On the contour C_l we are integrating over a small circle around the origin in γ'. It is not difficult to see that the leading term comes from expanding in γ', higher order corrections to the term we give being $O(\omega/\mu)$;

$$F=\frac{1}{(2\pi)^{\frac{3}{2}}(\beta t_l)^{\frac{1}{2}}}\frac{(-1)^l}{\gamma'}\times\exp\left[-\left(\frac{x^2+y^2}{2\gamma'}\right)\right]+O(\omega/\mu). \quad \text{(A.35)}$$

Proceeding similarly for the rest of the integrand, we obtain finally

$$g_1(\mathbf{r};\omega)=\frac{\pi}{(2\pi)^{\frac{3}{2}}\beta}\sum_{l=-\infty}^{\infty}{}'\frac{(-1)^l e^{\beta\mu t_l}}{(\beta t_l)^{\frac{1}{2}}\sin\pi t_l}\frac{1}{2\pi i}\times\oint\frac{\exp\{\gamma'\mu-(x^2+y^2)/2\gamma'\}}{\gamma'}d\gamma'. \quad \text{(A.36)}$$

The last integral is again well known.[15] Substituting its value and that of t_l, we finally obtain (up to correction terms of order ω/μ)

$$g_1(\mathbf{r};\omega)=\frac{\omega^{\frac{3}{2}}}{2\pi}\sum_{l=1}^{\infty}\frac{(-1)^l\sin(2\pi l\mu/\omega-\pi/4)}{l^{\frac{1}{2}}\beta\omega\sinh(2\pi^2 l/\beta\omega)}\times J_0\{[2\mu(x^2+y^2)]^{\frac{1}{2}}\}. \quad \text{(A.37)}$$

When g_1 is put into (A.23) it will yield the oscillatory terms in the proper self-energy part. Comparing g_0 from (A.32) with g_1, we see that g_1/g_0 is of the order of $(\omega/\mu)^{\frac{3}{2}}$, which is the estimate we used in obtaining the general formula for the DHVA oscillations in the thermodynamic potential. It is easy to trace down the origin of the $\omega^{\frac{3}{2}}$ dependence of g_1: it arises from the f_s^- factor in (A.1). This same kind of dependence comes in higher order skeleton diagrams from the sums on the true propagators, for ζ_l near μ, and therefore it is easy to see that this $\omega^{\frac{3}{2}}$ dependence is quite general.

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Dielectric Constant, Density, Expansion Coefficient, and Entropy of Liquid He^3 Under Pressure Below 1°K*†

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By measuring the resonant frequency of an LC circuit containing a capacitor filled with liquid He^3, the dielectric constant of the liquid was measured from 0.14 to 1°K at several pressures from 0.2 to 29.5 atmospheres. From these measurements the density, expansion coefficient at constant pressure, and the change in entropy on compression from saturated vapor pressure to higher pressures were determined. α_p was found to be negative and, hence, $(\partial S/\partial P)_T$ positive for all pressures below a certain temperature which increased monotonically with pressure, confirming behavior found by Brewer and Daunt. A minimum in the melting curve of He^3 was found at $T=0.32\pm0.01$°K and $p=29.1\pm0.1$ atm. The results are compared with those obtained by other methods.

I. INTRODUCTION

THE present experiment is an outgrowth of an earlier set of measurements on the heat transport properties of liquid He^3 in which it was found from the convection behavior that a maximum in the density occurs at 0.5°K for a pressure of $\frac{2}{3}$ atmosphere.[1]

In this paper we report experimental measurements of the density, ρ, the thermal expansion coefficient at constant pressure, α_p, and entropy of compression of liquid He^3 at temperatures from 0.14 to 1.0°K and at pressures from 0.2 to 29 atmospheres. Because the changes in density below 1°K are small, a sensitive method of measurement was required. The limited amounts of He^3 available likewise restricted the size of the liquid sample. Both of these requirements were satisfactorily met in a measurement of the dielectric constant, ϵ, of the liquid as described in Sec. II. The density of the liquid was found from ϵ, using the Clausius-Mossotti relation,

$$\frac{\epsilon-1}{\epsilon+2}\frac{M}{\rho}=\frac{4\pi}{3}A, \quad (1)$$

where M is the molecular weight and A is the molar polarizability. $\alpha_p=(1/V)(\partial V/\partial T)_p$ was likewise determined at several different pressures. The entropy of compression at any pressure p could than be found from the relation

$$S_p-S_{\text{sat}}=-\int_{\text{svp}}^{p}V\alpha_p dp, \quad (2)$$

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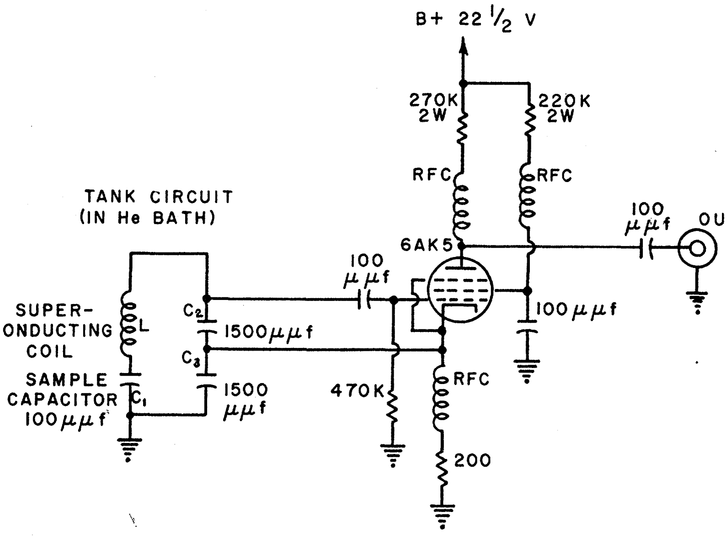
* Assisted by the National Science Foundation.

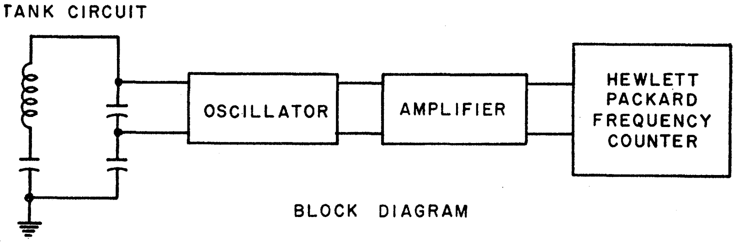
† Part of this work has been submitted to Yale University by D. M. L. in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

‡ Now at Cornell University, Ithaca, New York.

[1] D. M. Lee and H. A. Fairbank, Phys. Rev. **116**, 1359 (1959).



OSCILLATOR SCHEMATIC



BLOCK DIAGRAM

FIG. 1. The oscillator circuit.

where S_p is entropy at pressure p and S_{sat} is the entropy at saturated vapor pressure (svp). Preliminary reports of the results near saturated vapor pressure (0.2 atm) were reported earlier.[2,3]

Recently, Brewer and Daunt[4] have measured the change in temperature on adiabatic expansion of liquid He^3 in the pressure range 0.15°K to 1.15°K at pressures up to 22 atmospheres from which they determined α_p and $S_p - S_{\text{sat}}$. Preliminary results of a direct measurement of the density of liquid He^3 at the saturated vapor pressure below 1°K have also been reported by Taylor and Kerr.[5] Their results as well as some by Sherman and Edeskuty[6] and Grilly and Mills[7] above 1°K are compared with the present work in Sec. IV.

In the course of these measurements it was established that a minimum in the melting curve of He^3 occurs at 0.32 ± 0.01°K and 29.1 ± 0.1 atm.[8] Values of the melting pressure as a function of temperature were obtained from 0.32 to 0.8°K in reasonable agreement with earlier work.

II. APPARATUS

The dielectric constant of liquid He^3 was determined from the resonant frequency of an LC circuit in which part of the capacitance was a coaxial cylindrical capacitor containing the liquid. A simple Clapp oscillator circuit shown in Fig. 1 was used in these experiments. Because of the two large capacitors, C_2 and C_3, the resonant frequency is insensitive to changes in the lead capacitance, so that the tank circuit can be placed at a considerable distance from the oscillator tube without reducing the stability. It was thus possible to place the entire tank circuit in the cryostat. Instabilities due to mechanical vibrations were minimized by rigidly mounting all parts of the tank circuit. The resonant frequency of approximately 5 Mc/sec was measured to 0.1 cycle per second by a Hewlett Packard 524B electronic counter and was stable to better than 1 part per million per hour. The low oscillator plate voltage was required to maintain a low power level in the tank circuit.

The essential features of the cryostat used in these experiments are shown in Fig. 2. The assembly shown

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[2] D. M. Lee, J. D. Reppy, and H. A. Fairbank, Bull. Am. Phys. Soc. **3**, 339 (1958).

[3] D. M. Lee and H. A. Fairbank, Phys. Fluids **2**, 582 (1959).

[4] D. F. Brewer and J. G. Daunt, Phys. Rev. **115**, 843 (1959).

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[8] D. M. Lee, H. A. Fairbank, and E. J. Walker, Bull. Am. Phys. Soc. **4**, 237 (1959).

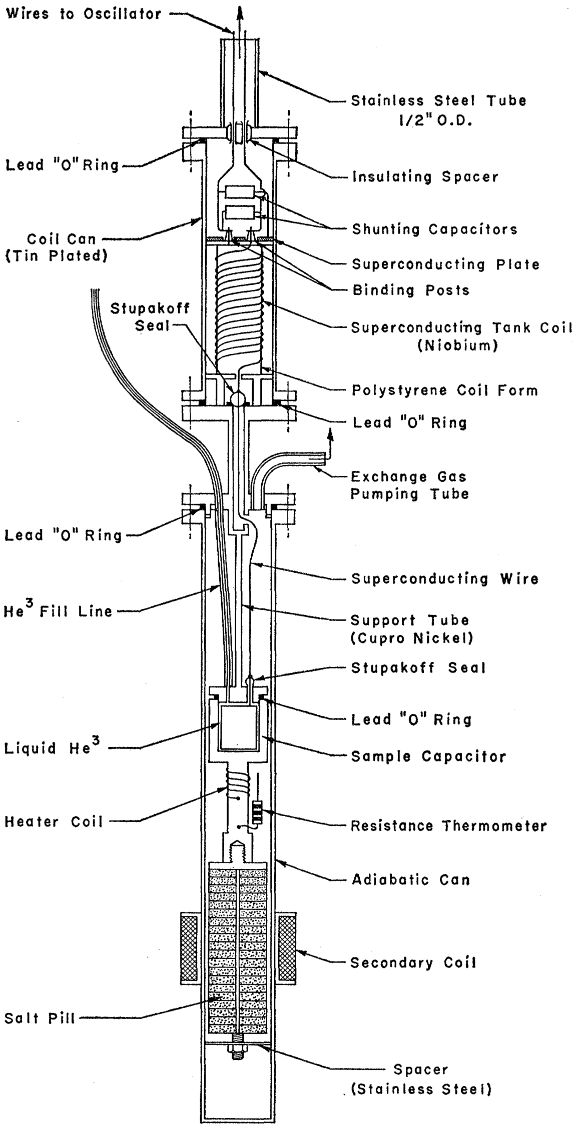

FIG. 2. The lower portion of the cryostat used for the dielectric constant measurements.

in the figure was immersed in liquid helium. The upper can, which was tin-plated inside, contained the superconducting oscillator tank coil (about 60 turns of 0.010-in. niobium wire wound on a polystyrene form) and two mica shunting capacitors. The use of superconducting components minimized Ohmic heating, and, consequently, the tank circuit had a high "*Q*." The upper can was isolated from the lower one by means of a Kovar-glass feed-through so that there were two independent exchange gas systems. He^4 gas at a few mm Hg pressure was introduced into the upper can before each run to maintain the circuit elements in thermal equilibrium with the external helium bath. The lower can contained the He^3 sample capacitor and a compressed powder salt pill consisting of about 40 grams of potassium chrome alum for magnetic cooling. Thermal contact between the salt refrigerant and the He^3 sample was achieved by screwing the salt pill shank (electrolytic copper) tightly into a threaded hole in the sample capacitor. The temperature was measured by an IRC 270-ohm carbon composition resistor or a Speer 12-ohm resistor calibrated against the susceptibility of the salt pill as determined by the ballistic method. The salt and sample capacitor could be thermally connected to or thermally isolated from the external helium bath by the introduction or removal of He^4 exchange gas.

Figure 3 shows the details of the He^3 sample capacitor whose parts were carefully machined from electrolytic copper to allow a gap width of 0.0035-in. between the inner and outer cylinders. Thin Teflon strips were wrapped around the inner cylinder to prevent it from short circuiting to the outer cylinder. The electrical lead to the inner cylinder was threaded through and soldered to the hollow central stem of a Kovar-glass seal. The upper flange was attached to the top of the outer cylinder with 12 2–56 brass screws compressing a lead "O" ring gasket. This arrangement made assembly and disassembly rather convenient. The capacitor was superfluid tight and was able to withstand 40 atmospheres with zero leakage.

The He^3 was condensed into the sample chamber

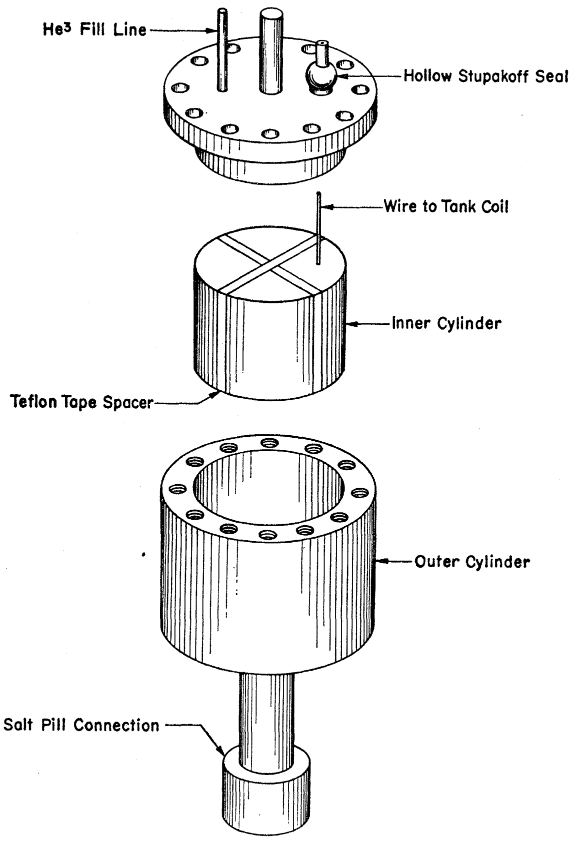

FIG. 3. Details of the He^3 sample capacitor.

through a 0.011-in. i.d., 0.030-in. o.d. cupro-nickel tube soldered to the top flange of the sample capacitor. The upper end of this fill line was connected to a gas handling system. In the measurements performed at pressures near the saturated vapor pressure, this gas handling system consisted of a glass Toepler pump and a mercury manometer for measuring the pressure. In the measurements at elevated pressures, a pressurizing system identical with that described by Walker and Fairbank[9] was used.

The He^3 used in these experiments had a purity of better than 99.9%.

III. PROCEDURE

The procedure of a typical run was as follows: The tank circuit was cooled to about 1.2°K and the resonant frequency measured, first with the coxial capacitor empty and then with the capacitor filled with liquid He^3. With the He^3 at constant pressure the condenser was then cooled to about 0.15°K by adiabatic demagnetization of the attached paramagnetic salt pill. As the capacitor slowly warmed to the temperature of the bath, the resonant frequency of the tank circuit and the temperature of the capacitor, as determined by the resistance thermometer, were simultaneously measured at frequent intervals.

In order to obtain values of the dielectric constant from the raw frequency data, it was necessary to calibrate the apparatus carefully. In the resonant circuit shown in Fig. 1 the shunting capacitors C_2 and C_3 were almost exactly equal and will henceforth be denoted by $C=C_2=C_3$. The sample capacitance C_1 will be written in the form $C_1=C_0+k\epsilon$. In this expression, $k\epsilon$ represents that part of the capacitance which depends on the dielectric constant ϵ of the liquid helium sample and is equal to some constant k when the sample is absent. C_0 is the stray capacitance to ground resulting from the Teflon tape spacers, the Stupakoff seal, and the leads. The dielectric constant is related to the resonant frequency of the tank circuit by the equation

$$f^2=[C+2(C_0+k\epsilon)]/[4\pi^2LC(C_0+k\epsilon)], \qquad (3)$$

where L is the self-inductance of the circuit.

From this equation ϵ can be determined from the frequency provided that the constants C, L, C_0, and k are known. Using a capacitance bridge, C and C_0+k were measured directly, the latter being the capacitance of the empty condenser. The two remaining equations needed to evaluate these four constants were obtained by inserting in Eq. (3) the resonant frequency of the tank circuit when the sample capacitor was empty and when the sample capacitor was filled with liquid He^3. In the latter case it was necessary to use the value of ϵ for He^3 computed from the Clausius-Mossotti equation, the known density of liquid He^3 [6] and a molar polarizability of 0.123 as measured by Peshkov[10] at optical frequencies.

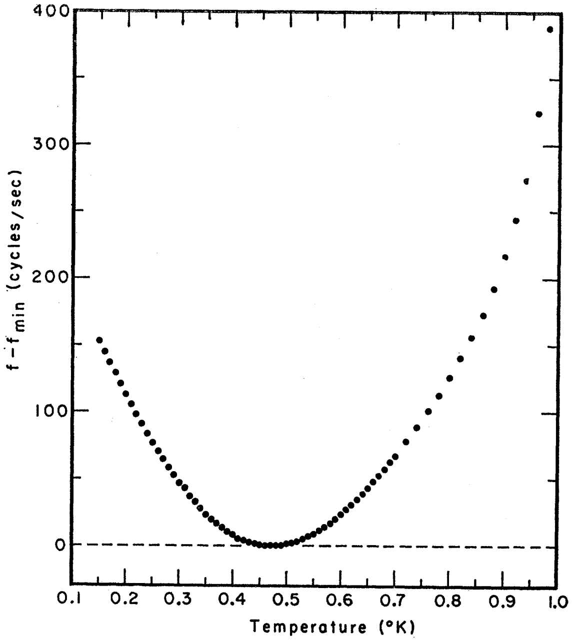

FIG. 4. The resonant frequency of the LC circuit plotted against temperature for the He^3 sample at a pressure of 0.2 atm.

To check this calibration, a separate experiment was performed in which the change in resonant frequency was observed when liquid He^4 was added to the empty capacitor at 1.2°K. The dielectric constant of He^4 computed from this frequency change was found to be 1.0567 in the run of December 26–28, 1958, and 1.0571 in the run of January 17–29, in good agreement with the value of 1.0569 at 1.4°K as measured by Maxwell, Chase, and Millett.[11] (The change in ϵ of He^4 between 1.4°K and 1.2°K is a negligible correction.)

IV. RESULTS

The resonant frequency of the LC circuit in a typical constant pressure warm-up is plotted against temperature in Fig. 4. The data are represented on the graph as a variation in frequency from the minimum frequency, which occurs near 0.5°K for this pressure. It follows from Eq. (3) (since the changes in ϵ are very small) that the change in ϵ corresponding to a change in f is given by

$$\Delta\epsilon=8\pi^2(L/k)f\Delta f(C_0+k\epsilon)^2. \qquad (4)$$

The change in density then follows from the relationship

$$\Delta\rho/\rho=3\Delta\epsilon/[(\epsilon-1)(\epsilon+2)], \qquad (5)$$

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[9] E. J. Walker and H. A. Fairbank, Phys. Rev. **118**, 913 (1960).

[10] V. P. Peshkov, Soviet Phys.-JETP **6**, 645 (1958).

[11] E. Maxwell, C. E. Chase, and W. E. Millett, *Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry, Madison, Wisconsin, August 30, 1957*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, Wisconsin, 1958), p. 53.

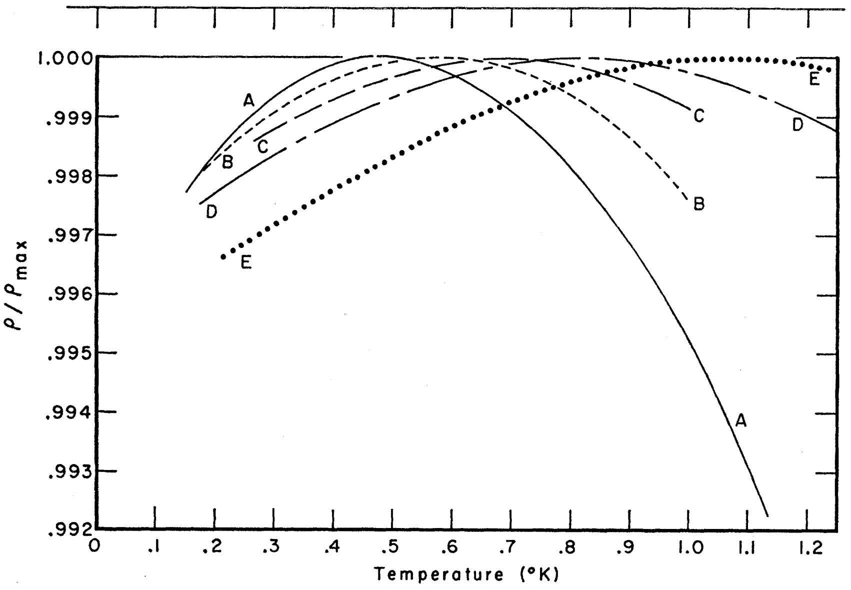

FIG. 5. The ratio of the density of liquid He^3 to the maximum density of the liquid as a function of temperature at five constant pressures as follows: Curve A—0.2 atm, Curve B—2.9 atm, Curve C—5.8 atm, Curve D—9.7 atm, Curve E—19.7 atm.

obtained from the Clausius-Mossotti equation [Eq. (1)]. These equations are valid for changes in temperature made at constant pressure but not for changes of pressure. This is because the very slight deformation of the capacitor with an increase in pressure decreases the capacitance by an amount of the order of 10% of the increase due to the charge in liquid density. Therefore, in each run at constant pressure we normalized our densities (and dielectric constants) to the density values of Sherman and Edeskuty[6] at 1°K. The values of $\Delta\epsilon$ obtained at constant pressure from Eq. (4) will still contain a small error since the values of k and C_0 used were obtained at only the lowest pressure. However, since k and C_0 change by a maximum of a few tenths of a percent with pressure, the resulting error in $\Delta\epsilon$ is less than 1%. Our results at five different pressures are given in Fig. 5.

The thermal expansion coefficient at constant pressure, $\alpha_p=(1/V)(\partial V/\partial T)_p=-(1/\rho)(\partial\rho/\partial T)_p$ was determined from the above data and is plotted in Fig. 6 for several pressures.

The entropy of compression was then calculated from Eq. (2) as a function of pressure for temperature from 0.2°K to 1.0°K and the results are plotted in Fig. 7. The absolute entropy can be obtained from the entropy at saturated vapor pressure. The specific heat at

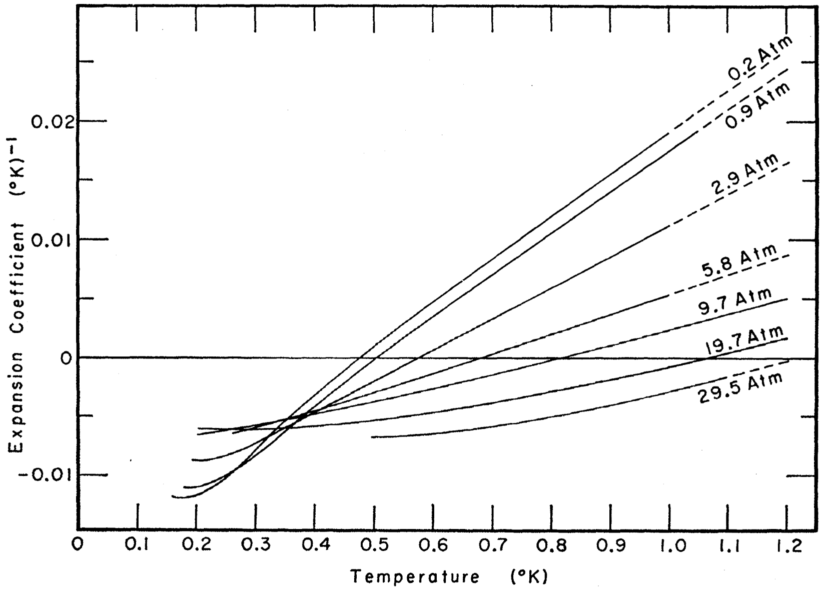

FIG. 6. The thermal expansion coefficient at constant pressure of liquid He^3 as a function of temperature.

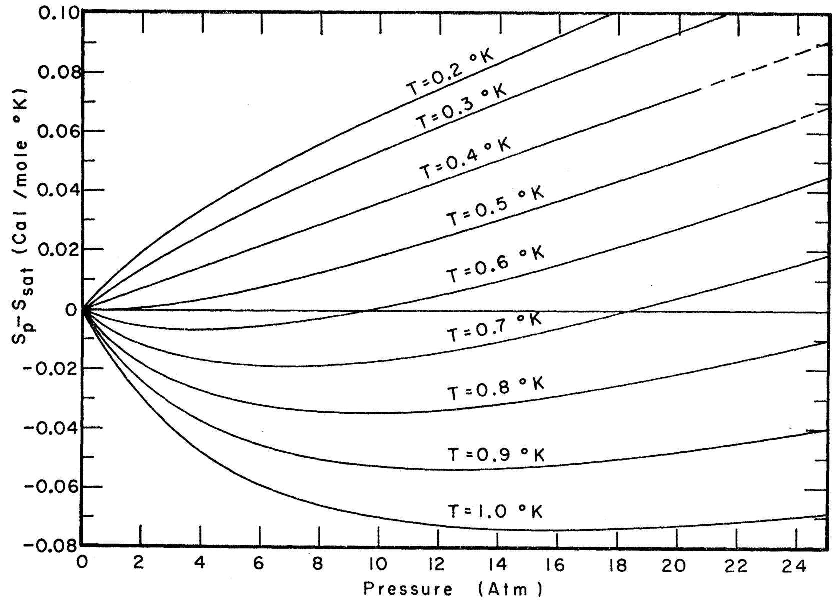

FIG. 7. The change in entropy of liquid He^3 from the value at saturated vapor pressure at a function of pressure at constant temperature.

saturated vapor pressure, C_{sat}, has been measured from 0.085°K to 0.75°K by Brewer, Daunt, and Sreedhar,[12] from 0.23 to 2°K by Abraham, Osborne, and Weinstock,[13] and from 0.54°K to 1.7°K by Roberts and Sydoriak.[14] Each of these authors calculated entropies at the saturated vapor pressure S_{sat} from the observed values of C_{sat} but used different methods to obtain absolute entropies. Brewer *et al.* linearly extrapolated C_{sat} to 0°K. Roberts and Sydoriak normalized entropy values to an absolute value at 0.5°K computed from vapor pressure data, and Abraham *et al.* to a value of S_{sat} at 1.5°K computed from heat of vaporization data. The results of Brewer *et al.* and Roberts and Sydoriak are in excellent agreement; however, the values of Abraham *et al.* are about 0.1 cal/mole degree higher throughout the temperature range of the overlap of data. The source of this discrepancy which exceeds the combined estimates of error is not yet explained. Following Brewer and Daunt we have taken the values of Brewer *et al.* for S_{sat} below 0.75°K and those of Abraham *et al.*, reduced by 0.105 cal/mole degree, above 0.75°K to bring the data into agreement at this temperature. Using these values of S_{sat} and our values of $S_p - S_{sat}$ from Fig. 7, S_p has been calculated and is given at four pressures as a function of T in Table I.

In evaluating the accuracy of these measurements the possibility of systematic errors in the frequency, pressure, and temperature deserve discussion. Although the oscillator frequency can be measured to a high precision (±0.1 cycle/sec), there is the distinct possibility of an error in $\Delta\epsilon$, $\Delta\rho$, and α_p arising from a frequency drift unrelated to the change in the properties of the He^3 sample. To check on this possibility, the temperature of the tank circuit was maintained constant at about 1°K on several occasions for periods of about one hour and the frequency monitored. A maximum variation of only a few cycles/sec was observed which appeared to be a random variation rather than a steady drift. A check which simulated more closely the conditions of the experiment was made on a demagnetization

TABLE I. Entropy of liquid He^3 in cal/mole deg as a function of pressure in atmospheres and temperature in °K. The values at vapor pressure are those given by Brewer and Daunt (see text). The values of Brewer and Daunt at the other pressures are larger than our values by the amount shown in parenthesis at each p and T.

$T \diagdown p$	svp	5.0	10.0	15.0	22.0
0.200	0.766	0.806	0.831	0.854	
		(0.007)	(0.018)	(0.035)	
0.300	1.042	1.073	1.095	1.116	1.144
		(0.006)	(0.015)	(0.027)	(0.037)
0.400	1.253	1.271	1.289	1.307	1.333
		(0.008)	(0.014)	(0.023)	(0.029)
0.500	1.426	1.432	1.444	1.460	1.484
		(0.006)	(0.012)	(0.018)	(0.023)
0.600	1.573	1.566	1.574	1.585	1.607
		(0.008)	(0.013)	(0.018)	(0.020)
0.700	1.703	1.684	1.686	1.695	1.712
		(0.007)	(0.013)	(0.016)	(0.018)
0.800	1.822	1.792	1.788	1.792	1.805
		(0.006)	(0.011)	(0.015)	(0.018)
0.900	1.933	1.890	1.881	1.881	1.888
		(0.009)	(0.010)	(0.013)	(0.017)
1.000	2.036	1.982	1.966	1.962	1.964
		(0.008)	(0.009)	(0.010)	(0.013)

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[12] D. F. Brewer, J. G. Daunt, and A. K. Sreedhar, Phys. Rev. **115**, 836 (1959).

[13] B. M. Abraham, D. W. Osborne, and B. Weinstock, Phys. Rev. **98**, 551 (1955); Physica **24**, 132 (1958); B. Weinstock, B. M. Abraham, and D. W. Osborne, Suppl. Nuovo cimento **9**, 310 (1958).

[14] T. R. Roberts and S. G. Sydoriak, Phys. Rev. **93**, 1418 (1954); E. F. Hammel, *Progress in Low-Temperature Physics* (North-Holland Publishing Company, Amsterdam, 1955), Vol. 1, p. 78.

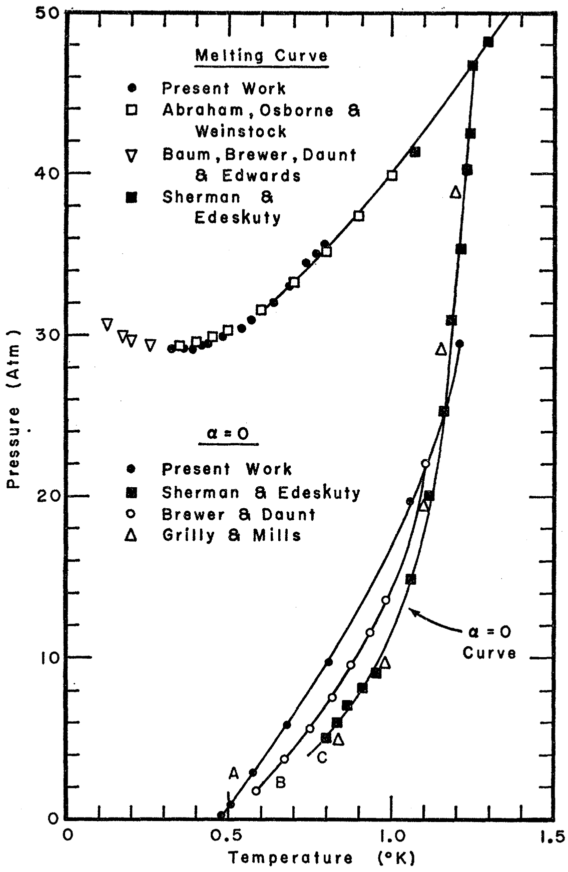

FIG. 8. Lower curve: the pressure at which α_p changes sign vs the temperature. Upper curve: the melting pressure of liquid He^3 vs temperature.

and subsequent warm-up with the helium in the capacitor at a pressure of about 36 atmospheres. Below about 0.8°K the capillary line was blocked with solid helium, thus keeping the helium in the cavity at constant density. Under this constant density condition, the maximum variation in frequency observed as the temperature changed was 1 cycle/sec. Thus frequency drift would not appear to be an important source of error.

Inasmuch as the compressibility of the liquid is large and the expansion coefficient relatively small, even a small drift in pressure during the warm-ups at constant pressure could introduce a large relative error in α. For pressures below 1 atmosphere the pressure was constant to better than 0.001 atmosphere as measured by a Hg manometer. At higher pressures no drift was observed although a change of ±0.05 atmosphere (the least count of the Heise Bourdon gauge) might conceivably have gone undetected. The error in α from this source is estimated to be less than ±0.001/°K.

Another possible error is introduced in the measurement of temperature by the thermal boundary resistance at the walls of the He^3 cavity. Using our previous estimates of the thermal boundary resistance,[1] the temperature of the He^3 would be about 0.002°K above that of the resistance thermometer at 0.2°K and the calculated $|\alpha_p|$ would be 2% too small. If the boundary resistance is indeed larger than our estimates, as suggested by Jeener and Seidel,[15] this correction would be correspondingly larger; but, in any event, the error from this source should be negligible above about 0.5°K because of the rapid decrease in boundary resistance with increasing temperature. The error in the temperature due to all causes is estimated to be no more than ±3%.

The estimated error in α_p due to all sources is estimated to be less than ±0.002/°K. The random error is much smaller.

V. DISCUSSION

By measuring the change in temperature on adiabatic expansion of liquid He^3, Brewer and Daunt[4] were able to determine values of α_p and $S_p - S_{\text{sat}}$ over much of the range of temperature and pressure of our data reported here. The *PVT* data of Sherman and Edeskuty[6] above 1°K allow a comparison near 1°K. Taylor and Kerr[5] have made a preliminary report of direct density measurements of the liquid at saturated vapor pressure below 1°K.

The general features of our data are consistent with the above measurements although there are systematic differences in detail. In Fig. 8 the pressure at which α_p changes sign is plotted as a function of temperature and compared with the data of other workers (Curves *A*, *B*, and *C*). The points of Sherman and Edeskuty[6] and Grilly and Mills[7] below 1°K were obtained by extrapolation of data obtained above 1°K.

In Fig. 9, α_p is plotted against the pressure at 1°K and at 0.5°K and compared with the data of Sherman and Edeskuty[6] and Brewer and Daunt,[4] respectively. Our values of α_p are, in general, above those of Brewer and Daunt; and, consequently, our values of $S_p - S_{\text{sat}}$ fall below those of Brewer and Daunt. (See Table I.) In view of the systematic nature of these discrepancies, it would appear that the source lies in systematic errors. Brewer and Daunt estimate the over-all random error in their values of α_p to be within ±0.0004/°K. They discuss also the possibility of systematic errors due to irreversibilities in the expansions of the liquid but present arguments for believing these errors not to be large. We believe that the random error in α_p is small but, as previously mentioned, cannot rule out an over-all error as large as ±0.002/°K, the principal source being a possible undetected small pressure drift. Thus the values of α_p and S_p as measured by these two quite different methods are consistent within the combined estimated error over most of this range of temperature and pressure.

Brewer and Daunt have discussed the implications of their measurements of α_p and S_p in this low-temperature range; although our results are slightly different,

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[15] J. Jeener and G. Seidel (private communication).

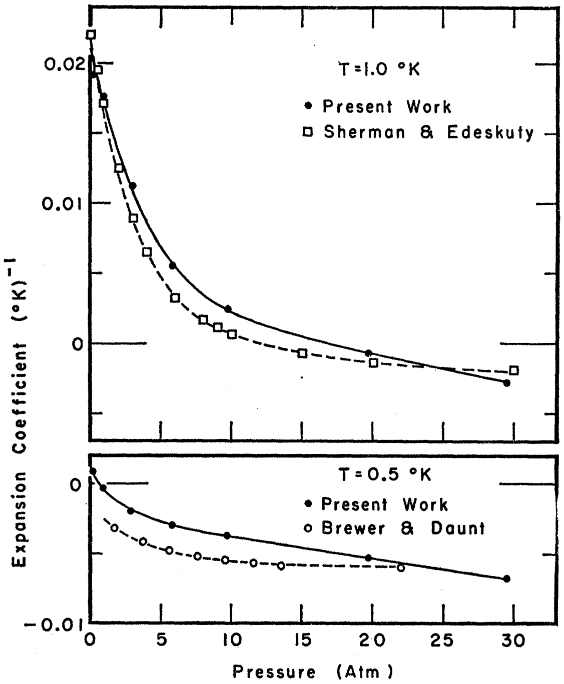

FIG. 9. The thermal expansion coefficient as a function of pressure at 1°K and 0.5°K.

this does not make any significant difference to their discussion.

VI. THE MELTING CURVE MINIMUM

The apparatus of this experiment provided a simple method of establishing the existence and position of the minimum in the melting curve of He^3.[8] At a constant temperature the pressure applied to the liquid He^3 capacitor was slowly increased and the frequency of the *LC* circuit monitored. At temperatures below 0.32°K the frequency steadily increased with increasing pressure until the pressure reached 29.1 atmospheres. Above this pressure no further increase in the frequency was observed, indicating a constant liquid density in the capacitor due to a solidification and blockage in the capillary fill line. Above 0.32°K as the pressure was raised the frequency took a sudden jump of several thousand cycles, indicating solidification of the He^3 in the capacitor. We infer, therefore, a minimum in the melting curve at $T=0.32\pm0.01$°K and $p=29.1\pm0.1$ atm. Recently, Baum, Brewer, Daunt, and Edwards[16] and Sydoriak, Mills, and Grilly[17] have reported direct measurements of the p and T at melting which extend below the minimum. Baum *et al.* find the minimum at $T=0.32$°K and $p=29.3\pm0.1$ atm, and Sydoriak *et al.* report the minimum at $T=0.330\pm0.005$°K and $p=28.91\pm0.02$ atm. Our values of the melting pressure and temperature above the minimum are shown in the upper curve of Fig. 8 along with representative values of other investigators.

ACKNOWLEDGMENTS

We wish to thank George Zimmerman, Sheldon Elliott, John Reppy, and Roger Walmsley for helpful suggestions and assistance with measurements.

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[16] J. L. Baum, D. F. Brewer, J. G. Daunt, and D. O. Edwards, Phys. Rev. Letters **3**, 127 (1959).

[17] S. G. Sydoriak, R. L. Mills, and E. R. Grilly, Phys. Rev. Letters **4**, 495 (1960).