Measurements along the Melting Curve of He^3 at Millikelvin Temperatures*

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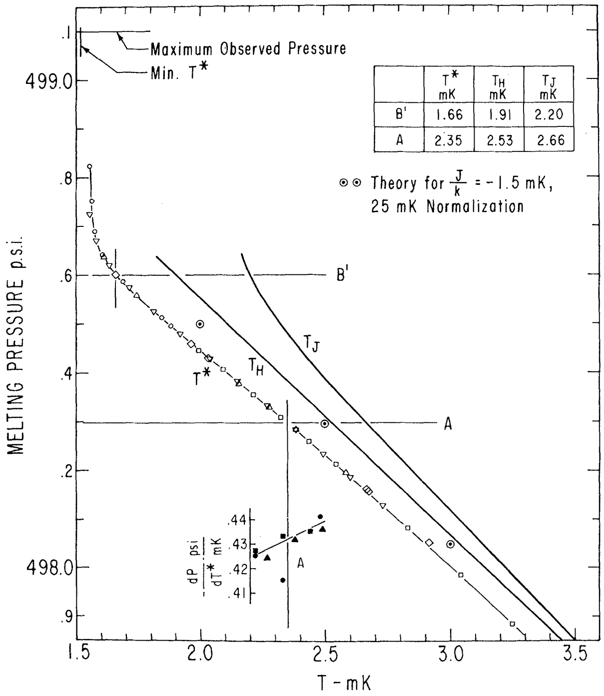
The melting curve of pure He^3 has been measured in the region of the "A" and "B" phenomena discovered by Osheroff, Richardson, and Lee. Within a 2% precision there is no effect of the "A" phenomenon on the slope of the equilibrium melting curve. This indicates that "A" is not related to the properties of solid He^3. Also, we describe a new effect on thermal relaxation of a powdered-cerium-magnesium-nitrate thermometer immersed in He^3.

In a recent Letter, Osheroff, Richardson, and Lee[1] reported some features designated "A" and "B" on the pressurization curve (pressure versus time) of He^3 in an adiabatic compressional cooling cell. The "A" feature, or phenomenon, was originally interpreted[1,2] as a transition in the solid. This interpretation is supported by some thermal-time-constant measurements,[3] but the recent measurements of Osheroff *et al.*[4] and interpretations by Leggett[5] and by Anderson and Varma[6] suggest that both the "A" and "B" phenomena are related to the liquid. In the present work we report equilibrium measurements of the He^3 melting curve in a region which includes the "A" and "B" phenomena. In this region the entropy of solid He^3 effectively determines the slope of the melting curve, so our measurements are sensitive primarily to solid entropy changes.

Our measurements were made using the adiabatic compression device described by Johnson and Wheatley.[7] A Straty-Adams[8] pressure-sensing capacitor was located at its bottom end. Our thermometer was a 1-mg pellet of powdered cerium magnesium nitrate (CMN) (grain size <37 μm) in the form of a right circular cylinder with diameter equal to height, located at the end of a transverse appendage 2 mm diam × 1.5 cm long located between the compression chamber and the capacitor. Some details of the apparatus, as well as typical illustrations of the "A" and "B" phenomena as they occur in our apparatus, have already been given.[9] The static magnetization of the CMN was measured with an rf-biased superconducting quantum-interference device in a trapped field of ca. 0.3 G using the techniques detailed by Giffard, Webb, and Wheatley.[10] Calibration of the CMN thermometer was made in the range 14–45 mK as described by Johnson *et al.*[11] using the theoretical melting curve slope derived from the solid exchange parameter J/k $= -1.5$ mK and a difference in molar volumes between liquid and solid of 1.31 $cm^3/mole$ as suggested by Grilly.[12] In an earlier version of this apparatus[9] we used a copper-foil Johnson noise thermometer like that of Webb, Giffard, and Wheatley[13] in place of the CMN thermometer.

Equilibrium melting-curve data for several runs are shown in Fig. 1 on the CMN magnetic temperature scale labeled T^*. Pressures labeled A, B', and "maximum observed pressure" refer to features on our pressure-versus-time curves. These subtle features are described in Refs. 1, 3, and 9. Quite possibly the much decreased dependence of T^* on pressure at the lowest temperatures reflects magnetic ordering in the CMN rather than an unexpected characteristic of the melting curve. Nothing striking occurs along the melting curve at B' (or B) although its location close to the temperature region in which the CMN begins to order limits us to qualitative observations. We had three runs in which pairs of points were grouped just above and just below "A" so that a local dP/dT^* could be calculated just above, just below, and across the "A" phenomenon. The results are shown inset near the bottom of Fig. 1. The slope dP/dT^* changes smoothly through "A," any discontinuity being less than 2%.

We have also shown as smooth curves in Fig. 1 the melting curve for two other temperature scales measured (T_J) and derived (T_H) by Webb, Giffard, and Wheatley[13] in relation to the powdered-cylinder–CMN magnetic temperature T^*. The curve labeled T_J is the melting curve with T^* corrected to the Johnson noise temperature T_J as given in Ref. 13. The curve labeled T_H[13] is initially based on a correction to T_J using the thermodynamic data of Hudson[14] and co-workers for the heat capacity of CMN but modified (maximum adjustment 0.05 mK) so that $T_J^4 - T_H^4$ is constant, as it would be if in the calibration ex-



	T^* mK	T_H mK	T_J mK
B'	1.66	1.91	2.20
A	2.35	2.53	2.66

FIG. 1. Melting curve of He^3 at millidegree temperatures with pressure plotted against (1) the magnetic temperature T^* of a cylinder of powdered CMN, (2) the corresponding Johnson noise temperature T_J as found in Ref. 13, and (3) a temperature T_H derived in Ref. 13 and based on thermodynamic CMN heat-capacity data of Hudson and co-workers. A and B' refer to phenomena discovered by Osheroff, Richardson, and Lee. Also shown are several melting-curve points (circles with dots) *calculated* from the simple theory for solid-He^3 entropy as discussed in the text. Insets show the slope of the melting curve near A and the temperatures of A and B' on the various temperature scales.

periments the noise thermometer were held above the surrounding He^3 by a constant heat leak. Details of this T_H temperature scale will be given elsewhere by the authors of Ref. 13. Thus the curve labeled T_H is thought to be our best estimate of the melting curve. Temperatures of A and B' on the various scales are given in an inset to Fig. 1. In the earlier measurements by Johnson, Paulson, and Wheatley of the A and B phenomena by noise thermometry,[9] it was suggested that a heat leak to the thermometer kept the B phenomenon from being measured accurately and that the temperature of A was 2.6 ± 0.1 mK. Osheroff and co-workers[1,4] have suggested that A lies near 2.7 mK and B below 2 mK. Our best estimates for temperatures of the B' and A phenomena along the melting curve are 1.95 ± 0.15 and 2.6 ± 0.1 mK, respectively.

The slope of the melting curve ($dP/dT = \Delta S/\Delta V$, where ΔS and ΔV are the molar entropy and volume differences between liquid and solid He^3) is dominated by the solid entropy. At A the liquid molar entropy is only $0.013R$. From the slope at 1.9 mK (T_H scale) we find $0.52R$ for the molar entropy difference between solid and liquid. The maximum observed pressure in this experiment, relative to A, lies between values given in Refs. 1 and 3. If our maximum observed pressure is taken to be the maximum melting curve pressure then one concludes that below 1.9 mK the *average* entropy difference between solid and liquid would be $0.26R$ or about half that derived above from the melting curve at 1.9 mK. Thus, as suggested in Refs. 1 and 3, it appears that the solid entropy is rather higher than expected by simple theory (see, for example, Ref. 11) for a bcc antiferromagnet with nearest neighbor interactions described by a single parameter J which is chosen to fit higher-temperature $(\partial P/\partial T)_V$ data.[15] The circles with central dots in Fig. 1 represent a melting-curve calculation[11] using this simple theory for the solid-He^3 entropy with $J/k = -1.5$ mK and $\Delta V = 1.31$ cm^3/mole. The pressure normalization is 489.59 lb/in.^{2} at 25 mK, as measured in these experiments. Thus, the simple theory predicts the melting pressure increasing more slowly with decreasing temperature than actually observed, indicating that the solid-He^3 entropy is decreasing less rapidly with decreasing temperature than the simple theory predicts.

During the measurements new transient phenomena with the CMN thermometer were also observed. In moving from one equilibrium melting curve point to another the pressure could be changed very rapidly and then stabilized rapidly (time scale of 10 sec), the thermometer lagging behind. The approach of the thermometer to equilibrium could then be measured, a single time constant τ for each pressure (temperature) accurately describing the results. These time constants were found to be reproducible from run to run, rather to our surprise since we had expected the CMN to be covered with an irreproducible amount of solid He^3. Furthermore, at higher temperatures τ increased with decreasing T, as it would if limited by boundary or internal CMN thermal resistance rather than He^3 thermal diffusivity. The time-constant data, shown for several runs as an inset to Fig. 2, are characterized by a relatively slow increase of τ

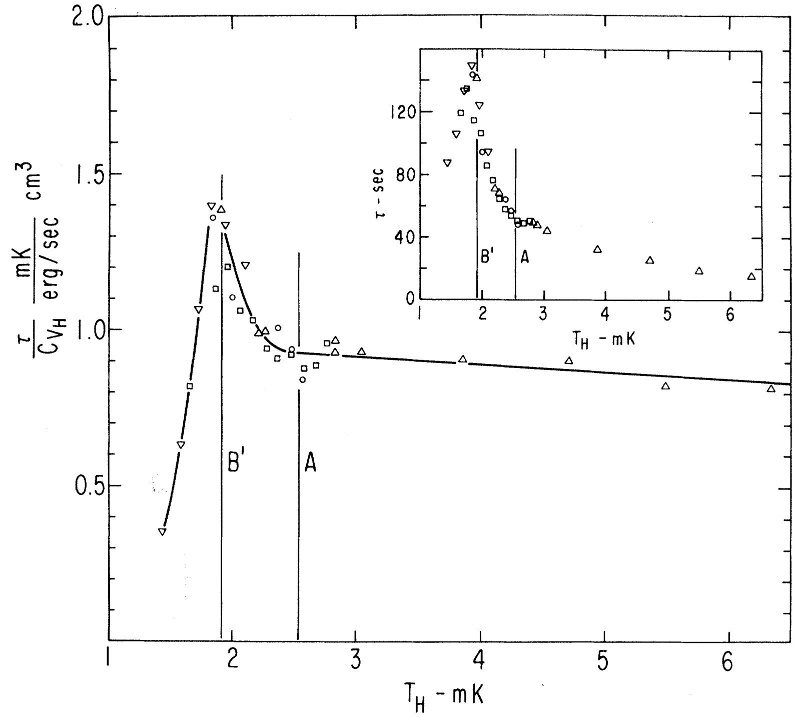


FIG. 2. Ratio of CMN-thermometer thermal time constant τ to CMN heat capacity per unit volume $(C_V)_{\mathrm{H}}$ as function of temperature T_{H} (see text). The thermometer is immersed in the He^3 in the adiabatic compression device. A and B' refer to phenomena discovered by Osheroff, Richardson, and Lee. The inset shows the time constants from which the ratios were derived.

with decreasing T_{H} until A is reached. Below A the time τ increases rapidly with decreasing T_{H} until, just below B, τ drops rapidly. Values of T_{H} below 1.9 mK were obtained by extrapolating the T_{H} melting curve and hence are qualitative only. Note that at all temperatures the times τ are remarkably short. We initially supposed the effect might reflect only the heat capacity of CMN; but Hudson's specific-heat data,[14] on which we based the T_{H} scale, show no maximum in the temperature region given. Examination of the results of Huntzicker and Shirley[16] leads to a similar conclusion. We thus divided τ by $(C_V)_{\mathrm{H}}$, the heat capacity per unit volume as obtained from Hudson's measurements, to obtain a quantity proportional to a thermal resistivity. The result is shown in Fig. 2. There appear to be three regions. Above A, $\tau/C_{V_{\mathrm{H}}}$ changes little with temperature. This is not expected but is somewhat similar to behavior in CMN-He^3 contact observed by Bishop *et al.*,[17] who interpreted a more complicated temperature dependence in terms of a surface magnetic resistance in series with an internal CMN resistance. Between A and B, $\tau/C_{V_{\mathrm{H}}}$ increases rapidly; and then below B, $\tau/C_{V_{\mathrm{H}}}$ decreases rapidly. It is not possible for us to say whether this effect is caused by either liquid or solid He^3 or a combination of the two. It may even be caused by the CMN. Under usual circumstances energy is magnetically coupled to CMN in this temperature range.[17,18] If the effect is due to the He^3, then it suggests that the magnetic coupling becomes progressively less effective as T drops between A and B but that below B the magnetic coupling to the CMN becomes much more effective. Whether or not the effect is related to He^3 properties, it is of considerable technical importance to the future of experimental work with He^3 in this temperature range.

We are indebted to Dr. R. P. Hudson for communicating his measurements of CMN heat capacity to us prior to their publication. We particularly wish to express our appreciation to Mr. R. A. Webb for his contributions in establishing the T_{H} scale in relation to the T_{J} scale. We thank Mr. Arthur Pontau for his assistance in taking and analyzing some of the data.

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Observation of a Second-Order Phase Transition and Its Associated P-T* Phase Diagram in Liquid He^3†

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

In measurements along the melting curve of He^3, using an adiabatic compressional cooling device, Osheroff, Richardson, and Lee[1] discovered two features on the He^3 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 He^3. Later experiments on magnetic properties by Osheroff *et al.*[3] strongly suggested the interpretation of both A and B as liquid-He^3 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 discontinuity but no divergence in the specific heat as a function of temperature, which occurs over a wide range of liquid-He^3 pressure. The transition was not observable in our apparatus for a He^3 pressure of 1.9 lb/in.^{2} (1 atm = 14.696 lb/in.^{2}).

The He^3 specific-heat cell is of standard design[8] containing a mixture of 10.78 g powdered cerous magnesium nitrate (CMN) and 1.23 cm^3 of liquid He^3, 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,