Absence of the Low-Temperature Specific-Heat Anomaly in bcc ^{3}He

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The specific heat of bcc ^{3}He has been measured along three isochores for temperatures T between 50 mK and the melting curve. The data for $T < 0.3$ K can be described well by the function $C_V = \alpha T^{-2} + \gamma T^3$ and are inconsistent with the existence of the low-temperature specific-heat anomaly reported previously by others.

Over the past fifteen years the specific heat at constant volume C_V of bcc ^{3}He has been measured by several groups[1-4] in the temperature range extending from below 0.3 K to the melting curve. *All* of these measurements have shown, in addition to the lattice specific heat proportional to T^3, an anomalous excess contribution for temperatures $T \lesssim 0.5$ K. In each experiment the anomalous component was considered to be a real effect and not due to experimental difficulties. In addition to the lattice, the only other known contribution to the specific heat at low temperatures is due to the ordering of the nuclear spins. Although this term has a T^{-2} temperature dependence and thus becomes increasingly significant as the temperature is lowered, its size is too small to explain the observations. Several other possible sources for the excess specific heat have been suggested.[4-9]

The most recent measurements of C_V by Castles and Adams[4] extend down to 65 mK and can be described, at each of the several molar volumes considered, by the function[10]

$$C_V = \alpha T^{-2} + \beta T + \gamma T^3. \qquad (1)$$

In this Letter I present new high-precision specific-heat data along three isochores for $T > 50$ mK which are significantly different from all of the earlier results. At 100 mK I measure a specific heat which is 30% less than that reported by Castles and Adams. The present results show no anomalous behavior. For $0.05\text{ K} < T < 0.30$ K, and at each molar volume, they can be well described by Eq. (1) with β set equal to zero.

The data were obtained using the standard heat-pulse technique with temperature increments equal to approximately 5% of T for most of the data. Two germanium thermometers in separate ac resistance bridges were used to span the complete temperature range and were calibrated against the magnetic susceptibility of cerium magnesium nitrate below 0.9 K.

The calorimeter was machined from oxygen-free high-conductivity copper and had an internal volume of nominally 9 cm^3. There were twelve 0.15-cm-diam copper rods passing through the sample volume to aid in making thermal contact to the solid helium sample, but still leaving the chamber with a fairly open geometry. It was hoped that the open geometry would help in reducing both the number of crystal defects and also the size of density gradients in the solid helium samples. The emply calorimeter had a specific heat which was within 5% equal to that of the equivalent mass of pure copper. The calorimeter was rigidly supported below the mixing chamber of a dilution refrigerator on three graphite rods. To counterbalance the residual heat leak of 2 nW into the cell, a fine copper wire linked the calorimeter to the mixing chamber. A mechanical heat switch was used to cool the sample chamber initially to less than 50 mK. The stainless-steel filling capillary between the mixing chamber and the calorimeter was 8 cm long and had an i.d. of 0.005 cm and an o.d. of 0.015 cm.

Between 20 and 40 h were required to solidify the sample completely at constant density. No further annealing was performed. The ^{3}He sample had a ^{4}He impurity level of 2.40 ppm.

The low-temperature specific heat results are shown in Fig. 1. For $T \lesssim 0.1$ K the specific heat is due mainly to the nuclear spin contribution proportional to T^{-2} while at higher temperatures the lattice contribution proportional to T^3 dominates. At the lowest two densities the scatter in the data is typically a few tenths of a percent. At the highest density it is about an order of magnitude larger mainly because the thermal relaxation times, which increased rapidly with increasing density, had become very long (4 min at 65 mK) for $T \lesssim 0.2$ K. The smoothed results of Castles and Adams at 24.40 cm^3/mol are also shown in the figure. The two sets of measurements are in agreement at $T \approx 0.065$ K and for $T \gtrsim 0.3$ K; however, in the region of the minimum in the curves

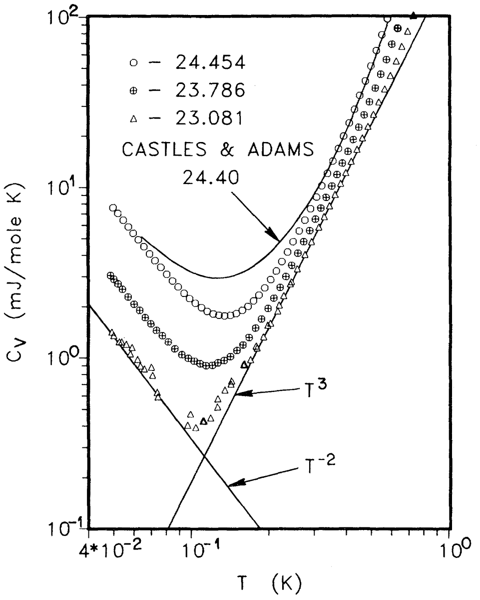

FIG. 1. Specific heat of bcc ^{3}He versus temperature. The numbers are the molar volumes in cubic centimeters. The smooth curve was determined using data given in Ref. 4.

there is a large discrepancy. Although not shown in the figure, a similar difference exists at the other molar volumes.

Castles and Adams were able to describe their results by Eq. (1) with β approximately equal to 9, 8, and 7 mJ/mol K^2 at molar volumes of 24.40, 23.81, and 22.91 cm^3, respectively. I also fitted the data below 0.3 K with the same function and obtained values of 0.02 ± 0.05, 0.14 ± 0.05, and -0.9 ± 0.3 mJ/mol K^2 for nearly corresponding molar volumes of 24.454, 23.786, and 23.081 cm^3, respectively. The two sets of parameters differ by an order of magnitude in absolute value. Although the standard error bars on the present values of β do not include zero for each of the molar volumes, the deviation from zero is not considered to be significant. Small systematic errors in the measurements or the fact that other terms due to the lattice and proportional to higher powers of T should have been included in the fitting function could account for this difference. I thus have no evidence for any contribution to the specific heat other than that attributable to the nuclear spins or to the phonons. Table I gives the best-fit parameters when the data are fitted with Eq. (1) with β set equal to zero. In place of the α and γ values, the exchange energy $|J|/k_B$ and Debye temperature at 0 K, Θ_0, are listed. These parameters were calculated from α and γ using the relations

$$|J|/k_B = (\alpha/3R)^{1/2}, \tag{2}$$

and

$$\Theta_0 = (12R\pi^4/5\gamma)^{1/3}, \tag{3}$$

where k_B is Boltzmann's constant and R is the gas constant. The uncertainties quoted for each of the values in the table are standard errors in the parameters. The additional systematic errors are difficult to estimate, but we feel they are less than 0.5% for all of the parameters except for the value of $|J|$ at 23.081 cm^3/mol. At this density the long thermal relaxation times encountered at low temperatures may be responsible for a considerably larger systematic error. The values of $|J|$ given in the table are generally in good agreement with those determined by others from different types of measurements.[11,12] The Debye temperature for the largest molar volume is in excellent agreement with the value determined from sound velocity measurements[13] in single crystals of ^{3}He at 24.45 cm^3/mol, namely 18.66 ± 0.34 K. The values of Θ_0 are also in agreement with the values determined from the previous specific heat measurements by others[1-4] if their data below approximately 0.5 K are disregarded.

TABLE I. The nuclear exchange energies and Debye temperatures resulting from fits of the specific heat data below 0.3 K to the function $C_V = \alpha T^{-2} + \gamma T^3$.

Molar volume (cm^3)	$\lvert J \rvert/k_B$ (mK)	Θ_0 (K)
24.454	0.8756 ± 0.0006	18.91 ± 0.02
23.786	0.5435 ± 0.0008	20.52 ± 0.03
23.081	0.359 ± 0.007	21.97 ± 0.09

Another representation of the present specific heat results is given in Fig. 2 where I have plotted $\Theta(T)$ calculated from the data using the equation

$$C_V - \alpha T^{-2} = (12\pi^4/5)R[T/\Theta(T)]^3. \tag{4}$$

If what remains after subtracting the spin-ordering contribution from the total measured specific heat of the sample is due to the lattice alone, then $\Theta(T)$ should be nearly constant for sufficiently low temperatures ($T/\Theta_0 \lesssim 0.02$). This is the behavior observed in most crystalline solids, including hcp ^{4}He.[14] The present data are consistent with this expectation, but contrary to the rapid decrease in $\Theta(T)$ with decreasing tempera-

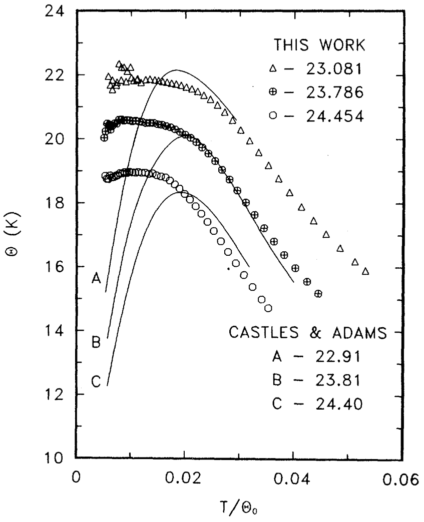

FIG. 2. The Debye thetas for bcc ^{3}He versus reduced temperature. The numbers are the molar volumes in cubic centimeters. The smooth curves were determined using the data of Castles and Adams tabulated in Ref. 4 with Θ_0 set equal to the maximum value of $\Theta(T)$.

ture that has been reported on the basis of all of the previous C_V measurements[1-4] on bcc ^{3}He. The small decrease in $\Theta(T)$ in the present data for $T/\Theta_0 \lesssim 0.008$ is within experimental uncertainty. Also shown in the figure are the smoothed results of Castles and Adams which are qualitatively typical of the earlier measurements.

It is concluded that the present specific heat data are inconsistent with the existence of the long-standing low-temperature specific-heat anomaly in bcc ^{3}He and thus that the anomaly observed by others is not due to some intrinsic property of this quantum solid.

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

[2] H. H. Sample and C. A. Swenson, Phys. Rev. 158, 188 (1967).

[3] R. C. Pandorf and D. O. Edwards, Phys. Rev. 169, 222 (1968).

[4] S. H. Castles and E. D. Adams, Phys. Rev. Lett. 30, 1125 (1973), and J. Low Temp. Phys. 19, 397 (1975).

[5] C. M. Varma, Phys. Rev. Lett. 24, 203, 970(E) (1970).

[6] H. Horner, J. Low Temp. Phys. 8, 511 (1972).

[7] R. A. Guyer, J. Low Temp. Phys. 6, 251 (1972).

[8] I. E. Dzyaloshinskii, P. S. Kondratenko, and V. S. Levchenkov, Zh. Eksp. Teor. Fiz. 62, 1574 (1972) [Sov. Phys. JETP 35, 823 (1972)].

[9] D. S. Greywall, Phys. Rev. B 11, 4717 (1975).

[10] Although Eq. (1) can be used to describe the data of Ref. 4, this does not imply that βT is the anomalous contribution to the specific heat. It has been shown (Ref. 9) that if γ is required to be consistent with sound velocity measurements then the excess specific heat of Ref. 4 has a Schottky-like temperature dependence.

[11] M. F. Panczyk and E. D. Adams, Phys. Rev. 187, 321 (1969).

[12] R. C. Richardson, E. Hunt, and H. Meyer, Phys. Rev. 138, A1326 (1965).

[13] D. S. Greywall, Phys. Rev. B 11, 1070 (1975), and Phys. Rev. B 13, 1065 (1976).

[14] W. R. Gardner, J. K. Hoffer, and N. E. Phillips, Phys. Rev. A 7, 1029 (1973).

New Mechanism for a Phonon Anomaly and Lattice Distortion in Quasi One-Dimensional Conductors*

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It is shown that for sufficiently strong repulsive electron-electron interactions a quasi one-dimensional conductor has a phonon anomaly with wave vector component $4k_F$ and an associated phase transition to a new kind of correlated charge-density wave state and lattice distortion, which can account for recent x-ray experiments on tetrathiafulvalene-tetracyanoquinodimethane. In the ordered state, the charge-density excitations are solitons and there is a gap in their spectrum.

In this Letter, it is shown that there is a new kind of correlated electron-phonon state in quasi one-dimensional conductors which may explain the $4k_F$ phonon anomaly discovered by Pouget *et al.*[1] by x-ray scattering from tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ). These