: Scope output in which Rabi Oscillations are observed (in Channel 2). Channel 1 shows the shape of the  $H_1$  RF pulse required. to generate the oscillations.

You can use a careful measurements of  $\Omega$  as a function of the peak-to-peak voltage  $V_{pp}$  of the  $H_1$  pulse, if you know that the  $H_1$  frequency is a little below the true resonance  $\omega_0$ , to get an especially precise measurement of  $\omega_0$ . Find what you think is the best value of  $\omega = \omega_0$ , and the deliberately de-tune it low by a couple of kHz. Then measure  $\Omega^2$  vs  $V_{pp}^2$ , and use a linear fit to a plot of this data find the de-tuning of your transverse field frequency from resonance. What quantity from the plot gives you the detuning from true resonance? From this you should be able to derive a third, most precise and accurate value for the value of  $\omega_0$  and therefore  $g_F$ .

Repeat this measurement for  ${}^{87}$ Rb and compute it's  $g_F$ , as well as the ratio of the two g-factors.

#### 4.5 Measuring the Earth field and the $H_1$ coil parameters

To calculate  $g_F$  from your measurements, you will need to know the Earth field. We will measure this using a gaussmeter, which requires the optical pumping assembly to be powered down. For this reason, it is best to perform this measurement after you have completed all measurements on the rubidium sample.

Turn off all the electronics except the gaussmeter. After the electronics are off, consult the TA for help in removing the optical pumping assembly from the aluminum cylinder. Carefully set the assembly aside in a secure place. Turn on the Bell 9500 gaussmeter and allow it to warm up for a few minutes. Check that the range is on "autorange". It should autorange into the 3 gauss range.

Take the gaussmeter probe and insert it into the ZERO GAUSS CHAMBER. Perform the probe zero procedure as follows: find this procedure by pressing the PROGRAM / ENTER button and then using the arrow buttons. Select CHANGE and then press the ENTER button. After the zeroing procedure is complete (takes about 15 seconds), select RUN on the menu and press the ENTER button. Insert the probe into the piece of white foam. Slide this piece of foam into the aluminum housing, and record the reading of the Earth field. Note how the reading is affected by changes in the cylinder's position. The probe is a Hall effect device which is designed to give a maximum amplitude reading when the field is oriented along the probe axis. (Other probes are designed for transverse measurements, e.g., the one used in the NMR experiment.) After recording the field, remove the foam from the housing and remove the probe from the foam. Turn off the

gaussmeter.

#### 4.6 Extension Ideas

- Compute the magnetic field strength of the  $H_1$  field as a function of position across the Rb cell, and integrate over the volume of the cell to obtain the average field strength in the cell. To do this, you will need to measure the resistance and geometry of the  $H_1$  coil. The resistance can be measured by simply using a DMM on the BNC that connects the pulser to the coil. Measure the geometry of the coil by gently inspecting the insert with a ruler or other measurement device you should be able to see the  $H_1$  wires running along the sides of the cylinder. Use this to compute the expected Rabi oscillation frequency as a function of  $H_1$  pulse voltage. Compare to your result from Section 4.4.
- Write the rate equation for coherent population transfer between the m = 2 and m = 1 states, combined with decoherent repopulation of the m = 2 state due to repumping. Solve the equation in the steady state to obtain a prediction for how the equilibrium m = 2 population (the level to which the Rabi oscillations dampen out) varies as a function of  $H_1$ . Measure the dependence with the apparatus and compare.
- Work out the Rabi Oscillation theory for a 5-level system. Compare with your data.
- Investigate how the depumping dip shape changes as you vary the sign and steepness of the slope of the  $H_0$  ramp.
- Set up the pulser to generate a pulse sequence of specified widths, chosen to be  $\pi$  or  $\pi/2$  pulses as appropriate, similar to the procedure for the pulsed NMR experiment. Attempt to measure something akin to the pulsed NMR spin-lattice and spin-spin relaxation times. Interpret your results.
- Develop the theory and a procedure to use Rabi oscillations to determine an optimal orientation of the apparatus relative to Earth magnetic field.
- Develop a physical model for how the transparency of the system changes with voltage in the  $H_0$  solenoid when  $\vec{H}_{sol} \approx -\vec{H}_{earth}$ . Compare to your data.

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