

Nuclear Spin Relaxation in Liquid He³†

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The nuclear spin thermal relaxation time, T_1 , of He³ nuclei has been measured between 1.2° and 3.0° in pure liquid He³ under its saturated vapor pressure. The measured times are those characteristic of the bulk liquid. T_1 is 300 seconds at 1.2° and increases to 550 seconds at 3.0°. These results are in fair agreement with the Bloembergen, Purcell, and Pound theory. For a 12% solution of He³ in He⁴ under its saturated vapor pressure, T_1 is about 2000 seconds and does not show any sharp change at the lambda point. In pure He³ gas at 4.2°K and 1000-mm pressure, T_1 is at least 1000 seconds. In dilute solutions at 1.25°K, T_1 is at least 90 minutes for a 3.5% solution and at least 120 minutes for a 1.7% solution.

THE nuclear spin thermal relaxation time, denoted by T_1 , has been measured in pure liquid He³ under its saturated vapor pressure, between 1.2° and 3.0°K. The measured relaxation times in this experiment appear to be approximately those characteristic of the bulk liquid; these measurements do not appear to have been significantly affected by relaxation occurring at the walls.

Early observations made in this laboratory^{1,2} on the nuclear resonance in liquid He³ were carried out with the primary goal of measuring the nuclear magnetic susceptibility. Attempts to measure simultaneously the value of T_1 were frustrated by the discovery that the measured T_1 depended markedly on the container used. The conclusion was drawn³ that in at least some of the containers the relaxation process was being artificially enhanced by processes occurring at the walls, and that

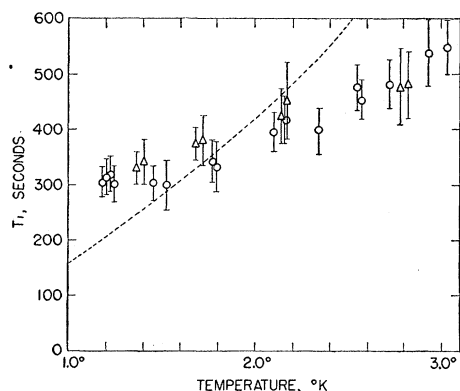


FIG. 1. Spin relaxation time, T_1 , in pure liquid He³ under its saturated vapor pressure. Circles represent measurements in small bulb (bulb A). Triangles represent measurements in large bulb (bulb B). Dashed line is the BPP expression, calculated from density and viscosity data.

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¹ Fairbank, Ard, Dehmelt, Gordy, and Williams, *Phys. Rev.* **92**, 208 (1953).

² Fairbank, Ard, and Walters, *Phys. Rev.* **95**, 566 (1954).

³ Fairbank, Ard, and Walters, *Conférence de Physique des Basses Températures, Paris, 1955* (Centre National de la Recherche Scientifique and UNESCO, Paris, 1956), p. 14.

the true T_1 , which would be measured in a very large sample, was greater than or equal to the longest observed time. It has been realized since the work of Bloembergen, Purcell, and Pound⁴ that T_1 measurements can give important information about atomic motions in liquids. Because of the great interest in the properties of liquid He³, the experiments reported here were begun with the specific goal of determining the value of T_1 characteristic of the bulk liquid.

One approach to the problem of measuring T_1 without complicating wall effects would be to use a container which is so large that in a time comparable to T_1 most atoms do not have time to diffuse to the walls, and thus will not be relaxed by the walls even if the walls can relax He³ atoms with 100% efficiency. However, the diffusion data of Garwin and Reich^{5,6} show that unless a sample volume greater than several hundred mm³ is used, most of the atoms will diffuse to the walls within a few hundred seconds, and it has previously been shown¹ that T_1 is at least 200 seconds at 1.2°. The alternative approach used here was to make T_1 measurements in containers of various geometries but made of the same material. In this case, if approximately the same T_1 is observed in all containers, then one can be reasonably confident that T_1 has not been artificially shortened by wall relaxation processes. Pyrex containers were used since the longest previously measured relaxation times¹ had been obtained in Pyrex bulbs.

In each run the sample, stored at room temperature, was passed through a purifying trap cooled by liquid helium and condensed into a bulb blown in the end of a long Pyrex capillary which extended down into the helium Dewar. The nuclear magnetic resonance absorption signals were observed in a field of about 9000 gauss. The observed line widths were in all cases determined by the inhomogeneity (approximately 0.1 gauss) of the external magnetic field. T_1 was measured by the "direct" method of saturating the nuclear spin system with a strong rf field, then decreasing the rf field strength to a value low enough to cause no appreciable

⁴ Bloembergen, Purcell, and 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.* (to be published).

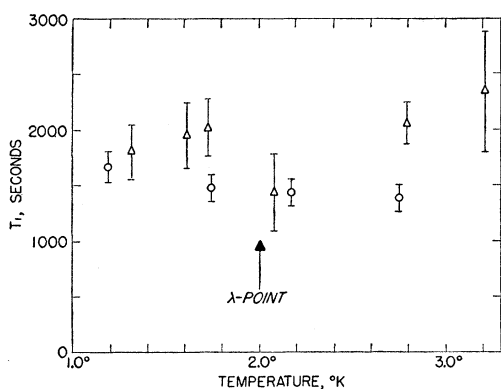


FIG. 2. Spin relaxation time, T_1 , in a 12.3% solution of He^3 in He^4 , liquid under its saturated vapor pressure. Circles represent measurements in small bulb (bulb A). Triangles represent measurements in large bulb (bulb B). The arrow indicates the λ point of the solution.

saturation, and then watching the amplitude of the signal recover toward its equilibrium value. The measured T_1 is the time constant characteristic of this exponential recovery.

Figure 1 shows the experimental results for pure He^3 in two different containers. Both containers were made of Pyrex as described. Bulb A was approximately spherical, with inside diameter approximately 5 mm. The measurements on bulb A were made with the bulb full of liquid. Bulb B was approximately spherical, with inside diameter approximately 10 mm. The measurements on bulb B were made with the bulb approximately $\frac{2}{3}$ full of liquid. Each point in Fig. 1 represents one observation of the recovery of the resonance signal. The error flags indicate roughly the uncertainty resulting from trying to fit exponentials with various time constants to the experimental plots of signal voltage vs time.

Figure 1 also shows the results of applying to He^3 the BPP theory. The expression used was

$$T_1 = 5kT / (6\pi^2 \gamma^4 \hbar^2 \eta N),$$

which is one form of the BPP expression for ordinary liquids,⁷ with the coefficient slightly modified according to the work of Kubo and Tomita⁸ and Torrey.^{9,10} Values of N , the number of atoms per cm^3 , were taken from the work of Kerr,¹¹ and values of η , the viscosity coefficient, from Zinov'eva.¹² The experimental results are clearly in good agreement with the BPP result, as far as the order of magnitude of T_1 and the sign of the temperature dependence are concerned.

⁷ E.g., E. R. Andrew, *Nuclear Magnetic Resonance* (Cambridge University Press, Cambridge, 1955), p. 117.

⁸ R. Kubo and K. Tomita, *J. Phys. Soc. Japan* **9**, 888 (1954).

⁹ H. C. Torrey, *Phys. Rev.* **92**, 962 (1953).

¹⁰ H. C. Torrey, *Suppl. Nuovo cimento* **9**, 95 (1958).

¹¹ E. C. Kerr, *Phys. Rev.* **96**, 551 (1954).

¹² K. N. Zinov'eva, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **34**, 609 (1958) [translation: *Soviet Phys. JETP* **34** (7), 421 (1958)].

T_1 measurements on pure liquid He^3 have also been made by Garwin, Kan, and Reich,^{6,13} and also by Careri, Modena, and Santini.¹⁴ The results obtained by both of these groups disagree with those reported here.

A preliminary measurement in He^3 gas at 4.2°K and 1000-mm pressure yielded a T_1 of about 1000 seconds. This measurement has not yet been repeated in a larger container to see if wall effects influenced the result. Bloembergen¹⁵ has given a theoretical expression for T_1 which should be applicable to a monatomic gas such as He^3 . For the stated experimental conditions, Bloembergen's formula yields $T_1 \cong 5000$ seconds, if the distance over which the interaction between two atoms is approximately constant is taken as 2.5 Å. The agreement with the experimental result is reasonably satisfactory, since the theoretical formula is sensitive to the value chosen for this distance.

Some measurements on the relaxation time of He^3 nuclei in He^3 - He^4 solutions have also been made, in addition to those previously reported¹⁶ which showed that in a 3.5 mole % solution, T_1 was at least 90 minutes at 1.25°K and in a 1.7% solution at least 120 minutes. These measurements on the 3.5% and 1.7% solutions were made in a Pyrex bulb of approximately 7-mm inside diameter, and have not been repeated in larger containers. Figure 2 gives results for a 12.3 mole % solution of He^3 as a function of temperature. Measurements were made in bulbs A and B , with the bulb full of solution in each case. If wall relaxation processes dominated, one would expect the markedly different geometries to give very different T_1 's. Hence, although the relaxation times measured in bulb B are consistently longer than those measured in A , the bulk relaxation times for this solution are probably not much longer than those shown. The striking feature of these results is the absence of any marked change as the temperature is changed through the λ point (approximately 2.0°). The diffusion coefficient of the He^3 nuclei, to which T_1 is closely related in the BPP theory, is known^{5,6} to change by several orders of magnitude as the temperature of a solution is changed through the λ point.

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¹³ Garwin, Kan, and Reich, *Proceedings of the National Science Foundation Conference on Low-Temperature Physics and Chemistry*, Baton Rouge, Louisiana, 1955 (unpublished), p. 11.

¹⁴ G. Careri (private communication); Careri, Modena, and Santini, *Nuovo cimento* **13**, 207 (1959).

¹⁵ N. Bloembergen, thesis, Leiden, 1948 (unpublished), p. 107.

¹⁶ R. H. Romer and W. M. Fairbank, *Bull. Am. Phys. Soc. Ser. II*, **4**, 150 (1959).