

inadequacy of the simple analytical Molière screening factor.<sup>13</sup> The experimental cross sections and particularly the shapes of the experimental cross-section curves in Figs. 1 and 3 show good agreement within the experimental errors (except for copper at 50 keV where the discrepancies are not understood) with the theoretical curves for gold and copper given by the phase-shift calculations of Lin, Sherman, and Percus.<sup>7</sup> The results for tin in Fig. 2 show that the Born approximation calculations with Molière screening break down at 50 keV, but give reasonably accurate results at the higher energies.

## ACKNOWLEDGMENTS

The authors wish to thank Dr. Shin-R Lin for his kind cooperation in evaluating the screened cross section given by the Lin, Sherman, and Percus phase-shift calculations for the parameters used in these measurements; Dr. N. Sherman and Dr. J. Percus for helpful discussions of their results prior to publication; and Dr. J. Coyne for providing the FORTRAN code that was used to evaluate the unscreened Mott exact cross section formula given in Ref. 2. Also, the authors are grateful to Dr. J. Percus for the use of the computer facilities at the Courant Institute of Mathematical Sciences of New York University.

Polarization of He<sup>3</sup> Gas by Optical Pumping

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(Received 5 August 1963)

The process of He<sup>3</sup> nuclear polarization by metastability exchange with optically pumped metastable He<sup>3</sup> atoms is described and experimental details given. Phenomenological theories are presented which explain the optical signals and the time variation of the polarization. The polarization is measured both optically and by nuclear magnetic resonance. Relaxation of the nuclear spins by diffusion through magnetic field gradients is discussed. When gradients are small, nuclear relaxation times as long as 4000 sec have been measured. The maximum polarization achieved was  $40 \pm 5\%$  in He<sup>3</sup> gas at a pressure of one mm Hg.

## I. INTRODUCTION

OPTICAL pumping techniques have been successfully applied to produce orientation of the magnetic moments of a number of atomic species. In particular, the alkali metals, mercury, and the helium isotope of mass four have been investigated in considerable detail by this method.

Colegrove and Franken first reported the alignment by optical pumping of He<sup>4</sup> atoms in the  $2^3S_1$  metastable state.<sup>1</sup> These studies were later extended by Schearer.<sup>2</sup> In their experiments, metastable atoms were produced by an rf discharge in a Pyrex bulb containing He<sup>4</sup> gas at low pressure. In the steady state, approximately one millionth of the He<sup>4</sup> atoms were in the desired  $2^3S_1$  state. Orientation of the magnetic moments of the metastable atoms was accomplished by  $2^3S-2^3P$  pumping light from a helium lamp. Transitions between the three equally spaced magnetic sublevels of the  $^3S_1$  state were induced by an rf magnetic field at the resonant frequency and detected by monitoring the transmitted pumping light.

Consider now the differences one must expect in attempting to polarize He<sup>3</sup> metastables by the same procedure. As in the case of He<sup>4</sup>, the pumping light will tend to produce unequal populations of the  $^3S_1$  magnetic sublevels, and the equilibrium polarization reached will depend on the rate at which these atoms are removed from a particular sublevel by various collision processes. For both He<sup>4</sup> and He<sup>3</sup> the most rapid pertinent collision process is the transfer of metastability from atom to atom, and it is this mechanism that plays a fundamentally different role in the two gases. He<sup>4</sup> has nuclear spin zero and a nondegenerate ground state; thus, metastability exchange collisions cannot alter the magnetic quantum number of the metastable atom, since angular momentum must be conserved. In contrast, He<sup>3</sup> has a nuclear spin  $I = \frac{1}{2}$  and a ground-state Zeeman doublet; metastability exchange can in this case result in a  $\pm 1$  change in the metastable magnetic quantum number, with a corresponding  $\mp 1$  change in the ground-state nuclear magnetic quantum number. Since exchange collisions occur more rapidly than the metastables can absorb photons, the He<sup>3</sup> metastables will be rapidly mixed among their available Zeeman levels, with simultaneous loss of angular momentum to ground-state atoms. It was first thought that this exchange process would

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<sup>1</sup> F. D. Colegrove and P. A. Franken, *Phys. Rev.* **119**, 680 (1960).<sup>2</sup> L. D. Schearer, *Advances in Quantum Electronics* (Columbia University Press, New York, 1961), pp. 239-251.

short circuit the optical pumping and result in an undetectably small metastable polarization. However, it must be remembered that the angular momentum lost by the metastables is transferred to the ground-state atoms. If the ground-state spin-lattice relaxation time is long enough so that no appreciable angular momentum "leak" occurs, the ground-state system will act as a huge "flywheel" attached to the metastable system. The optical pumping process then results in orientation, or polarization, of not only the metastables but also of the entire ground-state system. If the transmitted pumping light is monitored, three resonances will be observed when an rf magnetic field is applied at right angles to the dc field: the two Zeeman resonances of the  $2^3S_1$  metastable state at about 1.9 Mc/sec-G and 3.8 Mc/sec-G, and the ground-state resonance at 3.2 kc/sec-G.

The fact that the ground-state  $He^3$  atoms can be polarized by optical pumping is of considerable importance to nuclear physics. Targets of  $He^3$  gas polarized by the process described here are already being used in nuclear scattering experiments with particle beams from the Rice University tandem Van de Graaf accelerator.<sup>3</sup>

Historically, the development of a method for polarizing  $He^3$  nuclei has long been a major objective of a number of laboratories because of the recognized importance of such a capability to nuclear physics. A number of unsuccessful attempts were made several years ago by one of us (GKW) to polarize liquid  $He^3$  by the Overhauser and "solid-" effect techniques. A series of definitive tests for  $He^3$  polarization were run using charred hydrocarbons, irradiated plastics, mechanically damaged crystals,<sup>4</sup> and metallic sodium as paramagnetic surfaces placed in intimate contact with the liquid  $He^3$ . Though in most cases the foreign surfaces so introduced controlled the  $He^3$  spin-lattice relaxation,<sup>5</sup> and the appropriate microwave transitions were irradiated to satisfy the conditions for the Overhauser and solid effects involving  $He^3$ -wall interactions, the polarization results were always negative for reasons still not completely understood.

By the use of radioactive sources, an unsuccessful attempt was made also to produce ions and/or metastables in liquid  $He^3$  in sufficient quantity to control the  $He^3$  nuclear relaxation<sup>5</sup> and hence allow Overhauser polarization.

Bouchiat, Carver, and Varnum<sup>6</sup> first reported the use of an optical pumping technique to polarize  $He^3$ . They relied upon the weak dipole-dipole interactions between optically pumped rubidium vapor at  $10^{-3}$ -mm

pressure and  $He^3$  buffer gas at 2.8 atm as the polarization mechanism. However, competing  $He^3$  relaxation processes not involving interaction with the rubidium atoms predominated and limited their attainable polarization to 0.01%, too small to be useful in nuclear scattering experiments. It is the much stronger spin coupling between the optically pumped species and the ground state, provided by the metastability exchange process, which allows us to achieve  $He^3$  nuclear polarization as high as 40%. Preliminary reports of this work have been published,<sup>7,8</sup> and a subsequent paper will deal with the temperature dependence of the metastability exchange cross section.

In the next section a simplified theory will be presented which adequately describes most of the observed phenomena. Those portions of a complete phenomenological theory which must be considered will also be discussed. Following this, a description of the experimental apparatus will be given. The remaining sections will be devoted to experimental procedures and results, including a discussion of relaxation effects and transient phenomena, and a brief indication of the uses that may be made of this process.

## II. THEORY

### A. General Considerations and Definitions

The pertinent  $He^3$  energy levels are shown in Fig. 1. If a weak electric discharge is maintained in a low pressure  $He^3$  gas, a small fraction of the atoms will be in excited states or ionized. The excited atoms of immediate interest are those in the long-lived  $2^3S_1$  metastable state. The numbers of metastable and ground-state atoms/cc are denoted by  $n$  and  $N$ , respectively. Right-hand circularly polarized pumping light incident upon the sample along a weak applied magnetic field excites transitions between the  $^3S_1$  and  $^3P_0$  states with the selection rule  $\Delta m = +1$ . (A parallel treatment applies, of course, for light polarized in the opposite sense and  $\Delta m = -1$ .) The pumping light excites atoms from the metastable  $m = -\frac{1}{2}$  and  $m = -\frac{3}{2}$  sublevels to the  $^3P_0$  state, from whence they decay back with equal probabilities to all  $^3S_1$  sublevels. The net result is a transfer of atoms from the sublevels of low magnetic quantum number ( $m = -\frac{1}{2}, -\frac{3}{2}$ ) to those of high magnetic quantum number ( $m = +\frac{1}{2}, +\frac{3}{2}$ ).

The time that would characterize the approach to equilibrium in the optical-pumping process if no other mechanisms acted to mix the metastable atoms among the available magnetic sublevels is called  $\tau_p$ . With reference to Fig. 2, similar definitions can be made for the other interaction times. The metastable spin-lattice relaxation time  $\tau_r$  represents those processes which tend to short circuit the optical pumping by driving

<sup>3</sup> G. C. Phillips, R. R. Perry, P. M. Windham, G. K. Walters, L. D. Schearer, and F. D. Colegrove, Phys. Rev. Letters **9**, 502 (1962).

<sup>4</sup> G. K. Walters and T. L. Estle, J. Appl. Phys. **32**, 1854 (1961).

<sup>5</sup> G. K. Walters, *Helium Three* (Ohio State University Press, Columbus, Ohio, 1960), p. 37.

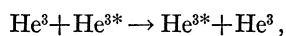
<sup>6</sup> M. A. Bouchiat, T. R. Carver, and C. M. Varnum, Phys. Rev. Letters **5**, 373 (1960).

<sup>7</sup> G. K. Walters, F. D. Colegrove, and L. D. Schearer, Phys. Rev. Letters **8**, 439 (1962).

<sup>8</sup> L. D. Schearer, F. D. Colegrove, and G. K. Walters, Phys. Rev. Letters **10**, 108 (1963).

the metastable sublevel populations toward their thermal-equilibrium distribution, such as collisions with the container walls, impurities, or extraneous discharge products.  $T_r$  will denote the ground-state nuclear magnetic relaxation time which would result from processes not involving interactions with metastables.

Coupling the metastable and ground-state magnetic sublevels are collisions involving metastability exchange,



where \* denotes an atom in the  $^3S_1$  metastable state. Since the incident and emerging ground-state atoms may be in different spin states, this mechanism allows the transfer of angular momentum between metastable and ground-state systems and acts to keep their respective polarizations equal. Equilibrium between the two systems is established in a characteristic time  $\tau_2 \approx 1/\sigma v N$ , where  $\sigma$  is the cross section for metastability exchange and  $v$  is the He<sup>3</sup> thermal velocity. Another constant  $T_2$  characterizes the approach of the ground-state polarization to that of a somehow fixed metastable polarization. These two time constants are related by the density ratio,  $T_2/\tau_2 = N/n$ ; physically,  $\tau_2$  and  $T_2$  are of the order of the times of existence of metastable and ground-state atoms, respectively, before undergoing the exchange process.

The densities of the two magnetic sublevels of the ground state of He<sup>3</sup> are  $N_1$  for  $M = -\frac{1}{2}$  and  $N_2$  for  $M = +\frac{1}{2}$ , such that  $N_1 + N_2 = N$ . The ground-state

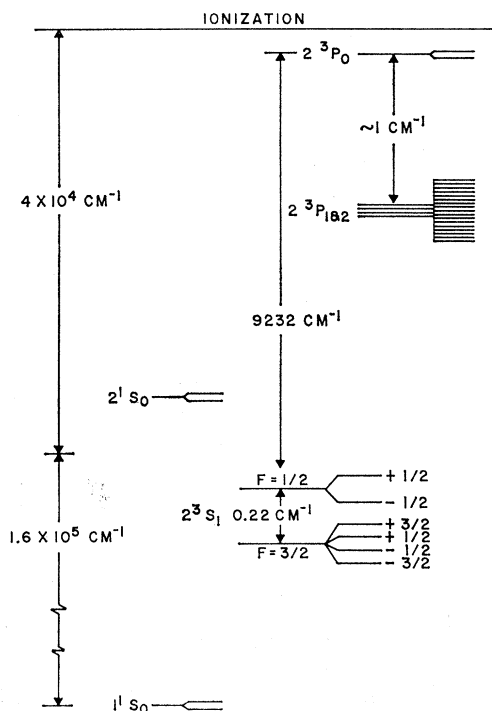


FIG. 1. Energy levels of He<sup>3</sup> atom in external magnetic field (not to scale).

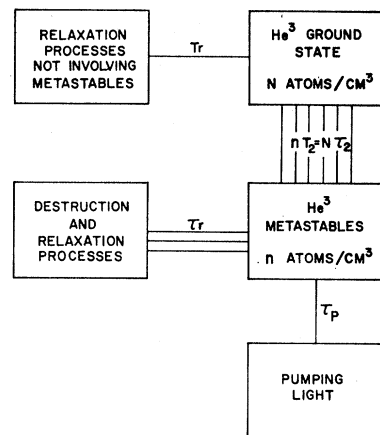


FIG. 2. Schematic diagram indicating the interactions determining the polarizations of metastable and ground state He<sup>3</sup> atoms. Symbols are defined in the text.

polarization is then  $P = (N_2 - N_1)/N$ . The metastable  $2^3S_1$  state is split into  $F = \frac{1}{2}$  and  $F = \frac{3}{2}$  hyperfine levels. In a magnetic field, there are then a total of six Zeeman sublevels. A general treatment of the eight nonlinear rate equations coupling each of these six sublevels with one another and with the ground state becomes extremely complex. However, the phenomenological theory developed in the next section is concerned only with the interrelationships of the various time constants, and no significant loss in generality occurs in treating the metastable state as a two-sublevel system, with densities  $n_1$  and  $n_2$  and polarization  $p = (n_2 - n_1)/n$ .

In Sec. IIC, where the relationship between polarization and optical absorption by the metastable state is derived, it will be necessary to take account of all six metastable sublevels.

## B. Polarization Time Dependence

In terms of the relaxation times defined above, the coupled rate equations governing the metastable and ground-state polarizations are

$$\frac{dp}{dt} = \frac{1-p}{\tau_p} - \frac{p}{\tau_r} + \frac{P-p}{\tau_2}, \quad (1)$$

$$\frac{dP}{dt} = \frac{p-P}{T_2} - \frac{P}{T_r}, \quad (2)$$

where the very small polarizations that result from thermal equilibrium Boltzmann factors are neglected.

The steady state polarizations found by setting the derivatives equal to zero are

$$p_0 = \left[ 1 + \frac{\tau_p}{\tau_r} + \frac{\tau_p}{\tau_2} \left( \frac{T_2}{T_r + T_2} \right) \right]^{-1} \quad (3)$$

and

$$P_0 = p_0 T_r / (T_r + T_2). \quad (4)$$

If there were no coupling between the metastable and ground-state systems,  $\tau_2$  and  $T_2$  would be infinite, and

the polarizations would become  $p_0=1/(1+\tau_p/\tau_r)$  and  $P_0=0$ . Thus, the third term of Eq. (3) represents the degradation in polarization caused by the coupling between the two systems. Comparison of that term with the other terms indicates that the polarization degradation will be negligible if  $N/T_r \ll n(1/\tau_r+1/\tau_p)$ , i.e., if the angular momentum "leak rate" out of the ground state is substantially less than the angular momentum "input rate" from the metastable  $\tau_r$  and  $\tau_p$  processes. Here we have used the approximation  $T_2 \ll T_r$  which is always found to be valid experimentally. In this approximation, Eq. (4) reduces to  $P_0=p_0$  so that the ground-state and metastable polarizations are equal.

Letting  $1/\tau=1/\tau_2+1/\tau_r+1/\tau_p$  and using the approximations  $\tau \ll T_2 \ll T_r$ , the transient solution to the coupled rate equations is

$$p = Ae^{-t/T_p} + Be^{-t/\tau} + p_0, \quad (5)$$

$$P = A \frac{\tau_2}{\tau} e^{-t/T_p} - B \frac{\tau}{T_2} e^{-t/\tau} + P_0. \quad (6)$$

Here the "pumping time" for the coupled systems is defined by

$$\frac{1}{T_p} = \frac{\tau}{T_2} \left( \frac{1}{\tau_p} + \frac{1}{\tau_r} \right) + \frac{1}{T_r}. \quad (7)$$

Since the time constant for the second term in Eqs. (5) and (6) is very short,  $T_p$  characterizes the approach of the coupled systems to equilibrium.  $A$  and  $B$  are constants which depend on the initial conditions.

The results of this phenomenological development will be used in discussing the experimental results in Sec. IV.

### C. Optical Signal

One of the principal advantages of a more general theory than the preceding one would be to relate the magnitude of the optical signal to the polarization of the  $\text{He}^3$ . An adequate theory for all experimental conditions would require knowledge of the relative transition probabilities between the six  $2^3S_1$  sublevels and the eighteen  $2^3P$  sublevels in addition to the detailed results of each of the dozen types of collisions between metastables and ground-state atoms. No attempt will be made to solve the problem with this much generality. Instead, an approximate solution will be found under two restrictions that are met in a large number of experiments.

At ordinary temperatures  $\tau_2$  is very much shorter than  $\tau_p$  or  $\tau_r$ . The metastables are so tightly coupled to the ground-state atoms that the population distribution of atoms in the metastable sublevels can be regarded as being determined almost entirely by collisions with the polarized ground-state atoms.

It is important to note that the metastable atoms are

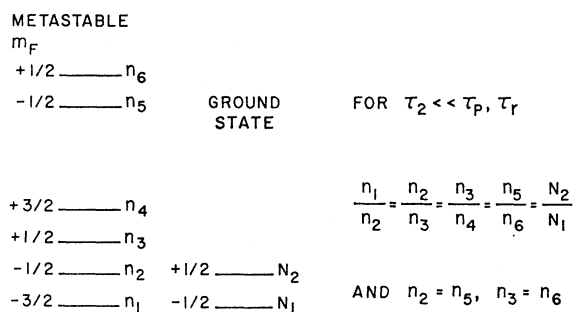


FIG. 3. Relationship of energy level populations between metastable and ground-state atoms at equilibrium under conditions of tight coupling.

generally in well defined spin states, with no memory of past exchange collisions, as they enter into an exchange process. This means that  $\Delta\omega\tau_2 \gg 1$ , where  $\Delta\omega \simeq 4 \times 10^{10}$  sec is the hyperfine splitting of the  $^3S_1$  state. In these experiments,  $\tau_2$  is typically  $10^{-5}$  to  $10^{-7}$  sec so that the condition is easily satisfied. Therefore, the sole factor determining the allowed spin states of the atoms emerging from an exchange collision is that the total angular momentum of the colliding particles, and its projection along the applied magnetic field, be conserved during the collision.

Anderson *et al.*<sup>9</sup> have given the general form of the steady-state solution for spin-exchange problems in which the relaxation times of the spin systems are much longer than the interaction time. In our case if the densities of atoms in the various magnetic sublevels are designated as shown in Fig. 3, the level populations will be given in terms of the ground-state polarization  $P$  by the ratios

$$n_1/n_2 = n_2/n_3 = n_3/n_4 = n_5/n_6 = N_1/N_2 = (1-P)/(1+P).$$

These relations yield, subject to the condition that  $n_1+n_2+n_3+n_4+n_5+n_6=n$ ,

$$\begin{aligned} n_1 &= [(1-P)^3/(6+2P^2)]n, \\ n_2 = n_5 &= [(1+P)(1-P)^2/(6+2P^2)]n, \\ n_3 = n_6 &= [(1+P)^2(1-P)/(6+2P^2)]n, \\ n_4 &= [(1+P)^3/(6+2P^2)]n. \end{aligned} \quad (8)$$

We now make a second simplifying assumption, that the resonant radiation from the helium lamp induces transitions between the  $2^3S_1$  sublevels and the  $2^3P_0$  sublevels only and does *not* cause transitions to the  $2^3P_1$  and  $2^3P_2$  levels. This condition is realized experimentally by deriving the pumping radiation from a  $\text{He}^4$  lamp instead of a  $\text{He}^3$  lamp. The reason for this can be seen by considering the spectra of Fig. 4 taken from the very thorough work of Fred, Tomkins, Brody, and Hamermesh concerning the optical spectrum of  $\text{He}^3$ .<sup>10</sup>

<sup>9</sup> L. W. Anderson, F. M. Pipkin, and J. C. Baird, Jr., Phys. Rev. **120**, 1279 (1960).

<sup>10</sup> M. Fred, F. S. Tomkins, J. K. Brody, and M. Hamermesh, Phys. Rev. **82**, 406 (1951).

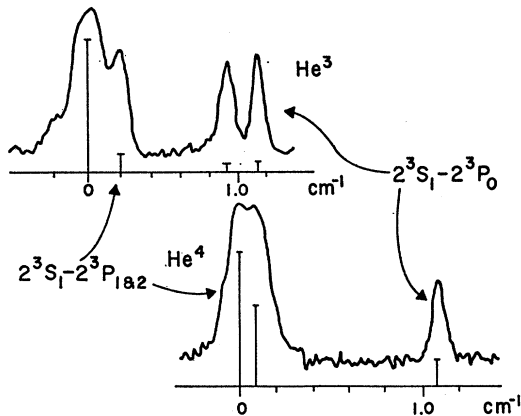


FIG. 4. Structure and isotope shift for the  $2^3S_1-2^3P$  spectral line of helium ( $10\,830\text{ \AA}$ ) from Fred *et al.* (See Ref. 10.)

The relative shifts of the emission lines of He<sup>3</sup> and He<sup>4</sup> are such that the strong component of radiation from He<sup>4</sup> (due to transitions from the  $2^3P_1$  and  $2^3P_2$  levels) induces only  $2^3S_1-2^3P_0$  transitions in He<sup>3</sup>. Since the lamps used in our experiments are more intense than the discharges used by Fred *et al.* the spectral lines from our lamps are somewhat broader than those of Fig. 4. However, spectrographs of our brightest lamps show that the broadening is not sufficient to cause the He<sup>4</sup> lamp to excite a significant number of the lower energy transitions. In practice He<sup>4</sup> lamps are not always used and the differences observed when the lamp contains the mass three isotope are discussed in a later section.

Under these assumptions, then, a solution for the optical signal as a function of polarization can be obtained for the case of small total absorption of light, i.e., when there is a linear relationship between the light absorbed and the number of absorbing atoms in the light path. Typically about 2% of the  $1\text{-}\mu$  light from a He<sup>4</sup> lamp is absorbed by the He<sup>3</sup> metastables so that this condition is satisfied in our experiments. If right-hand circularly polarized radiation from the He<sup>4</sup> lamp is directed along the magnetic field, only transitions in which  $\Delta m$  changes by  $+1$  can occur and thus only levels 1, 2, and 5 can absorb radiation. With the absorption probabilities designated  $a$ ,  $b$ , and  $c$ , respectively, the resonance radiation absorbed by the metastables is

$$I = an_1 + bn_2 + cn_5 \\ = n[(1-P)^2/(6+2P^2)][a(1-P) + (b+c)(1+P)].$$

Since, as discussed above,  $T_r \gg T_2$  and  $\tau_2$  is much shorter than any other lifetime limiting process for a metastable spin state, the metastable and ground state polarizations are tightly "locked" together. Depolarization of either spin system results in depolarization of the other. Therefore if the polarization of the sample is destroyed by some means such as a strong rf magnetic

field applied at the ground-state resonant frequency or at either of the metastable resonant frequencies, then the metastable sublevels are equally populated and the resonance radiation absorbed is  $I_0 = \frac{1}{6}n(a+b+c)$ . A normal resonance signal with saturating rf would be  $\Delta I = I_0 - I$ . A measure of the polarization of the He<sup>3</sup> gas can then be obtained by comparing the resonance signal with the total radiation absorbed.

$$\frac{\Delta I}{I_0} = \frac{P}{3+P^2} \left[ 6 - 2P - 3(1-P)^2 \left( \frac{b+c-a}{a+b+c} \right) \right]. \quad (9)$$

This relationship between the optical signal and polarization is accurate under the conditions specified in the derivation.

The electric dipole transition probabilities ( $2^3S_1-2^3P_0$ ) given in Table I were computed from infor-

TABLE I. Relative electric dipole transition probabilities between magnetic substates of the  $2^3S_1-2^3P_0$  transitions in He<sup>3</sup>. The values include the approximately 10% mixture of  $2^3P_1$  wave functions in the  $2^3P_0$  wave functions.

	$2^3S_1$	$2^3P_0$	
		$+\frac{1}{2}$	$-\frac{1}{2}$
$F = \frac{3}{2}$	$+\frac{3}{2}$	0.28	0.0
	$+\frac{1}{2}$	0.19	0.10
	$-\frac{1}{2}$	0.10	0.19
	$-\frac{3}{2}$	0.0	0.28
$F = \frac{1}{2}$	$+\frac{1}{2}$	0.15	0.30
	$-\frac{1}{2}$	0.30	0.15

mation contained in the work of Fred *et al.*<sup>10</sup> The accuracy of the relative absorption probabilities  $a$ ,  $b$ , and  $c$  obtained from this table is limited by the uncertainty of the relative illumination of  $F = \frac{1}{2}$  and  $F = \frac{3}{2}$  levels of the metastables. Since the  $2^3S_1-2^3P_{1,2}$  emission line of the He<sup>4</sup> lamp is quite broad relative to the  $2^3S_1-2^3P_0$  absorption lines of He<sup>3</sup>, it will be assumed that  $F = \frac{1}{2}$  and  $F = \frac{3}{2}$  levels are equally illuminated.

For equal illumination the absorption probabilities become  $a = 0.28$ ,  $b = 0.10$ , and  $c = 0.30$  and  $(b+c-a)/(a+b+c)$  is approximately  $\frac{1}{6}$ . Therefore Eq. (9) becomes

$$\Delta I/I_0 \cong P(11-2P-P^2)/(6+2P^2). \quad (10)$$

This curve is plotted in Fig. 5 and has been verified by nuclear magnetic resonance for polarizations up to 25% at room temperature and over the pressure range studied (see Sec. IVB).

### III. EXPERIMENTAL APPARATUS

The components necessary for optically polarizing the He<sup>3</sup> gas and for monitoring this polarization either optically or by nuclear magnetic resonance techniques are illustrated schematically in Fig. 6.

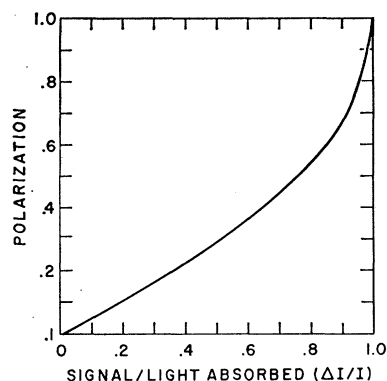


FIG. 5. Polarization as a function of the fractional change in light absorbed when the polarization is destroyed.

The lamps used in most experiments are simply pyrex tubes (about 4-mm i.d.) filled with pure  $\text{He}^4$  gas to a pressure of about 4 mm Hg. Under operating conditions, helium diffuses into the walls and the pressure will drop significantly in small lamps after a few hours operation. For this reason all lamps are built with a chamber at one end of the tube to act as a reservoir. These lamps are excited by capacitively coupling about 10 W of 50-Mc/sec rf energy into the tubes.

For experiments in which very high light intensity was desired, a quartz disk-shaped lamp was used. The disk was 3 cm in diameter with inside separation of 1 mm and was excited by 100 W of radiant power at 2450 Mc/sec provided by a Raytheon diathermy generator Model CMD5.<sup>11</sup>

Light from the lamps is circularly polarized by passing it first through Polaroid type HR linear polarizer for the infrared and then through a  $\frac{1}{4}$ -wave material with axes oriented at  $45^\circ$  to the axis of the linear polarizer. The  $\frac{1}{4}$ -wave material used was cellophane 0.001 in. in thickness.

The discharge cells (generally Pyrex spheres 5 cm in diam) were filled with  $\text{He}^3$  over a range of pressures from 0.1 to 10 mm Hg. Purity of the helium gas is important. The criteria for purity is that the spin-lattice relaxation time of the metastables ( $\sim 10^{-4}$  sec)<sup>1</sup> must not be appreciably shortened by collisions with impurities. A reasonable cross section for relaxation of a metastable by an impurity is  $\sigma \approx 10^{-14}$  cm<sup>2</sup> and the density of impurity atoms allowable can be determined from  $1/\tau_r \gg \sigma n$ . At room temperature then,  $n$  should be much less than  $10^{-13}$  atom/cm<sup>3</sup> and the partial pressure of impurities should be much less than  $10^{-4}$  mm Hg.

We used commercially available, spectroscopically pure  $\text{He}^4$  gas. The  $\text{He}^3$  gas was obtained from the Monsanto Mound Laboratory and was purified by flowing it through a liquid helium trap. Prior to filling an absorption cell, the walls must be thoroughly cleaned since ion bombardment of dirty walls in the weak discharge will drive out impurities. Therefore, in

<sup>11</sup> The lamps described are similar in design to those used by C. Cohen-Tannoudji (private communication).

addition to heating the walls under vacuum ( $\sim 10^{-6}$  mm Hg) it was also found necessary to admit helium, excite a bright discharge and pump out the resulting impure gas before finally refilling the cell with clean helium. When the walls of the container are well cleaned, the weak discharge has the opposite effect and tends to further improve the purity of the gas by driving impurities into the walls.

The electronic equipment is relatively simple but will be mentioned for completeness. A 50-Mc/sec oscillator is used to excite the electrodeless discharge in the cell. The frequency was chosen for convenience and has no significance other than that it is not near a  $\text{He}^3$  or  $\text{He}^4$  resonant frequency for the magnetic fields used.

The pumping light is monitored by a lead sulfide detector and the signal can be displayed in any of the usual ways. When the optical signal is used, the rf-magnetic field may be provided by conventional oscillators and the coil configuration is not important.

Nuclear magnetic resonance experiments were done using an Anderson bridge<sup>12</sup> tuned to 800 kc/sec. The rf coil consisted of two 21-turn windings on a 5.3-cm-diam cylinder. Each winding was 0.6 cm long and they were spaced 4.7 cm apart in order to pass the beam of pumping light. This coil had an inductance of 100  $\mu\text{h}$ . Signals were detected and amplified by a National HRO-60 radio receiver and recorded on an X-Y plotter. The 800-kc/sec input to the bridge was provided by a well shielded, crystal-controlled oscillator.

For optical detection of polarization or resonance signals, the dc magnetic field was provided by the earth's field or Helmholtz coils. The nuclear magnetic resonance experiments were performed in fields of 250 G inside a solenoid wound for this purpose. The solenoid was 10 in. in diam and was end-corrected to sixth order. It was patterned after coils studied by

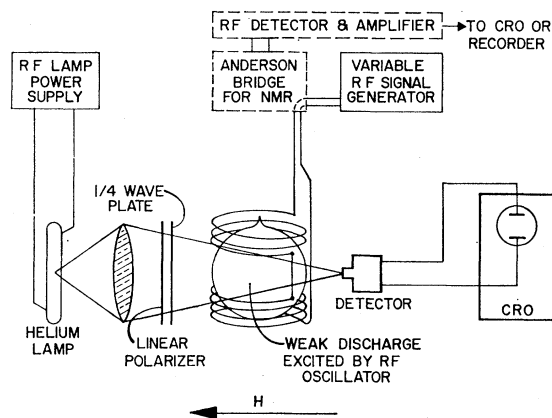


FIG. 6. Schematic of equipment used in performing resonance experiments on optically pumped  $\text{He}^3$ .

<sup>12</sup> H. L. Anderson, Phys. Rev. **76**, 1460 (1949).

Garrett.<sup>13</sup> The solenoid was wound with two layers of No. 14 wire. The field variation over the 5-cm discharge-cell was about 0.015 G.

#### IV. EXPERIMENTAL RESULTS

##### A. Time Constants and Transient Phenomena

Many of the time constants described in Sec. II can be measured by observing transient changes in the transmitted light when changes are made in the fields applied to the discharge cell. The rate of change of the polarization is accurately reflected in the rate of change of the transmitted light for relatively small polarization ( $\sim 10\%$ ) where the optical signal is approximately linear in polarization. All the measurements described here were done at room temperature. The metastable ground-state interaction times,  $\tau_2$  and  $T_2$ , are temperature-dependent and become quite long at low temperatures. This interaction will be the subject of a later paper.

The metastable relaxation time  $\tau_r$  has been measured in the earlier optical pumping experiments with He<sup>4</sup> and was found to be about  $2 \times 10^{-4}$  sec.<sup>1</sup> The technique used was simply that of observing the change in transmitted light when a saturating rf field at the Larmor frequency was suddenly removed and the metastables allowed to approach their equilibrium alignment. The metastable pumping time  $\tau_p$  can be estimated from measured light intensity and the resonant photon-absorption cross section  $\lambda^2/2\pi$ .<sup>1</sup> The pumping time is observed to become nearly equal to  $\tau_r$  for the brightest lamps and is larger for lamps of lower intensity. Therefore,  $\tau_p$  is estimated to be between  $10^{-4}$  sec and several msec.

The time constant  $\tau_2$ , which is roughly the time between exchange collisions of a metastable atom with ground-state atoms, can be obtained by measuring the metastable resonance linewidth. These measurements were made by chopping rf ( $\sim$  few Mc/sec) at a low audiofrequency and sweeping the magnetic field slowly through the resonance condition. The signal from the detector was amplified, passed through a narrow band phase detector, and recorded. Both the  $F = \frac{3}{2}$  and  $F = \frac{1}{2}$  metastable resonances at 1.9 and 3.8 Mc/sec, respectively, have been observed in this way. The measured value of  $\tau$  at room temperature was  $3 \times 10^{-6}$  sec at a pressure of 0.15 mm Hg. From the definition of  $\tau$  and the above values of  $\tau_r$  and  $\tau_p$  it can be seen that  $\tau \approx \tau_2$  for pressures above 0.1 mm Hg. Since  $1/\tau_2 = \sigma v N$ , where  $\sigma$  is the cross section for metastability exchange and  $v$  is the thermal velocity of the atoms,  $\tau_2$  will be inversely proportional to the pressure. At room temperature, we find  $\sigma \approx (4.2 \pm 0.5) \times 10^{-16}$  cm<sup>2</sup>.

The time constant  $T_p$  which describes the approach of the system to equilibrium can be most easily measured by destroying the polarization with a saturating

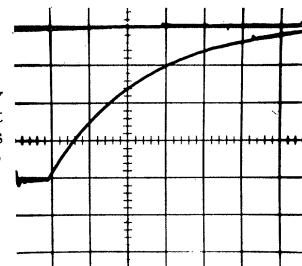


FIG. 7. Oscilloscope display of change in transmitted light intensity as polarization builds up in time  $T_p$ . Time base is 5 sec/division.

rf magnetic field at the Larmor frequency of the He<sup>3</sup> nuclei ( $\sim 3.2$  kc/sec) and then observing the exponential change in light intensity when the rf is suddenly turned off. The signal to noise obtained in such experiments can be seen from Fig. 7. The quantity  $T_p$  was found to depend upon pressure and discharge intensity as expected. Measured values varied from a few seconds in bright discharges to almost 100 sec for the weakest discharges. At a pressure of 1 mm Hg. and under usual operating conditions,  $T_p$  is about 30 sec. For intense pumping light, where  $\tau_p$  is as short as  $\tau_r$ ,  $T_p$  will also depend upon lamp brightness.

Figure 8 shows the change in transmitted light that occurs when a strong oscillating magnetic field is suddenly applied at the Larmor frequency of the metastables. The sharp drop when the oscillating field is applied is due to the term in Eq. (5) with the short time constant and the slow decay is a measure of  $T_p$ . These equations must be modified in this case by introducing the effect of the oscillating magnetic field,  $H_1$ , in inducing transitions among the  $^3S_1$  sublevels. This is done by replacing  $\tau$  by  $\tau^*$ , where  $1/\tau^* = 1/\tau + 1/\tau_{rf}$ . The average time between "transitions" due to the oscillating magnetic field is related to  $H_1$  by the equation

$$1/\tau_{rf} = (1/\tau^2 + \gamma^2 H_1^2)^{1/2} - 1/\tau.$$

The definition of  $T_p$  is then

$$\frac{1}{T_p} = \frac{1}{T_r} + \frac{\tau^*}{T_2} \left[ \frac{1}{\tau_p} + \frac{1}{\tau_r} + \frac{1}{\tau_{rf}} \right].$$

In the limit  $H_1 \rightarrow \infty$ ,  $\tau^*$  approaches  $\tau_{rf}$  which goes to zero; hence  $T_p$  approaches the value  $T_2$  (since  $T_2 \ll T_r$ ).  $T_2$  can be determined by plotting  $T_p$  as a function of  $1/H_1$  and extrapolating to  $1/H_1 = 0$ . Since  $1/T_2 = \sigma v n$ ,  $T_2$  will depend upon the number of metastables and therefore on the character of the discharge. Values of  $T_2$  ranged from 30 msec in bright discharges to 1 sec in the weakest discharge. With  $v = 1.6 \times 10^5$  cm/sec and  $\sigma = 4.2 \times 10^{-16}$  cm<sup>2</sup> at 300°K, the density of metastables is between  $n = 5.0 \times 10^{11}$  and  $1.5 \times 10^{10}$  atoms/cm<sup>3</sup>.

The measurement of  $T_r$  in the absence of a discharge is of particular interest and will be discussed in a separate section.

It is interesting to note that even though the light beam does not interact directly with the ground-state

<sup>13</sup> M. W. Garrett, J. Appl. Phys. 22, 1091 (1951).

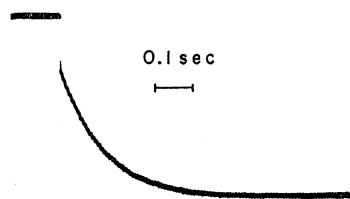


FIG. 8. Change in transmitted light when an oscillating magnetic field partially destroys the metastable polarization.

atoms, it can in most instances be used to monitor the magnitude and direction of the net magnetic moment of the  $\text{He}^3$  nuclei. The reason for this is the very close coupling between the metastables and the ground state atoms. In one Larmor period of the nuclei, the metastable state is exchanged many times. For instance Fig. 9 shows the optical signal observed when a strong oscillating magnetic field at the nuclear Larmor frequency is suddenly applied at  $90^\circ$  to the steady field. Considered in the reference frame rotating at the Larmor frequency, the macroscopic nuclear magnetic moment precesses about  $H_1$  becoming alternately parallel and antiparallel to the light beam, thereby modulating the relative metastable level populations, and therefore the light intensity, at the frequency  $\gamma H_1$ . At the same time the net magnetic moment decreases exponentially as the individual moments become dephased in the time  $T_2^*$  where  $1/T_2^* = 1/T_r + 1/T_2 + \gamma \Delta H_1$ , where  $\Delta H_1$  is the inhomogeneity of the rf field. The beat signal therefore has the form

$$\exp(-t/T_2^*) \cos \gamma H_1 t.$$

The tight coupling of the metastables to the ground state also makes possible "crossed-beam" experiments where the nuclear magnetic moment precessing about the  $z$  direction is observed by a light beam in the  $x-y$  plane. This includes free precession experiments in which the direction of the field is suddenly changed and experiments in which the precessing moment is driven in phase with an rf magnetic field.

A crossed-beam system patterned after Bell and Bloom<sup>14</sup> gave accurate measurements of  $T_p$  and  $T_2$ . In this configuration the optical axis is  $45^\circ$  to the magnetic field ( $z$ ) axis. An rf magnetic field is applied with its axis in the  $x-y$  plane and the external field swept through resonance. For very weak discharges resonance linewidths of 0.3 cps were observed corresponding to  $T_2 \approx 1$  sec. If the polarization is initially zero and periodic fast sweeps are made through the resonance

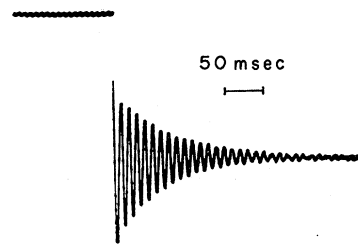


FIG. 9. Change in transmitted light upon sudden application of oscillating magnetic field at nuclear Larmor frequency.

<sup>14</sup> W. E. Bell and A. L. Bloom, *Phys. Rev.* **107**, 1559 (1957).

condition, the gradual buildup of polarization can be observed as a function of time as shown in Fig. 10. This method provided the most convenient means for measuring  $T_p$  under varying conditions of pressure and discharge intensity.

### B. Nuclear Spin Relaxation in $\text{He}^3$ Gas

Nuclear spin-lattice relaxation times for the  $\text{He}^3$  ground state were measured both in the presence and in the absence of the electric discharge. With the electric discharge on, the dominant relaxation process is via exchange collisions with  $2^3\text{S}_1$  metastables. The ground-state relaxation time due to metastability exchange is just the pumping time [Eq. (7)] one observes in the optical pumping experiment.

The relaxation time  $T_r$  in the absence of the discharge is measured by polarizing the sample, turning off the discharge, and monitoring the decay of the  $\text{He}^3$  nuclear magnetic resonance signal with time. For magnetic fields too low for nuclear magnetic resonance measurements, optical detection can be used to measure

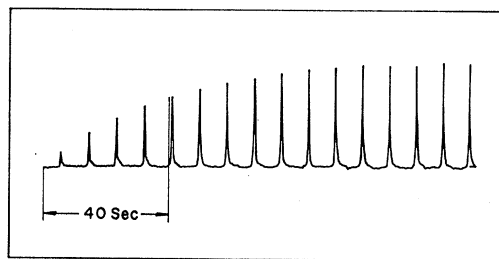


FIG. 10. Growth of precessing macroscopic moment in "crossed-beam" experiment shown by successive passes through resonance.

$T_r$  by a sampling technique. After the  $\text{He}^3$  has been optically pumped, the discharge is turned off for a period of time  $t$ . Upon reigniting the discharge, the strength of the residual optical signal is quickly measured before appreciable repumping takes place. Under these conditions,  $T_r \approx t(\ln S_0/S)^{-1}$ , where  $S$  and  $S_0$  are the magnitudes of the optical pumping signals after time  $t$  and at zero time, respectively.

The relaxation time  $T_r$  due to dipole-dipole interactions for  $\text{He}^3$  at 1 mm pressure is about  $10^9$  sec, using the BPP theory of nuclear relaxation.<sup>15</sup> The experimentally observed values of  $T_r$  were approximately 600 sec when measured by nuclear magnetic resonance in a solenoid field of 240 G, and about 4000 sec when measured by the optical-pumping method in a Helmholtz-coil field of about 4 G. The 600-sec relaxation time is probably determined by diffusion of the  $\text{He}^3$  atoms through the solenoid magnetic-field gradients,

<sup>15</sup> N. Bloembergen, E. M. Purcell, and R. V. Pound, *Phys. Rev.* **73**, 679 (1948). See also J. A. Pople, W. G. Schneider, and H. J. Bernstein, *High-Resolution Nuclear Magnetic Resonance* (McGraw-Hill Book Company, Inc., New York, 1959), Chap. 9.



a mechanism which will be discussed in greater detail later. On the other hand, the 4000-sec relaxation time occurs at low magnetic fields where gradients were not a problem. The relaxation mechanism in this case is due to interactions of the He<sup>3</sup> with the Pyrex surface of the container during collisions. This interpretation is further substantiated by the fact that the relaxation time is shorter for smaller sample containers.

It is interesting, and somewhat puzzling, that our relaxation times at 1 mm pressure are of the same order as those obtained by Bouchiat, Carver, and Varnum for He<sup>3</sup> at 2.8 atm.<sup>6</sup> They also attribute the relaxation to wall collisions, but for our results to be compatible with theirs one must require a wall relaxation mechanism that is highly pressure-dependent, since, at 2.8 atm, the wall collision rate for He<sup>3</sup> atoms is much smaller than at 1 mm pressure.

An interesting relaxation effect is observed whenever the He<sup>3</sup> samples are studied in the presence of appreciable magnetic field gradients. As a He<sup>3</sup> dipole undergoes Brownian motion throughout the container volume, it sees a randomly fluctuating magnetic field due to the gradient. Representing the mean square transverse field, averaged over either space or time, as  $H_t^2$  and applying the BPP theory<sup>15</sup> with an autocorrelation function  $g(\tau) = H_t^2 \exp(-t/\tau_c)$  to obtain an expression for the relaxation time  $T_\theta$  due to field gradients, we find (in the limit that  $\tau_c$  is much less than the Larmor period)

$$T_\theta = (2\gamma_{\text{He}^3} \langle H_t^2 \rangle_{\text{av}} \tau_c)^{-1}. \quad (11)$$

It seems natural to use the average time between collisions as the correlation time  $\tau_c$ , since the motion of an atom is appreciably randomized on each collision. In our experiments of 1 mm Hg pressure,  $\tau_c \simeq 3 \times 10^{-7}$  sec. Using Eq. (11), it can be seen that  $H_t \sim 0.03$  G is sufficient to reduce  $T_\theta$  to about 1 sec.

The functional form of the expression for  $T_\theta$  was verified by using a small current-carrying coil as a variable magnetic dipole to establish a magnetic field gradient over the He<sup>3</sup> sample volume. It was found, as expected, that  $T_\theta$  varies inversely as the square of the dipole moment and directly as the pressure. Also, an estimate of the absolute magnitude of  $H_t^2$ , together with  $\tau_c \simeq 3 \times 10^{-7}$  sec at 1 mm pressure, yields calculated values for  $T_\theta$  which agree approximately with experiment.

### C. Polarization Measurements

The He<sup>3</sup> polarization may be determined optically by comparing the total light absorbed by the unpolarized discharge  $I_0$  with the difference  $\Delta I$  in light absorbed by the discharge tube when it is unpolarized and when it has reached its equilibrium polarization. The approximate relationship between this ratio and the polarization is given by Eq. (10). The signal  $\Delta I$  may be obtained by suddenly destroying the polarization with an rf magnetic field at the nuclear resonant

frequency and recording the voltage change at the light detector. In practice it was found more convenient to measure  $2\Delta I$  by rotating the external field 180°. This was done rapidly (with respect to  $T_p$ ), but adiabatically (slow with respect to a Larmor period). The total light absorbed  $I_0$  is found simply by measuring the detector signal when the discharge is turned off and subtracting from this the small signal due to light from the discharge when the lamp is off.

While such measurements of polarization can be made quickly and with a minimum of additional equipment, the analysis of the optical signal is subject to some uncertainty. The derivation of Eq. (10) is correct only when the total absorption is small so that the absorption can be assumed directly proportional to the number of atoms in the light beam. Nonlinearities in the detector could also introduce error. While neither of these sources of error are serious for measurements made to an accuracy of  $\pm 20\%$ , it was considered necessary to make accurate measurements of polarization using nuclear magnetic resonance techniques and to compare these with the optical measurements.

Performing nuclear magnetic resonance experiments on He<sup>3</sup> gas at pressures near 1 mm Hg presents interesting problems. Since the polarization is high, the net number of excess spins aligned in the field is comparable to ordinary liquid samples at room temperature and several hundred G. It would therefore seem appropriate to compare the adiabatic fast passage signal of the He<sup>3</sup> with that of a known liquid sample. However, it was found very difficult in practice to obtain fast passage conditions in the He<sup>3</sup>, due to field inhomogeneities and rapid diffusion of the atoms throughout the sample bulb. The adiabatic condition is satisfied if  $(1/H_1)(dH_0/dt) \ll \gamma H_1$ , where  $H_1$  is the rf field strength and  $dH_0/dt$  is the sweep rate of the steady  $H_0$  field.<sup>15</sup> Consider now the effect of a field gradient  $\Delta H_0$  over the sample volume. If the time taken to traverse the resonance is longer than the time  $t_0$  that a typical He<sup>3</sup> atom takes to diffuse a distance of the order of the sample dimensions, then the resonating He<sup>3</sup> nuclei will experience the full range of  $H_0$  values during each time interval  $t_0$ . The adiabatic condition then becomes  $(1/H_1)(\Delta H_0/t_0) \ll \gamma H_1$ . In these experiments,  $t_0 \simeq 10^{-8}$  sec and  $\Delta H \simeq 15$  mG, thus demanding  $H_1 \sim 0.1$  G or greater. However, because of space limitations inside the solenoid and in order to allow passage of the pumping light through the He<sup>3</sup> cell,  $H_1$  was of necessity very inhomogeneous over the sample volume; hence,  $H_1$  fields of the magnitude required to satisfy the adiabatic condition quickly destroyed the He<sup>3</sup> polarization as a result of the inhomogeneity relaxation mechanism discussed in Sec. IVB. [The inhomogeneity relaxation formula, Eq. (11), should apply directly for oscillating magnetic fields of any frequency less than  $1/\tau_c$ .]

Rather than modify the equipment to satisfy the adiabatic fast passage conditions, we determined the He<sup>3</sup> polarization by comparing slow passage nuclear

magnetic resonance signals<sup>16</sup> with those from protons in a sample of benzene with identical geometry.

The procedure followed is one in which the frequency in the bridge remains constant (800 kc) and the nuclear magnetic resonance absorption curves are obtained at the resonant value of the field for each sample. The benzene linewidth is determined by the inhomogeneity ( $\gamma\Delta H$ ) while the He<sup>3</sup> line is narrowed by diffusion since in this case  $t_0 < 1/(\gamma\Delta H)$ . Therefore, the effective number of absorbing spins in the He<sup>3</sup> sample is determined by a comparison of the total area under the two resonant curves. In order that such a comparison be valid, the saturation of the resonances must be small for He<sup>3</sup> ( $\gamma H_1)^2 \ll 1/(T_1 T_2)$ , and the sweep must be slow;  $\gamma dH_0/dt \ll (\text{linewidth})^2$ .

The strength of the nuclear resonance signal will depend on the coil geometry, filling factor, rf magnetic field strength and frequency, and the imaginary part of the complex nuclear magnetic susceptibility. In these measurements, samples of identical size and shape are used in the same bridge circuit which is operated at a constant frequency and power level ( $H_1$  level). Therefore, all but the susceptibilities are identical for He<sup>3</sup> and benzene. The ratio of the signal amplitudes  $V_{\text{He}^3}$  and  $V_{\text{H}}$ , from He<sup>3</sup> and benzene protons, respectively, will simply be the ratio of the imaginary susceptibilities for He<sup>3</sup> and benzene.

$$V_{\text{He}^3}/V_{\text{H}} = \chi_{\text{He}^3}''/\chi_{\text{H}}'' = \gamma_{\text{He}^3} M_{\text{He}^3} g_{\text{He}^3}(\omega) / \gamma_{\text{H}} M_{\text{H}} g_{\text{H}}(\omega).$$

Here,  $\gamma$  denotes the appropriate nuclear gyromagnetic ratio,  $M$  the macroscopic magnetic moment of the sample, and  $g$  the shape function normalized in each case so that  $\int_0^\infty g(\omega) d\omega = 1$ . By measuring the areas,  $A_{\text{He}^3}$  and  $A_{\text{H}}$ , under the resonance lines, the shape functions integrate out, so that

$$A_{\text{He}^3}/A_{\text{H}} = \gamma_{\text{He}^3} M_{\text{He}^3} / \gamma_{\text{H}} M_{\text{H}}. \quad (12)$$

The macroscopic magnetic moment of the benzene sample is determined by the thermal Boltzmann factor

$$M_{\text{H}} = N_{\text{H}} \mu_{\text{H}}^2 H_{\text{H}} / kT,$$

where  $\mu_{\text{H}} = I \hbar \gamma_{\text{H}}$  is the proton magnetic moment,  $N_{\text{H}}$  is the density of protons in benzene, and  $H_{\text{H}}$  is the dc magnetic field strength. The He<sup>3</sup> macroscopic magnetic moment is given by

$$M_{\text{He}^3} = \mu_{\text{He}^3} (N_2 - N_1) = \mu_{\text{He}^3} NP.$$

Substituting the expressions for  $M_{\text{H}}$  and  $M_{\text{He}^3}$  into Eq. (12) and solving for  $P$ , we obtain

$$P = (A_{\text{He}^3}/A_{\text{H}}) [\gamma_{\text{H}}^2 N_{\text{H}} \mu_{\text{H}}^2 H_{\text{H}} / \gamma_{\text{He}^3}^2 N kT]. \quad (13)$$

Since all the quantities in the brackets are known,  $P$  can be determined from measurements of the areas under the benzene and He<sup>3</sup> resonance signals.

In these experiments, all measurements were made at room temperature (300°K) and at a fixed rf frequency

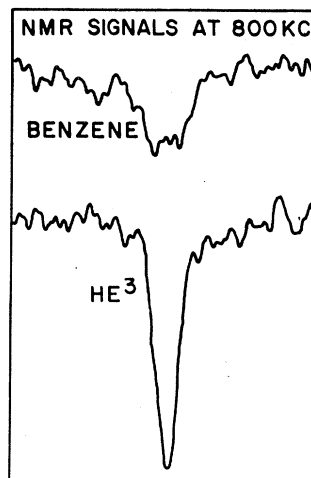


FIG. 11. NMR signals for benzene and He<sup>3</sup>.

of 800 kc. For a He<sup>3</sup> pressure of 1 mm Hg, Eq. (13) reduces to

$$P = (0.137) A_{\text{He}^3} / A_{\text{H}},$$

so that the He<sup>3</sup> and benzene nuclear magnetic resonance signals are of comparable strength for the range of He<sup>3</sup> polarizations (10%–40%) obtained. Figure 11 shows two resonance signals used in determining polarization of a He<sup>3</sup> bulb at a pressure of 1 mm Hg.

The polarization reached at equilibrium depends strongly upon the pressure and discharge intensity. For a given light intensity and pressure, the polarization increases steadily as the 50-Mc/sec electrodeless discharge is made weaker until the level of excitation is reached at which the discharge extinguishes. There is every reason to believe that in the pressure range studied higher polarizations could be obtained with weaker discharges. The reason for this appears to be an increase in the relaxation times of the metastables and/or ground state He<sup>3</sup> for weaker discharges. When  $\tau_2 \ll \tau_r$ ,  $\tau_p$  the equilibrium polarization is seen from Eqs. (3) and (4) to be

$$P \cong 1/\tau_r [N/(nT_r) + 1/\tau_r + 1/\tau_p]^{-1}. \quad (14)$$

For a fixed light intensity and pressure,  $\tau_p$  and  $N$  are constant. The relaxation time  $T_r$  cannot be measured in the presence of the discharge but is surely greater than  $T_p$  ( $\sim 40$  sec in weak discharges). Also  $n$  is known to decrease slightly for small decreases in discharge intensity, and for weak discharges is between  $10^{10}$  and  $10^{11}$  atoms/cm<sup>3</sup>. The achievable polarization depends upon the magnitude of  $1/\tau_p$  relative to the two relaxation terms of Eq. (14).

Using the techniques described above, the polarization has been measured as a function of pressure. The results are shown in Fig. (12) with the vertical scale in arbitrary units. The light intensity was held constant. While no adequate measure of discharge intensity is readily available when the pressure is to be varied, some control was obtained by using the same means of

<sup>16</sup> F. Bloch, Phys. Rev. 70, 460 (1946).

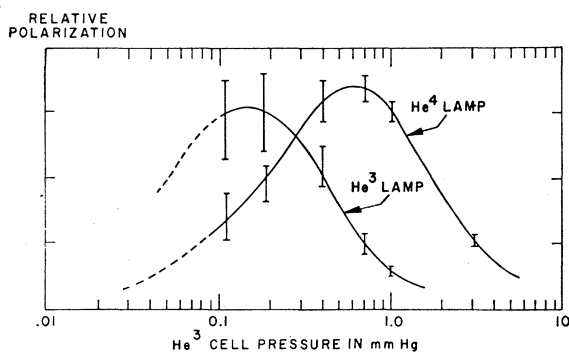


FIG. 12. Relative polarization measured by NMR as a function of the cell pressure for He<sup>3</sup> and He<sup>4</sup> pumping radiation.

excitation and always operating with the weakest self-sustaining discharge.

The falloff in polarization at low pressures is due to a steadily decreasing  $\tau_r$  as a result of the destruction of metastables at the walls. Higher polarizations could be obtained at low pressures by increasing the size of the vessel until the diffusion time to the walls is somewhat less than the metastable relaxation time due to other processes ( $\tau_r \approx 2 \times 10^{-4}$  sec). Diffusion coefficients for metastable helium atoms have been measured by Phelps and Molnar.<sup>17</sup>

It can be seen from Eq. (14) that the polarization will decrease nearly linearly with increasing  $N$  at higher pressures when the term  $N/(nT_r)$  becomes dominant, provided, of course, that  $n$ ,  $T_r$ , and  $\tau_r$  change less rapidly with pressure. Of possibly greater importance is the change in the character of the discharge at pressures above a few mm Hg. At these pressures it becomes increasingly difficult to maintain a uniform discharge throughout the cell. Regions of relatively bright discharge occur near the electrodes and probably cause shortening of  $\tau_r$  and/or  $T_r$ .

The difference between the two curves of Fig. 12 is due to the difference in spectral characteristics of the He<sup>3</sup> and He<sup>4</sup> lamps. It can be seen from Fig. 4 that He<sup>3</sup> metastables will be optically pumped by absorption of both components of resonant radiation when a He<sup>3</sup> lamp is used, but only by  $2^3S_1-2^3P_0$  transitions when a He<sup>4</sup> lamp is used. The effect of pumping with different ratios of these components at various pressures and discharge conditions of the absorption cell has been discussed for optical pumping of He<sup>4</sup> in Refs. 1 and 2. The effects of absorbing only one component of resonant radiation will be qualitatively the same in the case of He<sup>3</sup> metastables.

The references considered two distinct conditions for optical pumping: "mixing," which occurs at pressures of approximately 1 mm Hg and above, and "no mixing" which occurs at pressures below a few tenths of a mm Hg. In the first case a He atom excited from the

$2^3S_1$  state to the  $2^3P$  state undergoes several disorienting collisions before reradiating and thus has equal probability of radiating from each of the  $P$ -state levels independent of the level to which it was originally excited. In the no-mixing case the atom generally suffers no collisions in the  $P$  state before reradiating.

The significance of these two cases lies in the fact that when there is no mixing, both components of resonance radiation are effective and produce the same sense of polarization in the metastables. In the mixing case, radiation causing  $2^3S_1-2^3P_{1,2}$  transitions becomes considerably less effective in pumping and even tends to produce polarization of the opposite sense to that produced by the other component of resonance radiation. The pumping effect of the  $2^3S_1-2^3P_0$  transitions is almost the same for the mixing situation as for no mixing.

The net result is that He<sup>3</sup> lamps, will be more effective in the pumping process at low pressures where both components of the resonant light are absorbed and both are efficient in polarizing the metastables. At pressures above a few tenths of a mm, He<sup>4</sup> lamps will be more effective since the stronger of the two spectral lines is absorbed and is efficient in polarizing metastables, while only the weaker component of the He<sup>3</sup> lamp remains effective in the polarizing process.

The highest polarization measured by nuclear magnetic resonance was  $40 \pm 5\%$  in a 5 cm-diam Pyrex sphere with the He<sup>3</sup> gas pressure at 1 mm Hg. The light from two He<sup>4</sup> disk-shaped lamps described in Sec. III was directed on the sphere along the magnetic field from opposite directions and with opposite circular polarization.

The long relaxation time ( $T_r \sim 4000$  sec) suggests the possibility of obtaining this polarization at much higher pressures by compressing the gas from a very large cell into a small volume. Experiments requiring these higher pressures could be done on a cyclic basis during the periods that the He<sup>3</sup> nuclei were relaxing.

## V. APPLICATIONS

For some applications it is necessary to produce high-nuclear polarization in an environment somewhat different from the usual Pyrex sphere. One such application is in the design of a suitable cell in which to polarize He<sup>3</sup> for use in nuclear scattering experiments.<sup>3</sup>

The scattering cell requires transparent windows to pass  $1\text{-}\mu$  resonance radiation and other very thin windows to pass a beam of particles from an ion accelerator. In addition, provision has to be made to detect the particles scattered at a desired angle with respect to the primary beam. The scattering cell has to be vacuum tight and clean. Even in the presence of the discharge the amount of impurity driven from the walls must be small.

The field gradients seen by the He<sup>3</sup> nuclei as they

<sup>17</sup> A. V. Phelps and J. P. Molnar, Phys. Rev. **89**, 1202 (1953).

diffuse through the cell must be sufficiently small so that the nuclei are not caused to relax by the process discussed in Sec. IVB. This precludes the use of ferromagnetic materials in the cell construction, and requires that the materials have no small magnetic impurities that may be near the inside surface of the cell. For construction of the polarized  $\text{He}^3$  scattering cells used with the Rice University Van de Graaf accelerator,<sup>3</sup> materials were selected by measuring their effect on the  $\text{He}^3$  nuclear relaxation time  $T_r$  in the absence of a discharge. A sample of each material to be tested was placed inside a Pyrex cell in which the  $\text{He}^3$  polarization experiment could be performed. Since all that is required of  $T_r$  is that it be much longer than the pumping time  $T_p$  (typically about 30 sec), materials were accepted as suitable for cell construction if the tested  $T_r$  was greater than 600 sec. Brass, aluminum, copper, and Havar foils<sup>18</sup> appeared to have no detrimental effect on  $T_r$ . Ortec surface-barrier particle detectors were also tested and found acceptable. The relaxation time was considerably shortened, however, when the cell contained pieces of "nonmagnetic" type 303 stainless steel or any ferromagnetic materials such as thin nickel films.

The completed cell was machined from a single piece of brass. Vacuum seals were made around the Havar metal films and Pyrex windows by pressing them tightly against indium O-rings. The valve through which the system is evacuated and filled is a specially modified Veeco valve (R385) in which the valve seat and O-ring were replaced by indium and the magnetic parts replaced with brass.<sup>19</sup> Special care was taken to keep the distance from the center of the cell to the furthest point within the chamber as small as possible. This minimizes the relaxation effect of diffusion through field gradients that exist in the laboratory. The particle detectors were mounted inside the chamber on the first target cell, but

<sup>18</sup> Havar is the trade name of Hamilton Watch Company, Lancaster, Pennsylvania for a high-tensile strength spring steel alloy.

<sup>19</sup> F. D. Sinclair, *Rev. Sci. Instr.* **34**, 437 (1963).

on later cells the scattered particles passed through Havar foils to detectors outside.

When the cell was completed, it was placed on a vacuum system and baked in an oven at 135°C for a day or two. Higher temperatures could not be used without melting the indium seals. Before filling with  $\text{He}^3$ , the usual procedure was followed of running bright  $\text{He}^4$  discharges in the cell to clean the walls. In spite of these procedures the cell walls continued to outgas slowly. The cells remained sufficiently clean to perform experiments for a week or two. At this time they were again pumped out and refilled. The life of the cells was successfully extended by leaving an ion gage with an integral titanium getter attached to the filling tube. By opening this to the cell when it was not in use, the cells remained clean for several weeks.

The weak electrodeless discharge for producing metastables is created within the cell by fixing electrodes to the glass windows. 10–20% polarization could be obtained with two bright disk lamps. The reason for the somewhat lower polarization than in Pyrex cells is believed to be due to the less favorable distribution of the discharge in the metal chamber. In Pyrex spheres the discharge tends to concentrate in the center of the bulb when it is very weak, while in the metal cell, the discharge is drawn toward the walls. This is an example of the importance of the discharge level to the attainment of high polarization.

Another application of the  $\text{He}^3$  optical pumping process is in magnetometry, and will be reported elsewhere.<sup>20</sup>

#### ACKNOWLEDGMENTS

We wish to thank P. A. Franken, G. C. Phillips, M. de Wit, and C. A. Barlow, Jr. for valuable discussions related to various portions of this work. The authors are especially appreciative of the excellent technical assistance provided by Frederick D. Sinclair.

<sup>20</sup> L. D. Schearer, F. D. Colegrove, and G. K. Walters, *Rev. Sci. Instr.* (to be published).