Vapor Pressures of He³-He⁴ Mixtures*

S. G. Sydoriak and T. R. Roberts

Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico (Received December 7, 1959)

Vapor pressures, P_x , of He³-He⁴ mixtures ranging in liquid mole fraction, X, from 0.1 to 0.9 have been measured between 0.6° and 2.4°K versus the vapor pressure, P30, of liquid He3. Except for sharp breaks in the vicinity of the lambda and stratification temperatures of some of the mixtures, P_x/P_s^0 is found to vary slowly and smoothly with X and with temperature. Contrary to much of the work of other authors, there are no breaks at the He⁴ lambda temperature.

A comprehensive smoothed table of P_x/P_{3^0} is derived. Using this as a reference it is possible, for the first time, to intercompare all of the previously existing data on P_x . The data of some authors are in excellent agreement with our table but other data are in serious quantitative disagreement near 1.2°K and much previous data are in qualitative disagreement at the He⁴ lambda temperature.

INTRODUCTION

TAPOR pressures of He³-He⁴ mixtures have been reported by a number of workers. The most extensive series of measurements are those of Esel'son and Berezniak.¹ These workers and Sommers² discuss most of the early literature data.

An important practical use of such measurements is the correlation and smoothing of data into tables of vapor pressure, P_x , as a function of both X, the liquid mole fraction of He³, and temperature, T. Using such tables, the unknown composition of a liquid mixture may be determined from a measurement of its vapor pressure and temperature. For example, Sommers gives a smoothed table of $\log P_x$ for X between 0.00 and 0.13 and T between 1.2 and 2.2°K. In the present work vapor pressures were measured for nine liquid concentrations ranging in steps of 0.1 from X=0.1 to 0.9. The temperature ranged from 0.6 to 2.4°K and was determined from $P_{3^{0}}$, the vapor pressure of pure He³. The ratio, $R = 1000 P_x / P_{3^0}$, is found to be a slowly varying function of T and has been used to tabulate and graphically represent and analyze our data. A smoothed table of R as a function of X and T is presented and compared with results of other workers.

A second possible result of such measurements is the determination of the stratification temperature, $T_s(X)$, at which a liquid He³-He⁴ mixture first begins to separate into two layers having different compositions.³ Since the phases are in thermodynamic equilibrium, their vapor pressures are necessarily identical. Weinstock, Osborne, and Abraham⁴ applied this principle of coincidence of vapor pressures in an unsuccessful first attempt to demonstrate stratification, failing only

because the theoretical prediction⁵ on which they based their experiment gave a much too high value of T_s . Using a He³ bath to reach considerably lower temperatures, we demonstrate here the validity of the method and find it possible to determine T_s with moderate precision except near the consolute temperature, where the precision is very poor. A more precise determination of $T_s(X)$ has been made by first sound measurements.^{6,7}

A third result of careful measurements of P_x is the determination of the temperature, $T_{\lambda}(X)$, at which a He³-He⁴ mixture becomes superfluid by observing the temperature at which dP_x/dT is discontinuous.^{8,9} References to theoretical papers on the lambda transition in mixtures are given by Esel'son, Kaganov, and Lifshitz¹⁰ and by de Boer and Gorter.¹¹ In the present paper the determinations of $T_{\lambda}(X)$ from vapor pressure measurements are compared with and are shown to be in good agreement with most of the recent measurements of $T_{\lambda}(X)$ by other methods.^{6,7,12,13}

By the use of vacuum-jacketed pressure-sensing tubes and He³ vapor pressure thermometry it is felt that these measurements have minimized errors in X due to fractionation and errors in T due to He⁴ film reflux. The existence of such errors in previous measurements is made clear by a careful analysis of data appearing in the literature. For example, several sets of mixture data have shown breaks or irregularities in the slope of

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⁸ D. W. Osborne, B. M. Abraham, and B. Weinstock, Proceedings of the International Conference on the Physics of Very

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 ³ G. K. Walters and W. M. Fairbank, Phys. Rev. 103, 262-263

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 P_x versus T at the He⁴ lambda temperature.^{1,14-16} No such effect is apparent in our own or certain other measurements.^{4,17} Possible experimental causes of these inconsistencies are considered in the comparison of the various sets of data.

A fifth result of these measurements will be the subject of a separate paper. Wansink¹⁸ has calculated vapor composition and various thermodynamic functions from vapor pressure and density data for liquid concentrations up to 7% He³. Similar calculations are being performed for the entire range of compositions measured in the present experiments.

Finally, we may point to the fact that measurements of P_x have been of use^{2,19} in the evaluation of the various solution theories. As more extensive measurements of P_x become available and are analyzed the more recent theories of Chester^{20,21} and of Prigogine²²⁻²⁷ will be subject to similar tests of applicability.

EXPERIMENTAL

Some details of the experimental apparatus have been described elsewhere.^{28,29-31} We shall elaborate here only on those details which we believe pertinent to the accuracy of measurements of temperature and vapor pressure. Figure 1 is a schematic diagram of the interior of the Dewar. The chambers and tubes below the Dewar lid in the figure are surrounded by a vacuum. Thermal equilibrium is aided by putting the liquid He³ coolant, the He³ vapor pressure thermometer, and the mixture liquid, into a massive copper block having $\frac{1}{16}$ -inch thick walls separating the three liquids from each other and from the vacuum space.

Not shown at the top and bottom of the mixture cell are horizontal quartz crystals²⁹ used in these experi-

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- ³¹ S. G. Sydoriak and T. R. Roberts, Phys. Rev. 106, 175-182 (1957).



FIG. 1. He³ Dewar. A copper block, a, is hollowed out to leave $\frac{1}{16}$ -inch thick walls between a 15 cm³ annular well for liquid He³ b; a 1.9 cm³ cell for mixture liquid, c; and a 0.1 cm³ He³ vapor pressure bulb, d. The block is suspended in a separate vacuum space surrounded by copper, e, maintained $\ge 1.1^{\circ}$ K by liquid He⁴, f, in an eccentric annulus of 5 liters capacity. A $\frac{3}{16}$ -inch diameter constriction, i, minimizes film flow. The He⁴ pumping line has a diameter of 1 inch at j and increases in steps (not shown) to 6 inch diameter enroute to a 10 horsepower fore pump in the basement beneath the laboratory. The pumping and pressure-sensing tubes for He³, g, k, and n, have inside diameters of 2.7, 6.1, and 24.9 mm, respectively, in order that calculated pumping and thermomolecular pressure gradients be negligible in the lowest section and roughly the same in the middle and In the key sections of each line. Each section is about 30 cm long. A liquid nitrogen pot (not shown) keeps radiation shield, m, at To have a state of the main vacuum jacket (not shown) is sealed to the Dewar lid, p, by means of O rings. Section k of the mixture pressure-sensing tube was 1.4-mm i.d. in all experiments. In series A experiments tubes l and o were the same, 2.7-mm i.d. In series B runs l and o were 6.1- and 24.9-mm i.d., respectively.

ments as a liquid level indicator. When the liquid level reached the upper crystal it became possible to transmit sound between the crystals. Except for X=0.200 data, pressure measurements were carried out only under these conditions, which corresponded to the cell being at least 84% full. The minimum liquid volume for sound signals was 1.6 cm³.

For a number of reasons to be discussed later two sets of graduated pressure sensing tubes were used. Below the liquid He⁴ heat sink both sets were the same, consisting of 0.145-cm i.d. tubing. Above this level set A had an inside diameter of 0.270 cm all the way to room temperature while set B was enlarged to 0.608-cm i.d. up to the liquid nitrogen level and to 2.490-cm i.d. above this level. Each of the three sections was approximately 30 cm long. The He³ pressure-sensing tubing was identical in size with the set B mixture-sensing tubes.

Vapor Pressure Measurement Details

The use of He³ instead of He⁴ vapor pressure thermometry eliminates the possibility of a pressure gradient due to return flow of a vaporized He II film to the mother liquid. Also, due to the much higher vapor pressures of He³ and the large diameters of the pressure-sensing tubes the calculate d^{32} thermomolecular pressure ratio, $P_T/P_{300^{\circ}K}$ is nowhere less than 0.995 in the range of our measurements. The Weber-Schmidt thermomolecular pressure ratio equation has been found to fit within an accuracy of a fraction of a percent for experimental values as low as 0.500.32,33 Had He⁴ thermometry been used P_{cold}/P_{warm} would have fallen below 0.5 at 0.8°K.

In the mixture pressure measurements, when using the set A, small bore tubes, P_{cold}/P_{warm} was never less than 0.911, well above the experimentally proven range of the Weber-Schmidt equation. In further justification of the calculation of $P_{\rm cold}/P_{\rm warm}$, it was found that good agreement was obtained when using the large bore, set B, tubes.

Manometers and McLeod gauges used were similar to those described elsewhere,³¹ except that the mixture oil manometer was of smaller bore, 7 mm, in order to reduce the noxious volume of the mixture system. The oil manometers were frequently calibrated against mercury, at about 90 mm Hg, and were in turn used to check the McLeod gauge calibration at 3 mm Hg (43-mm oil). Detailed calibration of the McLeod gauges was carried out down to 0.100 mm Hg, using a traveling microscope clamped to the case of the oil manometer to obtain an accuracy of 0.002 mm Hg. Readings of the manometers during the actual vapor pressure measurements were done using a level slide-rule hairline in front of the manometer and a mirrored scale behind. With this system independent scale readings of the same pressure by two observers usually agreed to within 0.1 mm which was two or three times less than the scatter of the data from run to run.

Errors Due to Film Reflux Etcetera

A pressure gradient associated with reflux of an evaporating mobile film is not a likely source of error in our experiments because the high concentration of He³ in the vapor phase effectively prevents rapid diffusion toward the liquid surface of He⁴ vaporized from the film.³⁰

Actually a much more likely cause of serious error in pressure measurements involving He⁴ or He³-He⁴ mixtures is seldom mentioned in the literature. We refer to the likelihood of an appreciable temperature difference between the liquid and the walls of its container, due to heat entering the liquid via condensation at its

surface. For example, consider a cylindrical He⁴ vapor pressure bulb of diameter, D, and height, h, connected to a vacuum jacketed pressure sensing tube of the same diameter. At 1°K, the total heat flux due to condensation of the refluxing film will be 0.7D milliwatt or greater, depending on the surface over which the film flows.^{34,35} This heat must flow across a boundary separating the liquid from the rest of the apparatus, the area of the boundary being πDh if we neglect the area of the bottom of the bulb. The temperature drop across the liquid-solid boundary will therefore be³⁶

$$\Delta T = Q/0.0222AT^2 = (0.010/h)$$
 degrees,

if *h* is in centimeters.

The results of this calculation were mentioned some time ago.³⁷ The above details are included here for the purpose of calling renewed attention to the importance of providing an adequately large refrigerated wall surface in a He⁴ vapor pressure bulb.

The above calculation is also pertinent to the discussion of our measurements of P_x . For reasons not yet completely understood there was an appreciable heat influx to our mixture liquid beginning at pressures of the order of 3 mm Hg and increasing rapidly with decreasing pressure.³⁰ For a 40% He³ liquid mixture this anomalous heat leak had risen to 0.076 milliwatt by 0.7° and 0.093 milliwatt by 0.6°K. Taking 8 cm² for the surface area of the sides and bottom of the mixture cell we estimate the temperature of this mixture to have been in excess of the He³ thermometer by 0.0008° and 0.0014°K when at 0.7°K and 0.6°K, respectively. For X > 0.4 the effect was less. For X < 0.4the anomalous heat leak was not measured.

Liquid Concentration

From time to time the ratio X/(1-X) of He³ to He⁴ molar concentration of each of the nine stored mixtures was measured on a Nier-Consolidated mass spectrometer, to an accuracy of $\pm 1\%$. The accuracy of the measurement of X therefore varied linearly from $\pm 0.9\%$ at X=0.1 to $\pm 0.1\%$ at X=0.9.

Fractionation of a liquid He³-He⁴ mixture can cause its concentration to differ considerably from the known average concentration of the input gas. As an example, we note in the data of reference 1 that for X=0.10the vapor concentration, X_v , will be 0.34 at 2.4°K and will rise to 0.80 at 1.4°K. If, as is usually the case, only the average concentration of the input gas is known, then X will be known exactly only if the mass and composition of the vapor in the system are known. To minimize the uncertainty in liquid concentration due to fractionation, we have chosen a Dewar design

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³⁷ William E. Keller, Phys. Rev. 98, 1571-1575 (1955).

and operating procedure such that formation of He^3 rich vapor is kept to a minimum. As a check on the efficacy of these measures we have repeated many of the measurements with a much larger pressure-sensing tube and with differing volumes of vapor in the cell itself.

Evaporation of the mixture is prevented by following the procedure of starting a run by filling the cell at a high temperature and going, usually monotonically, to lower temperatures during the run. If a check point at a significantly higher temperature is needed the cell is emptied completely, the gas collected in the storage can is mixed thoroughly by pumping it at 8 liters/min for about half an hour out one port of a 12-liter storage can and into another port, and a new run is then started.

The procedure of progressively reducing temperature will not necessarily be sufficient to prevent appreciable vaporization of mixture liquid in an ordinary immersiontype Dewar. Some apparatus used elsewhere for mixture vapor pressure measurements has consisted of unjacketed pressure-sensing tubes immersed in liquid He⁴. With such a Dewar the high efflux of vapor accompanying a reduction in temperature will cool the entire pressure sensing tube and may conceivably result in a transient increase in m_g , the mass of vapor present in the tubing. Ultimately a new and lower equilibrium value of m_g will exist because of the reduced pressure, but there will be an increase in the He³ concentration of a large portion of the vapor mass every time the bath temperature is changed.

A second and possibly more serious cause of difficulty in measurements done with such an immersion-type He⁴ Dewar is the increase in vapor volume which will occur when in the He II region when the temperature is reduced. At progressively lower temperatures the film will rise to that higher level on the outside of the pressure sensing tubing at which the increased conduction heat leak just balances the increase in film flow available for vaporization. Hence a net increase in m_a and resultant decrease in X may occur despite the decreased pressure. We shall see later that measurements done elsewhere with unjacketed mixture pressuresensing tubes show peculiarities near the He⁴ lambda temperature which can be interpreted as above in terms of the effect of the He II film on m_g and hence on the He³ concentration in the liquid.

These difficulties cannot arise in the Dewar of Fig. 1 because the mixture pressure-sensing line is in a vacuum, it is surrounded by a constant temperature vacuum jacket and it makes thermal contact only with heat sinks held at fixed temperatures greater than the highest cell temperature during a run.

By the above described design and procedure, mixing of high He³ content vapor and low He³ content input gas is limited to that which can occur by the slow process of diffusion. There will be a tendency to counteract even this slow mixing process because of the condensation of vapor near the liquid surface which precedes each arrival at a new, lower pressure, data point. Direct experimental evidence for the effectiveness of these measures for avoiding errors in X will be given in the discussion of results.

RESULTS

Results of the measurements are given in Table I along with the deviation of each data point from a smoothed table (Table II) whose origin will be described later. In 20 of the runs the set A (small bore) sensing tubes were used. In later runs mixture pressures were transmitted to the gauges via the set B tubes.

Despite the order of magnitude enlargement of tubing cross sections after the first 20 runs it is seen that there is no systematic change in the fit of the data to the smoothed table. As further evidence for the avoidance of errors in X due to fractionation we call attention to the runs for X=0.200, in which the fraction of cell volume occupied by vapor was 16%, 18%, and 23%, respectively, in the three runs. We note that there is no systematic shift in the column of deviations, even for the higher pressures, where fractionation is most likely to have an effect on the liquid concentration.

Graphical representation of that portion of the data above T_{\bullet} is given in Fig. 2 as a plot of P_x/P_3^0 versus T. This is seen to be a most satisfactory way of presenting such data because the ratio P_x/P_3^0 changes so slowly with temperature; e.g., for several mixtures we observed a 200-fold change in P_x but for no mixture was there more than a twofold change in P_x/P_3^0 . Using this as an ordinate all of the data can be displayed with high sensitivity on a single figure. For purposes of smoothing the data this is of considerable advantage over the use of several figures covering different ranges of pressure.

The lambda line, $R_{\lambda} = f(T_{\lambda})$, is plotted as a dashed line in Fig. 2. In constructing this line we have used for T_{λ} results consistent with second sound velocity measurements,¹² and measurements of first sound velocity and attenuation.^{6,7} For mixtures of 70 and 80% He³, T_{λ} 's were taken from our tentative thermal conductivity measurements.³⁰ R_{λ} 's were obtained by interpolating our data on R = f(T) to the temperature T_{λ} .

Where the lambda line intersects the solid lines drawn through the data we find a more or less abrupt change in slope. The discontinuity is made more apparent by plotting deviations from T_{λ} and from $(P_x/P_3^0)_{\lambda}$ as in Fig. 3. We note that there are abrupt changes in slope at T_{λ} for low values of X but that for X>0.5 the change in slope is not so striking.

As described by de Boer and Gorter³⁸ the change at the lambda point in the specific heat of the liquid at constant pressure, ΔC_P , can be calculated from the change in slope of the vapor pressure at T_{λ} provided we also know P_3 and P_4 , the partial pressures of He³ and He⁴. Preliminary values of ΔC_P have already been

³⁸ J. de Boer and C. J. Gorter, Physica 18, 565-568 (1952).

TABLE I. ΔR is the observed value of R minus that calculated using Columns (a), (b), and (c) of Table II, where R is 1000 times the ratio of mixture vapor pressure, P_x , to pure He³ vapor pressure, P_3^0 . The temperature, T_E , is derived from P_3^0 , using the scale of reference 31. Subheadings give the He³ mole fraction, X. Asterisks denote readings taken in the region of stratification, for which Column (a) of Table II is used to calculate ΔR . Between runs the cell is completely emptied. The beginning of a new run is designated by symbol "A" for runs made with the set A, small bore, pressure sensing tubes and by "B" for runs with the set B, large bore, tubes. One quarter of the entries represent a single measurement of P_x and P_3^0 and three quarters are the average of two such readings taken several minutes apart in time and within a few millidegrees in temperature.

P_x	P_{3}^{0}	T_E	ΔR	P_x	$P_{3}{}^{0}$	T_E	ΔR	P_x	P_{3}^{0}	T_E	ΔR
	X = 0.10	00			X = 0.30	01			X = 0.39	98	
$\begin{array}{c} 120.99 \ A \\ 107.02 \\ 91.58 \\ 75.68 \\ 62.81 \\ 56.16 \\ 50.83 \\ 47.30 \\ 42.12 \\ 38.48 \\ 35.04 \\ 28.18 \\ 22.85 \\ 18.17 \\ 14.27 \\ 11.07 \\ 8.31 \\ 5.84 \\ 4.23 \\ 4.20 \end{array}$	$\begin{array}{c} x = 0.10\\ 367.4\\ 332.3\\ 292.1\\ 249.8\\ 214.4\\ 195.2\\ 179.6\\ 169.2\\ 152.1\\ 139.0\\ 126.5\\ 102.0\\ 82.76\\ 65.40\\ 50.74\\ 38.51\\ 28.01\\ 18.70\\ 12.81\\ 18.70\\ 12.81\\ \end{array}$	2.573 2.499 2.406 2.303 2.206 2.149 2.100 2.066 1.906 1.908 1.908 1.908 1.800 1.704 1.604 1.504 1.404 1.301 1.185 1.090	$\begin{array}{c} \cdots \\ \cdots \\ 1 \\ -1 \\ -1 \\ -2 \\ -2 \\ -2 \\ -2 \\ -1 \\ -1$	$\begin{array}{c} 82.66\\ 67.58\\ 56.74\\ 44.44\\ 51.34\\ 40.87\\ 37.18\\ 33.94\\ 30.62\\ 25.20\\ 27.97\ A\\ 19.49\\ 15.46\\ 11.54\\ 8.31\\ 5.76\\ 3.40\\ 1.878\\ 1.298\\ 0.295\end{array}$	$\begin{array}{c} X = 0.36\\ 179.4\\ 148.6\\ 125.6\\ 98.56\\ 113.9\\ 90.42\\ 81.35\\ 72.45\\ 63.80\\ 50.35\\ 57.12\\ 37.00\\ 28.06\\ 19.84\\ 13.51\\ 8.81\\ 4.78\\ 2.449\\ 1.602\\ 1.602\end{array}$	2.099 1.993 1.904 1.784 1.854 1.744 1.646 1.646 1.593 1.500 1.549 1.391 1.302 1.201 1.102 1.006 0.889 0.784 0.726	$\begin{array}{c} 0 \\ -2 \\ -2 \\ -2 \\ -1 \\ -2 \\ 2 \\ 1 \\ 0 \\ 1 \\ 1 \\ 0 \\ 0 \\ 0 \\ -5 \\ 3 \\ \end{array}$	$\begin{array}{c} 53.87\\ 43.73\\ 38.88\\ 34.74\\ 31.05\\ 34.73\\ 30.94\\ 27.58\\ 24.68\\ 21.80\\ 19.51\\ 14.75\\ 10.91\\ 7.62\\ 6.28\\ 4.90\\ 3.81\\ 2.95\\ 2.296\\ 4.20\end{array}$	$\begin{array}{c} X = 0.39 \\ 101.49 \\ 82.39 \\ 73.08 \\ 65.06 \\ 57.81 \\ 65.29 \\ 57.67 \\ 50.44 \\ 43.97 \\ 37.74 \\ 33.09 \\ 23.80 \\ 16.74 \\ 11.05 \\ 8.85 \\ 6.67 \\ 5.01 \\ 3.77 \\ 2.867 \\ \end{array}$	1.798 1.792 1.650 1.601 1.554 1.602 1.553 1.501 1.451 1.398 1.354 1.354 1.252 1.156 1.055 1.007 0.950 0.897 0.849 0.806	$\begin{array}{c} 3\\ 2\\ 1\\ 0\\ -2\\ -2\\ -2\\ -4\\ 1\\ 0\\ -1\\ -1\\ 0\\ 1\\ -1\\ 1\\ -1\\ 1\\ -1\\ 1\\ -1\\ 1\\ -1\\ 1\end{array}$
3.40 2.126 1.221	9.88 5.56 2.924	0.916	$-2 \\ -4 \\ -3$	0.935 0.672 42.65 B	0.769	0.682	5 1 4	1.038 1.145 1.872	1.950	0.752 0.703*	3 2 0
0.707	1.520	0.720	2	40.91 40.00	90.07 88.02	1.742	1	1.072	X = 0.50	0.771	Ŭ
$\begin{array}{c} 89.54 \ B\\ 74.44\\ 6.89\\ 4.66\\ 2.680\\ 1.657\\ 0.987\\ 141.42 \ B\\ 118.57\\ 98.98\\ 82.67\\ 78.90\\ 75.81\\ 68.04\\ 56.66\\ 49.88\\ 46.86\\ 44.94\\ 43.20\\ 41.76\\ 40.00\\ 38.38\\ 33.66\\ 26.92\\ 20.12\\ 14.54\\ 9.97\\ 49.23 \ B\\ 41.76\\ 31.68\\ 21.15\\ 12.58\\ 2.702\\ 1.627\\ 0.857\\ 0.411\\ 0.271\\ \end{array}$	$\begin{array}{c} 232.4\\ 196.4\\ 196.4\\ 13.42\\ 8.42\\ 4.37\\ 2.484\\ 1.371\\ 336.2\\ 288.6\\ 248.4\\ 213.0\\ 205.6\\ 198.5\\ 179.8\\ 150.6\\ 134.6\\ 126.7\\ 121.8\\ 117.6\\ 112.7\\ 107.1\\ 102.0\\ 87.60\\ 67.21\\ 47.97\\ 32.73\\ 20.86\\ 133.7\\ 112.9\\ 81.50\\ 51.40\\ 27.48\\ 4.43\\ 2.465\\ 1.176\\ 0.511\\ 0.316\\ \end{array}$	$\begin{array}{c} 2.257\\ 2.153\\ 1.101\\ 0.996\\ 0.874\\ 0.785\\ 0.708\\ 2.507\\ 2.400\\ 2.300\\ 2.202\\ 2.180\\ 2.100\\ 2.000\\ 1.940\\ 1.920\\ 1.940\\ 1.940\\ 1.940\\ 1.940\\ 1.940\\ 1.888\\ 1.870\\ 1.849\\ 1.824\\ 1.801\\ 1.729\\ 1.614\\ 1.482\\ 1.350\\ 1.215\\ 1.936\\ 1.295\\ 0.876\\ 0.784\\ 0.689\\ 0.602\\ 0.561\\ \end{array}$	$\begin{array}{c} -3 \\ -2 \\ 0 \\ 0 \\ 2 \\ 0 \\ \cdots \\ 7 \\ 4 \\ 1 \\ 0 \\ 3 \\ -1 \\ 1 \\ -4 \\ -1 \\ 1 \\ 1 \\ 2 \\ -1 \\ 0 \\ -2 \\ -2 \\ -4 \\ 1 \\ -5 \\ -5 \\ 1 \\ \cdots \end{array}$	$\begin{array}{c} 39.15\\ 38.28\\ 37.40\\ 35.90\\ 35.33\\ 33.99\\ 37.30\\ 41.94\\ B\\ 41.55\\ 40.76\\ 39.84\\ 40.64\\ 39.90\\ 38.92\\ 38.08\\ 37.42\\ 36.76\\ 35.91\\ 34.62\\ 37.00\\ 28.15\\ 20.05\\ 10.80\\ 19.87\\ B\\ 9.32\\ 4.67\\ 3.54\\ 1.819\\ 0.992\\ 0.730\\ 0.633\\ 0.548\\ 0.464\\ 0.384\\ 0.313\\ 137\\ 32\\ A\end{array}$	$\begin{array}{c} 86.13\\ 84.02\\ 81.91\\ 77.80\\ 76.20\\ 72.66\\ 81.83\\ 93.22\\ 92.45\\ 90.45\\ 88.29\\ 90.24\\ 88.41\\ 86.18\\ 84.09\\ 82.27\\ 80.36\\ 78.03\\ 74.50\\ 81.29\\ 57.68\\ 38.09\\ 18.25\\ 37.92\\ 15.44\\ 6.88\\ 5.01\\ 2.351\\ 1.186\\ 0.848\\ 0.720\\ 0.613\\ 0.514\\ 0.420\\ 0.340\\ X=0.39\\ 251.2\\ \end{array}$	1.722 1.710 1.699 1.678 1.647 1.699 1.758 1.754 1.754 1.754 1.754 1.754 1.754 1.754 1.754 1.754 1.754 1.753 1.744 1.733 1.743 1.743 1.744 1.733 1.744 1.733 1.744 1.733 1.744 1.733 1.744 1.733 1.744 1.752 1.711 1.691 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.696 1.553 1.401 1.178 1.658 1.658 1.658 1.658 1.656 1.553 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.401 1.178 1.658 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.555 1.656 1.557 1.556 1.557 1.556 1.557	$\begin{array}{c} 2\\ 2\\ 3\\ 3\\ 3\\ 2\\ -2\\ -3\\ -1\\ -2\\ -2\\ -2\\ -1\\ 1\\ 1\\ 1\\ 1\\ 0\\ 0\\ 2\\ 3\\ 0\\ 0\\ 1\\ 0\\ 2\\ 4\\ -5\\ 0\\ -1\\ 2\\ 0\\ -1 \end{array}$	$\begin{array}{c} 134.66\ A\\ 136.56\\ 110.20\\ 91.52\\ 75.72\\ 60.31\\ 50.04\\ 39.64\\ 31.12\\ 27.25\\ 23.78\\ 20.76\\ 20.42\\ 17.78\\ 15.59\\ 13.28\\ 8.87\\ 7.06\\ 3.85\\ 27.31\ A\\ 4.53\\ 0.571\\ 0.523\\ 0.832\\ 1.251\\ 1.815\\ 2.632\\ 155.8\ A\\ 17.42\ A\\ 11.33\\ 7.77\\ 5.74\\ 2.98\\ 135.06\ A\\ 103.11\\ 86\ 07\\ \end{array}$	$\begin{array}{c} 216.4\\ 220.4\\ 179.0\\ 149.5\\ 124.3\\ 99.14\\ 82.34\\ 65.18\\ 50.94\\ 44.40\\ 38.50\\ 33.32\\ 32.66\\ 28.04\\ 23.97\\ 19.88\\ 12.50\\ 9.66\\ 4.92\\ 44.41\\ 5.90\\ 0.638\\ 0.952\\ 1.467\\ 2.175\\ 3.255\\ 248.6\\ 27.47\\ 16.64\\ 10.85\\ 7.71\\ 3.74\\ X=0.59\\ 195.99\\ 151.06\\ 126.31\\ \end{array}$	2.212 2.224 2.098 1.996 1.898 1.787 1.702 1.602 1.505 1.454 1.404 1.356 1.350 1.302 1.254 1.202 1.084 1.202 1.084 1.454 0.926 0.657* 0.665* 0.716* 0.665* 0.665* 0.716* 0.827 2.300 1.295 1.154 1.051 0.979 0.848	$\begin{array}{c} 3\\ -1\\ -1\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\ 0\\$
117.35 A 98.79	X = 0.30 249.6 212.4	01 2.303 2.200	$-1 \\ 1$	$115.53 \\ 97.09 \\ 80.49 \\ 66.54$	213.0 180.2 150.5 125.2	$2.202 \\ 2.102 \\ 2.000 \\ 1.902$	2 2 1 2	$\begin{array}{c} 69.69 \\ 56.36 \\ 44.49 \\ 34.26 \end{array}$	$102.42 \\ 83.00 \\ 65.55 \\ 50.50$	$ 1.802 \\ 1.705 \\ 1.604 \\ 1.502 $	$ \begin{array}{c} 0 \\ -1 \\ 0 \\ -1 \end{array} $

$ \begin{array}{c c c c c c c c c c c c c c c c c c c $												
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	P_x	$P_{3^{0}}$	T_E	ΔR	P_x	P_{3}^{0}	T_E	ΔR	P_x	$P_{3^{0}}$	T_E	ΔR
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		X = 0.59	97			X = 0.69	99			X = 0.79	19	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	24.87	36.47	1.386	0	21.20	27.98	1.301	3	2.86	3 39	0.832	-3
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	20.62	30.07	1.323	Ŏ	15.24	20.02	1.204	1	1.951	2.290	0.774	7
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	17.52	25.46	1.272	-2	10.67	13.91	1.109	ī	1.369	1.599	0.726	-1
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	14.90	21.46	1.223	ō	8.55	11.08	1.057	$-\overline{1}$	1.004	1.159	0.688*	Ō
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	12.50	17.81	1.172	ŏ	6.98	8.96	1.010	ō	0.695	0 790	0.665*	-4
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	10.55	14.82	1.125	1	5.38	6.85	0.955	-1	0.619	0.704	0.634*	-10^{-1}
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	8.60	11.92	1.073	õ	4.12	5.21	0.904	-3	12.07 B	14.47	1.119	4
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	7.20	9.75	1.028	ž	3.24	4.06	0.861	-2	7.28	8 75	1 005	-3
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	5.88	7.77	0.980	2	2.724	3.385	0.832*	ō	5.65	6.77	0.953	-3
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	4.43	5.71	0.920	1	134.72 A	176.6	2.090		4.31	5.17	0.903	-4
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	3.40	4.26	0.869	2	131.71	172.8	2.077		3.16	3.77	0.849	$-\overline{6}$
	2.584	3.188	0.822*	0	98.54	130.2	1.922	-1	2.714	3.216	0.824	-2
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	1.932	2.325	0.776*	1						0.220		-
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	1.386	1.622	0.728*	4		X = 0.70	0			X = 0.89	7	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	1.014	1.168	0.688*	1	94.77 A	125.1	1.902	0	137.72 A	150.9	2.003	-1
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.709	0.800	0.646*	2	100.39	132.8	1.933	-2	115.73	127.2	1.910	-1
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	19.55 A	28.51	1.307	-1	108.82	144.1	1.976	-4	92.76	101.93	1.800	0
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	13.96	20.02	1.203	0	123.47	162.4	2.039	0	73.10	80.42	1.691	-1
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	9.94	13.93	1.110	0	111.38	146.4	1.985	2	59.05	64.99	1.600	0
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	19.70 A	28.79	1.310	-3	113.34	149.0	1.994	2	45.92	50.64	1.502	-1
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	9.88	13.89	1.109	-3	116.28	153.7	2.011	•••	34.85	38.47	1.404	0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	6.25	8.36	0.995	-1		V. 0.70	0		25.44	28.09	1.302	-2
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	3.82	4.85	0.892	1		A = 0.79	6		18.16	20.11	1.205	-1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	33.94 B	49.91	1.497	1	130.75 <i>B</i>	156.5	2.022	• • •	12.54	13.84	1.108	0
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	23.38	34.05	1.363	2	67.76	81.49	1.697	1	8.00	8.80	1.006	2
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	17.34	25.07	1.268	1	20.13	24.30	1.259	1	4.61	5.07	0.899	0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	11.74	16.72	1.155	-4	6.27	7.50	0.973	-1	21.69 A	23.94	1.254	0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	8.44	11.67	1.068	0	5.16	6.16	0.934	-2	18.23	20.17	1.206	-2
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	5.88	7.78	0.980	1	3.49	4.15	0.865	4	2.428	2.660	0.796	2
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	4.46	5.73	0.921	3	2.728	3.232	0.825	5	1.350	1.478	0.730	-2
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	3.24	4.04	0.860	6	0.836	0.952	0.665*	2	0.751	0.819	0.649	-2
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	0.547	0.608	0.619*	3	1.792 B	2.107	0.762	-3	0.555	0.600	0.618	0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$		Y = 0.60	18		1.197	1.383	0.708	8	0.451	0.484	0.597	-3
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		A = 0.09			0.710	0.802	0.647*	2	0.916	1.002	0.671	-5
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	13.63 B	17.80	1.172	4	122.91 B	147.7	1.990	-1	7.94 A	8.76	1.005	-1
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	6.93	8.90	1.008	2	73.30	88.23	1.733	0	4.70	5.17	0.902	2
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	7.18	9.25	1.016	2	0.355	0.387	0.577*	3	2.309	2.532	0.788	1
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	21.46 B	28.17	1.303	7	0.670	0.753	0.640*	4	1.200	1.317	0.702	-5
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	10.76	14.01	1.111	2	1.071	1.242	0.695*	0	0.983	1.073	0.678	-2
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	5.22	6.66	0.950	-1	1.197	1.392	0.709	1	0.751	0.821	0.649	-5
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	3.92	4.94	0.894	1	1.358	1.582	0.725	1	0.566	0.615	0.620	-8
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	3.16	3.92	0.850	-0	1.515	1.768	0.739	2	0.461	0.494	0.599	5
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2.73	3.38	0.832*	3	1.722	2.017	0.757	1	2.158	2.366	0.779	1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2.5/1	3.100	0.821*	3	2.249	2.041	0.794	3	3.372 A	3.716	0.846	-2
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2.102	2.540	0.789*	4		X = 0.79	9		2.472	2.704	0.798	3
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.185	1.305	0.707*	0	124 00 4	160.0	2 0.24	6	1.920	2.097	0.762	4
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.193	1.390	0.710	0	134.82 A	100.2	2.034	0	1.58/	1.737	0.737	-1
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	0.755	0.828	0.030	3	115.25	105.0	1.942	4	1.304	1.494	0.718	-1
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	0.013	0.084	0.031*	4	88.01	105.4	1.817	2	1.234	1.352	0.706	-2
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.031	1.188	0.090	2	09.23	83.17	1.700	1	0.915	0.997	0.670	3
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.045	1.959	0.755	1	54.51	03.39	1.003	2	2.592 A	2.842	0.805	1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$		X = 0.69	9		41.59	20.27	1.500	-3	1.480	1.605	0.727	9
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	125 25 1	164.6	2 050	1	31.4/ 32 20	30.00 78 10	1 202	-2	0.608	0.662	0.627	-7
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	123.23 A	1 020	2.030	1	23.30	20.10	1.505	2	0.296	0.309	0.558	• • •
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1 1 1 2 2	1.029	0.074	1	13.74.71	16 67	1.434	1	$6.27 \ B$	6.89	0.956	3
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2.404	2.734	0.735	1	0.07	10.07	1.155	0	1.622	1.773	0.740	1
2,700 0.711 0.001 1 0.01 0.00 0.971 1 0.000 0.300 0.326 0.562 0.000	2.030	2.419	0.705	1	5 51	6 58	0.047	1	0.502	0.539	0.608	3
	0.859	0.984	0.669*	-3	4.20	4,97	0.896	3	0.309	0.326	0.562	

TABLE I.-Continued.

calculated from our earlier results.²⁸ A recalculation is under way which will make use of the more complete vapor pressure data depicted in Fig. 3 and which will avoid the use of existing experimental vapor composition data,^{1,2} whose accuracy is not sufficiently high for this purpose. concentration, X_L and X_H . Since they are in thermodynamic equilibrium their Gibbs free energy and hence their vapor pressures, P_s , must necessarily be identical. Furthermore, all other liquids having an average concentration anywhere in the range X_L to X_H will have a vapor pressure equal to P_s at this same temperature since they will also be separated into two layers having the concentrations X_L and X_H . Figure 2 shows

At its stratification temperature, T_s , a mixture will separate into two conjugate phases of low and high He³

TABLE II. Smoothed values of $R = 1000P_X/P_s^0$, at stratification temperatures [Column (a)], at lambda temperatures [Column (b)], and at 0.1°K temperature intervals [Columns (c)], for values of X, the He³ mole fraction. Parentheses enclose values of T_s below the range of our measurements and the somewhat uncertain interpolations near the consolute temperature. In Columns (c) asterisks separate He II and He I phases and the letter "S" designates the region of stratification. Question marks denote our unpublished and provisional values of T_λ obtained from heat flow experiments which have not yet been checked against the possibility that the superfluidity of the conjugate of a stratified mixture was inadvertently being detected. For purposes of extrapolation of Columns (c) it should be remembered that for each value of X the curve of R versus T terminates at T_s . When interpolating near T_λ one should also note that the slope changes abruptly at T_λ , except for X=0, for which there is an inflection at T_λ .

X	(a <i>Ts</i>) Rs	(b) T)	R	0.6°	0.7°	0.8°	0.9°	1.0°	(c). 1.1°	Values	s of <i>R</i> 1.3°	at the f 1.4°	ollowir 1.5°	ng ten 1.6°	iperat 1.7°	ures: 1.8°	1.9°	2.0°	2.1°	2.2°	2.3°
0.00 0.02 0.04 0.06 0.08	(0.00) (0.08) (0.16) (0.22) (0.27)		2.1735 2.145 2.116 2.087 2.058	186 205 223 240 260	1 166 290 395 480	2 128 230 319 405	4 106 195 277 356	8 95 173 249 319	14 89 160 228 292	22 89 152 214 273	31 91 150 205 259	43 98 150 201 250	56 107 154 200 246	71 117 161 204 245	87 129 170 209 245	104 142 179 214 247	122 156 189 221 249	140 170 199 228 253	157 184 210 234* 257*	174* 199* 221* 241 263	190 211 232 252 272	205 225 245 264 284
0.10 0.12 0.14 0.16 0.18	(0.31) (0.35) (0.38) (0.42) (0.45)		2.028 1.998 1.967 1.936 1.904	279 298 316 334 352	558 629 687 735 775	478 547 605 652 692	425 487 541 586 623	384 440 490 532 567	352 405 450 490 522	328 377 418 455 485	309 354 392 426 456	296 337 371 403 432	287 324 356 385 411	281 315 345 371 395	279 309 334 360 381	277 303 327 350 369	277 301 323 343 360	278 299* 318* 336* 352*	279* 298 319 337 355	285 305 324 342 359	293 313 331 348 365	302 322 339 357 374
0.20 0.22 0.24 0.26 0.28	0.477 0.507 0.536 0.563 0.588	922 909	1.873 1.840 1.807 1.773 1.738	368 386 402 419 436	805 834 859 879 899	726 751 771 792 810	654 681 704 724 742	597 625 648 669 687	551 579 601 621 640	513 538 561 582 599	482 505 528 548 565	456 480 500 519 536	434 455 476 493 510	416 436 454 471 486	401 420 435 452 465	387 403 418 431* 443*	375* 389* 403* 418 435	369 386 403 420 435	373 390 406 423 439	378 393 410 427 443	384 400 416 433 448	391 407 423 438 454
0.30 0.32 0.34 0.36 0.38	0.614 0.636 0.658 0.679 0.700	898 889 880 871 863	1.704 1.667 1.630 1.592 1.553	453 472 490 507 526	5 5 5 5 5 5	824 835 847 856 863	757 771 782 793 802	703 718 730 740 749	656 670 683 694 704	616 630 642 654 664	581 596 608 620 630	551 565 578 589 599	524 537 549 560 570	500 512 522 532* 540*	477 485* 496* 506 520	454* 470 486 501 516	451 467 483 499 515	452 468 484 500 516	455 472 487 503 519	459 476 492 507 523	464 479 496 511 527	470 485 501 516 532
$\begin{array}{c} 0.40 \\ 0.42 \\ 0.44 \\ 0.46 \\ 0.48 \end{array}$	0.718 0.737 0.753 0.769 0.783	854 846 839 833 827	$1.514 \\ 1.472 \\ 1.430 \\ 1.387 \\ 1.342$	545 564 581 600 617	5 5 5 5 5 5	5 5 5 5 5 5	807 812 816 818 820	757 763 768 772 775	713 720 726 731 734	673 681 687 693 697	638 646 652 658 662	606 614 620 626* 630*	577 584* 590* 596 607	547* 560 572 584 597	536 550 565 578 593	530 545 561 576 592	530 546 562 578 593	532 548 564 579 595	536 551 567 583 598	539 554 570 585 601	542 558 574 588 604	547 563 578 593 608
0.50 0.52 0.54 0.56 0.58	0.796 0.809 0.822 0.832 0.841	821 817 811 806 803	$\begin{array}{c} 1.297 \\ 1.250 \\ 1.203 \\ 1.154 \\ 1.105 \end{array}$	635 653 671 690 709	5 5 5 5 5 5 5	5 5 5 5 5 5 5	821 S S S S	777 778 780 781 782	738 740 742 744 746	701 703 706 708* 711*	666* 670* 672* 681 689	634 644 654 665 677	618 630 643 656 669	610 624 638 652 667	607 621 636 650 666	607 621 636 651 666	608 623 638 653 667	609 625 640 654 668	612 627 642 656 671	616	619	623
0.60 0.62 0.64 0.66 0.68	0.848 (0.86) (0.87) (0.87) (0.86)	800	1.055 1.005 0.954 0.904 0.876?	728 748 766 786	5 5 5 5 5 5 5	5 5 5 5 5 5 5 5 5 5	S S S S S S S	783 785 785* 787* 789	747* 749* 754 762 770	717 725 735 745 756	699 710 723 735 747	689 701 716 729 743	683 696 712 726 740	681 696 711 726 740	681 696 711 726 741	682 697 711 727 742	682 697 713 728 743	683 698 713 728 743	684 699 715 729 745			
0.70 0.72 0.74 0.76 0.78	0.836 0.811 0.781 0.754 0.724	805 815 828 839 852	0.840? 0.820? 0.800? 0.777? 0.753?		5 5 5 5 5 5	5 5 5 5 5	S 825 832 840	793 799 807 817 829	779 788 798 809 822	767 780 792 805 818	761 774 788 802 816	756 771 785 801 815	755 770 784 798 814	755 770 785 800 815	756 770 786 800 816	756 771 787 802 817	757 772 787 802 818	758 773 788 803 819	759 774 789 804 820			
0.80 0.82 0.84 0.86 0.88	$\begin{array}{c} 0.695 \\ 0.667 \\ 0.636 \\ 0.604 \\ 0.570 \end{array}$	863 877 890 903 920	0.723? 0.695? 0.654? 0.620? 0.580?		S S 905 917	861 871 880 892 904	849 860 871 884 898	841 852 865 880 894	835 848 862 876 892	831 846 860 875 892	829 844 859 876 891	829 844 860 876 892	829 845 860 876 892	831 846 862 878 893	831 848 863 879 895	832 848 864 879 896	833 848 864 879 896	834 849 865 880 896	834 850 866 881 897			
0.90 0.92 0.94 0.96 0.98	0.531 (0.48) (0.43) (0.36) (0.26)		0.542?		930 942 956 970 983	918 933 947 964 981	912 928 943 961 980	910 927 942 960 979	908 925 941 959 979	908 925 942 960 980	908 926 944 962 980	909 927 945 963 981	909 927 945 963 981	910 927 946 964 981	911 928 946 964 981	912 929 946 964 981	912 929 946 964 982	913 929 946 964 982	914 931 947 964 982			

that the various mixtures do indeed appear to be approaching the same values of P_x/P_{3^0} as they approach the region of stratification. For the mixtures of low He³ concentration the rate of approach is sufficiently abrupt to serve as a fair determination of the value of T_s . However, for mixtures whose concentration is close to that of the consolute the approach is exceedingly gradual; e.g., the ratio $P_{x=0.6}/P_{x=0.5}$ is less than 1.020 and yet significantly greater than 1.000 for a quarter of a degree.

Figure 4 shows that portion of the data taken in the region of stratification, using values of T_s obtained from our sound velocity determinations.⁶ We find as expected, that within the scatter of the measurements

all the points fall on the same curve. This is very nearly a straight line whose equation is:

$$P_s/P_{3^0} = 1.214 - 0.502T_s$$
 for $0.58 < T_s < 0.82^{\circ}$ K.

A smoothed table of P_x/P_3^0 was constructed as follows: the solid curves of Fig. 2 were read off every 0.1°K and these values of P_x/P_3^0 were replotted versus X. Smooth curves were drawn (Fig. 5) through these points, except where abrupt changes in slope occurred at X_{λ} , and the curves were read off every 0.02 in X. As a final smoothing, a curve for each value of X was plotted versus T. Columns (c) in Table II were obtained from these curves. For convenience we tabulate $R \equiv 1000P_x/P_3^0$. Curves of R versus T terminate at the



FIG. 2. Present results of vapor pressure measurements on nine He^3-He^4 mixtures of He^3 mole fraction, X, are shown as a plot of the ratio of mixture vapor pressure, P_X , to measured He^3 vapor pressure, P_3^0 . The lambda line is shown in dashes. Measurements below the stratification temperature are omitted from this figure.

stratification temperature. As a mixture is cooled below T_s the pressure ratio, R_s , will be given by Column (a) in Table II. Abrupt changes in dR/dT may also occur at the lambda temperature. Values of T_{λ} and R_{λ} are given in Column (b) of Table II.

Table II was used to obtain the deviations column of Table I, $\Delta R = R_{obs} - R_{calc}$. The overall fit of the table to the data is satisfactory. For the 301 data points in the range of the table the average ΔR is +0.11. A

TABLE III. Our data on each mixture are compared to Table II with regard to the fit (average deviation from the table) and scatter (average deviation disregarding the sign of the deviation).

Mole fraction (X)	$\begin{array}{c} \text{Observations} \\ (n) \end{array}$	Fit $(\Sigma \Delta R/n)$	Scatter $(\Sigma \Delta R /n)$	Percent scatter $(100\Sigma \Delta R / \Sigma R)$
0.1	20	-1.1	1.4	0.45
0.2	34	-0.1	1.8	0.39
0.3	59	+0.2	2.0	0.35
0.4	26	+0.5	1.5	0.24
0.5	28	+0.2	1.3	0.19
0.6	31	+0.2	1.3	0.18
0.7	23	+0.6	1.9	0.24
0.8	38	+0.8	2.5	0.29
0.9	42	-0.5	2.3	0.25



FIG. 3. Data in the vicinity of the lambda temperature, T_{λ} , of each mixture, are shown here on a greatly expanded scale. T_{λ} for the two uppermost curves is based on our preliminary and provisional determinations of T_{λ} for these mixtures. Arrows designate the temperature at which a mixture reaches its stratification temperature.

measure of the precision of the measurements is given by the sum of ΔR without regard to sign. We find $\sum |\Delta R|/301=2.1$ in the units of the table. This can be

TABLE IV. The fit and scatter of our data in each of eighteen regions of temperature. The data in the region of stratification have been excluded from this tabulation.

T(°K)	Observations (n)	Fit $(\Sigma \Delta R/n)$	Scatter $(\Sigma \Delta R /n)$	Percent scatter $(100\Sigma \Delta R / \Sigma R)$
2.3 ± 0.05	6	+1.2	2.8	0.62
2.2	9	+0.8	1.7	0.37
2.1	7	-0.6	1.1	0.28
2.0	16	+0.3	1.9	0.29
1.9	15	-0.6	1.5	0.28
1.8	14	+0.0	1.6	0.31
1.7	31	+0.5	1.5	0.28
1.6	15	-0.1	1.2	0.22
1.5	14	-0.1	1.1	0.20
1.4	14	+0.1	0.9	0.14
1.3	19	+0.4	1.4	0.21
1.2	16	+0.2	1.3	0.19
1.1	18	+0.2	1.1	0.16
1.0	23	+0.2	1.4	0.19
0.9	23	+0.7	2.1	0.27
0.8	26	-0.1	2.8	0.34
0.7	22	+0.3	3.4	0.40
0.6	13	-1.8	3.4	0.37

expressed as a percentage scatter with respect to the average value of $R: 100 \sum |\Delta R| / \sum R = 0.31\%$. This is about equal to the sum of the precisions of the two vapor pressure measurements entering into a determination of R.

In Table III we show the over-all fit, scatter, and percent scatter for each group of concentrations. The data for X=0.1 is seen to be the poorest, both as to fit and as to percentage scatter.

Table IV shows the over-all fit, scatter, and percent scatter for 0.1°K intervals of temperature above T_s . The scatter is seen to increase at low temperatures, in the region where McLeod gauges are being used to measure both P_x and P_{3^0} . The poor fit and large scatter at the highest temperatures are probably due to allowing insufficient time for equilibration after charging the cell.

In Table V we have separately grouped the observations made in the region of stratification, for which R_{cale} is taken from Column (a) of Table II. The fit and scatter are seen to be comparable to those obtained above for the nonstratification region, in which Columns (c) of Table II were used.

 TABLE V. The fit and scatter of all data points taken in the region of stratification.

$T(^{\circ}\mathrm{K})$	Observations	Fit	Scatter	Percent scatter
0.8±0.05	11	+0.5 +0.5 +0.7	1.6	0.20
0.7	14		1.9	0.22
0.6	14		2.6	0.29

Table II should be useful for determining a liquid concentration *in situ* from a measurement of vapor pressure and temperature. Use of a He³ vapor pressure thermometer would, of course, be most convenient for this purpose, since P_x/P_3^0 is given by the measurement directly. Since the T_E scale³¹ was used in deriving Table II it would, in principle, be best to use this scale in deriving a temperature from a measured P_3^0 . However, at $1.8^\circ dR/dT$ is so small that the error due to using the T_L or some comparable future He³ scale would be nil: The maximum effect on R would be only half the least count of the table. At lower temperatures dR/dT is higher. Thus at 1°K the error would be equal to the least count if the T_L scale were used.

If a He⁴ vapor pressure thermometer is used it will be necessary to derive P_{3^0} from the measured P_{4^0} . To do this rigorously the appropriate inter-related He⁴ and He³ temperature scales should be used. Since the T_E He³ scale is based directly on the T_{55E} He⁴ scale,³⁹ the latter should be used to convert P_{4^0} to a temperature and the T_E He³ scale should then be used to obtain P_{3^0} . These operations can be described by the formula:

$$P_4^0 \rightarrow T_{55E} \equiv T_E \rightarrow P_3^0.$$



FIG. 4. Results of measurements in the region of stratification. The symbols \blacktriangle , \bigtriangleup , \Box , \ominus , \bigcirc , \bigcirc , and \blacksquare designate concentrations $X_{av}=0.3, 0.4, 0.5, 0.6, 0.7, and 0.8$ of the input mixture, respectively. The fact that the points are all on the same curve is a demonstration of the occurrence of stratification. T_c is the consolute temperature.

For the convenience of users of Table II who may not have a copy of the T_{55E} scale, we present in Table VI a compilation of P_4^0/P_3^0 derived by the above formula. A T_E scale in millidegree steps is available from the authors on request.

COMPARISON WITH RESULTS OF OTHER WORKERS

The results of other workers can be compared to our own by reference to Table II. We are seen, Fig. 6, to be in good agreement, below 1.4° K, with the results of Weinstock, Osborne, and Abraham⁴ on 20.3 and 25.5 mole percent He³ mixtures. In this temperature region our calculated curve passes through their data points nicely. At temperatures above 1.4° K our curve begins to deviate, until it is up to 4.5% above their data. This is appreciably greater than the spread in their data, which is only about 2%. It is interesting to note, however, that their data show pronounced breaks in



FIG. 5. The ratio of mixture to pure He³ vapor pressures as a function of the He³ mole fraction of the mixture. Dashed lines are in the region of stratification.

³⁹ William E. Keller, Nature 178, 883-887 (1956).

Т	+0.00	0.01	0.02	0.03	0.04	0.05	0.06	0.07	0.08	0.09
0.80	4.10	4.42	4.76	5.11	5.48	5.86	6.26	6.68	7.12	7.58
0.90	8.05	8.54	9.06	9.59	10.14	10.70	11.29	11.90	12.52	13.17
1.00	13.83	14.51	15.23	15.95	16.70	17.47	18.24	19.05	19.88	20.72
1.10	21.59	22.47	23.37	24.29	25.24	26.20	27.18	28.18	29.20	30.24
1.20	31.30	32.37	33.47	34.59	35.72	36.87	38.04	39.22	40.42	41.65
1.30	42.89	44.14	45.42	46.71	48.01	49.34	50.68	52.03	53.40	54.79
1.40	56.19	57.61	59.04	60.49	61.95	63.42	64.91	66.42	67.93	69.46
1.50	71.0	72.6	74.1	75.7	77.3	78.9	80.5	82.1	83.8	85.4
1.60	87.1	88.7	90.4	92.1	93.8	95.5	97.2	98.9	100.6	102.4
1.70	104.1	105.9	107.6	109.4	111.1	112.9	114.7	116.5	118.2	120.0
1.80	121.8	123.6	125.4	127.2	129.0	130.8	132.6	134.4	136.2	138.0
1.90	139.8	141.6	143.4	145.2	147.0	148.8	150.5	152.3	154.1	155.8
2.00 2.10 2.20 2.30	157.6 174.6 190.3 205.0	159.4 176.2 191.8	161.1 177.9 193.2	162.8 179.5 194.7	164.5 181.1 196.2	166.2 182.6 197.7	167.9 184.2 199.2	169.6 185.7 200.6	171.3 187.2 202.1	172.9 188.7 203.6

TABLE VI. The ratio $1000P_4^0/P_3^0$, for converting a measured He⁴ vapor pressure to a He³ vapor pressure at the same temperature The pressures were taken from comparable inter-related temperature scales: the 55E scale for P_4^0 and the E scale for P_3^0 .

slope at points which are in excellent agreement with our lambda line. This would tend to suggest that the discrepancy is not in the vapor pressures but in the He³ concentration. The trend of the discrepancy is consistent with occurrence of fractionation in the experiments under discussion, since this would have little effect on X at low temperatures, where m_q is small, but would have a progressively increasing effect at increasing temperatures. On the basis of this explanation the indication is that at T_{λ} fractionation had reduced the liquid concentration by about 1.5% in the experiments depicted in Fig. 6.

In Fig. 7 the circled data points show the data of Sommers² to be in somewhat better agreement with Table II but deviating in the opposite direction from the data of the previous figure. The squares show unpublished data of Kerr,⁴⁰ taken in conjunction with his density determinations. In general, his high-temperature points are on or above our table while his low-temperature points fall below. Agreement with



FIG. 6. Results of Weinstock, Osborne, and Abraham, reference 4, on two mixtures of high He³ content. Vertical bars connect the data points to curves calculated from Table II. Since He⁴ thermometry was used it was necessary in making the calculation to use appropriately inter-related He⁴ and He³ vapor pressure scales to convert from the measured P_4^0 to P_{3^0} .

our table is least satisfactory for high He³ concentrations but no consistent dependence of the discrepancy on X is evident. Triangles show the data of Peshkov and Kachinskii⁴¹ on a 93.6% mixture to be in excellent agreement with ours.

Figure 8 shows most of the data of Esel'son and Berezniak,¹ hereafter designated E-B. In the interest of clarity we have omitted data for X=0.008, 0.030, 0.226, and 0.563. Since this research provides the most extensive existing series of measurements on He³-He⁴ mixtures, a detailed discussion of them will be given.

The stated accuracy of the measurement of pressure was of the order of 0.5%. It is apparent from an examination of Fig. 8 that although for some of their mixtures the scatter of the points with respect to the best smooth curve one can draw through them is comparable to $\pm 0.5\%$, yet for other concentrations discrepancies of several percent are not infrequent. This is so much greater than errors one might reasonably expect in a pressure measurement that it appears more likely that the liquid concentration is not a constant throughout the measurements on a mixture of given average X. It is interesting to note, in this connection, that for nearly every concentration there is an abrupt apparent drop in X_{liquid} (or in R) when the bath temperature is lowered through the He⁴ lambda temperature. In the figure $T_{\lambda,0}$ is indicated by a vertical dashed line. The observed effect is qualitatively what one would expect for an unjacketed pressure sensing tube immersed in liquid He⁴, as outlined above: When surrounded by a mobile film the tube can have greatly increased noxious volume, resulting in increased fractionation and therefore in a reduction in the He³ concentration in the liquid. For mixtures in

 $^{^{40}}$ E. C. Kerr, tables of data by private communication; for experimental details see reference 12, pp. 158-163; Phys. Rev. **96**, 551-554 (1954); J. Chem. Phys. **26**, 511-514 (1957).

⁴¹ V. P. Peshkov and V. N. Kachinskii, J. Exptl. Theoret. Phys. (U.S.S.R.) 31, 720–721 (1956) [translation: Soviet Phys.—JETP 4, 607–609 (1957)].

the vicinity of X=0.1, R drops about 2%. This is equivalent to a decrease of about 6% in the value of X.

We have so far restricted the discussion to the scatter in the E-B data. As to a comparison of absolute magnitude, it can be said that most of the two groups of data are in good agreement, within the combined uncertainty in the determinations of X. They state their concentrations were known within 1.5-4% of the measured values. We estimate the uncertainty in our X's to vary linearly from $\pm 0.9\%$ of X at our lowest X to $\pm 0.1\%$ at our highest X. If we exclude the lowtemperature end ($T < 1.6^{\circ}$ K) of the E-B high-concentration data ($X \ge 0.734$), then all their remaining data is found to be in agreement with ours to within 5% of X and the existing discrepancies can be attributed almost entirely to stated uncertainties in X.

As to the above, excluded portion of their data, it is probably in error. It disagrees by over 10% in pressure (or in concentration) with all the existing data of similar X of three independent researches: ours, the measurements of Kerr,⁴⁰ and those of Peshkov.⁴¹ Furthermore, there is an apparent internal inconsistency between the E-B data on X=0.734 and 0.527, which



FIG. 7. The early results of Sommers, \bigcirc , reference 2; recent results of Peshkov, \triangle , reference 41; and unpublished measurements of Kerr, \Box , reference 40. He⁴ vapor pressure thermometers and He⁴ baths were used in all of these experiments.



FIG. 8. Results of Esel'son and Berezniak, reference 1. The vertical dashed line is at the lambda temperature of the He⁴ bath in which the experiment was conducted. Note the apparent discontinuity in P_X occurring near this temperature for most of the mixtures. Similar effects have been observed elsewhere, references 14-16, but were eliminated when the pressure-sensing tubes were completely vacuum jacketed, reference 17.

appear to be about to meet or cross at 1.4° K. In this connection it may be worth noting that the authors used a different technique for $X \ge 0.734$, in that the cell was only half filled. For lower concentrations the cell was nearly full.

In conclusion, it is worthy of note that smooth curves through the E-B data would apparently "break" very close to our lambda line. As discussed previously in the discussion of Fig. 6, this indicates the discrepancies between their results and ours in the vicinities of the λ points do not lie in the measurement of pressure or temperature but rather to the assignments of liquid concentration.

We are unable to include, in figures similar to the preceding ones, the early results of Daunt et al.,^{15,16} because they were not tabulated. A brief discussion will suffice. The differential pressure was measured between liquid He⁴ and mixture bulbs, using unjacketed pressure-sensing tubes. Particular attention was directed in one of these papers¹⁶ to discontinuities in slope

TABLE VII. Deviations of the smoothed table of Sreedhar and Daunt^a from our Table II, in units of $R=1000P_x/P_3^0$. The conversion $T_{58} \rightarrow P_4^0 \rightarrow T_{55E} \equiv T_E \rightarrow P_3^0$, was employed. The entries are in the sense $\Delta R = R_{\text{Sreedhar and Daunt}} - R_{\text{Table II}}$.

X	0.0191	0.0334	0.0651	0.1204
1.4 1.5 1.6 1.7 1.8 1.9 2.0 2.1 2.2 2.3	$ \begin{array}{r} -4 \\ 2 \\ 3 \\ -2 \\ 4 \\ 3 \\ 2 \\ 1 \\ 1 \\ 1 \end{array} $	0 4 5 4 3 4 3 1 1 1	$ \begin{array}{r} -4 \\ 2 \\ 0 \\ -5 \\ 3 \\ 4 \\ 3 \\ 3 \\ 2 \\ 1 \end{array} $	$ \begin{array}{r} -7 \\ -2 \\ 1 \\ -9 \\ 4 \\ 2 \\ -1 \\ -1 \\ -2 \\ \end{array} $
$rac{\Sigma \Delta R/n}{\Sigma \Delta R /n}$	1.1 2.3	2.6 2.6	0.9 2.7	-1.4 3.0

* See reference 17.

occurring at 2.17° K for all the solutions studied, and indeed, these were even more pronounced, by factors of 2 to 3, than the discontinuities we find in the E-B data on comparable concentrations. The authors stated that it was not clear whether this was a property of the solution itself or of the pure liquid He⁴.

There is a third possibility which should be considered. As explained above, the effect could be a capricious one due to fractionation occurring when the pressure sensing tube is cooled by a He II film. This explanation is indirectly substantiated by the fact that recent results of Sreedhar and Daunt¹⁷ show no such anomalies, despite the fact that again the differential pressure between liquid He⁴ and mixture bulbs was being measured. Significantly, in these measurements the tubing was completely vacuum jacketed, so that no He II-induced fractionation could have occurred.

The recent results of Sreedhar and Daunt¹⁷ do not include a table of the actual data. However, the authors found that their measurements are in good agreement with our Table II, and their smoothed tables do indeed show excellent agreement. The deviations of their table from ours is shown in Table VII. No large systematic deviations are evident. For each mixture the overall fit is quite good and the over-all fit of all the entries is remarkably good: The average ΔR is 0.8, which is only 0.27% of the average value of R. The agreement between their smoothed table and ours is thus quite similar to the agreement of our data with our Table II. There is therefore no need for a revision of Table II in the light of the recent results of Sreedhar and Daunt. This is particularly gratifying in view of the extensive graphic interpolations we made in the region of low concentrations.

The above comparisons with data of other workers is complete, to the best of our knowledge, except for some of the early isolated pieces of data on concentrations of the order of 1% He³.

In summary, it appears that almost all of the existing literature data are in excellent agreement with our lambda vapor pressure table, Column (b) of Table II. All of Columns (c) in Table II below 1.2° K remains to be confirmed. Above this temperature the data of Peshkov and Kachinskii provide adequate confirmation for $X \sim 0.9$ and the recent data of Sreedhar and Daunt provides adequate confirmation for $X \leq 0.12$. For other portions of Columns (c) (Table II) existing data are generally too uncertain as to X to provide a confirmation of the table.

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