

THE MELTING PRESSURE AND ENTROPY OF SPIN ORDERED SOLID ^3He

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Received 4 March 1980

We report measurements of the melting pressure of solid ^3He between 0.36 mK and 1.2 mK. At $1.030 \pm .005$ mK we observe a first order phase transition in the solid with a loss in entropy of $0.443R \ln^2$. Below ~ 0.6 mK the melting pressure varies as T^4 , in agreement with antiferromagnetic spin wave theory.

Nuclear spin interactions in solid ^3He near melting pressures are dominated by the actual exchange of atoms between nearby lattice sites. This unusual exchange mechanism results in a large and quite complex interaction which is not well understood [1]. It is known that solid ^3He near melting pressures orders antiferromagnetically near $T_N = 1.0$ mK [2–5], but details of the transition and of the ordered state have been lacking until quite recently [6,7]. We have performed the first direct, high resolution measurements of the melting pressure of solid ^3He from slightly above T_N to $0.35 T_N$. From these measurements more precise knowledge of the entropy of the solid phase has been obtained. Our results show a discontinuity in the entropy of solid ^3He at T_N equal to $0.443 R \ln^2$ where R is the gas constant. The limiting low temperature behavior is consistent with a model based on antiferromagnetic spin waves as the elementary excitations near $T = 0$. From the low temperature melting pressure data we are able to extract an average spin wave velocity for antiferromagnetic bcc ^3He near zero temperature.

To measure the melting pressure to well below T_N , we constructed a compression cell based on the design of Osheroff et al. [8] but one which contained a sintered silver heat exchanger with about 100 m^2 surface area. When attached to a copper nuclear demagnetization (CND) device, 20 cm^3 of liquid ^3He could be precooled to ~ 0.36 mK with this heat ex-

changer. A concentric cylinder displacement capacitor on the cell piston allowed us to measure and regulate changes in cell volume to about 10^{-6} cm^3 . A capacitance pressure transducer [9] allowed us to measure the melting pressure, P_{melt} , with a resolution of about $3 \mu \text{ B}$. A powdered platinum nmr thermometer allowed us to measure temperature. The thermometer and strain gauge were calibrated against the known properties of the superfluid phases using $T_c = 2.752$ mK and $P_{\text{melt}}(\text{B}') - P_{\text{melt}}(\text{A}) = 20.0 \text{ mB}$ [10]. The pulsed platinum thermometer was checked for linearity and possible offsets in its output by a variety of means and we believe any remaining non-linearities in our thermometry scale are less than 0.5 percent.

To measure P_{melt} the cell was first cooled to ~ 0.5 mK with the CND device. The cell pressure was then raised 5–10 mBar above the melting pressure, and a 2 erg heat pulse was applied to a small heater wire situated near the bottom of the ^3He chamber. From nmr studies we found this procedure would nucleate from ten to twenty seed crystals. The cell volume was then decreased until $\sim 0.03 \text{ cm}^3$ solid had been formed, after which the cell volume was held constant. The cell was then cooled to ~ 0.36 mK by lowering the field on the copper nuclear bundle, and the system was allowed to equilibrate for about three hours. During this time the solid would reach thermal equilibrium and a stable configuration with respect to melting and recrystallization (this last process is discussed in detail below). Data were obtained by raising the field on the copper nuclear bundle slightly,

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waiting until the melting pressure stopped changing, and then measuring P_{melt} and T simultaneously.

To measure P_{melt} accurately it was important that the distribution of solid as a function of elevation in the cell remain fixed: If the elevation of the solid changed, the pressure head of the liquid between the solid and the strain gauge would change, causing a variation in measured cell pressure at constant temperature. The hydrostatic liquid pressure head is about 0.1 mB/cm, and when combined with the high thermal conductivity of the liquid well below T_N , this causes solid to migrate to lower elevations in the cell rather rapidly. By waiting three hours at the lowest temperatures, we were able to ensure all solid migration had ceased.

A potentially more serious and unexpected problem was the tendency for the cell pressure to rise substantially above the melting pressure when solid was being grown. This rise was roughly proportional to the rate of compression, and most severe when only a single crystal of solid existed in the cell. When solid was grown at the rate of only $5 \times 10^{-5} \text{ cm}^3/\text{sec}$ the cell pressure could rise as much as 0.5 mB above the melting pressure. This was true even at the lowest temperatures where the melting curve was flat and in zero magnetic field. By measuring P_{melt} only when solid was not being formed, and by working with a polycrystalline solid sample, we could avoid this problem.

In the figure we show the results of three measurements of P_{melt} in which data were taken upon warming from ~ 0.36 mK in small steps. In all runs a magnetic field of 142 Oe was applied across the sample for thermometry purposes. This small field should not influence the results. Although the melting pressures were reproducible from run to run to within 0.03 mB, data for each warm up were offset so that the melting pressure at 0.36 mK was the same for each run. In the figure we have plotted only $P_{\text{melt}}(T=0) - P_{\text{melt}}(T) (= \Delta P)$. The melting pressure at $T=0$ was obtained by extrapolation of the melting pressure plotted against T^4 as shown in the insert to the figure.

The solid ordering temperature, T_N , can be seen very clearly in the melting curve data; being denoted by an abrupt change in slope. We determined the melting curve slope just above T_N to be 27.2 ± 0.5 mB/mK, and the melting curve slope just below T_N

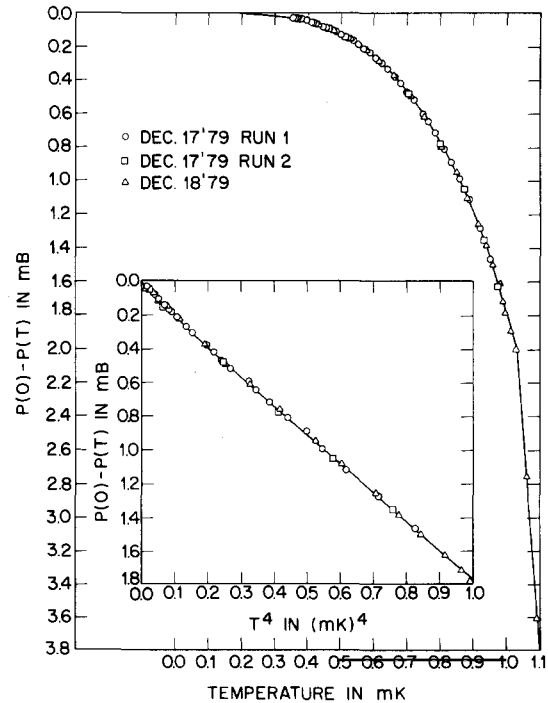


Fig. 1. The difference in melting pressure $P(T=0) - P(T)$ plotted against T (main figure) and T^4 (insert). Data from three separate runs are plotted. The solid lines are given by the function $\Delta P = 0.58 T^8 - 1.2 T^6 + 2.4 T^4 - 0.002$.

to be 7.7 ± 0.4 mB/mK. These slopes can be used to determine the solid entropy by using the Clausius-Clapeyron equation and noting that the liquid entropy is negligible compared to the solid entropy below 1 mK. Using $V_l - V_s = 1.309 \text{ cm}^3/\text{mole}$ as determined by Grilly [11], we find $S_s(T_N >) = 0.618 R \ln^2$ and $S_s(T_N <) = 0.175 R \ln^2$. In three separate runs we found $T_N = 1.030 \pm 0.005$ mK when we fixed T_c for the superfluid transition at melting pressure to be 2.752 mK. This change in entropy corresponds to a latent heat of 2.62 mJ/mole. Undoubtedly, this large latent heat, coupled with the poor thermal conductivity of the solid near T_N , ensures that upon warming ordered solid, thermal equilibrium will be maintained uniformly throughout most of the sample at T_N for quite some time.

The dispersion relation for antiferromagnetic spin waves is linear over a broad range of energies $\hbar\Omega_0 \ll \hbar\omega \ll J$, where Ω_0 is the zero field resonant frequency. In particular, this inequality should be

satisfied for thermal spin waves in solid ^3He well below T_N to the lowest temperatures attainable in our experiment. One therefore expects $S_s \propto T^3$ be analogous to lattice phonons. Since $S_1 = 0$, integration of the Clausius-Clapeyron equation shows that ΔP should vary as T^4 . In the insert to the figure we have plotted ΔP against T^4 . As can be seen, the melting pressure varies nearly as T^4 all the way to T_N , except for a gradual decrease in slope of about 25%. If one fits the data below 0.55 mK to the form $\alpha + \beta T^4$, one finds $\alpha = 2.09$ and $\beta = +0.003$. One can use this coefficient of T^4 to determine a weighted average spin wave velocity from the relationship:

$$C_v = \frac{4\pi^2}{15} k_B \left(\frac{k_B T}{\hbar} \right)^3 \left\langle \frac{1}{c^3} \right\rangle. \quad (1)$$

Here, C_v is the specific heat of the solid and c is the spin wave velocity. Setting $\alpha = 2.09$ yields $\langle 1/c^3 \rangle = 1.67 \times 10^{-3} (\text{sec/cm})^3$, and hence the weighted average spin wave velocity is 8.4 ± 0.4 cm/sec.

One can attempt to fit the entire set of data to various functional forms, although none of them have any theoretical motivation. The functions $\Delta P = 1.60 T^4 + 0.2 T^2 - 0.02$ and $\Delta P = 0.58 T^8 - 1.2 T^6 + 2.4 T^4 - 0.002$ both fit the data well over the entire temperature range; the latter function somewhat better. The positive coefficient of T^2 in the former function indicates the curvature at low temperatures seen in the insert cannot be caused by a heat leak into the thermometer. The best fit to the data near T_N is $\Delta P = 1.71 T^4 + 0.070$, although there appears to be a slight increase in slope very near T_N . The absence of any large increase in slope near T_N is consistent with the strong first order nature of the phase transition.

In conclusion, we have measured the melting pressure of solid ^3He to well below the spin ordering tem-

perature, and find behavior strongly indicative of a first order phase transition at 1.03 mK to an antiferromagnetic state. The change in entropy at T_N can ultimately be used to determine the change in the molar volume of the solid at the first order transition once T_N is followed to pressures above the melting pressure. The temperature dependence of ΔP near zero temperature should ultimately provide a crucial test for any Hamiltonian which is constructed to explain the nature of the ordered phase.

We wish to thank W.O. Sprenger for his technical assistance, and M.C. Cross for many stimulating and useful discussions.

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