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Spin relaxation of optically pumped rubidium.

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SPIN RELAXATION OF OPTICALLY PUMPED RUBIDIUM

BY

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A

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ABSTRACT

This Thesis is intended to be a self-contained guide to the theory and experimental procedures of optical pumping in one of its most usual applications. A particular study is made of optical pumping of rubidium vapor and results are given stating the most efficient temperature range in which to produce large populations of optically oriented rubidium atoms. The thermal relaxation time for optically orientated rubidium for a particular system is given and the effect of trapped resonance radiation is considered in the interpretation of the experimentally obtained curves.

ACKNOWLEDGEMENTS

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I. INTRODUCTION

In the past fifteen to twenty years, rapid strides have been made in the field of resonance phenomena. Electronic techniques have long been employed in the field of spectroscopy, but especially since World War II, when the physicist was forced to improve his techniques, research in the fields of resonance spectroscopy and resonant interactions, as examined by approximately coherent electromagnetic waves, have led to new techniques.

Considerable attention has been given to the interaction of resonant radiation with atoms aligned by a magnetic field. The experiments were fundamental in that they experimentally demonstrated the validity of selection rules for electron transitions and, in addition, provided an effective method of studying the Zeeman effect for hyperfine structures.

The experiment performed here was a partial duplication of H. G. Dehmelt's experiment, "Slow Spin Relaxation of Optically Polarized Sodium Atoms."[1] Rubidium was substituted for sodium and the relaxation was studied as a function of:

(1) Intensity of the exciting light source

(2) Temperature of the rubidium resonance cell.

From this information a qualitative understanding was gained of the optical pumping and relaxation processes and quantitative information was obtained for these processes in the particular system used in this experiment.

The rubidium resonance cell used was a half liter spherical flask that contained a very small vapor pressure of rubidium and a relatively large pressure of argon serving as a buffer gas. With this arrangement, it was possible to minimize rubidium - rubidium and rubidium-cell wall collisions to such a degree that the slow spin relaxation of the optically polarized rubidium atoms could be studied and accurate relaxation times of this process could be determined.

The object of this work was to determine relaxation times as a function of the above mentioned variables and to understand the mechanisms which permitted this process to take place.

II. REVIEW OF THE LITERATURE

Interest was created in spin relaxation processes when Whittke and Dicke[2] performed some experiments dealing with paramagnetic resonance absorption of free atoms diffusing in an inert gas.

Dehmelt[1] studied sodium atoms diffusing in argon gas, where relaxation due to sodium-sodium collisions was minimized by employing very low sodium pressures (about 10^{-7} mm of Hg). He used argon pressures from 1 to 40 cm. of Hg. In carrying out the experiments, optical pumping by circularly polarized resonance radiation was used to create an orientation of the sodium atoms which then was monitored by measuring the transmission of the pumping radiation through the sample. By suddenly reversing a small axial magnetic field, the polarization of the atoms could be made to reverse too. From the decay rates of this inverted polarization, Dehmelt found that the relaxation times found for his samples were significantly shorter than the average wall diffusion times. Dehmelt suggested that this shortening of the relaxation time could be explained by considering sodium-argon collisions.

Bell and Bloom,[3] in a paper that discussed two rf magnetic resonance experiments, discussed the optical pumping and relaxation processes and presented several simplifying assumptions that they had used in their calculations.

W. Franzen[4] reported a new optical pumping method for studying the spin relaxation process. In this method, the pumping radiation, consisting of circularly polarized D_1 resonance radiation, is suddenly turned off and then turned on again after a known time interval. The

relaxation which took place during the interval of darkness caused the rubidium vapor to be more opaque to the pumping radiation, at a rate which was determined by the relaxation time. From his work, Franzen was able to calculate deorientation cross sections between aligned rubidium atoms in the ground state and neon, argon, krypton, and xenon. He also was able to deduce the diffusion coefficients for rubidium in neon and argon.

III. DISCUSSION OF PRINCIPLES

This experiment dealt with the Zeeman effect for hyperfine structure in weak fields. The weak field case simply implied that the external field used was much smaller than the atomic field and thus the external field was weak enough to preserve the coupling between I, the nuclear spin in units of $\hbar$, and J, the total angular momentum of the extranuclear electrons in units of $\hbar$, and yet produced sufficient energy level splitting so that degeneracy was removed.

In the discussion to follow, a non-rigorous quantum mechanical approach was taken as the physics of the problem tended to be clarified by such a treatment. A large portion of the material used in this discussion was found in a more complete fashion in the references.[5,6]

It will be advantageous to summarize some of the results that have been found to be useful for the case under consideration. Since the experiment was concerned with the Zeeman effect for hyperfine structure, consideration was given to the spectral notation that was used to denote the various energy states of an atom, as well as, the selection rules that applied for the various energy transitions of an atom.

The quantum number L is associated with the resultant orbital angular momentum of the electrons and the quantum number S is associated with the resultant spin of the electrons surrounding an atom. Then in order to obtain the total angular momentum vector J of the extranuclear electrons, one must add vectorially to L an additional integral or half integral spin vector S. According to quantum theory, L and S cannot be oriented with respect to each other in any arbitrary

direction. The vector addition of L and S is such that the different possible values of their vector sum have integral differences. Therefore:

$$J = (L + S), (L + S - 1), \cdots, |L - S|$$

Now in the same fashion that L and S are combined to give the total angular momentum J of the extranuclear electrons, J and I must similarly be combined to give a resultant F, which is the total angular momentum of the whole atom. As with L and S, the quantum number F can take the values:

$$F = (J + I), (J + I - 1), \cdots, |J - I|$$

It is because of the magnetic moment of the nucleus that a coupling between J and I results and produces a precession about the total angular momentum vector F. Figure 1 is an illustration of the precession of angular momentum vectors about the total angular momentum F. The solid line ellipse shows the precession of I and J about F. The dotted-line ellipse shows the much faster precession of L and S about J taking place at the same time.[5]

Due to this precession, a small energy difference between states with different F exists. Since the magnetic moment of the nucleus is about 2000 times smaller than that of the electrons, the precession of I and J about F is about 2000 times slower than that of L and S about J and correspondingly the energy differences are this much smaller. It is these small differences that are observed in the hyperfine structure of the spectral lines.

In a magnetic field, a space quantization of F takes place. The quantum number M_F of the component of the angular momentum in the field direction can take only the following values:

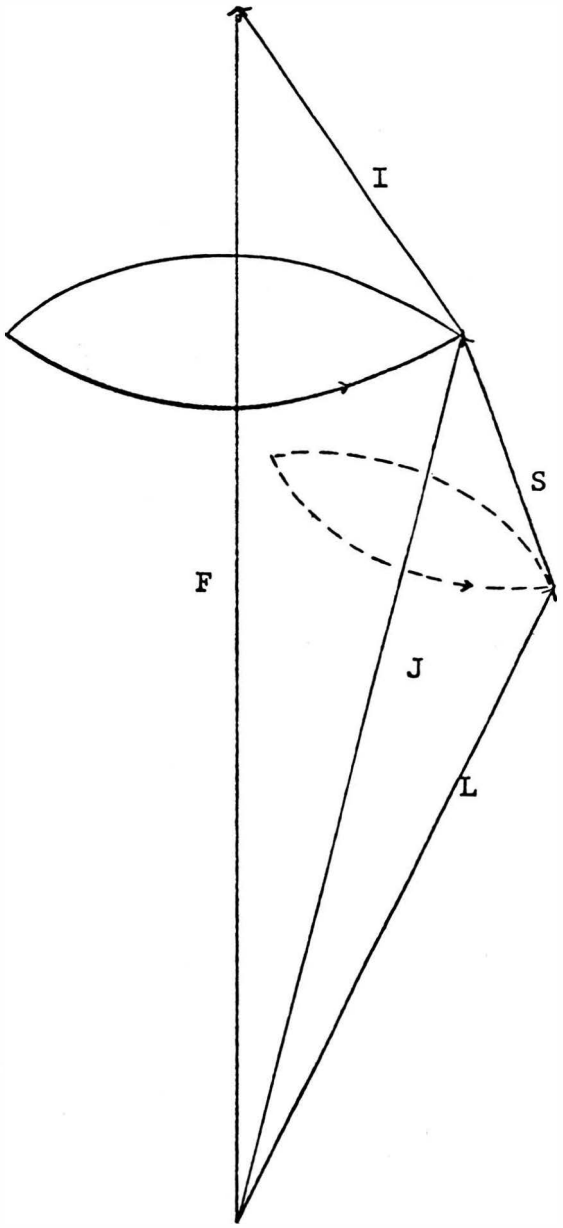


Figure 1

$$M_F = F, F - 1, F - 2, \cdots, - F$$

where the $2F + 1$ values of M_F correspond to states of different energies in a magnetic field.

In the process of absorption or spontaneous emission of photons of resonance radiation, the following transition rules hold for atoms changing energy states:

$$\Delta L = \pm 1, \Delta J = 0, \pm 1, \Delta F = 0, \pm 1 \text{ (but not } F = 0 \rightarrow F = 0)$$

$$\text{and } \Delta M_F = 0, \pm 1 \text{ (but not } M_F = 0 \rightarrow M_F = 0 \text{ for } \Delta F = 0)$$

It is now useful to consider spectroscopic notation that will be used later in this discussion. The energy state of an atom is often denoted by a term symbol that is developed as follows: The letters S, P, D, F, correspond to resultant orbital angular momentum of 0, 1, 2, 3 respectively. A prefixed superscript, the so called multiplicity, is determined by the formula $2S + 1$, where S is the resultant electron spin. A subscript, the quantum number J, is added and the term is defined. As an example of this notation, the ground state of rubidium has an electron spin equal to 1/2, the L quantum number equals 0, and J equals 1/2. Thus in spectral notation, this energy state would be the $^2S_{1/2}$ term (read the doublet S one half term).

Now that some of the fundamentals applying to spectra have been reviewed, a look at some of the particular properties of rubidium would be in order. Naturally occurring rubidium is composed of two isotopes, Rb 85 whose approximate abundance is 72% of all the rubidium, and Rb 87 which makes up the remaining 28%. Rb 85 has a nuclear spin $I = 5/2$, while the naturally occurring radioactive isotope, Rb 87, has a nuclear spin $I = 3/2$.

As mentioned above, for both isotopes in the ground state, the rubidium atoms are in the $^2S_{1/2}$ energy state. Now the allowed excitation transitions are those where L and J change by $\Delta L = 1$, $\Delta J = 0, +1$ (first excited state), so for the first excited state one has: $S = 1/2$, $L = 1$, $J = 3/2$ or $1/2$. In spectral notation the terms $^2P_{3/2}$ and $^2P_{1/2}$ correspond to the first excited states. Transitions from these excited states to the ground state are responsible for the two resonance lines (D - lines) in the rubidium spectrum. (Fig. 2)

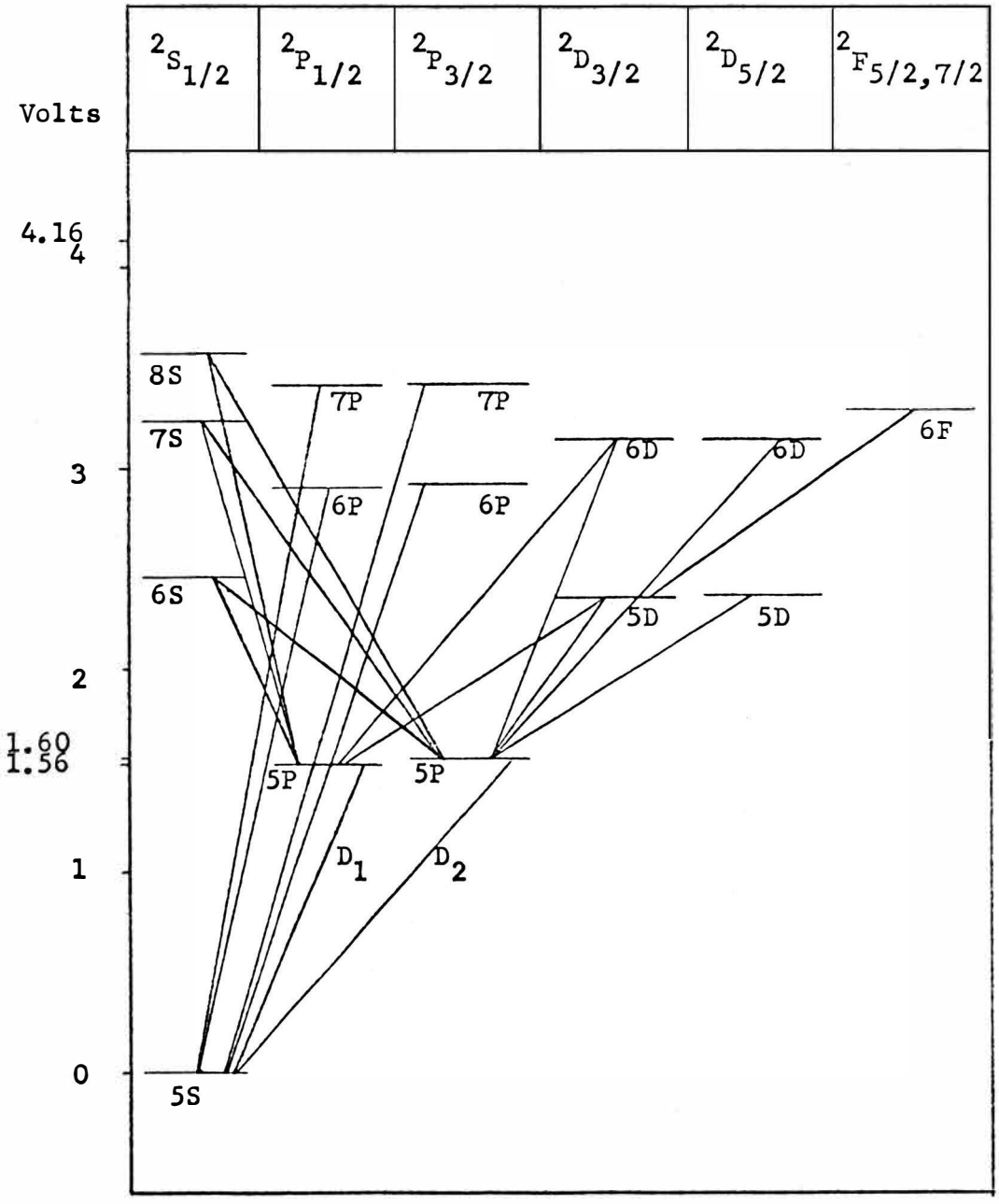
As has been explained, the Zeeman hyperfine structure of rubidium, or any other atom for that matter, results when the atom is subjected to an external magnetic field. This magnetic splitting is caused by the total angular momentum vector F precessing about the direction of the magnetic field. Figures 3 and 4 illustrate this energy level splitting in Rb 87 and Rb 85 respectively. The difference in the diagrams is caused by the different nuclear spins.

Below is summarized the various energy levels splittings Rb 87 and Rb 85 respectively for the ground and first excited states.

Rb 87	$I = 3/2$	$J = 1/2$	hence $F = 2,1$	$M_F = -F, \cdots, -F$
	$I = 3/2$	$J = 3/2$	hence $F = 3,2,1,0$	$M_F = -F, \cdots, -F$
Rb 85	$I = 5/2$	$J = 1/2$	hence $F = 3,2$	$M_F = -F, \cdots, -F$
	$I = 5/2$	$J = 3/2$	hence $F = 4,3,2,1$	$M_F = -F, \cdots, -F$

In this experiment it was necessary to isolate one or the other of the two D lines in the rubidium spectrum (see Fig. 2). The 7948 A line was chosen since it had few lines close to it and was easier to isolate.

A spherical bulb containing a low vapor pressure of rubidium and a relatively high pressure of the buffer gas argon was placed in



Rubidium Energy Level Diagram

Figure 2

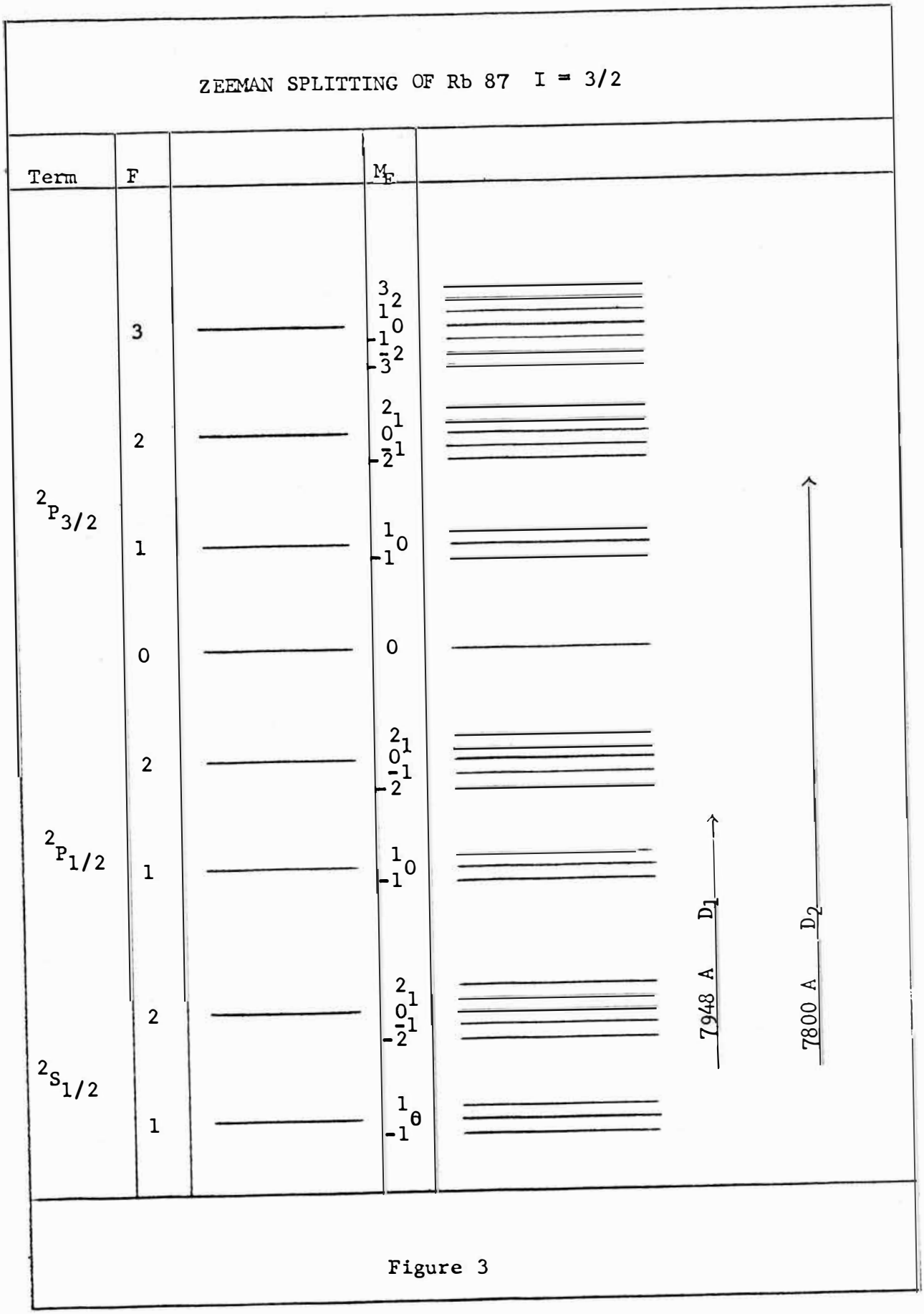

ZEEMAN SPLITTING OF Rb 87 I = 3/2
Term
F
M_F
$^2P_{3/2}$
$^2P_{1/2}$
$^2S_{1/2}$
3
2
1
0
2
1
2
1
3 2 1 0 -1 -2 -3
2 1 0 -1 -2
1 0 -1
0
2 1 0 -1 -2
1 0 -1
2 1 0 -1 -2
1 0 -1
7948 A D_1
7800 A D_2

Figure 3

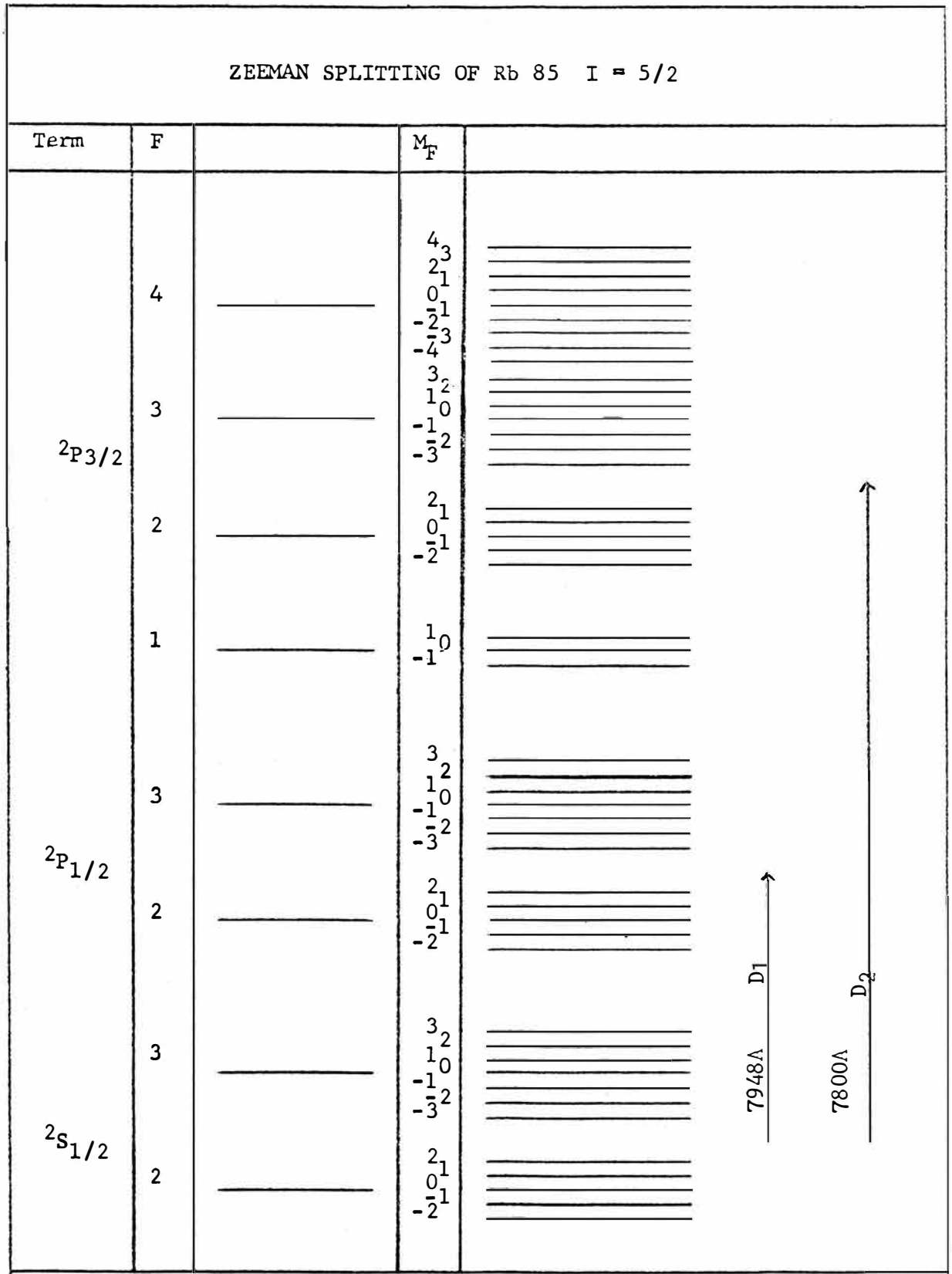

ZEEMAN SPLITTING OF Rb 85 I = 5/2
Term
F
M_F
$^2P_{3/2}$
$^2P_{1/2}$
$^2S_{1/2}$
4
3
2
1
3
2
3
2
4 3 2 1 0 -1 -2 -3 -4
3 2 1 0 -1 -2 -3
2 1 0 -1 -2
1 0 -1
3 2 1 0 -1 -2 -3
2 1 0 -1 -2
3 2 1 0 -1 -2 -3
2 1 0 -1 -2
7948Å
D_1
7800Å
D_2

Figure 4

the center of a set of rectangular coils. Passing a current through these coils permitted the vertical component of the earth's field to be continuously cancelled and the horizontal component to be reversed at will. Thus the horizontal component of the earth's field, or the reversed horizontal component of the earth's field, provided the necessary magnetic field to split the energy levels of the rubidium into their Zeeman hyperfine structures.

Incident upon the rubidium gas in the cell was circularly polarized resonant radiation from a rubidium light source that was axial to the direction of the magnetic field. For all practical purposes only the D_1 component, which had been isolated by filtering the rubidium light source and then circularly polarized, needed to be considered. For convenience, it was assumed that all light was right circularly polarized. Now examine the transition rules which were obeyed under these conditions.

In the absorption process the selection rules are:

$\Delta L = +1$, $\Delta J = 0$, $\Delta F = 0, \pm 1$, (not $F = 0 \rightarrow F = 0$)

and $\Delta M_F = +1$ (For right circularly polarized light)

For circularly polarized incident light, if the direction of observation, the direction of the magnetic field, and the direction of the primary radiation all coincide; then only the transitions corresponding to $\Delta M_F = +1$ or $\Delta M_F = -1$ are excited, depending on the sense of rotation of the electric vector of the primary light.[7]

The process of spontaneous emission following absorption follows the rules:

$\Delta L = \pm 1$, $\Delta J = 0$, $\Delta F = 0, \pm 1$, (not $F = 0 \rightarrow F = 0$)

and $\Delta M_F = 0, \pm 1$

It is the difference between the process of absorption and emission that is responsible for the optical pumping process.

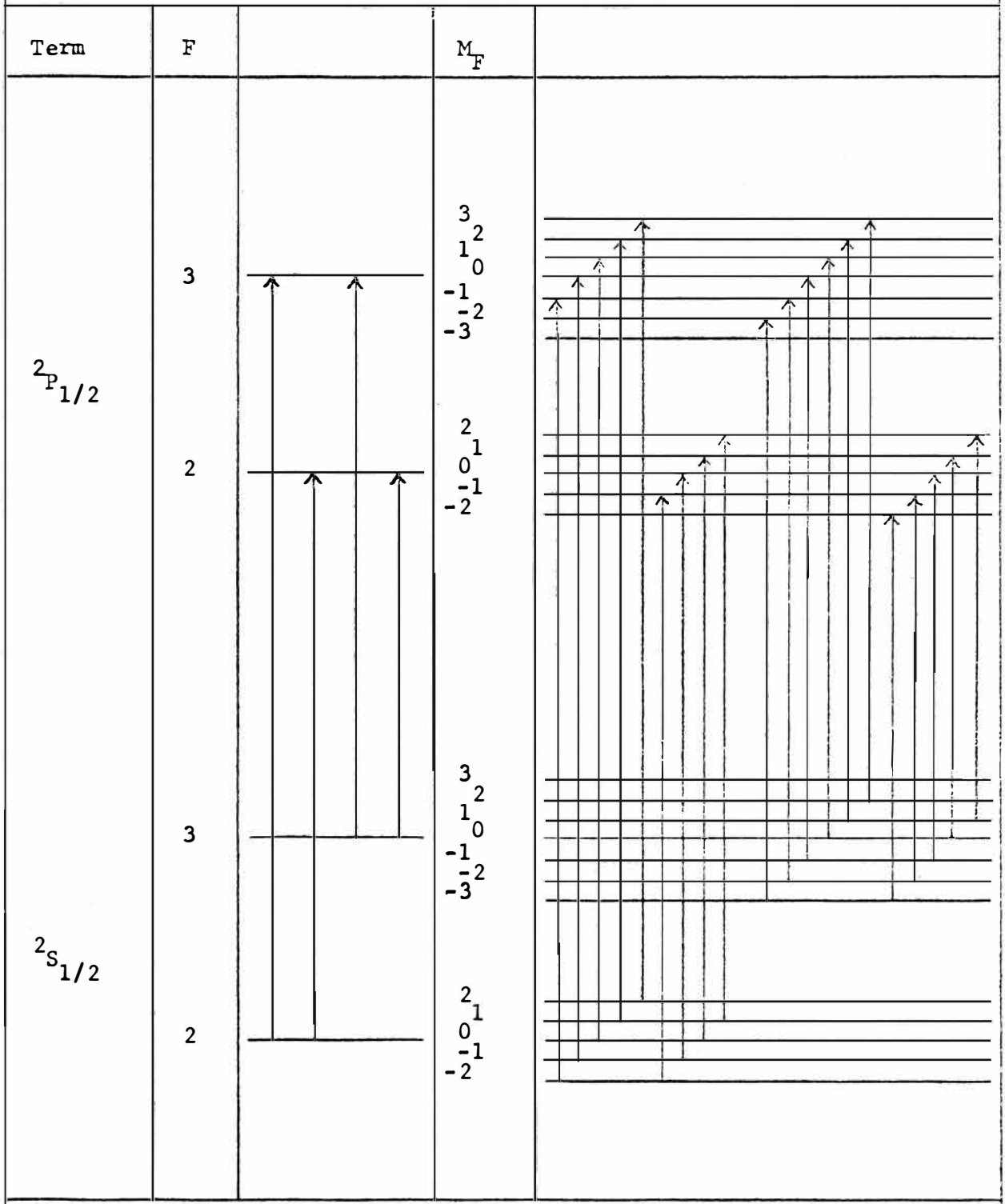
The name pumping is given to any change in the thermal-equilibrium populations of the sublevels that is brought about by an outside cause. The process of optical pumping is associated with a change of the orientation of the angular momenta of the atoms. Pumping of atoms between Zeeman sublevels is accompanied by a change of the magnetization of the gas.[8]

If reference is made to figures 5, 6, and 7, it can be seen that certain levels will be preferentially populated at the expense of others, provided that these population differences can be maintained after the pumping process has produced them. **Figure** 7 shows the possible transitions to the ground state for Rb 87, after being excited by the right circularly polarized D_1 light from a rubidium lamp (Fig. 6). **Figure 5** shows the absorption transitions for the D_1 line assuming that the incident radiation is right circularly polarized.

The **purpose** behind insuring a large difference in the intensities of the D lines is that the D_1 line pumps to large positive M_F values in the ground state when $F = 2$, while the D_2 line pumps to large negative M_F values for the same state. Thus they work in opposite directions which is an undesireable situation. Figure 7 illustrates the concentration in several sublevels which results from optical pumping. Since this is the fundamental principle behind this experiment, it is certainly worthy of further discussion.

It has been noted that right circularly polarized radiation can only produce transitions where M_F changes by $\Delta M_F = +1$. This is due

ABSORPTION TRANSITIONS FOR THE D_1 (7948) LINE Rb 85 I = 5/2
ASSUMING INCIDENT RIGHT CIRCULARLY POLARIZED LIGHT

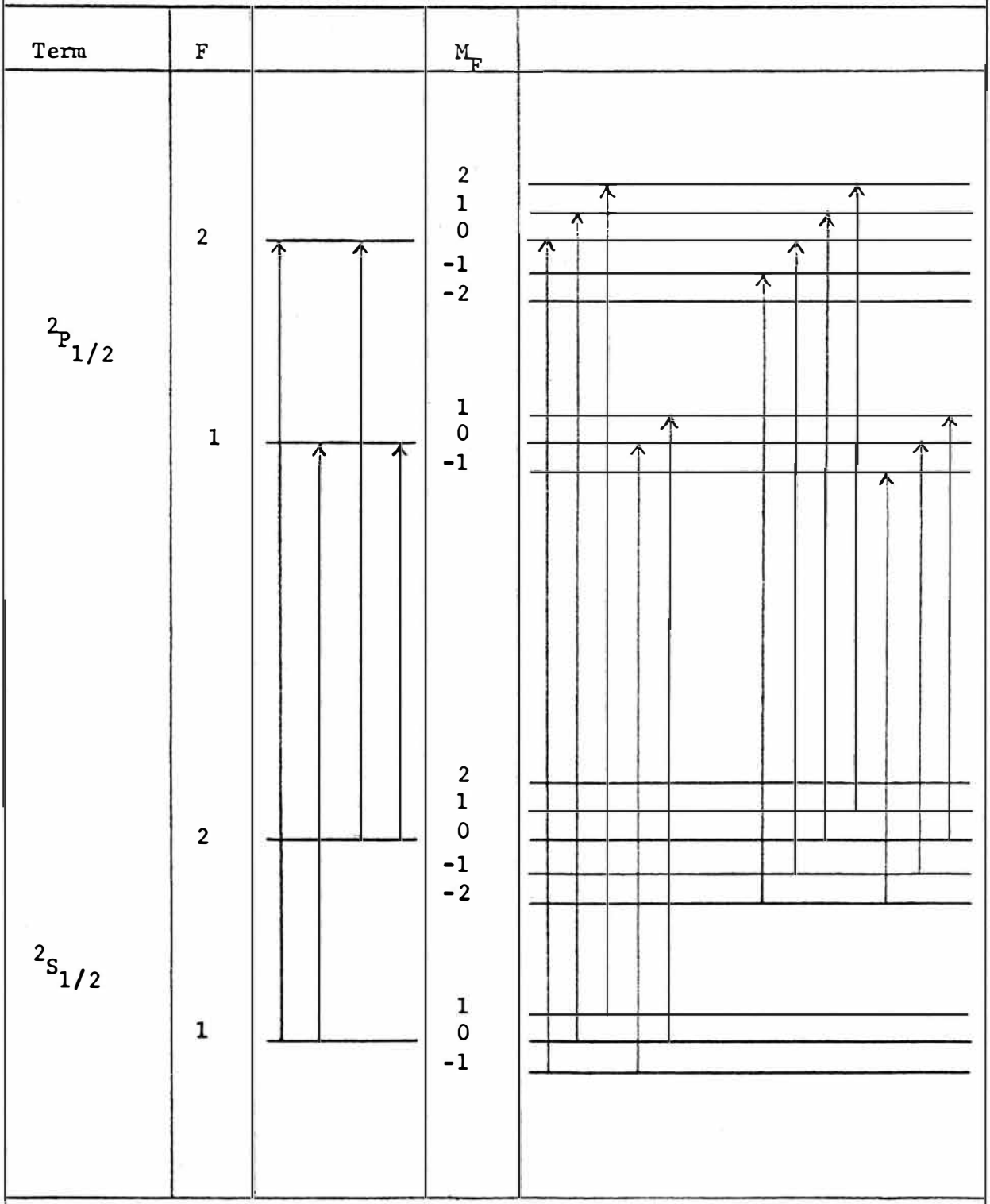


Selection rules obeyed in absorption process-
$\Delta L = +1$, $\Delta J = 0$, $\Delta F = 0, \pm 1$ (not $0 \rightarrow 0$), $\Delta M_F = +1$

Figure 5

ABSORPTION TRANSITIONS FOR THE D_1 (7948) LINE Rb 87 I= 3/2

ASSUMING INCIDENT RIGHT CIRCULARLY POLARIZED LIGHT

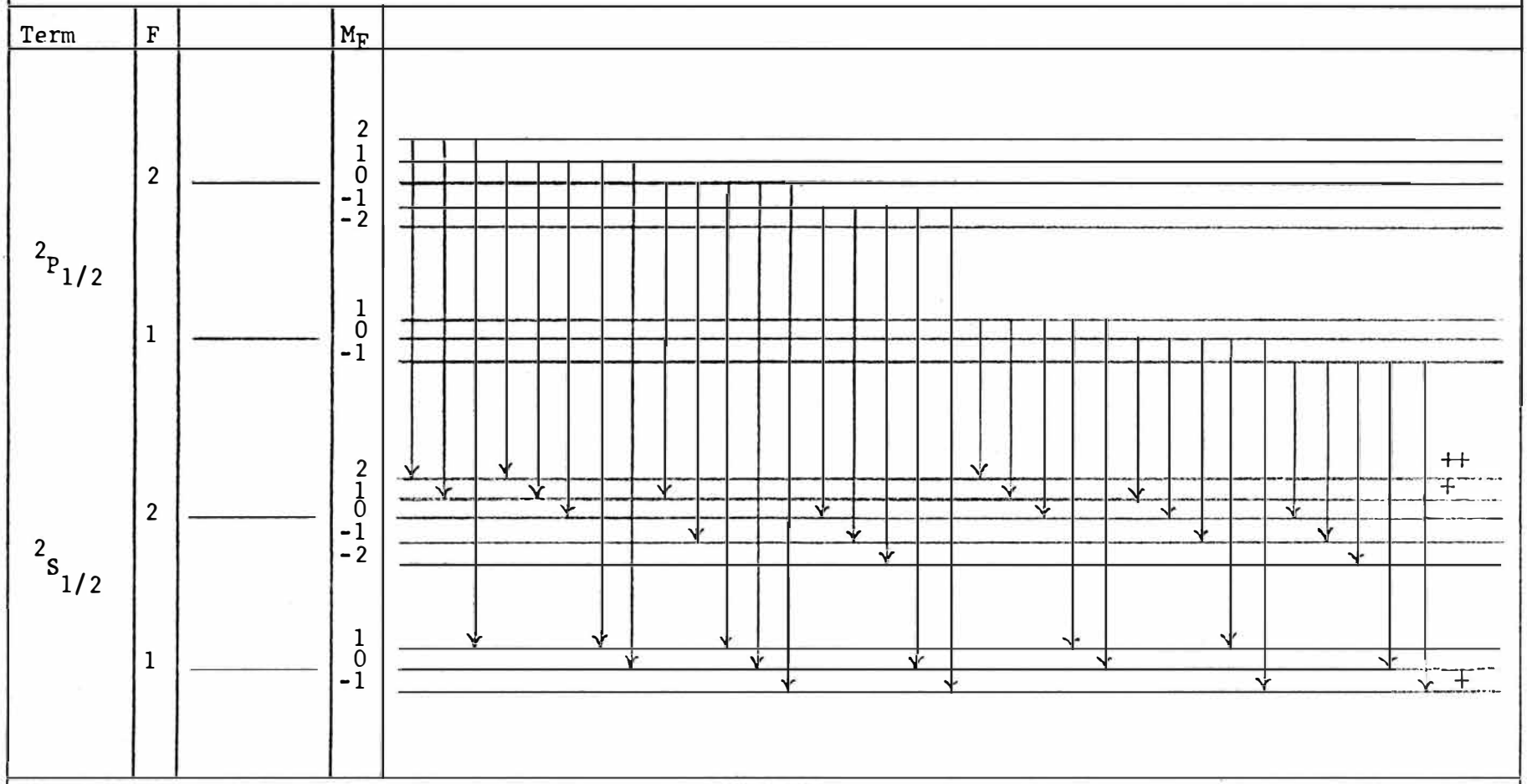


Selection rules obeyed in absorption process-

$\Delta L = +1$, $\Delta J = 0$, $\Delta F = 0, \pm 1$, (not $0 \rightarrow 0$), $\Delta M_F = +1$

Figure 6

EMISSION TRANSITIONS FOR THE D_1 LINE OF Rb 87 $^2P_{1/2} - {}^2S_{1/2}$



Selection rules obeyed in the process of spontaneous emission -
$\Delta L = +1$, $\Delta J = 0$, $\Delta F = 0, \pm 1$, (not $0 \rightarrow 0$), $\Delta M_F = 0, \pm 1$, (not $0 \rightarrow 0$ when $\Delta F = 0$)

FIGURE 7

to a transfer of the photon's angular momentum, but on returning to the ground state M_F can change by $\Delta M_F = 0, \pm 1$ with all changes probable. It is not required that these probabilities be equal, only that there exists some finite, non-zero probability for each of the transitions. This assumption is justified in part by the fact that an excited atom has no memory of the type of radiation used to excite it, thus there is no reason for the atom to show a preference in its mode of returning to the ground state.[3]

It is seen in the above discussion that optical pumping provides a relatively simple technique for concentrating the atoms of the gas in one of the Zeeman sublevels. In the cases where it can be employed, optical pumping is a decidedly simpler and usually a more precise method for exploring the fine and hyperfine structures of atoms in a magnetic field. By one of several techniques this preferential population of energy states may be detected and thereby something may be learned of the structure induced by the field.

IV. THE EXPERIMENT

The experiment performed was an investigation of the rate at which rubidium atoms respond to a reversal of a magnetic field. The optical pump was actuated in the following manner: light from a rubidium lamp was passed through a lens to produce a parallel beam of light which was passed through an interference filter to remove unnecessary or damaging lines in the rubidium spectrum. The filtered light, primarily D_1 radiation, passed through a polarizer and quarter wave plate in proper combination to give circularly polarized light. This beam was passed through the rubidium cell where it interacted with the rubidium gas in the presence of a weak magnetic field that was axial to the beam of light. Finally the resulting light was passed through a second lens and into a photocell. The output of the photocell was amplified and displayed on an oscilloscope screen from which the data was photographed. See Figure 8 for a block diagram of this arrangement. Very important to this arrangement was the set of rectangular coils that surrounded the absorption cell. These coils permitted the horizontal component of the earth's field to be very rapidly reversed while the vertical field remained cancelled at all times. It was the moment of field reversal that was of interest.

The optical pumping process has been previously described and now it is advantageous to consider not only the optical pumping process but also the repopulation process that is of interest at the moment of field reversal. The pumping process produced transitions from the $^2S_{1/2}$ hyperfine Zeeman ground state sublevels to the $^2P_{1/2}$ hyperfine

BLOCK DIAGRAM OF EXPERIMENTAL SET-UP

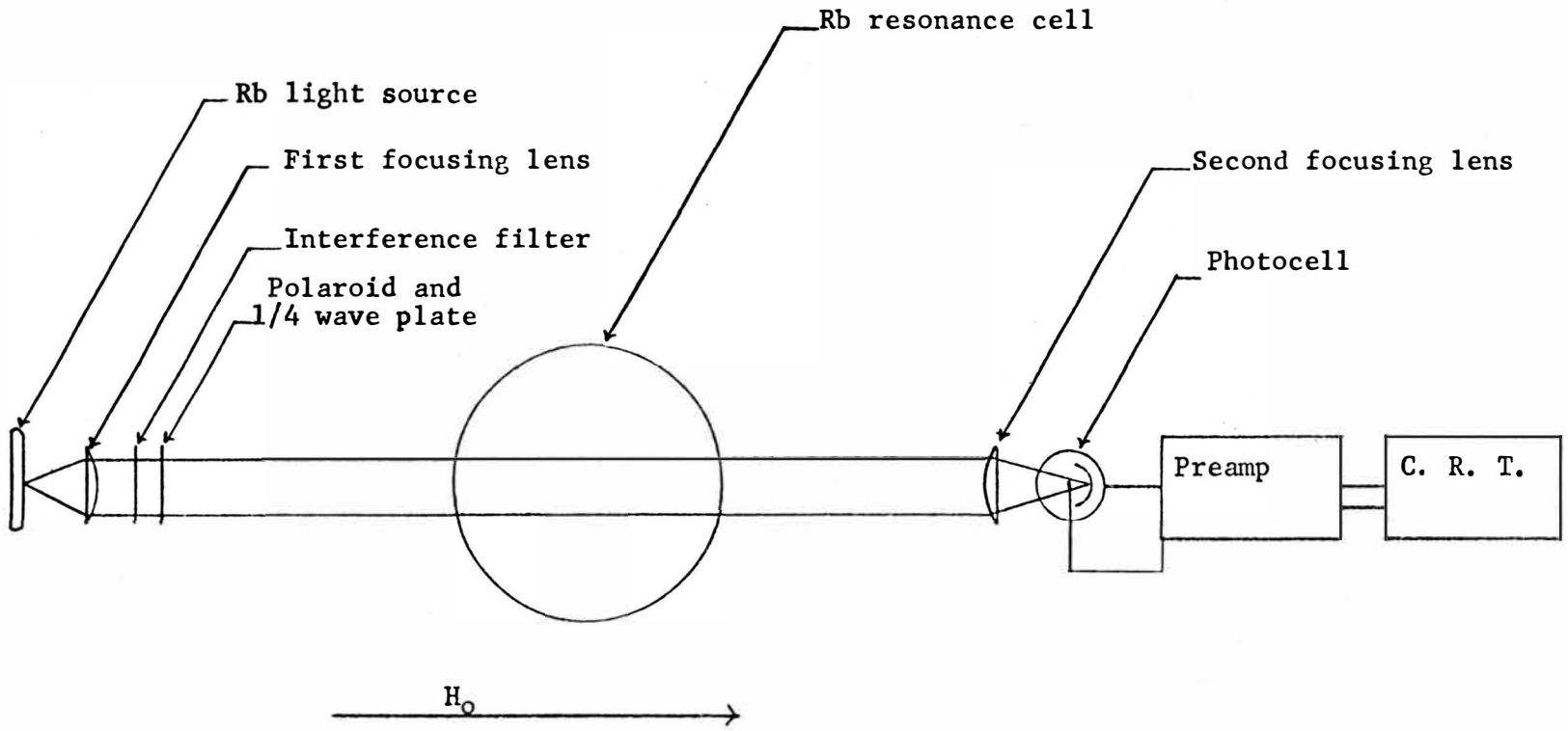


Figure 8

Zeeman sublevels for which ΔM_F changed by + 1. The transition probabilities from the various ground state Zeeman sublevels to an excited state varied for the various ground state sublevels.

A discussion of some work done by W. Franzen[4] is now in order. Franzen varied experimental parameters as the partial pressure of the buffer gas as well as the partial pressure of the alkali metal vapor so that the relative frequencies of the different types of collisions leading to relaxation could be changed and thereby studied separately.

The relaxation process is a deorientation process between the aligned ground state rubidium atoms and other rubidium atoms, the buffer gas, and the cell wall. If it were not for the incident pumping light, the large population differences between the Zeeman hyperfine sublevels could not be maintained and the aligned atoms would relax to thermal equilibrium.

The relaxation of a partially orientated alkali metal vapor in a buffer gas would be expected to take place by electron exchange between colliding atoms, as discussed by Wittke and Dicke.[2] Experimental evidence for the influence of electron exchange on the width of the hyperfine resonance line in Rb 87 has been obtained by a Princeton group.[9] From this data Franzen calculated the average time between collisions of rubidium atoms in a pure rubidium atmosphere. This time was 6.5×10^{-3} sec. and was of interest since the assumed temperature and pressure of the rubidium for the calculation were close to those used by the author.

When Franzen observed the relaxation times without a buffer gas, he found the relaxation times to be about eighteen times longer than the mean time between rubidium - rubidium collisions. An explanation

of this result was based on a suggestion made by Bender.[10] In a non-radiative binary collision, of which a spin exchange collision between two rubidium atoms is an example, the sum of the components of the total angular momenta of the colliding atoms in the direction of the applied magnetic field must be the same before and after the collision. This means, in the first place, that in a collision between two atoms which are both in the higher energy hyperfine state ($F = I + 1/2$) and for which the magnetic quantum number has either its maximum or minimum value ($M_F = F$ or $-F$), spin exchanges cannot affect the spin orientations of the two atoms. In the second place, if two atoms collide which are in the same hyperfine state both before and after the collision, their combined light absorption probability for D_1 radiation will remain unaffected by the collision. For Rb 87 with $I = 3/2$, the absorption probability for D_1 radiation in the $F = 2$ state is proportional to $2 - M_F$; in the $F = 1$ state it is proportional to $2 + M_F$. (See table I).[3] For two atoms, both of which are in the $F = 2$ state, the combined absorption probability is therefore proportional to $4 - (M_F^{(1)} + M_F^{(2)})$ while if they are both in the $F = 1$ state, it is proportional to $4 + (M_F^{(1)} + M_F^{(2)})$. If $(M_F^{(1)} + M_F^{(2)})$ is invariant, the amount of light absorbed by the two atoms in the same F state does not change as a result of the collision.

The relaxation process must conserve angular momentum among all of the particles, electrons, and nuclei involved in the collision between an aligned alkali atom and an inert gas atom. Bernheim[11] shows, using first order time dependent perturbation theory, that the interaction energies between alkali atoms and buffer gas atoms are of the order of the magnitude of the hyperfine interaction in atoms.

F	M_F	D_1 $^2S_{1/2}$-$^2P_{1/2}$	D_2 $^2S_{1/2}$-$^2P_{3/2}$
2	2	0	6
	1	1	5
	0	2	4
	-1	3	3
	-2	4	2
1	1	3	3
	0	2	4
	-1	1	5

Ground State Absorption Probabilities For $\Delta M_F = +1$

Table I

Because of the relatively high buffer gas pressure (2.65 cm of Hg) the average time between Rb-Argon collisions approaches the life time of the excited state (1.5×10^{-8} sec.) of rubidium.

It is therefore likely that considerable deorientation and loss of phase memory occurs for these excited atoms before they return to the ground state.[3] This allows one to make the simplifying assumption that all ground states have equal probabilities of being repopulated by the atoms returning to the ground state. This allows one to consider the pumping process entirely in terms of optical excitation from the various ground state sublevels.

Since the optically excited state has a short life time, it can be treated macroscopically in terms of an equivalent relaxation process between ground state sublevels. Table 1 gives the unnormalized relative transition probabilities ($\triangle M_F = +1$) for the D_1 and D_2 lines in the rubidium spectrum. The pumping rate for any level is obtained by weighting the intensities of the D_1 and D_2 components in the incident pumping light by the coefficients given.[3]

Assume that the pumping process has reached an equilibrium condition, that is, positive M_F sublevels in the $F = 2$, $^2S_{1/2}$ state have been overpopulated at the expense of the other sublevels in that state. This equilibrium will be considered a dynamic equilibrium, that is the rate at which atoms were being pumped to the large positive M_F sublevels in the $F = 2$, $^2S_{1/2}$ level, equals the rate at which atoms returned to the negative sublevels.

Now reversing the axial magnetic field produced a nearly instantaneous change in the positive and negative M_F levels. Immediately after

the field reversal, the negative levels became the overpopulated levels and thus the probability of the gas absorbing the incident light became relatively high. If reference is made to Figures 6 and 7, a better understanding of this process can be gained. When the gas was in equilibrium with the incident light, large positive M_F populations existed in the $^2S_{1/2}$, F = 2 level. Immediately after field reversal, the highly populated positive states became highly populated negative states and from table 1, for the F = 2, $^2S_{1/2}$ level, it is seen that the probability of an atom accepting a right circularly polarized photon ($\Delta M_F = +1$) and becoming excited is much higher for the negative M_F sublevels than for the positive M_F sublevels. Thus immediately after the field was reversed the rubidium atoms absorbed a portion of the incident light. As the atoms were again pumped to the positive M_F sublevels the rubidium vapor became more and more transparent to the incident light. This occurred because the positive M_F sublevels (F = 2, $^2S_{1/2}$) had a relatively small possibility of being excited by the incident radiation. The Zeeman splitting was still of the same magnitude an instant after the field was reversed, since the magnitude of the field has not been changed. The absorption reduced the intensity of the transmitted light and a dip was observed on the oscilloscope screen corresponding to this decrease in intensity.

Immediately after the field has been reversed, the oscilloscope trace appeared similar to the trace shown, (fig. 9) where A denotes the equilibrium level and 0 denotes the minimum intensity of the transmitted light. The dotted line is drawn tangent to the curve at 0. It will be assumed that the rate at which the intensity (Y-axis) builds up to the equilibrium level is an exponential and the point 0 is

OSCILLOSCOPE TRACE OF LIGHT ABSORPTION

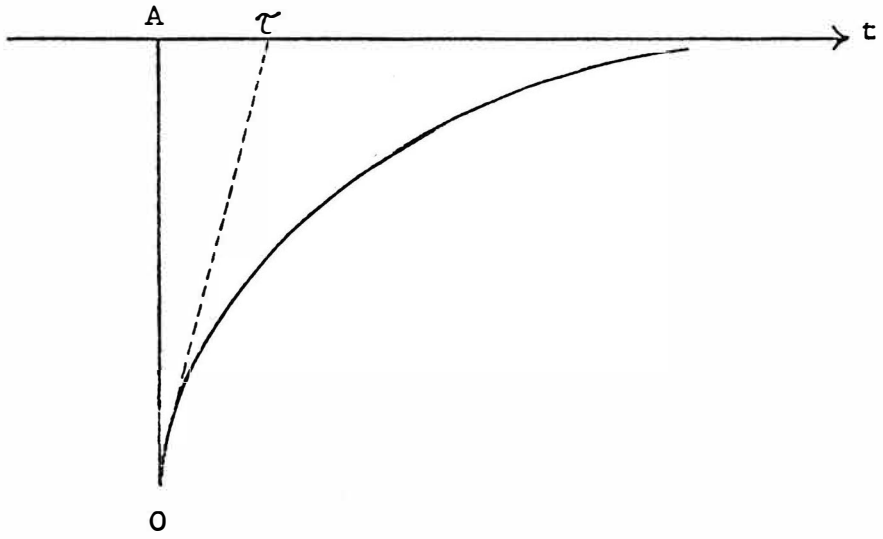


Figure 9

chosen as the origin of a coordinate system where the X-axis is the time axis. The curve is then given by the equation

$$Y = A(1 - e^{-t/b})$$

where b is a constant to be determined and is the overall pumping time. Now the slope of the exponential decay is given by

$$M_{t = o} = A/b$$

Now consider the dotted line which is drawn tangent to the curve at $t = 0$. The point slope form of an equation of a line is given as:

$$y - y_o = M (t - t_o)$$

and when $y = 0$ and $t = 0$, $y_o = Mt_o = (A/b) t_o$

so that when $y = A$ and $t = b = \tau$, we have

$$y = A (1 - e^{-t/\tau})$$

At $t = \tau$, we have $y = A (1 - e^{-1})$

so that at time $t = \tau$, y has increased to the fraction 1/e of its final value. It should also be noted that the repopulation rate $(1/\tau)$ is the sum of two processes, the relaxation rate, and the pumping rate. Stated mathematically:

$$1/\tau = 1/\tau_R + 1/\tau_P$$

The pumping rate is the rate at which the atoms would be reorientated if there were no competing relaxation processes.

Employing τ as a measure of the time required for the intensity to return to the equilibrium level, we may visualize τ as the time required for 1/e of the orientations of the magnetic moments to be reversed and again assume their earlier alignment with respect to the field. Restating the situation, the field has been nearly instantaneously reversed with the atoms momentarily maintaining the same orientation as before the field was reversed. The preferential

orientation has been reversed or the populations of the (+) and (-) levels have been exchanged and equilibrium is the state corresponding to a return to the earlier preferential alignment of the magnetic moments of the atoms with respect to the magnetic field.

As has been stated, the atoms can change their ground state orientations by two different processes: a pumping process and a relaxation process. Lifetime against hyperfine-structure transitions of isolated atoms in the ground state are very long. For alkali-metal atoms they are of the order of 10^6 years,[8] thus any relaxation due to this process may be neglected. It was the aim of this experiment to measure the relaxation rate of rubidium under a specific set of circumstances.

Since the aim of the experiment was to measure one of the two competing processes, one must be minimized so that the other is the predominant process. Since relaxation rates were of interest, light intensities were kept low enough to minimize the pumping process. This is an unusual approach, for the actual mechanism used to detect the effect was minimized and actually was extrapolated out of the data to obtain the value of interest.

The pumping rate corresponds to the reciprocal of the time required for an atom in the negative state to be excited by an incident photon and to fall back to the positive state. It should be realized that not all atoms decaying decay to a positive state and that some atoms in a positive state will be pumped to lower states but this is all part of the pumping process.

The relaxation process is a more direct process in that it requires no incident light, hence no preferentially excited states.

In the relaxation process, through one of several mechanisms, the magnetic moment of the atom is completely reversed.

It has been found that an alkali metal atom loses its orientation in a magnetic field with just one collision with the wall of a glass cell.[12] Thus if it is desirable, as it was in the present experiment, to preserve the orientation, it is necessary to minimize wall collisions. By introducing a buffer gas such as argon, which has a symmetric charge distribution, the alkali metal atoms are forced to take a devious path to get to the cell wall and thus the rate of cell wall collisions is greatly reduced.

The buffer gas has a deorienting effect on the "pumped" rubidium atoms in the ground state, but the advantage gained by reducing cell wall collisions outweighs this disadvantage. The buffer gas deorienting effect is certainly a contributing factor to the relaxation time found but at the pressure used in this experiment it does not produce a detrimental effect. Dehmelt[1] found that increasing the argon pressure from 3 cm Hg to 40 cm Hg decreased the relaxation time for sodium by a factor of about ten.

Since the relaxation rate is the quantity of interest, consideration will now be given to its determination. The pumping rate varies with light intensity, while the relaxation rate should remain constant if all other variables remain constant, i.e. buffer gas pressure, rubidium vapor pressure, etc. Now one has that the repopulation rate equals the pumping rate plus the relaxation rate

$$1/\tau = 1/\tau_P + 1/\tau_R$$

and from the assumption that $1/\tau_R$ = constant while $1/\tau_P \rightarrow 0$ ($\tau_P \rightarrow \infty$)

as $I \rightarrow 0$, it is seen that $1/\tau \rightarrow 1/\tau_R$ as $I \rightarrow 0$ when I refers to incident light intensity. Hence if $1/\tau$ versus I is plotted and the curve is extrapolated to $I = 0$, the intercept on the $1/\tau$ axis will give directly $1/\tau_R$, the relaxation rate. Thus if data are taken with variable intensities of light, the relaxation time can be found.

V. THE APPARATUS

As has been stated previously, the optical pump worked in the following manner: light from a rubidium light source was made parallel by a lens, passed through an interference filter to remove all but the D_1 line of the rubidium spectrum and then circularly polarized by a polarizer and quarter wave plate. The resulting light was passed through a cell containing rubidium at a vapor pressure of approximately 10^{-5} mm of Hg pressure and argon buffer gas at a pressure of 26.5 mm of Hg pressure. The resulting transmitted beam of light was detected by a photocell whose output was amplified and displayed on the screen of an oscilloscope. The data taken was in the form of photographs of the trace on the oscilloscope screen. Plate I shows the experimental equipment.

Since the optical pumping process was a very difficult effect to detect, it is worth while to examine some of the particular circuits and equipment used. Listed below are the principal pieces of apparatus:

A. Tektronix Oscilloscope Type 531A.

B. Tektronix Low Level Preamplifier Type 122.

C. Tektronix Oscilloscope Camera Type C-12.

D. Bell Gaussmeter Type 120.

E. L and N Potentiometer.

F. Two sets square Helmholz coils with controls.

G. Photocell (1P40).

H. Light source and power supply.

I. Rubidium resonance cell.

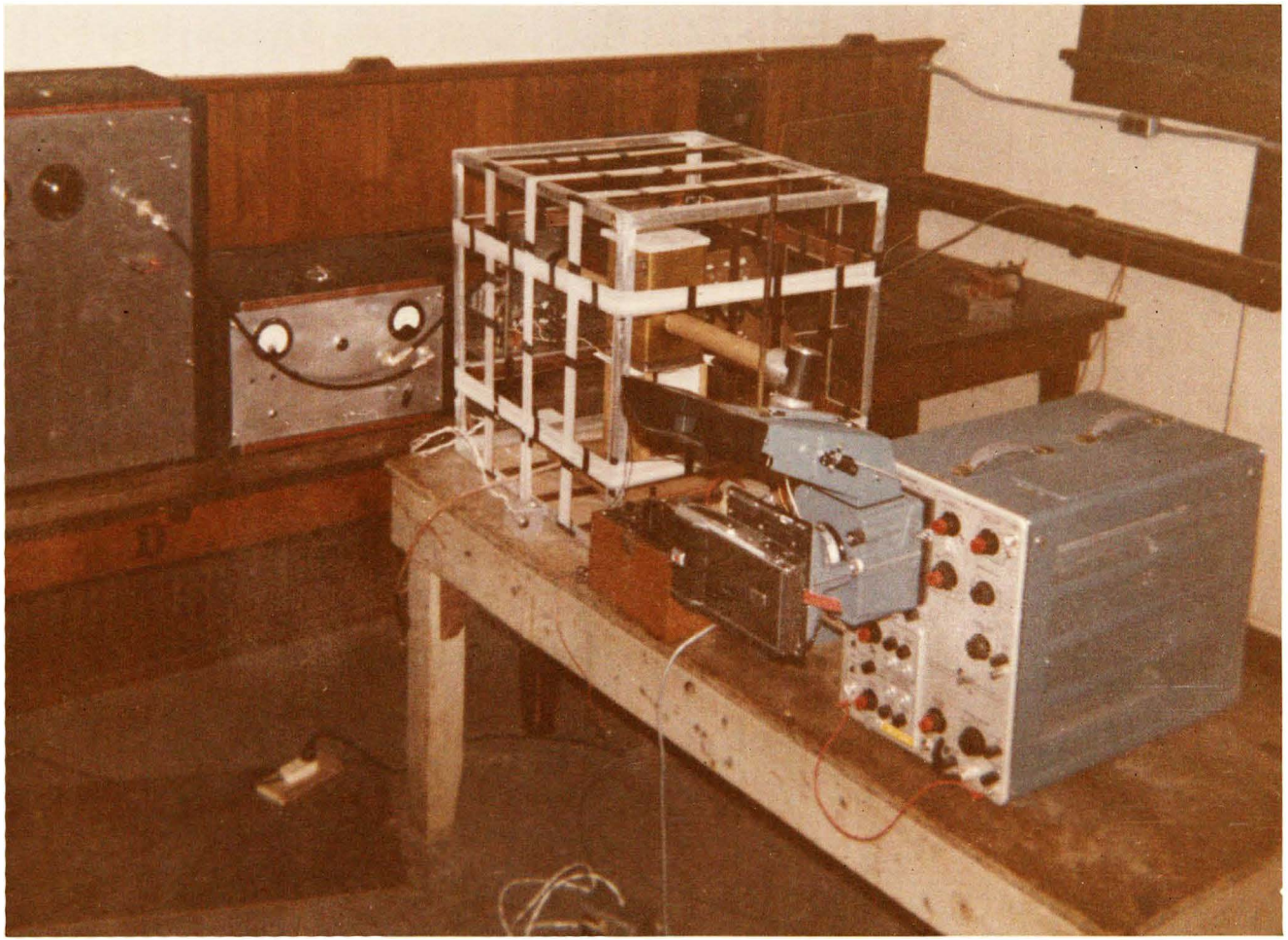


Plate I. The Optical Pump

J. Mercury diffusion vacuum system.

K. Optical accessories, 2 - 2" focal length lenses, polaroid, 1/4 wave plate (mica), and interference filter.

The resonance cell, which was the heart of the experiment, was prepared by the author on a mercury diffusion vacuum system. The 500 milli-liter pyrex flask was cleaned very thoroughly before it was attached to the vacuum system. Plate II shows the cell (near middle of picture) attached to the vacuum system. After the cell had been attached to the vacuum system, all glassware that had been added was evacuated and then heated periodically over a period of thirty-six hours, until subsequent heatings did not drive any more absorbed gases from the glass. The release of gases from the hot glass was easily detected from the readings on the vacuum pressure gauges. When it was felt that the glassware was sufficiently free of water vapor, gases, and impurities, the system was opened and an open capsule of rubidium, approximately one gram, was inserted. The system was rapidly sealed and again evacuated until the pressure had reached a value around 1×10^{-5} mm of Hg. Several oxides formed on the rubidium even though precautions were taken to minimize them, but most of the oxides were reduced by heating the metal in the vacuum. The remaining oxides were stable enough not to present any problem.

Near the upper left hand corner of Plate II, the 1.1 liter flask containing pure argon can be seen. The vacuum system was flushed with argon several times in an attempt to minimize the remaining nitrogen and oxygen in the system. At the top of Plate II, right of center, a cold trap containing dry ice and acetone can be seen. All

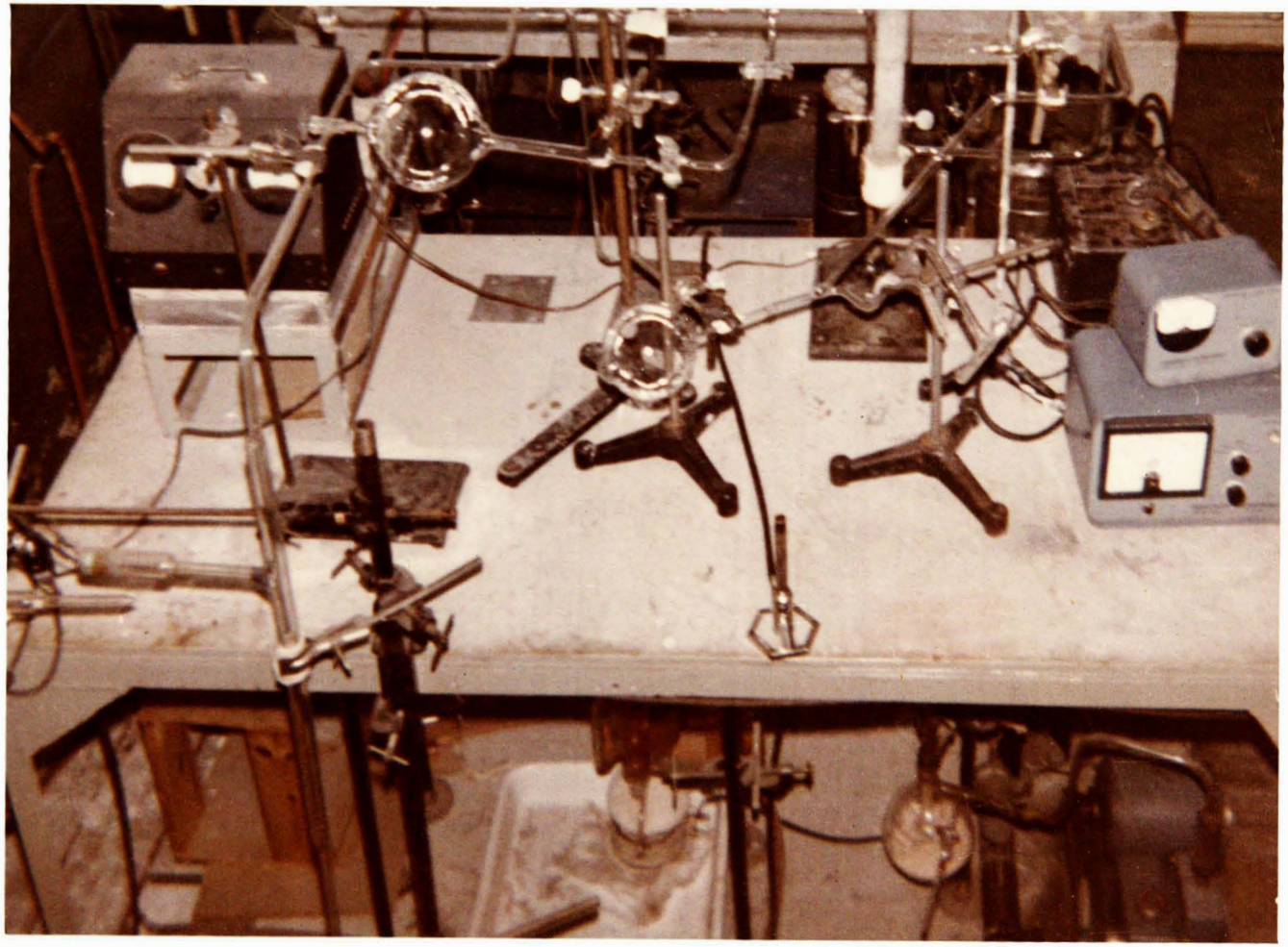


Plate II. Resonance cell on vacuum system

of the glassware in front of this cold trap was heated in an attempt to drive out any mercury that might have diffused into it.

A few hundredths of a gram of rubidium was then distilled into the resonance cell. Time was given the system to reach its minimum pressure, 7×10^{-6} mm of Hg. The top part of the vacuum system was then sealed from the pumps by means of a stopcock and the buffer gas was added to a pressure of 26.5 mm of Hg. The cell was then sealed off from the vacuum system and removed.

After the cell had been removed from the vacuum system, two chromel-alumel thermocouples were attached to it so that the cell temperature could be determined at any time. The resonance cell was mounted in a brass "can" and placed in the center of the Helmholz coils (see Plate I). After the thermocouples were attached to the potentiometer the resonance cell was ready for use. A Bunsen burner was situated under the resonance cell so that it could be heated gently. Since there was an excess of rubidium in the cell, it was assumed that the vapor pressure of rubidium could be found by measuring the temperature.

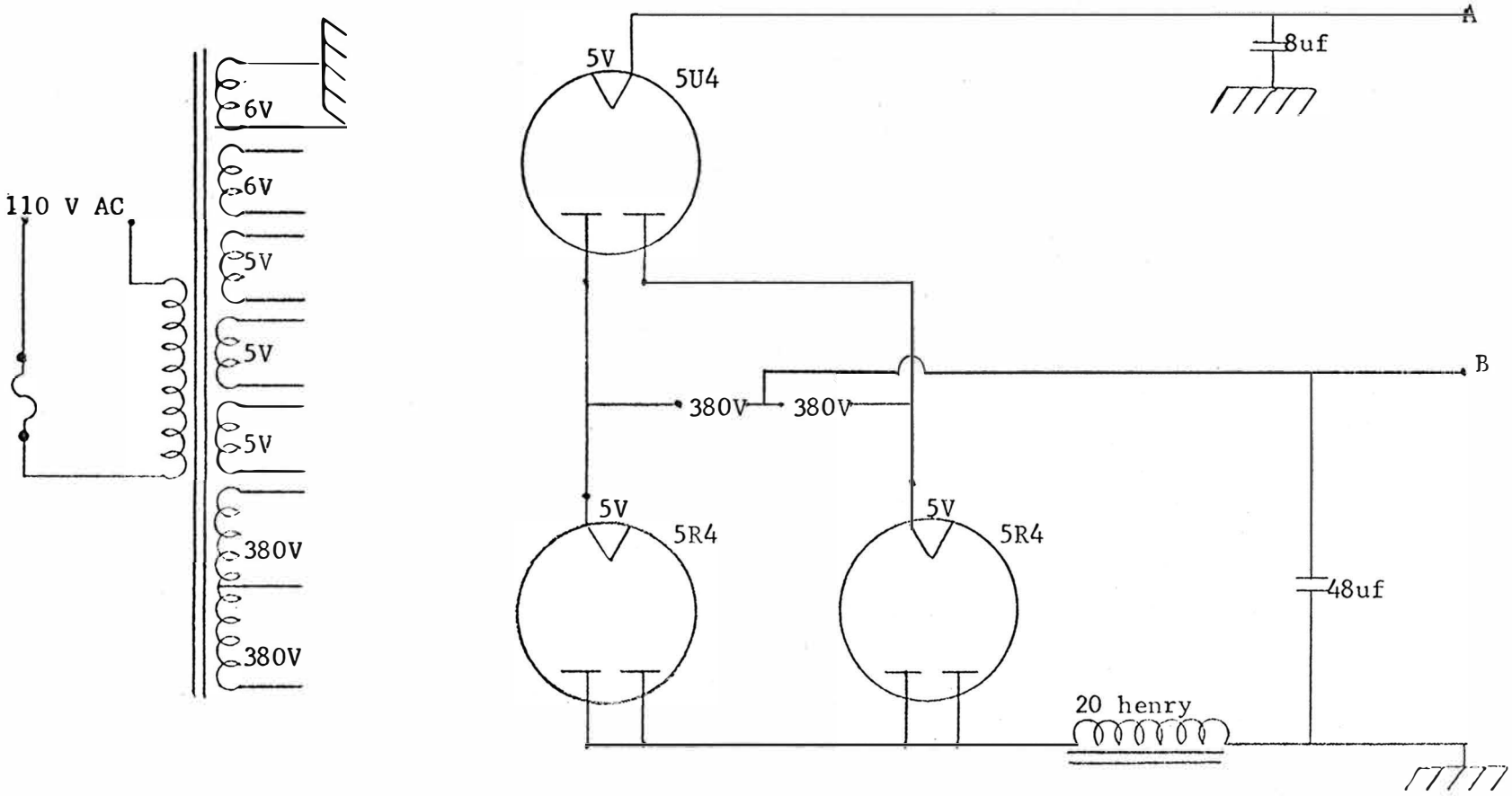
The greatest problem encountered was developing a light source that would have a relatively large intensity of the D lines and yet would be stable enough to allow the pumping effect to be detected over the random fluctuations in the light source. The design selected was an electrodless, rf excited discharge. The discharge tube was 3.5" long with a constricted central portion 1.5" long having an inside diameter of .16". The end portions of the discharge tube had inside diameters of .36". An attempt was made to select a size that

would give a fairly large surface area of the discharge and yet would minimize the undersirable effect of self reversal of the resonant D - lines.

The light source was prepared on the mercury diffusion vacuum system with a few hundredths of a gram of rubidium distilled into the light source. Then argon was added to a pressure of 2×10^{-1} mm of Hg before sealing and removing the source from the vacuum system. The argon served as a discharge starter, for at room temperature the rubidium vapor pressure was too small to sustain a discharge, but the argon discharge rapidly raised the temperature of the system and the rubidium very quickly became the predominating discharge.

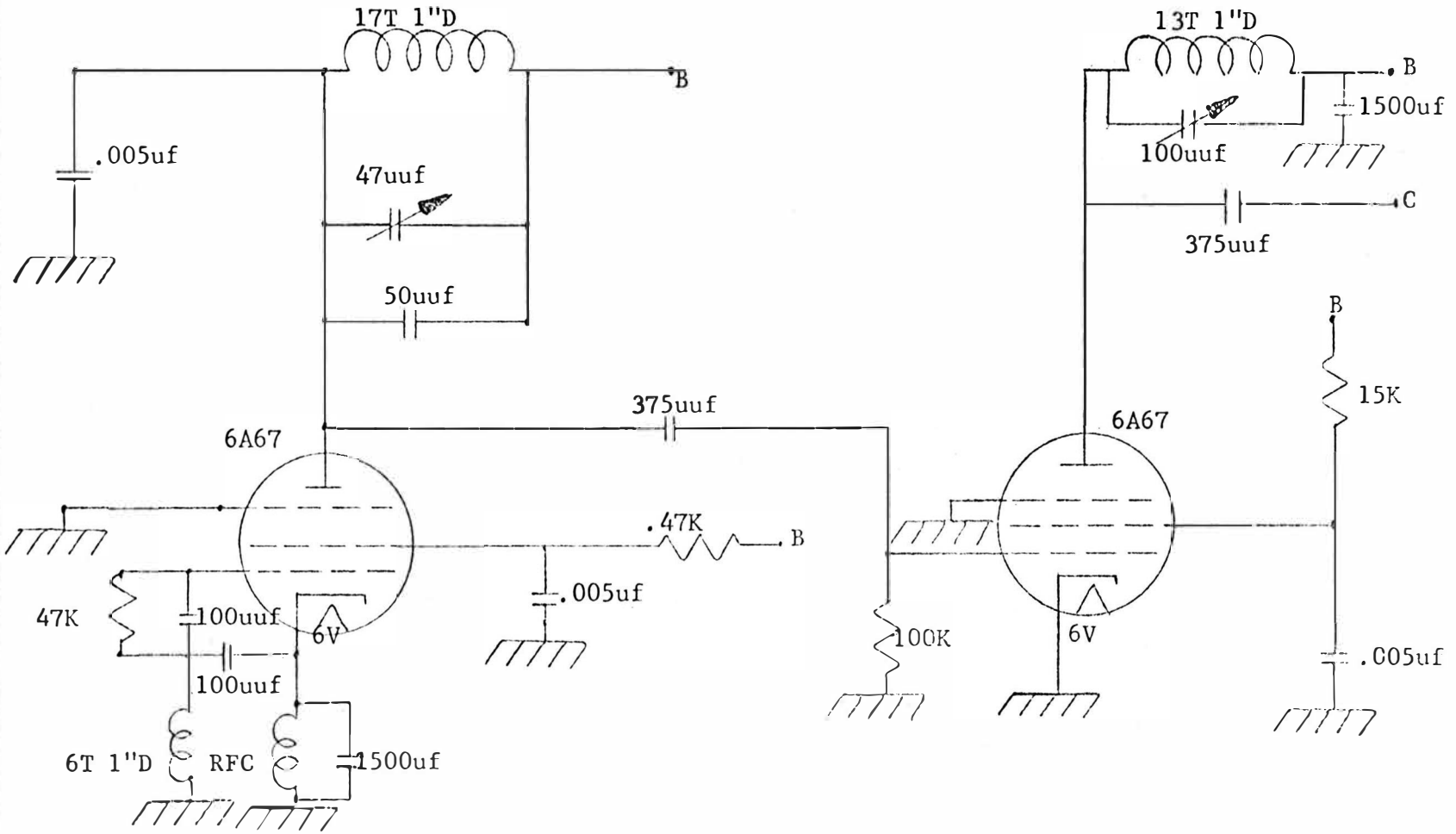
Figures 10, 11, 12, and 13 are the schematic diagrams of the circuits which were used to power the source. Figures 10, 11, and 12 represent different parts of one circuit and the letters indicate joining connections. The large number of stages in the oscillator circuit were necessary to keep the oscillator oscillating when the lamp fired. Before the discharge fired in the rubidium lamp, the coil around the light source presented a high impedence to the oscillator, but as soon as the lamp fired, the impedence dropped almost instantaneously to a much lower value. This drop imposed unusual requirements on the oscillator. Without the buffering or protective stages, the change in load when the lamp fired would detune the oscillator to such a degree that the oscillator would stop operating.

The circuit was designed to operate at a frequency of nine megacycles. This frequency was chosen somewhat arbitrarily; the main requirement being that the frequency be sufficiently high so as not to interfere with the optical pumping signal.



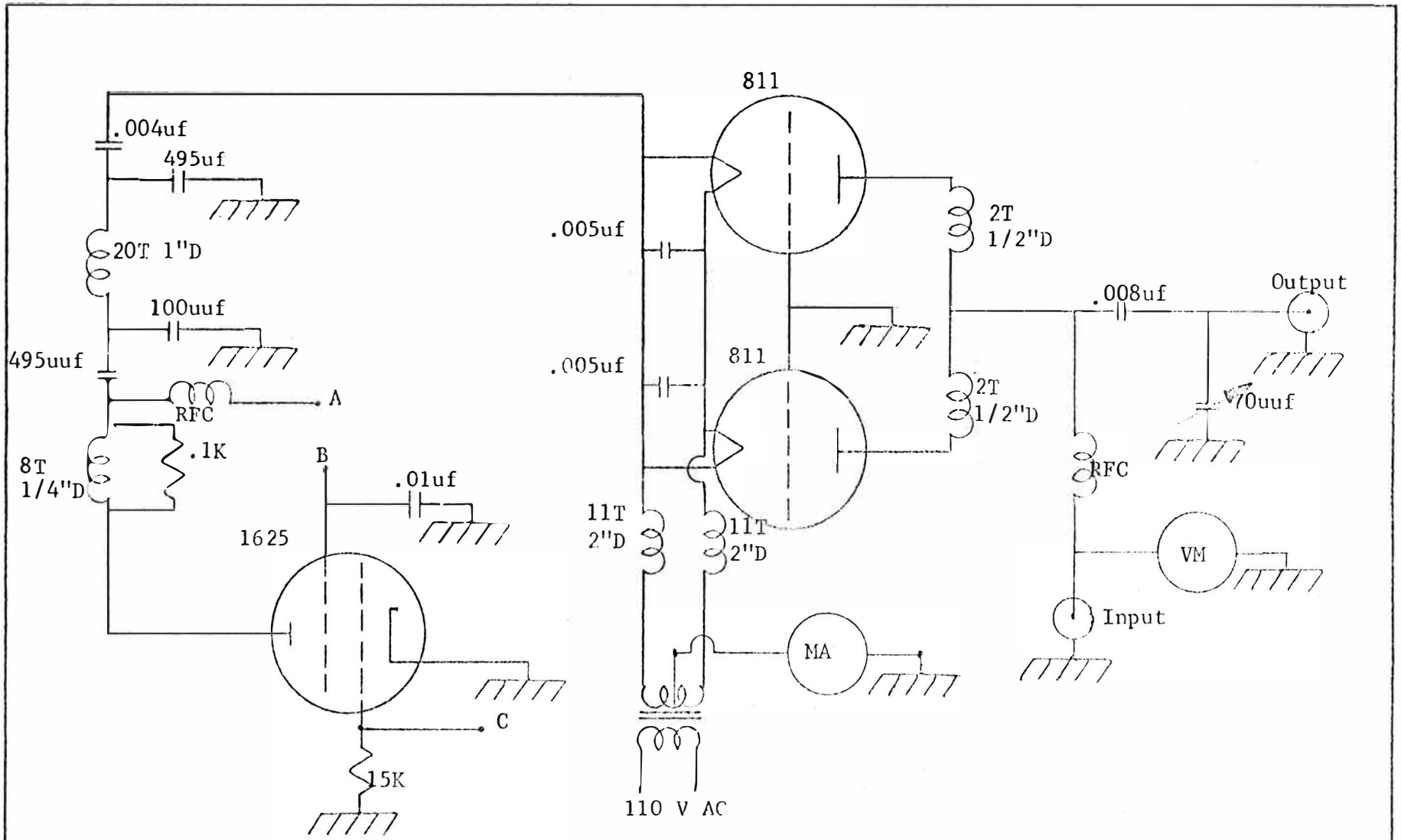
Power Supply For Oscillator

Figure 10



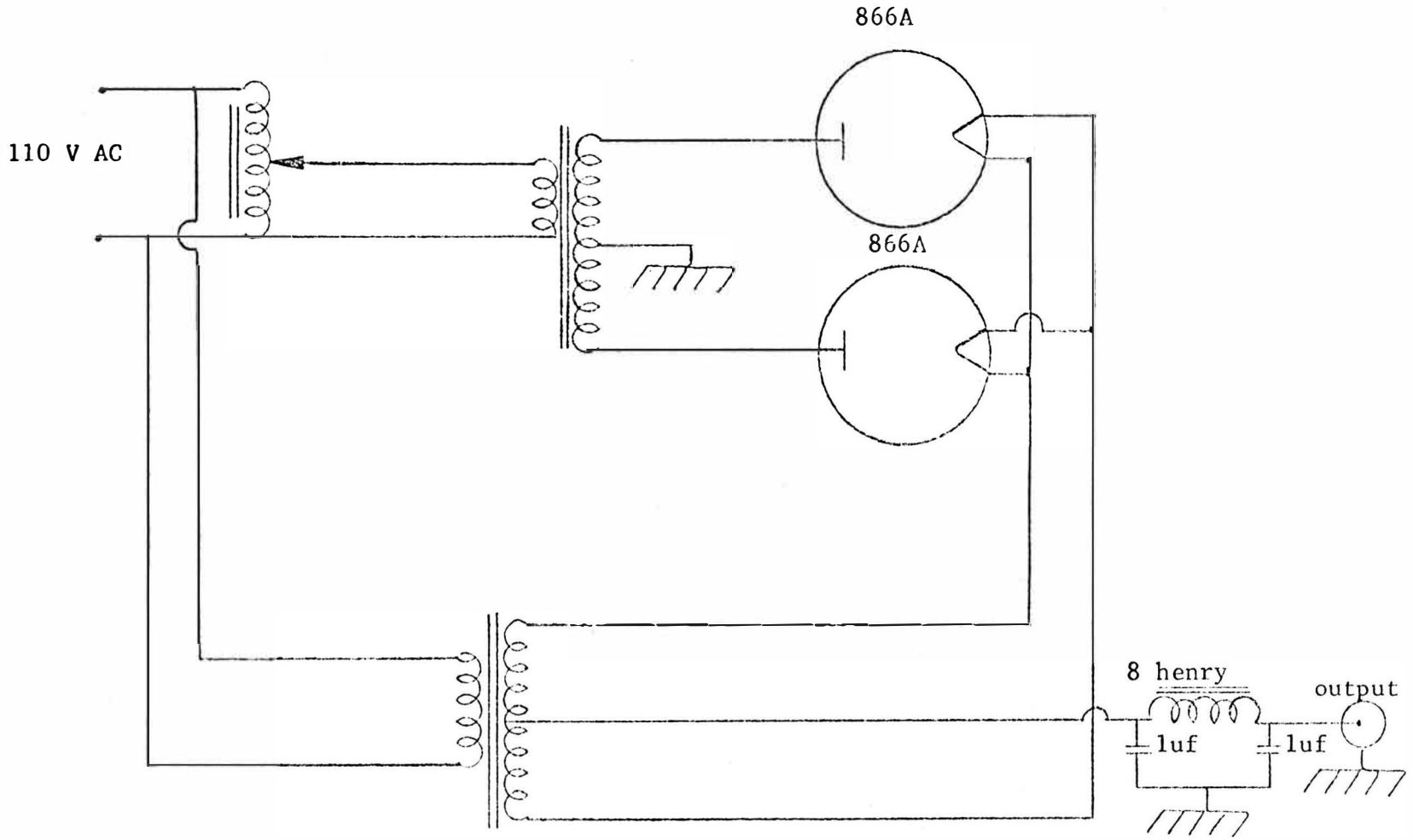
Oscillator and Buffer Amplifier Stages

Figure 11



Driver and Final Amplifier Stages

Figure12



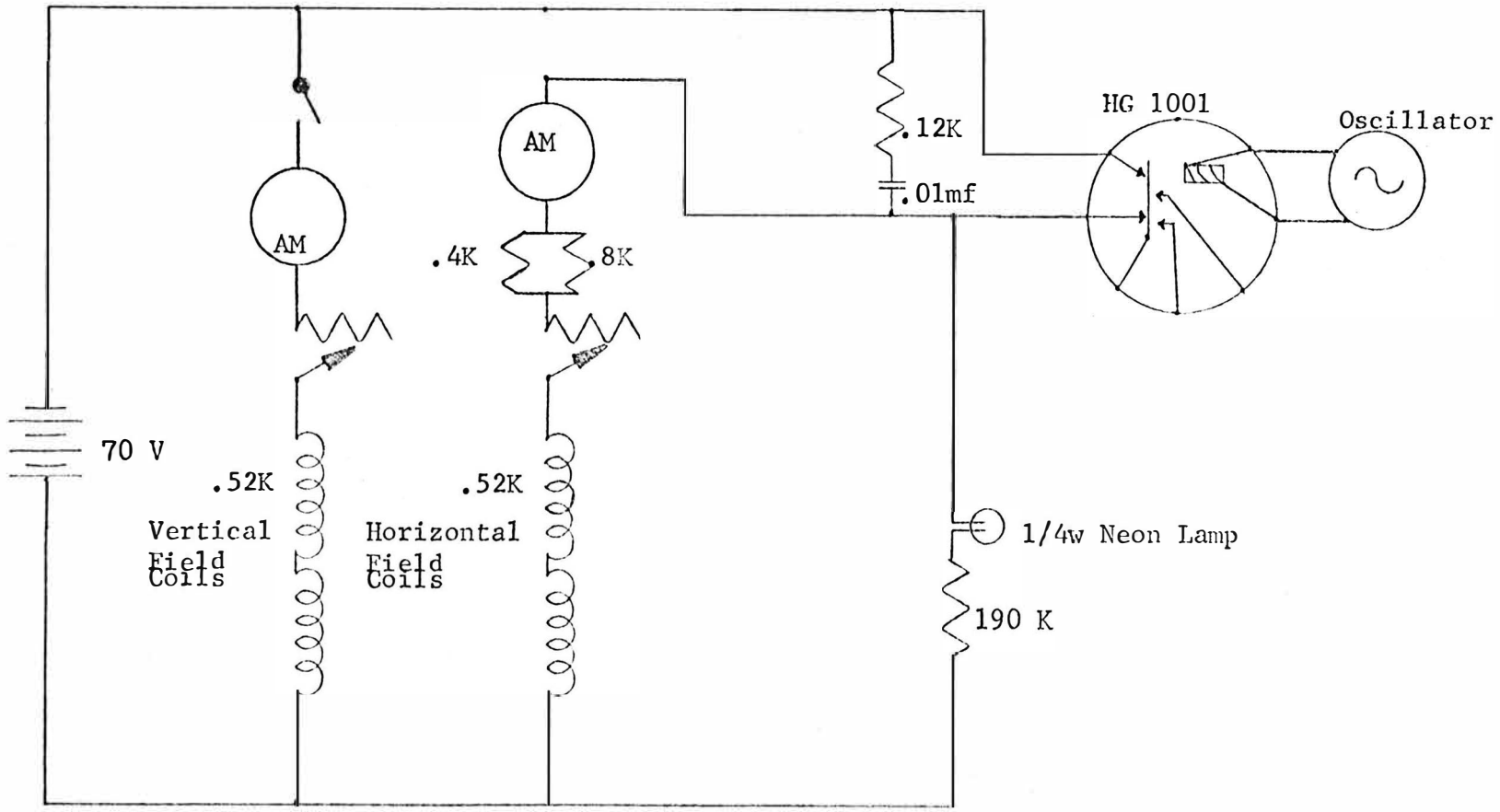
Power Supply

Figure 13

Figure 13 shows the high voltage D. C. supply that powers the final amplifier stage of the oscillator. The oscillator was designed to be operated at any power level from a few watts to about three hundred watts. It was found that best operation of the light source occurred at power outputs around thirty-five watts. It was also found helpful to pass a cool stream of air through the light source housing. This keeps the light source temperature in a range that produced good operating characteristics.

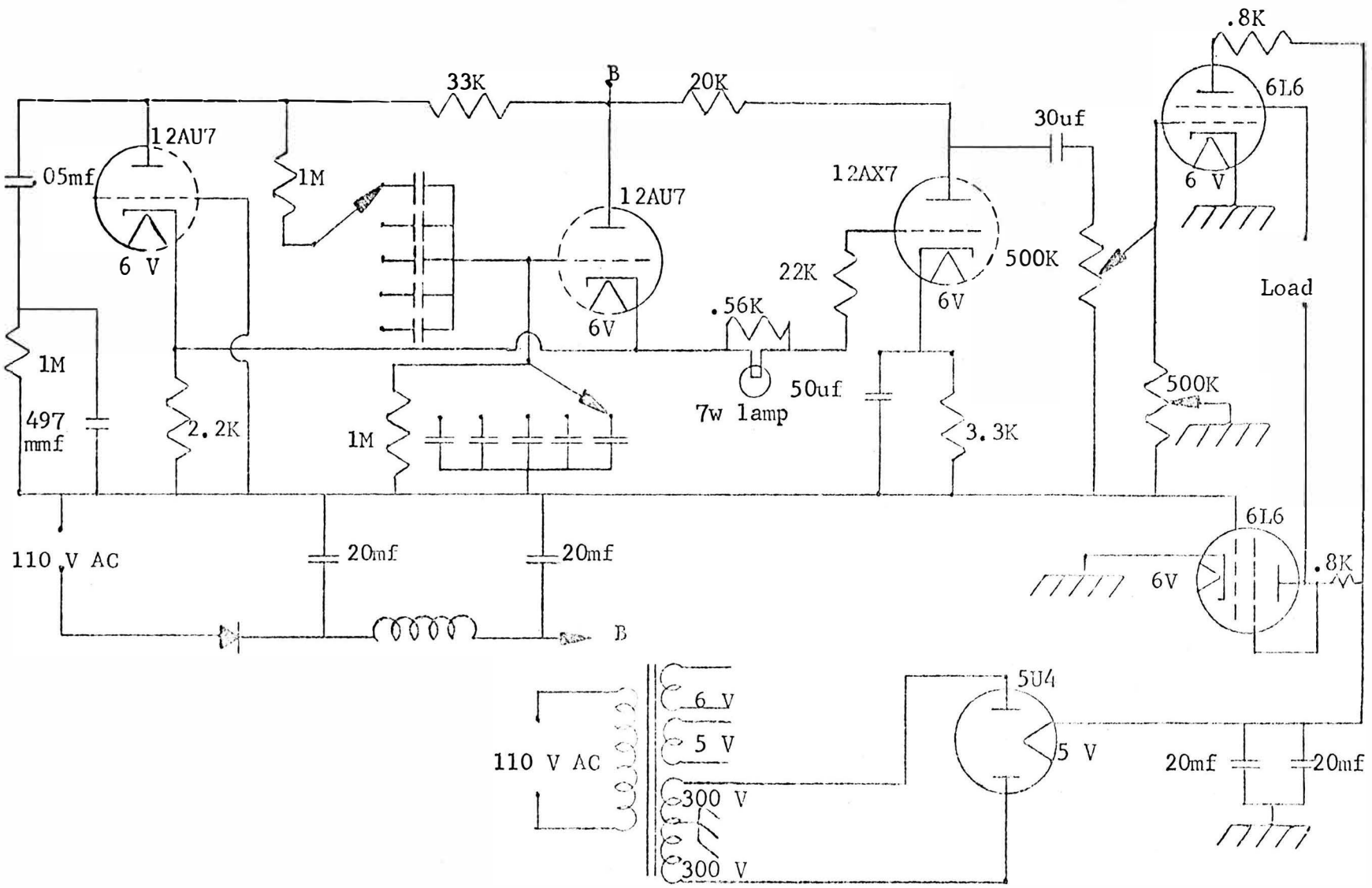
Figure 14 shows the control system for the magnetic fields. The battery supply was a series of six volt storage cells which provided a very stable source of direct current for periods of several hours. Plate I shows the coils which were two feet on a side with a distance of one foot between each of the two coils comprising the set. The coils in the horizontal planes were adjusted so as to just cancel the earth's vertical field (.127 amps). The coils in the vertical planes were situated so that the earth's horizontal field was perpendicular to their plane, and the current passing through them (.107 amps) was adjusted such that it exactly reversed the direction of the earth's field in the center of the coils, but it did not change the magnitude of the field. Only the two outside coils were used, the center one in the vertical plane was not connected.

Figure 15 is the schematic diagram of the oscillator used to drive the mercury wetted relay that turned the horizontal field coils on and off. The oscillator was designed to produce a sine wave of frequency 6 cycles per second to 0.1 cycles per second. A small box containing a pilot light can be seen in Plate I between the table edge and the magnetic field coils. Its purpose was to tell the operator



Controls For Magnetic Fields

Figure 14



6 to .1 cps Oscillator
Figure 15

the direction and moment of field reversal. When the light was on, there was current passing through the horizontal field coils and when the light was off, there was no current in these coils. The field reversed in about 10^{-5} seconds and this was one limitation on the accuracy of the data taken.

Time was required for the resistors to reach equilibrium before attempting to make fine adjustments on the field strength. As with all resistors, they slowly heated and their values changed, thus it was important to give them at least fifteen minutes to stabilize. The Bell "120" Gaussmeter provided a fairly accurate method of calibrating the fields. The Gaussmeter was very sensitive below .01 gauss and while the scale gave readings to .001 gauss, it is doubtful that it had an accuracy greater than .01 gauss. The field used was about .5 gauss and reversing the field to an accuracy of .01 gauss gave a field reversal within 2% which was sufficient for this experiment.

The band pass width on the preamplifier was made as narrow as possible to filter out unwanted signals and yet pass the signal of interest. Even though the light source had a much lower noise level than the commercial Osram spectral lamp, it still had a considerable amount of random fluctuations.

The oscilloscope camera used Polaroid film, this proved to be convenient, for it was known within seconds if all adjustments had been made properly. The camera had the added advantage that the trace being photographed could be viewed while it was being photographed. A little practice was required in synchronizing the camera shutter with the field reversal, but the practice was time well spent since shorter exposures had less random fluctuations on them.

The following procedures are suggested as a count down guide for operating the optical pumping equipment:

A. Begin heating the resonance cell. Best results are obtained when the cell is heated to a high temperature, about 100° C, and then let fall slowly to the working temperature range.

B. Connect the battery supply for the magnetic fields.

C. Turn on the oscillator that operates the field switch.

D. Turn on power supply and oscillator for the light source. Give mercury rectifiers about fifteen minutes to warm up.

E. Connect photocell battery.

F. Turn on the preamplifier.

G. Turn on the oscilloscope.

H. Turn up variac on the power supply, tune the oscillator and fire the rubidium lamp. (peak voltage)

I. Turn on the cooling air supply for rubidium lamp.

J. Check and adjust all variables.

VI. EXPERIMENTAL DIFFICULTIES

The effect that resulted from the reversal of the magnetic field was an extremely difficult process to study experimentally. Since the vehicle used to communicate information about the relaxation process was the transmitted light beam, it was necessary to develop a sensitive detection system to observe this phenomon.

Originally, the detection system used to monitor the light beam was a 1P40 photocell, whose output was amplified by a 100 gain miniature preamplifier. The amplification of the preamplifier was found to be too low to detect the decrease in transmitted light intensity that occurred when the field was reversed.

It was then decided to use a Tektronix's type 122 preamplifier for several reasons. First, this preamplifier could be operated as either a hundred or a thousand gain preamplifier. Second, it allowed one to choose narrow frequency pass bands. Third, if one wanted to develop a system to sample the light beam before it interacted with the resonance cell, the random fluctuations in the light source could be eliminated from the oscilloscope trace.

When the type 122 preamplifier was installed, nothing could be detected. After a considerable amount of investigation, it was found that the preamplifier could not operate with a direct current input greater than one tenth of a volt and the photocell had a direct current output of several volts. This problem was rectified by using a capacitor output on the photocell so that its output showed only changes in the intensity of the transmitted beam.

The next problem encountered was that of stray light. Room lights produced such a large effect that it was found necessary to work in the dark. Even working with room lights out did not prove satisfactory since the pilot lights on the electrical components, reflections from the oscilloscope screen, and the glow from vacuum tubes produced large amounts of background noise on the oscilloscope trace. It finally became necessary to enclose the optical system. The photocell, light source, and resonance cell were already enclosed in metal containers, so it was only necessary to use cardboard mailing tubes between these pieces of apparatus to complete the enclosure. This arrangement can be seen in Plate I.

With a detection system that was sensitive to 10^{-5} volts, careful grounding was necessary. The best rule to follow was to ground everything. All the electrical components, which consisted of the aluminum frame for the field coils, the housings for the light source and photocell, and the switching circuit for the magnetic fields, were grounded. It was found by carefully grounding everything in sight that the background fluctuations were reduced by a factor of twenty or more. Even with these precautions, it was still easy to detect someone working with electrical components within a few hundred feet. It was found best to take data late at night.

The light source probably created the most time consuming problem. The commercially available Osram lamp when operated on direct current as suggested,was completely useless. The operating characteristics were poor for two reasons. First, the lamp fluctuated so much that one could never find the small signal produced when the field was

reversed. Second, the lamp produced resonant lines that were very badly self reversed. Self reversal in spectral lamps became appreciable when the discharge column was more than a millimeter from the outer glass wall of the discharge tube and when the temperature of the lamp ran high.

The rubidium light source described in the discussion of the apparatus minimized these poor features of commercial spectral lamps. In this lamp the discharge took place close to the glass cell, it was air cooled to prevent pressure broadening of the resonance line and it was excited by rf energy since a rf discharge was found to give more stable operation.

Another problem was in the method used for producing circularly polarized light. A quarter wave plate, when set perpendicular to the beam of polarized light with its principal direction at 45^{o} to the azimuth of polarization, retards one component of the polarized light until its phase is 90^{o} behind the phase of the other component and it thus produces a circularly polarized light beam. It was discovered that the polaroid which was used first was transparent to the infrared exciting light, thus the light was not being polarized. A polaroid effective in the infrared was substituted for the defective one and a commercial mica quarter wave plate was used for the 7948A line.

The vacuum system used to evacuate the resonance cell proved to be unsatisfactory. The best vacuum that was attainable was 7×10^{-6} mm of Hg pressure and the vapor pressure of the rubidium in the resonance cell is only 3×10^{-6} mm of Hg pressure at 55^{o}C. Thus it can be readily seen that impurities created a problem. Most of the impurities

were flushed out by evacuating the system, then filling the system with several thousand times more pure argon than there was residual gas present in the cell and finally re-evacuating the system. The argon served to pump out a large portion of the residual gases with it, but a problem remained. The resonance cell had been exposed to the atmosphere for a long time before it was attached to the vacuum system and it was difficult to drive the absorbed gases from the walls of the cell. If they were not removed, these gases would continue to slowly escape from the walls of the cell and contaminate the cell after it had been sealed from the vacuum system. The glassware was repeatedly outgased by heating while the cell was attached to the vacuum system until no rise in pressure could be observed from the repeated heatings.

The other problem in constructing the cell was the possibility of mercury contamination. At room temperature mercury has a vapor pressure of about 10^{-3} mm of Hg pressure. The gauges on the vacuum system would not record the mercury pressure because they were situated between cold traps that froze out the mercury. In order to eliminate this Hg contamination, a second cold trap was inserted between the resonance cell being constructed and any source of mercury. This was done and all the glassware around the resonance cell was heated in an attempt to drive any mercury back to the cold trap.

It was assumed that the vapor pressure of the rubidium could be found by measuring the temperature of the resonance cell. It was of interest to study the relaxation effect as a function of rubidium vapor pressure (or temperature), thus it was important to develop a method of controlling the temperature of the resonance cell. One

method attempted was circulating water whose temperature was thermostatically controlled around the cell. The cell was housed in a brass tank fitted with two windows that allowed the incident beam of resonance radiation to pass through the water and cell. While the cell temperature could be controlled very accurately and uniformily, several other problems arose. First, bubbles formed and interrupted the incident beam and these caused fluctuations in the transmitted light beam intensity. The water apparently absorbed and scattered some of the transmitted light. Finally, the windows used in the tank were strained causing depolarization of the circularly polarized light. It was also felt that the water would be a depolarizing agent on the light. When all these factors were considered, it was decided to discard the water system and use the method described in the apparatus section.

The resonance cell itself had been annealed to remove all strains. Any strains in the glass that the light beam traversed before it interacted with the rubidium gas in the cell had a possibility of depolarizing the light.

The magnetic fields also posed several problems. The problem of calibrating them was resolved when a gaussmeter was obtained. The second problem was that of controls. The field windings were energized by a series of six volt storage cells and the current passing through the coils was adjusted by variable resistors. The temperature of the resistors heated and changed value. The best method found to minimize this problem was to use as few batteries as possible. This decreased the voltage drop across the resistors, decreased the power that had to be dissipated by the resistors, and thus minimized the temperature and resistance changes.

The next problem was that connected with the rapid reversal of the horizontal magnetic field. Since the horizontal component of the earth's field was used in one direction and then the total magnetic field along the direction of the earth's horizontal component was reversed by passing a current through two coils, it appeared that a simple switch would suffice to turn the current on and off. A problem arose in that there was a spark caused by the switch contacts on opening and closing and this was seen by the detection system and finally appeared as a spurious optical pumping trace on the oscilloscope. Neither grounding nor shielding the switch eliminated the problem. It was for this reason that the complicated automatic switching system described was developed. The shielded mercury wetted relay, type HG1001, produced no problems.

VII. RESULTS

Table III is a tabulation of two sets of data. First, the optical pump was operated with all variables held constant except the intensity of the light incident on the resonance cell. The light intensity was varied by adjusting the power to the light source. Six photographs were taken of the repopulation signal on the oscilloscope with six different values of power input to the light source. An example of the relaxation signal, as viewed on the oscilloscope is shown in Plate III. Since the light intensity during optical pumping could not be measured, it was necessary to measure it as a separate set of data. This was done by setting the rf discharge supply at the same operating conditions as was used in the pumping experiment, then monitoring the light intensity as it was interrupted by a camera shutter. Six photographs were taken of the oscilloscope trace produced by the photocell monitoring the interrupted light beam. Since the shutter was operated the same way each time it was used, the pulse heights, recorded photographically, were proportional to the intensity of the interrupted light beam. It was only necessary to have relative values of intensity since the intensity was extrapolated to zero when it was plotted versus $1/\tau$. An example of the pulses recorded when the shutter was opened and closed is shown in Plate IV.

Table III give the data obtained from six photographs of the relaxation signal as a function of the rubidium resonance cell temperature. All variables were kept constant except the cell temperature. The cell itself was heated to about $100^{\circ}C$ and then its temperature

Data Set I			
Rubidium cell temperature °C	Relative light intensity	Pulse height 10^{-2} volts	τ 10^{-3} sec.
64	1.00	17.0	3.25
64	1.13	17.2	3.50
64	1.27	17.5	3.40
64	1.35	16.8	3.25
64	1.51	16.2	2.80
64	1.75	14.5	2.25

Table II

Data Set II			
Rubidium cell temperature °C	Relative light intensity	Pulse height 10^{-2} volts	τ 10^{-3} sec.
87	1	4.2	1.75
76	1	6.5	3.50
64	1	6.0	3.10
62	1	5.6	2.84
59	1	5.1	2.20
48	1	2.1	2.00

Table III

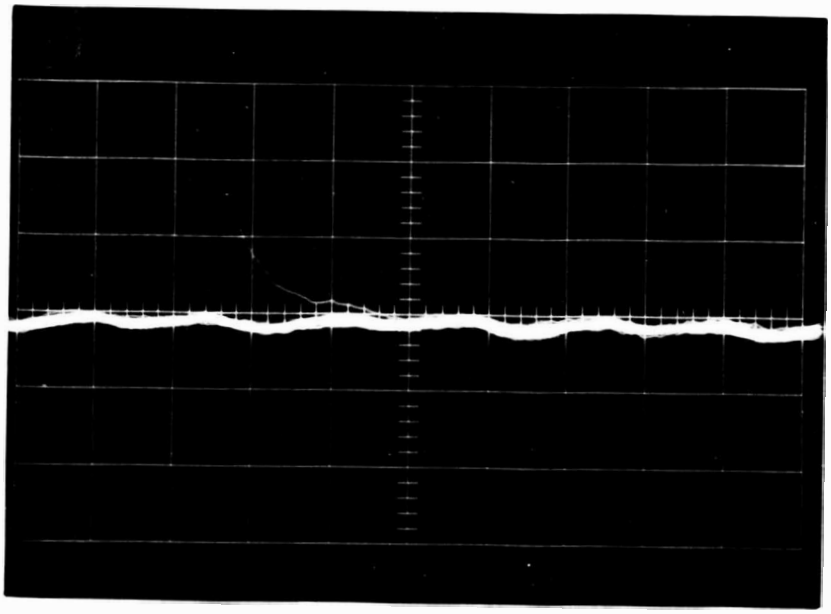


Plate III. Relaxation signal

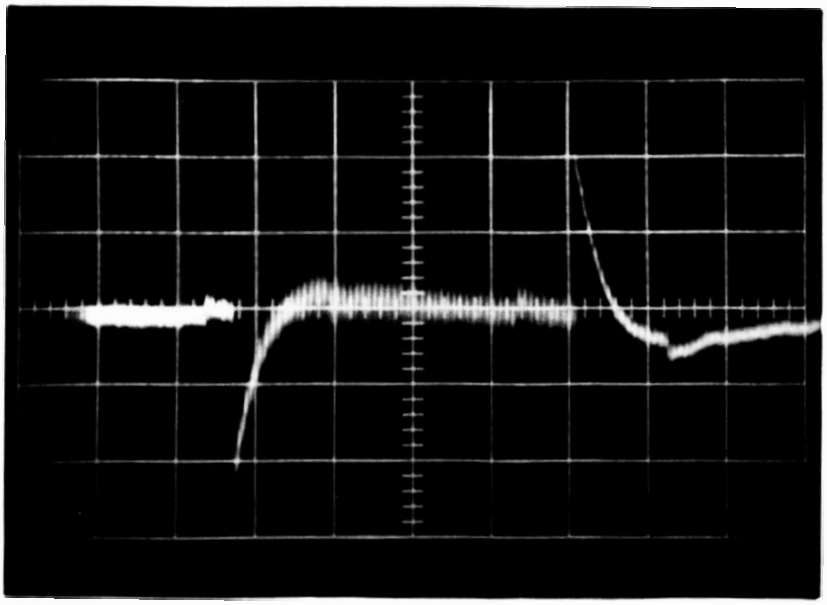


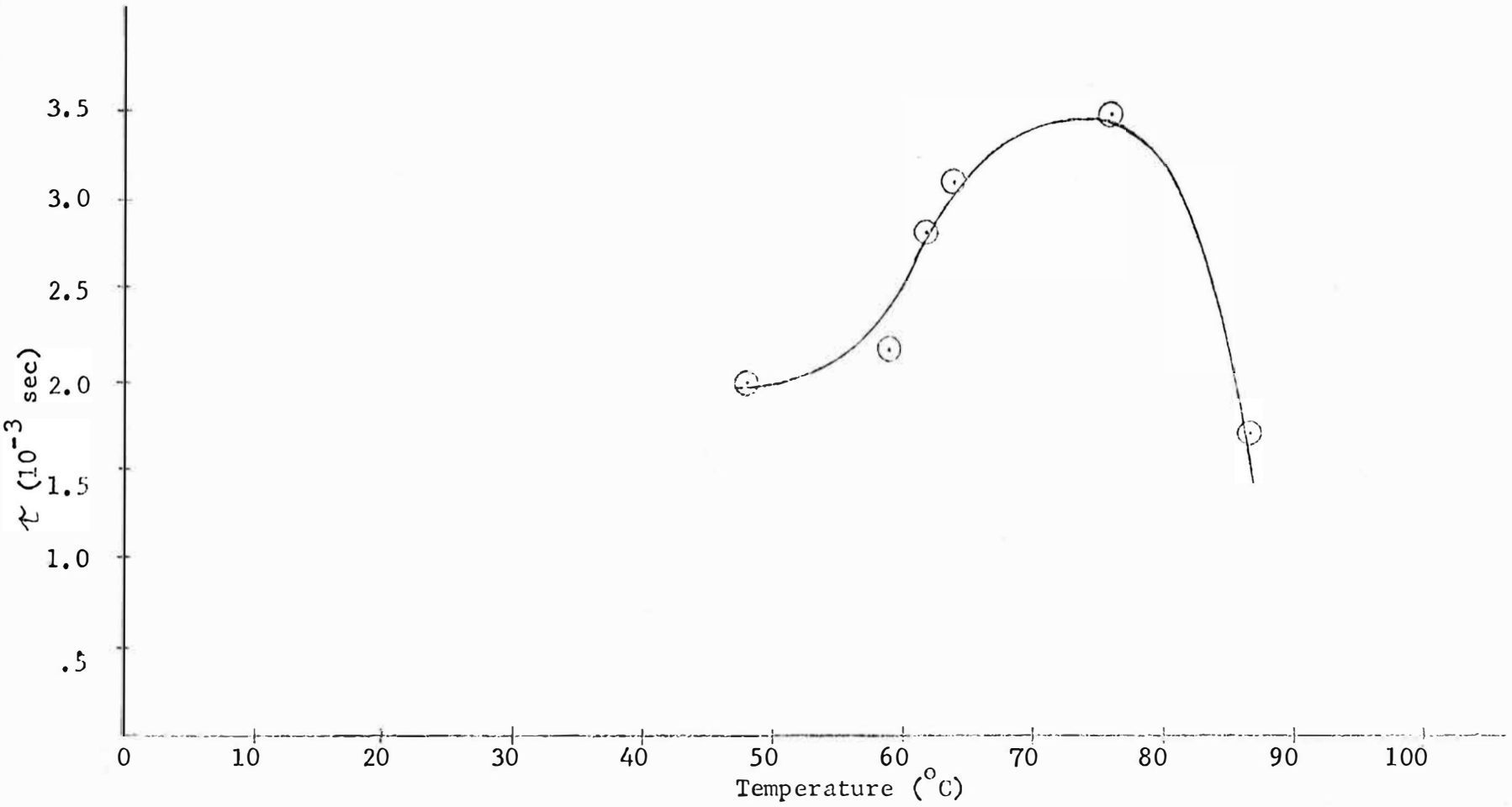
Plate IV. Intensity data

was allowed to drop slowly. When the relaxation trace was observed, the data were taken. The temperature readings were obtained from the thermocouples attached to the cell, using an ice bath reference junction and a potentiometer to record the potential developed.

Figure 16 is a graph of the total relaxation time versus temperature and was obtained from the data in table III. Figure 17 was also plotted from the data in table III and is a plot of the pulse height versus temperature of the resonance cell.

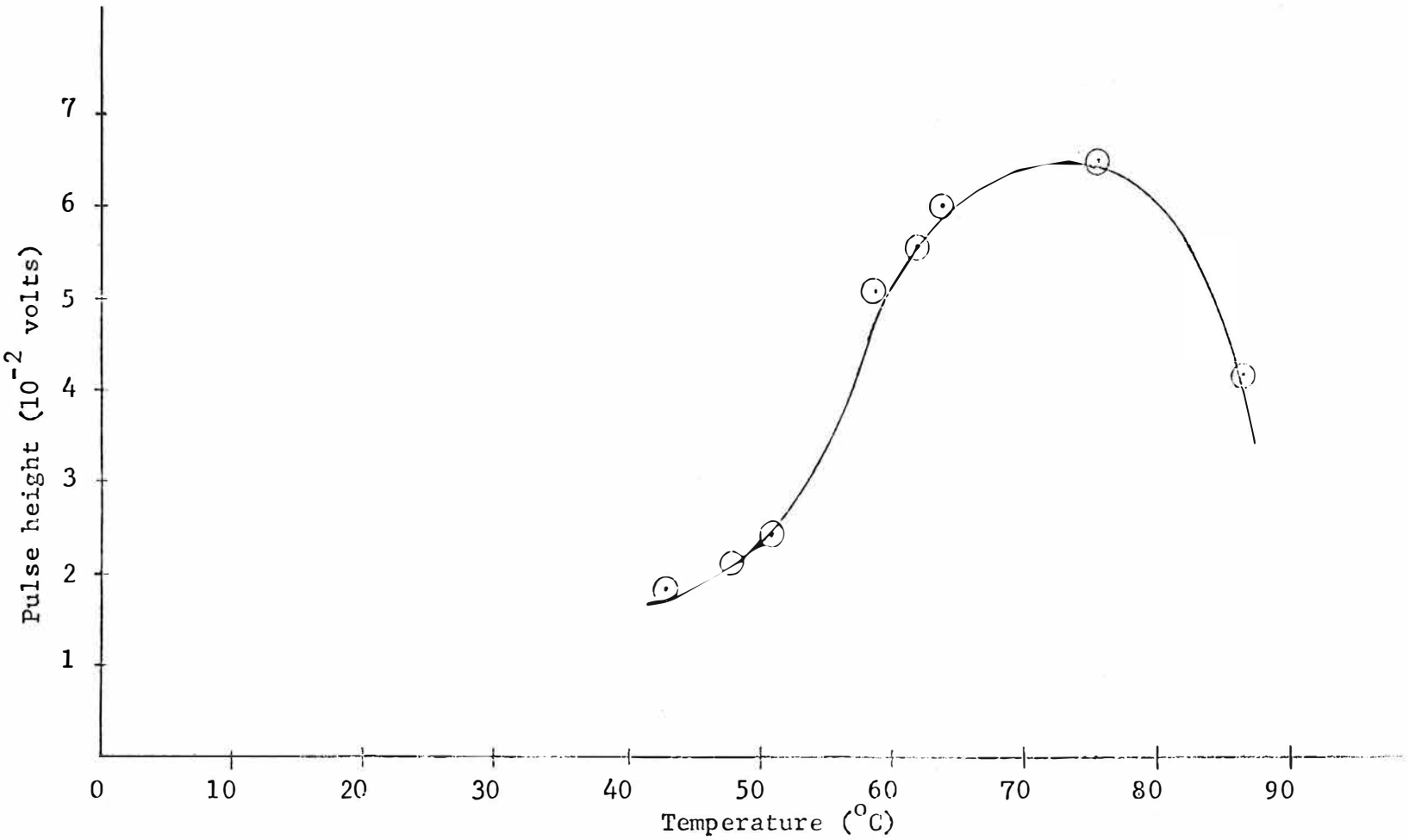
Figure 18 is a plot of the relative intensity of the rubidium light source versus $1/\tau$. This graph was extrapolated to zero and the thermal relaxation time for zero light intensity for the system was found to be 3.72×10^{-3} seconds.

The method used to determine τ from the photographs was that described earlier. The Polaroid photographs of the oscilloscope traces were rephotographed on 35 mm film and projected on a large screen. It was felt that more accurate measurements of τ and the pulse height could be obtained in this way.



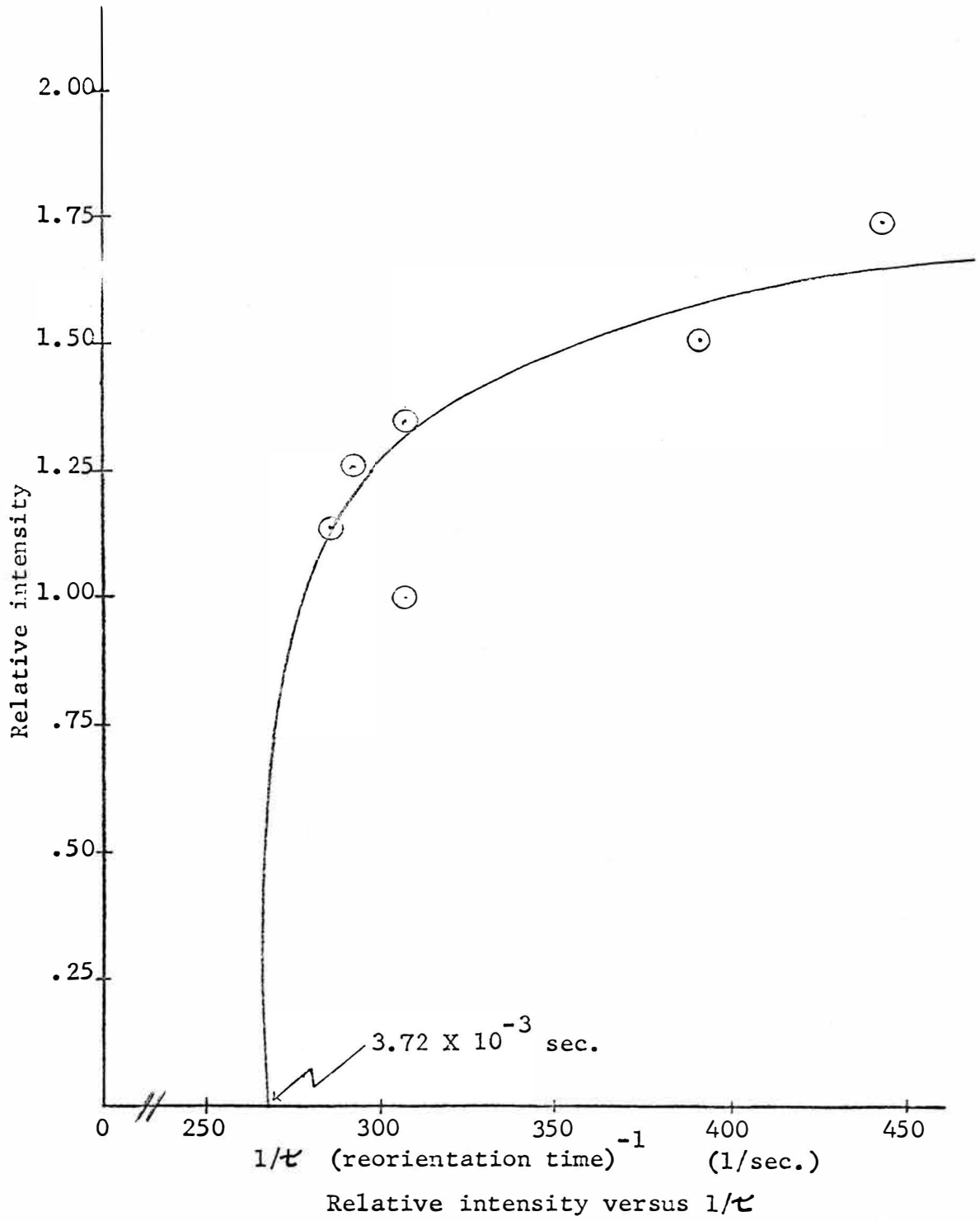
τ versus temperature

Figure 16



Pulse height versus temperature

Figure 17



Relative intensity versus $1/\tau$

Figure 18

VIII. DISCUSSION OF RESULTS

From the plot of intensity versus the reciprocal of the repopulation time, Figure 18, it was determined that the thermal relaxation time for zero light intensity was 3.72×10^{-3} seconds. The portion of the curve for the highest intensities did not level off completely. Since increasing the light intensity shortened the repopulation time, it was assumed that the incident intensity was not high enough to completely pump the levels to their maximum population differences.

To make the extrapolation to zero intensity more accurate, it would be desireable to have more points in the low intensity range. In fact, as explained earlier, it was only at the low intensities of the incident light that the relaxation process became important. For reasonably large values of incident light, the reorientation caused by optical pumping would be too fast to make measurements that would contain a significant factor due to the relaxation time.

Figure 17 is a graph of the relative intensity of the absorbed light measured from the magnitude of the dip in light intensity versus the temperature of the resonance cell for a constant intensity of incident light. This dip in the intensity, caused by the rubidium atoms absorption of the transmitted beam, was proportional to the degree of orientation and the number of atoms pumped. From this graph it was concluded that the optimum resonance cell temperature for maximum absorption was about 74 degrees centigrade. The shape of the curve may be explained in the following way:

The basic effect of the temperature was to control the vapor pressure and hence the density of the alkali atoms. At low temperatures the vapor pressure was low and therefore there were fewer atoms which could be pumped and consequently the absorption of the incident light was low. Then as the temperature increased, an increase in the absorption was noted as more rubidium became available for pumping. As the temperature and hence the density of rubidium atoms continued to increase, another effect became significant. Photons became trapped in the gas resulting in a decrease in the absorption of the transmitted beam. This could be explained by considering the effect of the trapped resonance radiation. The incident right circularly polarized light could only produce transitions from the ground state corresponding to $\Delta M_F = +1$, but the re-emitted radiation from the rubidium atoms contained all components, right and left circularly polarized as well as linearly polarized light. When the atoms reached a temperature of approximately 74 degrees centigrade, their density became great enough to increase significantly the probability of re-absorbing this internally emitted radiation. The result was deorientation of the Zeeman hyperfine levels. Thus when the magnetic field was reversed, the probability of populating the more highly absorbing negative states became less that it was before trapping of resonance radiation became a large effect. Thus the amount of the incident light absorbed at the moment of field reversal became smaller with increasing temperature.

Figure 16 is a graph of the repopulation time versus cell temperature plotted from the same set of data as Figure 17. This curve

shows that a maximum repopulation time occurs at approximately 74 degrees centigrade. It is felt by the author that the processes which tended to keep the atoms from reorientating were at a maximum at this temperature. Above 80 degrees centigrade the relaxation time apparently decreased rather suddenly. This could be explained in a fashion similar to the discussion given for Figure 17. The density of atoms increased and absorption of the emitted photons in the resonance cell increased. Since the atoms excited by the incident right circularly polarized light could emit all Zeeman hyperfine components, deorientation or depopulation could be caused by the left circularly and linearly polarized components. In a resonance cell that has a relatively small percentage of the atoms oriented, one would expect the repopulation time to be short. For a temperature below the maximum value, the repopulation time was small because there was a small density of rubidium atoms and thus the pumping time became short.

In terms of the rate equation:

$$1/\tau = 1/\tau_P + 1/\tau_R \quad \text{or, rearranging,} \quad \tau = \tau_R\tau_P/\tau_P + \tau_R$$

it can be seen that if the relaxation processes tending to deorientate the aligned atoms remains essentially constant and the pumping time is increased, as was the case when the density of rubidium atoms increased with increasing temperature, the total reorientation or repopulation time for the alkali atoms will increase. Then if it is assumed that the pumping time leveled off because of trapped resonance radiation at high rubidium temperatures and relaxation processes became a significant factor, where the relaxation time would decrease with increased imprisonment of resonance radiation, these effects would cause a

decrease in the total repopulation time for high rubidium temperatures. Thus on the basis of the rate equation, the shape of the curve can then be explained.

It would appear from Figure 16 that a leveling off occurred around 60 degrees centigrade, but it was difficult to make any definite conclusions from this curve since the number of plotted points was too small to justify completely the shape of the curve.

IX. CONCLUSIONS

The light source developed for this work proved to be superior to the commercial Osram lamp, however, there is still room for improvement. The signal to noise ratio in some of the data photographs was so poor that no information could be derived from them. There are two possible approaches for improving this situation.

The full capabilities of the Tektronix preamplifier were not fully exploited. By building a second detection system identical to the present one, the random fluctuations in the light source could be eliminated from the oscilloscope traces. In optical pumping experiments the desired signal is superimposed on the undesirable signal light source noise. By connecting one input of the preamplifier to the source of desired signal and the other to the undesired noise signal, the undesired noise signal will be attenuated in the preamplifier by outphasing. Thus if one detection system is operated as in the experiment and the other detection system samples the light beam before it interacts with the gas in the resonant cell, most of the random fluctuations in the oscilloscope trace will be removed.

The second possibility for improving the signal to noise ratios would be to construct a spherical light source as suggested by Brewer.[13] This would not be very difficult since the power requirements for his type of light source would be satisfied by the electrical components in the power supply used to operate the author's light source.

A large portion of the deorientation of the aligned rubidium atoms in the experiment was caused by cell wall collisions. Since it

is not the deorientation caused by collisions with the cell wall that is usually of interest, it would be useful to consider using a long saturated hydrocarbon chain substance for wall coatings. Care must be taken in choosing wall coating materials to be certain that the substance used is pure and has a very low vapor pressure in the temperature ranges of interest.

The experimental work reported in this thesis points out the necessity of considering the effect due to trapped resonance radiation within the resonance cell. This phenomenon is worthy of future investigation. Franzen[4] and others have studied the relaxation processes caused by collisions between aligned rubidium atoms and the cell wall and buffer gas. He did this by using cylindrical geometry and minimizing the effect of trapped resonance radiation. It would be of interest to investigate the effect of trapped resonance radiation and complete the study of the relaxation processes.

BIBLIOGRAPHY

1. H. G. Dehmelt (1957), Phys. Rev., 105, 1487.

2. J. P. Wittke and R. H. Dicke (1956), Phys. Rev., 103, 670.

3. W. E. Bell and A. L. Bloom (1957), Phys. Rev., 107, 1559.

4. W. Franzen (1959), Phys. Rev., 115, 850.

5. G. Herzberg (1944), Atomic Spectra and Atomic Structure, Dover Publications, New York.

6. H. G. Kuhn (1962), Atomic Spectra, Academic Press, New York.

7. Mitchell and Zemansky (1934), Resonance Radiation and Excited Atoms, MacMillan Co., New York.

8. G. V. Skrotshii and T. G. Izyumova (1961), Soviet Physics Uspekhi, 4, 177.

9. Dicke, Carver, Alley and Van der Ven (1957), Final Report to the U. S. Army Signal Corps Engineering Laboratory, (Unpublished).

10. P. L. Bender (1958), Proceedings of the 12th Annual Frequency Control Symposium, Asbury Park, New Jersey, (Unpublished).

11. R. A. Bernheim (1962), Journal of Chemical Physics, 36, 135.

12. J. Brossel (1960), Quantum Electronics, (a symposium), Columbia University Press, New York.

13. R. G. Brewer (1961), The Review of Scientific Instruments, 32, 1356.

14. R. G. Brewer (1962), Journal of Chemical Physics, 37, 2504.

15. A. Kastler (1957), Journal of the Optical Society of America, 47, 460.

16. P. Pringsheim (1949), Fluorescence and Phosphorescense, Interscience Publishers Inc., New York.

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