where ζ is the transform variable. By taking the Laplace transform of (7.5), we obtain

$$(\rho_0 T_0/\beta)[\zeta\bar{\theta}-\theta(x,0)]-(\gamma T_0/\beta) \times[\zeta\bar{e}_{ii}-e_{ii}(x,0)]+(\tilde{\kappa}/T_0)\nabla^2\bar{\theta}=0, \quad (7.8)$$

where $\theta(x,0)$ and $e_{ii}(x,0)$ are the temperature and dilatation at $t=0$. It may be deduced from (7.6) and (7.8), and the fact that conservation laws are the same whether the medium has viscosity or not, that: *Theorem of Correspondence: The solution of the Laplace transform of basic equations of thermo-visco-elasticity is the same as that of the thermoelasticity with the transformed body forces* $\bar{f}_i$ *and surface tractions* $\bar{\sigma}_i$ *in the latter replaced by* $\tilde{f}=\bar{f}_i-\sigma_{ji,j}{}^0$ *and* $\tilde{\sigma}_i=(\bar{\sigma}_{ij}-\sigma_{ij}{}^0)n_j$, *and* λ, μ *and* κ *replaced by* $\tilde{\lambda}$, $\tilde{\mu}$ *and* $\tilde{\kappa}$.

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Nuclear Spin Relaxation in Liquid He^3. II*

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The nuclear spin thermal relaxation time, T_1, of He^3 nuclei in pure liquid He^3 at its saturated vapor pressure has been measured at temperatures between 0.8°K and 3.1°K, at static magnetic fields from 1560 gauss to 12 200 gauss. No dependence of T_1 on the static magnetic field was observed. The measured relaxation times increase gradually from 300 seconds at 0.8°K to 650 seconds at 3.1°K. These measured relaxation times do not appear to have been significantly shortened by wall relaxation processes. These results, which do not agree with measurements made elsewhere, are in good agreement with the short correlation time form of the Bloembergen, Purcell, Pound theory of spin relaxation in liquids.

INTRODUCTION

Recently we reported measurements[1] of the thermal spin relaxation time, T_1, of He^3 nuclei in pure liquid He^3 at its saturated vapor pressure. Previous measurements in this laboratory[2,3] had yielded different T_1 values depending on the container used. It had been concluded from these earlier measurements that in at least some of the containers used the relaxation time was being artificially shortened by wall relaxation or possibly by some impurity. The combination of the long relaxation time in liquid He^3 and the large diffusion coefficient makes it difficult to avoid wall effects by simply using a very large sample. In these circumstances, the longest T_1 measured at any given temperature, pressure, etc., probably lies closest to the value characteristic of the bulk liquid, unless there is some systematic error which gives spuriously long T_1 values. The safest procedure seems to be to construct containers of a material believed to be relatively ineffective at relaxing He^3 spins; if then approximately the same results are obtained in two containers of quite different sizes, one has presumably measured something close to the "true" T_1.

Fortunately, it was discovered early that Pyrex surfaces do not relax He^3 spins very fast.[2] The measurements reported in I were therefore made in two Pyrex bulbs of different sizes and the results obtained from the two bulbs were in good agreement with each other. All the data in I were taken in a static magnetic field of 9000 gauss and seemed to be in reasonable agreement with the BPP[4] theory of spin relaxation in liquids, provided that one makes the assumption that the correlation time for the motion of the atoms ($\tau_c \cong r^2/10D \cong 10^{-12}$ seconds, where r, a typical inter-atomic distance, is a few A, and $D \cong 10^{-4}$ cm^2/sec.[5,6]) is much less than the Larmor period in available magnetic fields ($2\pi/\omega_0 \gtrsim 2\times10^{-8}$ seconds). The short τ_c approximation would appear to be an extremely good one for He^3, particularly at high temperatures ($\sim$3.0°K).

Shortly after the completion of the measurements reported in I, the subject was re-opened by the report[7] of a previously unsuspected magnetic field dependence of T_1. Low and Rorschach, making measurements at

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[1] R. H. Romer, Phys. Rev. **115**, 1415 (1959), hereafter referred to as I.

[2] W. M. Fairbank, W. B. Ard, H. G. Dehmelt, W. Gordy, and S. R. Williams, Phys. Rev. **92**, 208 (1953).

[3] W. M. Fairbank, W. B. Ard, and G. K. Walters, Phys. Rev. **95**, 566 (1954).

[4] N. Bloembergen, E. M. Purcell, and R. V. Pound, Phys. Rev. **73**, 679 (1948), hereafter referred to as BPP.

[5] R. L. Garwin and H. A. Reich, Bull. Am. Phys. Soc. **3**, 133 (1958).

[6] R. L. Garwin and H. A. Reich, Phys. Rev. **115**, 1478 (1959).

[7] F. J. Low and H. E. Rorschach, post-deadline paper presented to the American Physical Society, April 30, 1959.

the saturated vapor pressure, at 2.0°K, in a $\frac{1}{8}$-in. i.d. cylindrical nylon container, using a different technique, found T_1 values which rose from less than 50 seconds at low fields ($\sim$1 gauss) to about 175 seconds at 3000 gauss and 400 seconds at 13 500 gauss. (The T_1 value reported in I for 2.0°K and 9000 gauss is approximately 400 seconds.) Since one result of the BPP theory in the short τ_c approximation is that T_1 should be nearly field independent,[8] the existence of a strongly field dependent T_1 at temperatures as high as 2.0°K, where one might expect the BPP theory to be at least approximately valid, would require a drastic revision of our ideas of atomic motions in liquid He^3. Because Low and Rorschach had no internal evidence for the absence of wall relaxation in their experiment, it seemed important to measure T_1 at various fields in large Pyrex bulbs. We have accordingly made T_1 measurements at fields between 1560 gauss and 12 200 gauss, at temperatures from 0.8°K to 3.1°K. In view of the very large discrepancies between the results of various workers in this field, the presentation of the results will be preceded by a detailed description of the experimental procedures used.

EXPERIMENTAL PROCEDURE

A. Cryogenics

At the start of each run, the He^3 sample, stored at room temperature, was condensed into the low-temperature apparatus via a purifying trap cooled by liquid He^4 (4.2°K). The data above 1°K were obtained by condensing the He^3 sample into a Pyrex bulb, blown at the end of a long Pyrex capillary. No special cleaning or baking procedure was used on the Pyrex bulb. The sample bulb was either immersed directly in the liquid He^4 bath or else was inside a copper can in the He^4 bath, the sample being cooled by a small amount of He^4 exchange gas. Temperatures above 1°K were determined from measurements of the vapor pressure of the He^4 bath and of the vapor pressure of the He^3 sample. Agreement between the two temperature determinations gave a cross-check on the purity of the sample (better than 99% He^3).

The major discrepancies between our data and those of other workers are in the temperature range above 1°K; therefore the main emphasis was put on obtaining further results in this temperature range. However, some data near 0.8°K were obtained with a rudimentary He^3 pumping system, which consisted of a 7-mm i.d. Pyrex sample bulb, blown at the end of a glass pumping lead which tapered from 2-mm i.d. near the sample bulb to 8-mm i.d. near the top of the flask. The lower part of the pumping lead, which was thermally shorted to the He^4 bath about 30 cm above the sample bulb, projected into a can which could either be evacuated or filled with He^4 exchange gas. The bulb and the lower part of the pumping lead were filled with liquid He^3 at 1.32°K; upon then removing the exchange gas and then pumping on the He^3, the temperature of the He^3 in the bulb was reduced. The temperature reached was estimated from the height of the nuclear resonance signal relative to its height at 1.32°K, by means of the susceptibility data.[3] [The nuclear resonance coil surrounded the bulb and the lowest few cm of the pumping lead. Because of the field gradient of the magnet, the nuclear resonance signal from the He^3 in the pumping lead occurred at a different time than did the signal from the He^3 in the sample bulb as the magnetic field was swept through the resonance. In estimating the temperature reached, and in measuring T_1, the height of the main signal (from the bulb) was used, while the shape and size of the subsidiary signal (from the pumping lead) served to monitor the amount of liquid left above the bulb.] In the one run made with this apparatus, the temperature reached was limited by a large heat leak ($\sim$500 ergs/sec), due probably to residual exchange gas or to a radiation leak.

B. Nuclear Magnetic Resonance

A simple bridgeless nuclear resonance spectrometer[9] was used. An rf oscillator fed, through a high impedance, the tuned circuit containing the sample, which was in turn connected to the input of an rf amplifier. The rectified output of the rf amplifier was direct coupled (via a battery which bucked out most of the rectified rf carrier) to a high gain oscilloscope. The amplitude of the He^3 resonance absorption signal was then observed by sweeping the magnetic field slowly through the resonance value. The rf coil was external to the Pyrex sample bulb.

At each different magnetic field, the rf oscillator was set at the appropriate Larmor frequency. The sample coil was tuned by inserting the appropriate length of cable between the flask and the amplifier. In this way it was possible, with one of the sample coils used, for instance, to obtain nuclear resonance signals at Larmor frequencies between 5 Mc/sec and 40 Mc/sec without having to make any changes in the low-temperature part of the apparatus. (On some occasions, a tuning condenser was placed directly in parallel with the coil. A word of warning is in order here. The ceramic condensers used, Centralab type TCZ, have a remarkably temperature independent capacitance from 300°K to below 1°K; however, they were found to be strongly

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[8] The important result that T_1 should be approximately field independent if $\omega_0\tau_c \ll 1$ can be seen from the following considerations. The spin relaxation of a given nucleus is assumed to be caused by the fluctuating local magnetic fields produced by the magnetic moments of other atoms as they move about; specifically by the Fourier components of these local fields at frequencies near ω_0. τ_c is a measure of the time interval during which the local field at a nucleus is approximately constant. The Fourier spectrum of the local fields is therefore "white" up to frequencies of the order of $1/\tau_c$, leading to a relaxation time independent of ω_0 if $\omega_0 \ll 1/\tau_c$.

[9] J. Hatton and B. V. Rollin, Proc. Roy. Soc. (London) **199**, 222 (1949).

magnetic at low temperatures and must be kept several inches from the sample.)

The filling factor for the He^3 in the sample coil was kept small to avoid corrections for radiation damping effects, which are often appreciable for He^3. If these effects are not kept small, then the change in the rectified output of the rf amplifier upon passing through the nuclear resonance is no longer strictly proportional to the nuclear spin susceptibility.[10,11]

Most of the T_1 measurements were made by first sweeping the external magnetic field rapidly back and forth through the resonance (about once per second, with an amplitude of about 1 gauss), simultaneously applying an rf magnetic field sufficiently large to "saturate" the nuclear resonance signal. Conditions were usually such that the signal was nearly saturated within 10 or 20 seconds. This saturating rf field was usually left on for a time equal to several times T_1. In order to observe the growth of the signal, the rf field was then reduced and the rapid field sweep turned off. The external magnetic field was then set about 1 gauss above the resonance field, and then, typically once every minute, the external field was swept slowly through the resonance value at about 0.1 gauss/sec. The field sweep was derived from the saw-tooth oscilloscope sweep, which was manually triggered. The oscilloscope trace was photographed on each passage through resonance with decreasing magnetic field; the passages through resonance which occurred with rapidly increasing magnetic field just after each downward sweep were ignored. The amplitudes of successive photographs were then plotted as a function of time and analyzed to find T_1. The growth curve was assumed to be a simple exponential; there is no experimental evidence that it was not.

In order to make sure that the rf magnetic field used in observing the growth of the signal was not itself large enough to saturate the signal appreciably, many of the recoveries were repeated with different rf voltages applied to the sample coil during the observation of the growth of the signal. It was found, as expected, that when the rf field was quite large, the measured T_1 decreased with increasing rf field, but that when the rf field was kept sufficiently small, the measured T_1 was independent of the rf field strength.

It should be emphasized that during the observation of any one recovery the rf field was kept constant, and the rate of change of the external field was the same on each photographed passage through resonance. Thus, even though the amplitude of the signal may depend on these parameters, the time constant characterizing the growth of the observed signal should be the correct time constant describing the growth of the nuclear spin magnetization.

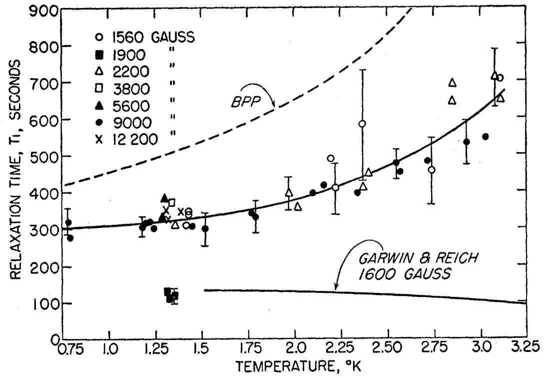

FIG. 1. Spin relaxation time, T_1, in pure liquid He^3 under its saturated vapor pressure, as a function of temperature, at various values of static magnetic field. Solid curve is a fit of the data. Dashed curve is a plot of the BPP expression [Eq. (2)]. See text for a discussion of the 1900 gauss data. The lower solid line is taken from reference 6 and represents T_1 measurements made at 1600 gauss and 2.38 atmospheres pressure.

Several measurements were made by two other methods. These methods differ from the first one described only in that different means were employed to produce the nonequilibrium spin magnetization at the beginning of an observation; the method of observing the recovery toward equilibrium was in all cases the same. One of these methods was to sweep through the resonance once under adiabatic fast passage conditions.[12] This produced an inversion of the spin level populations, and when the rf field was then reduced to the observing level, a stimulated emission signal (of sign opposite to that produced by an absorption signal) was observed. This emission signal decreased rapidly to zero and then built up as an ordinary absorption signal. Another method consisted of turning the magnet off for about 20 minutes, then suddenly turning the magnet on to the desired field and watching the absorption signal grow. This last method involved no manipulation of the electronics during a measurement. Measurements made by all three methods were in agreement with each other.

RESULTS

The results are shown in Fig. 1, which also includes the data from I. Each point represents a single T_1 measurement. Typical error estimates are shown. Each point is accurate to at best about ±10%, and for many points the uncertainty is substantially larger. The uncertainty arises from the ambiguities in fitting the observed voltage *vs* time plots with exponentials of various time constants. The signal jitter which produces this ambiguity probably arises from amplitude fluctuations in the rf oscillator output. The smooth curve in Fig. 1 lies within experimental error of all our measurements except for those made in one bulb discussed below. Several different spherical Pyrex bulbs were

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[10] C. R. Bruce, R. E. Norberg, and G. E. Pake, Phys. Rev. **104**, 419 (1956).

[11] G. K. Walters, Ph.D. thesis, Duke University, 1956 (unpublished).

[12] F. Bloch, Phys. Rev. **70**, 460 (1946).

used, in addition to the two Pyrex bulbs (5-mm and 10-mm i.d.) described in I. Most of the new data were taken in a 12-mm i.d. bulb, blown at the end of a long glass capillary. The points near 0.8°K were obtained in a 7-mm i.d. Pyrex bulb, as described previously. We have no evidence that wall effects were absent in the 0.8°K data; the true T_1 may be higher than the value shown.

At 1900 gauss, in one bulb, 8.5-mm i.d., we obtained T_1 values much shorter than any others we have measured. (See Fig. 1.) Unfortunately, this bulb was broken before further data could be obtained from it. However, since the only difference of which we are aware between the data obtained from this bulb and from all the others lies in the different containers, we believe that these short T_1 values result from some contamination. These points do show that our electronics is capable of measuring T_1's shorter than 300 seconds. This particular bulb had more opportunities to become contaminated than any other. For instance, the inside of the tube leading to this bulb (and only this bulb) had been coated with Aquadag, and flakes of Aquadag were later discovered to have fallen into the bulb. (This tube and bulb were designed, but never used, as a modified version of the He^3 pumping system previously described. The Aquadag was added, at a bend in the tube above the sample, in order to trap out high-temperature radiation.) There is no proof that it was the Aquadag that caused the short T_1 values, but the fact remains that all the T_1 values obtained from those bulbs which we have been careful to keep clean lie within experimental error of the smooth curve in Fig. 1. We conclude that our measurements give no evidence of any field dependence of T_1. The consistency of our data (in particular the agreement between the T_1 values obtained from different bulbs) leads us to believe that wall relaxation has not significantly shortened our results and that the smooth curve in Fig. 1 is a good representation of the relaxation time characteristic of the bulk liquid.

The line width, $\sim$0.1 gauss, was in all cases due to the inhomogeneity of the magnet. The magnetic field at which the resonance in liquid He^3 occurs was measured at 1.3°K and 1560 gauss by comparing the He^3 resonance frequency with the frequency of the proton resonance in mineral oil. The shift from the result expected from room temperature measurements on He^3 gas[13] was less than 1 part in 10^4.

DISCUSSION

Our T_1 measurements are in agreement with the short correlation time form of the BPP theory, in so far as that theory predicts a field independent T_1. A quantitative comparison of our data with the BPP theory can now be made. In I we compared our data with the BPP expression (for short τ_c), as modified by Torrey[14,15] and by Kubo and Tomita,[16] which involves the viscosity coefficient:

$$1/T_1=(6\pi^2/5)\gamma^4\hbar^2N\eta/kT \tag{1}$$

Reasonably good agreement was shown to exist between Eq. (1) and our results, although the right-hand side of Eq. (1) has a stronger temperature dependence than do our experimental T_1 values. Torrey's results were actually expressed in terms of the diffusion coefficient, and Eq. (1) was obtained from Torrey's expression by means of the Stokes relationship: $3\pi\eta aD/kT=1$. The T_1 expression as given by Torrey[15] is

$$1/T_1=(2\pi/5)\gamma^4\hbar^2N/aD, \tag{2}$$

and this expression has been plotted in Fig. 1, with a= atomic diameter = 2.7 A, the diffusion coefficient as measured by Garwin and Reich,[5,6] extrapolated by us to the saturated vapor pressure, and N, the number density of He^3 atoms, as measured by Kerr.[17] Equation (2) was actually derived by Torrey for the case in which the size of the elementary step in the diffusion process is comparable to or smaller than the atomic diameter. More exactly, Eq. (2) is supposed to hold under the approximation $\langle r^2\rangle\ll 6a^2$, where $\langle r^2\rangle$ is the mean squared flight distance in the diffusion process. Careri *et al.*[18] have suggested that another expression given by Torrey, applicable to the case in which the size of the elementary step in the diffusion process is large in comparison with the atomic diameter, specifically $\langle r^2\rangle\gg 6a^2$, may be a more reasonable one for the case of He^3. This expression is obtained by multiplying the expression for $1/T_1$ given in Eq. (2) by the factor $5\langle r^2\rangle/12a^2$. $\langle r^2\rangle$ is not directly known, but it seems reasonable that for an "open" liquid like He^3 (volume $\sim$38 cc/mole) the factor $5\langle r^2\rangle/12a^2$ would have a value somewhat greater than unity and would be roughly temperature independent. If this is correct, then the use of this alternative expression would decrease the calculated T_1 values slightly, leaving the temperature dependence approximately unchanged. Probably neither approximation regarding the relative values of $\langle r^2\rangle$ and $6a^2$ is a good one for liquid He^3. Seidel[19] has recently extended Eq. (2) to situations in which the parameter $\langle r^2\rangle/6a^2$ is taken into account in a first approximation, without making the extreme assumption $\langle r^2\rangle/6a^2\gg 1$. Using for $\langle r^2\rangle$ the values which result if one considers the He^3 atoms as forming a cubic lattice of the proper density, Seidel calculates approximate T_1 values as follows: 1°K, 310 seconds; 2°K, 450 seconds; 3°K, 800 seconds. Another discussion of spin relaxation in liquids, based on the BPP approach, has recently been given by

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[13] H. L. Anderson, Phys. Rev. **76**, 1460 (1949).

[14] H. C. Torrey, Phys. Rev. **92**, 962 (1953).

[15] H. C. Torrey, Suppl. Nuovo cimento **9**, 95 (1958).

[16] R. Kubo and K. Tomita, J. Phys. Soc. (Japan) **9**, 888 (1954).

[17] E. C. Kerr, Phys. Rev. **96**, 551 (1954).

[18] G. Careri, I. Modena, and M. Santini, Nuovo cimento **13**, 207 (1959).

[19] G. Seidel (private communication). The author wishes to thank Dr. Seidel for permission to quote these results.

Oppenheim and Bloom.[20] If applied to liquid He^3, their calculations yield a formula like our Eq. (2) but with a slightly different coefficient. We conclude that the short correlation time form of the BPP theory is in quite good agreement with our results.

The reason for the use of Eq. (1) in I was simply that the required data are all known,[17,21] and it was not necessary to make any choice for the atomic diameter or to make an extrapolation of the experimental diffusion coefficient data. It should be noted that the Stokes relationship *is* approximately correct for liquid He^3; i.e., the ratio $3\pi\eta aD/kT$ has the value 2.3 at 1.2°K and the value 1.3 at 3.0°K, for example. However, the deviations from the Stokes relationship are such that the temperature dependence of our T_1 data appears to be closer to the BPP expression when Eq. (2) is used than when Eq. (1) is used.

Although our data do indicate that the BPP theory provides a good description of spin relaxation in liquid He^3, over a temperature range of four to one, this conclusion must be regarded as a tentative one in view of the discrepancies between our data and the data obtained by other workers, to be described below. Even if one can conclude that the BPP theory is applicable to He^3 above 0.8°K, it would be extremely interesting to carry out T_1 measurements at temperatures well below the spin degeneracy temperature. In this quantum region, something more complicated than the single-atom BPP picture may well be needed. In particular, the concept of a diffusion coefficient in a quantum liquid, where identity effects are important, seems quite different from what it is at high temperatures; the relationship between measured D and T_1 values might well be quite different from that given by the BPP formula. If wall effects are still a problem at these low temperatures, such measurements will be extremely difficult, since it will be necessary to cool large quantities of He^3, preferably to temperatures below 0.3°K which is probably as low a temperature as can be reached with He^3 pumping.

Our data are clearly in disagreement with those of Low and Rorschach.[7] Although the discrepancy might originate in the different methods of measurement, new measurements,[22] made after those reported in reference 7, indicate that at least some of the short relaxation times observed by Low and Rorschach may be due to wall effects. Careri, Modena, and Santini[18] have reported T_1 measurements made at the saturated vapor pressure, in a magnetic field of 4316 gauss, using glass bulbs smaller than ours. Their results are in agreement with ours at 1.2°K, but their T_1 values are much shorter at higher temperatures. Their measurements were made by quite a different method than the ones we have used. Garwin and Reich[6] have reported T_1 measurements (see Fig. 1) made at pressures of 2.38 atmospheres and above, at 1600 gauss, in a 6.6-mm i.d. Araldite container with a 2.1-mm o.d. copper tube down the center. Unless there is a much stronger pressure dependence between the saturated vapor pressure and 2.38 atmospheres than is indicated by their higher pressure data, their results do not agree with ours.

ACKNOWLEDGMENTS

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[20] M. Bloom (private communication); I. Oppenheim and M. Bloom (to be published).

[21] K. N. Zinov'eva, J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 609 (1958) [translation: Soviet Phys. JETP **34**, (7), 421 (1958)].

[22] F. J. Low and H. E. Rorschach (private communication).