

of this argument may not be as convincing as has been believed. Hasted<sup>35</sup> measured the detachment cross section for  $O^-$  with rare gas atoms and found this cross section to be independent of the manner in which the negative ions were produced. The two methods of production of  $O^-$  were by electron bombardment of  $O_2$  and by presumed thermal dissociation of  $N_2O$  to  $N_2$  and  $O$  followed by radiative attachment to  $O$ . Recently, it was found,<sup>21,22</sup> however, that  $O^-$  can be produced from  $N_2O$  by very low-energy electrons (peak at 0.7 eV) and that these ions may well be in the ground state of  $O^-$ . Thus, Hasted may have used identical  $O^-$  ions for his two experiments.

Recently, Dunn<sup>7</sup> showed that the angular distribution of the particles in dissociative attachment may be far from isotropic. In fact, depending on the intermediate state excited, the distribution could be sharply peaked in either the forward or perpendicular direction. Thus,

<sup>35</sup> J. B. Hasted, Proc. Roy. Soc. (London) **A222**, 74 (1954).

an instrument with a limited angular acceptance angle could discriminate against certain ionic species. This phenomenon may be the cause for much of the discrepancy between various experiments.

The possibility that kinetic-energy measurements in oxygen containing gases are affected by some, as yet unknown, difficulty cannot be dismissed. Further verification of the electron affinity of oxygen in different types of experiments is needed before one can state with certainty that an excited state of  $O^-$  exists.

#### ACKNOWLEDGMENTS

The author wishes to acknowledge frequent discussions with the Atomic Physics Group of the Westinghouse Research Laboratories. In particular, the author is indebted to R. K. Curran and A. V. Phelps for many helpful suggestions. The author is glad to acknowledge the technical assistance of J. Kearney who also took many of the data reported in this paper.

## Nuclear Magnetic Resonance in Gaseous $He^3$ †

K. LUSZCZYNSKI, R. E. NORBERG, AND J. E. OPFER\*

*Washington University, Saint Louis, Missouri*

(Received May 21, 1962)

Relative nuclear magnetic susceptibility, self-diffusion coefficient  $D$ , and spin lattice relaxation time  $T_1$  (characteristic of wall relaxation) were measured in  $He^3$  gas between 1.7 and 4.2°K in the range of pressure between 0.08 and 56 atm. These results were derived from measurements of free induction decays and spin echoes at 20.00 Mc/sec. Values for the density of the gas were deduced on the assumption that the gas obeys Curie's law. At moderate densities  $D$  was found to be inversely proportional to the density, while  $T_1$  was found to increase with the density of the gas. Experimental results obtained for the self-diffusion coefficient are compared with theoretical values. Details of experimental procedure are described.

#### INTRODUCTION

THE investigation of the transport processes of  $He^3$  gas and its equation of state at low temperatures has many interesting aspects, since we expect to find the properties of  $He^3$  gas to be affected appreciably by quantum mechanical effects.<sup>1</sup> Out of the four transport coefficients of viscosity, thermal conduction, thermal diffusion, and self-diffusion we can measure the coefficient of self-diffusion  $D$ , using nuclear magnetic resonance techniques.

The spin-echo technique<sup>2-4</sup> of nuclear magnetic resonance is particularly well suited to measurements of

self-diffusion in media containing magnetic nuclei in sufficient concentration. Also the amplitude of the nuclear magnetic signal, under certain well specified conditions, is proportional to the nuclear magnetic susceptibility of the sample. If the relationship between the susceptibility and density of the sample is known, it is then possible to derive relative values for the density, which, in turn, can be converted into absolute values by comparison with an absolute determination of the density at any suitable point. It is thus possible, in principle, to construct an equation of state of the sample, in this case  $He^3$  gas, and evaluate its virial coefficients. Results obtained from measurements of this type are reported here.

#### EXPERIMENTAL DETAILS

Nuclear magnetic resonance signals were observed at 20.00 Mc/sec, that is, in a field of  $H_0=6180$  G produced by a 12-in. electromagnet with a 3-in. gap.

The sample container was in the form of a nylon

† This work was supported by a research grant from the U. S. Army Research Office (Durham) and by an equipment loan contract from the Office of Naval Research.

\* National Science Foundation Cooperative Graduate Fellow.

<sup>1</sup> J. de Boer, *Progress in Low-Temperature Physics*, edited by C. J. Gorter (North Holland Publishing Company, Amsterdam, 1957), Vol. 1, p. 381 and references contained therein.

<sup>2</sup> E. L. Hahn, Phys. Rev. **80**, 580 (1950).

<sup>3</sup> H. Y. Carr and E. M. Purcell, Phys. Rev. **94**, 630 (1954).

<sup>4</sup> R. L. Garwin and H. A. Reich, Phys. Rev. **115**, 1478 (1959).

cylinder with a 0.25-cm internal diameter and a 0.5-cm length. The effective length of the sample container could be adjusted by means of a nylon piston fitting inside the cylinder. In this way, the sample volume could be changed and the sample could be confined to a region of optimum homogeneity of the radio-frequency field  $H_1$ . This arrangement is not essential to the experiment and, in fact, once the optimum volume of the sample is found the piston is locked so that the volume remains constant during the experiment: however, adjustment of the size of the sample from the top of the experimental probe was found to be very convenient.

The sample cell was sealed to a metal tube via a brass-nylon differential contraction seal.<sup>5</sup> The sample cell is shown schematically in Fig. 1. The nylon piece is screwed finger tight onto the brass plug until it just forms a vacuum tight seal at room temperature. A very thin layer of Apiezon-N grease on the sealing surfaces makes the adjustment of tightness less critical. As the cell is cooled, nylon contracts more than brass and a very tight high-pressure seal is thus formed. The cell has been tested up to 2000 psi at 77°K. The cell has been subjected subsequently to many temperature cycles between 1 and 300°K over a period of some six months and no evidence of a leak has been observed.

The sample container was surrounded by a brass can filled with He<sup>4</sup> exchange gas to provide the thermal contact with the liquid He<sup>4</sup> bath. The temperature of the sample was determined by means of carbon resistor thermometers used in conjunction with a Wien bridge oscillator,<sup>6</sup> calibrated against vapor pressure measurements of He<sup>4</sup>. Pressures over the He<sup>3</sup> sample below one atmosphere were measured with a mercury manometer, while pressures in excess of one atmosphere were determined with the aid of Bourdon gauges, connected directly to the sample tube. In order to reduce the "dead volume" associated with the sample cell, the high-pressure gauge was filled with glycerin and separated from the He<sup>3</sup> gas by a Teflon diaphragm. The He<sup>3</sup> gas was compressed in a mercury-oil Toepler pump.

The self-diffusion coefficient  $D$  was measured by means of the spin-echo technique.<sup>2-4</sup> A pulse sequence consisting of  $90^\circ-\tau-180^\circ-T-180^\circ$  gives rise to two echoes separated by a time interval  $t=2(T-\tau)$ . The ratio of the amplitudes of the two echoes is given by

$$R = \exp[-(\gamma G)^2 D t^3 / 12],$$

when the effect of the spin relaxation time is neglected. In our case the spin relaxation time  $T_2$  is very much longer than the "diffusion time" and the effect of  $T_2$  is therefore negligibly small. If the magnetic field gradient  $G$  is known, the self-diffusion coefficient  $D$  can be readily derived from a plot of  $\log R$  vs  $t^3$ . When a Carr-Purcell<sup>3</sup> sequence is applied to the sample, an estimate of the minimum value of  $T_2$  can be made. We have used

<sup>5</sup> F. J. Low and H. E. Rorschach, Phys. Rev. **120**, 1111 (1960).  
<sup>6</sup> B. J. Sandlin and J. C. Thompson, Rev. Sci. Instr. **30**, 659 (1959).

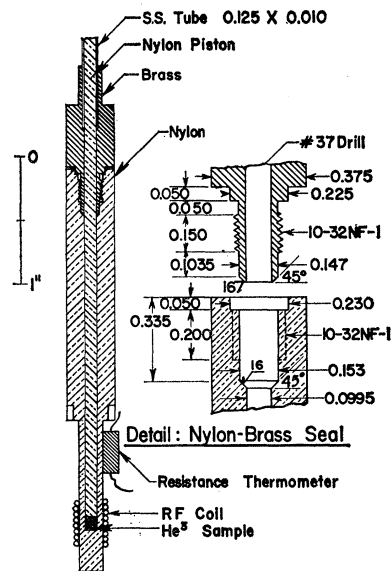


FIG. 1. Sample cell and a detail of the metal-plastic seal. The seal is made between the bevel surfaces.

this sequence also to verify that there was no appreciable convection in the sample during measurements.

The field gradient  $G$  was produced by a pair of coaxial coils attached to the pole pieces of the electromagnet. A power supply, current regulated to  $\pm 0.1\%$ , was used in conjunction with the gradient coils. Two independent methods were used to determine the field gradient  $G$ .

The normalized shape of the nuclear free precession signal in a cylindrical sample subjected to a linear field gradient  $G$  along  $H_0$  and perpendicular to the cylinder axis is given by

$$Y(t) = 2J_1(X)/X, \quad (1)$$

where  $J_1(X)$  is the first-order Bessel function,  $X = \gamma G r t$ ,  $\gamma$  is the gyromagnetic ratio of the sample,  $r$  is the radius of the cylinder, and  $t$  is the time from the origin. For example, from measurements of the time,  $t_{\text{null}}$ , at which the first null in  $Y(t)$  occurs it is possible to deduce  $G$ . This method gave  $G = 2.59$  G/cmA for a He<sup>3</sup> sample at low temperatures. Similar measurements were made with a glycerin sample at room temperature in a sample container, whose linear dimensions were increased by a factor of 2. These gave a value of  $G = 2.66$  G/cmA. The gradient can also be calculated approximately from the geometry of the system. Since the gradient coils are placed on the pole pieces of the magnet, it is necessary to take into account the effect of the magnetic material adjacent to the coils. If it is assumed that the pole faces are infinite in extent, the magnetic field in the magnet gap may be calculated by the method of images. Thus, a pair of real coils separated by a distance  $d$  leads to a series of virtual coil pairs with a successively increasing separation approximately in multiples of  $d$ . The field at the midpoint between the real coils is therefore in the

the form of  $\sum_{1,2,3,\dots} H(nd)$ . This series converges rapidly and a reasonably accurate value for the field associated with the coils is obtained by taking the first four terms of the series. An experimental determination of the field produced by the coils connected in the same sense gave a value of 20.6 G/A whereas the value calculated in the manner described is 20.4 G/A. (The field calculated for the coils in air is 8.9 G/A.) The rather close agreement between the calculated and experimental results is almost certainly accidental. The calculated value of the gradient produced by the coils connected in the opposite sense is 2.54 G/cmA. Determination of the absolute error in  $G$  is rather difficult in any case and at best we can only estimate it to be  $\pm 3\%$  on the basis of extensive measurements of this quantity. The value of  $G=2.66$  G/cmA was taken in our calculation of the self-diffusion coefficient. It is interesting to note that the gradient calculated for the same pair of coils in air is 1.65 G/cmA.

In determining the spin lattice relaxation time  $T_1$  a pulse sequence consisting in principle of two  $90^\circ$  pulses was used. The amplitude of the free induction signal following the second pulse was measured as a function of pulse separation  $\Theta$ . Since the amplitude of this signal is given by

$$y(\Theta) = y(\infty)[1 - \exp(-\Theta/T_1)], \quad (2)$$

$T_1$  can be determined from a plot of  $\log[1 - y(\Theta)/y(\infty)]$  vs  $\Theta$ .

The infinity value of the signal  $y(\infty)$  is also a measure of the volume susceptibility of the sample, if its volume is kept constant. Precautions were taken to insure a satisfactory stability of the apparatus. In addition, periodic checks were made with a standard signal to

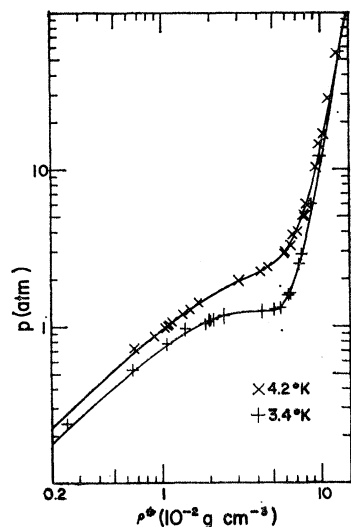


FIG. 2. The density of  $\text{He}^3$  gas  $\rho^*$  (derived from the susceptibility data) as a function of pressure at 3.4 and 4.2°K. Measurements of the density obtained in this experiment are normalized to those of Peshkov (reference 8) at  $p=1.110$  atm,  $T=3.40^\circ\text{K}$ , where  $\rho=2.12 \times 10^{-2}$  g/cm $^3$ . The solid lines represent the empirical formulas given by Eqs. (4) and (5).

correct for any long term drift in the gain of the receiving amplifiers.

The  $\text{He}^4$  content as measured in a room-temperature sample taken from our  $\text{He}^3$  storage container was less than 0.1%. This measurement was made with a Veeco (MS9)  $\text{He}^4$  leak detector modified to detect both  $\text{He}^3$  and  $\text{He}^4$ . Any remaining gas impurities were trapped in a long annular tube immersed in liquid  $\text{He}^4$  through which the  $\text{He}^3$  had to pass before reaching the sample cell.

## RESULTS AND DISCUSSION

The measurements quoted below were made between 1.7 and 4.2°K in the pressure range between 0.08 and 56 atm with corresponding density values between  $0.2 \times 10^{-2}$  to  $13.2 \times 10^{-2}$  g/cm $^3$ .

In deriving the density of the gas from measurements of volume susceptibility, we have assumed that the amount of  $\text{He}^3$  absorbed in the wall of the sample container is small compared to the bulk of the sample, and secondly that the susceptibility of the gas obeys Curie's law; that is to say, the molar susceptibility is independent of density and is inversely proportional to the absolute temperature. Absolute values of density were obtained by normalizing our results to an absolute measurement<sup>7,8</sup> at 3.40°K,  $p=1.110$  atm, and  $\rho=2.12 \times 10^{-2}$  g/cm $^3$ . The density so derived is denoted by  $\rho^*$ . Naturally, if our assumptions are correct then  $\rho^*$  should be the true density of the gas. The results are summarized in Figs. 2 and 3. Figure 3 also includes Peshkov's results<sup>8</sup> at 3.4°K with an extrapolation of his data to higher densities according to his formula<sup>8</sup>:

$$[(p/\rho) - 27.35T] = -2.3 \times 10^3 \rho + 1.8 \times 10^4 \rho^2, \quad (3)$$

where  $p$  is in atmospheres,  $\rho$  is in g/cm $^3$ , and  $T$  is in °K. Values of the density of  $\text{He}^3$  deduced from this equation agree with Keller's absolute density measurements<sup>7</sup> to better than 1%. This deviation is smaller than our experimental error and therefore we have used Peshkov's formula for *interpolation* purposes in normalizing our density measurements to those of Peshkov. We should like to emphasize that in each experimental run we redetermine  $y(\infty)$  at the reference point at 3.40°K and  $p=1.110$  atm, so that all our density determinations are normalized to the same point. The choice of this point was somewhat arbitrary; however, it was selected for the following reasons: It corresponds to a fairly high density which results in a good signal to noise ratio; it lies in the region where  $p$  is still almost directly proportional to  $\rho$ ; and finally, because it is at a temperature higher than the critical temperature so that there is no possibility of obtaining any errors arising from the presence of liquid  $\text{He}^3$ .

It is quite clear from Fig. 3 that higher virial coefficients have to be included in Eq. (3) in order to

<sup>7</sup> W. E. Keller, Phys. Rev. **98**, 1571 (1955).

<sup>8</sup> V. P. Peshkov, Soviet Phys.—JETP **6**, 645 (1958).

describe our data in the region where the density is greater than  $4 \times 10^{-2} \text{ g cm}^{-3}$ . In principle, these higher virial coefficients could be determined from the present data and we could use the resulting equation of state of He<sup>3</sup> gas to determine a number of thermodynamic quantities for the gas in a manner analogous to that used by Michels<sup>9</sup> for gaseous xenon.

Unfortunately, the relatively low accuracy of our measurements does not allow us to make accurate determinations of the virial coefficients. We estimate that our probable experimental error in  $\rho^*$  is of the order of  $\pm 2\%$  for  $\rho^*$  in the range between  $1 \times 10^{-2} \text{ g/cm}^3$  and  $5 \times 10^{-2} \text{ g/cm}^3$  and that the error increases to  $\pm 5\%$  outside this range. The probable error increases even

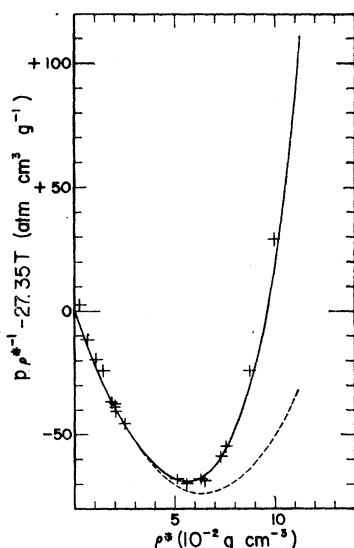


FIG. 3. Relationship between pressure and density of He<sup>3</sup> gas at 3.40°K; +, experimental points, this research; solid line, empirical formula, Eq. (5); dashed line, Peshkov's formula, Eq. (3).

further for very low gas densities. Within the experimental error the results obtained for  $\rho^*$  fit the following empirical formulas (see Fig. 2), at 4.2°K:

$$[(p/\rho^*) - 27.35T] = -2.3 \times 10^3 \rho^* + 1.97 \times 10^4 \rho^{*2} + 5.7 \times 10^7 \rho^{*6}, \quad (4)$$

and at 3.4°K:

$$[(p/\rho^*) - 27.35T] = -2.3 \times 10^3 \rho^* + 1.84 \times 10^4 \rho^{*2} + 6.6 \times 10^7 \rho^{*6}. \quad (5)$$

At temperatures below the critical point our results are adequately described by Eq. (3). These results do not indicate any systematic deviation from Curie's law greater than the experimental error in the region where the density of He<sup>3</sup> gas (vapor) is known.<sup>10</sup> The signal-to-noise ratio of our apparatus can be improved so that

<sup>9</sup> A. Michels, T. Wassenaar, and P. Louwse, *Physica* **20**, 99 (1954).

<sup>10</sup> In a private communication Dr. R. H. Romer pointed out that  $\rho^*$  may not be equal to  $\rho$  at 3°K in the density region below  $1 \times 10^{-3} \text{ g/cm}^3$ . [R. H. Romer, *Bull. Am. Phys. Soc.* **7**, 76 (1962)].

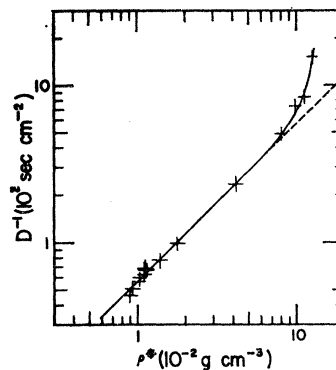


FIG. 4. Relationship between the coefficient of self-diffusion  $D$  and density of He<sup>3</sup> gas at 4.2°K.

it should be possible to reduce our experimental error and extend the present measurements to much lower densities. In the high-density region, *absolute* measurements of He<sup>3</sup> gas density, for example, at 3.40°K and pressures in excess of 1 atm would be quite valuable, because it would then be possible with the aid of the present data for  $\rho^*$  to check whether He<sup>3</sup> obeys Curie's law for high densities of the gas. We may anticipate the answer to this question on the basis of results obtained in liquid He<sup>3</sup>. Our measurements of the molar susceptibility of liquid He<sup>3</sup> at 2°K, where absolute density values are available,<sup>11</sup> show that the susceptibility is independent of density; specifically, the molar susceptibility is found to be constant to within  $\pm 1\%$  for an increase in the density of the liquid from  $7.9 \times 10^{-2} \text{ g/cm}^3$  to  $11.6 \times 10^{-2} \text{ g/cm}^3$ .

The results obtained for the self-diffusion coefficient  $D$  are summarized in Figs. 4 and 5. Figure 4 shows

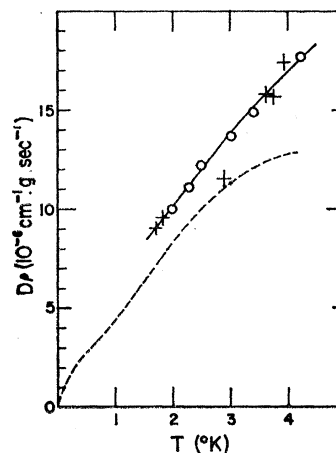


FIG. 5. Comparison of experimental and theoretical results for  $D\rho$  in He<sup>3</sup> gas in the normal density region. The dashed curve, theoretical results, (reference 1) second approximation; O, experimental points (average of several measurements made at different values of  $\rho^*$ ); +, experimental points (single measurements.)

<sup>11</sup> R. H. Sherman and F. J. Edeskuty, *Ann. Phys. (New York)* **9**, 522 (1960).

( $1/D$ ) as a function of  $\rho^*$ , at 4.2°K. Similar curves were obtained at other temperatures.  $D$  is found to be inversely proportional to  $\rho^*$ , that is,  $D\rho^*$  is independent of  $\rho^*$ , for values of  $\rho^*$  less than  $6 \times 10^{-2}$  g/cm<sup>3</sup>. As the density of the gas is increased further, the coefficient of self diffusion  $D$  decreases rapidly. The bend in  $D$  vs  $\rho^*$  plot occurs approximately at the same value of  $\rho^*$  for all temperatures between 3.4 and 4.2°K. Below the critical point the density is less than  $4 \times 10^{-2}$  g/cm<sup>3</sup> and the plots of  $D$  vs  $\rho^*$  are quite linear. The slope (i.e.,  $D\rho^*$ ) of the linear portion of the graph is temperature dependent. This temperature dependence of  $D\rho^*$  is shown in Fig. 5, where the calculated values<sup>1</sup> for  $D\rho$  are also plotted for the sake of comparison.

In order to make this comparison, we recall some of the basic assumptions made in the theoretical approach. The theoretical results refer to a region of "normal" density. In this region the density is on the one hand high enough to make the mean free path small compared to the dimensions of the sample container, i.e., Knudsen phenomena can be neglected, and on the other hand the density is so low that the effects due to triple collisions are negligibly small, or in other words, the mean free path remains large compared with distances characteristic of the atomic interaction. A very rough estimate of the mean free path shows that measurements reported here should refer for the most part to the region of normal density. For example, as the density changes from  $0.1 \times 10^{-2}$  to  $6 \times 10^{-2}$  g/cm<sup>3</sup>, the mean free path in He<sup>3</sup> goes from about 700 to 12 Å, respectively. Since the radius of the He<sup>3</sup> atom is about 2.5 Å, we should expect agreement between our experimental data and the theoretical results, if the density of the gas is not too high.

In the region of normal densities the self-diffusion coefficient is shown<sup>1</sup> in first approximation to be given by

$$D_1 = 3kT/[8mn\Omega^{1,1}(T)], \quad (5)$$

where  $m$  is the atomic mass of He<sup>3</sup>,  $n$  is the total number density of atoms,  $k$  is Boltzmann's constant,  $T$  is the

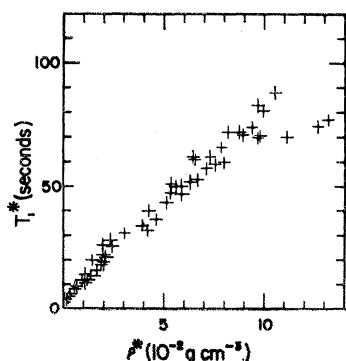


FIG. 6. Spin-lattice relaxation time  $T_1^*$  for He<sup>3</sup> gas as measured in a cylindrical nylon sample container (i.d. 0.25 cm, length 0.5 cm) in the temperature range between 1.7 and 4.2°K. At constant density, in the range of densities measured here, the temperature dependence of  $T_1^*$  is smaller than the apparent scatter in  $T_1^*$  values.

absolute temperature, and  $\Omega^{1,1}(T)$  is the "total" cross section of two He<sup>3</sup> atoms, which is independent of density in the normal density region. Hence, it follows that  $D$  should be inversely proportional to the density of the gas at constant temperature. This is clearly demonstrated in Fig. 4.  $D\rho^*$  is independent of density up to  $\rho^* \cong 6 \times 10^{-2}$  g/cm<sup>3</sup>. Any further increase in the density of the gas reduces diffusion rapidly, indicating that the mean free path becomes comparable to the interatomic distance, that is, the gas is now outside the normal density region. The dependence of  $D\rho^*$  and  $D\rho$  on temperature for normal densities is illustrated in Fig. 5. The experimental results generally fall higher than the calculated values of  $D\rho$ . Also  $D\rho^*$  apparently increases more rapidly with temperature than predicted. The reason for this discrepancy is not clearly understood. On the one hand, it is possible that the constants of the interaction potential used in the calculation of the scattering cross sections are in error, and, on the other, we should remember that the coefficient of self-diffusion, as derived from nuclear magnetic resonance experiments, may be subject to some systematic errors, especially in the external field gradient. However, the disagreement between the theoretical and experimental results is outside the experimental error. The maximum probable error in  $D$  is estimated to be  $\pm 6\%$ . We conclude, therefore, that the theoretical calculations should be examined more closely. It is worth noting here that a comparison of the viscosity data<sup>1</sup> shows that the experimental values fall below those derived from theory. This trend is in qualitative disagreement with the discrepancy of the self-diffusion data.

Figure 6 summarizes the experimental results for the spin lattice relaxation time, for temperatures between 1.7 and 4.2°K, plotted against the density of the gas  $\rho^*$ . It appears that this relaxation time is almost independent of temperature and that it increases with the density of the gas. For a monatomic gas we should expect  $T_1 \propto 1/\rho$ , while for a diatomic gas  $T_1 \propto \rho$  is predicted.<sup>12</sup> Since we have to dismiss the possibility of a large amount of He<sup>3</sup> molecules<sup>13</sup> being present in the gas, we conclude that the spin relaxation time is determined here by the rate of diffusion of He<sup>3</sup> atoms to the walls of the sample cell where the atoms are relaxed. The spin lattice relaxation time measured here is denoted by  $T_1^*$  to distinguish it from the bulk relaxation time  $T_1$ . We find that within the experimental error the recovery curve  $y(\Theta)$  is governed by a single time constant as indicated in Eq. (2). This is a little surprising, especially in view of the fact that the shape of the free precession signal depends on the waiting time  $\Theta$ , that is, to say, the time interval  $t_{\text{null}}$ , Eq. (1), increases with

<sup>12</sup> N. Bloembergen, *Nuclear Magnetic Relaxation* (W. A. Benjamin, New York, 1961) and A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, Oxford, England, 1961).

<sup>13</sup> N. Bernardes and H. Primakoff, *J. Chem. Phys.* **30**, 691 (1959).

$\Theta$  as long as  $\Theta \lesssim T_1^*$ . For example, for  $\rho^* = 7.3 \times 10^{-2}$  g/cm<sup>3</sup> and  $\Theta/T_1^* = 0.03$  it is found that  $t_{\text{null}} = 0.20$  msec in an external field gradient of 5.32 G/cm, whereas for  $\Theta/T_1^* = 4.0$ ,  $t_{\text{null}} = 0.27$  msec. This means that for  $\Theta \ll T_1^*$  the signal is more characteristic of a cylindrical shell than a cylinder. Such a behavior is again consistent with wall relaxation. In this connection it is important to note that in determining the field gradient  $G$  from the shape of the nuclear signal, waiting times  $\Theta \gg T_1^*$  must be used, to avoid large errors in  $G$ .

It should be noted that  $1/T_1^*$  is not a linear function of the self-diffusion coefficient  $D$ , especially at the two extremes of density values. In the region of low-density values an extrapolation of  $T_1^*$  to  $\rho^* = 0$ , has a finite intercept on the  $T_1^*$  axis of the order of 2 sec; this should be compared with the plot of  $(1/D)$  vs  $\rho^*$  which goes through the origin. If such an extrapolation is permissible, it would appear that  $T_1^*$  remains finite even though the mean free path of the atoms is much larger than the dimensions of the sample cell. A behavior of this type would be consistent with a situation where the atoms spend a finite time on the wall of the container before they are relaxed. By extending the present measurements to lower densities, it should be possible to add greater detail to this picture. In the region of high-density values,  $T_1^*$  does not reflect the rapid decrease of the self-diffusion coefficient as the density is increased. In fact, Fig. 6 shows that  $T_1^*$  begins to level off above  $\rho^* = 10 \times 10^{-2}$  g/cm<sup>3</sup>. This can be explained in terms of the effect of the bulk relaxation time, which is inversely proportional to the density of the gas.<sup>12</sup> On the basis of this argument we find that the bulk spin-lattice relaxation time in He<sup>3</sup> gas is equal to about 300 sec for  $\rho^* = 13.2 \times 10^{-2}$  g/cm<sup>3</sup> at 3.4°K. In any case, since the wall relaxation is dependent presumably on the nature of the wall surfaces and impurities imbedded there,  $T_1^*$  may vary from one experimental run to another.

Our experiments do not show any pronounced fluctuations in  $T_1^*$  in the same sample cell, which was used throughout this set of experiments.  $T_1^*$  should also depend on the geometry of the sample container. A similar sample container made out of a different material would most likely show a different  $T_1^*$  in the region of moderate densities. These measurements of  $T_1^*$  though not of particular interest in themselves in so far as they represent wall relaxation, may however throw some light on the spin lattice relaxation time measurements in liquid He<sup>3</sup>, reported in the literature.<sup>14</sup> This question will be discussed elsewhere.<sup>15</sup>

### CONCLUSIONS

The experimental results for  $D\rho$  reported here fall higher than the theoretical results.<sup>1</sup> In addition, our measurements show that  $D\rho$  increases with temperature more rapidly than predicted by de Boer.<sup>1</sup> Our results on the volume nuclear magnetic susceptibility of He<sup>3</sup> gas do not indicate, outside the experimental error, any deviations of the magnetic susceptibility from Curie's law in the low-density region where absolute measurements for the density of the gas are available.<sup>7,8</sup> Absolute measurements of the density of the gas at high pressures in the vicinity of the critical point would be quite valuable, because it would then be possible to see whether He<sup>3</sup> continues to obey Curie's law as the density of the gas is increased.

### ACKNOWLEDGMENT

We would like to express our thanks to L. Vossel for his expert technical assistance with the cryogenic apparatus.

<sup>14</sup> G. K. Walters, *Proceedings of the Second Symposium on He<sup>3</sup>* (Ohio State University Press, Columbus, Ohio, 1960), p. 37.

<sup>15</sup> J. Gaines, K. Luszczynski, and R. E. Norberg (to be published).