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Pressure Measurement of Solid ³He through the Magnetic Ordering Temperature

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The pressure of solid ³He has been measured as a function of temperature and molar volume down to and through its nuclear ordering temperature. Pressure measurement above the ordering temperature reveals that besides the known $1/T$ term there are a significant negative term in $1/T^2$ and a positive term in $1/T^3$. The observed discontinuous change in pressure through the ordering temperature is discussed in connection with the first-order phase transition.

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Solid ³He shows a peculiar nuclear magnetism because of its unique direct exchange interaction, although it has a simple nuclear spin $\frac{1}{2}$ and a bcc crystal in the low-pressure phase. The direct exchange interaction depends strongly on the lattice spacing, i.e., molar volume of solid ³He. In this respect pressure measurements of solid ³He have provided valuable data on the exchange interaction.¹ So far the investigated temperature range, however, has been limited to above 13 mK because of the long relaxation time. We have low-

ered the temperature range down to 0.47 mK, and present here the first measurements of the pressure of solid ³He in that temperature range. Higher-order terms than the first-order term of $1/T$ have been observed in the pressure versus temperature relation above the ordering temperature T_N and new information on the ordering has been obtained from the pressure measurements in the ordered state.

Cooling was performed by means of adiabatic demagnetization of a copper bundle. A pulsed Pt

NMR thermometer was used to measure the temperature of a ^3He cell. The thermometer was calibrated against a nuclear orientation thermometer of ^{60}Co in hcp ^{59}Co single crystal and thereafter corrected for the ordering temperature of solid ^3He on the melting curve.² A Straty-Adams-type capacitance pressure sensor³ was used.

Samples were formed at constant volume by using the blocked capillary method. The ^3He used had a ^4He content of 3 ppm. The cell was packed with a sintered silver sponge composed of 700-Å particles. The available volume was 0.16 cm³ with a surface area of 5.8 m². Since the gap between the sintered material and the diaphragm of the capacitance sensor has a strong influence on the relaxation time, we made the gap as small as possible by compressing the indium O ring between them.

We concentrate on a sample with molar volume of 24.19 cm³ unless stated otherwise. We refer to the results of different molar volumes when necessary. The results of pressure measurements are shown in Fig. 1. We deal with the raw data of pressure for the present, although a pres-

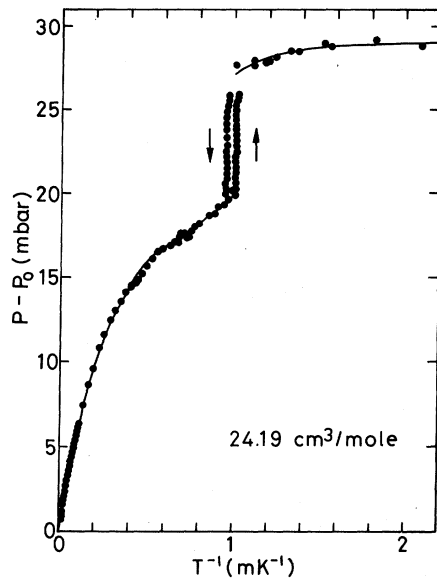


FIG. 1. Pressure vs inverse temperature. P_0 indicates the pressure at high temperature which is not associated with nuclear magnetism. Data are a composite of three runs. Arrows indicate the direction of measurements. A first-order phase transition is evidenced by the discontinuous change in the pressure at $T_N=1.01$ mK. The line above T_N indicates a least-squares fit of $P - P_0 = 61.4/T - 77.2/T^2 + 36.0/T^3$ mbar (T in mK). The line below T_N shows $P - P_1 = 2.2T^4$ (mbar).

sure correction is required as explained in detail later. Data were taken after the complete relaxation of the pressure to equilibrium. The results were fitted up to the third-order term in the range above the ordering temperature by

$$P = P_0 + A/T + B/T^2 + C/T^3,$$

where P_0 , A , B , and C are constants independent of temperature. A , B , and C are 61.4 mbar mK, -77.2 mbar mK², and 36.0 mbar mK³, respectively. This is the first case in which pressure measurements yield terms of higher order than the $1/T$ term. The measured volume dependences of the coefficients are $A \propto V^{38 \pm 2}$, $B \propto V^{61 \pm 3}$, and $C \propto V^{78 \pm 8}$.

The discontinuous change in pressure, ΔP , in Fig. 1 shows the nature of the first-order phase transition, which is confirmed by the rapid decrease of the ^3He signal at 250 kHz in an NMR cell installed separately. The ordering temperature T_N was precisely determined as explained in the following. The time derivative of pressure, dP/dt , through the ordering temperature was plotted against a number of set temperatures, T , of the cell. According to the relation $dP/dt \propto (T_N - T)/R_K$, T yields a precise T_N when dP/dt is interpolated to zero in the plots of dP/dt vs T , where R_K is the Kapitza resistance between the sample and the cell. T_N of samples with molar volumes 24.19, 24.07, 23.85, and 23.72 cm³ were 1.01, 0.93, 0.77, and 0.69 mK, respectively, which varied as $V^{19.1 \pm 0.2}$.

The time evolution of the pressure through the phase transition is shown in Fig. 2 together with

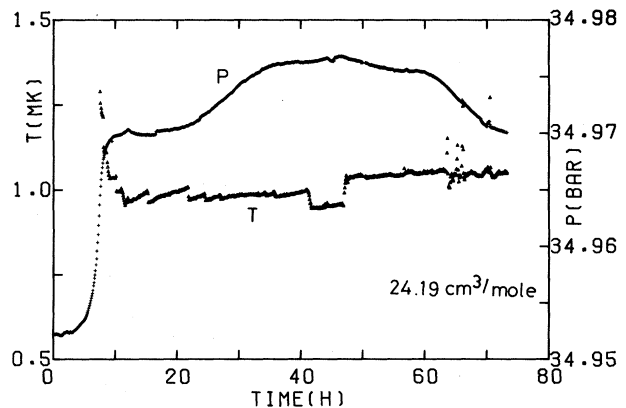


FIG. 2. Pressure and temperature vs time. This run is concentrated on the change in pressure through ordering. The pressure rise at 0.99 mK and the pressure depression at 1.02 mK correspond to passing through the phase transition from above and below T_N .

the temperature. After the temperature was set at 0.99 mK, 25 h were needed to finish the transition completely. This long time is due to the release of the latent heat associated with the first-order phase transition, and is essentially different from the short relaxation time on the order of an hour in temperature regions above and below T_N . As shown in Fig. 2, little change in pressure was observed in the first 14 h after setting the cell temperature below T_N . This duration is the time in which a large portion of the solid ^3He that is contained in the sintered sponge releases its latent heat and its temperature becomes lower than T_N . After the temperature of the solid ^3He in the sponge decreases below T_N , the pressure in a portion of the solid ^3He which is active with the pressure sensor begins to change. Note that the solid helium in the sponge hardly contributes to the pressure because of difficulty in transmission of pressure in the sponge with small pores. The same situation occurred in setting the temperature of the cell above T_N , where the pressure hardly changed in 47 to 60 h in Fig. 2 until the temperature of the helium in the sponge rose above T_N .

The value of ΔP was determined to be the difference between two inflection points in Fig. 2. ΔP varies as $\Delta P \propto V^{19}$, indicating that ΔP varies as a first power of the exchange interaction. This is in strong contrast to the fact that the temperature variation of pressure above T_N is mainly dominated by the second power of the exchange interactions. Therefore, information on the exchange interaction might be deduced from ΔP if a theoretical expression for ΔP were available. If one calculates the entropy difference through the ordering, ΔS , from the Clausius-Clapeyron relation $\Delta P = -\Delta S(dT_N/dV)$ together with $\Delta P = 6.0$ mbar, one gets $\Delta S = 0.133R \ln 2$. As explained below, the entropy change will be used to correct the pressure for change in volume of the cell.

The pressure in the ordered state is expressed as $P = P_1 - DT^4$, where P_1 and D are constants independent of temperature. $D = 2.2 \pm 0.3$ mbar mK^{-4} was obtained. The temperature dependence of pressure and the volume dependence of D as $V^{-60 \pm 10}$ are in agreement with the antiferromagnetic spin-wave theory. Since we know the volume dependence of the pressure in temperature regions above and below T_N , we can obtain the entropy at any temperature by integration of the pressure with volume followed by differentiation with temperature. The entropy at high temperature is calculated to be $0.338R \ln 2$, which is the

sum of the entropy below T_N , $0.065R \ln 2$, the entropy change through ordering, $0.133R \ln 2$, and the entropy above T_N , $0.140R \ln 2$, respectively. It seems that there is no new ordering below T_N obtained in this work since the present T_N is consistent with T_N measured on the melting curve² and we have lowered the temperature down to $T/T_N = 0.47$. A small entropy at high temperature suggests a correction to the pressure measurements. The small change in pressure might be because the volume constancy in pressure measurements is not maintained as a result of a small effective volume for the pressure sensor. The relation between the measured slope dP/dT and the desired constant-volume slope $(\partial P/\partial T)_v$ is⁴

$$\left(\frac{\partial P}{\partial T}\right)_v = \frac{dP}{dT} \left(1 + \frac{1}{\kappa V} \frac{dV}{dP}\right),$$

where κ is the compressibility of helium. If the whole volume of helium in the cell contributed to the pressure, the last term would give a 2.2% correction. However, because of small pores in the present silver sponge, only a small portion of the cell close to the pressure sensor is active and the effective volume V is small, so that the correction term is larger. If the entropy at high temperature should be $R \ln 2$, the correction factor to pressure measurements has to be scaled to 2.95. The entropy discontinuity ΔS is then scaled to $0.394R \ln 2$. Incidentally, ΔS for different molar volumes between 24.07 and 23.72 cm^3 is $(0.127-0.132)R \ln 2$, which scales to $(0.376-0.390)R \ln 2$. The entropy discontinuity through the ordering turns out to be almost constant with decreasing molar volume.

With use of the notation of the four-spin exchange model,⁵ the high-temperature expansion yields the pressure-versus-temperature relation:

$$P = \frac{R}{8} \left(\frac{1}{T} \frac{\partial e_2}{\partial V} + \frac{1}{3T^2} \frac{\partial e_3}{\partial V} + \frac{1}{6T^3} \frac{\partial e_4}{\partial V} \right).$$

If our measurements are scaled to the correct value of pressure, $e_2 = 11 \text{ mK}^2$, $e_3 = 25 \text{ mK}^3$, and $e_4 = 18 \text{ mK}^4$ are obtained by volume integration of the pressure coefficients A , B , and C whose volume dependences are known. These values are in agreement with $e_2 = 13.2 \text{ mK}^2$ and $e_3 = 24.1 \text{ mK}^3$ of Halperin *et al.*⁶ and $e_2 = 12 \text{ mK}^2$ and $e_3 = 28 \text{ mK}^3$ of Dundon and Goodkind.⁷ Note that e_2 and e_3 of these authors are corrected to molar volume 24.2 cm^3 . $e_2 = 11 \text{ mK}^2$ and $e_3 = 25 \text{ mK}^3$ derived in the present study do not yield the solution for the exchange interactions within the framework of

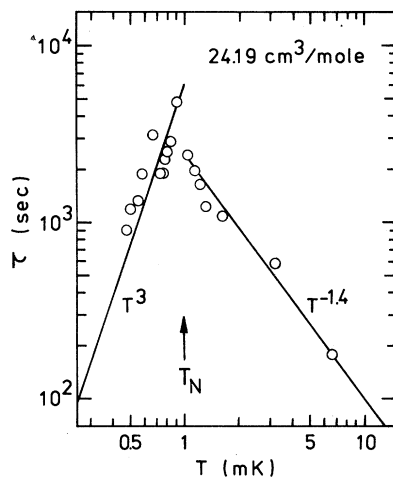


FIG. 3. The relaxation time to the pressure equilibrium vs temperature. The arrow indicates the ordering temperature.

the four-body exchange interaction model when combined with $\theta = -2.6$ mK and $B = -2.7$ mK².⁸ However, $J_1 = -0.20$, $J_2 = -0.48$, $J_3 = -0.20$, $K_P = -0.40$, and $K_F = -0.36$ mK are the closest values to the solutions and they give $e_2 = 12$ mK², $e_3 = 21$ mK³, $\theta = -3.4$ mK, and $B = -2.1$ mK².

The relaxation time τ to the pressure equilibrium is shown in Fig. 3. The relaxation time in the region T_N to 10 mK varies as $2400T^{-1.4}$ sec (T in mK), which yields a Kapitza resistance of the order of $R_K = 10^6$ K m²/W by using $\tau = R_K C$, where C indicates the specific heat calculated by the present values of e_2 , e_3 , and e_4 . In the ordered state τ varies as $(6000 \pm 2000)T^3$ sec, which

yields a Kapitza resistance of the order of 10^6 K m²/W because the specific heat in the ordered state behaves as T^3 .

In conclusion, we have presented the first measurements of the pressure of solid ³He through the ordering temperature. We have obtained higher-order terms than the first-order $1/T$ term in the pressure versus temperature relation, and the pressure discontinuity associated with the first-order phase transition. We have also investigated the pressure in the ordered state and the Kapitza resistance between solid ³He and metal particles.

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