**ADVANCED UNDERGRADUATE LABORATORY**

**EXPERIMENT 5, Rb**

# Optical Pumping in Rubidium

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## Purpose

The object of this experiment is to measure the Zeeman splitting of the hyperfine structure components of the ground state of rubidium in a known weak magnetic field. From this measurement, the corresponding g-factor, $g_F$ is determined and hence the nuclear spin.

In the method used, the technique of "optical pumping" produces a non equilibrium distribution of atoms among the Zeeman sublevels of the atomic ground state. A Magnetic resonance experiment is then performed to measure the frequency (energy) difference between adjacent sublevels.

## Introduction

By "optical pumping", we mean the use of light to change the population of a set of energy levels of a system from the Boltzman distribution at the temperature of the experiment. That is, a system of atomic spin orientations is "pumped" from an equilibrium distribution over magnetic sub-states into a non-equilibrium distribution in which a large majority of spins are aligned in a given direction.

Consider the alkali atoms with their $^2S_{1/2}$ ground state and $^2P_{1/2}$ and $^2P_{1/2}$ excited states. A transition from the $^2P_{1/2}$ state to the $^2S_{1/2}$ state involves an energy sufficient to give rise to optical radiation. Since the first excited state $^2P_{1/2}$ is a $P$ state with orbital angular momentum $L = 1$ and since the total electronic spin $S = 1/2$, there are, in this state, two possible values of the total electronic angular momentum, $J = 3/2$ and $J = 1/2$. Due to the spin-orbit interaction, these two states of different total angular momentum differ in energy relative to the ground state which gives rise to the doublet structure in the spectra of the alkali metals. A familiar example of this is the pair of sodium $D_1$ and $D_2$ lines.

Due to the existence of a nuclear spin of the atom, all states, ground and excited, will be individually split into a number of substates which are designated as hyperfine states. Each of these hyperfine states has a slightly different energy arising from the interaction of the electron with the nuclear magnetic dipole moment and electric quadrupole moment. The different energies leading to this hyperfine structure come from the dependence of the interaction energy on the various quantum-mechanically allowed orientations of the nuclear angular momentum $I$ relative to the total electronic angular momentum $J$. According to the rule of quantization, the allowed combinations are such that the total angular momentum $F$ takes on the values $F = I+J, I+J-1, …, |I-J|$. Each of these levels may, in turn, be split again by the application of a weak external magnetic field.

This splitting arises from the different energies associated with different orientations of a given F with the external magnetic field and is directly proportional to the magnetic field in the low field limit. The only orientations of $F$ allowed are those which give projections $m_F = F, F-1, …, -F$ along the direction of the magnetic field.

Transitions between levels of different $F$, the hyperfine transitions, will involve absorption or emission of magnetic dipole radiation, with $\Delta F = 0, \pm 1$ only. Transitions between different $m_F$ levels with $F$ remaining constant comprise the Zeeman transitions with $\Delta m_F = 0, \pm 1$ only.

In this particular experiment, a vapour of free rubidium atoms, $^{85}$Rb with nuclear spin $I = 5/2$ and $^{87}$Rb with nuclear spin $I = 3/2$ are placed in a weak magnetic field which creates the Zeeman splitting of the hyperfine structure. A schematic energy level diagram for $^{87}$Rb is given in Figure 1.

We now consider the effect of shining circularly polarized resonance radiation of the $D_1$ type ($\lambda = 794.8$ nm) through the vapour, (assume that the $D_2$ has been filtered out). The free rubidium atom will

absorb the radiation and jump into the $^2P_{1/2}$ excited state. If the light is left circularly polarized, (i.e. all photon angular momenta are parallel to the direction of propagation), then absorption of a photon must lead to a net gain of one unit of angular momentum, if the light travels in the direction of the external magnetic field vector.

Absorption of a quantum of $D_1$ radiation by an atom in the $F = 1$, $m_F = -1$ substate can only lead to a substate with $m_F = 0$ in the $^2P_{1/2}$ excited state. Doppler broadening of the resonance radiation will in general allow transitions to either $F = 2$ of $F = 1$ hyperfine levels of the excited state. When this state reradiates to the ground state, the dipole selection rules permit $m_F = 0$, $\pm 1$ with equal probability, so that the atom has a probability of 2/3 to return to the ground state with a larger value of $m_F$ than it had originally. Figure 2 gives a schematic illustration of this process.

$$5\,^2P_{\frac{1}{2}}$$

$$5\,^2S_{\frac{1}{2}}$$

$m_f$

F=2
+2
+1
0
-1
-2

F=1
+1
0
-1

F=2
+2
+1
0
-1
-2

F=1
+1
0
-1
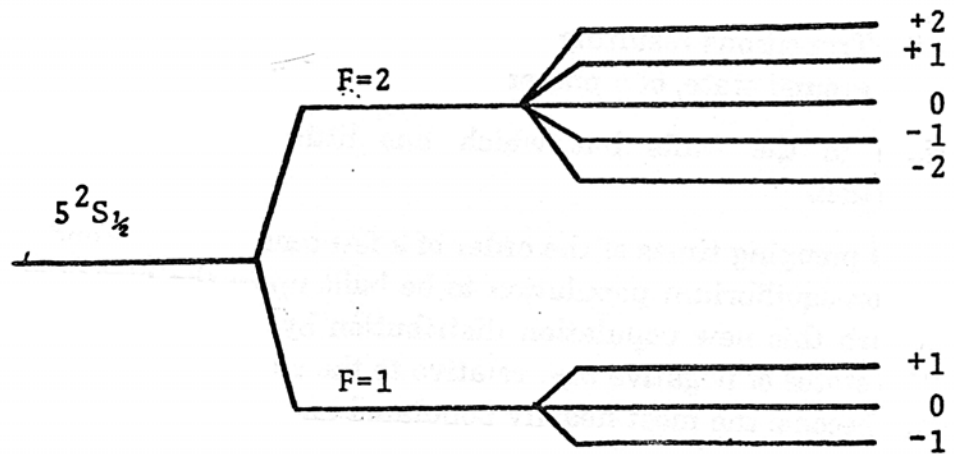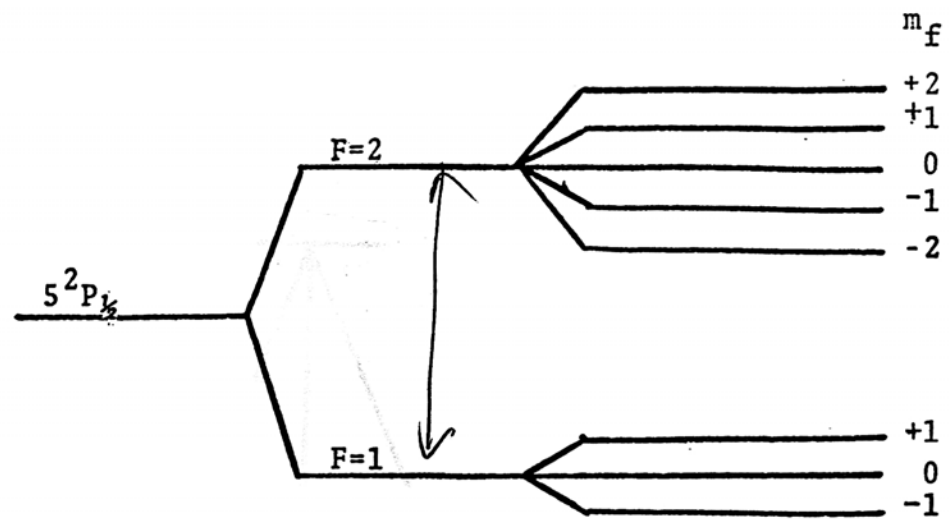
Atomic
State

Hyperfine
Structure

Zeeman Splitting in
Weak Magnetic Field

Figure 1: Energy levels of $^{87}Rb$ (I = 3/2).

This absorption and re-radiation can continue repeatedly, and if, between these occurrences, the $m_F$ level populations are not relaxed, soon all the atoms will be "pumped" into the $F = 2$, $m_F = 2$ ground state level giving complete alignment of the atomic spins. When the atom reaches this level, it can no longer absorb $D_1$ radiation. Experimentally, however, this ideal situation is never reached since atom-atom and atom-wall collisions cause mixing of $m_F$ levels in ground and excited states. The effect of atom-wall collisions may be reduced by the introduction of an inert buffer gas such as helium which increases the time taken for a rubidium atom to diffuse to the walls but which has little effect in producing collisional depolarization.
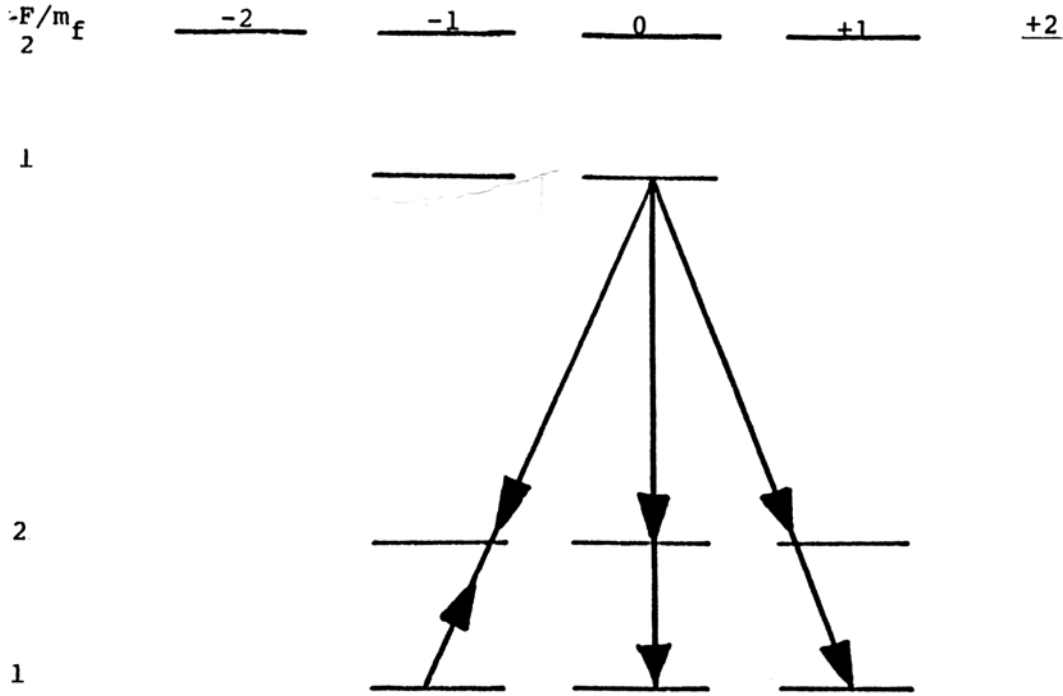
Figure 2: Transitions resulting from absorption by an atom in the $F = 1$, $m_F = -1$ ground state, of a photon carrying a +1 unit of angular momentum.

Optical pumping times of the order of a few tens of milliseconds, are sufficient to allow a non-equilibrium population to be built up in the higher $m_F$ sub-states. We may disturb this new population distribution by reversing the external magnetic field. The states of negative $m_F$, relative to the axis defined by the light beam, will suddenly become the most heavily populated and strong light absorption will occur as these are pumped back toward positive values. A photomultiplier monitoring the transmitted light intensity serves as the detector of optical pumping. This method, however, will only show the presence of atomic alignment.

If, when the vapour is optically pumped, an rf signal of proper frequency to cause Zeeman or hyperfine transitions is applied to the vapour, light absorption will increase because the non-equilibrium population has been disturbed. Thus, optical detection of Zeeman hyperfine structure transition frequencies is made possible.

The magnetic moment $\mu_F$ is proportional to the angular momentum $\vec{F}$ where the proportionality factor is $\gamma_F$ the gyromagnetic ratio.

$$\vec{\mu}_F = \gamma_F \vec{F} \tag{1}$$

The gyromagnetic ratio is often written as

$$\gamma_F = g_F \left( \frac{e}{2m} \right) \tag{2}$$

for historical reasons since $e/2m$ is the gyromagnetic ratio for a classical spinning electron. (An electron considered classically would be a uniform spherical distribution of mass and charge.) The value $g_F$ is called the g-factor or spectroscopic splitting factor.

The energy of interaction between the magnetic moment and the magnetic field is

$$E = -\vec{\mu}_F \cdot \vec{B} = -\gamma_F \vec{F} \cdot \vec{B} = -\gamma_F F_z B \tag{3}$$

where $F_z$ is the component of $\vec{F}$ along the magnetic field. For the ground state of $^{87}$Rb where $J = 1/2$ and $I = 3/2$, $F$ can have values 2 and 1. For $F = 1$, the components of $F$ along the magnetic field $\vec{B}$ can have values $\hbar$, 0 and $-\hbar$. Thus, from (3), $E$ can have values $\gamma_F \hbar B$, 0 and $-\gamma_F \hbar B$. So for weak fields the separation between the adjacent states, $\Delta E$ is constant and is given by

$$\Delta E = \gamma_F \hbar B \tag{4}$$

The same arguments give the same result as (4) for other values of $I$ and $F$.

For absorption of rf, the energy separation of the states must be equal to the energy of the incident radio-frequency (rf) photons. Using (4) we have

$$h\nu = \gamma_F \hbar B \tag{5}$$

and from (2)

$$h\nu = g_F \left( \frac{e}{2m} \right) \hbar B \tag{6}$$

or

$$g_F = 4\pi \frac{m\nu}{eB} \tag{7}$$

In books on atomic spectra like White (p 374) or Kuhn (p 348), $g_F$ is given by

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} - g_I \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} \tag{8}$$

Equation (8) may be simplified if we ignore the second term. Recall that observed hyperfine structure separations are many times smaller than fine structure separations. This means that the $g_1$ factor for the nucleus must also be very small compared with the $g_J$ factor for the extranuclear electrons. Another way of looking at this, is that if the nuclear spin is due to one or more particles with the mass of protons, then the nuclear $g_1$ factors should be in the neighbourhood of a thousandth of those for electrons. Penselin et al. have shown that the ratio $g_1/g_J$ is -0.00015 and -0.00050 for the $^{85}$Rb and $^{87}$Rb isotopes respectively. Ignoring $g_1$, equation (8) becomes

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

where

$$g_J = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)} \qquad (10)$$

Thus, by measuring the magnetic field and the resonant frequency $v$ and using (7), (9) and (10) one can determine the nuclear spin $I$.

For a more complete discussion of optical pumping see the book "Optical Pumping" by Bernheim or the extensive review article of Skrotskii and Izyjumova in which $^{87}$Rb and $^{23}$Na** are discussed in detail

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** Since $I = 3/2$ for $^{23}$Na and $^{87}$Rb, both have the same hyperfine structure.

## Apparatus

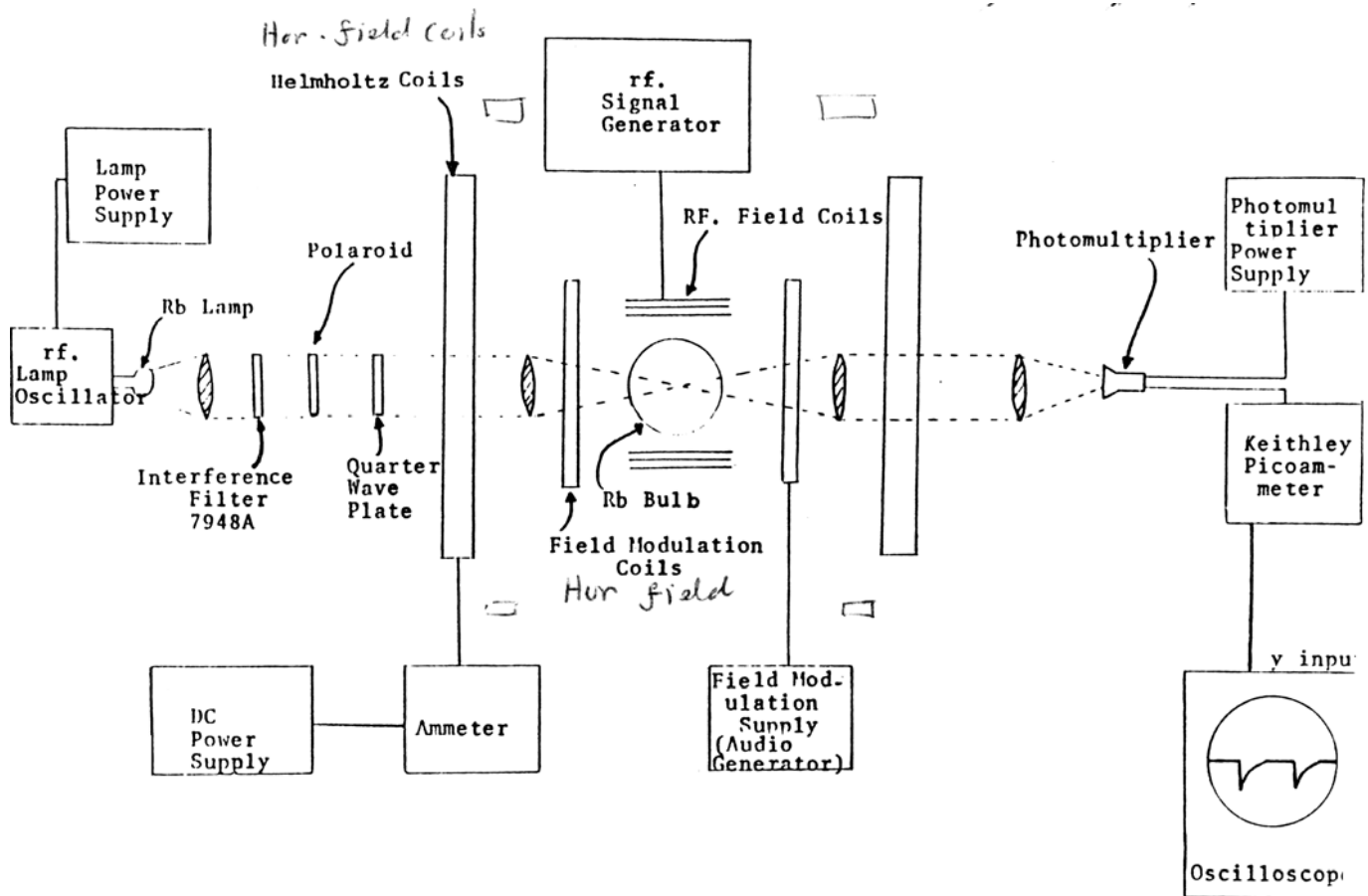A schematic diagram of the apparatus is given in Figure 3.



Figure 3: Schematic Diagram of the Optical Pumping Apparatus.

### Rubidium Vapour

A pyrex bulb contains metallic rubidium whose vapour pressure at room temperature is of the order of $10^{-5}$ torr. The bulb also contains about 60 torr of helium which acts as a buffer gas, increasing greatly the time required for a rubidium atom to diffuse to the walls of the bulb. This is necessary since collisions with the walls cause very strong mixing of the different $m_F$ levels, and hence tend to destroy the spin alignment. Collisions with helium atoms, on the other hand, have little tendency to produce depolarization.

### Rubidium Lamp

The lamp is an rf electrode-less discharge with very narrow lines. The lamp tube contains a drop of rubidium and a carrier gas of about 10 mm Hg pressure. The tube sits inside the tank coil of a push-pull oscillator operating at about 10 MHz and provides part of the load. The discharge is initially in the argon but as the system heats up, the rf power is absorbed mainly by the rubidium.

The pressure of the rubidium in the lamp is controlled by the temperature of the base of the lamp. The temperature of the base is controlled by current through a section of heating tape. If the temperature of the base is too low then the rubidium pressure will be correspondingly low and so also will be the intensity of the rubidium excitation lines. If the rubidium pressure is too high then self absorption in the

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lamp will cause the lines to broaden which effectively reduces the optical pumping intensity and, in addition, the lamp will become unstable.

In ordinary discharge lamps, excitation of the rubidium takes place throughout the whole volume. A photon emitted from the centre of the lamp has a high probability of being reabsorbed before escaping from the lamp. Re-absorption is most likely for the centre of the Doppler broadened line and at high enough pressures this self broadening may even result in self reversal where the output line from the lamp appears as two closely spaced lines (really it is one line with part of the centre missing). In the rf lamp, the power is absorbed in a very narrow skin depth layer of metallic rubidium vapour. Since excitation takes place at the surface of the lamp, each photon emitted has a high probability of escaping without being reabsorbed. The emission lines thus closely match the narrow absorption lines. If one looks down on the discharge tube, the ring discharge appears as a bright pink glow near the glass.

**Interference Filter**

The interference filter is a narrow bandpass filter which transmits the 794.7 nm component of the resonance doublet but eliminates the 780.0 nm component. This is necessary to maximize the effect of optical pumping. A discussion of this is given by de Zafra.

**Circular Polarizer**

The circular polarizer consists of a sheet of polaroid followed by a quarter-wave plate. For a discussion of this see Jenkins and White p 556.

**Oscillatory rf Field**

The oscillatory rf field is produced within the small pair of coils which surround the rubidium bulb by an alternating current from a variable frequency rf signal generator.

**Field Modulation**

Field modulation permits the resonance to be displayed on the oscilloscope. An A.C. current from the audio signal generator is applied to the inner pair of Helmholtz coils to modulate the steady magnetic field $\vec{B}$. If the resonance condition is fulfilled, this modulation causes the resonance to occur twice during each period of the modulating field.

## Procedure

1.      LAMP

All settings for the lamp have been optimized for maximum intensity and need not be altered. Turn on the LAMP OSCILLATOR SUPPLY. Turn the LAMP HEATER SUPPLY to 50. The lamp should reach equilibrium in about 30 minutes although it will be usable in about 15 minutes.

This process can be speeded up so that the lamp will be stable in about 15 minutes by initially turning up the LAMP HEATER SUPPLY to 90 for a maximum of five minutes. If it is left at 90 too long then too much rubidium will diffuse from the base into the lamp tube. The lamp will become unstable and it will take some time for the rubidium to diffuse back into the base.

2.      OPTICAL ALLIGNMENT AND PHOTOMULTIPLIER

The most important factor in this experiment is the light intensity.  Unless the optical components have been moved noticeably out of place one should assume that the optics have been suitably lined up.  Insertion and removal of the rubidium absorption cell can cause small misallignment.  One can obtain maximum intensity readings by small vertical and horizontal adjustments of the photomultiplier.

There are two filters inside the photomultiplier assembly.  The first is a Wratten filter (89A) which has a high transmission for 794.7 nm but which has a sharp cutoff in the visible.  The second is a dark red glass filter (587) which also reduces intensity by a factor of 10.  With this combination one may perform the experiment with the room lights on.

The supply voltage to the photomultiplier should be 800 V or less since the maximum current through the R.C.A. 4832 photomultiplier is $10^{-7}$ ampere.

When measuring small changes in the photomultiplier current one may use the current suppress to eliminate a large part of the signal and then turn to a more sensitive scale to observe changes.

WHEN MAKING ANY CHANGES IN SETTINGS OF THE EQUIPMENT, ALWAYS TURN THE PICOAMMETER SCALE TO A HIGH SETTING TO AVOID DRIVING THE NEEDLE OFF SCALE.

3.      CIRCULARLY POLARIZED LIGHT

Circularly polarized light may be obtained by employing the following procedure.

The $D_1$ line emitted by the lamp is unpolarized and the interference filter which transmits only the $D_1$ line does not polarize the light.  A piece of polaroid is in the assembly with the interference filter and although the assembly can be rotated, its orientation (at least before the quarter wave plate is considered) is irrelevant.  When the absorption cell is in the beam, the intensity seen by the photomultiplier will change by a few per cent as the quarter wave plate is rotated.  The degree of circular polarization will be a maximum when the intensity is either a maximum or a minimum.  Which is it and why?  In order to see the small changes as the quarter wave plate is rotated one should use the zero suppress on the picoammeter.

Confirm that you have circularly polarized light by placing another piece of polaroid in front of the photomultiplier and rotating it.  You will notice that the light is not 100% circularly polarized.

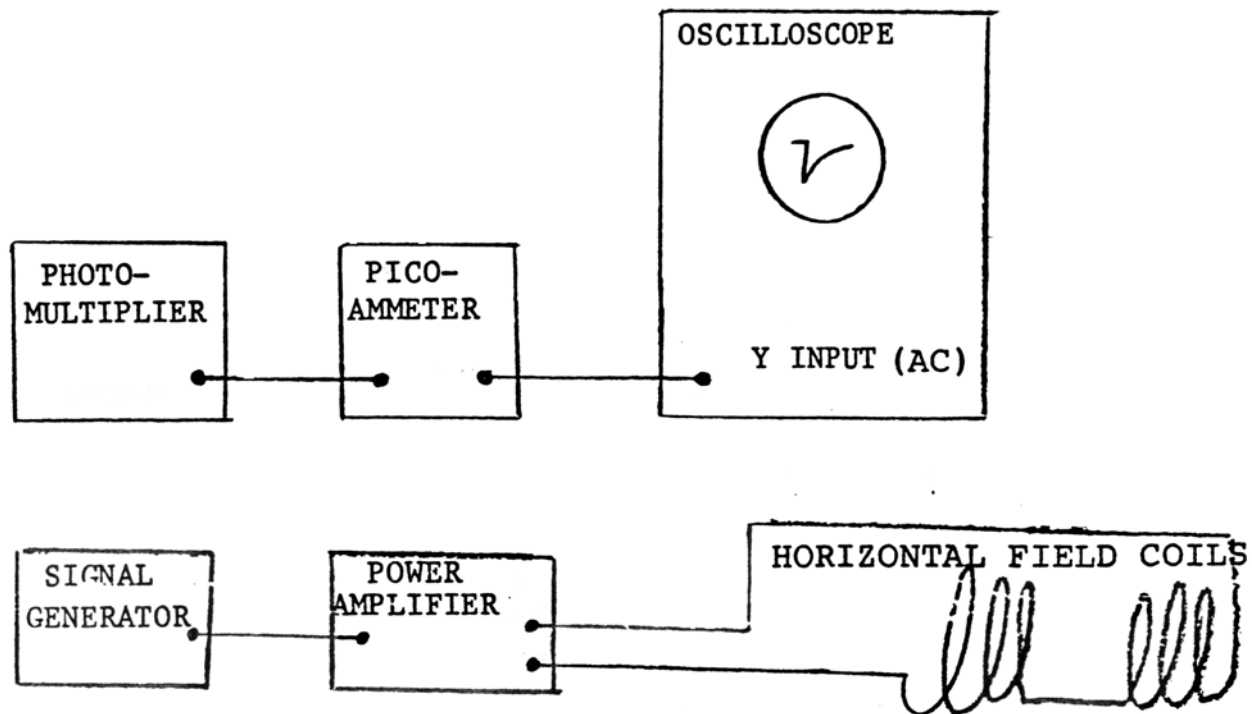4.      PRELIMINARY OBSERVATION OF OPTICAL PUMPING

**Figure 4: Schematic of Equipment for Preliminary Observations.**

Connect the equipment as shown in figure 4. A reproducible display can be obtained by triggering the oscilloscope with the SYNC OUT of the signal generator. Apply a square wave signal of 0.8 A amplitude and frequency of about 0.6 Hz to the horizontal field coils and observe the resonance as the field passes through zero.

5.    DETERMINATION OF ZERO FIELD

Optical pumping is most pronounced when the light incident on the absorption cell is circularly polarized with the direction of propagation parallel to the external magnetic field. In the apparatus, the earth's magnetic field is at some angle to the direction of propagation of light and this angle can be reduced by eliminating the vertical component of the earth's magnetic field with the VERTICAL FIELD coils. As the vertical component is reduced, the optical pumping increases and the absorption cell will become more and more transparent to the circularly polarized light as in figure 5.
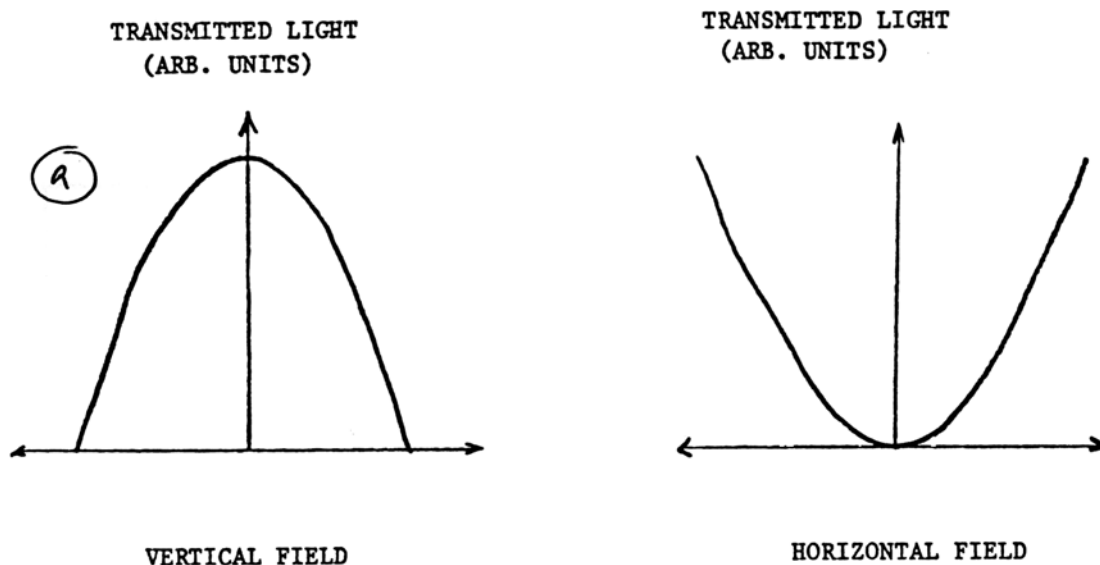
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Figure 5: Transmitted Intensity as a Function of Magnetic Field

As the magnetic field along the direction of propagation decreases due to the current through the HORIZONTAL FIELD coils, the magnetic $m_F$ sublevels become closer and closer together until in zero field they are degenerate. This means that the optical pumping of the $m_F = 2$ level is not as effective and hence the amount of transmitted circularly polarized light will decrease. It should be noted that even in zero field the $F = 2$ level will be pumped relative to the $F = 1$ level even though the $m_F$ levels are degenerate.

With the picoammeter still on a sensitive scale, connect the D.C. power supplies to the HORIZONTAL FIELD and VERTICAL FIELD coils. By referring to figure 5, determine the current through the VERTICAL FIELD and HORIZONTAL FIELD coils which produce zero field and record these values. They will be used later to zero the gaussmeter when measurements of magnetic field are made.

6.    DETECTION OF OPTICAL PUMPING AT ZERO FIELD USING THE MODULATION COILS

With zero magnetic field at the absorption cell, connect the output from the power amplifier to the modulation coils and observe the resonance as the field passes through zero. This is similar to step 4, the preliminary observation of optical pumping. Now vary the current, first through the HORIZONTAL FIELD coils and then through the VERTICAL FIELD coils. Notice that the size of the resonance signal decreases. Why?

Measure the approximate time that it takes to pump the system after the direction of the magnetic field has changed. What determines the pumping time in the apparatus?

7.    DETECTION OF OPTICAL PUMPING AT NON ZERO FIELDS USING RF MIXING

a.    **The AC Method**

Turn the current through the HORIZONTAL FIELD coils up to about 250 ma so that the field is several times the earth's magnetic field.  The optical pumping signal should disappear.  Why?  Turn on the rf generator and increase the rf frequency until the optical pumping signal appears again.

As the frequency is increased further, the signal should disappear and then reappear at a higher frequency.  The signal at the higher frequency which corresponds to the $^{87}$Rb isotope will be weaker since the $^{87}$Rb isotope comprises 28% of natural rubidium as compared to 72% for $^{85}$Rb.

If the two separate resonances cannot be picked out then it may be necessary to reduce the amplitude of the modulating field.  Why is this necessary?

### b.      **The DC Method**

Once one understands the physics of what is happening, a more accurate method of obtaining the resonance frequency may be used.  As the resonance frequency is approached, the steady DC current decreases because the amount of light transmitted through the cell decreases.  The reason for this is that at resonance the optically pumped $m_F = 2$ state is depopulated by the rf and thus more optical pumping takes place with the concomitant absorption of light.  By varying the rf frequency one should observe two minima in the DC current corresponding to the two rubidium isotopes.  The relative magnitudes of the two dips should correspond to the natural abundances of the rubidium isotopes.

8.      DETERMINATION OF THE NUCLEAR SPIN OF THE RUBIDIUM ISOTOPES

At this point one may proceed to section (10) and measure the magnetic field as a function of current or one may continue by assuming that magnetic field is proportional to current.

For various magnetic fields, (or currents in the coils), both in the direction of the earth's field and opposite it, measure the two resonant frequencies corresponding to the two rubidium isotopes.  Plot frequency versus magnetic field and determine the $g_F$ values from the slopes of the lines.  By using equation (9), determine $I$.  Having determined $I$, compare your values of $g_F$ with the calculated values obtained using equation (9).  Do they agree within experimental error?  How did you obtain your experimental error?  One should obtain a least squares fit to the experimental data.  Do the two lines intersect at the value of current which is supposed to give zero field?  If not, why not?  If all measurements of frequency were 10 kHz too high, how would this affect your results?

9.      COMBINED RESONANCES

Adjust the current in the HORIZONTAL FIELD coils until the absorption cell is in zero field.  Change the input to the power amplifier from a square wave to a triangular wave of about 0.6 Hz.  Note that the SYNC OUT from the function generator comes at a different point in the period depending on whether the output is a triangular or square wave.  Adjust the power amplifier output to the MODULATION COILS to maximum current (about 800 ma).  Turn the rf frequency to its maximum value of 600 KHz.  With the time base on the oscilloscope set at 50 ms (variable), adjust the triggering and the variable time base on the oscilloscope until a reproducible pattern appears on the oscilloscope which will probably look like that in figure 6.
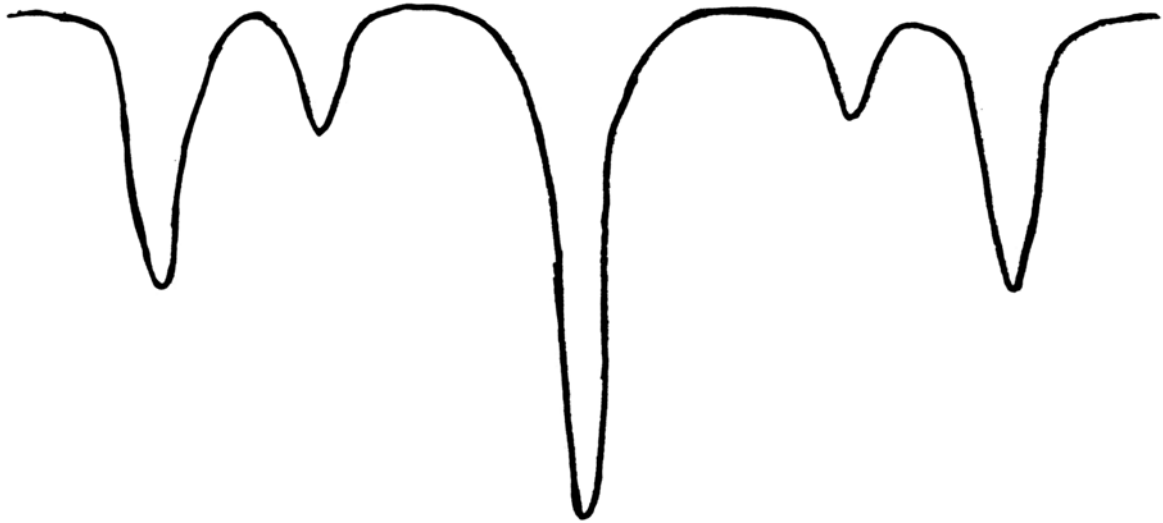
**Figure 6: Absorption Versus Magnetic Field for a Fixed rf Frequency.**

Draw a sketch of what you observe and be able to explain it. Be able to explain the horizontal spacing of the spikes as well as their relative heights. Lower the frequency of the rf oscillator. What happens? Why?

10.     DEPENDENCE OF MAGNETIC FIELD ON CURRENT

Being extremely cautious, remove the absorption cell and store it in a safe place. Remove the support stand and replace it with the jig to hold the gaussmeter probe. Adjust the current in the Helmholtz coils to the value which you know gives zero field and zero the gaussmeter making sure the meter reading remains zero as the Hall effect probe is rotated. Plot magnetic field both along and against the horizontal component of the earth's magnetic field as a function of current.

**QUESTIONS**

1.     Using classical arguments, explain the interaction of the rf field with the magnetic moments which precess about the constant magnetic field along the axis of the apparatus (i.e. along the direction of propagation of the light).

> HINT: The arguments are the same as for other resonance experiments such as electron spin resonance, nuclear magnetic resonance or nuclear quadrupole resonance.

2.     In this experiment you are working with the ground state of rubidium. For this state evaluate equation (10) without the substitution of numerical values for $J$ and $S$.

3.     Using the results of question (2) show that equation (9) becomes

$$g_F = \frac{2S}{I+S} \qquad\qquad (11)$$

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4.  Recall that $g_s$ for a free electron is 2 (or 2.0023 after corrections for relativistic effects). Kusch and Taub had suggested and Dehmelt has shown that $g_J$ for the ground state of sodium and the lighter alkali metals and $g_s$ for a free electron are identical to within their very small experimental error. Kusch and Taub have also shown that for the ground state of rubidium, $g_J$ = 2.0024 which is almost identical to $g_s$. Keeping in mind your answer to question (2), and how you obtained it, comment on the meaning of this.

5.  The splitting between the $\Delta m_F$ levels is approximately but not exactly equal so that in principle one should observe four resonant frequencies for each of the isotopes. For example, Driscoll states that for $^{87}$Rb, in a field of 12 gauss, the four $F = 2$ lines are spread over a range of about 64 kHz. Extrapolate from this data to the magnetic fields in your experiment and discuss the possibility of observing separate $F = 2$ resonances for a single isotope.


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