

# Optical Pumping of Helium in the $^3S_1$ Metastable State\*†

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The alignment of  $\text{He}^4$  atoms in the ( $n=2$ , metastable)  $^3S_1$  state is described. Metastable atoms are produced by an rf discharge in a glass tube containing a few mm of pure helium, and the one micron pumping light ( $2^3P-2^3S$ ) is provided by a helium lamp. A resonance signal is obtained from radio frequency disorientation by monitoring the transmitted pumping light. The double maximum line shape of this signal for strong rf magnetic fields is discussed. Included also is a discussion of the angular dependence of the signal when unpolarized light is used and an explanation of the inversion of the resonance signal for certain densities of the metastable helium atoms.

The measured relaxation time of the oriented metastable atoms in the discharge is about  $2.5 \times 10^{-4}$  second and the pumping time is about a millisecond. A method is proposed and initial measurements are given for the cross section for destruction of metastable helium atoms by collision with foreign gas atoms. The application of optical pumping in helium to the measurement of weak magnetic fields is also discussed.

## I. INTRODUCTION

THE technique of optical pumping<sup>1</sup> has been applied successfully to the alkali metals and mercury to produce an orientation of the magnetic moments of these atoms.<sup>2</sup> In these experiments polarized resonance radiation is passed through a vapor of the metal contained in a vessel usually filled with an inert buffer gas. However, there is difficulty in applying the technique to other elements because of problems associated with the requisite light sources, vapor pressures, and relaxation times.

We recently reported the successful application of optical pumping techniques to helium in the  $^3S_1$  metastable state.<sup>3</sup> The associated phenomena are similar to those observed in the alkali and mercury experiments but there are several distinctive features. In order to provide a self-contained report, our description of these phenomena will include some calculations that parallel those already developed in investigations of the alkalis.

## II. ALIGNMENT OF HELIUM

Figure 1 is a schematic illustrating the basic components necessary for the production and orientation of an ensemble of helium metastables as well as a means for detecting this orientation. The relevant energy levels of a helium atom in an external magnetic field are shown in Fig. 2. Helium atoms are excited by electron

collisions in the discharge tube shown in Fig. 1. Many of the atoms that are excited to higher energy levels or are ionized decay back to the  $^3S_1$  state. Since the radiative decay to the  $^1S_0$  ground state is doubly forbidden, atoms live in this state until they are quenched by some nonradiative process. For the discussion to follow, the  $^3S_1$  state is sufficiently long-lived ( $\sim 10^{-4}$  second) to be considered as the ground state of a new atom which is contained in a buffer gas of ground-state helium atoms.

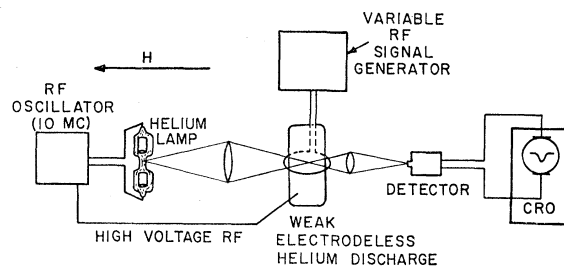


FIG. 1. Schematic diagram of the apparatus.

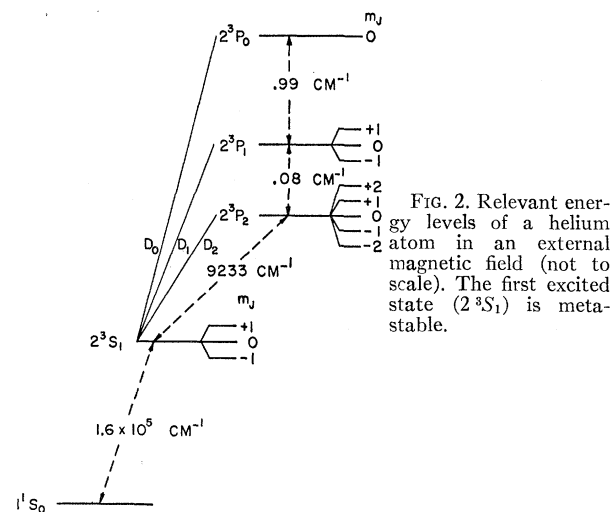


FIG. 2. Relevant energy levels of a helium atom in an external magnetic field (not to scale). The first excited state ( $2^3S_1$ ) is metastable.

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<sup>1</sup> A. Kastler, *J. phys. radium* **11**, 255 (1950).

<sup>2</sup> For review articles the reader is referred to: A. Kastler, *J. Opt. Soc. Am.* **47**, 460 (1957); and A. Kastler, *Report of Scientific Research* (Jouve, Paris, 1958).

<sup>3</sup> P. A. Franken and F. D. Colegrove, *Phys. Rev. Letters* **1**, 316 (1958).

Transitions are induced in these metastable atoms between the  $2^3S_1$  state and the  $2^3P$  states by the absorption of resonance radiation from the bright helium lamp. If there is sufficient mixing in the  $P$  states, a steady state will be reached in which there will be more atoms in those  $^3S_1$  sublevels having the smallest absorption probabilities. The resulting unequal distribution of atoms in the three sublevels will be called an orientation of the helium atoms. This orientation will be referred to as an alignment if the  $m=+1$  and  $m=-1$  sublevels are equally populated, and a polarization if the ensemble of atoms has a net magnetic moment. Unpolarized light is used most frequently and can only produce an alignment.

TABLE 1. Relative emission and absorption probabilities between magnetic sublevels,  $m'$ , of the  $2^3P$  states and the sublevels,  $m$ , of the  $^3S_1$  state of helium.  $D_2$ ,  $D_1$ , and  $D_0$  refer to radiation emitted or absorbed in the transitions  $^3P_2-^3S_1$ ,  $^3P_1-^3S_1$ , and  $^3P_0-^3S_1$ , respectively.

(a) Relative spontaneous emission probabilities											
$D_2$						$D_1$				$D_0$	
$m' \backslash m$	2	1	0	-1	-2	$m' \backslash m$	1	0	-1	$m' \backslash m$	0
+1	6	3	1	0	0	+1	3	3	0	+1	2
0	0	3	4	3	0	0	3	0	3	0	2
-1	0	0	1	3	6	-1	0	3	3	-1	2
(b) Relative absorption probabilities											
Unpolarized light directed along the $z$ axis											
$D_2$						$D_1$				$D_0$	
$m' \backslash m$	2	1	0	-1	-2	$m' \backslash m$	1	0	-1	$m' \backslash m$	0
+1	6	0	1	0	0	+1	0	3	0	+1	2
0	0	3	0	3	0	0	3	0	3	0	0
-1	0	0	1	0	6	-1	0	3	0	-1	2
Unpolarized light in the $xy$ plane											
$D_2$						$D_1$				$D_0$	
$m' \backslash m$	2	1	0	-1	-2	$m' \backslash m$	1	0	-1	$m' \backslash m$	0
+1	3	3	$\frac{1}{2}$	0	0	+1	3	$\frac{3}{2}$	0	+1	1
0	0	$\frac{3}{2}$	4	$\frac{3}{2}$	0	0	$\frac{3}{2}$	0	$\frac{3}{2}$	0	2
-1	0	0	$\frac{1}{2}$	3	3	-1	0	$\frac{3}{2}$	3	-1	1

To detect the orientation of the helium atoms, an rf magnetic field is applied at right angles to the constant field. When the frequency of this rf field is near the resonant frequency representing the energy separation between sublevels,  $\nu = eH/2\pi mc \cong 2.8$  Mc/sec gauss, transitions occur which tend to equalize the populations of these sublevels. The number of atoms in the more strongly absorbing sublevels is thereby increased and more of the pumping light is absorbed. Therefore the intensity of light reaching the detector shown in Fig. 1 decreases at the resonance condition. This resonance signal can be displayed on an oscilloscope as in Fig. 3 by varying the frequency or magnetic field and observing the associated change in light intensity.

The effect of resonance radiation on the metastable

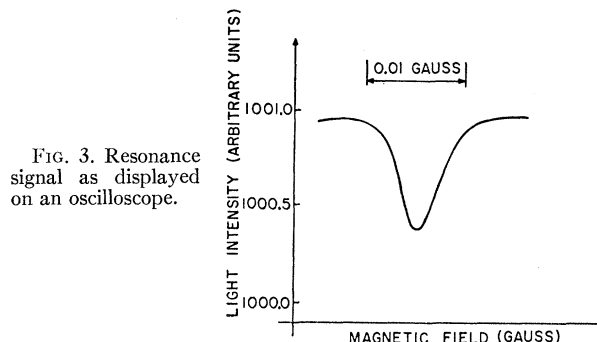


FIG. 3. Resonance signal as displayed on an oscilloscope.

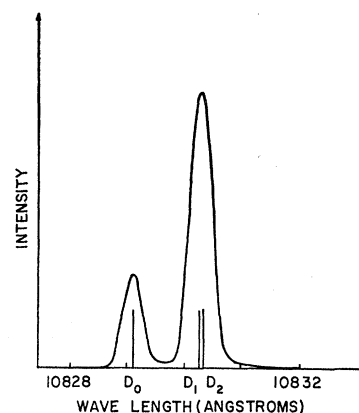


FIG. 4. Spectral profile of the  $2^3P-2^3S$  helium lines.

helium atoms can be analyzed in terms of the relative probabilities for absorption of radiation and the relative probabilities for spontaneous emission from the nine sublevels of the  $^3P$  states to the  $^3S_1$  state. Table I gives these emission probabilities and absorption probabilities for light along the magnetic field and at right angles to the field.<sup>4</sup> Resonance radiation emitted or absorbed by transitions between the  $2^3P_2$  and  $2^3S_1$  states is called  $D_2$  light, that between  $2^3P_1$  and  $2^3S_1$  is  $D_1$  light, and between  $2^3P_0$  to  $2^3S_1$  is  $D_0$  light.

### A. Helium Resonance Radiation

The width of the  $^3P-^3S_1$  resonance lines emitted by most bright helium discharge tubes is of the order of one third of a wave number. Since the separation between the  $^3P_2$  and  $^3P_1$  levels is only about  $0.08 \text{ cm}^{-1}$ , the  $D_2$  and  $D_1$  spectral lines are completely unresolved. The  $^3P_0$  level is one wave number from the  $^3P_2$  and  $^3P_1$  levels, and thus the  $D_0$  line is completely resolved. Figure 4 is a spectral profile of these lines for a helium Geissler tube obtained with a grating spectrometer having a resolution of about a quarter of a wave number.

In discussing the effect of this resonance radiation upon helium atoms, it will be possible to consider  $D_2$  light and  $D_1$  light to be equal in intensity whenever

<sup>4</sup> See E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1951), Chap. 3.

the total light absorption is small. Therefore an arbitrary intensity ratio will be given as  $D_2:D_1:D_0=1:1:K$ . For a more dense ensemble of metastable helium atoms in which an appreciable fraction of the resonance radiation has been absorbed, the absorption Doppler width must be considered. For the one-micron line of helium at room temperature, this is about 1800 Mc/sec or  $0.06\text{ cm}^{-1}$ . Since the  $D_1$  and  $D_2$  absorption lines are separated by only  $0.08\text{ cm}^{-1}$ , it is still a fair approximation to assume that  $D_1$  light and  $D_2$  light are always equally intense even though they are absorbed at different rates.

For most purposes, then, the resonance radiation will be considered as composed of just two components,  $D_3$  and  $D_0$ . The relative probabilities for absorption of  $D_3$  light will be the sum of the probabilities for  $D_2$  light and  $D_1$  light.

From statistical weight considerations it might be expected that the intensities of these two lines would be in the ratio  $D_3:D_0=8:1$ . Spectrographs of bright helium discharges show, however, that the  $D_3$  line is normally only two to three times more intense than the  $D_0$  line.

### B. Rate Equations Without Relaxation

The alignment process for  $^3S_1$  helium may be analyzed in a fashion similar to the treatment of alkali polarization by Dehmelt.<sup>5</sup> It will be convenient to develop the necessary equations in detail.

For an ensemble of  $N$  metastable helium atoms in the discharge tube of Fig. 1, let the number of atoms in each of the substates  $m=+1, 0$ , and  $-1$  be  $n_+$ ,  $n_0$ , and  $n_-$ , respectively, where  $N=n_++n_0+n_-$ . When resonant radiation from the helium lamp is incident upon this ensemble, the populations of the three levels will in general become unequal due to the different rates at which atoms make transitions between the  $^3S_1$  state and the  $^3P$  states. An equilibrium distribution will then be reached in which as many atoms leave a particular level as return to it. The rate at which atoms leave and return to a level will depend upon the transition probabilities and relaxation effects.

Two distinct situations arise in considering the equilibrium populations. At low pressures of the buffer gas (in this case ground-state helium) there is very little disorientation of the orbital angular momentum with respect to spin and a fixed direction during the comparatively short lifetime of the atom in the  $^3P$  states. However at higher pressures (beginning in the range of several mm Hg pressure) the  $P$  states may be considered completely mixed before decay to the  $^3S_1$  state.

For complete mixing, an atom which has been excited to one of the  $^3P$  sublevels will be equally likely to decay

from any  $^3P$  sublevel. It can therefore be seen from the transition probabilities that atoms will decay to the three  $^3S_1$  sublevels with equal probability regardless of the level from which they were originally excited. Let the relative absorption probabilities of the  $m=+1, 0$ , and  $-1$   $^3S_1$  sublevels be denoted by  $R_+$ ,  $R_0$ , and  $R_-$ , respectively. For the moment relaxation phenomena will be neglected and the appropriate rate equations are:

$$dn_+/dt = -CR_+n_+ + (C/3)(R_+n_+ + R_0n_0 + R_-n_-), \quad (1a)$$

$$dn_0/dt = -CR_0n_0 + (C/3)(R_+n_+ + R_0n_0 + R_-n_-), \quad (1b)$$

$$dn_-/dt = -CR_-n_- + (C/3)(R_+n_+ + R_0n_0 + R_-n_-). \quad (1c)$$

where  $C$  is a constant characteristic of the light intensity such that  $CR_+$ ,  $CR_0$ , and  $CR_-$  give the absolute absorption rates for the  $+1, 0$ , and  $-1$  sublevels. At equilibrium  $dn_+/dt = dn_0/dt = dn_-/dt = 0$  and the populations of the three sublevels are in the ratio

$$n_+:n_0:n_- = 1/R_+:1/R_0:1/R_-. \quad (2)$$

For unpolarized light directed along the axis of the magnetic field the relative absorption probabilities can be obtained from Table I, and for a light component ratio  $D_3:D_0=1:K$  the equilibrium populations are

$$n_+:n_0:n_- = 6:(5+K):6. \quad (3)$$

Similarly, for unpolarized light in the  $xy$  plane,

$$n_+:n_0:n_- = (10+2K):(11+K):(10+2K). \quad (4)$$

It can be seen that  $n_+$  and  $n_-$  are always equal for unpolarized light so that the populations can be conveniently described by an alignment parameter,  $A$ , such that

$$A = (n_+ - n_-)/(2n_+ + n_0), \quad (5)$$

where the notation  $n_{\pm}$  means  $n_+$  or  $n_-$ . For the case of no relaxation effects, an expression for the alignment  $A = \bar{A}$  is readily obtained from Eq. (2):

$$\bar{A} = (R_0 - R_{\pm})/(2R_0 + R_{\pm}). \quad (6)$$

$\bar{A}$  represents the maximum alignment that can be obtained for given absorption rates  $R_{\pm}$  and  $R_0$ .

### C. Relaxation Phenomena

Relaxation effects can be introduced phenomenologically into the rate equations by assuming that each of the three sublevels have the same mean lifetime  $\tau_R$ . Then the rate of decay due to relaxation is given by  $(dn_+/dt)_{\text{relax}} = -n_+/\tau_R$ , etc. It is further assumed that the rate at which each of the three levels is populated, as new metastable atoms are formed, is the same and hence equal to  $N/3\tau_R$  where  $N = n_+ + n_0 + n_-$ .

<sup>5</sup> H. G. Dehmelt, Phys. Rev. **109**, 381 (1958).

Therefore the rate equations (1) become

$$\begin{aligned}\frac{dn_+}{dt} &= -CR_+n_+ + \frac{C}{3}(R_+n_+ + R_0n_0 + R_-n_-) \\ &\quad + \left(\frac{N}{3} - n_+\right)\frac{1}{\tau_R}, \\ \frac{dn_0}{dt} &= -CR_0n_0 + \frac{C}{3}(R_+n_+ + R_0n_0 + R_-n_-) \\ &\quad + \left(\frac{N}{3} - n_0\right)\frac{1}{\tau_R}, \\ \frac{dn_-}{dt} &= -CR_-n_- + \frac{C}{3}(R_+n_+ + R_0n_0 + R_-n_-) \\ &\quad + \left(\frac{N}{3} - n_-\right)\frac{1}{\tau_R},\end{aligned}\quad (7)$$

and at equilibrium

$$(CR_+ + 1/\tau_R)n_+ = (CR_0 + 1/\tau_R)n_0 = (CR_- + 1/\tau_R)n_-, \quad (8)$$

so that

$$n_+ : n_0 : n_- = (CR_+ + 1/\tau_R)^{-1} : (CR_0 + 1/\tau_R)^{-1} : (CR_- + 1/\tau_R)^{-1}.$$

In the case when unpolarized light is used, the equilibrium alignment  $A_I$  is

$$A_I = \frac{C(R_0 - R_{\pm})}{C(R_{\pm} + 2R_0) + 3/\tau_R}.$$

The "pumping time" may be defined as an average of the pumping times out of the levels  $n_{\pm}$  and  $n_0$ :

$$\tau_P = 3/(CR_{\pm} + 2CR_0). \quad (9)$$

Therefore

$$A_I = \bar{A}\tau_R/(\tau_R + \tau_P). \quad (10)$$

A general treatment for the case of no mixing or partial mixing in the  $P$  states is algebraically complex. For the simple case in which relaxation effects are neglected and the incident radiation is unpolarized and directed along the  $z$  axis, the equilibrium populations of the three levels are in the ratio  $n_+ : n_0 : n_- = 9 : (2 + 2K) : 9$ .

Similarly, for unpolarized light in the  $xy$  plane, the equilibrium populations would be  $n_+ : n_0 : n_- = (13 + 4K) : (20 + 2K) : (13 + 4K)$ . Comparison with (3) and (4) shows that when  $K < 1$  the pumping is more efficient for this case of no mixing.

#### D. Angular Dependence

The resonance signal is obtained by monitoring the light passing through the ensemble of metastable helium atoms. In general, the atoms in the three magnetic sublevels absorb light at different rates which results in unequal populations. The signal is then the difference in the light absorbed under these equilibrium conditions and the light absorbed when an rf field forces

equal population of the levels. The absorption probabilities and thus the population differences will depend upon the character of the incident light and upon its direction relative to the magnetic field.

For unpolarized light the dependence of the signal upon the direction of resonance radiation relative to the magnetic field may be readily found with the following three simplifying assumptions:

1. The light is strictly parallel;
2. There is complete mixing in the  $P$  states;
3. The rf is sufficiently strong to force equal population of the three magnetic sublevels.

For some complexion of light (arbitrary ratio  $D_2 : D_1 : D_0$ ) and for some particular direction relative to the  $z$  axis, let the relative absorption probabilities of the three  $^3S_1$  levels be  $R_+$ ,  $R_0$ , and  $R_-$ . In the case of unpolarized light  $R_+ = R_-$  and  $n_+ = n_-$ . The light absorbed when the rf is not on will be proportional to the number of atoms in each level multiplied by the relative absorption probability for that level:  $\Delta I = C(2n_{\pm}R_{\pm} + n_0R_0)$ . The sublevel populations  $n_{\pm}$  and  $n_0$  can be found from Eq. (8) and the condition  $N = 2n_{\pm} + n_0$ . Then

$$\Delta I = CN \frac{2R_{\pm}(CR_0 + 1/\tau_R) + R_0(CR_{\pm} + 1/\tau_R)}{2(CR_0 + 1/\tau_R) + (CR_{\pm} + 1/\tau_R)}.$$

When the rf is on and mixes the levels such that  $n_{\pm} = n_0 = N/3$ , the light absorbed will be  $\Delta \bar{I} = CN(2R_{\pm} + R_0)/3$ . The signal will be the difference between the light absorbed with rf and without:

$$S = \Delta \bar{I} - \Delta I = \frac{2}{3}CN \frac{(R_{\pm} - R_0)^2}{[2R_{\pm} + R_0 + 3/(C\tau_R)] - (R_{\pm} - R_0)}.$$

Since the intensity of light is proportional to the square of its electric vector, parallel light of intensity  $I$  directed at an angle  $\theta$  to the  $z$  axis is equivalent to light along the  $z$  axis of intensity  $I \cos^2\theta$  plus light in the  $xy$  plane of intensity  $I \sin^2\theta$ . While neither  $R_{\pm}$ ,  $R_0$  nor their difference depend upon the total light intensity, these quantities do depend upon the relative light intensities of the components along the  $z$  axis and in the  $xy$  plane. Therefore the difference in relative absorption probabilities for the angle  $\theta$  is

$$(R_{\pm} - R_0)_{\theta} = (R_{\pm} - R_0)_z \cos^2\theta + (R_{\pm} - R_0)_{xy} \sin^2\theta.$$

It can be seen from Table I that  $(R_{\pm} - R_0)_z = -2(R_{\pm} - R_0)_{xy}$ . Thus  $(R_{\pm} - R_0)_{\theta} = (R_{\pm} - R_0)_z (\cos^2\theta - \frac{1}{2} \sin^2\theta)$ . Since the total relative probability for a transition  $(2R_{\pm} + R_0)$  is independent of the angle  $\theta$ , the angular dependence of the signal is given by

$$S_{\theta} = \frac{2}{3}CN \frac{(R_{\pm} - R_0)_z^2 (\cos^2\theta - \frac{1}{2} \sin^2\theta)^2}{[2R_{\pm} + R_0 + 3/(C\tau_R)] - (R_{\pm} - R_0)_z (\cos^2\theta - \frac{1}{2} \sin^2\theta)}.$$

Under ordinary circumstances  $(R_{\pm} - R_0)_z$  is much less than  $(2R_{\pm} + R_0 + 3/C\tau_R)$  and the angular dependence will be

$$S_{\theta} \propto (\cos^2\theta - \frac{1}{2} \sin^2\theta)^2 \propto (3 \cos^2\theta - 1)^2.$$

We have verified the general features of this angular dependence experimentally.

### E. Apparatus

The equipment necessary for the optical pumping of helium is relatively simple and is for the most part comprised of standard components as indicated in Fig. 1. A few of the more important items will be discussed briefly together with an indication of the possible variations.

1. *Lamps.* Commercially available spectral lamps made by the Osram Company in Germany were used in the initial experiments. However, it was found that lamps with better noise characteristics could be made with two hollow, cylindrical, aluminum electrodes mounted on tungsten rods and enclosed in a glass tube with a narrow constriction between the electrodes (see Fig. 5). These were made in all sizes; but if the lamps were to be operated with a power input near 10 watts, it was found necessary to use electrodes larger than  $\frac{3}{4}$  inch in diameter to avoid sputtering. The lamp used in most of the experiments had electrodes  $1\frac{1}{2}$  in. in diameter and  $1\frac{5}{8}$  in. long. This lamp has been in use for a year with no sign of deterioration. The lamps were filled with helium at a pressure of from 2 to 5 mm Hg and excited at about 6 Mc/sec.

2. *Discharge Tubes.* The discharge tube defines the region in which metastable helium atoms are created and pumped. A great variety of these tubes have been used successfully. Three models are shown in Fig. 6. They range in size from an effective pumping region less than  $\frac{1}{2}$  in. thick up to regions  $2\frac{3}{4}$  in. thick. Good resonance signals have been obtained in these tubes at helium pressures of from  $5 \times 10^{-2}$  mm Hg up to 30 mm Hg. The discharge was generally excited by high-voltage rf fields near 10 Mc/sec, although dc excitation has also been successful. For rf excitation, electrodes were not necessary. Both the intensity of the discharge and the pressure affect the production of metastables, which in turn affects the signal as described in Sec. V.

In the early phases of this investigation, considerable work was done in an effort to obtain extremely pure helium in the discharge tubes. It was discovered later, however, that the electrodes in a helium discharge tube are very efficient getters of impurities. Tubes without

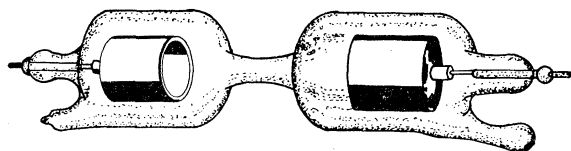


FIG. 5. Lamp used as a source of resonance radiation.

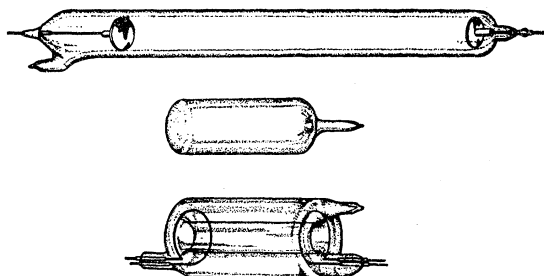


FIG. 6. Examples of discharge tubes.

electrodes can be cleaned up easily by flashing a getter in each sealed off tube.

3. *Detectors.* A variety of detectors were found suitable for monitoring the 10 000 Å helium resonance radiation. The one most frequently employed was a lead sulfide photosensitive conductor (Kodak Ektron Detector). When faster response times were needed a Texas Instruments *n-p-n* diffused silicon photo-duodiode (Type 1N2175) or an S-1 response photo tube were used.

4. *Electronics.* Standard components and circuitry were used throughout. The lamp and discharge tube were excited by a push-pull Hartley type oscillator. High voltage was taken off the tank coil for the weak discharge and a pickup coil provided a higher current for the lamp. The weak rf magnetic field used to disorient the metastable helium atoms was supplied by a General Radio 0.5-to 50-Mc/sec unit oscillator connected to a single loop of wire around the discharge tube.

The output of the detector was amplified by a Tektronix type 122 low-level preamplifier and fed either directly to an oscilloscope or through a narrow-band amplifier to a recorder. Field sweep was provided by 24-in. Helmholtz coils with current supplied by a standard audio oscillator. Other windings on these coils provided a constant field for experiments at other than the earth's field.

## III. PUMPING AND RELAXATION TIMES

### A. Pumping Time

The intensity of the pumping light measured at the discharge tube with an S-1 response vacuum phototube is about 0.5 mw/cm<sup>2</sup>. For one-micron radiation (10 000 cm<sup>-1</sup>) this represents  $2.5 \times 10^{15}$  photons/cm<sup>2</sup> sec. However, these photons are spread over a frequency range of about 10 000 Mc/sec (0.3 cm<sup>-1</sup>) due to Doppler and pressure broadening, while each atom absorbs only photons within its natural line width of  $\Delta\nu = 1/2\pi\Delta t \cong 1.5$  Mc/sec where  $\Delta t$  is the lifetime of an atom of the  $2^3P$  state. Therefore the effective number of photons per square cm per second at the discharge tube is  $\mathcal{N} = 2.5 \times 10^{15} \times 1.5 / 10\,000 \cong 4 \times 10^{11}$  photons/cm<sup>2</sup> sec.

The reciprocal of the pumping time will then be the "cross section,"  $\sigma$ , for absorption of resonance radiation

by an atom multiplied by the intensity of the light. This cross section is of the order of  $2\pi\lambda^2$  which, for one-micron radiation, is approximately  $1.6 \times 10^{-9}$  cm<sup>2</sup>. Thus the pumping time is about  $\tau_P = 1/(\sigma\mathcal{N}) \cong 1.6 \times 10^{-3}$  sec.

### B. Relaxation Time

The lifetime of helium in the  $2^3S_1$  state has recently been measured as a function of pressure by Phelps and Molnar<sup>6</sup> who observed lifetimes as long as 20 milliseconds after the discharge was turned off. However, the significant quantity for our experiment is the lifetime of the metastable helium atoms in a particular magnetic sublevel of the  $2^3S_1$  state while the discharge is on. The helium atoms aligned by optical pumping provide a convenient means for making such measurements.

From Eqs. (5), (6), (7), and (8) an approximate expression may be obtained for the rate of change of the alignment when  $A \ll 1$ :

$$dA/dt = (\bar{A} - A)/\tau_P - A/\tau_R,$$

where  $\tau_P$  is the pumping time,  $\tau_R$  is the relaxation time and  $\bar{A}$  is the equilibrium alignment in the absence of relaxation effects. The equilibrium value,  $A_I$  [Eq. (10)] will then be reached in a characteristic time  $\tau$ :

$$dA/dt = (A_I - A)/\tau,$$

where

$$1/\tau = 1/\tau_P + 1/\tau_R. \quad (11)$$

The characteristic time,  $\tau$ , can be measured by applying an rf magnetic field at the resonant frequency to obtain a nonequilibrium value of  $A$ , and then abruptly turning off the rf field. In practice it was found easier to make a sudden change in frequency of the rf field by adding capacitance to the tank circuit of the rf oscillator by means of a mercury relay. The return to equilibrium when the frequency is suddenly shifted off resonance is observed by monitoring the intensity of the transmitted pumping light. The light absorbed by the metastable atoms is

$$\Delta I = C(2R_{\pm}n_{\pm} + R_0n_0).$$

Equation (5) and the relation  $N = 2n_{\pm} + n_0$  give this absorption in terms of the alignment

$$\Delta I = CN[R_0 + 2(R_{\pm} - R_0)(A + 1)/(A + 3)].$$

Since  $A$  is in general a very small number,

$$\Delta I \cong CN[R_0 + \frac{2}{3}(R_{\pm} - R_0)(1 + \frac{2}{3}A)]. \quad (12)$$

It can be seen from (12) that  $d\Delta I/dt$  is proportional to  $dA/dt$  and thus the light intensity will change with the same time constant as the alignment.

Figure 7 is a reproduction of an oscilloscope trace used to measure  $\tau$ . The rf magnetic field is initially at the resonance frequency and is suddenly shifted in

frequency by 200 kc/sec. The maximum values of  $\tau$  obtained were about  $2 \times 10^{-4}$  sec. For a pumping time of  $1.6 \times 10^{-3}$  sec, the relaxation time,  $\tau_R$ , is found from Eq. (11) to be about  $2.3 \times 10^{-4}$  sec.

### C. Signal Amplitude

The preceding measurements may be checked by calculating the magnitude of the signal from the measured values of  $\tau$  and  $\tau_P$  for comparison with the signal obtained experimentally. The maximum signal amplitude is compared with the total light absorbed rather than the total light reaching the detector. The light absorbed by the metastable helium atoms is easily determined by comparing the light reaching the detector when the discharge is on with the light at the detector when the discharge is off.

The resonant radiation absorbed at equilibrium is found from Eq. (12) to be

$$\Delta I \cong CN(2R_{\pm} + R_0)/3 + 4CN(R_{\pm} - R_0)A_I/9 \quad (13)$$

and the light absorbed when the alignment is zero (for instance, when the rf magnetic field is very strong) is

$$\Delta \bar{I} = CN(2R_{\pm} + R_0)/3 \quad (14)$$

The maximum signal is just the difference between (13) and (14). Therefore the ratio between the maximum signal and the total amount of light absorbed is

$$\frac{\Delta I - \Delta \bar{I}}{\Delta \bar{I}} = -\frac{4}{3} \frac{R_{\pm} - R_0}{2R_{\pm} + R_0} A_I.$$

Equations (6), (10), and (11) give this ratio in terms of  $\tau$  and  $\tau_P$ :

$$\frac{\Delta I - \Delta \bar{I}}{\Delta \bar{I}} = \frac{4(R_{\pm} - R_0)^2}{(2R_{\pm} + R_0)(R_{\pm} + 2R_0)} \frac{\tau}{\tau_P}. \quad (15)$$

For unpolarized light directed along the  $z$  axis and an assumed component intensity ratio of  $D_{\pm}:D_0=2$ , the relative absorption probabilities are  $R_{\pm}=11$  and  $R_0=12$ . Therefore with the values  $\tau=2 \times 10^{-4}$  sec and

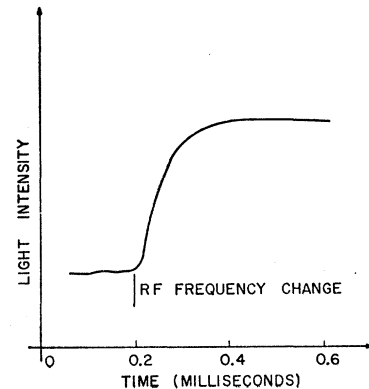


FIG. 7. An oscilloscope trace used in the measurement of the time constant,  $\tau$ .

<sup>6</sup> A. V. Phelps and J. P. Molnar, Phys. Rev. **89**, 1202 (1953).

$$\tau_P = 1.6 \times 10^{-8} \text{ sec},$$

$$(\Delta I - \Delta \bar{I})/\Delta \bar{I} \cong 1.4 \times 10^{-4}. \quad (16)$$

This ratio was determined experimentally using a discharge tube with a helium pressure of 5 mm Hg. When the detector output was 0.3 volt with the discharge off and 0.15 volt with the discharge on, the signal was  $4 \times 10^{-5}$  volt output and the ratio of signal to light absorption is thus

$$(\Delta I - \Delta \bar{I})/\Delta \bar{I} \cong 2.7 \times 10^{-4}.$$

This is in fair agreement with the predicted result (16).

### D. Impurity Effects

It is possible to obtain an estimate of the cross section for quenching of metastables by impurities by letting known quantities of a foreign gas into the system and observing the change in the optical pumping signal. From Eqs. (11) and (15), it can be seen that the signal is proportional to the relaxation time,  $\tau_R$ , as long as  $\tau_P \gg \tau_R$ . Thus if the signal is reduced by one-half when impurities are added, the relaxation time due to collisions with this gas is equal to  $\tau_R$ . Therefore the number of quenching collisions per metastable atom per second with impurities is  $1/\tau_R \cong \sigma V \mathcal{N}$ , where  $\sigma$  is the cross section for quenching,  $v$  is the mean velocity of atoms in the gas, and  $\mathcal{N}$  is the number of foreign gas atoms per cubic centimeter. It was found that the signal was reduced to one-half its original value when about  $10^{-3}$  mm Hg of any impurity was admitted. A partial pressure of  $10^{-3}$  mm Hg represents about  $3.5 \times 10^{13}$  atoms/cm<sup>3</sup>. At a temperature of 300°K the velocity of a helium atom is approximately  $10^5$  cm/sec. Combining these values with a  $\tau_R$  of  $2.3 \times 10^{-4}$  sec gives a cross section  $\sigma \cong 10^{-15}$  cm<sup>2</sup>.

The impurities admitted were argon, xenon, air, and He<sup>3</sup>. This latter gas acts as an impurity because of its nuclear magnetic moment. When a metastable He<sup>4</sup> atom in the  $m = +1$  magnetic sublevel exchanges electrons and metastability with a ground state He<sup>4</sup> atom, angular momentum is conserved and the atom originally in the ground state is now in the sublevel  $m = +1$ . However, when the exchange is with a He<sup>3</sup> atom, the electronic magnetic moment precesses about the resultant of the electronic and nuclear magnetic moments and thus the "memory" of the  $z$  component of electronic spin with respect to the magnetic field can be rapidly lost.

By measuring electron-ion densities, Biondi has calculated the cross section for destruction of metastable helium atoms by collision with argon atoms and obtained a value  $(9.3 \pm 0.8) \times 10^{-17}$  cm<sup>2</sup>.<sup>7</sup> Due to the difficulty of measuring the impurity pressures accurately in our dynamic system our measurements were somewhat crude. However we believe our measurement is significant to within a factor of five, which suggests

<sup>7</sup> M. A. Biondi, Phys. Rev. **83**, 653 (1951).

that a real discrepancy exists and merits further investigation.

### IV. RESONANCE LINE SHAPE

When the rf field is small, the predominant factor contributing to the breadth of the resonance line is the inhomogeneity of the earth's magnetic field in the laboratory. The field gradients appear to be a few milligauss per centimeter in most of the accessible regions of Randall Laboratory. Therefore atoms in different parts of the discharge tube may be in fields that differ by as much as 5 milligauss. This represents a line width of 14 kc/sec. In order for line shape effects due to the rf magnetic field to be predominant, it is then necessary to have line widths somewhat larger.

Other effects contributing to the line width will usually be negligible under these conditions. The natural line width will depend upon the lifetime of an atom in a sublevel of the  $^3S_1$  state. This lifetime will be determined by all the various collision processes in which the metastable atom is removed from the  $^3S_1$  state or in which the spin angular momentum is changed. The lifetime in one of the substates ordinarily appears to be between  $10^{-4}$  and  $10^{-3}$  sec. The line width,  $\Delta\nu$ , for a lifetime  $\tau$  is  $\Delta\nu = 1/\pi\tau$  and therefore the natural line width is between 3200 and 320 cps.

In this section all line-broadening effects with the exception of the rf magnetic field will be neglected. The most striking feature of the rf broadening is that under some circumstances the center of the line is diminished by about 10%, so that a double maximum, or "splitting," occurs. This splitting of the resonance line arises for the same reasons as the similar line shape discussed by Brossel and Bitter<sup>8</sup> in their work on the excited  $^3P_1$  state of mercury. However, the analysis here is somewhat different since the  $^3S_1$  state of helium is considered as a ground state and the alignment of these metastable atoms is detected in a different manner.

To find the line shape it is necessary to determine the dependence of the light absorption upon the population differences between the  $^3S_1$  sublevels. These differences depend in turn upon the manner in which transitions are induced between the sublevels by the rf magnetic field.

The problem is one of an atom in a time-dependent magnetic field which is the resultant of a constant field  $H_0$  and a very much smaller oscillating field  $H_1$  at right angles to  $H_0$ . The effect of this oscillating field upon the atom is very nearly the same as that of a field rotating in a plane perpendicular to  $H_0$ .<sup>9</sup> For such a system Rabi<sup>10</sup> and Majorana<sup>11</sup> have given expressions for the probability  $P(J, m, m', t)$  that a system of total angular momentum  $J$  which is initially in a state described by  $m$  will at a time  $t$  later be in a state  $m'$ .

<sup>8</sup> J. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952).

<sup>9</sup> F. Block and A. Siegert, Phys. Rev. **57**, 522 (1940).

<sup>10</sup> I. I. Rabi, Phys. Rev. **51**, 652 (1937).

<sup>11</sup> E. Majorana, Nuovo cimento **9**, 43 (1932).

These probabilities are polynomials of the form  $\sum_i C_i [f(\omega, H_1) \sin^2(\sim \gamma H_1 t)]^i$  near the resonant frequency, where  $f(\omega, H_1)$  is a function of the strength and angular frequency  $\omega$  of the rf magnetic field  $H_1$ .

When the field  $H_1$  is strong enough to clearly observe the resonance line splitting the period of these polynomials is much shorter than  $\tau_P$  and  $\tau_R$ . Therefore an atom in a given magnetic sublevel will make many transitions to the other two sublevels during the period of time it spends "in" the given state. It is possible then to use the *average* probabilities,  $P(J, m, m')_{av}$ , which can be interpreted as the average length of time an atom "in" the state  $m$  spends in the state  $m'$ .

$$P(+1, 0)_{av} = P(0, +1)_{av} = P(-1, 0)_{av} \\ = P(0, -1)_{av} = x - \frac{3}{4}x^2 \equiv P_0,$$

$$P(+1, -1)_{av} = P(-1, +1)_{av} = (3/8)x^2 \equiv P_1,$$

where  $x = 1/(1 + \delta^2)$  and  $\delta = (\omega - \omega_0)/(\gamma H_1)$ . For example, an atom which is excited to the  $m = +1$  sublevel of the  $^3S_1$  state will spend a fraction  $P_0$  of its time in the  $m = 0$  sublevel and a fraction  $P_1$  of its time in the  $m = -1$  sublevel.

In the following treatment a "thin" layer of metastable helium atoms will be assumed so that multiple scattering of resonant radiation can be neglected and  $R_+$ ,  $R_0$ , and  $R_-$  will be constants throughout the sample. In addition, all other line-broadening effects will be considered negligible compared with the broadening due to the rf magnetic field. In particular,  $\tau$  is assumed infinite. Only the case of complete mixing in the  $^3P$  states will be treated.

The rate Eqs. 1(a), (b), (c) become:

$$\begin{aligned} dn_+/dt &= -Cn_+\Phi_+ + (C/3)(n_+\Phi_+ + n_0\Phi_0 + n_-\Phi_-), \\ dn_0/dt &= -Cn_0\Phi_0 + (C/3)(n_+\Phi_+ + n_0\Phi_0 + n_-\Phi_-), \\ dn_-/dt &= -Cn_-\Phi_- + (C/3)(n_+\Phi_+ + n_0\Phi_0 + n_-\Phi_-), \end{aligned}$$

where

$$\begin{aligned} \Phi_+ &= (1 - P_0 - P_1)R_+ + P_0R_0 + P_1R_-, \\ \Phi_0 &= (1 - 2P_0)R_0 + P_0(R_+ + R_-), \\ \Phi_- &= (1 - P_0 - P_1)R_- + P_0R_0 + P_1R_+. \end{aligned}$$

These  $\Phi$  functions are simply the relative probabilities for absorption of a photon when the rf magnetic field is operative. At equilibrium

$$n_+\Phi_+ = n_0\Phi_0 = n_-\Phi_-, \quad (17)$$

and the total light absorbed will be

$$\Delta I = C(n_+\Phi_+ + n_0\Phi_0 + n_-\Phi_-) = 3Cn_+\Phi_+.$$

The number  $n_+$  may be found from (17) and the expression for the total number of atoms in the ensemble  $N = n_+ + n_0 + n_-$ . Therefore the light absorbed is

$$\Delta I = 3CN\Phi_+\Phi_0\Phi_-/(\Phi_+\Phi_0 + \Phi_+\Phi_- + \Phi_0\Phi_-). \quad (18)$$

Equation (18) constitutes a complete solution for the light absorption as a function of the frequency and

magnitude of the rf magnetic field. This function represents the resonance line shape when the rf magnetic field is so strong that other line-broadening effects are negligible. The line shape will in general depend upon the values  $R_+$ ,  $R_0$ , and  $R_-$ , and for a particular set of these constants a plot of light absorption versus  $(\omega - \omega_0)/\gamma H_1$  may be obtained.

When unpolarized light is used for optical pumping the analysis becomes particularly simple. From an inspection of the relative transition probabilities between  $^3S$  and  $^3P$  states, it can be seen that  $R_+ = R_-$  for any direction of the light. In general,  $R_+$  and  $R_-$  differ from  $R_0$ . It is convenient to describe this difference by  $3\epsilon R$  so that  $R_+ = R_- = R(1 - \epsilon)$  and  $R_0 = R(1 + 2\epsilon)$ . Then the  $\Phi$  functions become

$$\begin{aligned} \Phi_+ &= \Phi_- = (1 - P_0)R_+ + P_0R_0 = R(1 - \epsilon + 3\epsilon P_0), \\ \Phi_0 &= (1 - 2P_0)R_0 + 2R_+P_0 = R(1 + 2\epsilon - 6\epsilon P_0), \end{aligned}$$

and the light absorbed is

$$\Delta I = CNR \frac{1 + \epsilon - 2\epsilon^2 - 3\epsilon P_0 + 12\epsilon^2 P_0 - 18\epsilon^2 P_0^2}{1 + \epsilon - 3\epsilon P_0}. \quad (19)$$

The signal observed will be the difference between the light absorbed at a particular frequency of the rf magnetic field [Eq. (19)] and the light absorbed when the effect of the rf is negligible, i.e., when  $(\omega - \omega_0)/\gamma H_1$  is infinite. For this condition  $P_0 = 0$  and

$$(\Delta I)_{\delta \rightarrow \infty} = CNR(1 + \epsilon - 2\epsilon^2)/(1 + \epsilon).$$

Therefore the signal is

$$S = \Delta I - (\Delta I)_{\delta \rightarrow \infty} = 6\epsilon^2 CNR \frac{(2 + \epsilon)P_0 - 3(1 + \epsilon)P_0^2}{(1 + \epsilon - 3\epsilon P_0)(1 + \epsilon)}. \quad (20)$$

For  $\epsilon$  less than  $\sim 0.1$  the magnitude of the signal is approximately proportional to  $\epsilon^2$  but the line shape is relatively insensitive to  $\epsilon$ . Under the usual conditions in which unpolarized resonance radiation is directed along the  $z$  axis, a reasonable value of  $\epsilon$  is  $\approx 0.01$  for which the points plotted in Fig. 8 were computed.

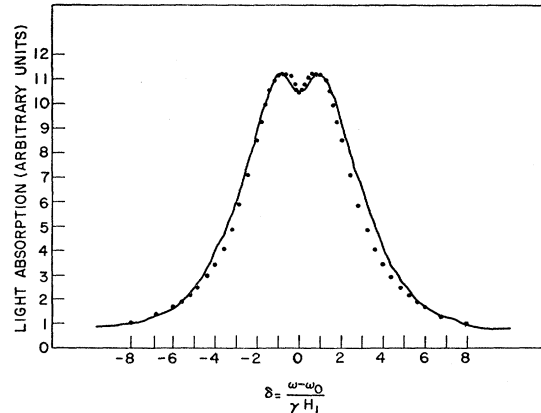


FIG. 8. Experimental resonance line shape for a strong rf magnetic field. The points are from the theoretical line shape.



The experimental curve of Fig. 8 was obtained by setting the frequency at 1.6 Mc/sec and sweeping the field at about 0.05 gauss per minute. The rf was chopped at 79 cps and the light signal passed through a narrow-band amplifier, rectified, and recorded. The separation between the peaks is about 6 milligauss and the width at half maximum is 0.02 gauss. Since the field inhomogeneity over the dimensions of the discharge tube is about one-quarter of the line width, the experimental curve is broader and does not have as sharp a minimum between the peaks as Eq. (20) predicts.

It can easily be shown that for any value of  $\epsilon$  the resonance signal has only the two humps and that the separation of these two maxima depends only upon the strength of the rf field. The extremum points of the signal curve are at  $dS/d\delta = (\partial S/\partial P_0)(\partial P_0/\partial x)(\partial x/\partial \delta) = 0$ . One extremum is  $\partial x/\partial \delta = 0 = -2\delta/(1+\delta^2)^2$ . Therefore  $\delta = (\omega - \omega_0)/\gamma H_1 = 0$  or  $\omega = \omega_0$ . Another is at  $\partial P_0/\partial x = 1 - 3x/2 = 0$  or  $\omega = \omega_0 \pm \gamma H_1/\sqrt{2}$ .  $\partial S/\partial P_0 = 0$  can be shown to provide no new real roots.

It can be seen from this analysis that for strictly unpolarized light the separation between the two maxima will be  $\sqrt{2}\gamma H_1$ . This separation is independent of  $H_0$  provided only that  $H_0 \gg H_1$ . The degree of mixing in the  $^3P$  states, the imprisonment of radiation, and the intensity ratio  $D_2:D_1:D_0$  affect only the effective value of  $\epsilon$  and therefore will not influence this separation.

## V. INVERSION OF THE RESONANCE LINE

It is often possible to find values for the exciting voltage and pressure of the discharge tube such that the resonance signal becomes inverted. Specifically, for certain densities of metastables the absorption of light in the discharge tube *decreases* at resonance. This inversion can be explained in terms of the changing ratio  $D_3/D_0$  of the component light intensities. This ratio is usually greater than unity for the resonance radiation incident upon the metastables in the front of the tube; but since  $D_3$  light is more strongly absorbed than  $D_0$  light, it is possible for the  $D_3/D_0$  ratio to become less than unity at some point within the discharge tube. Under certain circumstances this can produce an inverted resonance signal due to the sensitivity of the alignment process to the  $D_3/D_0$  ratio.

In this discussion the absorption of radiation which has been scattered once or more will be neglected, as its primary effect is that of a relaxation process. The diffusion of the atoms will also be neglected. This is equivalent to considering the atoms to be contained in

a rigid lattice but is justified by the small distances traveled by an atom between absorption of photons. These two effects and general relaxation phenomena will tend to reduce any signal obtained but will not eliminate the phenomenon of resonance line inversion. Complete mixing in the  $^3P$  states will be assumed.<sup>12</sup>

Consider resonance radiation passing through an ensemble of helium atoms in the  $^3S_1$  state (see Fig. 9). Let the distance of penetration into the ensemble of atoms be  $x$ . The two components of radiation will be denoted by the subscripts 0 and 3 so that the initial intensity ( $x=0$ ) will be  $D=D_0+D_3$  and the intensity at any point within the gas will be given by  $I(x)=I_0(x)+I_3(x)$ . If the number of atoms per unit distance in the states  $m=+1, 0$ , and  $-1$  is  $n_+(x)$ ,  $n_0(x)$ , and  $n_-(x)$ , respectively, then the total number of atoms per unit distance encountered by the light beam is

$$N = n_+(x) + n_0(x) + n_-(x), \quad (21a)$$

where  $N$  is independent of  $x$ . When the rf is on, at the resonant frequency, let these same quantities be given by  $\bar{I}(x) = \bar{I}_0(x) + \bar{I}_3(x)$  and

$$N = \bar{n}_+(x) + \bar{n}_0(x) + \bar{n}_-(x). \quad (21b)$$

Let

$$I_3:I_0 = 1:K(x). \quad (22)$$

In terms of  $K(x)$  and the absorption rates for  $D_3$  and  $D_0$  light ( $^3R_{\pm,0}$  and  $^0R_{\pm,0}$ ) the relative optical absorption probabilities for a metastable atom are

$$R_+ = {}^3R_+ + K(x){}^0R_+,$$

$$R_0 = {}^3R_0 + K(x){}^0R_0,$$

$$R_- = {}^3R_- + K(x){}^0R_-,$$

Since  $K(x) = 1$  and  $R_+ = R_0 = R_-$  when  $I_3 = I_0$ , it follows that the relative transition probabilities obey the relation:

$${}^3R_+ + {}^0R_+ = {}^3R_0 + {}^0R_0 = {}^3R_- + {}^0R_- = F, \quad (23)$$

where  $F$  is just a constant.

Referring to Fig. 9, the change in light intensity in an interval  $dx$  is proportional to the light intensity and the number of atoms in each level multiplied by the relative absorption probability for that level. Considering the two components of resonance radiation as separate beams of light the change in light intensity may be written

$$dI_0 = -C({}^0R_+n_+ + {}^0R_0n_0 + {}^0R_-n_-)I_0dx, \quad (24a)$$

$$dI_3 = -C({}^3R_+n_+ + {}^3R_0n_0 + {}^3R_-n_-)I_3dx, \quad (24b)$$

where  $I_0$ ,  $I_3$ ,  $n_+$ ,  $n_0$ , and  $n_-$  are functions of  $x$ . The

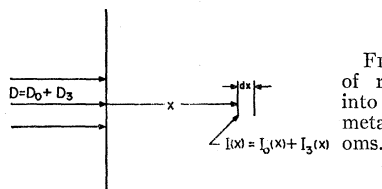


FIG. 9. Penetration of resonance radiation into an ensemble of metastable helium atoms.

<sup>12</sup> Note added in proof.—Observation of the line inversion under conditions of low discharge tube pressure (less than 1 mm Hg) by Laird Scheerer and Joe Rice at Texas Instruments Incorporated has brought our attention to the fact that the inversion effect is much larger for cases of *incomplete* mixing. In addition, they have found that the use of circularly polarized light at low pressure can produce an inversion even in cases where the fraction of resonant light absorbed is very small. (Private communication.)

corresponding equations may be written for the case when the rf is on:

$$d\bar{I}_0 = -C(^0R_+\bar{n}_+ + ^0R_0\bar{n}_0 + ^0R_-\bar{n}_-)\bar{I}_0 dx, \quad (25a)$$

$$d\bar{I}_3 = -C(^3R_+\bar{n}_+ + ^3R_0\bar{n}_0 + ^3R_-\bar{n}_-)\bar{I}_3 dx. \quad (25b)$$

A useful relation may be derived immediately by adding (24a) and (24b) or (25a) and (25b) using (21) and (23):

$$dI_0/I_0 + dI_3/I_3 = -CNF dx = d\bar{I}_0/\bar{I}_0 + d\bar{I}_3/\bar{I}_3.$$

Integration and application of the initial conditions  $I_0 = \bar{I}_0 = D_0$  and  $I_3 = \bar{I}_3 = D_3$  yields

$$I_0 I_3 = \bar{I}_0 \bar{I}_3 = D_0 D_3 e^{-CNF x}.$$

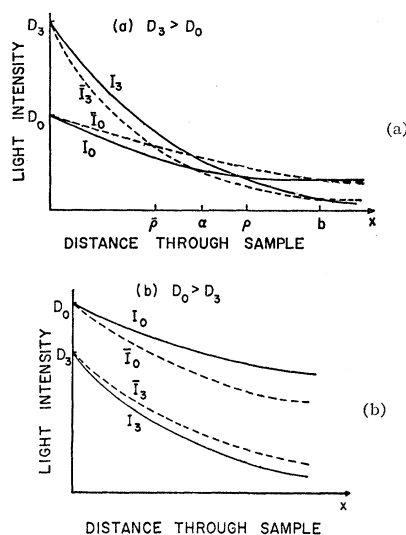


FIG. 10. Qualitative behavior of the intensity of the two components of resonant radiation as they are absorbed by the ensemble of metastable helium atoms for the two initial conditions  $D_3 > D_0$  and  $D_0 > D_3$ . The curves  $I_0$  and  $I_3$  represent the change in intensity when there is no disorienting rf magnetic field, and  $\bar{I}_0$  and  $\bar{I}_3$  represent the intensities with the disorienting rf field on. Since at every point  $I_0 I_3 = \bar{I}_0 \bar{I}_3$ , then the total intensities will be related by the condition  $I_0 + I_3$  is greater than, equal to, or less than  $\bar{I}_0 + \bar{I}_3$  whenever  $|I_0 - I_3|$  is greater than, equal to, or less than  $|\bar{I}_0 - \bar{I}_3|$ .

This general relation holds throughout the ensemble of atoms and is independent of the relative strengths of the light components or the strength of the rf magnetic field. It expresses the fact that the products of the component light intensities are equal and have a simple exponential dependence on  $x$ .

Figure 10(a) shows the change of light intensity for each of the components of resonant light as they penetrate the ensemble of metastable helium atoms both with the rf "on" and "off." The curves represent the solutions of Eqs. (24) and (25). These linear differential equations may be reduced to algebraic equations involving one light intensity ( $I_0$ ,  $I_3$ ,  $\bar{I}_0$ , or  $\bar{I}_3$ ) and  $x$  which can only be solved numerically. However,

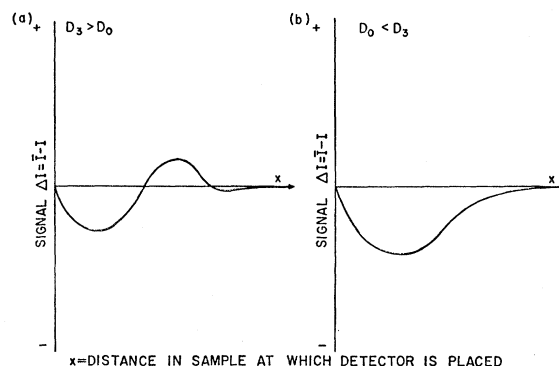


FIG. 11. Dependence of the signal upon the distance through which the resonant light has passed for the two initial conditions. Note. The notation " $D_0 < D_3$ " at the right-hand side uppermost part of the figure should read " $D_0 > D_3$ ."

the essential features of the curves in Fig. 10(a) may be demonstrated without recourse to numerical analysis.

In order to exhibit the resonance signal inversion assume, for the present, that the intensity of each component decreases as shown in Fig. 10(a). The signal is the difference between the total light intensity with the rf on and the intensity with the rf off,  $\bar{I} - I$ . A normal signal is one in which  $I > \bar{I}$  and an inverted signal one in which  $I < \bar{I}$ . In view of the relation  $I_0 I_3 = \bar{I}_0 \bar{I}_3$ , it can be shown that the intensity  $I = I_0 + I_3$  is greater than, equal to, or less than  $\bar{I} = \bar{I}_0 + \bar{I}_3$  depending upon whether the absolute difference  $|I_0 - I_3|$  is greater than, equal to, or less than  $|\bar{I}_0 - \bar{I}_3|$ .

Referring then to Fig. 10(a), it can be seen that for a value of  $x$  between 0 and  $a$ ,  $|I_0 - I_3|$  is greater than  $|\bar{I}_0 - \bar{I}_3|$  and the signal will be normal; between  $a$  and  $b$ ,  $|I_0 - I_3|$  is less than  $|\bar{I}_0 - \bar{I}_3|$  and the signal will be inverted; and greater than  $b$ ,  $|I_0 - I_3|$  is greater than  $|\bar{I}_0 - \bar{I}_3|$  and the signal will be normal. This can be pictured qualitatively as in Fig. 11(a).

It should be noted that if the complexion of the light is such that initially the more strongly absorbed component is weaker, as is usually true in the case of alkali optical pumping, the curves of light intensity with and without rf never cross [Fig. 10(b)]. Therefore the inversion of the resonance line does not occur, as illustrated in Fig. 11(b). In helium this would correspond to  $D_0$  light being initially stronger than  $D_3$  light, while in fact  $D_3$  is generally from two to three times more intense than  $D_0$ .

As mentioned earlier, it is not necessary to find the exact solutions of Eqs. (24) and (25) to demonstrate the relevant features of Fig. 10(a). For helium the  $D_3$  component of light is more strongly absorbed than the  $D_0$  component regardless of the state of polarization or direction of the resonant radiation relative to the magnetic field as can be seen from the table of relative absorption probabilities. In fact

$$^3R_+ > ^0R_+, \quad ^3R_0 > ^0R_0, \quad \text{and} \quad ^3R_- > ^0R_-.$$

From (22) and (24) it is seen that

$$\begin{aligned} dK/K &= dI_0/I_0 - dI_3/I_3 \\ &= -C[n_+(^0R_+ - ^3R_+) \\ &\quad + n_0(^0R_0 - ^3R_0) + n_-(^0R_- - ^3R_-)]dx. \end{aligned}$$

Therefore, since  $K$ ,  $n_+$ ,  $n_0$ , and  $n_-$  are all intrinsically positive,  $dK/dx > 0$ . In particular,  $dK/dx$  has a positive finite value even when  $I_0 = I_3$ , and the ratio  $I_0/I_3$  is therefore a monotonically increasing function of  $x$ . Similarly, it can be shown that a quantity defined by  $\bar{K} = \bar{I}_0/\bar{I}_3$  also obeys the relation  $d\bar{K}/dx > 0$ . It can be seen from these relations that  $I_0$  crosses  $I_3$  at some point  $\rho$ , and  $\bar{I}_0$  crosses  $\bar{I}_3$  at a point  $\bar{\rho}$ . The essential features to be proved are then

1.  $I_0$  is less than  $\bar{I}_0$  to the point  $x = \rho$ .  $I_3$  is greater than  $\bar{I}_3$  to the point  $x = \rho$ .
2.  $\rho > \bar{\rho}$ .
3.  $I_0$  crosses  $\bar{I}_0$  beyond point  $\rho$ .  $I_3$  crosses  $\bar{I}_3$  beyond point  $\rho$ .

Each of these features may be easily proved by examining the differential Eqs. (24) and (25). For example the slopes of the curves  $I_0$  and  $\bar{I}_0$  are given by

$$\begin{aligned} dI_0/dx &= -C(^0R_+n_+ + ^0R_0n_0 + ^0R_-n_-)I_0, \\ d\bar{I}_0/dx &= -C(^0R_+\bar{n}_+ + ^0R_0\bar{n}_0 + ^0R_-\bar{n}_-)\bar{I}_0. \end{aligned}$$

Whenever  $I_3 > I_0$  the level populations are related in the *same* manner as the absorption coefficients for  $I_0$  light (e.g., if  $^0R_+ > ^0R_0 > ^0R_-$ , then  $n_+ > n_0 > n_-$ ). Therefore since the total number of metastables is constant and the difference in level populations is always greater without rf than with, the factor  $(^0R_+n_+ + ^0R_0n_0 + ^0R_-n_-)$  is greater than the factor  $(^0R_+\bar{n}_+ + ^0R_0\bar{n}_0 + ^0R_-\bar{n}_-)$  in the region  $x = 0$  to  $x = \rho$  where  $I_3 > I_0$ . In this region, then,  $I_0$  must remain less than  $\bar{I}_0$  since at any point where  $I_0 = \bar{I}_0$ ,  $I_0$  would have a smaller slope than  $\bar{I}_0$ . Similarly from Eqs. (24b) and (25b) it can be shown that  $I_3 > \bar{I}_3$  to the point  $x = \rho$ .

The other features of the curves in Fig. 10(a) necessary to prove the inversion of the resonance line may be argued in an analogous manner.

#### IV. MAGNETOMETER

The apparatus described in this paper can be adapted for the measurement of small magnetic fields. It should be noted that the alkali optical pumping techniques can also be adapted for these purposes.<sup>13</sup>

The accuracy of a helium magnetometer depends primarily upon the width of the resonance line. The minimum line width attained in the course of this work was about 5 milligauss due to the inhomogeneity of the earth's magnetic field in our laboratory. In a

<sup>13</sup> W. E. Bell and A. L. Bloom, Phys. Rev. **107**, 1559 (1957).

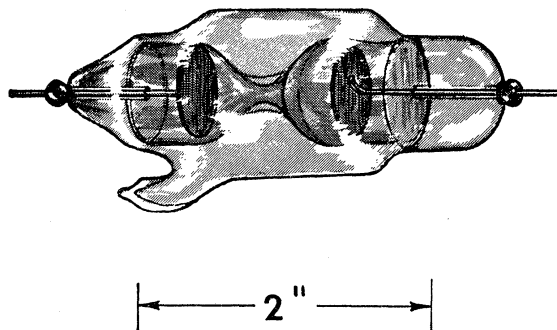


FIG. 12. Concentric discharge tube and lamp.

perfectly homogeneous field the line width would be determined primarily by the lifetime of the metastable atoms. For a typical lifetime of  $2 \times 10^{-4}$  sec, the line width would be about  $\Delta\nu = 1/\pi\tau \cong 1600$  cps  $\cong 5 \times 10^{-4}$  gauss. Standard narrow-band techniques make it possible to find the center of the resonance line to one part in a hundred. Therefore it should be possible to measure magnetic fields with an accuracy of  $5 \times 10^{-6}$  gauss. It may be possible to increase the metastable lifetimes with a corresponding increase in sensitivity.

For most purposes it would be desirable to have a compact "sensing head"—in this case the discharge tube, lamp, detector, and a loop of wire to supply the rf magnetic field. A great reduction in size has been achieved by the use of a cylindrical arrangement of these elements instead of the linear arrangement of Fig. 1. The lamp is surrounded by a concentric discharge tube and two or more detectors placed in a ring around the tube. This can be done in such a way that regardless of the orientation of the tube, not more than one detector is in a position such that a line from the lamp to the detector makes an angle  $\cos^{-1}(1/\sqrt{3})$  with respect to the magnetic field. With this arrangement there will be no orientation in which the signal is zero. The wire providing the rf magnetic field can be so arranged that there is always a component at right angles to the constant field in some part of the discharge tube. A geometry of this type has also been operated successfully with the lamp and discharge tube in the same glass envelope. Several sizes of lamps and discharge tubes have been used with the smallest combination being 3 in. long and  $1\frac{1}{2}$  in. in diameter. One embodiment of this design is illustrated in Fig. 12.

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