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A MEASUREMENT OF THE SPIN RELAXATION TIME OF
OPTICALLY PUMPED RUBIDIUM VAPOR

by

Robert Oliver Breault

A thesis presented to the
Faculty of the School of Graduate
Studies in partial fulfillment
of the
Degree of Master of Arts

Western Michigan University
Kalamazoo, Michigan
June 1964

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CHAPTER I

THE PROBLEM AND ATOMIC ENERGY LEVELS

In 1949 Francis Bitter proposed a method for optically detecting radio frequency resonance. That same year Alfred Kastler, Ecole Normale Supérieure, Paris, France, introduced a technique, which he called "Optical Pumping", of concentrating atoms in some of the Zeeman sublevels of one of the hyperfine state levels of the ground state. These two experiments are the basis of research involving optical methods of atomic orientation and of magnetic resonance.

I. THE PROBLEM

Statement of the problem. It was the purpose of this study to measure the spin relaxation times of rubidium vapor atoms oriented by polarized resonance absorption with neon acting as a buffer gas.

Method of approach. The optical pumping technique was used for alignment. The pumping radiation, consisting of circularly polarized D_1 resonance radiation, was suddenly shut off and then turned on again in some specified time t . The relaxation of the vapor while the radiation was off, caused the vapor to become more nearly opaque. The

rate of development of the opacity then determined the relaxation time which could thus be measured. In optical pumping no transport of material is involved, but rather a "pumping" of atomic spin orientations from an equilibrium distribution over magnetic substates into a nonequilibrium distribution in which a preponderance of spins is aligned in a given direction, i.e., parallel to the direction of propagation of the circularly polarized light.

Characteristics of vapor. There are two elements, sodium and rubidium, on which optical pumping may most easily be accomplished. Of these, rubidium pumping is hampered by the lack of intense light sources, but otherwise presents distinct advantages over sodium pumping. These are (a) sufficient vapor pressure (1.5×10^{-6} mm) for good pumping at 55°C , in contrast to the 140°C needed for sodium, and (b) the large separation of the D_1 and D_2 lines (7947 Å and 7800 Å, respectively) allowing efficient removal of the D_2 line by the use of interference filters.¹

II. ATOMIC ENERGY LEVELS

Energy levels due to R-S coupling. Rubidium, $Z = 37$ is an alkali metal which has thirty-six core electrons with a single electron in its outer most (0) shell.

According to the Bohr model "only those orbits are permissible for which the angular momentum of the electron is an integral multiple of \hbar ", and only those electrons which are outside the closed shells can be considered as contributing to the angular momentum. As the electron orbits, it has an associated angular momentum called the orbital angular momentum given by quantum mechanics to be: $\hbar\sqrt{l(l+1)}$ where $l = 0, 1, 2, 3, \dots$. An electron in the ground state of rubidium may then be represented by $n = 5$ and $l = 0$, with the first excited state as $n = 5, l = 1$.

As the electron of the rubidium atom revolves, it also spins or rotates about its axes. The angular momentum of the electron due to its spin, $\hbar\sqrt{s(s+1)}$, has the spin quantum number s and the value of one-half. Since rubidium has only a single outer orbital electron, the projection of \vec{s} with respect to a vertical magnetic field vector \vec{H} has as a value for parallel spin (vector pointing up), $m_s = +1/2$ and for anti-parallel spin (vector pointing down), $m_s = -1/2$.

In the calculation of the total electronic angular momentum, j , for one electron the vector sum of \vec{s} and \vec{l} is used, i.e., $\vec{j} = \vec{l} + \vec{s}$. The vectorial precession about \vec{j} is due to the magnetic interactions of the magnetic moments. Since in the case of rubidium atoms, the total spin angular momentum, $\vec{S} = \sum_i \vec{s}_i$, and the total orbital

angular momentum, $\vec{L} = \frac{h}{2\pi} \vec{l}$, is for one electron then the total angular momentum of the electron, \vec{J} , is $\vec{J} = \vec{s} + \vec{l}$.

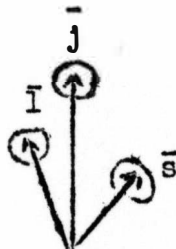


FIGURE 1

VECTORIAL REPRESENTATION OF SPIN-ORBIT COUPLING

For the ground state designation, $n = 5$, $L = 0$, $S = 1/2$ and thus $J = 1/2$.

In the historical development of spectroscopy, the notation for electron configuration was given as:

$$L = 0, 1, 2, 3, 4, \dots$$

$$S \quad P \quad D \quad F \quad G \quad \dots$$

The number of possible values of J , due to the multiplicity of the state is written as a superscript and the total angular momenta J written as a subscript. For rubidium, $5^2S_{1/2}$ represents the ground state and $5^2P_{3/2}$, $5^2P_{1/2}$, the first excited states. The energy difference of the two P-states arise because of the spin-orbit interaction. These two states of different total electronic angular momentum differ in energy and give rise to the familiar doublet structure in the spectrum of rubidium.

Because of the existence of nuclear spin, the ground state and all other states may be additionally split into a number of substates, the hyperfine states. These states have slightly different energies, arising from the interactions of the electron (magnetic moment) with the nuclear magnetic dipole moment. The discreteness of the energy states of this hyperfine structure arises from the quantum-mechanically allowed orientations of the nuclear angular momentum \bar{I} and the total electronic angular momentum \bar{J} . According to the rules of quantization, the allowed combinations are such that the total angular momentum of the atom, \bar{F} , takes on the integral values $F = |I + J|, |I + J - 1|, \dots, |I - J|$ only.² For rubidium 87, $I = 3/2$ and for rubidium 85, $I = 5/2$. The development in the next section is for rubidium 87 and the development of rubidium 85 is similar. The addition of the total electronic angular momentum, J , to the total nuclear angular momentum, I , is then given by $\bar{F} = \bar{I} + \bar{J}$ where; $F = 1, 2$ for $J = 1/2$, and $F = 1, 2, 3$ for $J = 3/2$.

Energy levels due to external field for $I = 3/2$.

A still further splitting of energy levels (the Zeeman splitting of the hyperfine structure levels) may be produced by application of an external magnetic field. This splitting arises from the different energies associated

with different orientations of the magnetic moments of a given F with the external field \vec{H} and is directly proportional to the magnitude of \vec{H} in the low field limit. In a very weak magnetic field, the vector \vec{F} precesses about the direction of the magnetic field.³ The projection of \vec{F} on the direction of \vec{H} , which is m_F , can assume values differing by unity and ranging from F to $-F$, giving $2F + 1$ values.

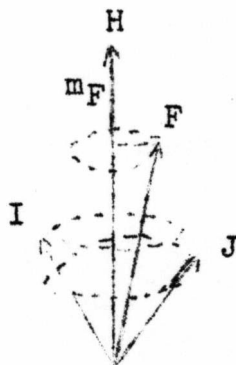


FIGURE 2

COUPLING, PRECESSION AND PROJECTION OF
TOTAL ANGULAR MOMENTUM VECTOR F .

The energy levels for the ground state and first excited states of a rubidium atom with nuclear spin $I = 3/2$ are shown on the following page. Transitions between different F levels, the so called hyperfine transitions, will involve absorption or emission of magnetic dipole radiation, with the attendant rule $\Delta F = 0, \pm 1$. Transitions between different m_F levels,

(with F remaining constant) comprise the Zeeman transitions, with the attendant rule $\Delta m_F = 0, \pm 1$. It should be noted that on this diagram the spacing between energy levels is not to scale, and for simplicity the Zeeman splitting of the $5^2P_{3/2}$ state is not shown.

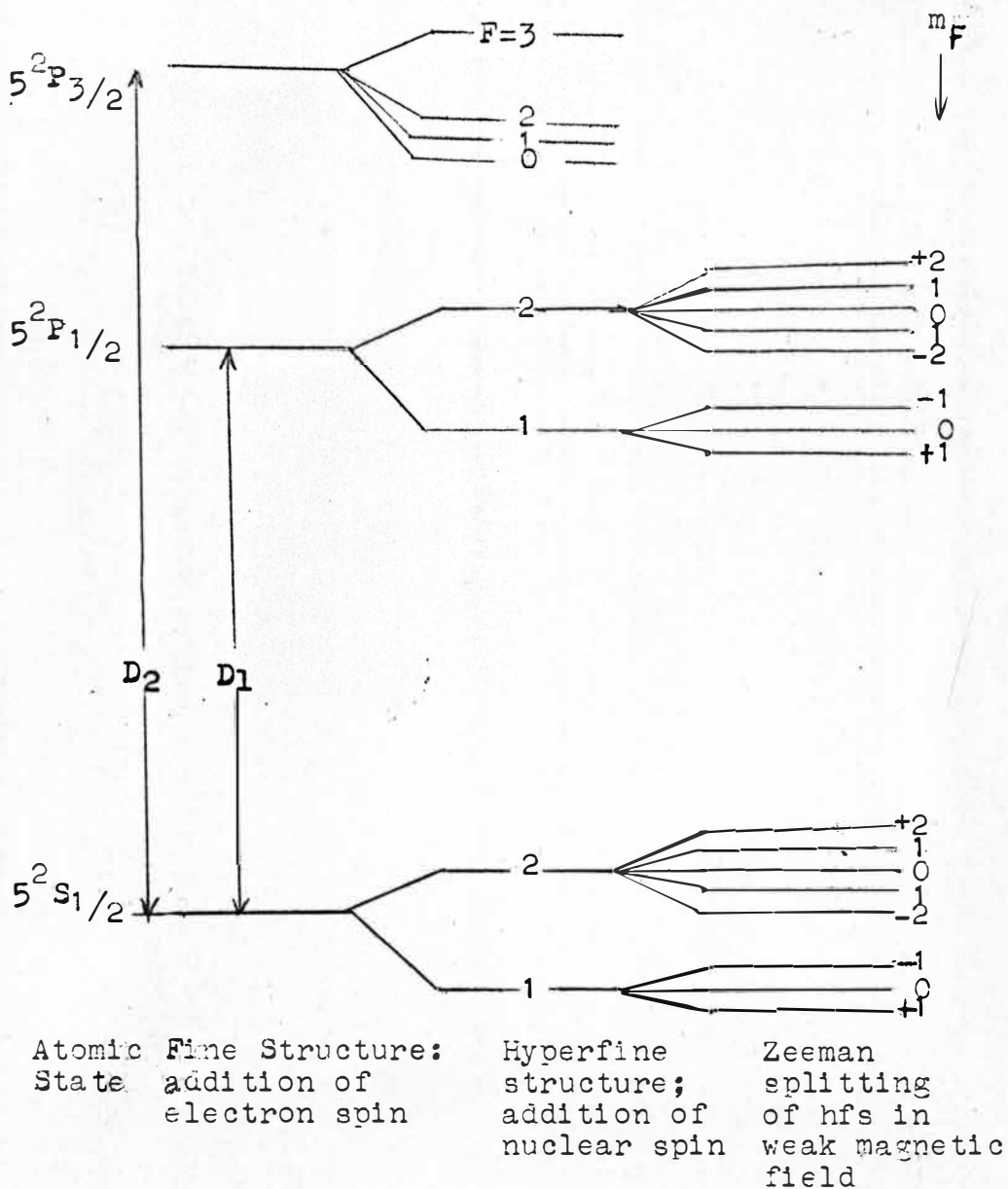


FIGURE 3

ATOMIC ENERGY LEVELS FOR RUBIDIUM 87
IN A WEAK MAGNETIC FIELD
(Not according to scale)

CHAPTER II

THE OPTICAL PUMPING PROCESS

The efficiency of alignment by the optical pumping technique involves three factors: (1) transition probability; (2) occupation probability; (3) circular polarization of the resonant radiation.

I. TRANSITION PROBABILITY

Atomic systems may be described as existing in stationary states, each of which corresponds to a definite value of the energy, and thus characterized by a quantum number of the energy. When two or more states have the same energy the level is called degenerate, and the number of states with the same energy is the multiplicity of the level. Transitions between energy states may occur with attendant emission or absorption of energy as radiation, or with the transfer of energy to or from another system. The frequency of the radiation emitted or absorbed by the system is given by Bohr's frequency relation, $h\nu = E_2 - E_1$, where E_2 and E_1 are the energies of the states between which the transition takes place and h is Planck's constant.

The level of the system with the lowest energy is

called the ground level, and every other level is an excited level. An atom in the ground level can only absorb radiation. When the atomic system is not in the ground level, it may spontaneously change to a lower level with the emission of radiation. The probability that an atom in level i will spontaneously change to the lower level j within a unit of time is called the spontaneous transition probability and is denoted by the Einstein coefficient A_{ij} . Between multiple levels the probability of all transitions becomes $\sum_i \sum_j A_{ij}$ summed over all state pairs involved. If there is a large collection (ensemble) of atomic systems and a_i is the number of the systems in the i^{th} level, the number of transitions to level j will be $\sum_i a_i A_{ij}$ per second. Spontaneous radiations will emerge from the atoms of the assembly in a random phase; therefore the assembly of independent gas atoms is said to emit this radiation as an incoherent source.⁴ In absorbing energy (or in transitions to higher energy levels) the atoms absorb photons from a beam of radiation producing an absorption spectrum in the transmitted light. In the reverse process (transitions to lower energy levels) the atoms emit photons producing an emission spectrum. In general, atoms in the form of a gas or vapor exhibit sharp emission or absorption lines.

A probability transition illustration. Popula-

tion redistribution in energy states due to absorption of resonance radiation may be demonstrated with an ensemble of atoms having three distinct energy levels, E_1 , E_2 , E_3 , where the energy difference between the levels is $h\nu = E_m - E_n$, $m > n$. When the ensemble of atoms is exposed to radiation of frequency $\nu = \frac{E_3 - E_1}{h}$ photons of light will be absorbed by atoms in state E_1 and transitions to level E_3 will occur.

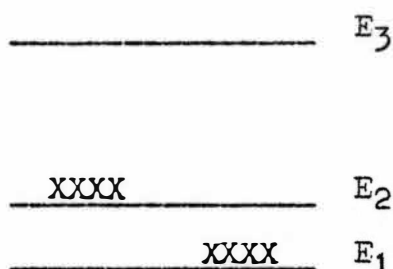


FIGURE 4

ENSEMBLE OF ATOMS IN THREE ENERGY LEVELS
WHERE X DESIGNATES A PARTICULAR ATOM

The average time that the atom remains in E_3 is the mean life for that particular state. Reradiation of the excited atom to a lower state, E_1 or E_2 , may occur with probabilities of the same order of magnitude. Since any atom after returning to E_1 may absorb photons of light and no atom in E_2 will absorb that frequency, given enough time the atom will terminate in the E_2 state and the material is then considered to be "optically pumped" from state E_1 to state E_2 via state E_3 . After

pumping, nearly all of the atoms will be found in E_2 if the pumping rate is substantially greater than the spontaneous transition rate $E_2 \rightarrow E_1$.

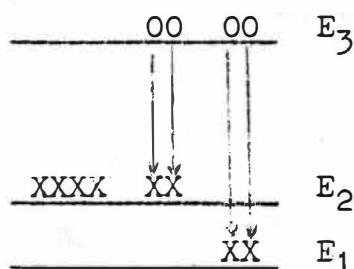


FIGURE 5

TRANSITION PROBABILITY FROM HIGHER TO LOWER ENERGY LEVELS

Detection of the pumped state. A simple method for the detection of the pumped state was developed by H. G. Dehmelt⁵ at the University of Washington, and is the technique used in this problem. The method depends on the fact that the transparency of the sample to the light beam varies with the population of state E_1 . As atoms are removed from state E_1 , the material can absorb less and less of the pumping radiation, and more of the radiation passes through reaching a maximum when state E_1 is empty. If some of the atoms are suddenly returned to state E_1 , transmission light will again be absorbed and the intensity of the transmitted beam will drop sharply.

II. CIRCULARLY POLARIZED RADIATION

If the pumping radiation is circularly polarized, i.e., the intrinsic photon angular momenta are parallel to the direction of propagation, then absorption of a photon by an atom will lead to a net gain of one unit of angular momentum by the absorbing atom in the direction of photon motion.

Absorption of left circularly polarized quantum of D_1 radiation by an atom in the $F = 1$, $m_F = -1$ substate, can thus lead to a substate with $m_F = 0$ in the $5^2P_{1/2}$ excited state. When this state reradiates to the ground state, the dipole radiation rules permit $\Delta m_F = 0, \pm 1$ with equal probability, so that the atom when returning to the ground state has a probability of two-thirds to return to a state with a greater value of m_F than it originally had. If no appreciable redistribution of the population in the various m_F levels occurs between absorption and reradiation of the atom, in time an excess of atoms would be "pumped" in the $F = 2$, $m_F = +2$ ground state level, giving alignment of the atomic spins.

Orientation and mixing of the m_F levels of $5^2S_{1/2}$.

A partial orientation of the rubidium vapor by optical pumping with circularly polarized resonance radiation

may be obtained even if both D_1 and D_2 lines exist in the pumping radiation. If the D_2 (7800 Å) line is filtered out, a larger orientation is possible because the mechanism controlling the orientation process is independent of the degree of random reorientation in the excited state.⁶ If left circularly polarized (σ^+) radiation is utilized, the selection rule, $\Delta m_F = +1$ applies to absorption. The highest value m_F may attain in the $5^2S_{1/2}$ ground state is $m_F = I + 1/2$. Since radiation inducing transition to the $5^2P_{3/2}$ state (D_2 line) has been filtered out, the $m_F = I + 1/2$ substate of $5^2S_{1/2}$ cannot undergo transitions because for $m_F = +2$ a $\Delta m_F = +1$ would require a $m_F = +3$ state which is not permitted. Had pumping to the $5^2P_{3/2}$ state not been prohibited, in addition to the mixing of the magnetic sublevels, absorption of D_2 radiation by the ground substates would cause a decrease in the polarization of the sample and thus reduce the efficiency of the technique.

Except for the $F = 2$, $m_F = +2$, a nonabsorbing level, all the remaining $4I + 1$ m_F states of the $5^2S_{1/2}$ level for rubidium 87 may absorb the circularly polarized resonance radiation. W. Franzen and A. G. Emslie have reported:

If the pumping radiation is sufficiently intense and the relaxation time is sufficiently long, the entire population will be found in the single nonabsorbing $m_F = +2$ sublevel of the ground state. This condition corresponds to complete orientation of the alkali atoms, with the nuclear and electron spins both aligned along the axes of quantization.⁶

Occupation probability of state k. It has been shown that the occupation probability $p_k(t)$ of an atom in the k^{th} state may be obtained by a solution of eight simultaneous differential equations of the form, for rubidium 87 only, of

$$\dot{p}_k = - \sum_{j=1}^8 (b_{kj} + w_{kj}) p_k + \sum_{i=1}^8 (b_{ik} + w_{ik}) p_i$$

$k = 1, 2, \dots, 8$, where b_{ij} is the probability per unit time that an atom in substate i of the ground $5^2S_{1/2}$ state has undergone a transition to a substate j of the $5^2S_{1/2}$ state by absorption and re-emission of a photon, and w_{ij} is the probability per unit time of the relaxation occurring between states i and j . The diagonal terms, $j = k$ and $i = k$, represent the transition of an atom in the k^{th} state of the ground state to an excited state and then back to the k^{th} state giving no change in the net occupational probability, hence diagonal terms should be omitted from the sum.

Absorption probability of state k. Those terms of the preceding sum that affect the total absorption

probability, assuming that the occupational probabilities of the levels are equal do not include the diagonal term transitions because of the discriminatory nature of the circularly polarized incident radiation. Since the absorption probability per unit time has been given as $\sum_j b_{kj}$ for an atom in the k^{th} state, the total absorption probability of all states will be $\sum_k \sum_j b_{kj} p_k$. Since $p_k = 1/8$, for rubidium 87, the average absorption probability per atom per unit time is $A_{kj} = 1/8 \sum_{kj} b_{kj}$.

Relaxation probability of state k . The collision of a rubidium atom with another rubidium atom, causing an exchange of electrons with opposite spin, and the collision of an atom with the wall of the container, producing a reorientation are the causes of the relaxation. For rubidium 87, atoms that are in the $F = 2$, $m_F = +2$ state there is equal probability of transition to any of the other seven magnetic substates. Thus the probability that relaxation will occur is given by $1/8 T$ where 8 is the number of possible substates and T is the relaxation time.

CHAPTER III

SPIN EXCHANGE COLLISIONS AND RELAXATION

When the pumping radiation passing through the absorption cell is abruptly cut off, the rubidium vapor begins to lose its polarization due to collisions with the cell walls and with other gas atoms. An inert gas, such as neon, has little tendency to produce depolarization during collisions and may be used as a "buffer gas" whose function is to increase the time required by the rubidium atoms to diffuse to the walls of the chamber.

I. ATOMIC POPULATION

Number of available atoms. The chemical definition for the heat of dissociation (dissociation energy), D , of a diatomic molecule AB is the work (ev) required to dissociate the molecule from the lowest level of the electronic ground state into two distinct normal atoms, $A + B$. This energy has been shown⁷ to be $.46 \pm .05$ ev for Rb_2 . A calculation of the translational kinetic energy for a rubidium atom gives, $KE = \frac{3}{2}kt = .0417$ ev at $323^\circ K$. By using the bond dissociation energy one can determine how many of the particles are atoms and

how many remain in the molecular state. By letting $x_0 = kT/D$, then for the rubidium vapor in this problem,

$$f = 1 + \frac{2}{\sqrt{\pi}} x_0 e^{-x_0^2} - \frac{2}{\sqrt{\pi}} \int_0^{x_0} e^{-x^2} dx = 1$$

With $f = 1$, then fifty per cent of all the particles have energies sufficient to become dissociated into atoms. By taking Avogadro's number, N_0 , as the number of molecules per gram-mole then the number density of an ideal gas at standard temperature and pressure is:

$$\rho = \frac{N_0}{V} = \frac{6.023 \times 10^{23}}{22.4 \times 10^3} = 2.7 \times 10^{19} \frac{\text{particles}}{\text{gram-cm}^3}$$

The vapor pressure, given in Chapter I, is 1.5×10^{-5} cm, at 300°K , so that the particle density inside the absorption cell, at this specific vapor pressure is $n = 2.7 \times 10^{19} \frac{(1.5 \times 10^{-5})}{76} = 5.3 \times 10^{12}$ particles. Since it has been determined that two-thirds of these particles are atoms and one-third are diatomic molecules, there are 5.3×10^{12} atoms/cm³ and 2.65×10^{12} molecules/cm³.

Absorption by vibrational molecular modes. By elementary mechanics, a particle of mass μ , acted on by a linear restoring force, $-kx$, vibrates with a frequency

$$\nu = \frac{1}{2\pi} \sqrt{\frac{k}{\mu}}$$

Thus the frequency of oscillation for a diatomic molecule is $\nu = \frac{1}{2\pi} \sqrt{\frac{2\bar{a}^2 D}{\mu}}$ where the propor-

tionality constant $k = 2\bar{a}^2 D$ has been expanded in a Taylor series of $(r-r_e)$, the interatomic displacement. ⁸

Since the characteristic temperature of vibration is

$\theta_v = \frac{h}{k} \nu$ then $\theta_v = \frac{h}{2\pi\mu} \sqrt{\frac{2a^2D}{\hbar^2}}$ where D is the dissociation energy and a is a constant determined by the curvature about the minimum of the potential curve. Applying the values of h , k , μ , and $\theta_v = 100^\circ\text{K}$, which is the characteristic temperature needed for vibrational energy level excitation, it can readily be seen that the wavelength $\lambda = \frac{c}{\nu} = \frac{ch}{k\theta_v} = 1,430,000 \text{ \AA}$ is located in the extreme part of the infra-red and hence has no effect on a S-1 (8000 \AA) spectral response detector tube, and the presence of a large number of rubidium diatomic molecules in the sample should have no effect on the atomic system and may be ignored.

II. SPIN EXCHANGE COLLISIONS

There are three kinds of depolarizing collisions that may occur, (1) a rubidium atom may collide with the absorption cell wall and may be assumed to rebound from the wall with an equal probability of being in any of the eight magnetic substates of the ground state, thus contributing no net polarization: (2) or the collision of a rubidium atom with an inert buffer gas atom causing very little, if any, depolarization because of the spherical symmetry of the inert gas atoms: (3) or a rubidium atom may collide with another rubidium atom and exchange electron-spin coordinates with it.

In this third type of depolarization, L. W. Anderson and A. T. Ramsey⁹ of the University of Wisconsin have found that the polarization of the sample is not destroyed by the spin-exchange collisions but instead is merely redistributed because the z component of the angular momentum is a constant of the motion. Because spin-exchange collisions with buffer gas atoms have small probability and the buffer atoms inhibit diffusion to the walls, the addition of an inert gas increases the relaxation time of the rubidium atom. The amount of increase is dependent on the buffer gas pressure. Figure 6 shows the relationship between the relaxation time of rubidium and five different buffer gases, as determined by W. Franzen.¹⁰ He has interpreted the results such that at lower buffer-gas pressures the buffer-gas inhibited the diffusion of rubidium to the wall of the container, thus causing a rise in the curve. The decrease of the curve at higher buffer-gas pressures is due to disorienting collisions of the rubidium with the buffer-gas.

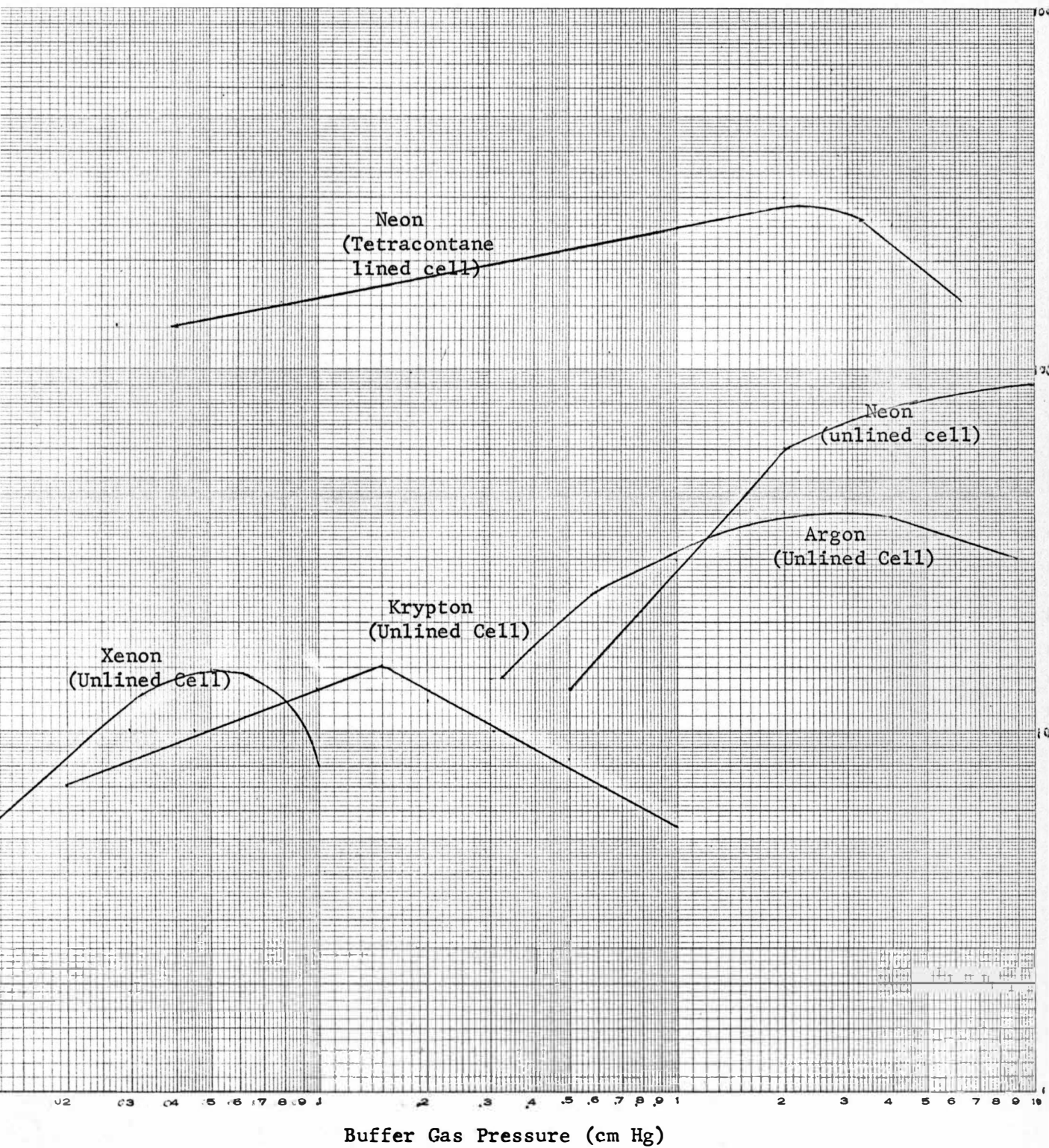
III. RELAXATION PROCESS

Spin-flip reorientation. The Hamiltonian describing the spin-flip transition probability of an alkali atom's outer s ($l = 0$) electron has been computed

using the application of first-order time-dependent perturbation theory by R. A. Bernheim¹¹ of Columbia University, and has been shown to be $H_{\text{eff}} = \sum_j \gamma_j s_j J$

where J is the total angular momentum of the colliding pair of atoms about their center of mass, s_j is the spin of electron j and γ_j is a constant which may be evaluated by using the proportionality theorems used by Van Vleck¹² in his treatment of coupling of angular momentum in molecules. Since it has been assumed that the disorientation arises from a spin-flip of the rubidium atom's single outer electron during collision, the total electronic angular momentum, J , becomes the common factor associated with the disorientation and relaxation processes.

Fig. 6. Rubidium vapor spin relaxation time as a function of buffer gas pressure under various conditions.



CHAPTER IV

EXPERIMENTAL APPARATUS

The experimental apparatus used in this problem is shown in block diagram form in Figure 7. Resonance radiation was directed through a circular polarizing filter and a D_1 line filter onto the surface of an absorption cell containing rubidium vapor. The transmitted light was then detected by a photocell whose output signal was directed into a low noise, d-c coupled cathode follower (Appendix A) and shown on an oscilloscope.

The excitation lamp. A small electrodeless bulb containing rubidium and argon was inserted in the coil loop of a radio frequency oscillator. The oscillator was designed to produce fifty watts of r-f power at one hundred megacycles. It contained a 829B dual pentode transmitting tube connected to a parallel resonance line with a single loop in the end. The physical advantage of the single loop is that there is a small voltage drop across the loop end of the line with large current surges producing magnetic fields around the bulb, thus increasing the stability of the light source.

A d-c connected heating coil placed inside the

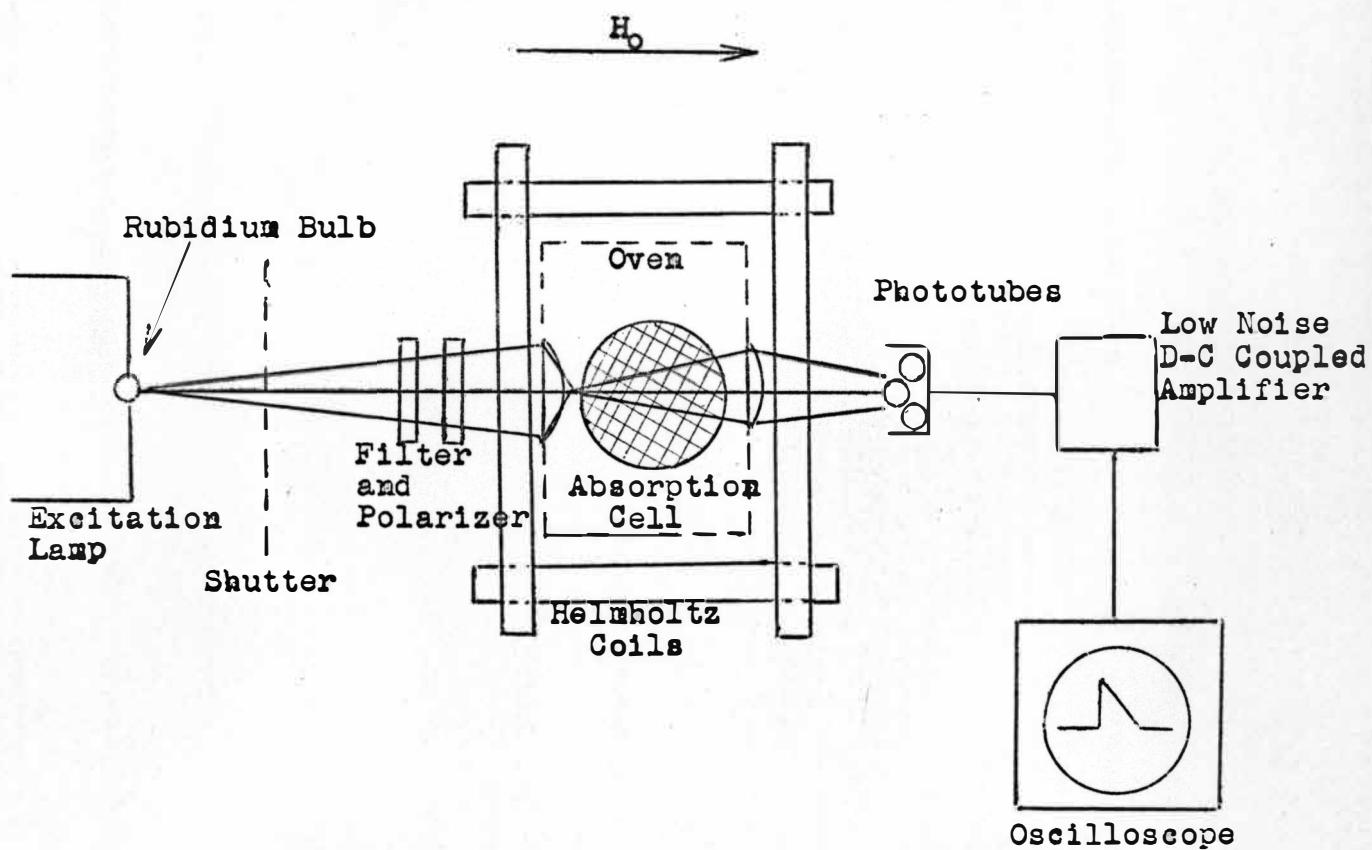


FIGURE 7

THE BLOCK DIAGRAM OF THE
EXPERIMENTAL APPARATUS

bulb chamber provided regulated heating of the rubidium bulb. The optimum temperature giving the best results was found to be $170^{\circ} \pm 20^{\circ}\text{C}$. The operation of the lamp outside the specified temperature limits was found to cause weaker light intensities and increased light fluctuations.

The shutter. A **Packard** No. 6 shutter, purchased from Michigan Photo Shutter Company, Kalamazoo, Michigan, was used to stop the light flow and to allow the rubidium atoms to decay in the dark. The shutter has a four inch diameter and was connected by a thin wire to a solenoid placed a distance of four feet away.

Filters. Two distinct filters were used: a D_1 line filter which permitted D_1 radiation to pass through but filtered out the D_2 (7500 Å) line, and a circular polarizing filter. The circular polarizer was purchased from the Polaroid Corporation, Cambridge, Massachusetts. It consists of a polaroid HN7 plastic laminated linear polarizer and a $210 \pm 20 \text{ m}\mu$ retardation plate. Maximum circular polarization was achieved by allowing red light to pass through the linear polarizer and retardation plate and fall on the face of a highly reflecting metal sheet. The quarter wave retardation plate was then rotated with respect to the polarizer until the minimum

amount of reflected light came back through the circular polarizing filter.

Absorption cell. The absorption cell was spherical and made out of Pyrex glass. It contained both the neon buffer gas and the rubidium vapor. The neon was obtained in a spectroscopically pure grade from the Linde Air Products Company. The absorption cell was housed in an oven approximately one cubic foot in volume. The oven was made from a light asbestos pressboard, Martinite. To eliminate reflected light transmission, the inside of the oven was sprayed with black paint. The oven temperature was regulated by two electrically heated coils. A d-c source provided the power for the heating elements and reduced sixty cycle noise that would otherwise be introduced.

Helmholtz Coils. The absorption cell was located at the center of two pairs of orthogonal Helmholtz coils, as shown in Figure 8.

The horizontal pair of coils was used to compensate for the vertical component of the earth's magnetic field. Since the dip angle of the earth's field is of the order of 73° in Kalamazoo, Michigan, where this experiment was performed, the earth's field is nearly vertical. A dip needle placed at the center of the system determined

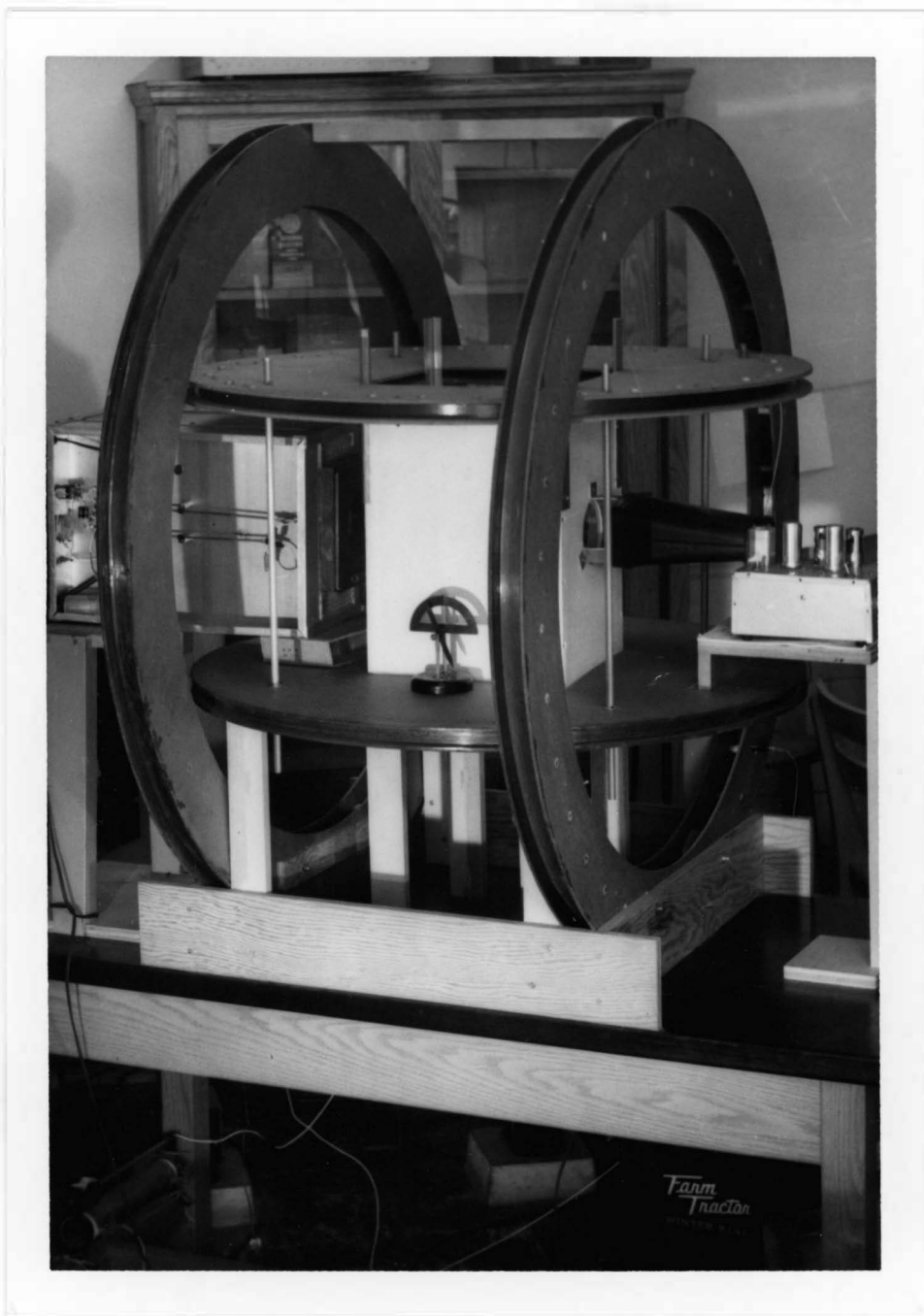


FIGURE 8

TWO PAIR OF ORTHOGONAL
HELMHOLTZ COILS

the value of the current needed through the coils to balance out the vertical component of the field. It has been shown¹⁰ that if the vertical component of the earth's field was not compensated for less efficient pumping alignment of the vapor would take place and the light intensity would return to the value of the unpolarized state when the shutter was opened.

The vertical pair of coils provided a static external magnetic field, \bar{H}_0 , through the absorption cell. The magnitude of which may be determined from: $H_0 = \frac{4\pi N I r^2}{10 (\frac{r^2}{4} + r^2)}^{3/2}$

where $N = 298$ turns and $r = 58.5$ centimeters. Applying the values for N and r , $H_0 = 7.3$ I.

The axis of the system was directed along a magnetic meridian of the earth's field inside the room.

Radio frequency field. The method used for the initial determination of the alignment was that of Zeeman transition resonance. A radio-frequency field was produced by a circular coil, four inches in diameter, placed around the absorption cell and orientated at right angles to H_0 . The radio-frequency was chopped at a rate of 15 cycles per second.

The r-f field produced by the coil was produced by a Type 1211-Unit Oscillator which was purchased from General Radio Company, West Concord, Massachusetts.

Detection of the signal. The light, after passing through the absorption cell, is monitored by three 918 phototubes whose current is proportional to the transmitted light. The interruption of the light by the shutter causes the photocurrent to drop to a lower value. When the shutter is opened the photocurrent rises to a value that is proportional to the transmitted light. This value will be somewhat less than the steady state condition optical pumping provides. The radiation then pumps the sample back to the steady state condition. The signal voltage is sensed by a cathode follower whose output matches the cable impedance. The signal was d-c coupled and displayed on the face of a Tektronix 503 oscilloscope.

Oscilloscope reproductions. Permanent images of oscilloscope tracings were obtained with the aid of a Dumont Type 271-A oscillograph-record camera. This camera, especially designed for obtaining oscilloscope pictures, was attached to a light hood which was clamped on the oscilloscope face. A lens speed of $f/3.5$ was used with a manual shutter. The film used was Panatomic X developed in Kodak D-76.

CHAPTER V

DATA, CALCULATIONS AND CONCLUSION

One of the fundamental calculations necessary to the unification of this problem was that of determining the values of the resonance frequencies for Zeeman transitions of rubidium 85 and rubidium 87.

Determination of the Zeeman transition frequencies.

In a weak magnetic field the spacing between the different magnetic states is given by the Landé splitting factor

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

which may be approximated as $g_F \hat{=} g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)}$

since $g_I \hat{=} \frac{1}{2000} g_F$. For $J = 1/2$, $F = 2$, $g_F = g_J \frac{1}{2I + 1}$

and the energy is given¹³ as $E = g_F \mu_0 H_0$ or the frequency

as $\nu = \frac{g_F \mu_0 H_0}{h}$, where μ_0 is one Bohr magneton, H_0 is

the external field and h is Planck's constant.

For rubidium 87, $g_J = 2$, $g_F = 1/2$,

$$\nu = \frac{1/2(9.273 \times 10^{-21})}{6.6 \times 10^{-27}} (H_0) = 7.04 \times 10^5 \times 1.82 =$$

1.28mc, where $H_0 = 7.3 I = 1.82$ gauss for $I = .25$ amperes.

For rubidium 85, $g_F = 1/3$, $\nu_{85} = 2/3 \nu_{87} = 850$ kc, which is the frequency of the Zeeman transitions. Experimentally

ν_{85} was found to be 525kc and $\nu_{87} = 825\text{kc}$. The difference in the values is within the precision of the ammeter.

The natural resonance frequencies for the hyperfine transitions have been found¹³ to be $\nu_{87} = 6834\text{mc}$ and $\nu_{85} = 3035\text{ mc}$.

Bloch equations. There are several methods for expressing the relaxation times in both nuclei in solids and atoms in a vapor. One of these methods was proposed by Felix Bloch in 1946. He derived a set of phenomenological equations which described the behavior of interacting nuclear paramagnets.¹⁴ Into these equations two relaxation times were introduced, T_1 representing the spin-lattice interaction and T_2 representing the spin-spin interactions. The underlying physical consideration is that when nonequilibrium of magnetic substates exists, the magnetization vector, \vec{M} , which is the magnetic moment per unit volume, may tend to precess around the external static magnetic field vector with an angular frequency $\bar{\omega}_L = \gamma \hbar B$. The relaxation time, T_1 , causes the nonequilibrium magnetization vector to tend exponentially in time toward a thermal equilibrium state M_0 . The z-component of \vec{M} will grow toward M_z directed along the z axis and with a magnitude determined by the random distribution of spins as given by the Boltzmann distribution function. The result is the Bloch

equation:

$$\frac{d\bar{M}}{dt} = (\bar{H} \times \bar{M}) - \left[\frac{(M_x + M_y)}{T_2} + \frac{(M_z - M_0)}{T_1} \right]$$

where M_0 is the magnetization vector at thermal equilibrium.

I. MEASUREMENT OF THE SPIN DEPHASING TIME T_2'

The mechanisms producing T_2' may most readily be understood if one considers the following argument.

When the rubidium atoms in the absorption cell are optically aligned to an external static field, the bulk magnetization vector of the magnetic moments of the atoms will be pointing along the direction of the external field vector \bar{H}_0 . If an r-f field, H_1 , is applied perpendicular to \bar{H}_0 , the bulk magnetization vector, \bar{M} , will precess around \bar{H}_1 . When \bar{M} is parallel to the direction of the light it corresponds to a polarized state $m_F = +2$ and cannot absorb radiation, but when \bar{M} precesses through an angle of 180° it is anti-parallel to the light flow (and \bar{H}_0) which corresponds to a radiation absorption condition of $m_F = -2$. Thus small sinusoidal fluctuations in the optical transparency of the vapor would be produced as \bar{M} periodically precesses around \bar{H}_1 . But constant amplitude fluctuation is a hypothetical situation since it assumes homogeneity of \bar{H}_1 through out the entire

absorption cell. Experimentally it was assumed that \bar{M} precesses around H_1 with a Larmor frequency $\omega_1 = \gamma H_1$. If there exists a field inhomogeneity, ΔH_1 , then a corresponding $\Delta \omega_1$ will occur and \bar{M} will tend to "fan out" (dephase) in a cone around it. The loss of phase coherence in the components of \bar{M} , due to the field inhomogeneity of H_1 , causes the sinusoidal representation of \bar{M} to decay. (Figure 9) The characteristic time of decay is T_2 .

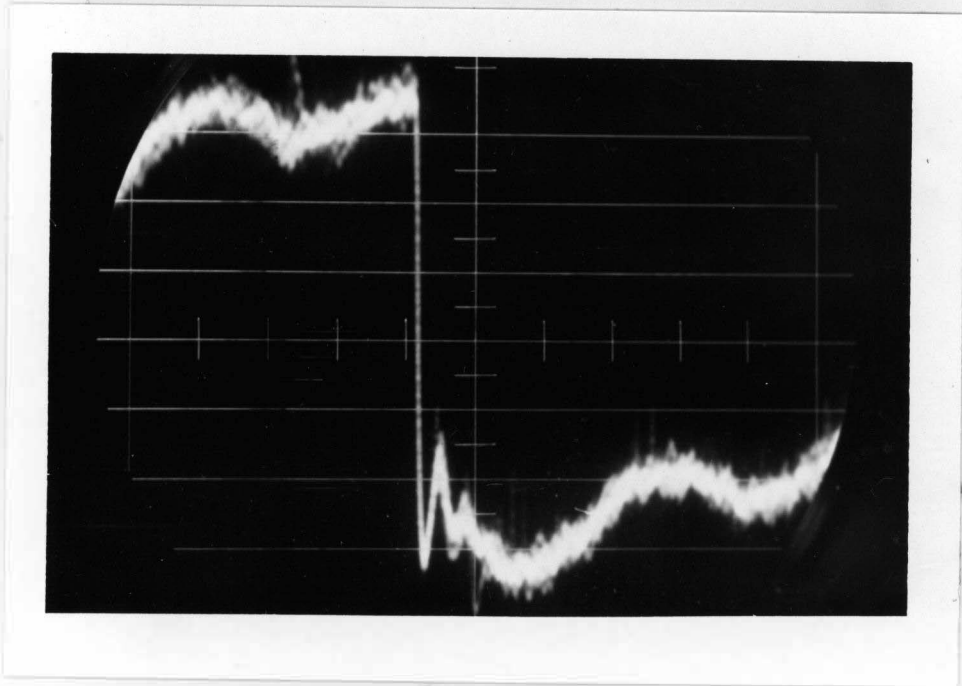


FIGURE 9

THE TRANSIENT MODULATION OF THE OPACITY OF AN OPTICALLY PUMPED GAS CELL WHEN A RESONANT R-F FIELD IS APPLIED TO THE BULB.

Since the decay curve of T_2 is exponential, it can be represented mathematically by $I = I_0 e^{-t/T_2}$, where I is the intensity (amplitude) and t is the time.

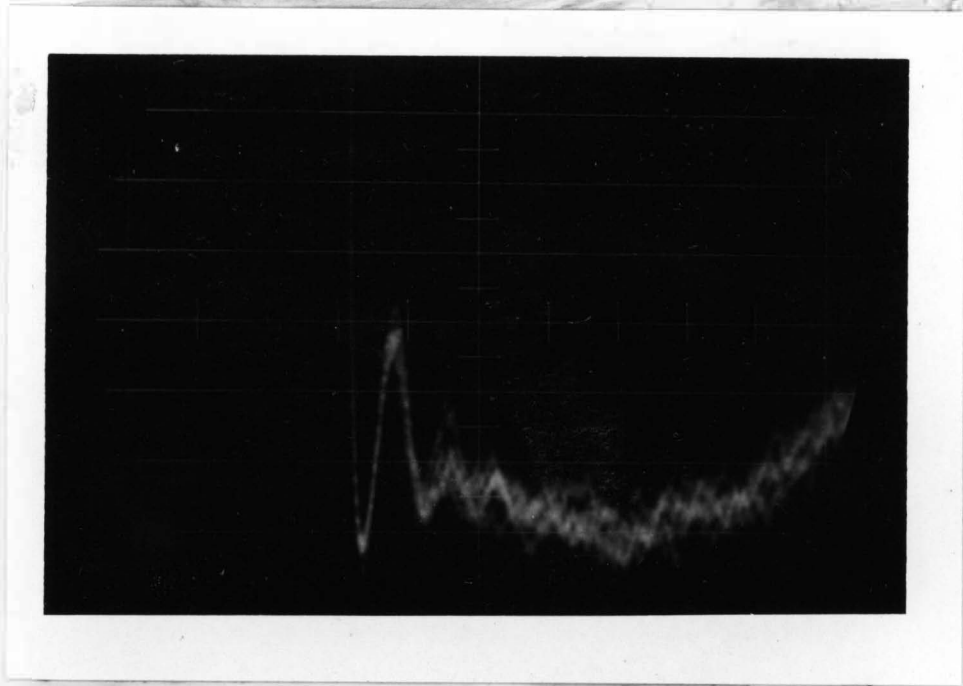


FIGURE 10

TIME INCREASES FROM LEFT TO RIGHT IN UNITS OF 25 msec/div.
THE PHOTOCELL OUTPUT INCREASES FROM BOTTOM TO TOP IN UNITS
OF 1 mV/div. CELL TEMPERATURE WAS 33°C.

The logarithm (base e) of the average decrement of the decay is given by $\bar{\delta} = \ln \left[\left(\frac{1}{3} (Y_1/Y_2 + Y_2/Y_3 + Y_3/Y_4) \right) \right]$. The spin dephasing time, T_2' , is then given by $T_2' = t^*/\bar{\delta}$ where t^* is the average time interval of the logarithmic decrements. With $t^* = 75$ msec, T_2' was found to be 82 msec.

II. MEASUREMENT OF THE SPIN-LATTICE RELAXATION TIME T_1

A family of curves showing the decay of the absorption cell transparency due to polarization decay is presented in Figure 11. The curve connecting the "recovery" traces represents the transparency of the rubidium vapor as function of the time the shutter is closed. This curve is exponential and is assumed to be proportional to the decay of the sample.

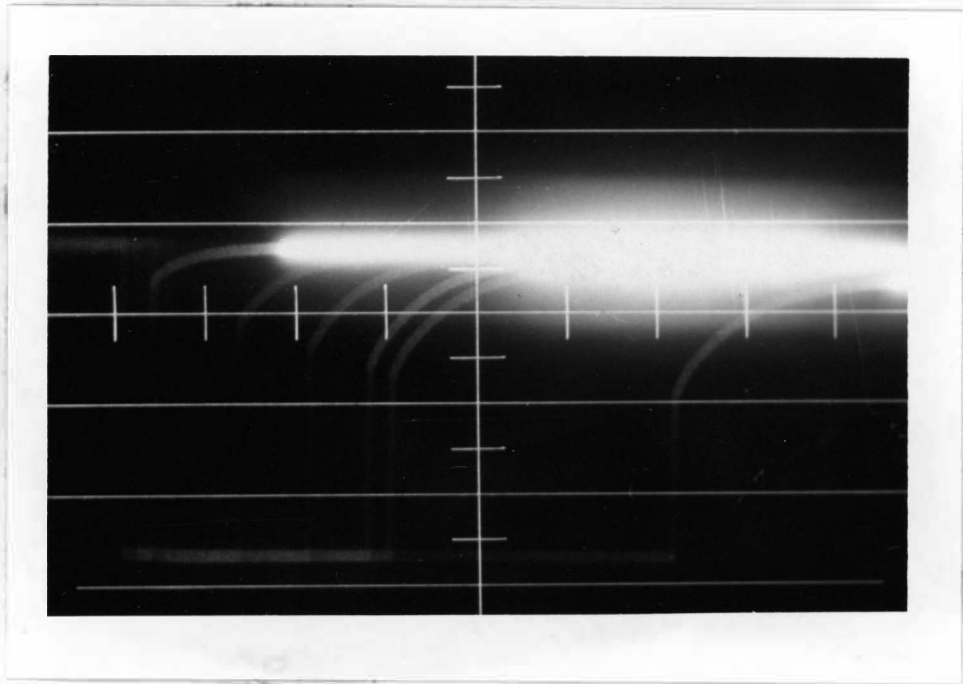


FIGURE 11.

THE DECAY CURVE OF THE RUBIDIUM VAPOR TRANSPARENCY. THIS PHOTOGRAPH WAS TAKEN UNDER THE FOLLOWING CONDITIONS:
 BUFFER GAS, NEON AT A PRESSURE OF .5cm OF HG: CELL
 TEMPERATURE, 55°C, OSCILLOSCOPE SWEEP USED, 500 msec/
 SCALE DIVISION

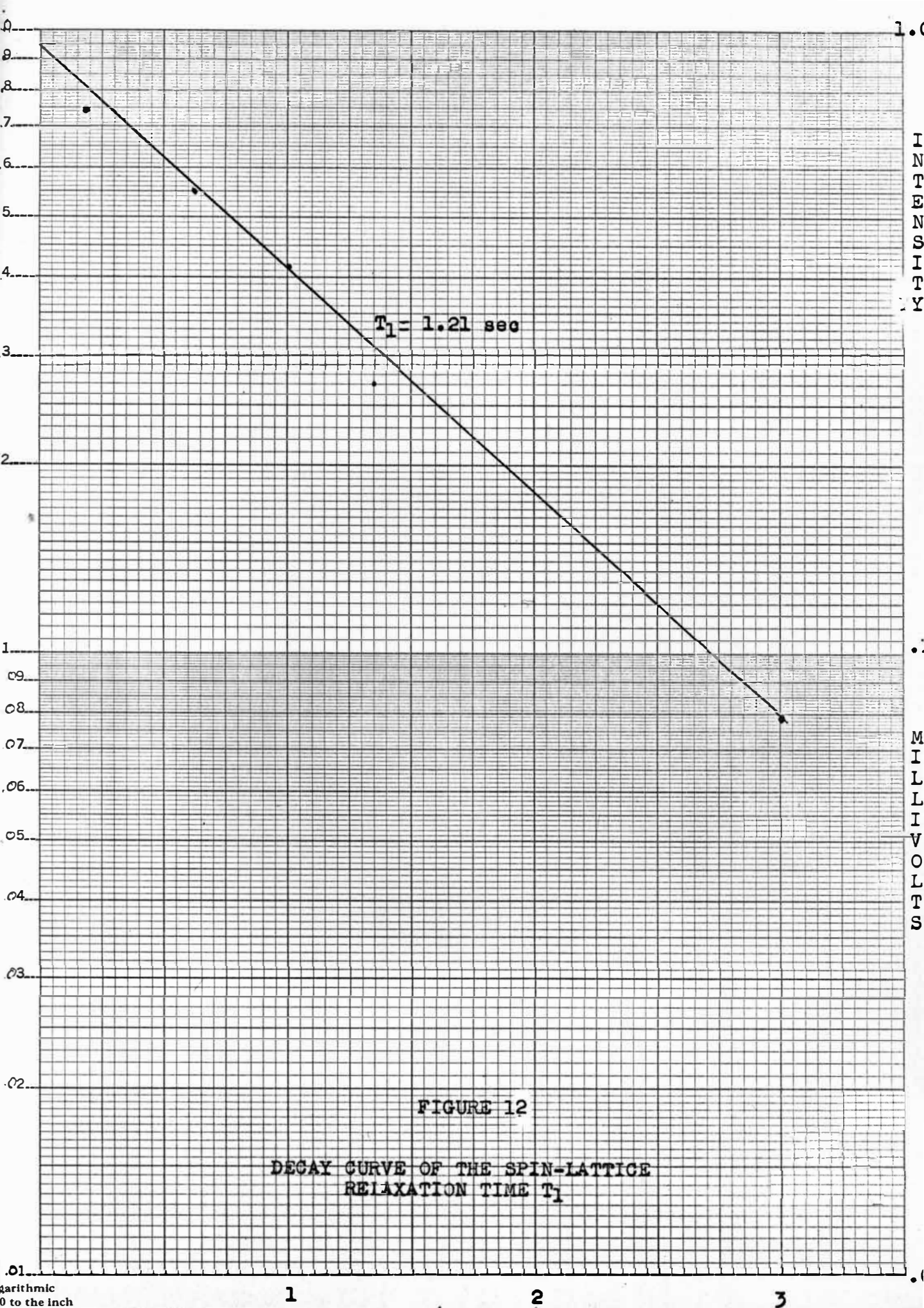
The decrease in the transparency of the cell is assumed to follow the exponential law $I = I_0(1 - e^{-\lambda t})$, where I is the decrease in the transparency of the cell and I_0 is the decrease in the transparency of the cell from the steady-state value to the limiting value which the curve approaches asymptotically. Figure 12 shows a plot of $\ln_0 \frac{(I_0 - I)}{I_0}$ verses time. The slope of this line is the decay constant λ . The average relaxation time, T_1 , is $T_1 = 1/\lambda$. At $t = 0$, $\frac{(I_0 - I)}{I_0} \neq 1$. This discrepancy is probably due to the loss in time accumulated in opening and closing the shutter. The measured value of the relaxation time, T_1 , using neon as a buffer gas, at a temperature of 55°C (which corresponds to a vapor pressure of 3×10^{-6} mm of Hg) was found to be 1.21 sec.

III. CONCLUSION

The object of this study was to determine the relaxation times of a rubidium vapor using neon as a buffer gas. The spin-lattice relaxation time T_1 , was found to be 1.21 sec at 55°C .

Proposed projects for future work include the determination of the spin-lattice relaxation time under various temperatures and with various densities. The determination of T_1 using other buffer gases has also

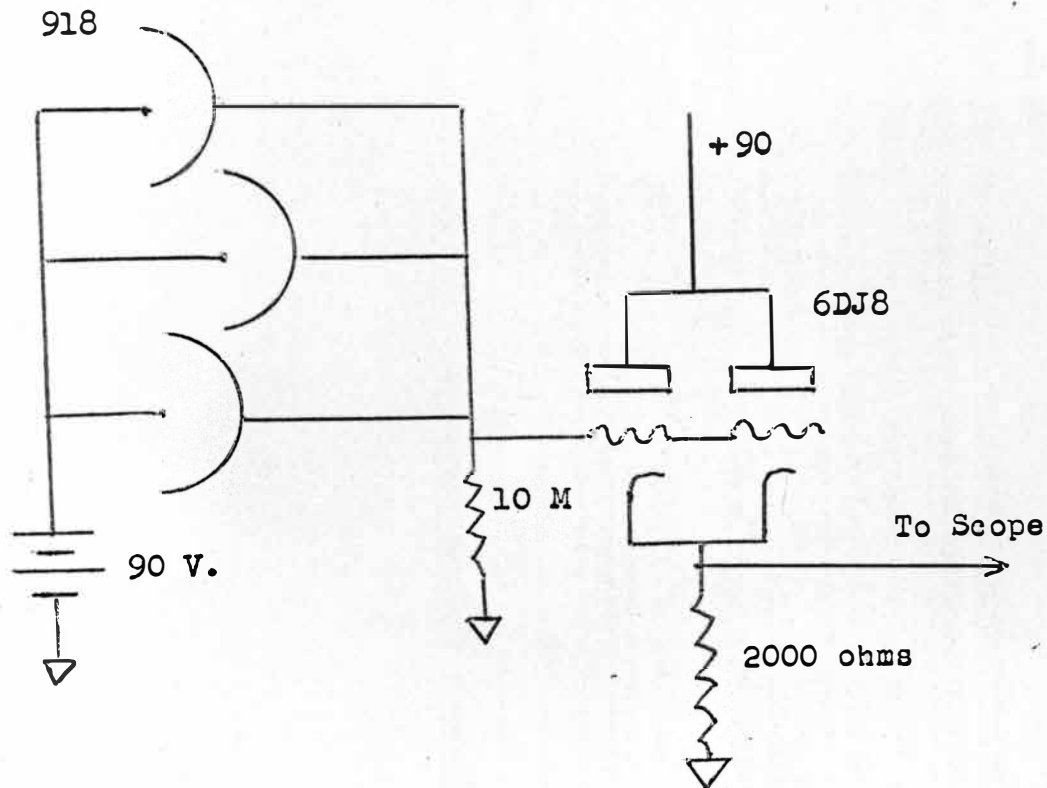
been suggested. By varying the experimental conditions the frequency of different types of collisions leading to relaxations may be controlled and the disorienting interactions may then be studied independently.



APPENDIX

APPENDIX A

The circuit diagram for the phototube detector used in this problem is shown below. It consists of three 918 phototubes and a d-c coupled cathode follower.



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