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NUCLEAR SPIN RELAXATION IN SOLID He³[†]

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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 He³ the nuclear susceptibility departs from Curie's law,[1-3] below $0.2°K$, indicating that some exchange interaction must be present in addition to the direct dipole-dipole interaction between atoms. Thus solid He³ is also found to exhibit some such peculiar properties. A further distinctive property of solid He³ is the α-β phase transition discovered by Grilly and Mills.[4] This transition has no analog in solid He⁴, 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 He³ 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 He³ entered the sample chamber. In this way, by passing a current pulse through the wire, the plug of solid He³ 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 α-β phase boundary above approximately $2°K$. Figure 1 shows the data at
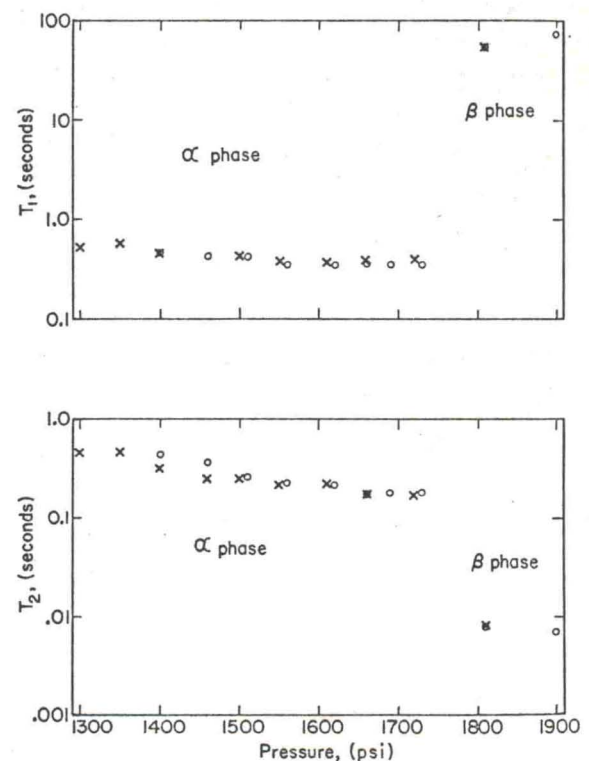

FIG. 1. Nuclear spin relaxation times in solid He³ as a function of pressure at $2.2°K$, indicating a discontinuity at the α-β phase boundary, discovered by Grilly and Mills (reference 4). Circles and crosses represent, respectively, data taken while increasing and decreasing the pressure.

$2.2°K$. 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 α 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
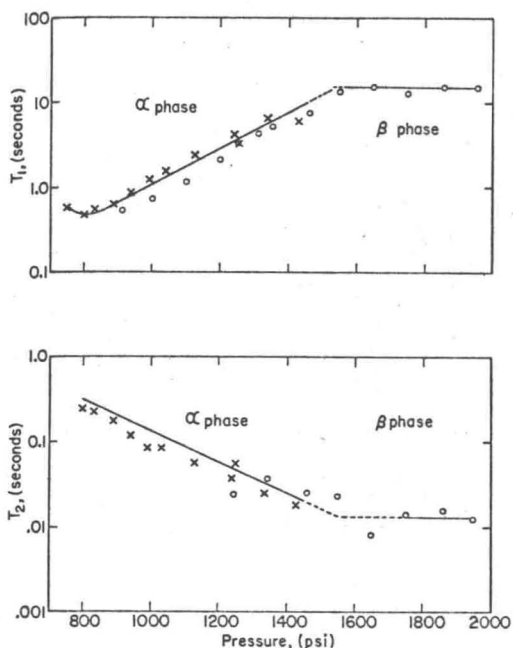
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FIG. 2. Nuclear spin relaxation times as a function
of pressure at 1.37°K, indicating a discontinuity in
their rate of change with pressure at the α-β phase
boundary. Circles and crosses represent, respective-
ly, data taken while increasing and decreasing the
pressure.

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

as the pressure changes near the liquid-solid
phase boundary. Furthermore, there appears to
be a discontinuity in the rate of change with pres-
sure 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^3 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 dif-
ficult with the apparatus at hand. Since the dis-
continuity 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 depend-
ence of the effect is planned.

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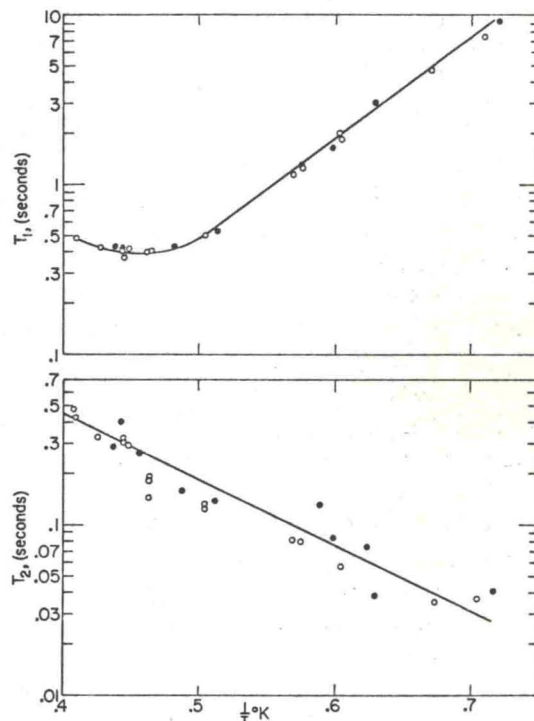istics predicted by the semiclassical relaxation

theories[5,6] (see Fig. 3). T_1 passes through a
minimum as the temperature decreases and ap-
proaches an exponential dependence on $1/T$ near
$T = 1.5$°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 dif-
fusion.[7] Thus $T_1 \propto 1/D$ and $T_2 \propto D$, where the dif-
fusion 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 densi-
ties of 0.151, 0.144, and 0.142 g/cm^3. The T_2
values were consistently 3°K lower, for reasons
which are not at present understood. The densi-
ties given above were estimated from the in-
formation 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 ± 0.05 second between these limits.

In summary, the following information about solid He³ is obtained from the results: (1) In the cases where there is a volume change associated with the α-β transition there are also large changes in the relaxation times. Since the relaxation in the α 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 α 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

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

and in the superconducting state,

$$C_s = C_q + a\gamma T_c \exp(-bT_c/T) + \alpha_s T^3 + \beta_s T^5,$$
$$T < 0.7T_c. \qquad (2)$$

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.[1] From the microwave resonance measurements of Hewitt and Knight[2] 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