# Low-Temperature Heat Capacity of Liquid He<sup>3</sup><sup>†</sup>

W. R. ABEL, A. C. ANDERSON, W. C. BLACK, AND J. C. WHEATLEY Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois (Received 27 January 1966)

The heat capacity of pure He<sup>3</sup> has been measured using a difference method from 6 to 50 m°K at 0.28 atm and from 4 to 30 m°K at 27.0 atm. In neither case is the ratio of heat capacity to temperature constant over the range of temperature of the measurements. Moreover, the high- and low-pressure heat capacities seem to have qualitatively different temperature dependences. The raw heat-capacity data down to a temperature [on the magnetic temperature scale valid for powdered cerium magnesium nitrate (CMN) in the form of a right circular cylinder with diameter equal to height] of 2 m°K at 0.28 atm and 4 m°K at 27.0 atm show no evidence for anomalous behavior. As a by-product of the measurements the heat capacity of the CMN cooling salt was also obtained.

#### I. INTRODUCTION

CEVERAL years ago the heat capacity of He<sup>3</sup> was D measured by Brewer, Daunt, and Sreedhar; Brewer and Keyston<sup>1</sup>; Strongin, Zimmerman, and Fairbank<sup>2</sup>; and by Anderson, Reese, and Wheatley,3 the lowest temperature work being that of the latter authors. The low-temperature heat-capacity data of Refs. 1 and 2, at low pressures, could be interpreted in terms of a limiting value of  $C/n_3RT$ , where  $C/n_3$  is the molar heat capacity of He<sup>3</sup>, of between 2.2 and 2.4°K<sup>-1</sup> and would, furthermore, be in agreement with a parabolic temperature dependence of this quantity. On the other hand it was found that the low-pressure and low-temperature experimental data of Ref. 3 could best be fitted by a temperature dependence of the form  $C/n_3RT = \alpha + \beta T$ , the value of  $\alpha$  being about 2.9°K<sup>-1</sup>. Since the heat capacity of Ref. 3 did not obey the temperature dependence either of a Fermi gas<sup>4</sup> or, according to the calculations of Richards,<sup>5</sup> of the finite-temperature Fermi liquid, they were not generally accepted. However, in recent work of Abel, Anderson, Black, and Wheatley<sup>6</sup> certain aspects of the heat-capacity measurements were repeated, and it was found that the discrepancy with the work of Refs. 1 and 2 was enhanced. All measurements before the present ones of the heat capacity of He<sup>3</sup> by the Illinois group are probably in error to some extent due to the fact that the "background" heat capacity of the calorimeter was not precisely known. This background was probably due to the powdered cerium magnesium nitrate (CMN) thermometer, which was mixed with the liquid He<sup>3</sup> in order to provide good thermal contact, and to the wire constituting the

<sup>5</sup> P. M. Richards, Phys. Rev. 132, 1867 (1963).

electrical heater,<sup>7</sup> which was also located in the He<sup>3</sup>. The measured heat capacity could only be corrected approximately for the effects of "background" since the background was not measured directly.

In view of the repetition in our recent experiment<sup>6</sup> of essentially the earlier results obtained in the laboratory, we decided to perform an experiment in which the influence of the calorimeter background heat capacity could be eliminated and the heat capacity of pure He<sup>3</sup> could be obtained. All that is required is that the heat capacity be measured in two separate measurements with the same thermometer and heater but with differing amounts of He<sup>3</sup> in the cell. In order that the contact between the He<sup>3</sup>, the heater, and the thermometer be exactly the same in the two different experiments, we decided to use a single cell which could be modified by adding an extra volume, for He<sup>3</sup> only, between one experiment and the next. It is also possible to obtain the heat capacity of pure He<sup>3</sup> under pressure using this method. This may be done by taking the difference between two heat-capacity measurements using the same cell, so that the background heat capacity cancels out, one measurement being at high pressure and the other at low pressure. The difference gives immediately the added heat capacity of pure He<sup>3</sup> due to applying pressure. It may be combined with the measurements suggested above, used to obtain the heat capacity of pure He<sup>3</sup> at low pressure, to obtain the heat capacity of pure He<sup>3</sup> at high pressure. These measurements have been carried out and are described in the present paper. A preliminary account of this work has already been given.8

P. W. Anderson<sup>9</sup> found that by plotting the older results for  $C/n_3RT$  versus  $\ln T$  he obtained a rather straight line from the lowest temperatures to over  $0.1^{\circ}$ K. This suggested that the heat capacity might be of the form  $C/n_3RT = A \ln(B/T)$ . He argued that, if this were so, perhaps He<sup>3</sup> is not a normal Fermi liquid. If He<sup>3</sup> were not a Fermi liquid then it would be neces-

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<sup>&</sup>lt;sup>1</sup> D. F. Brewer, J. G. Daunt, and A. K. Sreedhar, Phys. Rev. **115**, 836 (1959); D. F. Brewer and J. R. G. Keyston, Nature **191**, 1261 (1961).

<sup>&</sup>lt;sup>2</sup> M. Strongin, G. O. Zimmerman, and H. A. Fairbank, Phys. Rev. **128**, 1983 (1962).

<sup>&</sup>lt;sup>a</sup> A. C. Anderson, W. Reese, and J. C. Wheatley, Phys. Rev. **130**, 495 (1963).

<sup>&</sup>lt;sup>4</sup> E. C. Stoner, Phil. Mag. 21, 145 (1936).

 $<sup>^{6}</sup>$  W. R. Abel, A. C. Anderson, W. C. Black, and J. C. Wheatley, Physics 1, 337 (1965).

<sup>&</sup>lt;sup>7</sup> J. C. Ho, H. R. O'Neal, and N. E. Phillips, Rev. Sci. Instr. 34, 782 (1963).

<sup>&</sup>lt;sup>8</sup> W. R. Abel, A. C. Anderson, W. C. Black, and J. C. Wheatley, Phys. Rev. Letters **15**, 875 (1965). <sup>9</sup> P. W. Anderson, Physics **2**, 1 (1965).

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sary to re-examine various predicted properties for liquid He<sup>3</sup>, in particular the existence of a transition into a superfluid phase. Anderson's work stimulated a great deal of interest in this problem among theoretical physicists. In particular, Balian and Fredkin<sup>10</sup> have made a theory based on an assumed long-range interaction He<sup>3</sup> atoms and zero-sound phonons which leads to the expression  $C/n_3RT = A[\ln(B/T)]^{1/2}$ .

### **II. EXPERIMENTAL**

The cell used for the measurements is shown in Fig. 1. It is made principally from Epibond 100A.<sup>11</sup> The CMN is confined to a cylindrical region with diameter equal to height. Thermal contact with the He<sup>3</sup> in this cell is made by means of one thousand 0.08-mm diam (No. 40 AWG) Formex insulated copper wires arranged uniformly on a cylindrical surface somewhat larger than the bore of the cavity containing the He<sup>3</sup>. The general method of fabrication and support of this cell is given in Abel, Anderson, Black, and Wheatley,<sup>6</sup> who also describe how the above copper wires are connected to a zinc thermal switch of dimensions 20 mm long by 10 mm by 0.2 mm. The CMN thermometer is contained in a right circular cylindrical cavity with both diameter and height equal to 18.54 mm. Packed into this cavity

with the CMN is a helically wound, bifilar heater of 0.05-mm-diam Evanohm wire about 15-cm long. The ends of the heater were hard-soldered to lengths of 0.125-mm-diam Formex insulated copper wire. These copper wires were sealed through the wall with Epibond 121<sup>11</sup> and then soldered to pairs of 0.05-mm-diam leadcovered manganin wires used in a standard four-wire heater arrangement. The resistance of the copper wire was negligible at low temperatures compared with that of the Evanohm heater, which had a resistance of 94.4  $\Omega$  at low temperatures. The He<sup>3</sup> was introduced into the cell by a  $\frac{1}{64}$ -in.-o.d. by 0.003-in.-wall 70-30 cupronickel tube with a small cotton wool filter over the end inside the cell to prevent its being clogged with CMN particles. The CMN consisted of powdered single crystals grown using high-purity cerous nitrate from the Lindsay Chemical Company. All the powder which would pass through an NBS 40 sieve was accepted for packing into the cell. This powder consisted of grains ranging downward in size from about  $300-400 \mu$  and averaging about  $100-200 \mu$ . It was packed into the cell with a hand-held Teflon rod using some force. The total amount of CMN in the cell was 6.49 g which therefore, taking  $\rho = 2.0$  g/cm<sup>3</sup> for CMN, occupied 0.65 of the volume allocated to it, a greater fraction than in any previous heat capacity cell constructed by us. The CMN and heater arrangement was maintained in position by a cotton cloth filter<sup>12</sup> backed by an Epibond 100A filter support. The latter contained a square array of 45 holes which were 1.59-mm diam on 2.29-mm centers. The filter and support were sealed into the main cell at the place shown, using Epibond 121 only on the cylindrical edges. After this seal had cured, the cell was mounted in a lathe and a suitable counterbore made to leave the bottom of the support accurately perpendicular to the cell axis. The distance D was measured with precision. Assembly of the cell was completed by then sealing in a solid end plug, using Epibond 121 only along the cylindrical sides. The free volume of the cell at room temperature was about 1.93 cm<sup>3</sup>.

Upon completion of the first series of measurements with this cell, it was disassembled from the demagnetization apparatus and returned to the lathe. Using the known dimension D as a guide, it was possible to remove, by boring, all of the solid end plug without removing any of the filter support or in any way affecting the contents of the cell. The cell was then fitted with a new end plug having a cavity in one end of dimensions 5.59-mm deep and 18.54-mm diam. This end plug was sealed into the cell using Epibond 121 along its cylindrical surface only, thus completing reassembly of the cell.

The number of moles of He<sup>3</sup> contained in each of the two modifications of the cell was determined in separate experiments using a small test Dewar. The determinations were made by measuring the amount of gas which

<sup>&</sup>lt;sup>10</sup> R. Balian and D. R. Fredkin, Phys. Rev. Letters 15, 480 (1965). <sup>11</sup> Furane Plastics, Los Angeles, California.

<sup>&</sup>lt;sup>12</sup> Cut from a piece of white cotton broadcloth.

evaporated from the cell and filling tube, initially at 1.2°K and a pressure of approximately 50-cm Hg and with a well-defined height of liquid He<sup>4</sup> in the test Dewar, on increasing the temperature of these parts to room temperature. The volume of He<sup>3</sup> in the filling tube was determined in a separate experiment to be 3.4% of the total volume in the case of the cell without the extra He<sup>3</sup> volume and to be 2.2% in the case of the cell with the extra volume. The actual measurements of the amounts of gas evaporated were made using a constantvolume mercury manometer, a glass Toepler pump with accurately known volume as a function of height, a 2095-cm3 dead volume, and a mercury-in-glass thermometer. The numbers of moles in the cell at low temperatures and at 0.28 atm and 27.0 atm were obtained from the raw data using thermal expansion and compressibility corrections based on the experimental data of Sherman and Edeskuty<sup>13</sup> and of Boghosian, Meyer, and Rives.<sup>14</sup> The cell without the extra volume contained 0.0540 moles at 0.28 atm and 0.0742 moles at 27.0 atm with an accuracy of about 1 to 3%. This corresponds to a free volume of 1.97 cm<sup>3</sup>, about 2%greater than the free volume estimated above. The cell with the extra volume contained 0.0952 moles at 0.28 atm, from which by subtraction one finds that at 0.28 atm the number of moles in the extra volume is  $\Delta n$ =0.0412 moles. This corresponds to a volume of 1.504 cm<sup>3</sup>. From the geometrical measurements of the extra volume at room temperature one finds V = 1.510cm<sup>3</sup>. However, a 3% decrease in volume might be expected at low temperature due to the contraction of the epoxy, leading to an expected low temperature volume of 1.465 cm<sup>3</sup>. The "measured" volume of 1.504 cm<sup>3</sup>, close to the room temperature value, was, therefore, somewhat surprising. However, in further treatment of the heat-capacity data it was found that the calorimeter "background" heat capacity, which could be derived from the measurements, approached zero in a reasonable manner at high temperatures only if one assumed that the measured mole difference  $\Delta n$ was about 1.8% too high, an error not unreasonable on the basis of the errors in the measurements from which  $\Delta n$  was derived. If  $\Delta n$  were corrected accordingly, as it was in deriving values for the calorimeter "background" heat capacity, the resulting extra volume would be 1.478 cm<sup>3</sup>, in more reasonable agreement with the expected value. In any case the geometrical measurements are consistent with the mole measurements within 2 to 3%. Moreover, an error in  $\Delta n$  will not change the shape of the final heat-capacity curve.

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The temperature of the He<sup>3</sup> was determined by measuring with a 17-cps Cryotronics<sup>15</sup> mutual inductance bridge the magnetic susceptibility of the CMN powder in contact with the He<sup>3</sup>. The temperature scale was calibrated by means of the He<sup>4</sup> vapor-pressure scale between 1.1 and 2.2°K. The calibration constant was accurate to a few tenths of a percent. After disassembly and reassembly of the apparatus for the purpose of changing the end plug of the cell it was found that the calibration constant had shifted only 0.1%.

The general features of the surroundings of the heat capacity cell are the same as those illustrated in Fig. 3 of Ref. 6 except that there is no CMN guard as in that figure. The chromium potassium alum refrigerator contains about 170 g of salt. The general techniques and parameters for the adiabatic demagnetization are also given in Ref. 6.

In the present measurements the inductive component of the bridge was measured frequently as time progressed after final demagnetization. When dynamic equilibrium was reached under the action of the residual heat leak (about 1.5 erg/min), heater power was applied for an interval of time in excess of 30 sec, measured to within 0.1 sec by means of an Anadex model CF-200R counter-timer.<sup>16</sup> During the heating period only the current through the heater was measured in order to avoid possible unknown heating by the potentiometer during the balancing procedure necessary for measuring the potential. The resistance of the heater was measured at a different time. During and after the heating period, bridge readings were taken for a sufficient length of time to assure that the cell's temperature was again drifting under the action only of the heat leak. Then heat was applied again, and so on. At low temperatures the thermal equilibrium was quite rapid, as we shall indicate quantitatively later. However, at higher temperatures the heating curves developed a small overshoot, due to the He<sup>3</sup> located outside the region filled with CMN powder. The time constants of the recovery increased with increasing temperature as a result of the increasing heat capacity and the decreasing thermal conductivity of the He3. At 0.28 atm and 50 m°K  $(1 \text{ m}^{\circ}\text{K} \equiv 10^{-3} \text{ }^{\circ}\text{K})$  the equilibrium times were several thousand seconds in the cell without extra volume and several hours in the cell with extra volume. However, from the results of the measurements, it is clear that the background heat capacity is negligibly small at 50 m°K and above, so that in future experiments in that temperature range no provision for an extra volume need be made if an experimental apparatus similar to the present one is used.

### **III. TREATMENT OF EXPERIMENTAL DATA**

Problems of calorimetry have already been discussed in some detail in Ref. 6, particularly in the discussion of Fig. 10 of that paper. What one would like to obtain is the temperature change  $\Delta T$  due to adding heat Q at the average temperature  $\overline{T}$ . To obtain  $\Delta T$  one might

<sup>&</sup>lt;sup>13</sup> R. H. Sherman and F. J. Edeskuty, Ann. Phys. (N. Y.) 9, 522

 <sup>(1960).
 &</sup>lt;sup>14</sup> C. Boghosian, H. Meyer, and J. E. Rives, Phys. Rev. 146, 110 (1966). We are indebted to Professor Meyer for sending us these results prior to publication.

<sup>&</sup>lt;sup>15</sup> Cryotronics, Inc., West Main Street, High Bridge, New Tersev.

<sup>&</sup>lt;sup>16</sup> Anadex Instruments, Inc., Van Nuys, California.



FIG. 2. Raw heat-capacity data for the cell without the extra volume.

plot T as a function of time, including an adequate interval of dynamic equilibrium under the action of the heat leak both before and after the heating period. However, it frequently happens that the fore and after intervals of T versus time are not straight lines, in which case  $\Delta T$  is uncertain. The "equal-area" rule for determining  $\Delta T$  is not applicable since the CMN temperature does not necessarily have any simple relation to the He<sup>3</sup> temperature, which may itself be inhomogeneous.

Let us assume that during the fore and after intervals and the heating period, the residual heat leak  $\dot{Q}_r$  is constant. Then there is one case, that of a constant specific heat, in which the dynamic-equilibrium parts of the fore and after intervals of T versus time are parallel straight lines. If this is not the case, let us suppose that in the small temperature interval considered the heat capacity C is approximated by  $C=bT^n$ , where b and nare constants. Then

$$\dot{Q}_r = C\dot{T} = \frac{b}{n+1} \frac{d}{dt} (T^{n+1}); \quad (n \neq -1).$$
 (1)

If now one plots the experimental data as  $T^{n+1}$  versus time, one should find that the fore and after dynamic equilibrium intervals are represented by parallel straight lines. This allows an unambiguous extrapolation through the interval of time encompassing the heating period and subsequent return to dynamic equilibrium.  $\overline{T}$  and  $\Delta T$  are obtained from these extrapolated lines at a time on the plot near the middle of the total temperature change. The possible error introduced by the selection of this time is small if  $\Delta T/T \ll 1$ . In the particular case of a linear specific heat (nearly like the He<sup>3</sup> itself) one would plot  $T^2$  versus time and find  $\Delta(T^2)$ . The quantity which we plot is C/T, which is just

$$C/\bar{T} = Q/\bar{T}\Delta T = Q/(\frac{1}{2})\Delta(T^2), \qquad (2)$$

so that in this case the present method of data treatment leads to exactly the quantity desired. At low temperatures we used n=-2, at temperatures near the minimum in the heat capacity n=0, and at higher temperatures n=1.

The above procedure was used to reduce the data, different values of n being tried when the best procedure was not obvious from a rough calculation of the heat capacity. The scatter of the data was considerably reduced, the general quality being improved.

At higher temperatures the T-versus-time curves developed a slight overshoot and thermal relaxation times increased as noted above. This was more of a nuisance than a source of error in data analysis since in the temperature region involved the total changes in temperature even during the long relaxation times were small compared with the temperature change produced by the heat added electrically.

#### IV. RESULTS AND DISCUSSION

The raw heat-capacity data for the cell without the extra volume are shown on Fig. 2. The increase in heat capacity at low temperatures is due to the CMN and possibly the heater. The lowest temperature possible for good-quality data was about 2 m°K at 0.28 atm and  $4 \text{ m}^{\circ}\text{K}$  at 27.0 atm on the  $T^*$  scale. These temperatures were different because the added heat capacity of the He<sup>3</sup> at 27.0 atm did not allow it to be cooled as much as the 0.28 atm He<sup>3</sup>. In spite of the low temperature of the measurements there is little scatter in the experimental data, treated as indicated in Sec. III, primarily because of the small ( $\sim 1.5$  erg/min) heat leak and the small thermal time constants, which have been discussed in a separate publication.<sup>17</sup> It is evident that at neither pressure is there any departure from a smoothly varying dependence on the temperature, as might occur in a transition to the superfluid state. However, at 2 m°K the heat capacity of the He<sup>3</sup> is about 28 erg/m<sup>°</sup>K, or only 12% of the total, so that small changes would have been difficult to detect. This illustrates the impractica-

<sup>17</sup> W. R. Abel, A. C. Anderson, W. C. Black, and J. C. Wheatley, Phys. Rev. Letters 16, 273 (1966).



FIG. 3. Processed heat-capacity data divided by  $n_3RT^*$ .

bility of the present method for searching for a possible transition to the superfluid phase in He<sup>3</sup> much below the present temperatures. The He4: He3 ratio in the gas at room temperature was 10<sup>-5</sup>. It is expected that because of the phase-separation effect<sup>18</sup> the purity will be even higher at the low temperatures of the measurements.

Reduced heat-capacity data from all the measurements are shown in Fig. 3. The data for this figure were obtained as follows. In the case of the "small" cell, or cell without extra volume, one measured

- $C_s(0.28)$ , the heat capacity at 0.28 atm;
- $n_s(0.28)$ , the number of moles of He<sup>3</sup> in the cell at 0.28 atm;
- $C_s(27.0)$ , the heat capacity at 27.0 atm.

From the data of Sherman and Edeskuty13 and Boghosian, Meyer, and Rives<sup>14</sup> one can obtain  $n_s(27.0)/$  $n_s(0.28)$ , where  $n_s(27.0)$  is the number of moles of He<sup>3</sup> in the small cell at 27.0 atm, assuming no change in volume of the cell or CMN on applying pressure, a reasonable assumption for the present cell which had walls 5 mm thick. In the case of the "large" cell, or cell with extra volume, one measured

 $C_l(0.28)$ , the heat capacity at 0.28 atm;

 $n_l(0.28)$ , the number of moles in the cell at 0.28 atm.

If the  $C_b$  is the "background" heat capacity of the cell, presumably primarily due to the CMN, then one has

$$C_s(0.28) = C_b + n_s(0.28)c_3(0.28), \qquad (3)$$

$$C_{l}(0.28) = C_{b} + n_{l}(0.28)c_{3}(0.28), \qquad (4)$$

<sup>&</sup>lt;sup>18</sup> D. O. Edwards and J. G. Daunt, Phys. Rev. 124, 640 (1961).



FIG. 4. Plot of  $(C/n_3RT^*)^2$  versus  $\ln T^*$  for a pressure of 0.28 atm.

and

$$[C_{l}(0.28) - C_{s}(0.28)] / [n_{l}(0.28) - n_{s}(0.28)] = c_{3}(0.28),$$
(5)

where  $c_3(0.28)$  is the molar heat capacity of He<sup>3</sup> only at 0.28 atm. On Fig. 3 have been plotted  $C_s(0.28)/n_s(0.28)RT^*$ ,  $C_l(0.28)/n_l(0.28)RT^*$ , and

$$[C_l(0.28) - C_s(0.28)] / [n_l(0.28) - n_s(0.28)] RT^*,$$

where  $C \equiv Q/\Delta T^*$ . The quantities C/n have been divided by the gas constant R to render them dimensionless and by  $T^*$  to remove most of the temperature dependence. At temperatures for which  $C_b$  is negligible all three of these should reduce to  $c_3/RT$  in the absence of systematic errors in, for example, the temperature scale and the mole determination. This is, evidently, very nearly so for temperatures above about 30 m°K, though as indicated earlier close examination suggests that the measured value of  $n_l(0.28) - n_s(0.28)$  may be 1.8% too high. At low temperatures the nondifference data rise above the difference data as  $C_b$  becomes non-negligible.

The molar heat capacity of He<sup>3</sup> at 27.0 atm,  $c_3(27.0)$ , may also be obtained from the experimental data indicated above. The background heat capacity is eliminated by taking the difference

$$C_s(27.0) - C_s(0.28) = n_s(27.0)c_3(27.0) - n_s(0.28)c_3(0.28).$$
(6)

Solving this equation for  $c_3(27.0)$ , one finds

$$\frac{c_{3}(27.0)}{RT^{*}} = \frac{C_{s}(27.0) - C_{s}(0.28)}{RT^{*}n_{s}(0.28)[n_{s}(27.0)/n_{s}(0.28)]} + \frac{n_{s}(0.28)}{n_{s}(27.0)}\frac{c_{3}(0.28)}{RT^{*}}.$$
 (7)

P=0.28 atm				P = 27.0  atm	
T*	C/nRT*	T*	C/nRT*	T*	C/nRT*
(10 <sup>3</sup> °K)	(°K <sup>-1</sup> )	(10 <sup>-3</sup> °K)	(°K <sup>-1</sup> )	(10 <sup>-3</sup> °K)	(°K <sup>-1</sup> )
6.07	2.965	18.37	2.875	4.068	4.460
6.91	2.910	18.89	2.798	4.584	4.499
7.31	2.897	19.36	2.855	5.208	4.465
7.70	2.885	20.67	2.847	5.997	4.612
7.99	2.896	22.21	2.803	6.902	4.435
8.54	3.031	22.33	2.840	8.021	4.449
9.10	2.956	24.19	2.850	9.510	4.420
9.35	2.973	25.69	2.800	11.150	4.402
9.91	3.014	26.28	2.774	13.149	4.333
10.53	2.984	28.08	2.792	16.047	4.269
10.80	2.971	29.35	2.771	19.754	4.184
11.52	2.974	30.71	2.769	23.890	4.096
12.31	2.881	32.60	2.806	28.856	3.984
12.42	2.902	33.66	2.807	35.537	3.836
13.83	2.908	36.15	2.704		
14.36	2.891	37.79	2.741		
14.91	2.872	39.99	2.731		
15.79	2.878	42.19	2.633		
16.80	2.884	50.60	2.692		
17.67	2.857	51.00	2.658		

TABLE I.  $C/nRT^*$  for pure He<sup>3</sup> at two pressures.

For  $c_3(0.28)/RT^*$  one uses the smoothed values of the difference data at 0.28 atm shown on Fig. 3. The smoothed curve was extrapolated to 4 m°K to allow values of  $c_3(27.0)/RT^*$  to be derived from the values of  $C_s(27.0)-C_s(0.28)$  which exist down to that temperature.

As is indicated on Fig. 3 the best fits to the experimental values of  $c_3/RT^*$  versus  $T^*$  are, on that plot, a curve slightly concave upward at 0.28 atm and a straight line at 27.0 atm. At low temperatures the scatter increases because the He<sup>3</sup> heat capacity is a decreasing fraction of the total heat capacity. The extrapolations of these lines to  $T=0^{\circ}$ K lead to limiting values for  $C_3/n_3RT^*$  of 3.05 K<sup>o-1</sup> and 4.62 °K<sup>-1</sup> and for



FIG. 5. Plot of  $(C/n_3RT^*)^2$  versus  $\ln T^*$  for a pressure of 27.0 atm.



FIG. 6. Collected measurements of  $(C/n_3RT^*)$  for various authors, including the present work. The results of Brewer and Keyston were normalized to agree with those of Brewer, Daunt, and Sreedhar (Ref. 1) above  $0.12^{\circ}$ K.

and

 $m^*/m$  of 3.08 and 5.78 at 0.28 atm and 27.0 atm, respectively.

According to Balian and Fredkin<sup>10</sup> the limiting temperature dependence of the specific heat should be  $C/T = A[\ln(B/T)]^{1/2}$ . In Figs. 4 and 5 are plotted  $(C/nRT^*)^2$  versus  $\ln T^*$ . Within experimental scatter the 0.28-atm data do fit a straight line on this plot, though they do not define such a line. However, it is evident that the 27.0-atm data definitely do not fit a straight line on this plot. This was to be expected since they fit well a straight line on a linear plot.

We wish to emphasize that because of the scatter of the experimental data, we do not claim to have established the limiting temperature dependence of the heat capacity of He<sup>3</sup>. However, it does seem clear that  $c_3/T^*$ is continuing to rise as  $T^*$  falls, over the temperature range of the measurements. Table I gives a listing of the experimental data for the purpose of allowing others to work with it.

It may be argued that in spite of the general consistency of the present measurements they are not correct because the magnetic temperature  $T^*$  is not the same as the Kelvin temperature T, as has been assumed throughout, and that, in fact,  $c_3/T$  is a constant over the temperature range of the present measurements. To examine this argument, let us suppose that, in fact,

$$c_3(0.28) = \gamma_{0.28} T, \qquad (8)$$

$$c_3(27.0) = \gamma_{27.0}T, \qquad (9)$$

where  $\gamma$  is a constant and where the relationship between  $T^*$  and T is

$$T^* = Tf(T), \tag{10}$$

f(T) being some function approaching 1 at higher temperatures. The measured and plotted parameter is

$$C/T^* = Q/T^* \Delta T^*. \tag{11}$$

From the definition of  $T^*$  above, this quantity is

$$\frac{C}{T^*} = \frac{1}{f(T)[f(T) + Tf'(T)]} \left(\frac{Q}{T\Delta T}\right).$$
(12)



FIG. 7. Heat capacity of the calorimeter "background" divided by the number of moles of CMN,  $n_{\text{CMN}}$ , and the gas constant R.

Since  $Q/T\Delta T$  is proportional to  $\gamma$  introduced in Eq. (8) and Eq. (9) above, one has

$$[C(0.28)/T^*]/[C(27.0)/T^*] = \gamma_{0.28}/\gamma_{27.0} = \text{const.} \quad (13)$$

The experimental heat capacities do not obey this relation, however. Moreover, if f(T) in Eq. (10) were not unity in most of the present temperature range, then the diffusion coefficient in He<sup>3</sup> would not be proportional to  $T^{-2}$  since it has been found<sup>6,19</sup> that it is proportional to  $T^{*-2}$ . Actually the thermodynamic measurements on CMN certainly suggest  $T^* = T$  down to 10 m°K<sup>20</sup> and probably below.<sup>21</sup> So far as the relationship between  $T^*$  and T in CMN is concerned, recent measurements in this laboratory<sup>8,17,22,23</sup> have shown that the lowest magnetic temperature of a magnetic thermometer of powdered CMN in the form of a right circular cylinder with diameter equal to height is very different from that for a spherical single crystal. The latter consistently<sup>24-26</sup> leads to a minimum magnetic temperature of about 3.2 m°K. Our powdered CMN

- <sup>22</sup> A. C. Anderson, W. R. Roach, R. D. Sarwinski, and J. C.
   <sup>23</sup> A. C. Anderson, D. O. Edwards, W. R. Roach, R. E. Sarwinski, and J. C. Wheatley (unpublished).
   <sup>24</sup> J. M. Daniels and F. N. H. Robinson, Phil. Mag. 44, 630
- (1953). <sup>25</sup> F. Carboni and R. C. Sapp, Ann. Phys. (N. Y.) 33, 77 (1965).
- <sup>26</sup> R. B. Frankel, D. A. Shirley, and N. J. Stone, Phys. Rev. 140, A1020 (1965).

thermometers consistently<sup>22,23</sup> indicate a lowest magnetic temperature of about 1.8 m°K. Moreover, the temperature determined<sup>23</sup> from the diffusion coefficient of dilute solutions of He<sup>3</sup> in He<sup>4</sup> differs by at most a few tenths of a millidegree from the magnetic temperature, even at 2 m°K. Hence we conclude that temperature scales such as that of Ref. 26 are inapplicable to the present work. Moreover, it seems very likely that in the region of temperature above 6 m°K, corresponding to the heat capacity data of Table I, the magnetic and Kelvin temperatures are equal to one another.

A comparison between the present results and our earlier results<sup>3</sup> as well as those of other workers<sup>1,2</sup> is given in Fig. 6. The high-pressure results of the previous data have been corrected to a common pressure of 27.0 atm using the formula

$$(1/V_m T)(\partial C_p/\partial p)_T = -\alpha^2 - (\partial \alpha/\partial T)_p, \quad (14)$$

where  $V_m$  is the molar volume,  $C_p$  the molar specific heat, and  $\alpha$  the expansion coefficient. The expansion coefficient data of Anderson, Reese, and Wheatley<sup>3</sup> have been used for this purpose. The corrections are small. Taken as a whole the data of Fig. 6 represent a consistent picture. At a given pressure the different experimental results are shifted with respect to one another, but this is to be expected due to calibrational errors of the temperature scale and of the number of moles of He<sup>3</sup> in the heat-capacity cell. Near the low-temperature end of any given set of measurements the scatter increases and even smoothed data are not reliable. This is not surprising since the lowest temperature of the measurements is usually determined by the failure of the experiment to work satisfactorily. The general picture is one of a steadily increasing value of  $C/nRT^*$ as the temperature decreases. It is significant to observe that in the present work the quantity of CMN is eight times greater than in Ref. 3 while the quantity of He<sup>3</sup> was nine times greater for the small cell and sixteen times greater for the large cell. These increases serve to improve the accuracy of the present work relative to that in Ref. 3.

It is also possible to obtain the heat capacity of the "background" from the experimental data once a good estimation of  $c_3$  is made. In terms of experimental quantities one has, for example,

$$C_b = C_s(0.28) - n_3(0.28)c_3(0.28).$$
 (15)

In addition to the CMN the only probable source of calorimeter background is the Evanohm heater. If this heater were manganin, its heat capacity<sup>7</sup> would probably not be in excess of  $(3 \text{ erg/m}^{\circ}\text{K})$   $(10 \text{ m}^{\circ}\text{K/T})^2$  while the actual background is about ten times this figure. Hence, it seems reasonable to assume that  $C_b$  represents principally the 0.00849 moles of CMN in the cell and to treat it as such. The results for  $C_b/n_{\rm CMN}R$  are given in Fig. 7. Above 7 or 8 m°K the data follow a  $T^{*-2}$  law within experimental scatter, the value of  $CT^{*2}/nR$ 

<sup>&</sup>lt;sup>19</sup> A. C. Anderson, W. Reese, and J. C. Wheatley, Phys. Rev. 127, 671 (1962).

<sup>&</sup>lt;sup>20</sup> R. P. Hudson, in Temperature, Its Measurement and Control <sup>11</sup> Science and Industry, edited by F. Brickwedde (Reinhold Publishing Corporation, New York, 1962), Vol. 3, Pt. I, p. 51.
 <sup>21</sup> D. deKlerk, Handbuch der Physik, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 15, p. 117.
 <sup>22</sup> A. C. Anderson, W. R. Roach, R. E. Sarwinski, and J. C. Wheatley, Phys. Part Letters 16, 263 (1966).

being  $4.2 \times 10^{-6}$  K<sup>o2</sup>. This figure is even lower than the 5.4×10<sup>-6</sup> °K<sup>2</sup> mentioned by Hudson, Kaeser, and Radford<sup>27</sup> as the lowest value of  $CT^2/nR$  observed by them. Inclusion of the heat capacity of the heater can only lower the present value. If  $CT^2/nR$  for CMN is, in fact, considerably less than was originally supposed,<sup>24</sup>

27 R. P. Hudson, R. S. Kaeser, and H. E. Radford, in Proceedings of the Scientif International Conference on Low Temperature Physics, edited by G. M. Graham and A. C. Hollis Hallet (University of Toronto Press, Toronto, 1961), p. 100.

then it may not be surprising that lower temperatures with CMN are being obtained<sup>20,25-27</sup> than were obtained in the early work on this substance. If one simply assumes that  $T^* = T$  down to 2 m°K, one finds from the data on Fig. 7 that at 2 m°K, S/nR=0.25, where at high T one has S/nR = 0.693, S/n being the molar entropy. The molar entropy calculated above is undoubtedly in error, but it does encourage the speculation that considerably lower temperatures may still be obtained with this salt.

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# Theory of a Superfluid Fermi Liquid. II. Collective Oscillations\*

A. J. Leggett<sup>†</sup>

Department of Physics, University of Illinois, Urbana, Illinois (Received 3 February 1966)

The long-wavelength, low-frequency "collisionless" collective oscillations of a superfluid Fermi liquid are studied throughout the region  $0 < T < T_e$  with the help of a field-theoretic formalism developed in a previous paper. It is found that any collective mode (other than the density fluctuation) that is possible in the normal phase persists below  $T_c$  as an oscillation of the normal component, but disappears at some finite temperature which in general depends on the Landau interaction parameters  $F_0$ ,  $F_1$ ,  $Z_0$ , etc. The behavior of the densityfluctuation mode depends critically on  $F_0$ ; if the latter is negative, there is a well-defined collective oscillation only for  $T \ll T_c$ , while if it is small and positive, the oscillation is well defined for  $T \ll T_c$  and  $T - T_c \ll T_c$ but not in the intermediate region. If  $F_0$  is large and positive, the density-fluctuation collective mode exists throughout the whole region  $0 < T < T_e$  and its velocity is approximately temperature-independent.

## I. INTRODUCTION

WELL-KNOWN result of Landau's<sup>1-3</sup> theory of a  $oldsymbol{A}$  normal Fermi liquid is that various kinds of phonon-like<sup>4</sup> collective oscillations may be propagated in the liquid in the "collisionless" region.<sup>5</sup> In particular, if we define the quasiparticle interaction function f on the Fermi surface in the usual way and expand it in Legendre polynomials :

$$F(\mathbf{p},\mathbf{p}':\sigma,\sigma') \equiv \nu(0) f(pp',\sigma\sigma')$$
  
=  $\sum_{l} (F_{l} + Z_{l} \boldsymbol{\sigma} \cdot \boldsymbol{\sigma}') P_{l}(\cos\theta), \quad (1)$ 

[where  $\nu(0)$  is the density of states at the Fermi surface] then it follows from the Landau theory that there is a "collective" pole in the density autocorrelation function if  $F_0$ ,  $F_1 > 0$  and, in addition, a pole in the transverse-current autocorrelation function if  $F_1 > 6$ . (We assume for simplicity, as is usually done, that all  $F_1$  can be neglected for  $l \ge 2$ .) Similarly a pole appears in the spin-density autocorrelation function if  $Z_0>0$ , and so on. Oscillations of this type may be visualized as a coherent distortion of the Fermi surface, and are usually called zero sound; in the limit as the Landau parameters tend to their critical values the velocity of the excitation tends asymptotically to the Fermi velocity  $v_F$ .

In a superfluid Fermi system at absolute zero the situation is quite different. [We shall always in this paper assume that we are interested in the limit of long wavelengths  $(v_F k \ll \Delta)$  and in the "phonon-like" excitations only.] It was shown by Bogolyubov<sup>7</sup> and by Anderson<sup>8</sup> that in a superfluid neutral Fermi gas at T=0 there exists a density-fluctuation mode whose velocity is  $3^{-1/2} v_F$ , which is identical to the speed of "hydrodynamic" sound.<sup>5</sup> This feature was subsequently shown by Larkin and Migdal<sup>9</sup> to be true also of a

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<sup>&</sup>lt;sup>†</sup> On leave of absence from Magdalen College, Oxford, England. <sup>1</sup>L. D. Landau, Zh. Eksperim. i Teor. Fiz. 30, 1058 (1956)

<sup>&</sup>lt;sup>1</sup>L. D. Landau, Zh. Eksperim. i Teor. Fiz. **30**, 1058 (1956) [English transl.: Soviet Phys.—JETP **3**, 920 (1957)]. <sup>2</sup>L. D. Landau, Zh. Eksperim. i Teor. Fiz. **32**, 59 (1957) [English transl.: Soviet Phys.—JETP **5**, 101 (1957)]. <sup>3</sup>L. D. Landau, Zh. Eksperim. i Teor. Fiz. **35**, 95 (1958) [English transl.: Soviet Phys.—JETP **8**, 70 (1959)]. <sup>4</sup> By "phonon-like" we mean in this paper "without an energy gap in the limit  $k \rightarrow 0$ ."

<sup>&</sup>lt;sup>6</sup> For the distinction between "collisionless" and "hydro-dynamic" regions see, e.g. D. Pines and P. Nozières, *Theory of Quantum Liquids* (W. A. Benjamin and Company, Inc., New York, 1965).

<sup>&</sup>lt;sup>6</sup> This is a sufficient but not, in general, a necessary condition. <sup>7</sup> N. N. Bogolyubov, V. V. Tolmachev, and D. N. Shirkov, New Method in the Theory of Superconductivity (English transl.: Consultants Bureau Enterprises, Inc., New York, 1959). <sup>8</sup> P. W. Anderson, Phys. Rev. 112, 1900 (1958). <sup>9</sup> A. I. Larkin and A. B. Migdal, Zh. Eksperim. i Teor. Fiz. 44, 1703 (1963) [English transl.: Soviet Phys.—JETP 17, 1146 (1963)].