Melting Pressure and the Phase Diagram of Magnetically Ordered Solid ^{3}He

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From high-resolution measurements of the melting pressure of solid ^{3}He in applied fields, we determine the entropy and order of the magnetic phase transitions. We find the entropy discontinuity at the first-order antiferromagnetic transition to decrease as the field increases. No entropy discontinuity is seen at the paramagnetic to high-field transition, indicating that it is not first order.

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Magnetic ordering of the nuclear spins in solid ^{3}He was observed over ten years ago and several experiments since then have elucidated the nature of the transitions.[1-10] At melting pressure and in zero magnetic field, the solid undergoes a first-order transition near 1 mK.[1] The transition temperature and type of ordering depend on the applied field as shown in the field-temperature (B-T) phase diagram of Fig. 1 for the melting solid.[1-5] (Consistent with observations reported here, we show dB/dT continuous along the low-field phase boundary.)

Experiments have not yet given definitive information on the order of the transition along the various phase boundaries. The transitions to the low-field antiferromagnetic phase (LFP) from either the paramagnetic phase (PP) or the high-field phase (HFP) are accepted as first order. However, the PP-HFP transition has been reported as first order, second order, or perhaps only ordering by the field without a phase transition. Theoretical models have been devised to fit existing data and to make new predictions.[10,11] However, these have not been sufficiently refined to yield a unique phase diagram.

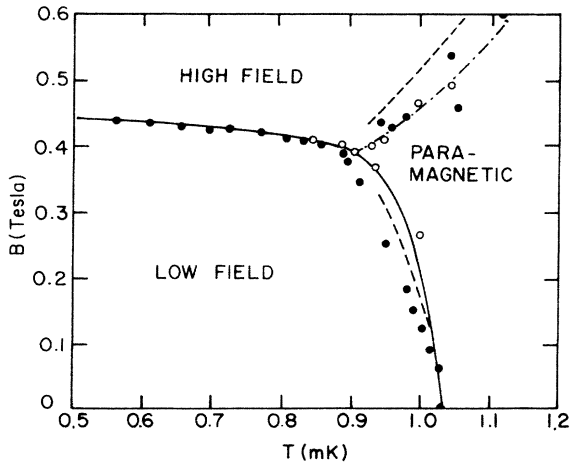

FIG. 1. The B-T phase diagram of magnetically ordered solid ^{3}He at melting pressure: Closed circles, Ref. 5; open circles, this work; dashed line, Ref. 2.

A sensitive way to obtain the entropy of the solid and then the order of the transition is by use of the melting pressure $P(T,B)$ and the Clausius-Clapeyron equation,

$$dP/dT = (S_s - S_l)/(V_s - V_l). \quad (1)$$

Here S_s, S_l, V_s, and V_l are the solid and liquid entropy and molar volumes, respectively. The objective of this experiment was to provide high-resolution $P(T,B)$ data in the region $0.2 < B < 0.5$ T, spanning the low- and high-field phases, and for a range of temperatures going well below the phase transitions.

We used the standard blocked-capillary confinement of a mixture of liquid and solid at melting, with a capacitive strain gauge for measuring the melting pressure.[12] If the cell is filled with an appropriate quantity of helium, a mixture of liquid and solid at melting pressure is maintained as the cell changes temperature. The cell, shown in Fig. 2, was designed to meet a number of special requirements for the measurement of $P(T,B)$ with high precision. The cell body was constructed of sterling silver and the strain gauge of BeCu. Eight 0.76-mm-diam pure silver wires were welded to the cell body to provide additional contact with the 700-Å packed Ag powder. Greater contact between the liquid helium and powder was achieved through eight 1-mm-diam holes drilled in the powder and a central 5-mm-diam hole. The surface area of the powder was 8 m^2 and the helium volume was 1.3 cm^3.

An initial filling of the cell with $v = 25.44$ cm^3/mole was chosen to give a very small fraction of solid ($\sim 6\%$) near 1 mK. The location of the solid in the open volume at the bottom of the cell was favored by gravity and the elevation of the melting pressure in small pores of Ag.[13] Very slow warming rates of $\dot{T} \lesssim 5$ μK/h, corresponding to growth of the solid at a rate of $\dot{V}_s \lesssim 2.5\times10^{-5}$ cm^3/h, assured that the liquid-solid interface remained in the open region of the cell. Thus, with the conditions of this experiment, the pressure registered by the strain gauge was the correct melting pressure, free of spurious effects. The cell could be cooled to below 500 μK by demagnetization of 0.6 mole of $PrNi_5$. After a slow cooling through the

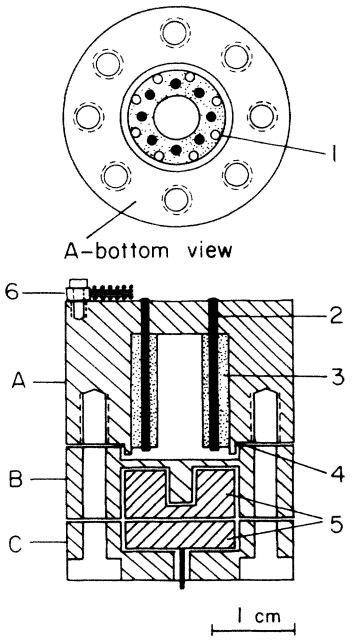

FIG. 2. Schematic view of the melting pressure cell: A, heat exchanger; B, diaphragm and movable plate (5); C, fixed plate holder. 1, holes; 2, Ag wires for thermal contact; 3, Ag packed powder; 4, indium seal; 5, capacitor plates; 6, Pt thermometer.

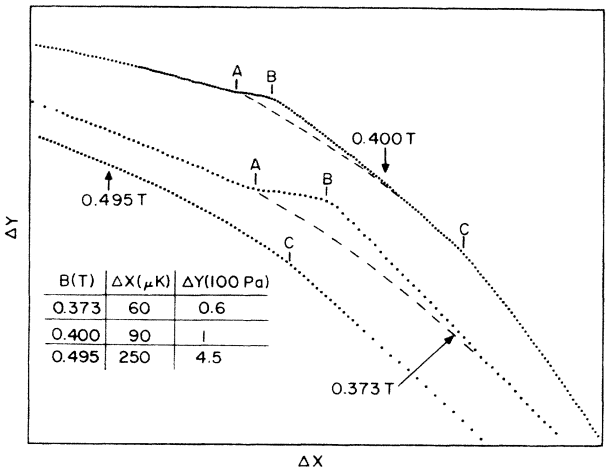

FIG. 3. Melting pressure relative to the A transition vs time or cell temperature for three fields. Note the different scales used for the three fields, with the range of temperatures and pressures as shown in the table. Points A mark the onset of the first-order LFP-PP transition, shown in Fig. 1 as open circles with the solid line. Points C indicate the HFP-PP transition, shown in Fig. 1 as open circles with the dot-dashed line. (Points B are out of thermal equilibrium.) The dashed lines illustrate the behavior of $P(T)$ during the first-order transition.

region $T \sim 1$ mK to locate the phase transitions, the demagnetization was later stopped and data were taken as the apparatus warmed because of the heat leak of ~ 1 nW. Also data were taken during slow cooling.

A Pt wire brush made with 25-μm-diam wires was screwed to the cell body to serve as the thermometer. Its susceptibility was determined by an NMR spectrometer operating at the frequency appropriate to the field applied to the ^{3}He. The thermometer was calibrated against the ^{3}He melting pressure at several temperatures near T_A, the superfluid transition. A timer produced small pulses ($\lesssim 5°$) at intervals of 10 to 30 min for the Pt susceptibility measurement. The integral of the Pt free-induction decay was computed and stored for later analysis. Because of scatter of $\sim 0.3\%$ in the Pt susceptibility, individual points were not used for determination of temperatures. Instead, use was made of the uniform warming rates to smooth the $T(t)$ data. The temperature assigned to each datum was determined from the $T(t)$ fit (a straight line for periods of 24 h or more). This procedure gave the smooth $P(T)$ required for taking derivatives dP/dT.

The pressure strain-gauge outputs were recorded continuously on a strip chart as well as digitally by a scanner that was triggered by the Pt pulses. We show in Fig. 3 the digitally recorded $P(t)$ (cell temperatures, which are linearly related to t, are shown) on warming for fields of 0.373, 0.400, and 0.495 T. At 0.373 T the solid underwent the first-order LFP-PP transition between points A and B. For 0.495 T the HFP-PP transition occurred in the vicinity of point C (below we will discuss the order of this transition). Both the LFP-HFP and the HFP-PP transitions are displayed (separated by about 30 μK) for the 0.400-T field.

It is important to understand the dynamical behavior of $P(t)$ as distinct from $P(T)$ in indicating the order of the various transitions. Because of the very large heat capacity of the $PrNi_5$, the warming rate of the cell body was unaffected by the entropy taken up by the solid ^{3}He in the phase transition. The pressure measured by the strain gauge indicates the temperature at the liquid-solid interface. Thus during a first-order transition there is a *sloping* plateau in $P(t)$, as the interface warms while the interior of the solid undergoes the transition. A second-order transition with a specific-heat discontinuity will produce a discontinuity in dP/dt although dP/dT is continuous. A rapid change in dP/dt will occur in the case of a λ-type transition. This "enhancement" in the behavior of dP/dt relative to dP/dT is well known from observations of the superfluid transitions in liquid ^{3}He.

As seen in Fig. 3, the plateau caused by the first-order transition was observed only when one of the phases was the LFP. For the HFP-PP transition, dP/dt always showed the rapid change characteristic of a λ

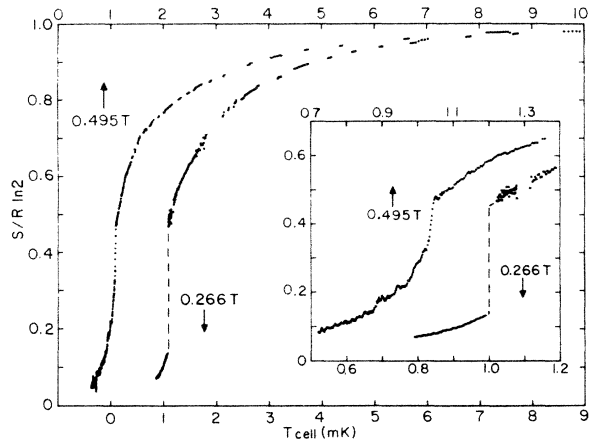

FIG. 4. The solid entropy vs temperature for two fields. The abscissas for the two different curves are displaced for clarity. An expanded view of the transition region is shown in the inset.

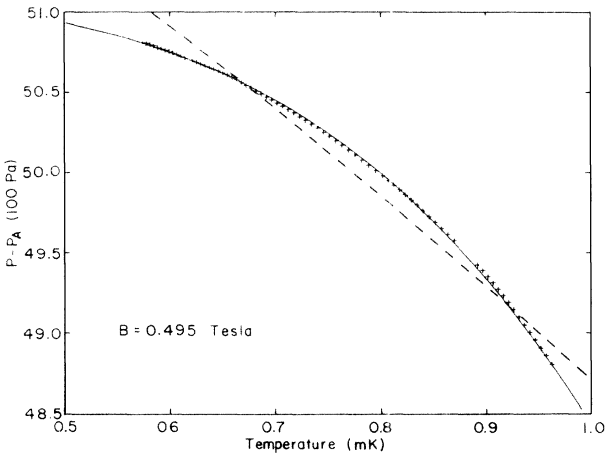

FIG. 5. Melting pressure vs temperature for $B = 0.495$ T: Crosses, measurements; solid line, $P - P_A = 51.092 - 2.66 \times T^4$; dashed line, free spins.

transition.[14] This is particularly apparent at fields not too near 0.400 T, so that the transition region is broader. Also, all of the phase transitions were observed under slow cooling conditions and had the same characteristics as on warming. We conclude that the HFP-PP transition is not first order for $0.4\ \mathrm{T} < B < 0.495$ T. However, we cannot exclude a transition with a small entropy discontinuity, $\Delta S/R\ \ln 2 \lesssim 0.05$, which would produce an indiscernible plateau in $P(t)$.

As we have discussed already, the temperature measured was that of the cell body. In the conversion from $P(t)$ to $P(T)$, the Kapitza resistance causes a temperature difference ΔT between the ^{3}He and the thermometer. Near T_C, the large specific heat produced time constants ~ 1 h and $\Delta T \lesssim 5\ \mu$K. This caused a small error in dP/dT in the vicinity of T_C, which is of little consequence here, since we make no use of the T dependence of S_s derived from dP/dT. As seen in Fig. 3 for $B = 0.373$ T, a temperature difference $\Delta T \approx 10\ \mu$K accumulates (between points A and B) at the first-order transition. During this interval and for a few hours afterwards, the ^{3}He and the thermometer are far from equilibrium and we make no quantitative use of $P(t)$. Behavior of $P(T)$ for the first-order phase transitions is illustrated schematically by the dashed lines in Fig. 3. Elsewhere we convert $P(t)$ to $P(T)$ using the measured warming rates dT/dt.

Upon taking dP/dT and substituting for all other known quantities in the Clausius-Clapeyron relation, Eq. (1), we have the entropy of the solid $S_s(T)$. The results for two fields are shown in Fig. 4. For $B = 0.266$ T, there is an entropy discontinuity $\Delta S/R \times \ln 2 = 0.35$ at the transition. This decreases with field and is only 0.10 at $B = 0.400$ T since most of the entropy has already been removed by the ordering in the HFP. Our values are consistent with $\Delta S/R\ \ln 2 = 0.44$ for $B = 0.014$ T, reported by Osheroff and Yu.[6] We show $S(T)$ continuous through the PP-HFP transition, consistent with the behavior of $P(t)$ discussed earlier. We emphasize that this does not rely on determining whether dP/dT is continuous but on the absence of the plateau in $P(t)$, a much more obvious feature.

Along the LFP boundary of Fig. 1, dB/dT is shown as continuous but with a rapid change near $B = 0.396$ T and $T = 0.90$ mK, where the PP-HFP transition meets the first-order transition line. (This is similar to the P-T phase diagram of ^{4}He where the λ line meets the melting curve.)

The low-temperature form of the specific heat gives important information about the magnetic structure of the HF phase or, indeed, if it is simply a highly polarized paramagnet. An antiferromagnetically ordered phase with a linear spin-wave dispersion has a T^3 specific heat, while a paramagnetic phase has an exponentially small specific heat. The T^3 specific heat gives a melting pressure $P(T) = P_0 - AT^4$, where A is related to the spin-wave velocity c. As shown in Fig. 5, we find that our results for $B = 0.495$ T, which go to $T/T_C = 0.52$, do fit quite well to the T^4 dependence with $A = 2.66$. Spin-wave velocities would be 6.2, 7.8, or 8.9 cm/sec for one, two, or three modes, respectively. Osheroff and Yu reported 8.4 cm/sec for the LFP.[6]

Also shown in Fig. 5, by the dashed line, is $P(T)$ for free spins in a field of 0.495 T. This is a very poor fit because the specific-heat maximum for the Schottky anomaly is at $T = 0.33$ mK and $P(T)$ is almost into the linear region. If we take an effective field as a fit

parameter in $P(T)$ for free spins, $B_{\text{eff}}=3.9$ T is obtained with a fit almost as good as $P\propto T^4$. However, ^{3}He as a fully polarized paramagnet would have an internal field $\sim 10^{-4}$ T; only an ordered state could give such an effective field.

The major conclusions of this work are that the HF phase is magnetically ordered, as indicated by the T^4 melting pressure, and that the transition between this and the paramagnetic phase is not first order.

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[14]Similar appearance would be caused by (1) a second-order transition with a temperature gradient or (2) a specific-heat anomaly without a phase transition. However, with our slow warming rates only small temperature gradients exist except at a transition with a latent heat. The second possibility is excluded by the T^4 dependence of the pressure, discussed later.