

tured by Union Carbide Corporation, Carbon Products Division, 270 Park Avenue, New York, N.Y.

²E. M. Lifshitz, *Zh. Eksp. Teor. Fiz.* 29, 94 (1955) [*Sov. Phys. JETP* 2, 73 (1956)].

³This assumption is clearly a simplification of a complicated problem and its validity is questionable when $\xi(T)$ becomes comparable to δ .

⁴V. L. Ginzburg and L. P. Pitaevskii, *Zh. Eksp. Teor. Fiz.* 34, 1240 (1958) [*Sov. Phys. JETP* 7, 858 (1958)].

⁵We wish to leave open the interpretation of δ , since the way it is obtained from the data it could include a number of effects such as surface impurities or modifications of the macroscopic wave function.

⁶C. H. Anderson and E. S. Sabisky, *Phys. Rev. Lett.* 24, 1049 (1970).

⁷P. Richmond and B. W. Ninham, *J. Low Temp. Phys.* 5, 177 (1971); E. S. Sabisky and C. H. Anderson, *Phys. Rev. A* 7, 790 (1973).

⁸The parameter α is shown in Refs. 6 and 7 to decrease slowly with increasing d , but by using the ratio in Eq. (2) this effect is, to a large extent, compensated.

⁹This value for α is higher than our previously reported values because of a variety of small corrections.

¹⁰J. A. Herb and J. G. Dash, *Phys. Rev. Lett.* 29, 846 (1972). Our values for the onset on Grafoil disagree

with those reported in this reference.

¹¹We are indebted to Dr. A. Goodman of RCA Laboratories for supplying the treated samples.

¹²H. W. Chan, A. W. Yanof, F. D. M. Pobell, and J. D. Reppy, in *Proceedings of the Thirteenth International Conference on Low Temperature Physics*, Boulder, Colorado, 1972 (to be published). Chan *et al.* reported an onset thickness of 10 Å for bare Vycor and 7 Å for neon preplated Vycor at 0.57 K. Using the results of the present paper and the calculated value of α for neon, we calculate a difference of 2 Å in the onset thickness in this case.

¹³For a listing of references see Ref. 12 or 10.

¹⁴This value for the length $\xi(T)$ agrees exactly with that estimated by Mamaladze for the characteristic length of bulk liquid helium when $\xi(T)$ is divided by the geometric factor π , which is the value given by Ginzburg and Pitaevskii (Ref. 4) assuming the superfluid density vanishes at both surfaces {Yu. G. Mamaladze, *Zh. Eksp. Teor. Fiz.* 52, 729 (1967) [*Sov. Phys. JETP* 25, 479 (1967)]}.

¹⁵J. R. Clow and J. D. Reppy, *Phys. Rev. Lett.* 16, 887 (1966).

¹⁶See, for example, R. A. Ferrell, N. Menyhard, H. Schmidt, F. Schwabl, and P. Szepfalussy, *Ann. Phys. (New York)* 47, 565 (1968), and references therein.

Low-Temperature Specific-Heat Anomaly of bcc ^3He

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(Received 16 April 1973)

The specific heat of bcc ^3He has been measured from 25 mK to near the melting curve, with particular attention given to the region of the low-temperature anomaly, $T \lesssim 500$ mK. Here we find that the specific heat has a term linear in T , in addition to the Debye T^3 term. From the specific heat at the lowest temperatures, for the larger-molar-volume samples, values of the exchange energy J in agreement with previous results are found.

Previous measurements on the bcc phase of ^3He have indicated that the specific heat C_v does not have the T^3 temperature dependence expected from the Debye theory at temperatures $T < 0.5$ K.¹⁻³ This "low-temperature anomaly" can be characterized by an excess specific heat. There is also an anomaly in bcc ^3He at temperatures $T > 0.5$ K which is characterized by excess specific heat, possibly due to vacancies in the lattice. The cause of the low-temperature anomaly is still open to speculation. Since an apparent low-temperature anomaly was seen in early ^4He data⁴ but not in more recent work,⁵ some doubt has persisted as to the validity of the effect in ^3He . However, subsequent measurements of $(\partial P/\partial T)_v$ ⁶

and of thermal conductivity⁷ substantiate the existence of the anomaly in bcc ^3He .

The object of this work⁸ was to make measurements of sufficient precision to show the temperature and volume dependence of the anomalous contribution. For comparison and as a check of our apparatus and procedure, two runs were made in hcp ^4He .

The cylindrical calorimeter, made of beryllium-copper and copper, has a sample volume of 0.903 cm³. It contains a copper wire brush of 0.06-mm-diam wires with an average spacing of 0.16 mm, which provides 250 cm² surface area for thermal contact to the sample. The stainless-steel filling capillary between the thermal

shield and the calorimeter is 10 cm long, with i.d. and o.d. of 0.004 and 0.008 in., respectively. A capacitance strain gauge, used to determine the molar volume V of each sample from its melting pressure⁹ and to calibrate the resistance thermometers against the melting pressure of ^3He ,¹⁰ was machined on the bottom of the calorimeter. Thermal contact between the calorimeter and the cooling source was provided by a Sn heat switch. The size of the switch varied from $A/L = 5 \times 10^{-3}$ to 6×10^{-5} cm, depending upon the temperature range of interest.

The heat capacity was measured by the standard heat-pulse method with a small correction for the fore- and afterdrifts. One difference from previous work on helium was in the data analysis. Following Collan *et al.*¹¹ a functional form was assumed for the specific heat, such as $C = aT^{-2} + bT + cT^3$. The fitting parameters a , b , and c were obtained from a least-squares fit to the data by equating the heat added during each pulse, Q , to the integral of the assumed functional form evaluated at the initial and final temperatures T_i and T_f . With the fitted parameters, calculated values of C and Q , C_{calc} and Q_{calc} , were found. The experimental points presented here give the specific heat at the midpoint of the heating interval and were obtained from $C_{\text{meas}} = C_{\text{calc}} \times Q_{\text{meas}}/Q_{\text{calc}}$. The results are largely independent of the particular form assumed for C as long as enough fitting parameters are used to give a good fit to the data. By fitting Q , T_i , and T_f directly, the need for a curvature correction is eliminated, allowing large temperature intervals to be used. Intervals of about 10 to 50% of T were used. An additional analysis using a polynomial least-squares fit with a standard curvature correction gave essentially the same results.

Except for the $V = 24.13$ -cm³/mole sample (all volumes in cm³/mole), which was from an early run, all of the samples were annealed near their melting point for several hours. However, the $V = 24.40$ sample, which melts at 35 mK, was used to calibrate the resistance thermometers below this temperature and was not reannealed. All the ^3He samples contained 2 ppm ^4He , except for the $V = 21.67$ sample, which had 10 ppm.

The heat capacities of the empty cell and of three samples of ^3He and one of ^4He are presented in Fig. 1. (Other samples studied have been omitted for clarity.) In evaluating the heat capacity of the $V = 24.13$ ^3He sample, the only one studied below 65 mK, a straight-line extrapolation of the heat capacity of the empty sample chamber

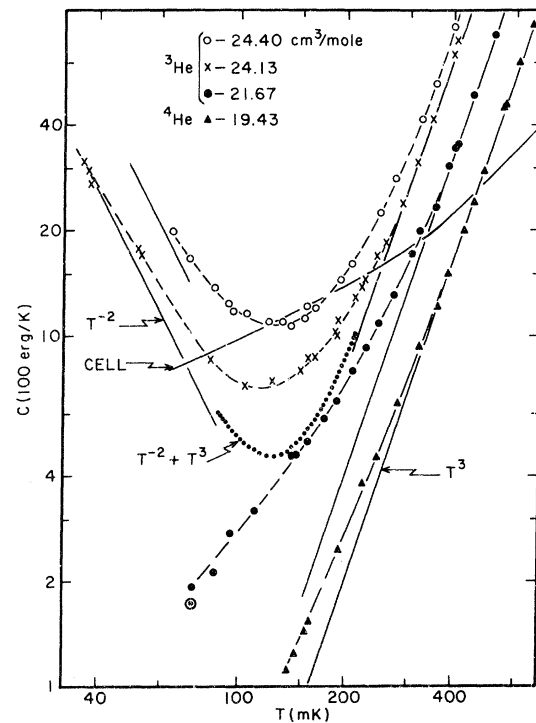


FIG. 1. Heat capacities versus temperature. The solid line marked "cell" represents the empty calorimeter. The dotted line, the solid lines marked T^{-2} and T^3 , and the encircled dot are discussed in the text.

was assumed, since it was not measured for $T < 65$ mK. The 24.40- and 24.13-cm³/mole samples show the large nuclear spin-ordering contribution, $C_s = 3R(J/kT)^2$, at the lowest temperatures (R is the gas constant and k is Boltzmann's constant). Because of the strong volume dependence of J , C_s is much smaller for the smaller-molar-volume samples. The T^{-2} lines drawn for $V = 24.40$ and 24.13 represent C_s for these samples with $|J|/k = 0.89$ and 0.67 mK, respectively. The latter value is in precise agreement with that of Panczyk and Adams,¹² while that for $V = 24.40$ is about 7% larger than would be found from extrapolating their results.¹³ This is the first determination of $|J|$ from specific heat measurements.

In Fig. 1, T^3 lines representing Debye behavior have been drawn tangent to the results for several samples. For the $V = 24.13$ sample, the dotted curve shows the sum of the T^{-2} and T^3 terms. The data fall well above this curve, with the difference representing the "excess" heat capacity. The slight effect of the spin term for the $V = 21.67$ sample is shown by the encircled point at 75 mK. Thus, with C_s decreasing as

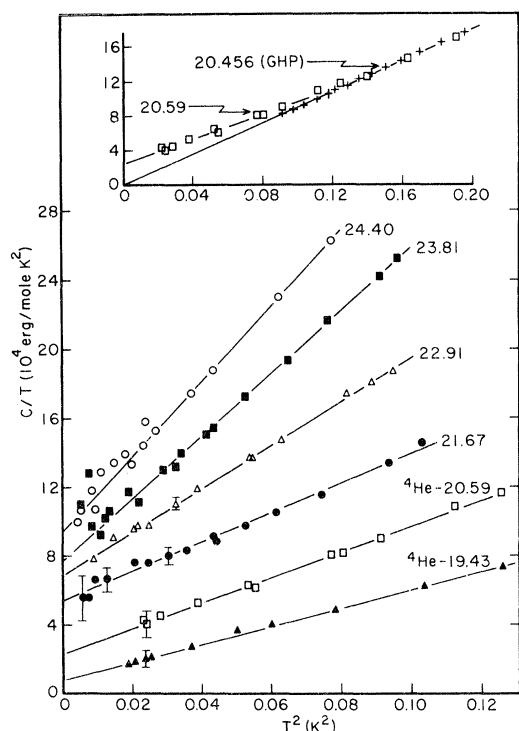


FIG. 2. C/T versus T^2 . Inset, 20.59-cm³/mole ⁴He data compared with 20.456-cm³/mole ⁴He data from Gardner, Hoffer, and Phillips (Ref. 5).

T^{-2} , most of the departure of the $V=21.67$ data from the T^3 line represents "excess" heat capacity.

To check the apparatus and procedure, ⁴He was studied since it has been established as showing T^3 behavior for T down to 300 mK. Our results for ⁴He at $V=19.43$ are shown in Fig. 1. A nearly- T^3 dependence exists in the range from 700 to about 350 mK. The gradual departure of the observed specific heat from the T^3 behavior below 350 mK is almost within our experimental uncertainty. We take the near- T^3 behavior of the ⁴He as evidence that the excess heat capacity of the ³He is not due to an experimental artifact. At present we are not certain whether the ⁴He has a slight excess heat capacity or if this is due to some experimental effect.

In order to identify the temperature and volume dependence of the excess specific heat, we show in Fig. 2 the specific heat of four ³He samples in the region of the anomaly and of two ⁴He samples, in the form of C/T versus T^2 . The T^{-2} spin contribution was first subtracted for the ³He samples. If the specific heat is given by $C=AT+BT^3$, this type of plot gives a straight line with inter-

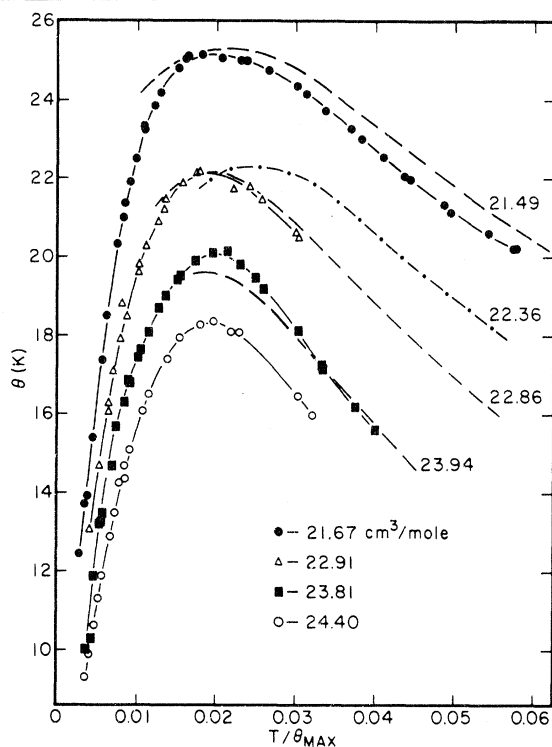


FIG. 3. Debye temperature Θ versus T/Θ_{\max} for bcc ³He. The dashed lines are the results of Sample and Swenson (Ref. 1) and the dot-dashed line the results of Pandorf and Edwards (Ref. 2).

cept $C/T=A$ and slope B . Our data in Fig. 2 fall on straight lines with nonzero intercepts indicating a linear contribution to the specific heat, in addition to the T^3 term, at the lowest temperatures.¹⁴ (A similar plot in the form C versus T^3 , to look for a constant term, yielded curved lines with zero intercepts.) The magnitude of the linear term decreases with decreasing molar volume in both ³He and ⁴He. However, the term is smaller in ⁴He than would be expected in ³He at the same V , based on extrapolation of the ³He results.

For comparison with our ⁴He data, results of Gardner, Hoffer, and Phillips⁵ for V close to that of one of our samples are shown in the inset of Fig. 2. The agreement is good above 350 mK, but our data begin to depart from the T^3 line at lower T , indicating a small linear term. Further comment on this behavior will be made later.

It is customary to represent the lattice specific heat by a T -dependent Debye temperature Θ . Shown in Fig. 3 are Θ 's calculated from our data for four bcc ³He samples, with the spin contri-

bution C_s first subtracted. Smoothed results of Sample and Swenson¹ and of Pandorf and Edwards² are shown for comparison. Where they overlap, the various results are reasonably consistent. For heavier rare-gas solids, Θ approaches a constant, Θ_0 , at temperatures less than $T/\Theta_0 \approx 0.02$. However, because of the low-temperature anomaly, Θ for bcc ³He does not approach a constant, but reaches a maximum, Θ_{max} , at $T/\Theta_{\text{max}} \approx 0.02$. This is particularly evident in our results where Θ is still decreasing at $T/\Theta_{\text{max}} \approx 0.004$. Greywall and Munarin¹⁵ have determined the elastic Θ_0 from sound velocity measurements on a sample with $V=21.64$ and found $\Theta_{0\text{elas}} = 19.5^{+2}_3$ K. In our measurements for $V=21.67$ a value for Θ has been found at the lowest temperature which is less than this.

As already mentioned, the slopes in Fig. 2 provide the coefficients of a T^3 term in the specific heat. *If one were to assume* that the linear contribution is not due to the lattice (see the discussion which follows on possible causes of the anomaly), then the remaining T^3 term would represent the Debye specific heat for the lattice. Calculated Θ_0 's from the slopes in Fig. 2 are 10–15% higher than the Θ_{max} of Fig. 3. We do not suggest that the slopes provide the correct Θ_0 's.

Several theoretical explanations for the low-temperature anomaly have been offered. Horner¹⁶ has calculated the phonon spectrum for bcc ³He at four molar volumes using the self-consistent harmonic approximation corrected for cubic anharmonicities. His results show a decrease in Θ at $T/\Theta_{\text{max}} \approx 0.02$ as a result of anomalous dispersion, but the magnitude of the effect is too small. Other calculations, using a different method for treating short-range correlations, show no anomalous dispersion. Our observed large effect would not rule out anomalous phonon dispersion as a cause for part of the excess specific heat. Guyer¹⁷ has suggested that a zero-point concentration of vacancies of about 10^{-5} would produce an excess specific heat of the right magnitude. However, he obtains a constant specific heat from the vacancies, which is contrary to our observed linear term (he also notes such a concentration of vacancies would be incompatible with existing NMR data). Allen¹⁸ has suggested that long-wavelength phonons from the helium will have a large transmission coefficient across the He-Cu boundary and that these phonons will excite the free electrons in the copper

to produce their characteristic T -dependent specific heat. The magnitude of this effect would depend on the amount of copper and of helium, the phonon mean free path, Θ_0 , and the number of atoms per unit cell. This effect might provide a basis for explaining the small linear term seen in ⁴He by us but not by Gardner, Hoffer, and Phillips, since the dimensions of their sample were much greater than ours.

We acknowledge the assistance of W. P. Kirk and useful discussions with K. R. Allen, R. A. Guyer, and L. H. Nosanow. We are particularly grateful to S. B. Trickey for extensive discussions and his continued interest in this work.

†Work supported in part by the National Science Foundation.

¹H. H. Sample and C. H. Swenson, *Phys. Rev.* **158**, 188 (1967).

²R. C. Pandorf and D. O. Edwards, *Phys. Rev.* **169**, 222 (1968).

³This has been discussed by S. B. Trickey, W. P. Kirk, and E. D. Adams, *Rev. Mod. Phys.* **44**, 668 (1972).

⁴J. P. Franck, *Phys. Lett.* **11**, 208 (1964).

⁵W. R. Gardner, J. K. Hoffer, and N. E. Phillips, to be published.

⁶P. N. Henriksen, M. F. Panczyk, S. B. Trickey, and E. D. Adams, *Phys. Rev. Lett.* **23**, 518 (1969).

⁷W. C. Thomlinson, *Phys. Rev. Lett.* **23**, 1330 (1969).

⁸Some of these results have been reported previously by S. H. Castles, W. P. Kirk, and E. D. Adams, in *Proceedings of the Thirteenth International Conference on Low Temperature Physics*, Boulder, Colorado, 1972 (to be published).

⁹E. R. Grilly, *J. Low Temp. Phys.* **4**, 615 (1971).

¹⁰R. A. Scribner and E. D. Adams, in *Temperature, Its Measurement and Control in Science and Industry*, compiled by the American Institute of Physics (Instrument Society of America, Pittsburgh, Penn., to be published), Vol. 4.

¹¹H. K. Collan, T. Heikkila, M. Krussius, and G. R. Pickett, *Cryogenics* **10**, 389 (1970).

¹²M. F. Panczyk and E. D. Adams, *Phys. Rev.* **187**, 321 (1969).

¹³Part of this difference may be due to some superheated liquid in the solid after melting the sample at 35 mK.

¹⁴Various other workers have noted a linear term in the specific heat of solid helium. However, our identification of this term in Fig. 2 is from data below 300 mK where it has become larger relative to the T^3 term.

¹⁵D. S. Greywall and J. A. Munarin, *Phys. Lett.* **31A**, 469 (1970).

¹⁶H. Horner, *Phys. Rev. Lett.* **25**, 147 (1970).

¹⁷R. A. Guyer, private communication.

¹⁸K. R. Allen, private communication.