

Optical Pumping

Optical pumping uses light to affect the internal degrees of freedom of a sample of atoms in a vapor. These internal degrees of freedom are angular momentum states of the atomic ground state, which are populated when light is absorbed and emitted. This experiment reveals many phenomena of quantum mechanics including angular momentum selection rules, the density matrix, and resonance.

Optical pumping is important to a wide variety of applications from the laser, to precise measurements of atomic hyperfine level splittings and atomic clocks, sensitive measurements of weak magnetic fields, and recently to enhance NMR and MRI. This experiment provides an array of measurement opportunities including measurement of optical depth, atomic electron polarization and relaxation rates, radio frequency electron paramagnetic resonance (EPR), fine structure splittings, nuclear moments, the isotopic composition of rubidium, and the investigation of resonance line shapes and their relation to exponential decay.

A solid state laser diode is used in the experiment. A similar laser is used for the Doppler Free Spectroscopy experiment. The laser is described in a separate Chapter.

Background

This section provides an essential theoretical treatment of the basic physics of optical pumping and magnetic resonance. We first explain how selected angular momentum states can be populated and probed with light. Next the rate equations for magnetic resonance are presented. Finally we discuss resonance. It is important to review the theory of the one electron hydrogen-like atoms, the atomic shell model, and the quantum mechanical theory of

angular momentum (including Clebsch-Gordon coefficients) in your Quantum Mechanics text.

How atoms absorb light

Light consists of photons which carry energy ($E = \hbar\omega$), momentum ($p = \hbar k = E/c$), and angular momentum ($j = 1$). Quantum mechanics provides for the additional degrees of freedom, labeled by m_j , for a system with angular momentum. For a photon, $m_j = \pm 1$. The $m_j = 0$ state cannot exist for a massless particle. The quantum mechanical description of a photon state is therefore made up of the product of momentum and angular momentum wave functions. The angular momentum eigenstates are called σ_+ ($m_j = +1$) and σ_- ($m_j = -1$). Linearly polarized light, labeled π , is a combination of the two circular polarizations given by:

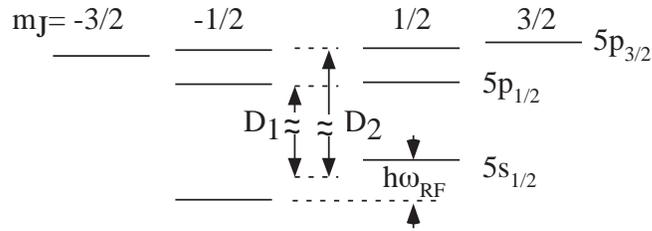
$$\pi = \frac{1}{\sqrt{2}}[\sigma_+ \pm \sigma_-]$$

Atoms are made up of electrons and the nucleus bound by coulomb and magnetic forces. The total angular momentum of an atom is made up of the electron spin and orbital angular momentum and the nuclear spin. We will consider alkali-metal atoms (Li, Na, K, Rb, or Cs) which have a single electron bound to a core consisting of a nucleus with charge Z and closed shells of $Z - 1$ electrons. For example rubidium has $Z = 37$ with shells closed shells $nl = 1s, 2s, 2p, 3s, 3p, 3d, 4s, 4p$. The spin orbit interaction pushes the $5s$ shell lower than the $4d$ and $4f$ shells, so that the rubidium atoms behaves like a single electron in the $5s$ ground state. The lowest excited states are the $l = 1$ $5p$ states. The quantum theory of angular momentum provides that these $5p$ states have total electron angular momentum $\vec{J} = \vec{L} + \vec{S}$ where \vec{L} is the orbital angular momentum and \vec{S} is the electron spin angular momentum. For the electron $s = 1/2$, and therefore $j = l \pm 1/2$. For the $l = 0$ $5s$ state, $j = 1/2$. For the $l = 1$ $5p$ state, $j = 1/2$ and $j = 3/2$. These two $5p$ states are separated in energy by the spin-orbit interaction $\vec{L} \cdot \vec{S}$.

Many nuclear isotopes have total angular momentum as well. Generally labeled by \vec{K} , it is the combination of the orbital spin angular momentum of the neutrons and protons. The nuclear shell model successfully accommodates the systematics of nuclear \vec{K} , which combines with the total electron angular momentum to form the total angular momentum of the atom $\vec{F} = \vec{J} + \vec{K}$. The naturally occurring isotopes of rubidium are ^{85}Rb with

$K = 5/2$ and ^{87}Rb with $K = 3/2$. The $j = 1/2$ rubidium ground states for the two isotopes are split by the hyperfine interaction into doublets with $F = K \pm 1/2$. The hyperfine interaction is the magnetic interaction of the nuclear magnetic moment with the electron magnetic moment. The hyperfine splitting, ΔW , is much smaller than the spin-orbit fine structure splitting and is expressed as a frequency, typically a few GHz.

Figure 1. The levels of a rubidium atom with no nuclear spin. The total angular momentum is $\vec{J} = \vec{L} + \vec{S}$. The $5s_{1/2}$ ground state and the lowest excited states are shown. The magnetic sublevels are split by $E = \hbar\omega_{RF}$ due to the Zeeman interaction discussed in the next section.



The relevant atomic structure of rubidium is summarized in figures 1 and 2. In figure 1 a hypothetical isotope with no nuclear spin is shown. The atomic levels are labeled in standard spectroscopic notation as nl_j . The rubidium ground state is $n = 5$, $l = 0$ (s -state), and $j = 1/2$, *i.e.* $5s_{1/2}$. The magnetic sub-levels are also illustrated in figure 1. There are $2j + 1$ magnetic sublevels with magnetic quantum number $m + j = -j, -j + 1, \dots, j$. For $j = 1/2$, there are two states with $m_j = -1/2, +1/2$. For $j = 3/2$ there are four states with $m_j = -3/2, -1/2, 1/2, 3/2$. In figure 2, all the levels for the two naturally occurring isotopes ^{85}Rb and ^{87}Rb are shown. Each magnetic level is labeled by quantum numbers n, l, j, F , and m , where $\vec{F} = \vec{J} + \vec{K}$.

When atoms absorb light, they must absorb both energy and angular momentum in the transition from the ground state to an excited state. The most basic, leading order process is called the electric dipole, or E1, transition and has the selection rules for the atom $\Delta l = \pm 1$, $\Delta F = \pm 1$ and $\Delta m = 0, \pm 1$, depending on the polarization of the light. For circularly polarized photon absorption $\Delta m = m_j$. For linearly polarized photons $\Delta m = 0$. These restrictions of $\Delta l, \Delta F$, *etc.* are called selection rules. Emission of light by F1 radiation has the same selection rules. Also if $\Delta m = \pm 1$, the photon emitted is circularly polarized, and if $\Delta m = \pm 0$, the emitted photon is linearly polarized, that is a superposition of σ_+ and σ_- .

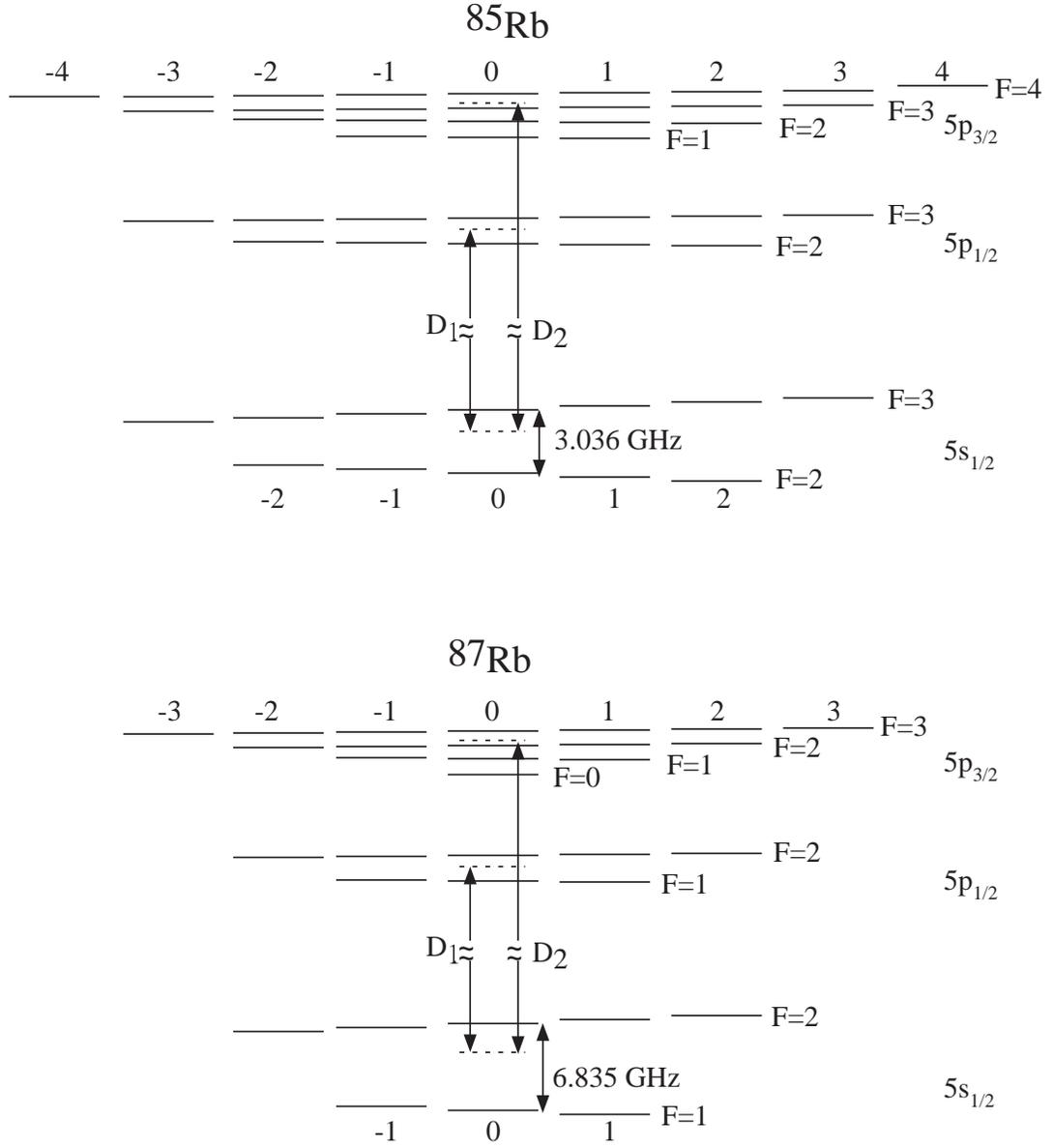


Figure 2. The ground state $5s_{1/2}$ and lowest excited state $5p_{1/2}$ and $5p_{3/2}$ energy levels of ^{85}Rb and ^{87}Rb . The magnetic sublevels are separated by the Zeeman interaction discussed in the next section.

The absorption of circularly polarized, σ_+ photons incident on ^{87}Rb atoms is illustrated by the solid lines in figure 3. The incident light is assumed to be tuned to the energy difference between the $F = 2$ ground state and the

$F' = 2$ $5p_{1/2}$ excited state. The solid lines show the allowed transitions for absorption of σ_+ light. The selection rule for photons with $m_j = +1$ is $\Delta m = +1$ as shown. For emission of light from the populated excited states, emission of σ_{\pm} and π photons may occur with relative probabilities given by the Clebsch–Gordon coefficients for the combination shown. These relative probabilities are also shown in figure 3. The dashed lines show the possible decays by E1 photon emission from each state. You can see that the incident σ_+ light causes transitions out of every state except for $m = +F$, and the probability for an initial state to be repopulated by decay from an excited state is less than unity. The result is that all but one state is depopulated so that after absorption of several photons, nearly all the population is in the $m = +F$ state. This is called depopulation pumping, and once the states with $m < +F$ are depopulated, the atoms do not absorb σ_+ photons. A sample of atoms that do not absorb photons is called transparent.

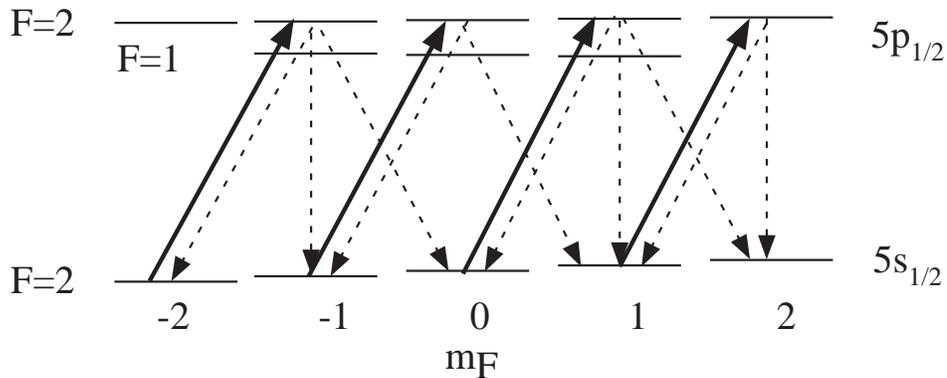


Figure 3. Illustration of optical pumping for D1 transitions from the ground state $F = 2$ level to the $5p_{1/2}$ states in ^{87}Rb . The solid lines show absorption of σ_+ photons and the dashed lines show decays from each of the $5p_{1/2}$ magnetic sublevels. All ground state sublevels are depopulated by absorption, except for the $m = +2$ level where the population builds up.

Rate Equations

We will derive the rate equations for optical pumping of a sample of atoms using the model of an atom with no nuclear spin with energy levels illustrated in figure 1. A sample of atoms with concentration $[N]$ is characterized by the concentrations of atoms in each magnetic sub level of the ground state $[N_+]$

and $[N_-]$, where

$$[N_+] = \rho_{++}[N] \quad \text{and} \quad [N_-] = \rho_{--}[N]$$

The probabilities ρ_{++} and ρ_{--} , where $\rho_{++} + \rho_{--} = 1$ uniquely describe the concentrations in the two levels of the ground state. These are called the diagonal elements of the density matrix, a $J \times J$ matrix that provides a mathematical description of the probabilities of finding each state or mixtures of states in an ensemble. Since $\rho_{++} + \rho_{--} = 1$ provides a restriction, a single quantity, usually the polarization $P = \rho_{++} - \rho_{--}$, describes the concentrations. More generally, the polarization is

$$P = \sum_{m_j=-J}^J m_j \rho_{m_j m_j}$$

where

$$\rho_{++} = \frac{1+P}{2} \quad \rho_{--} = \frac{1-P}{2}.$$

When σ_+ D1 light is incident on the sample of atoms, atoms with $m_j = -1/2$ absorb the light at a rate R , which depends on the intensity and frequency distribution (spectrum) of the light. Each atom that absorbs a photon of incident light is excited to a $5p_{1/2}$ state with $m_j = +1/2$, but the excited state lifetime is 30 ns or less, so the atoms quickly decay back to one of the ground state levels with probabilities α and $(1 - \alpha)$ for $m_j = +1/2$ and $m_j = -1/2$ respectively. For decay of $j = 1/2$ atoms in a vacuum, $\alpha = 1/3$ is related to the Clebsh-Gordon coefficient for the $j = 1/2$ atom and the $J = 1$ photon. Allowing for transitions between the ground state levels at a rate Γ rate equations for ρ_{++} and ρ_{--} are

$$\begin{aligned} \frac{d\rho_{++}}{dt} &= \alpha R \rho_{--} - \Gamma \rho_{++} \\ \frac{d\rho_{--}}{dt} &= -\alpha R \rho_{++} - \Gamma \rho_{--} \end{aligned}$$

From this, you can see that

$$\frac{d[\rho_{++} + \rho_{--}]}{dt} = 0 \quad \frac{dP}{dt} = \frac{d[\rho_{++} - \rho_{--}]}{dt} = \alpha R - \Gamma P$$

The time dependence of the polartzation is exponential with a time constant $(\Gamma + \alpha R)^{-1}$.

$$P(t) = \frac{\alpha R}{\alpha R + \Gamma} [1 - e^{-(\Gamma + \alpha R)t}]$$

You will measure this exponential time dependence.

Magnetic Resonance

Many mechanisms cause transitions between the ground state sub levels with $m_j = +1/2$ and $m_j = -1/2$. All of these mechanisms involve interaction of the atom's magnetic moment with a magnetic field, either applied or intrinsic. Of course the measurement of magnetic resonance requires applied magnetic fields, but there are many examples of intrinsic, unavoidable fields as well. For example, the rubidium vapor is contained in a glass cell, and the glass itself has a small component of paramagnetic iron atoms. When a rubidium atom moves near the wall, the rapidly varying field in the atom's rest frame can cause transitions between $m_j = +1/2$ and $m_j = -1/2$. This is called "wall relaxation." Other mechanisms involve a pair of rubidium atoms moving past each other. The magnetic dipole moment of one atoms causes a rapidly changing magnetic field in the rest frame of the other leading to spin flips and the phenomena of spin exchange. Spin exchange is an important effect because it is very rapid and affects the electron spins of atoms that may be in different hyperfine levels or even different isotopes.

Magnetic resonance involves the coupling of energy from the applied magnetic field to the atomic spin system, which has a natural frequency due to the Zeeman splitting of spin up and spin down atoms in the presence of an applied, static magnetic field \vec{B}_0 . The energy levels for an atom with $J = 1/2$ and two hyperfine levels $F = I \pm 1/2$ are given by the Breit–Rabi formula

$$E(F, m) = \frac{\Delta W}{2(2I + 1)} + m \frac{\mu_I}{I} B_0 \pm \frac{\Delta W}{2} \sqrt{1 + \frac{4m}{2I + 1} x + x^2} \quad x = \frac{(2\mu_0 - \mu_I/I)}{\Delta W} B_0$$

where ΔW is the hyperfine splitting of the two levels $F = J + 1/2$ and $F = J - 1/2$, and μ_0 is the Bohr magneton. The magnetic resonance transitions correspond to $\Delta F = 0$ and $\Delta m = \pm 1$. For B_0 of a few Gauss, $x \ll 1$, and the Breit Rabi formula can be used to show, for example that the corresponding frequency is

$$h\nu \approx \left[\frac{\mu_I}{I} \pm \left(\frac{1}{2} + \frac{\mu_J - \mu_I}{2I + 1} \right) \right] B_0 = h\gamma_A B_0$$

where γ_A is a gyromagnetic ratio for each isotope ^{85}Rb with $I_{85} = 5/2$ and ^{87}Rb with $I_{87} = 3/2$.

If an oscillating RF magnetic field (with frequency typically in the MHz range) is applied, with $\omega_{RF} \approx \omega_0$ the resulting transitions change Γ so that

$$\Gamma = \Gamma_0 + \Gamma_{RF}$$

where Γ_0 is the intrinsic combination of wall relaxation and other mechanisms, but does not include spin exchange, which conserves P , the rubidium polarization. The rate of RF induced transitions, Γ_{RF} depends on the frequency and magnitude of the applied RF magnetic field. Any change of Γ leads to a change of polarization with exponential time dependence. For example, switch the RF on, and the polarization decreases from P_0 (corresponding to Γ_0) to P (corresponding to $\Gamma = \Gamma_0 + \Gamma_{RF}$).

The phenomenon of magnetic resonance is observed by changing the frequency and magnitude of the applied RF magnetic field. The mathematics is identical to that of a driven LRC oscillator, where the damping rate constant is $\Gamma = R/2L$. The result is a frequency response that is given by the Lorentzian line shape (for a review, refer to the time dependent perturbation theory section your quantum mechanics text). When the RF is turned on, the frequency dependence is

$$P_0 - P = \Delta P \frac{\Gamma^2/4}{(\omega_{RF} - \omega_0)^2 + \Gamma^2/4}$$

One objective of this lab is measurement of this Lorentzian line shape.

The apparatus

The optical pumping apparatus is shown in figure 4.

The Light Source is either an electrodeless discharge lamp or a laser diode set up in the external cavity mode. The discharge lamp consists of a small spherical glass bulb containing rubidium and a vacuum tube powered high frequency RLC oscillator. The oscillator's inductor is a coil with a few turns wrapped around the rubidium bulb. The oscillating electromagnetic field produced in the bulb ionizes and accelerates the rubidium atoms producing a glow discharge and many spectral lines corresponding to transitions among the excited and ground states of the atoms of both isotopes. The D1 and D2 lines are by far the strongest in the discharge (the D2 is 2 times more intense), but only the D1 line is used. For this reason a narrow bandwidth

interference filter is used to transmit only the D1 line. Only a few μ watts of D1 light is provided by the lamp. The diode laser, in contrast produces a few to many milliwatts. The diode laser is described in a separate section. The light from the lamp is emitted in all directions, and a reflector helps direct the light through the rubidium bulb. A lens is used to collect the light into the photodetector. The laser beam is collimated with the aid of a small lens right next to the laser and no additional lenses are required. The “beam” from the lamp is practically invisible. The laser beam can be seen in a darkened room and with the aid of a fluorescence card or an infrared viewer.

Circular polarization is accomplished by passing linear polarized light through a quarter wave plate oriented at 45° with respect to the linear polarization. Light emitted by the lamp is unpolarized and a linear polarizer must be used. Light emitted by a laser is generally polarized, but it may be necessary or useful to use a linear polarizer with the laser as well.

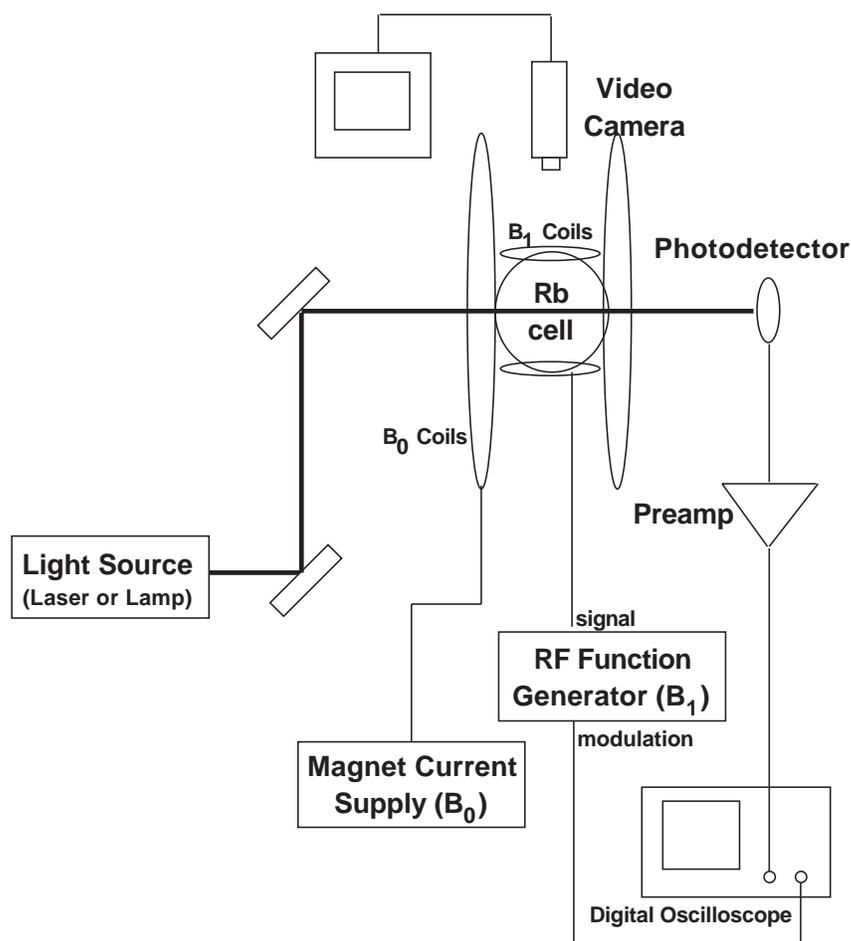
Two sets of magnetic fields are provided by symmetric pairs of coils. Each set is assembled in the Helmholtz configuration, which is chosen because it provides a more uniform magnetic field near the center of the pair. The Helmholtz configuration uses the spacing between coils to cancel the second derivative, with respect to the coil axis, of the magnetic field (*e.g.* $d^2B_0/dt^2 = 0$). The odd derivatives cancel due to symmetry, so the Helmholtz coil field is given approximately by

$$B_0(z) \approx B_0(0) + \frac{1}{16} \frac{d^4 B_0}{dt^4} z^4$$

The RF field, B_1 is provided by a separate set of coils driven by a function generator. The function generator provides a variety of waveforms (sinusoid, square wave, etc.) and the ability to modulate the frequency of amplitude of the signal. You should spend some time familiarizing yourself with the wave forms and modulation capabilities of the function generator. Use the digital oscilloscope with channel 1 connected to the signal output and channel 2 connected to the modulation output. Turn off all modulation and look at a pure sinusoidal wave form. Then try both amplitude and frequency modulation.

The optical pumping can be monitored by the transmission of light through the cell and by the fluorescence produced when the incident photons are absorbed. The transmission is measured by a silicon photodetector connected

to a preamp. A video camera is provided to provide a nice visual signal of fluorescence when the high powered laser is used.



Getting Started

You should set up the experiment with the laser. Begin by setting up the camera, focused at the center of the rubidium bulb. In a dark room the video display will show the laser beam scattering from the glass surfaces of the bulb. Adjust the camera aperture for the clearest picture.

The laser must be tuned to the D1 line. It should be close, but should be checked by the lab staff before beginning. The laser is easily tuned with the aid of a piezoelectric element inside the laser cavity. A high voltage power supply is used for tuning. Adjust the voltage up and down by about 10-20 volts (possibly more) to tune the laser on and off the line. The laser tuning has some features that will, at first, seem strange: there is significant hysteresis and the wavelength does not always tune smoothly, so there is not always a one-to-one correspondence of voltage and wavelength. With the magnetic field B_0 off, use the video fluorescence signal to tune the laser to the D1 wavelength. You should actually see up to four lines corresponding to the two hyperfine levels of each isotope. (See figure 2.) Some laser instability is to be expected and you may need to retune.

Once you have the laser set to the strongest D1 line, use the fluorescence to monitor the optical pumping and magnetic resonance signals.

Turn on the magnetic field B_0 and set the current to 40 mA. You should see (and be able to explain) the change in fluorescence when B_0 is turned off and on.

Turn on the RF with maximum amplitude and a sinusoidal signal with NO modulation. Set $f \approx 1.48$ MHz. Scan the RF frequency in 1 kHz steps to find the resonance for ^{85}Rb . Also find the ^{87}Rb resonance near 2.1 MHz. You'll notice that even though the laser is tuned to a transition in only one isotope, magnetic resonance signals from both are observed. This is due to the rapid spin exchange of electron polarization among all the atoms.

Finally observe the signal from the photodetector on the oscilloscope. The best way to do this is to amplitude modulate the RF – that is turn it off and on. Use a rate of about 5 Hz and 100% modulation depth. The oscilloscope should be triggered with the modulation output signal from the frequency generator. Use the size of this signal to fine tune the RF frequency, the laser wavelength, and the quarter wave plate position.

Now you are ready for quantitative study of the magnetic resonance phenomena.

Things to measure

You should measure the gyromagnetic ratios for ^{85}Rb and ^{87}Rb , the optical pumping time constants (Γ^{-1}) with RF off and on, and the Lorentzian line shapes as a function of RF power.

Gyromagnetic Ratios

You will need to measure and/or calculate the magnetic field strength B_0 . You can easily formulate the field due to the Helmholtz windings using the elementary result of the field along the axis of a loop applied to this geometry. Alternatively, use a Hall Probe Gauss meter to measure the field. This is complicated by the location of the glass bulb at the position of interest. Measuring the magnetic resonance frequencies at several values of B_0 should yield a linear relation. The slope is γ_A .

Time Constants

The optical pumping and magnetic resonance time constants are best observed by amplitude modulation of the RF. Use 100% modulation depth with square wave modulation. The photodetector signal is observed with the digital oscilloscope. Signal averaging using the AVERAGING function accessed from the DISPLAY button on the oscilloscope is helpful. The stored, averaged waveform can be downloaded for analysis, and a quick analysis is possible using the oscilloscope cursors. Measure the amplitude and time constants with RF both on and off (two states of the square wave). Do this at several RF amplitudes. It is also interesting to investigate the dependence on laser intensity. Use neutral density filters that each reduce the intensity of incident light by a factor of 2.

Line Shapes

The Lorentzian line shape is expected for any two state process because the coherence time constants of two states is limited. The relationship of the line width (full width at half maximum) and the time constants is given in the introductory section. The oscilloscope can be used to measure the line shape by frequency modulating the RF. Use the linear or triangle waveform with about 40 kHz span and a rate of about 5 Hz. Alternatively you can measure the AM modulated signal amplitude as a function of RF frequency. Measure the line shapes and extract the amplitude and line width as a function of RF power.

Caveats

The laser can be tricky and it is possible to damage the laser diode, the piezoelectric element, or the cavity alignment. Please have a lab supervisor help get you started.

The photodetector has a linear response (output current vs input power) over a broad range, but it does saturate at high power levels encountered with the laser. You can check the linearity using the neutral density filters.

Prelab questions

1. The Lorentzian line shape is the frequency domain Fourier Transform of a decaying exponential in the time domain. You will be measuring in both the time and frequency domains in this lab. Confirm this mathematical relationship.
2. How does a quarter wave plate produce circularly polarized light when placed with its optic axis at 45° with respect to the linear polarization?
3. What differences do you expect in the optical pumping phenomena and signals if σ_+ light is used instead of σ_- light?