

TEMPERATURE DEPENDENCE OF DEBYE THETA'S FOR bcc ^3He AND hcp ^4He †

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From measurements of $(\partial p/\partial T)_V$, values of $\Theta_D/\gamma^{1/3}$ have been obtained for bcc ^3He and hcp ^4He . Below $T/\Theta_{D\text{max}} \approx 0.02$ a decrease in Θ_D occurs for ^3He but not for ^4He .

Extensive measurements of the heat capacity of both bcc and hcp phases of ^3He and ^4He have been carried out by several investigators.¹⁻⁷ Thorough discussions of these results are given by Pandorf and Edwards (PE)⁶ and by Sample and Swenson (SS).⁵ In general it has been found that in the low-temperature limit the heat capacity of hcp ^4He can be represented by a Debye term only^{3,7} ($C_V \propto T^3$), whereas the heat capacity of bcc ^3He has an anomalous contribution¹ which can be represented by a term linear in T . Expressed in terms of a temperature-dependent Θ_D , the anomaly causes a decrease in Θ_D below $T/\Theta_{D\text{max}} \approx 0.02$.^{5,6} Similar behavior had been found in earlier ^4He work.^{1,2}

It was suggested by SS that the anomalous term might be an apparatus effect rather than a real contribution to the heat capacity. The anomaly was also observed by PE and a number of possible causes enumerated; however, no definite conclusions as to the origin of the effect were reached. Temperature-dependent Θ_D 's have been calculated for bcc helium,⁸ but no insight into the problem is provided since Debye behavior was assumed for $T \lesssim 0.5$ K.

The purpose of the present study was to investigate the temperature dependence of Θ_D through the relation

$$\left(\frac{\partial p}{\partial T}\right)_V = \frac{\gamma C_V}{V} = \frac{234\gamma R}{V} \left(\frac{T}{\Theta_D}\right)^3, \quad (1)$$

where γ is the Grüneisen parameter, R the gas constant, and V the molar volume of the sample. The second equality above follows from the Debye expression for C_V and is taken as the experimental definition of Θ_D . In this experiment the quantity measured is $(\partial p/\partial T)_V$ from which $\Theta_D/\gamma^{1/3}$ is obtained using Eq. (1).

Pressures on the solid were measured using a highly sensitive capacitive strain gauge⁹⁻¹¹ which detected changes in pressure as small as 2×10^{-6} atm. The strain-gauge measurement is fundamentally different from a specific-heat measurement and offers a more favorable situation for eliminating apparatus effects. Since there is no subtraction of the specific heat of the thermome-

ter and empty calorimeter, this source of error is removed. Also the "filling-capillary effect" resulting from the thermal conductivity of the solid helium within the capillary^{5,6} is eliminated.¹²

Measurements were made on five bcc ^3He samples and five hcp ^4He samples. The ^3He samples had a ^4He impurity of 20 ppm. Ordinary commercial helium was used for the ^4He samples. This had been run through a liquifier and repurifier at least once. In order to remove any impurities other than helium, the samples were passed through a liquid-helium cold trap containing a metal-membrane filter with 5- μ pores.

The constant-volume samples were obtained by the blocked-capillary technique with the densities determined from the melting point. Samples were annealed for 30 min at about 0.01 K below the melting point. (The annealing process could be observed through its effect on the sample pressure, and this time interval was determined to be adequate in the present situation.) After the annealing process further changes in temperature took place slowly. The lowest temperatures were obtained by adiabatic demagnetization. Temperatures were measured using a germanium or carbon resistance thermometer and a cerium-magnesium-nitrate magnetic thermometer,¹⁰ with the calibration against ^3He vapor pressure above 0.5 K.

The temperature dependence of the capacitance of the empty strain gauge was measured throughout the temperature interval of interest. There was a very slight temperature dependence below 0.4 K. A correction was applied to the data to account for this; however, the effect on $\Theta_D/\gamma^{1/3}$ was negligible.

Particularly for the low-density ^3He , there is a sizable contribution to $(\partial p/\partial T)_V$ due to exchange.¹⁰ In order to obtain $\Theta_D/\gamma^{1/3}$ representative of the lattice alone, this contribution had to be subtracted. This was done by extending the measurement of p vs T to ~ 20 mK where the lattice has negligible effect and $p \propto T^{-1}$ due to exchange. The exchange contribution was then extrapolated to higher temperatures and subtracted from the total pressure. The exchange energy is so small

for $V < 22 \text{ cm}^3/\text{mole}$ that this subtraction has negligible effect.¹⁰

After presentation of the results, we will give evidence that the effect found in ^3He is not due to ^4He impurities.^{13,14}

The data were taken as values of p and T at constant V . Several similar computer fits were used to obtain the slope $(\partial p/\partial T)_V$. These varied from a linear fit of groups of four adjacent, closely spaced, raw data points to a fit of ten points by a quartic in T . The character of the results varied only slightly with the type of fit used.

From the $(\partial p/\partial T)_V$, values of $\Theta_D/\gamma^{1/3}$ were computed using Eq. (1). These are shown in Fig. 1 for three volumes each for ^3He and ^4He , where $\Theta_D/\gamma^{1/3}$ is plotted versus T/Θ_0 using the Θ_{D0} 's (i.e., $\Theta_{D\text{max}}$) of SS and of Edwards and Pandorf (EP).³ If the appropriate values of γ are used,^{3,5} values of Θ_0 are obtained which are quite close to those of SS and EP. The most striking feature of the present results is that $\Theta_D/\gamma^{1/3}$ reaches a constant value for ^4He at low temperatures,¹⁵ while for ^3He there is a pronounced decrease of $\Theta_D/\gamma^{1/3}$ below $T/\Theta_{D0} \approx 0.02$. Part of the temperature dependence seen for $\Theta_D/\gamma^{1/3}$ is due to γ . However, the temperature dependence of γ is not very great,^{3,5,9} and to account for the decrease in $\Theta_D/\gamma^{1/3}$ below $T/\Theta_{D0} \approx 0.02$ an increase in γ of about 50% would be required. More to the point, γ cannot be temperature dependent unless Θ_D is also. (The converse is not true.) Therefore, at the lowest temperatures, most of the variation in $\Theta_D/\gamma^{1/3}$ should be due to Θ_D .

The maximum error in $(\partial p/\partial T)_V$ occurs at the lower temperatures where the slope is small.

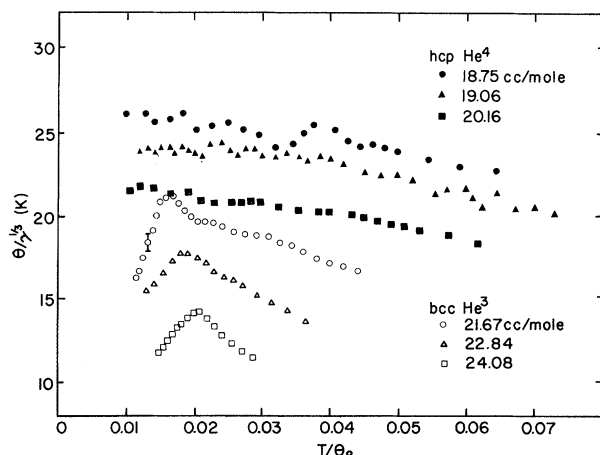


FIG. 1. Values of $\Theta_D/\gamma^{1/3}$ vs T/Θ_{D0} for various molar volumes of hcp ^4He and bcc ^3He .

An estimate of the error in $\Theta_D/\gamma^{1/3}$ resulting from pressure and temperature errors is shown by the vertical bar in Fig. 1. It is seen that this is not adequate to account for the decrease in $\Theta_D/\gamma^{1/3}$ for ^3He . (The scatter in the data for ^4He at $18.75 \text{ cm}^3/\text{mole}$ is a result of less precise temperature control for that particular run.)

We now examine the question of effects of ^4He impurities. For the usual commercially available ^3He with ^4He impurities of a few hundred parts per million, phase separation^{13,14} would occur at $T \sim 0.2 \text{ K}$. However for our samples with 20 ppm ^4He , phase separation would not occur until $T = 0.07 \text{ K}$.¹³ Measurements of excess pressure¹⁴ due to phase separation indicate that the total effect on the pressure, which would occur in the vicinity of 0.07 K , would be $5 \times 10^{-5} \text{ atm}$ or less. The decrease in Θ_D which occurs between $T \sim 0.3$ and 0.5 K corresponds to a change in pressure 10 times as great. Indeed, if as a result of critical fluctuations a significant effect due to phase separation should exist as high as 0.3 K , an increase in Θ_D would result since the contribution to $(\partial p/\partial T)_V$ due to phase separation is negative.¹⁴ Thus, we believe that all sources of spurious effects, such as that due to the apparatus, to nuclear spin ordering, or to ^4He impurities, have been eliminated. We are then led to the conclusion that the decrease in Θ_D for bcc ^3He as was found previously^{5,6} is a real effect associated with the lattice specific heat of pure ^3He .

The low-temperature behavior of Θ_D is in general given by

$$\Theta_D(T) = \Theta_D(0) \left[1 - \frac{20\pi^2}{21} \frac{a_4}{a_2} \left(\frac{kT}{\hbar} \right)^2 + \dots \right], \quad (2)$$

where a_2 and a_4 are integrals arising as coefficients in the series expansion of the phonon spectrum in powers of phonon frequency.¹⁶ For an fcc lattice $a_2 > 0$, and a_4 is negative only for solids which are quite anisotropic.¹⁷ If we assume similar behavior for the bcc and hcp lattices, then our results are to be expected if bcc ^3He is quite anisotropic while hcp ^4He is not.

Clear-cut evidence as to the elastic anisotropies of the various phases from sound-velocity measurements is not available. Extant sound-velocity results^{18,19} seem to indicate that the bcc phase of either isotope is less isotropic than the hcp phase,^{20,21} a result consistent with our interpretation of the sign of $d\Theta_D/dT$ for small T .

These results suggest a need for re-examination of the calculated low-frequency phonon spectrum⁸

for bcc ^3He as well as for further sound-velocity measurements.

If the above supposition is correct, then the decrease in Θ_D for helium is related to the crystal structure rather than to the particular isotope. The work is being extended to hcp ^3He to investigate the temperature dependence of Θ_D in this phase.

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¹E. C. Heltemes and C. A. Swenson, *Phys. Rev.* **128**, 1512 (1962).

²J. P. Franck, *Phys. Letters* **11**, 208 (1964).

³D. O. Edwards and R. C. Pandorf, *Phys. Rev.* **140**, A816 (1965).

⁴G. Ahlers, *Phys. Letters* **22**, 404 (1966).

⁵H. H. Sample and C. A. Swenson, *Phys. Rev.* **158**, 188 (1967).

⁶R. C. Pandorf and D. O. Edwards, *Phys. Rev.* **169**, 222 (1968).

⁷J. K. Hoffer, thesis, University of California, 1968 (unpublished). We are grateful to Professor N. E. Phillips for sending us a copy of this thesis.

⁸F. W. de Wette, L. H. Nosanow, and N. R. Werthamer, *Phys. Rev.* **162**, 824 (1967).

⁹G. C. Straty and E. D. Adams, *Phys. Rev.* **169**, 232 (1968).

¹⁰M. F. Panczyk, R. A. Scribner, G. C. Straty, and E. D. Adams, *Phys. Rev. Letters* **19**, 1102 (1967).

¹¹G. C. Straty and E. D. Adams, to be published.

¹²By use of a valve, this problem has been overcome by J. K. Hoffer, W. R. Gardner, C. G. Waterfield, and N. E. Phillips, in *Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968*, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1969), p. 453. In the strain-gauge measurements of Straty and Adams (Ref. 9) another capillary effect was mentioned. This referred to slippage of the plug in the capillary near the melting curve. This effect can be ruled out at temperatures well away from the melting curve.

¹³D. O. Edwards, A. S. McWilliams, and J. G. Daunt, *Phys. Rev. Letters* **9**, 195 (1962).

¹⁴M. F. Panczyk, R. A. Scribner, J. R. Gonano, and E. D. Adams, *Phys. Rev. Letters* **21**, 594 (1968).

¹⁵This is equivalent to the behavior of $T^{-3}(\partial p/\partial T)_V$ found previously in ^4He by J. F. Jarvis, D. Ramm, and H. Meyer, *Phys. Rev.* **170**, 320 (1968).

¹⁶A. A. Maradudin, E. W. Montroll, and G. H. Weiss, *Solid State Phys. Suppl.* **3**, 123 (1963).

¹⁷A. B. Bhatia and G. K. Horton, *Phys. Rev.* **98**, 1715 (1955).

¹⁸J. H. Vignos and H. A. Fairbank, in *Proceedings of the Eighth International Conference on Low Temperature Physics*, edited by R. O. Davies (Butterworths Scientific Publications, Ltd., London, England, 1963), p. 31.

¹⁹F. P. Lipshultz and D. M. Lee, *Phys. Rev. Letters* **14**, 1017 (1965).

²⁰J. Wilks, *Liquid and Solid Helium* (Oxford University Press, Oxford, England, 1967), p. 651.

²¹W. E. Keller, *Helium-3 and Helium-4* (Plenum Press, Inc., New York, 1969), p. 389.

NUCLEAR ANTIFERROMAGNETISM IN SOLID He^3 †

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The nuclear magnetic susceptibility of pure solid He^3 (1×10^{-5} He^4 impurity) at molar volumes of 23.3, 23.6, and 24.2 cm^3/mole has been measured between 0.2 and 0.04°K. Antiferromagnetic nuclear ordering is indicated with the Néel temperature T_N increasing from $(0.75 \pm 1) \times 10^{-3}$ °K at 23.3 cm^3/mole to $(2.4 \pm 1) \times 10^{-3}$ °K at 24.2 cm^3/mole . Special attention has been given to thermal equilibrium.

For many years the nuclear magnetic properties of solid He^3 have been under intensive theoretical¹⁻⁸ and experimental⁹⁻²³ investigation in hopes of studying nuclear spin ordering at experimentally realizable temperatures. In most solids the dominant interaction between spins is the dipole-dipole interaction, which should produce ordering temperatures of about 10^{-6} °K. However in solid He^3 the large zero-point energy en-

hances the spin-exchange interaction such that the ordering temperature should be on the order of 10^{-3} °K.^{1-8, 11-17}

Earlier nuclear susceptibility measurements^{9,10,13,20} have yielded various inconclusive results which are now thought to be due to poor thermal equilibrium in some cases and He^4 impurities in other cases. We have taken special precautions to circumvent these difficulties. Refrigeration was by