Density, Coefficient of Thermal Expansion, and Entropy of Compression of Liquid Helium-3 under Pressure below 1.2°K*

CHARLES BOGHOSIAN, HORST MEYER, AND JOHN E. RIVEST Department of Physics, Duke University, Durham, North Carolina (Received 17 January 1966)

The method of dielectric-constant measurement has been used to determine the density of liquid He³ under pressure between 0.07 and 1.2°K. It consists of measuring the resonant frequency of a tunnel-diode oscillator, the LC tank circuit of which includes a capacitor filled with the liquid He3. The tunnel-diode oscillator is kept at low temperature and is described and analyzed in detail. A description is given of the system necessary to keep the pressure of the He³ constant. From the density measurements, the coefficient of thermal expansion and the entropy of compression are derived. The results are compared with earlier work. They are found to be thermodynamically consistent with the data obtained by other methods.

I. INTRODUCTION

CEVERAL years ago, Lee, Fairbank and Walker¹ \mathbf{J} determined the density of liquid He³ down to 0.16°K from measurements of the dielectric constant and the use of the Clausius-Mosotti relation. The liquid was contained in the cylindrical capacitor of an LC circuit, the frequency of which was measured. At the time this work was published, similar measurements² were being carried out at Duke University on 99.5% liquid He³ down to approximately 0.05°K. In the temperature range above 0.2°K, both determination of the coefficient of thermal expansion α_p were in reasonable agreement, but the Duke results were more consistent with those of Brewer and Daunt³ derived by the method of adiabatic expansion. Below 0.08°K, Rives and Meyer² found that the coefficient of expansion under 0.18 atm could be represented by the expression $\alpha_p = -0.12 T(^{\circ}K)^{-1}$. However, it was found that at pressures above 15 atm, the coefficient of expansion became unexpectedly positive below a certain temperature. In view of more recent determinations of the coefficient of expansion from adiabatic expansion,^{4,5} it appeared that the results of Rives and Meyer at high pressures and below about 0.15°K were spurious. It was speculated that the erroneous result could be partly due to the 0.5% He⁴ impurity in the liquid because a phase separation could possibly account qualitatively for the measured effect. It was decided to repeat the experiment with the purest He³ available and with an improved apparatus in order to obtain more accurate data. This is part of a systematic effort in this laboratory to measure the density, the coefficient of thermal expansion and to calculate the related thermodynamic properties for pure He³ and He⁴ under pressure and for their

110

solutions. The results on liquid He⁴ that were obtained, and the discussion of the results in terms of Landau's theory will be presented in a subsequent paper.^{5a}

The present account describes the most important parts of a new cryostat for measuring the density, the procedure for taking data and the analysis of the results. In Sec. 3, the new results on liquid He³ are presented and various thermodynamic quantities are calculated from the data. The results are then compared with relevant data obtained by other authors.

II. EXPERIMENTAL PART

Measurements of the density by means of the dielectric constant are of interest because they are able to detect smaller changes in density than the method used for example by Sherman and Edeskuty.⁶ The achieved sensitivity in our experiments is comparable to that of the dilatometric method used by Atkins and Edwards7 and by Kerr and Taylor.8 The advantage of the dielectric method over the dilatometric one is that it can be used for measurements under pressure and can be more easily extended to temperatures below 0.3°K. However the cell has to be calibrated first at a given temperature by means of the data of another method.

The apparatus to be described requires only about 300 cm³ NTP He³ to fill the density cell and the pressure line. This facilitates the maintenance of a constant pressure and is convenient because only moderate quantities of He³ and He⁴, pure and in solution, need to be prepared.

The crucial assumptions in this experiment are the validity of the Clausius-Mosotti relation for helium and the independence of the polarizability A on density. The most extensive measurements to determine A are

^{*} This research has been supported in part by the Army Re-search Office and the National Science Foundation.

[†] Present address: Department of Physics, University of ¹ D. M. Lee, H. A. Fairbank, and E. J. Walker, Phys. Rev. 121,

^{1258 (1961).}

 ¹² J. E. Rives and H. Meyer, Phys. Rev. Letters 7, 217 (1961).
 ³ D. F. Brewer and J. G. Daunt, Phys. Rev. 115, 843 (1959).
 ⁴ A. C. Anderson, W. Reese, and J. C. Wheatley, Phys. Rev. 130, 495 (1963).

⁵ D. F. Brewer and J. R. G. Keyston, Nature 191, 1261 (1961).

¹⁴⁶

 $^{^{5}a}$ Note added in proof. The results on liquid He⁴ below 1.3°K are presented and discussed in the Ph.D. thesis of C. Boghosian, Duke University, 1965 (unpublished). Results above 1.3°K, with particular emphasis on the lambda line have been presented by D. Elwell and H. Meyer, Bull. Am. Phys. Soc. 11, 175 (1966). ⁶ R. H. Sherman and F. J. Edeskuty, Ann. Phys. (N. Y.) 9, 522

^{(1960).}

⁷ K. R. Atkins and M. H. Edwards, Phys. Rev. 97, 1429 (1955). ⁸ E. C. Kerr and R. D. Taylor, Ann. Phys. (N. Y.) 20, 450 (1962).

those of Edwards⁹ and of Chase, Maxwell and Millett¹⁰ on He⁴. Edwards, who studied A on liquid and gaseous He⁴ between 1.2°K and room temperature concludes that the polarizability is independent of density. The measurements of Chase, Maxwell and Millett indicated some systematic departures from a constant value of Aabove the lambda point. This, however, may be due to temperature inhomogeneities in the liquid. Their average value is A = 0.1230. For He³, Peshkov¹¹ found also A = 0.123. Since A is believed to be constant for He^4 for densities up to about 0.14 g/cm³, we will assume that it will stay constant for both liquid He³ and He⁴ under pressure. In this respect the He isotopes and perhaps also the gaseous hydrogen isotopes may be unique. The molecular packing in the liquid phase of these elements is appreciably smaller than in that of argon, where a detailed polarizability study was carried out.¹² There a noticeable change in A was observed in the gaseous and liquid phase.

It should be mentioned that the question of applicability of the Clausius-Mosotti formula has recently received detailed attention and that theoretically some density and temperature dependence of the polarizability even in nonpolar fluids is expected.18

The principle of the density measurement is the same as that used by Lee, Fairbank, and Walker.¹ The following parts constitute the major improvements in design: (1) A low rf level, stable oscillator entirely contained in the cryostat at low temperatures; (2) improved shielding against heat leaks for better isolation of the sample system at temperatures below 0.2° K; and (3) a thermostatically regulated pressure-control system external to the cryostat.

A. Cryostat

Figure 1 gives a schematic view of the cryostat and tank circuit. For more rigidity of the tank circuit, both the inductance and the density cell are mounted on the same support and are cooled by adiabatic demagnetization of a paramagnetic salt pill. A shield which can be kept at temperatures between 1.2 and 0.35°K reduces the stray heat input into the sample. The shield consists of copper 40 SWG wires of about 35 cm length, glued on a thin phenolic form and soldered at the top to a small liquid-He³ container. Its temperature can be regulated by pumping off the liquid and by means of a small heater. The density cell is made up of two parts machined from electrolytic copper and kept electrically isolated by means of thin Mylar spacers. The two parts are rigidly glued together at the top with Araldite. The



resulting seal was found to be leaktight and could stand pressures in excess of 35 atm at all temperatures below room temperature. By this construction it was hoped to achieve good thermal contact between both plates of the condenser.

The inductance consisted of NbZr3 copper-plated wire, wound on a lucite form. The capacitance of the cell was approximately 80 pF and the coil was so designed that the resonant frequency was approximately 15 Mc/sec. The electrical connection of the different parts of the tank circuits was by a screw-tightened joint or by soft solder. A 450- Ω Speer carbon resistor was introduced in a hole drilled in the external copper block of the capacitor, and was kept in thermal contact with the sample support by means of Apiezon oil. A small liquid-He³ pot in this block permitted temperature regulation down to 0.35°K.

The salt pill was made of approximately 30 g of small crystals of chrome methylammonium sulphate. These crystals were embedded between copper wires in an nylon container, and were in thermal contact with them by means of Castolite. A check of several pills made this way showed a very uniform distribution of salt, wires, and binder. A second carbon resistor was embedded in the salt pill in order to show how quickly the thermal equilibrium was established after demagnetization.

B. The Oscillator Circuit

A schematic diagram of the circuit used to measure the dielectric constant of the liquid is shown in Fig. 2. It makes use of a low-power tunnel diode as suggested to us by Dr. C. F. Kellers.^{14,15} The analysis of this circuit is given in Appendix A. The entire oscillator, except for the power supply, is at low temperatures. The external circuit is simply a battery power supply and an amplifier for increasing the strength of the rf signal to a large

⁹ M. H. Edwards, Can. J. Phys. 34, 898 (1956); 36, 884 (1958). ¹⁰ C. E. Chase, E. Maxwell, and W. E. Millett, Physica 27, 1129 (1961).

¹¹ V. P. Peshkov, Zh. Eksperim. i Teor. Fiz. 33, 833 (1957)

[[]English transl.: Soviet Phys.—]ETP 6, 645 (1958)]. ¹² G. O. Jones and B. L. Smith, Phil. Mag. 5, 355 (1960). A. Michels and A. Botzan, Physica 6, 586 (1949).

¹³ D. S. McQuarrie and H. B. Levine, Physica 31, 749 (1965), and private correspondence with Dr. McQuarrie.

¹⁴ The basic circuit for a tunnel-diode oscillator is given in the G. E. Tunnel Diode Manual. The stability of an LC tunnel-diode oscillator at 2.5 Mc/sec has been tested by Heybey (Ref. 15) under supervision of D. M. Lee. We thank Professor Lee for lend-

ing us a copy of this thesis. ¹⁵ O. W. G. Heybey, Master's thesis, Cornell University, 1962 (unpublished).



FIG. 2. Tunnel-diode circuit.

enough magnitude to drive the frequency counter. The large capacity C_2 shunts the transmission line and essentially isolates the frequency determining part of the circuit from the rest of the electronics. The tunnel diode, where almost all the power is dissipated, is thermally anchored to the main He⁴ bath at 1.2°K. The power then dissipated in the tank circuit was found to be negligible. The stray heating into the sample cooled to 0.06°K was found to be unaffected whether the oscillator was on or off.

The short-term stability of the oscillator (measurements every few seconds) was of the order of ± 2 cps. There was no detectable drift in the frequency over a long period of time if the temperature was held constant. Shifts in the frequency of less than 100 cps due to mechanical changes in the oscillator system were observed after the cryostat was slightly shaken during the process of turning large metal valves on the apparatus. Such shifts could be readily identified in a graph of the frequency versus temperature and could be mathematically compensated with a linear translation of the shifted portion of the curve.¹⁶ We will see later that the error in the absolute density due to a 100-cps uncertainty in the magnitude of the frequency is negligible compared with the error that already exists in the values used to normalize the density at 1.2°K.

The frequency of a tunnel-diode oscillator is a function of the bias current. Therefore it was important to provide a very stable source of current. It was found that the voltage-versus-time characteristic of a pair of mercury cells (Eveready E-42) in parallel was sufficiently level for low current drains to satisfy this requirement. The bias supply was kept in a styrofoam box in order to prevent any large changes in temperature. In early experiments, a simple transistorized amplifier was used for increasing the rf signal. Grounding problems, however, introduced feedback into the oscillator and changed the frequency by about 10 cps when certain electrical contacts for other parts of the apparatus were changed. A mixer-amplifier which could be located atop the cryostat was then designed. There, the 10-Mc/sec signal from the Hewlett-Packard counter was fed into the mixer and after elimination of undesirable harmonics the difference between the original oscillator frequency and the 10-Mc/sec standard was counted. This system worked very satisfactorily and is presently used on two other cryostats in this laboratory.¹⁷

C. The Pressure System

It is important that the pressure be kept as constant as possible. In order to determine the expansion coefficient to better than say 5% at all temperatures, the frequency had to be determined to within ± 3 cps. The pressure should always be regulated so that the uncertainty in the frequency due to fluctuations in the pressure was less than ± 1 cps. The system to be described satisfies these requirements and is simple to adjust.

The basic features of the control system are shown in Fig. 3. The reference system contains He⁴ gas held at constant volume and at a temperature regulated within 0.01 °C by means of a thermostatically controlled bath. The stability of the pressure is then $\Delta P/P=3\times10^{-5}$. The mercury column is narrowed at the top to a capillary of approximately 1-mm bore, which is convenient for visual monitoring of the column height. Changes in this height because of changes in the volume of the liquid He³ in the cell and in the capillary are compensated by adjusting the micrometer heads of two mercury pistons. Thus, the reference system pressure and sample pressure can be returned to their original values to within 0.2 mm Hg.

The pressure in the reference system was measured by a Helicoid Bourdon gauge calibrated with a Mansfield and Green Type 3K deadweight tester. The absolute pressure in the sample could then be read to better than 0.5 psi for pressures above 1 atm. For pressures below 1 atm, the reference system was evacuated and the absolute pressure was read to 0.1 mm with a cathetometer.

One has now to estimate the effects due to hydrostatic pressure of the liquid-He³ column in the capillary. This



 17 In the more recent versions of this system, the tunnel diode is mounted directly on the support holding the cell. The oscillator is then very compact and rigid. The short-time stability is then about 0.2 cps. The long-time drift is still not larger than about 1 cps every few hours.

¹⁶ During an actual period of data taking, no valves were turned and any frequency shifts occurred only rarely.

perature. We estimate the height of the He column above the density cell to be of the order of 25 cm, corresponding to a hydrostatic pressure of about 1.5 to 2 mm Hg. During an actual period of data taking between 0.07 and 1.2°K, which lasted approximately five hours, the height of the column was estimated to decrease by less than 5 cm, which would correspond to a change in pressure of 0.3-0.5 mm Hg. This is comparable to the stability of the external pressure regulation,¹⁸ but is only of a long-drift type.

D. Temperature Measurement

The magnetic susceptibility of the salt was used over the whole range of temperature for the actual experiments. The magnetic thermometer was calibrated between 2 and 0.7°K by means of the vapor pressure of the liquid He³ in the sample pot. For this salt, the relation between the magnetic and the absolute temperature has been obtained from direct calibration against the susceptibility of cerium magnesium nitrate¹⁹ down to 0.07°K. The two carbon resistors were calibrated against the vapor pressure of liquid He³ and the salt but were only used for monitoring the thermal equilibrium between the different parts of the sample support. We estimate the accuracy of the absolute temperature to be approximately 2 mdeg between 0.06 and 0.1°K and above this temperature between about 2 and 4 mdeg.

III. PROCEDURE OF THE EXPERIMENT AND EVALUATION OF THE RESULTS

After the whole cryostat was cooled to 4.2°K, the exchange gas was pumped out of the spaces inside the vacuum can. The temperature of the main bath was then lowered to 1.2°K. He³ was then introduced into the line leading to the sample pot, and after condensing, it cooled the sample container. When the salt pill was progressively magnetized at 1.2°K, the reflux of the liquid in the pot kept the temperature of the sample equal to that of the bath, so that no exchange gas had to be used. The temperature was further lowered to about 0.4°K by pumping on the liquid He³ in the pot and finally boiling it off completely. Then adiabatic demagnetization was carried out. Measurements of the frequency were started approximately 40 min after demagnetization and at temperatures above 0.06°K, when the rf frequency had become constant. The heating rate below 0.06°K was estimated to be approximately 10 ergs/min from alternate heating and drift measurements and decreased progressively as the temperature of the heat shield was approached.

An experiment was first performed where the frequency ν_0 of the tank circuit with empty density cell was measured. Such a "blank" experiment was necessary because the frequency was temperature-dependent, the slope increasing as the temperature dropped below 0.2°K. When the curve of ν_0 versus T was well established, He³ was introduced into the density cell. The frequency ν was measured for several pressures as a function of temperature. For each pressure, several adiabatic demagnetizations were carried out. The curves were in general reproducible to a few cycles, after a slight shift in frequency between the different experiments, due to moving the cryostat, had been taken into account. At 1.2°K, the pressure dependence of the frequency was determined.

As will be shown in the Appendix, the dielectric constant ϵ is obtained from the relation

$$\epsilon = \{ (\nu_0/\nu)^2 + K [(\nu_0/\nu)^2 - 1] \} \{ 1 + B(P) \}^{-1}, \quad (1)$$

where $K = C_a/C_s(0)$ and $B(P) = \lceil C_s(P) - C_s(0) \rceil / C_s(0)$. $C_s(P)$ and $C_s(0)$ are the capacities of the cell available to liquid He at pressure P and zero pressure. C_a is the capacity of the addenda. The coefficient B(P) reflects the distortion of the density cell under pressure and is assumed to be independent of temperature. The density ρ is related to the dielectric constant by the Clausius-Mosotti formula

$$\left[(\epsilon - 1)/(\epsilon + 2) \right] M/\rho = \frac{4}{3}\pi A , \qquad (2)$$

where M is the molecular weight and A is the molar polarizability. The value of A used is 0.1230 given by Chase, Maxwell and Millett.¹⁰ Sherman and Edeskuty⁶ have made a determination of the molar volume of liquid He³ as a function of pressure at temperatures above 1.0°K. Their data at 1.2°K were used for the calibration of the density cell and the constants Kand B(P) were determined to be 1.19 and $-2.2 \times 10^{-4}P$, respectively.²⁰ Such a determination of B(P) assumes that the polarizability is pressure-independent up to 30 atm.

The density-versus-temperature curve for each pressure was obtained by computer from the frequencyversus-temperature data using Eqs. (1) and (2). The coefficient of expansion was obtained by graphical differentiation of the average density curve resulting from several experiments. The entropy of compression was calculated by numerical methods from the expansion coefficient and the density values using the relation

$$S(P,T)-S(0,T) = -M \int_{P=0}^{P} \alpha_p \rho^{-1} dP.$$

¹⁸ For the experiments on liquid He⁴, a column of 25 cm corresponds to approximately 2.5-3 mm Hg. Each series of measure-ments, carried out between 0.5 and 1.4°K, lasted only about 30 min, and the change in pressure due to the decrease in height of the column was estimated to be only of the order of 0.1 to 0.2 mm Hg. ¹⁹ H. Meyer (unpublished).

²⁰ From experience with several density cells with varying wall thicknesses, it appears that the coefficient B(P) is not due to a distortion of the copper walls but rather to the shearing of the Araldite seal between the plates.

The temperature dependence of the compressibility can be obtained in two ways: (1) by differentiating the density-versus-pressure curves and then plotting the difference $k_T(T) - k_T(1.2^{\circ}\text{K})$ versus T. (2) by using the thermodynamic relation

$$(\partial k_T/\partial T)_P = -(\partial \alpha_p/\partial P)_T$$

and integrating $(\partial k_T/\partial T)_P$ as a function of T starting at 1.2°K. In practice the second method gave the least scatter and was therefore used preferentially. The smoothed results from this operation are presented in Sec. IV.²¹

The possible sources of errors must now be considered, as has already been done by Lee et al. for their early density experiment. Their greatest source of error, a "possible undetected small pressure drift" is considerably reduced in the present apparatus. Our frequency stability is somewhat better than theirs and possible problems due to stray heating inputs are smaller in our apparatus because no exchange gas was used for magnetization and because of the He³ shield. Therefore in the overlapping temperature range, that is, above about 0.2°K, we estimate our error in α_p to be about ± 0.001 (°K)⁻¹. Below this temperature region, the possibility of a systematic error in α becomes increasingly more significant with a decrease in termperature for the following reasons: (1) The relative uncertainty in the absolute temperature increases; (2) The equilibrium becomes more difficult to establish because of several thermal resistances within the sample holder particularly that at the boundary between the liquid and the cavity walls; and, (3) The slope $d\nu_0/dT$ increases continuously.²² This last effect made interpolation difficult between data points at the lowest temperatures reached. Interpolation was necessary throughout the temperature range of the measurements to form the "standard" frequency-versus-temperature tabulations used in the calculations.

It was mentioned in Sec. II that the absolute value of the frequency within about 100 cps is not too critical to the analysis of the data. The reason for this is that the frequency enters in the calculation of the density as the ratio of two large numbers of the order of 10^7 which differ by about 0.5×10^6 . An uncertainty of 100 cps in the frequency represents an error of less than 0.02% in

FIG. 4. Relative density of liquid He³.

the absolute density. The error in the density data given by Sherman and Edeskuty, used for normalization at 1.2° K, is 0.1%. We were actually able to compensate for the shifts in frequency to better than 50 cps by use of a frequency-versus-pressure calibration made before and after the experiment. Of course, this error had no effect on the coefficient of thermal expansion because this property is only a function of the temperature dependence of the relative density. The discontinuities in the frequency-versus-temperature curves due to these sudden shifts could easily be adjusted to within ± 2 cps, the oscillator stability.

The over-all error as a function of temperature is naturally difficult to calculate exactly because the different sources of error. To obtain a reasonable estimate, curves of the frequency ν_0 and ν versus T were drawn for the maximum possible source of error from the different contributions. The resulting α_p were calculated and the difference from the average value was noted. This error is plotted as an "error bar" for the curves below 0.2°K.

IV. RESULTS AND DISCUSSION

The density ratio, $\rho(T,P)/\rho(1.2^\circ,P)$, is plotted in Fig. 4, as a function of T for various pressures. In Figs. 5-9, the coefficient of expansion is plotted versus pressure for various representative values of T. The agreement with the data of Brewer and Daunt³ down to 0.2°K is very gratifying. Such good agreement between dielectric-constant measurements and adiabatic expansion was already obtained by Rives and Meyer² down to 0.3°K. As to be expected, the agreement between the various experimenters is good at temperatures above 0.5°K and deteriorates as the temperature is lowered. Nevertheless, it can be seen from Fig. 10 that the agreement with Anderson, Reese, and Wheatley⁴ and Brewer and Keyston⁵ at low temperatures is within the combined experimental error. The coefficient of expansion at standard pressures versus temperature is presented in Table I. The extrapolated

²¹ The smaller scatter is due to the fact that the differentiation $\partial \rho / \partial T$ is much smoother than $\partial \rho / \partial P$. Also k_T changes only little with T. Calculating the temperature dependence of k_T by the first method means subtracting one large quantity from another. The scatter introduced by carrying out the differentiation $\partial \alpha / \partial P$ is partly smoothed out by the subsequent integration $\int (\partial k_T / \partial T) dT$.

²² The reason for the increased drop in frequency as T decreases is not well established. Perhaps this has to do with a resistance minimum in a part of the tank circuit. Since the ν_0 -versus-T curve is approximately the same for different inductances (pure wire Cu, tinned Cu, and NbZr₃) it is possible that the effect is attributable to the cylindrical Cu capacitor. Experiments to study this effect will be undertaken shortly with the hope of reducing this temperature variation of ν_0 and extending the density measurements down to the 0.01°K region.



FIG. 5. Expansion coefficient of liquid He³ versus pressure at 1.0°K. • This research. \triangle Brewer and Daunt. \bigcirc Sherman and Edeskuty. \blacksquare Rives and Meyer. \square Lee, Fairbank and Walker. \blacktriangle Kerr and Taylor.



FIG. 6. Expansion coefficient of liquid He³ versus pressure at 0.6° K. • This research. \triangle Brewer and Daunt. \square Lee *et al.* \blacksquare Rives and Meyer. \blacktriangle Kerr and Taylor.



FIG. 7. Expansion coefficient of liquid He³ versus pressure at 0.4° K. • This research. \triangle Brewer and Daunt. \blacksquare Rives and Meyer. \Box Lee *et al.* \blacktriangle Kerr and Taylor. — Goldstein (partial spin contribution).

data at zero pressure is compared to data of previous investigators in Fig. 11. The excellent agreement with the data of Kerr and Taylor⁸ above 0.4° K is particularly pleasing. The systematic deviation between the two sets of measurements below 0.4° K may perhaps be due to difficulties with the temperature equilibrium in the sample of Kerr and Taylor, where more than 1 cm³ of the liquid had to be cooled. For temperatures below 0.09° K, we find approximately $\alpha_p = -(0.14 \pm 0.02)T$. This relationship should only be taken as an indication of the temperature dependence down to 0.06° K. At lower temperatures, deviations from this type of relation may be expected since Abel, Anderson, Black, and



FIG. 8. Expansion coefficient of liquid He³ versus pressure at 0.2° K. • This research. \triangle Brewer and Daunt. \blacksquare Rives and Meyer. \Box Lee *et al.* \blacktriangle Kerr and Taylor. — — Anderson, Reese, and Wheatley; and Brewer and Keyston. — — Goldstein's theory (partial spin contribution).



FIG. 9. Expansion coefficient of liquid He³ versus pressure at 0.1°K. • This research. — — Anderson *et al.*, Brewer and Keyston. — — Goldstein (partial spin contribution).



FIG. 10. Expansion coefficient of liquid He³ at pressures above 20 atm versus temperature. Solid line: this research. Dashed line: average of the data of Anderson, Reese and Wheatley and of Brewer and Keyston at 21.1 and 25.2 atm.

T^P	0	1	3	5	10	15	20	25	28 atm
1.20 °K	30.3	23.2	13.8	8.57	4.02	1.94	0.80	0.15	0.10
1.10	25.3	18.7	11.0	7.03	2.63	0.75	-0.20	-0.87	-1.17
1.00	20.7	15.1	10.4	5.55	1.43	-0.36	-1.35	-1.98	-2.23
0.90	16.8	12.0	7.4	3.88	0.02	-1.66	-2.56	-3.06	-3.23
0.80	12.9	8.7	4.1	1.88	-1.22	-2.66	-3.56	-4.10	-4.27
0.70	9.1	5.00	2.0	-0.12	-2.53	-3.72	-4.60	-5.13	-5.36
0.60	4.9	1.80	-0.6	-1.94	-3.88	-4.90	-5.57	-6.04	-6.30
0.50	-0.10	-1.80	-3.5	-4.10	-5.10	-5.82	-6.36	-6.80	-7.06
0.40	-5.2	-5.75	-6.2	-6.50	-6.86	-7.05	-7.19	-7.29	-7.35
0.30	-9.5	-9.35	-9.0	-8.8	-8.4	-8.12	-8.0	-8.0	-8.0
0.20	-12.5	-11.9	-11.1	-10.4	-9.4	-9.0	-8.8	-8.7	-8.6
0.10	-13.1	-12.4	-11.3	-10.5	-9.4	-9.0	-8.9	-8.8	-8.8
0.085	-12.4	-11.6	-10.5	-9.9	-8.8	-8.3	-8.2	-8.2	-8.2
0.07	-10.8	-10.0	-9.2	-8.7	-8.0	-7.5	-7.5	-7.4	-7.4

TABLE I. Coefficient of expansion of liquid He³ in units of 10^{-3} deg^{-1} (10^{-3} deg^{-1}). Smoothed values at standard pressures.

Wheatley²³ have shown that the specific heat deviates from a linear relation in T.

As can be seen from Table I, there is a minimum in α_p which occurs for all pressures in the vicinity of 0.12°K. The exact temperature of the minimum is difficult to locate because of the flatness of the minimum and the relatively large error in the expansion coefficient. The minimum could occur at any temperature up to about 0.2°K. The lowest temperature where the minimum can occur is determined by a thermodynamic relation. One has

$$\left(\frac{\partial \alpha_p}{\partial T}\right)_P = -\alpha_p^2 - \frac{1}{VT_m} \left(\frac{\partial C_p}{\partial P}\right)_P + \frac{\alpha_p}{T_m} = 0,$$

which implies that $\partial C_p / \partial P < V \alpha_p$ at the temperature of the minimum. The results of Brewer, Daunt and Sreedhar²⁴ on the specific heat of liquid He³ show that $\partial C_p / \partial P = 0$ at 0.16°K. From these results, we must



FIG. 11. Expansion coefficient of liquid He² near zero pressure versus temperature. Curve A: This research (P=0). Curve B: Kerr and Taylor (SVP). Curve C: Lee, *et al.* (0.2 atm). Curve D: Rives and Meyer (0.2 atm).

conclude that the minimum in α_p lies above 0.16°K at all pressures. The results of Anderson, Reese and Wheatley⁴ do not show the crossing of the constant pressure curves of the specific heat as clearly as do those of Brewer et al., but the crossover is smeared out over a temperature interval of approximately

0.14 < T < 0.25 °K.

Goldstein²⁵ assumes the coefficient of expansion to be given by the sum of the spin contributions $\alpha_{\sigma,p}$ and the nonspin contribution $\alpha_{n\sigma,p}$, the latter being always positive. The results of Goldstein's theory are included in Figs. 8–9. It must be remembered that these are the predicted values for $\alpha_{\sigma,p}$ and not the total α_p . Therefore the values of α_p shown here are a lower limit and the total calculated α_p is always more positive. From these figures it is apparent that the experimental results are partly in contradiction with Goldstein's theory, since the experimental curves at temperatures above 0.2°K tend to lie below the calculated α_p curves. At temperatures below 0.15°K however, there is good consistency between theory and experiment for all pressures.



FIG. 12. Density maximum of liquid He³. • This research. Rives and Meyer. \triangle Brewer and Daunt. \Box Lee *et al.* \bigcirc Sherman and Edeskuty. \times Grilly and Mills.

²⁵ L. Goldstein, Phys. Rev. 133, A52 (1964).

 ²⁸ W. R. Abel, A. C. Anderson, W. C. Black, and J. C. Wheatley, Physics 1, 337 (1965).
 ²⁴ D. F. Brewer, J. G. Daunt, and A. K. Sreedhar, Phys. Rev.

^{115, 836 (1959).}

	0	1	3	5	10	15	20	25	28 atm
1.20 °K	0	-1.19	-2.69	-3.54	-4.68	-5.20	-4.52	-5.50	-5.50
1.10	0	-0.98	-2.20	-2.91	-3.77	-4.06	-4.10	-4.00	-3.91
1.00	0	-0.80	-1.79	-2.37	-2.98	-3.06	-2.91	-2.63	-2.42
0.90	0	-0.64	-1.43	-1.86	-2.17	-2.01	-1.65	-1.18	-0.88
0.80	0	-0.48	-1.02	-1.26	-1.29	-0.93	-0.39	+0.25	+0.65
0.70	0	-0.31	-0.58	-0.65	-0.37	+0.20	+0.91	+1.72	+2.22
0.60	0	-0.16	-0.22	+0.11	+0.46	+1.25	+2.15	+3.10	+3.70
0.50	0	+0.04	+0.27	+0.57	+1.46	+2.43	+3.48	+4.56	5.23
0.40	0	+0.24	+0.75	+1.27	+2.54	+3.79	+5.01	6.20	6.91
0.30	0	+0.42	+1.20	1.92	3.53	5.00	6.37	7.19	8.46
0.20	0	+0.54	+1.51	2.37	4.24	5.86	7.38	8.85	9.71
0.10	0	+0.56	+1.57	2.46	4.37	6.01	7.53	8.99	9.85
0.085	0	+0.53	+1.47	2.30	4.04	5.56	6.96	8.24	9.07
0.07	0	+0.48	+1.32	2.06	3.64	5.01	6.28	7.50	8.22

TABLE II. Entropy of compression of liquid He³, $10^{2}{S(P,T)-S(0,T)}/R$.

We also note the qualitative agreement between our data at zero pressure and the theoretical result $\alpha_p = -0.08T$ predicted from the theory of Brueckner and Gammel.²⁶

Finally we show in Fig. 12 the temperature where $\alpha_p = 0$ is plotted against the pressure, and compare our results to work of previous authors. Again the agreement is best with the result of Brewer and Daunt.

The entropy of compression is tabulated for various pressures as a function of T in Table II. Because of the uncertainty still existing in the temperature dependence of C_p under pressure at very low temperatures a comparison with the entropy calculated from the specificheat data^{4,5} should still be considered somewhat tentative. However the influence of this uncertainty on the absolute value of S is probably small at temperatures above 0.1°K, and decreases further as the temperature increases. A representative comparison is given





10

К

FIG. 14. Entropy of compression of liquid

T=0.1 °K

In Figs. 13 and 14, where the entropy of compression is plotted as a function of pressure at different temperatures. As can be seen, the entropy of compression derived from density measurements is consistent with that calculated from specific-heat data.



²⁶ K. A. Brueckner and J. C. Gammel, Phys. Rev. **109**, 1023, 1040 (1958).

²⁷ M. Strongin, G. O. Zimmerman, and H. A. Fairbank, Phys. Rev. 28, 1983 (1962).

م \										
T	0	1	3	5	10	15	20	25	28 atm	
 0.5 °K	0.0015	0.508	2.04	2.94	5.12	7.32	9.36	11.23	12.47	
0.4	3.41	4.28	5.26	6.07	7.54	8.68	9.71	10.56	11.06	
0.3	9.09	9.01	8.75	8.73	8.73	8.83	9.18	9.72	10.05	
0.2	11.62	10.75	9.67	8.81	7.79	7.63	7.70	7.91	7.93	
0.1	9.96	8.99	7.49	6.52	5.35	4.99	5.00	4.99	5.05	
0.09	9.11	8.14	6.76	5.85	4.71	4.37	4.22	4.18	4.22	
0.08	7.95	7.04	5.83	4.95	4.06	3.89	3.65	3.61	3.55	
0.07	6.45	5.52	4.65	4.14	3 49	3 05	3.05	2 08	2 00	

TABLE III. Ratio of specific heats expressed as $(\gamma - 1) \times 10^4$.

The change in the coefficient of isothermal compressibility is shown in Fig. 15. The values of the compressibility at 1.2°K may be obtained from Sherman and Edeskuty.6

The ratio $C_p/C_v = \gamma$ has been calculated from the relation

$$u - 1 = T u_1^2 \alpha_p^2 / C_p$$

and is presented in Table III. The specific-heat data under pressure used are those of Brewer et al., 5,28 as they cover the widest temperature range. The velocity of sound u_1 has been taken from the data of Abel, Anderson and Wheatley.29

V. CONCLUSION

In conclusion our experiment has shown the consistency between the different types of measurements of α_p in liquid He³. Therefore it is believed that the assumption of a temperature-independent polarizability, which is essential to this experiment, is well confirmed. There are some systematic discrepancies between the results of different authors; some of these are thought to be due to difficulties inherent with work at very low temperatures, such as uncertainty in the absolute temperature scale, incomplete thermal equilibrium within the sample etc. The development of the constant-pressure control system, the application of a tunnel-diode circuit to density measurements and the reduction of the heat leak are the main improvement over the measurements of Lee, Fairbank and Walker who were the first to demonstrate the usefulness of this method to the problem of liquid He³.

ACKNOWLEDGMENTS

We are indebted to Dr. E. R. Hunt and J. F. Jarvis for their advice and help with the electronics. We are very grateful to Dr. R. C. Richardson and Dr. D. C. Rorer for help in several of the experiments. We are grateful to Professor H. A. Fairbank for his comments on the manuscript. One of us, (H. M.) has been supported by an A. P. Sloan Fellowship during the period in which this work was carried out.

APPENDIX A: TUNNEL-DIODE CIRCUIT ANALYSIS

The equivalent circuit for the tunnel-diode oscillator is given in Fig. 16 and the total impedance of this circuit can be written as the sum of two parts

$$Z = Z_i + Z_t, \tag{A1}$$

where the subscript *t* refers to the tank circuit composed of C_s , L and g_L , the equivalent conductance. Also we have

$$Z_i = (g_1 + j\omega C_2)^{-1} + (-g_d + j\omega C_1)^{-1}.$$
 (A2)

It is well known that the condition for constant amplitude oscillation is the vanishing of the real part of the total circuit impedance and the frequency is given by the vanishing of the imaginary part of Z. We have

$$\operatorname{Re}(1/Z_i) + g_L = 0, \qquad (A3)$$

$$\operatorname{Im}(1/Z_i) + \omega C_s - 1/\omega L = 0, \qquad (A4)$$

with

$$\operatorname{Re}(1/Z_{i}) = \frac{(g_{1} - g_{d})[-g_{1}g_{d} - \omega^{2}C_{1}C_{2} + \omega^{2}(g_{1}C_{1} - g_{2}C_{2})(C_{1} + C_{2})]}{(g_{1} - g_{0})^{2} + \omega^{2}(C_{1} + C_{2})^{2}},$$
(A5)

$$\operatorname{Im}(1/Z_i) = \frac{\omega g_1^2 C_1 + g_d^2 C_2 + \omega^2 C_1 C_2 (C_1 + C_2)}{(g_1 - g_d)^2 + \omega^2 (C_1 + C_2)^2}.$$
(A6)

From Eq. (A3) and (A5) we get the result that

$$g_d - g_L = (C_1/C_2)^2 g_1$$
 (A7)

from which it can be seen that the negative conductance of the diode must be slightly larger than the equivalent conductance of the tank circuit, the difference being given by the right-hand side of Eq. (A7). Because of this relation, the capacitances, C_1 and C_2 , must be chosen so that g_1 can be adjusted until this condition is met. It is difficult to calculate the actual difference, but

²⁸ D. F. Brewer, A. K. Sreedhar, H. C. Kramers, and J. G. Daunt, Phys. Rev. 110, 282 (1958). D. F. Brewer, J. G. Daunt, and A. K. Sreedhar, *ibid.* 115, 836 (1959).
²⁹ W. R. Abel, A. C. Anderson, and J. C. Wheatley, Phys. Rev. Letters 7, 299 (1961).



FIG. 16. Equivalent circuit diagram of tunnel-diode oscillator.

the order of magnitude can be estimated, which is somewhere between 10^{-9} and 10^{-10} . The "Q" of the tank circuit is given by $\omega C/g_L$. Since we know that $g_L \gg g_d$, a lower limit to the Q of the tank circuit can be determined. In the present case, $g_d = 4 \times 10^{-5}$ mho, $\omega = 10^8$ sec⁻¹ and C = 100 pF. Using these values, we find that the minimum Q of the tank circuit is approximately 250.

Equation (A6) gives the approximate relation

$$\omega^{2} = \frac{1 - (C_{1}g_{1}^{2} + C_{2}g_{d}^{2})L(C_{1} + C_{2})^{-2}}{L[C_{s} + C_{1}C_{2}(C_{1} + C_{2})^{-1}]}, \quad (A8)$$

where the approximations used are

$$(g_1 - g_d)^2 \approx 10^{-8} \ll \omega^2 (C_1 + C_2)^2 \approx 10^{-2}$$
 (A9)

$$g_d g_1 \approx 10^{-9} \ll C_1 C_2 \omega^2 \approx 10^{-4}$$
. (A10)

For the oscillator in the present experiment, the second term in the numerator of Eq. (A8) has the approximate value of 10^{-6} . The second term in the denominator, however, is much more significant and has the approximate value of 2×10^{-11} which is roughly 20% of the first term.

The value of the dielectric constant at a given temperature is determined from the relative magnitudes of the resonant frequency of the circuit when the cell is empty ω_0 and when the cell is filled with liquid helium.

The capacitor of the tank circuit is made up of two parts

$$C_s = C_s^0 + C_a, \qquad (A11)$$

where C_a is the value of the capacity of the part which does not come into contact with the liquid. C_s^0 is the value of that part of the cell which is available to the liquid at zero pressure. Under pressure, its value is C_s because of distortions. From Eq. (A8), it can be seen that the frequency of the tunnel-diode oscillator with and without liquid may be written as

$$\omega^2 = \frac{1 - GL}{L(\epsilon C_s + C_A)}, \qquad (A12)$$

and

$$\omega_0^2 = \frac{1 - GL}{L(C_s^0 + C_A)},$$
 (A13)

where

$$G = \frac{C_1 g_1^2 + C_2 g_2^2}{(C_1 + C_2)} \text{ and } C_A = C_a + \frac{C_1 C_2}{C_1 + C_2}, \text{ (A14)}$$

$$(\omega_0/\omega)^2 = (C_A + \epsilon C_s)(C_A + C_s^\circ).$$

If we let $C_s = C_s^0 [1+B(P)]$, where

$$B(P) = (C_s(P) - C_s^0) / C_s^0$$

and $K = C_A/C_s^0$, we obtain finally for the dielectric constant

$$\epsilon = \{ (\omega_0/\omega)^2 + K[(\omega_0/\omega)^2 - 1] \} \{ 1 + B(P) \}^{-1}$$

Thus knowledge of ω_0 , ω and the value of ϵ versus pressure at a convenient temperature will provide a calibration of the system. Then ϵ can be determined for all temperatures and pressures from the values of ω and ω_0 .

and