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New Magnetic Phenomena in Liquid He^3 below 3 mK*

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Magnetic measurements have been made on a sample of He^3 in a Pomeranchuk cell. Below about 2.7 mK, the NMR line apparently associated with the liquid portion of the sample shifts continuously to higher frequencies during cooling. Below about 2 mK the frequency shift vanishes, and the magnitude of the liquid absorption drops abruptly to approximately $\frac{1}{2}$ its previous value. These measurements are related to the pressure phenomena reported by Osheroff, Richardson, and Lee.

Pressure measurements along the melting curve of He^3 in compressional cooling experiments have indicated the possible existence of two phase changes occurring in the He^3 within the compression cell.[1] Osheroff, Richardson, and Lee referred to these pressure phenomena as A and B. A, believed to occur at about 2.7 mK, is characterized by an abrupt decrease in the rate of cooling in the cell during a period of time in which the rate of compression is held constant. The pressure at which A occurs, $P(A)$, is highly reproducible and does not display supercooling. B, occurring at a lower temperature, perhaps 2 mK, is characterized by a sudden drop in the cell pressure by a few ten thousandths of an atmosphere upon cooling, and by a brief hesitation in the pressure as it decreases upon warming, at $P(B')$. The pressure $P(B')$ is highly reproducible, and the B' phenomenon will not occur unless the B phenomenon has already occurred. The B effect, however, shows a great deal of supercooling (as much as 10^{-2} atm), depending upon how far below $P(B')$ the cell pressure has been lowered since last going through B'. The smaller this pressure difference, the smaller will be the degree of supercooling. Although the magnetic field dependence of $P(A)$ is small and comparable in sense and magnitude to the expected depression of the melting curve itself at 2.7 mK in magnetic fields, the pressure at which B' occurs increases sharply with increasing magnetic field, and the field dependence of the pressure difference $P(B') - P(A)$ can be represented by $[P(B') - P(A)]_{H=H_0} - [P(B') - P(A)]_{H=0} = +2.02$

$\times 10^{-3} H_0^2$ atm/(kG)^2. This clearly shows that the temperature of the B transition is strongly depressed by the application of a magnetic field.

We have subsequently carried out an extensive continuous-wave nuclear magnetic resonance survey of both the liquid and solid phases existing within the cell during compressions to the lowest temperatures, and have found unexpected behavior occurring within the liquid phase.

The cell in which these measurements were performed is identical to the cell described in Ref. 1 except that the platinum thermometer assembly was replaced by a specially constructed NMR coil and support plug. The coil form was 2.54 cm long and 0.48 cm in diameter. It was oriented vertically, and extended to within 0.5 cm of the BeCu strain-gauge diaphram. The bore of the NMR coil form was open at both ends. The upper portion of the coil was surrounded by a 1.9-cm-diam epoxy support plug. A nearly linear gradient in the applied external field H_0 could be created perpendicular to the external field, and parallel to the coil axis. In the gradient, only a thin layer of sample could satisfy the Larmor resonance condition at a given radio frequency. By sweeping the radio frequency, or equivalently H_0, the resonant layer could be moved up or down the length of the coil, providing a means to obtain a profile of the resonant absorption of the sample as a function of distance along the coil length.

In Fig. 1 is shown a series of idealized profiles obtained in the gradient field. The first profile was obtained prior to compression and is due to all liquid in the region of the coil. The irregularities in the all-liquid profile are due to variations in the field gradient and in the effective volume of helium seen by the coil at various distances along its length.

Fortunately, solid tended to nucleate at only one or two points within the region of the coil, and with sufficient care all further solid deposited within the coil could be caused to form at these places. (Presumably solid also formed outside the coil.) This allowed the separate observation of both solid and liquid signals in the single coil throughout the compression. The Fermi degenerate liquid He^3 susceptibility is nearly temperature independent in the low-temperature limit, and is much smaller than the susceptibility of an equivalent volume of solid He^3. In profiles b and c of Fig. 1 the solid signal can be seen to grow rapidly out of the liquid background, so that by frames d, e, and f the solid peaks are between 30 and 60 times the liquid signal, and are simply truncated on the figure.

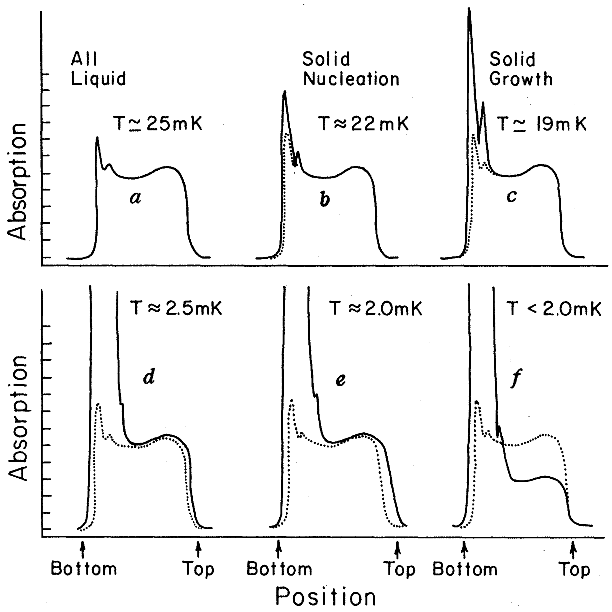

FIG. 1. Profiles of NMR absorption as a function of position along the NMR coil length at various stages during compression. Extrapolated melting-curve temperatures below 3 mK are only approximate. Dotted line, initial all-liquid profile.

No changes in the magnetic behavior of the solid were observed which seemed correlated with either the A or B transition. This might be a consequence of the long thermal relaxation times associated with the solid. In contrast, the liquid did display marked changes in its magnetic behavior at the A and B transitions. Profiles d–f of Fig. 1 show the liquid at successively higher pressures above $P(A)$, with the final frame having been recorded subsequent to the B transition. The initial liquid absorption profile is superimposed on all frames for reference. Notice that as the pressure increases from $P(A)$ to $P(B)$, the edge of the liquid signal seem to shift to the right, which corresponds to a shift to successively higher frequencies. Finally, at $P(B)$, the liquid profile suddenly shifts back to its original position and its magnitude drops nearly in half. These two changes in the behavior of the liquid susceptibility have been studied in detail.

To study more clearly the possibility that the resonant frequency of the liquid He^3 shifted as the cell pressure increased above $P(A)$, the gradient field was removed. In this arrangement, what was previously a position profile became a frequency profile. Figure 2 shows a series of

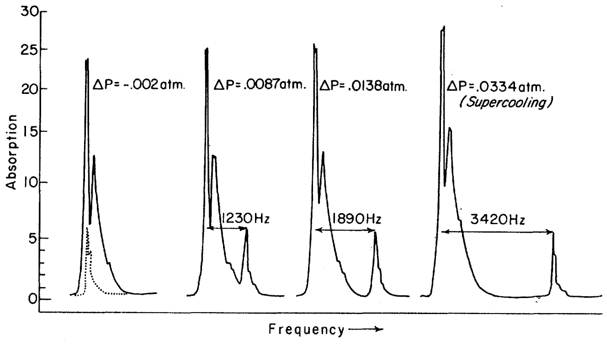

FIG. 2. Frequency profiles obtained at various pressures above $P(A)$ as indicated by $\Delta P = P(t) - P(A)$, showing the frequency splitting as measured in a 466-G magnetic field. Dotted line, the initial all-liquid profile.

frequency profiles obtained in a 466-G magnetic field at pressures above $P(A)$. Notice the nonlinear vertical scale. This allowed the simultaneous observation of the solid peaks and a smaller shifted signal. Superimposed on the first profile is a dotted curve indicating the all-liquid profile obtained prior to compression. For convenience, we have labeled the pressure for each profile using the notation $\Delta P(t) = P(t) - P(A)$. Notice that as ΔP increases in Fig. 2, a satellite peak splits off the main absorption line and moves to higher frequencies. Finally, once the cell pressure reaches $P(B)$, the satellite disappears. If the cell pressure were dropped below $P(B')$, however, the satellite would reappear.

There is strong evidence to suggest that the shifted line is, in fact, the liquid contribution to the NMR signal, and to suggest that the shift is a unique property of the liquid pressure and temperature and of the applied magnetic field H_0. The main evidences supporting this hypothesis are as follows: (a) The frequency shift is a unique function of cell pressure, and totally independent of the solid fraction within the chamber. (b) The magnitude of the shifted line is nearly constant, decreasing slightly as the solid fraction in the coil increases. (c) The shape of the shifted line closely resembles the shape of the initial all-liquid line, even when this shape is rather complex. (d) The line shape of the shifted signal does not distort or broaden even when the ratio of the splitting to the linewidth is as high as 15:1. (e) The nuclear relaxation time of the shifted peak is about 2 sec, nearly a factor of 10 longer than the relaxation time in the solid.

The resonant frequency and pressure (temperature) dependence of the splitting were studied in magnetic fields from 31 G to over 850 G. In all cases where the splitting could be measured absolutely, the frequency shift extrapolated to zero at $P(A)$. At each of several Larmor frequencies studied, a plot of $\Delta\nu$ versus ΔP was obtained. When the resulting plots were adjusted for all to extrapolate through the origin, all the data could be fitted to within ± 2% by a single function,

$$\nu_{\mathrm{liq}}^2 - \nu_{\mathrm{sol}}^2 = (4.956\times10^{11}\,\mathrm{Hz}^2/\mathrm{atm})\Delta P - (5.947\times10^{12}\,\mathrm{Hz}^2/\mathrm{atm}^2)(\Delta P)^2.$$

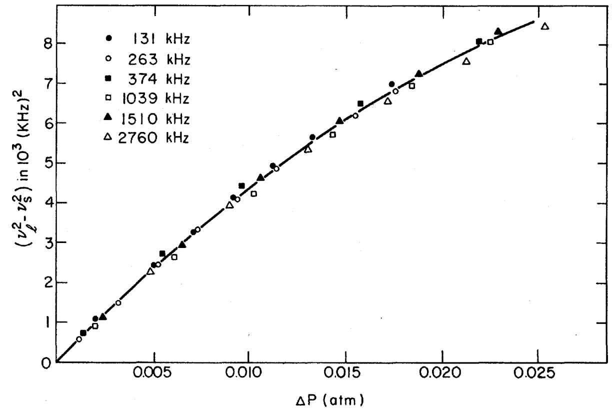

FIG. 3. Differences in the squares of the liquid and solid NMR resonant frequencies, adjusted as described in the text, as a function of ΔP. The solid curve corresponds to the best fit to all the data.

In Fig. 3 are shown the individual curves of $\nu_l^2 - \nu_s^2$ obtained at six different frequencies. The satellite line could not be seen in applied fields below 31 G, independent of the splitting and the resonant frequency. Because of the poor quality of the signals at the lowest frequencies, this result must be considered somewhat tentative.

In the gradient experiments it was found that the liquid susceptibility dropped to about one half its Fermi degenerate value precisely when the drop in cell pressure occurred at B, and returned to its original value precisely at B'. These pressure phenomena were also coincident with the disappearance and reappearance of the satellite line seen in the frequency profile experiments. The drop in susceptibility appeared to vary from about 40 to 60%, although as a result of the shift associated with the liquid profile below $P(B')$, this was difficult to measure accurately. No further changes in the liquid signal were seen at still higher pressures. Occasionally, a small region of liquid seemed to drop nearly to zero susceptibility at the B transition. The resulting profile in these cases seemed stable provided the cell pressure remained above $P(B')$.

Transient phenomena associated with the liquid behavior were observed by sweeping the radio frequency 20 times/sec and displaying the resulting position profile on an oscilloscope. Briefly, it was observed that (1) the drop in susceptibility at B always proceeded from the bottom of the cell upward, and the increase at B' proceeded downward. The rate at which this process occurred seemed proportional to the rate of compression. (2) The boundary between the two liquid phases was manifested by a wave with a trough and a peak. The shape of this wave is most likely explicable in terms of the frequency shift associated with the liquid susceptibility between A and B. (3) Between A and B, the liquid responded to the application into a wire-wound heater located at the bottom of the NMR coil of a 25–100-erg heat pulse in the form of a small wave traveling from the top of the coil downward, and disappearing about halfway through the coil. In the frequency profile experiments (as in Fig. 2) a similar heat pulse distorted the satellite line shape slightly. Both phenomena occurred in less than 0.5 sec. (4) At pressures above $P(B)$, a 25–100-erg heat pulse caused an immediate drop in the already lowered liquid susceptibility to nearly zero, and relaxation to the value of $\frac{1}{2}$ the Fermi degenerate level was manifested as a sharp wave front traveling upward from the bottom of the cell at a velocity of perhaps 1 cm/sec. Occasionally at the end of this process a small portion of the liquid profile would remain with zero susceptibility. This region, though stable in time, could be moved upward in the coil by the application of high levels of resonant rf power. (5) By the separate application of high levels of resonant rf power to the solid and satellite lines in the frequency profile, the nuclear relaxation time for the solid and satellite, respectively, were found to be about 0.2 and 2 sec. It was not possible to make a saturation measurement above $P(B)$ because the large amounts of rf power required simultaneously generated a heating pulse.

One interesting result concerning the solid was obtained in these experiments. By comparing the maximum solid peak heights observed in the gradient experiments to the corresponding liquid susceptibilities, we were able to estimate effective magnetic temperatures of the solid producing these peaks.[2] If the He^3 in the resonant layers producing these peaks was not all solid, or was not uniformly at the lowest temperatures, then these estimates of magnetic temperatures will actually be higher than the true magnetic temperatures to be associated with the coldest solid. Even so, our largest solid peaks had equivalent magnetic temperatures as low as 3.2 mK. Previous investigations down to 5 mK have shown that solid He^3 obeys the Curie-Weiss relation $T^* = T + \theta_{CW} = T + 3.2$ mK.[3] A simple extrapolation based on this relation is incompatible with our measured solid magnetic temperatures under any reasonable estimate of the actual temperature associated with that solid. This result suggests that below 5 mK the magnetic properties of solid He^3 deviate from their expected behavior. Additional indication of this is provided by recently published spin diffusion measurements[4] which show a dramatic *decrease* in the solid spin diffusion coefficient at the lowest temperatures.

It is a pleasure to thank Professor John D. Reppy for many valuable suggestions throughout the course of this work. We are also grateful to Professor John Goodkind and Dr. V. L. Vvedensky who pointed out the possibility that the anomalous features in the pressurization curve of Ref. 1 might be associated with phase changes in liquid He.[3] Finally, we wish to express our gratitude to Professor V. Ambegaokar, Professor N. W. Ashcroft, Professor M. E. Fisher, Dr. R. Giffard, Professor N. D. Mermin, Professor J. R. Schrieffer, Professor R. H. Silsbee, and Professor J. W. Wilkins for many interesting and informative discussions regarding the meaning of these results.

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