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### NUCLEAR SPIN RELAXATION IN SOLID He<sup>3</sup>†

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The liquid states of both helium isotopes have been of particular interest due to their peculiar macroscopic properties which have been attributed to quantum effects. In solid He<sup>3</sup> the nuclear susceptibility departs from Curie's law,<sup>1-3</sup> below 0.2°K, indicating that some exchange interaction must be present in addition to the direct dipole-dipole interaction between atoms. Thus solid He<sup>3</sup> is also found to exhibit some such peculiar properties. A further distinctive property of solid He<sup>3</sup> is the  $\alpha$ - $\beta$  phase transition discovered by Grilly and Mills.<sup>4</sup> This transition has no analog in solid He<sup>4</sup>, and its nature is not yet understood. Some relevant information contained in the original work of Grilly and Mills is that the volume change associated with the transition approaches zero at approximately 2°K. In the work to be described here, nuclear spin relaxation times were measured in solid He<sup>3</sup> such that they might yield additional information as to these properties.

Free-precession techniques were used to measure both the transverse and longitudinal relaxation times at 30.4 Mc/sec to an accuracy of roughly 10%. The measurements were made both as a function of pressure at constant temperature and as a function of temperature at constant volume. In order to allow pressure variation of the sample, a loop of resistance wire was placed inside of the capillary through which the He<sup>3</sup> entered the sample chamber. In this way, by passing a current pulse through the wire, the plug of solid He<sup>3</sup> which formed in the capillary could be melted momentarily and the solid in the sample chamber could come to pressure equilibrium with the vapor in the warmer part of the capillary.

Large discontinuities in both relaxation times were observed as the pressure was increased or decreased across the  $\alpha$ - $\beta$  phase boundary above approximately 2°K. Figure 1 shows the data at

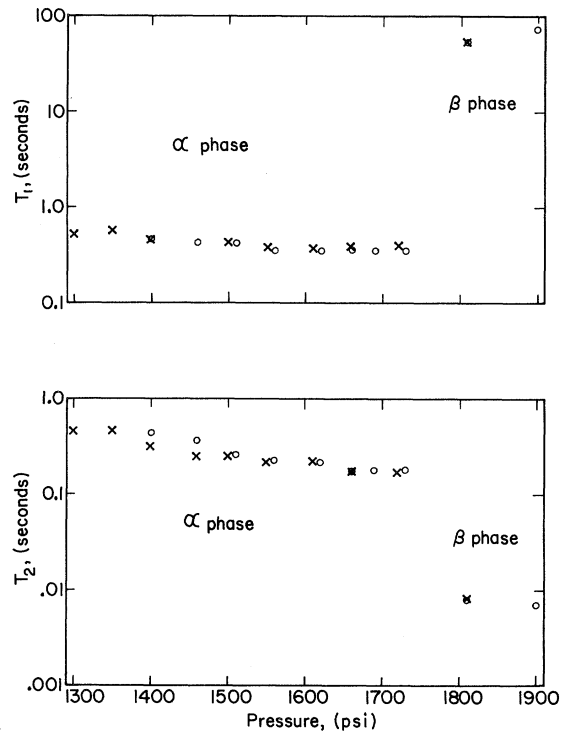


FIG. 1. Nuclear spin relaxation times in solid He<sup>3</sup> as a function of pressure at 2.2°K, indicating a discontinuity at the  $\alpha$ - $\beta$  phase boundary, discovered by Grilly and Mills (reference 4). Circles and crosses represent, respectively, data taken while increasing and decreasing the pressure.

2.2°K. The circles represent the data taken while increasing the pressure in steps and the crosses those taken while decreasing the pressure. At 1.37°K, however, the relaxation times displayed an exponential dependence on the pressure in the  $\alpha$  phase and no discontinuity at the phase boundary. This may be seen in Fig. 2, where it may also be seen that  $T_1$  passes through a minimum

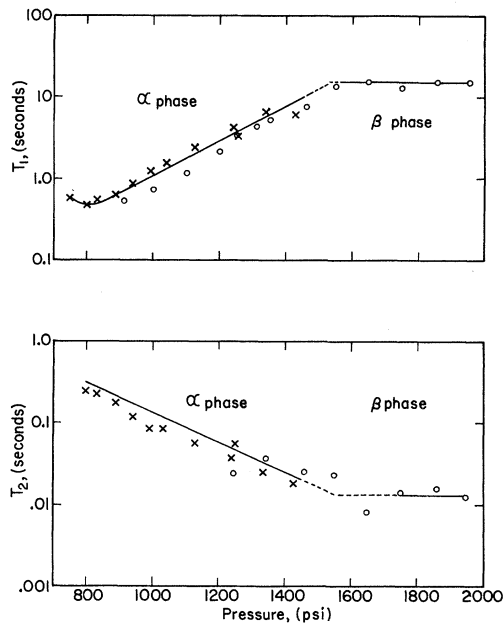


FIG. 2. Nuclear spin relaxation times as a function of pressure at 1.37°K, indicating a discontinuity in their rate of change with pressure at the  $\alpha$ - $\beta$  phase boundary. Circles and crosses represent, respectively, data taken while increasing and decreasing the pressure.

as the pressure changes near the liquid-solid phase boundary. Furthermore, there appears to be a discontinuity in the rate of change with pressure of  $T_1$  and  $T_2$ . The pressure was measured to the accuracy obtainable with an Ashcroft Bourdon gauge.

Results consistent with this picture have been obtained at both higher and lower temperatures. Measurements of  $T_1$  were made as low as 0.8°K, using a He<sup>3</sup> refrigerator, by both free-precession and steady-state methods. However, at this temperature  $T_1$  was 26 minutes at 1300 psi, and apparently still increasing exponentially with pressure. Consequently,  $T_1$  became so long as to make measurements at higher pressures difficult with the apparatus at hand. Since the discontinuity of the relaxation times vanishes near the temperature at which Grilly and Mills found the volume change approaching zero, a more detailed investigation of the temperature dependence of the effect is planned.

Several different constant-volume curves were measured. In general, in the temperature range investigated, the curves displayed the characteristics predicted by the semiclassical relaxation

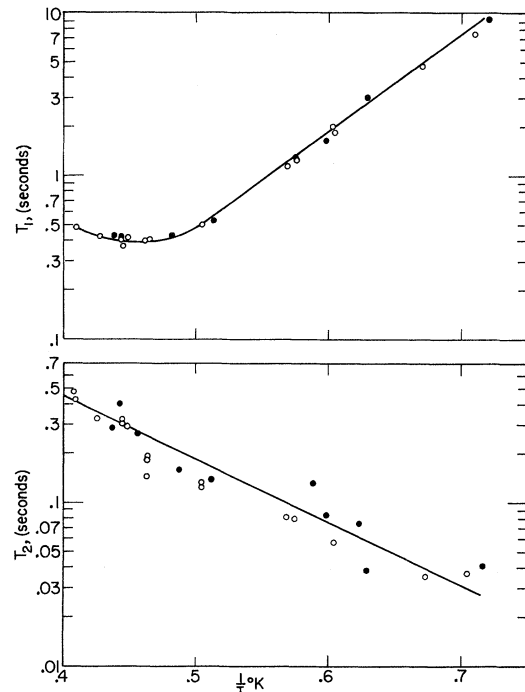


FIG. 3. Relaxation times as a function of the reciprocal of the temperature at a constant density of  $\rho = 0.144 \text{ g/cm}^3$ , indicating an exponential dependence at temperatures here represented. Open circles and dots represent data taken on two different runs.

theories<sup>5,6</sup> (see Fig. 3).  $T_1$  passes through a minimum as the temperature decreases and approaches an exponential dependence on  $1/T$  near  $T = 1.5^\circ\text{K}$ .  $T_2$  displays an exponential dependence on  $1/T$ . This information is interpreted as an indication that in these temperature regions the relaxation is due primarily to an activated diffusion.<sup>7</sup> Thus  $T_1 \propto 1/D$  and  $T_2 \propto D$ , where the diffusion coefficient  $D$  is of the form  $D = D_0 \exp(-E/kT)$ . Consequently, plotting the logarithms of  $T_1$  and  $T_2$  against the reciprocal of the temperature, the slope of the straight line part of the plot should be a measure of the activation energy  $E$ .

The energies determined from the  $T_1$  data in this way were 12.3°, 14°, and 12.5°K for densities of 0.151, 0.144, and 0.142 g/cm<sup>3</sup>. The  $T_2$  values were consistently 3°K lower, for reasons which are not at present understood. The densities given above were estimated from the information given by Grilly and Mills and the known temperatures and pressures at which the constant-volume curves were started.

With a different apparatus some preliminary results have been obtained down to 0.15°K. A

single measurement of  $T_1$  on the same constant-volume curve as that of Fig. 3 was about 40 minutes at 0.1°K. This is shorter by about 50 orders of magnitude than it would have been if it had continued to follow the exponential curve  $T_1 = 3 \times 10^{-4} \exp(14/T)$  of Fig. 3. One complete set of  $T_1$  measurements has been obtained between 0.7 and 0.14°K at a relatively low density (starting temperature and pressure 0.8°K and 500 psi). In this case  $T_1$  was about 20% higher at 0.14°K and 0.7°K than at intermediate temperatures but seemed roughly constant at  $0.55 \pm 0.05$  second between these limits.

In summary, the following information about solid He<sup>3</sup> is obtained from the results: (1) In the cases where there is a volume change associated with the  $\alpha$ - $\beta$  transition there are also large changes in the relaxation times. Since the relaxation in the  $\alpha$  phase in these cases is primarily due to diffusion, there must also be changes in the diffusion coefficient across the phase boundary. (2) In the cases where the volume change approaches zero, there are no changes in the relaxation time across the boundary and, consequently, if there are changes in the diffusion coefficients, there must be compensating changes in other relaxation mechanisms. Even in these cases there remains a discontinuity in the rate of change with pressure of the relaxation times at the phase boundary. (3) For temperatures above 1.37°K the  $\alpha$  phase behaves as an ordinary solid with a large diffusion coefficient.

That is, the relaxation is determined by an activated diffusion of the atoms through the lattice. However, at temperatures somewhat below 1°K the relaxation is much too fast to be explained by the classical diffusive motion of the Bloembergen, Purcell, and Pound theory.

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### SPECIFIC HEAT OF INDIUM BELOW 1°K\*

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Heat capacity measurements on two indium specimens indicate that the  $T^3$  term in the specific heat is smaller in the superconducting than in the normal state, contrary to the usual assumption that it is the same in both states. The expected temperature dependence in the normal state is

$$C_n = C_q + \gamma T + \alpha_n T^3 + \beta_n T^5, \quad (1)$$

and in the superconducting state,

$$C_s = C_q + a\gamma T_c \exp(-bT_c/T) + \alpha_s T^3 + \beta_s T^5, \quad (2)$$

$$T < 0.7T_c$$

In each case, the first term,  $C_q$ , is the contribution from the nuclei due to an electric quadrupole interaction similar to that found in rhenium.<sup>1</sup>

From the microwave resonance measurements of Hewitt and Knight<sup>2</sup> it follows that  $C_q = 9.0 \times 10^{-4}$  millijoule/mole deg, which is 11% of  $C_s$  at 0.35°K, the lowest temperature reached in the measurement, and decreases to less than 1% of  $C_s$  at 0.6°K. The second term in  $C_s$  is the electronic specific heat,  $C_{es}$ , of a superconductor at temperatures below  $0.7T_c$ , and is less than 1% of  $C_s$  below half a degree. In this term,  $a$  and  $b$  are quasi-universal constants, and  $T_c$  is the transition temperature of indium (3.4°K). A