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Observation of a Second-Order Phase Transition and Its Associated P-T* Phase Diagram in Liquid He³†

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A specific-heat "discontinuity" is observed in liquid He³ from 241 lb/in.² to the melting pressure in the temperature range 2–3 mK.

In measurements along the melting curve of He³, using an adiabatic compressional cooling device, Osheroff, Richardson, and Lee[1] discovered two features on the He³ pressure versus *time* curve at temperatures below 3 mK. These features were a sudden change of slope, feature A, and a sudden drop in pressure during compression, feature B. Feature A was originally thought to reflect a first-order transition[1] or a λ-type specific-heat anomaly[2] in solid He³. Later experiments on magnetic properties by Osheroff *et al.*[3] strongly suggested the interpretation of both A and B as liquid-He³ effects. This conclusion was somewhat obscured by the observation by Halperin *et al.*[4] of a maximum near A in the thermal time constant in a compressional cell containing 95% *solid*. Further support that A and B are *liquid* effects was given theoretically by Leggett[5] and by Anderson and Varma.[6] Also, measurements of the equilibrium melting curve by Johnson *et al.*[7] showed that the A feature does *not* reflect a solid transition. In the present work we measure the *liquid* specific heat and find a second-order phase transition, a discon-

tinuity but no divergence in the specific heat as a function of temperature, which occurs over a wide range of liquid-He³ pressure. The transition was not observable in our apparatus for a He³ pressure of 1.9 lb/in.² (1 atm = 14.696 lb/in.²).

The He³ specific-heat cell is of standard design[8] containing a mixture of 10.78 g powdered cerous magnesium nitrate (CMN) and 1.23 cm³ of liquid He³, where the CMN acts both as refrigerant and thermometer. The CMN was packed to a higher density than usual, with possibly some inhomogeneity. The magnetic temperature T* was calibrated in the range 0.34 to 4.2 K with an accuracy better than 1% using a germanium resistance thermometer. The heater was made of a short length of 0.05-mm-diam Evanohm wire and located well within the cell. A capacitive pressure gauge thermally attached to the mixing chamber of our precooling dilution refrigerator served to measure cell pressure at all pressures except those in which there was some solid in the cell.

Two types of experimental measurements were made, both in fields of less than 1 G. In one,

after an adequate foreinterval, an accurately measured amount of heat Q is introduced. Then, using an adequate afterinterval, the temperature increment ΔT^* caused by Q is determined. The resulting heat capacity on the magnetic scale is $C^* = Q/\Delta T^*$. Equilibrium time constants were less than 100–200 sec except at higher temperatures. A typical fractional magnetic temperature rise for a single specific-heat point near $T_c{}^*$ was 2%, though both this figure and the time and rate of heating were varied substantially. Because of the sharpness of the transition, both the actual value of the transition temperature and the specific-heat jump were usually obtained using a second type of measurement in which the cell contents were allowed to drift through the transition under the action of the residual heat leak while both T^* and $\dot{T}^* = dT^*/dt$ were measured. No evidence via either C^* or $\dot{T}^*$ was obtained for a transition at a low pressure of 1.9 lb/in.^{2}. Since the heat capacity of the CMN dominates the He^3 heat capacity in the temperature region involved, we display our C^* data by plotting the differential heat capacity $\Delta C^* \equiv C^*(P) - C^*(1.9$ lb/in.$^2)$. We then assume that in the temperature region for which the differences ΔC^* are precise, the low-pressure liquid-He^3 heat capacity

is known so that a total He^3 heat capacity can be computed. Smoothed values of $C^*(1.9$ lb/in.$^2)$ at $T^* = 2.2$, 2.1, 2.0, 1.9, 1.8, and 1.7 mK are respectively, $C^*(1.9$ lb/in.$^2) = 380.6$, 398.8, 425.4, 470.4, 548.8, and 700.0 erg/mK. The raw high-pressure data may be obtained from these figures.

An example of experimental data for a pressure of 484.5 lb/in.^{2}, 13.7 lb/in.^{2} below the melting pressure,[7] is shown in Fig. 1. For $T^* < 1.6$ mK the measured total heat capacity is in excess of 10^3 erg/mK, so that the differential heat capacity plotted is imprecise. Precision in the differential heat capacity increases with increasing temperature and is characterized by the experimental scatter. The rapid transition from one total heat capacity (He^3 plus CMN) is another is indicated on the inset in Fig. 1 which shows the temperature drift rate as a function of magnetic temperature (on an expanded scale) in the immediate vicinity of the transition. It is evident from the inset that the specific heat does not diverge in the vicinity of $T_c{}^*$. The half-width of the transition in this case is about 0.3% of $T_c{}^*$. This width is representative of all our data, though the width does increase as the residual heat leak increases.
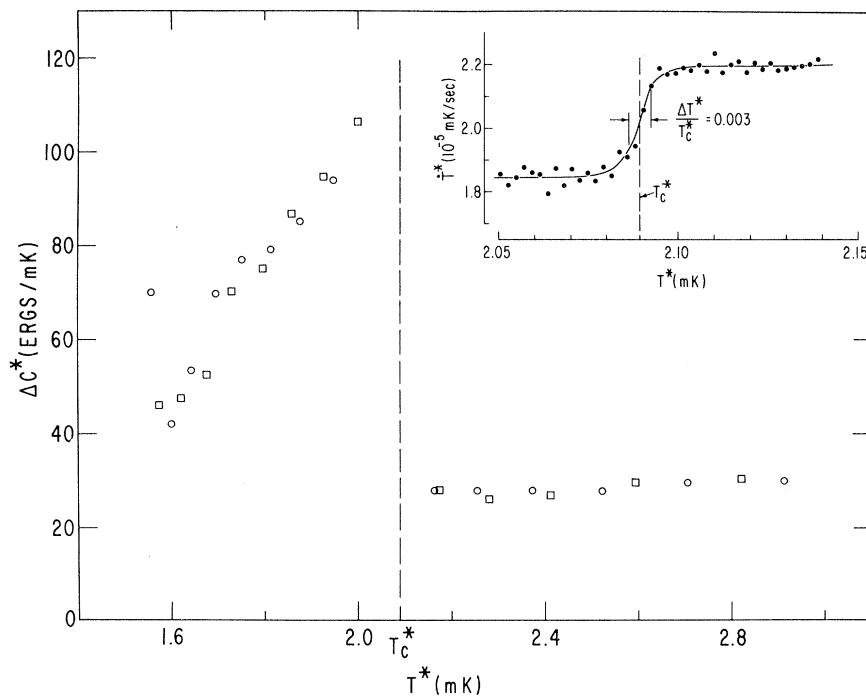
Measurements similar to those shown in Fig.

FIG. 1. Experimental difference between the total cell heat capacity for a liquid-He^3 pressure of 484.5 lb/in.^{2} and that for a pressure of 1.9 lb/in.^{2} on a magnetic temperature scale T^*. The inset shows on an expanded magnetic scale the time rate of change T^* in the vicinity of the specific-heat discontinuity.

1 were made over a range of pressures on the liquid, up to and including the melting pressure. Transition temperatures T_c^* were obtained from drift data like those shown in the inset to Fig. 1. These are shown as the curve labeled T_c^* in Fig. 2. At 241 lb/in.[2] the transition is nearly obscured by noise on the $\dot{T}^*$ plot.

The greatest uncertainty in the present measurements is in the temperature scale. Measurements in Ref. 7 of the equilibrium temperature of the pressure feature[1] A gave $T_A^* = 2.35$ mK on a CMN scale and a best estimate of 2.6 ± 0.1 mK for its absolute temperature. The T_c^* curve of Fig. 2 intersects the melting curve at a magnetic temperature of 2.10 mK, not 2.35 mK, so we cannot unambiguously assert that the second-order transition we observe is responsible for the A feature on the pressurization curve. But since A has been shown[7] not to be a solid effect and since we see no other anomalous thermal effects in the liquid at higher temperatures, we assume that our magnetic scale is distorted from that of Ref. 7, probably as a result of a higher packing density and greater inhomogeneity. Because of this distortion we are not free to use the noise temperature data of Webb, Giffard, and

Wheatley[9] to convert T^* to T. However, we can obtain an approximate T scale by using the measured ΔC^* and T_c and by making the following assumptions: (1) A magnetic temperature of 2.10 mK is 2.60 mK absolute; (2) for $T > T_c$ the difference heat capacity is linear in T with a proportionality constant equal, at a given pressure, to the value expected from higher-T measurements[10,11]; and (3) for $T^* < T_c^*$ the curve $\Delta C^*(T^*)$ for $T^* > T_c^*$ may be extrapolated to lower T^* by 10 or 15% of T_c^* without introducing serious error. We carried out this program for the data shown in Fig. 1. This $T^* - T$ relationship ($T^* > 1.9$ mK) is the same as that between another T^* and the Johnson noise temperature T (corrected for noise thermometer heating) determined in Ref. 9 if one adds 0.3 mK to our currently measured T^* values. For $T^* < 1.9$ mK the amount to be added to T^* to get agreement with Ref. 9 falls below 0.3 mK, and so the scale becomes more qualitative. This scale was then used to recompute the heat capacity data of Fig. 1 from about 0.3 mK below T_c^* to higher temperatures. The results are shown as a total molar heat capacity in Fig. 3, the calculated He[3] heat capacity at 1.9 lb/in.[2] being added to the mea-
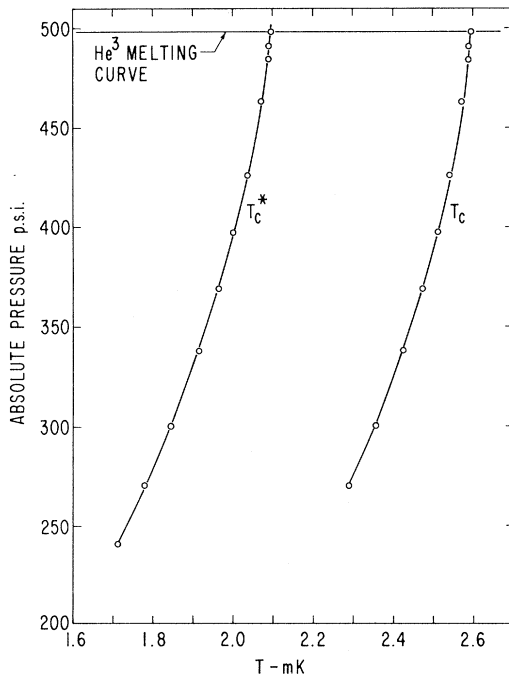

FIG. 2. Temperature of the specific-heat discontinuity as function of pressure for both a magnetic temperature scale T^* and a temperature scale T discussed in the text which is our best estimate of the absolute temperature.
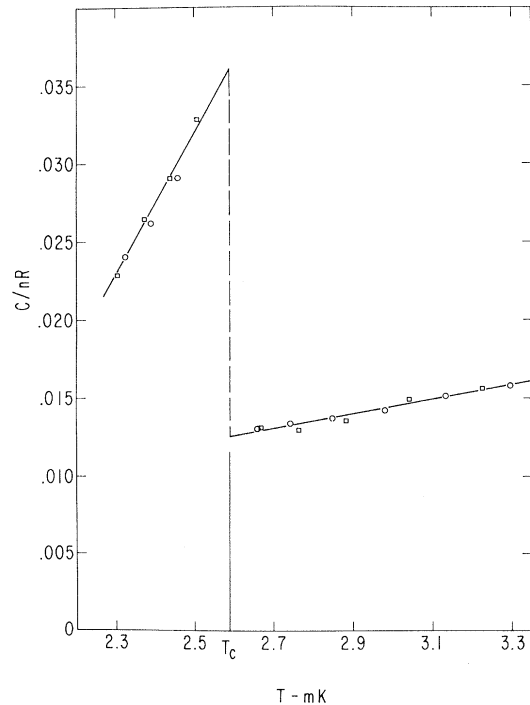

FIG. 3. Total molar heat capacity as a function of temperature of liquid He[3] at a pressure of 484.5 lb/in.[2] as deduced from the measurements and a T^*-T relation discussed in the text.

sured difference heat capacity at 484.5 lb/in.^{2}. The precision is represented by scatter, but the accuracy is limited by the uncertainties of the absolute temperature scale as indicated above. Above T_c the data fit a straight line passing through $T = 0$. If the data below T_c are fitted arbitrarily by a power law, then $C/nR = 2.9\gamma T_c(T/T_c)^4$ fits satisfactorily, γT_c being the molar heat capacity just above T_c. The data are fit equally well by $C/nR = 2.9\gamma T_c \exp\{-3.6[(T_c/T)-1]\}$. We wish to emphasize that both the representation of the data in Fig. 3 and the above fits must be used with great caution, owing to the uncertainty in the temperature scale. We present the data in this form because we believe that they represent our best, though imperfect, knowledge of the heat capacity of He^3 on an absolute scale in the vicinity of the transition.

The T-T^* scale derived as indicated above has been used to give our best estimate of the P-T_c phase diagram. This is shown in Fig. 2 as the curve labeled T_c.

The factor by which the He^3 heat capacity changes at the transition may be computed from the experimental data without assumptions about the temperature scale. In what follows let $\Delta C_<^*$ and $\Delta C_>^*$ be the differential heat capacities just below and just above T_c^*, let $\dot{T}_<^*$ and $\dot{T}_>^*$ be the drift rates just below and just above T_c^*, and let $C_>^*$ be the total (He^3 + CMN) heat capacity just above T_c^*. The magnitude of the specific heat jump was obtained using the $\dot{T}^*$ measurements from the relation $\Delta C_<^* - \Delta C_>^* = [(\dot{T}_>^*/\dot{T}_<^*) - 1]C_>^*$. We then computed the total He^3 heat capacity on the magnetic scale just above T_c^* from $\Delta C_>^*$ by assuming that the ratio of differential He^3 heat capacity to total He^3 heat capacity at P on the magnetic scale is the same as that which can be computed from experimental knowledge[10,11] of He^3 heat capacities at, say, $T = 5$ mK. This assumption takes the heat capacity of liquid He^3 as being linear in T below 5 mK with a coefficient which is a known function of pressure. The ratio of the total He^3 specific heat on the magnetic scale just below to just above T_c^* was then found to vary from near 2.4_5 at 338 lb/in.^{2} to near 2.6_5 at 491 lb/in.^{2}. This trend may reflect some systematic error in the measurements rather than a real pressure dependence. Using the specific-heat data of Fig. 3 we find 2.9 for this ratio, which is somewhat more than that obtained using the drift data. This discrepancy may partly re-

flect an incorrect extrapolation to T_c of the heat-capacity data. The specific-heat ratio can be compared with the slope ratio characteristic of the pressure feature A as given in Ref. 1, 1.8, and by the authors of Ref. 7, 2.1-2.3. The pressure feature A can be understood qualitatively, as pointed out by Leggett,[5] if there is a specific-heat discontinuity in the liquid at A. If the rate of conversion of liquid to solid in a compressional cell is constant and if there are no volume effects,[12] then, since dP_3/dT does not change at A,[7] in absence of external heat input the time rate of change of He^3 pressure $\dot{P}_3$ in inversely proportional to the total cell heat capacity. A discontinuity in liquid heat capacity then is reflected as a discontinuity in $\dot{P}_3$, but this discontinuity will be less than the liquid specific-heat discontinuity as a result of the contribution of the solid-He^3 heat capacity to the nonequilibrium measurement.

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