# Lambda-Temperatures of Solutions of He<sup>3</sup> in He<sup>4</sup> below 1°K\*

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Measurements have been made of the transition (lambda) temperatures at which solutions of He<sup>3</sup> in He<sup>4</sup> first show superfluidity by observation of the thermal isolation of a vessel which contained the solutions and which was cooled to low temperature by a paramagnetic salt. The incidence of superfluidity was accompanied by a high heat influx to the solution produced by a convection of film flow to the high temperature together with return flow in the vapor phase. The solution with the highest concentration, namely 89 percent He<sup>3</sup>, had a lambda-temperature of 0.38°K; solutions with smaller He<sup>3</sup> content (concentrations down to 42 percent He<sup>3</sup> were investigated) showed higher lambda-temperatures ranging up to 1.15°K. It was concluded that pure He<sup>3</sup> could not be superfluid above 0.25°K and most probably is not superfluid down to 0°K. From the results conclusions have been drawn regarding the free energy of pure He<sup>4</sup> in the liquid II state on the basis of the two-fluid model.

#### **1. INTRODUCTION**

N 1947 it was shown by Daunt, Probst, Johnston, Aldrich, and Nier<sup>1</sup> that in dilute solutions of He<sup>3</sup> in He<sup>4</sup>, the He<sup>3</sup> did not partake in superfluid flow. This was confirmed by another method, also using dilute solutions, by Lane, Fairbank, Aldrich, and Nier.<sup>2</sup> It had been suggested by Pollard and Davidson<sup>3</sup> and by Franck<sup>4</sup> that the criterion for the superfluid flow of the helium isotopes was whether they obeyed the Bose-Einstein statistics, in which the low temperature degeneration phenomenon was responsible, according to the theory of F. London,<sup>5</sup> for the occurrence of the superfluid properties of liquid He<sup>4</sup>. Although the early experiments<sup>1, 2, 2a</sup> on dilute mixtures of He<sup>3</sup> in He<sup>4</sup> lent considerable support to this view of the importance of the statistics, a contrary opinion was subsequently put forward by Landau and Pomeranchuk<sup>6</sup> who suggested that in very dilute solutions any solute would not partake in superfluid flow. Subsequent and more decisive evidence has been gained from the measurements of Osborne, Weinstock, and Abraham<sup>7</sup> on the flow properties of pure liquid He<sup>3</sup> which showed no sign of superfluidity at any temperature between 1.05°K and its boiling point<sup>8</sup>  $(3.2^{\circ}K)$ , thus favoring the basic theory of F. London. It was considered to be of interest to extend to lower temperatures the experimental tests for the superfluidity of He<sup>3</sup> and details of these observations down to a temperature of 0.25°K are given in this

paper. A second point of interest to which the experiments reported herewith are pertinent concerns the thermodynamics of solutions of He<sup>3</sup> in He<sup>4</sup>. From measurements of the vapor pressures of such solutions, it has recently been concluded by Taconis, Beenakker, Nier, and Aldrich<sup>9</sup> that for dilute solutions in the helium II phase the He<sup>3</sup> dissolves in the "normal" constituent of the liquid only, resulting in high concentrations of He<sup>3</sup> in the vapor phase of low temperatures.<sup>9a</sup> Some conclusions that can be drawn from this peculiar law for the solubility of He<sup>3</sup> in liquid helium II have been made by Stout,<sup>10</sup> de Boer,<sup>11</sup> and by de Boer and Gorter,<sup>12, 12a</sup> who have calculated the variation of the lambda-temperature,  $T_{\lambda}$ , with He<sup>3</sup> concentration, even up to 100 percent concentration. Since these theoretical calculations differed markedly from one another in the high concentration range, because of basically different assumptions regarding the free energy of liquid helium, it was of interest to investigate the variation of  $T_{\lambda}$  in He<sup>3</sup>+He<sup>4</sup> mixtures. Some results on this up to 28 percent He<sup>3</sup> concentration have been published by

<sup>\*</sup> Assisted by a contract between the ONR and The Ohio State University Research Foundation.

<sup>&</sup>lt;sup>1</sup> Daunt, Probst, Johnston, Aldrich, and Nier, Phys. Rev. 72, 502 (1947). See also, Daunt, Probst, and Johnston, J. Chem. Phys. 15, 759 (1947).
 <sup>2</sup> Lane, Fairbank, Aldrich, and Nier, Phys. Rev. 73, 256 (1948).

<sup>&</sup>lt;sup>2a</sup> See also Daunt, Probst, and Johnston, Phys. Rev. 73, 638

<sup>(1948).</sup> <sup>3</sup> E. C. Pollard and W. L. Davidson, Applied Nuclear Physics (John Wiley and Sons, Inc., New York, 1942), p. 183. <sup>4</sup> J. Franck, Phys. Rev. **70**, 561 (1946). <sup>5</sup> F. London, Nature **141**, 643 (1938) and Phys. Rev. **54**, 947

<sup>(1938).
&</sup>lt;sup>6</sup> L. Landau and I. Pomeranchuk, Comptes Rendus Acad. Sci. U.S.S.R. 59, 669 (1948).
<sup>7</sup> Osborne, Weinstock, and Abraham, Phys. Rev. 75, 988 (1949).
<sup>8</sup> See Sydoriak, Grilly, and Hammel, Phys. Rev. 75, 303 (1949).

<sup>&</sup>lt;sup>9</sup> Taconis, Beenakker, Nier, and Aldrich, Physica 15, 733 (1949) and Phys. Rev. 75, 1966 (1949).

<sup>&</sup>lt;sup>9a</sup> The marked differences between the values of the distribution coefficient  $(C_V/C_L)$  obtained by Taconis, Beenakker, Nier, and Aldrich and those obtained by facons, beenakker, Nier, and Aldrich and those obtained previously by Fairbank, Lane, Aldrich, and Nier [Phys. Rev. 73, 729 (1948)] and by Daunt, Probst, and Smith [Phys. Rev. 74, 495 (1948)], are undoubtedly partly due to the improved stirring of the liquid adopted by Taconis et al., thereby avoiding concentration gradients in the liquid with the lower concentration of  $He^3$  on the surface. It would, however, also be expected that the concentration gradients would be less marked at higher average concentrations of the liquid owing to the automatic stirring produced by differences in vapor pressure for different concentrations. Indeed calculations indicate that such an effect may contribute in large measure to the variations in the results of the three different experiments quoted above, their results indicating that without exception increasingly large values of  $C_V/C_L$  were observed for increasing average concentrations. Such an explanation seems more straight average concentrations. Such an explanation seems more straightforward than that based on the special assumptions put forward by F. London and O. K. Rice (Phys. Rev. 73, 1188 (1948)).
<sup>10</sup> J. W. Stout, Phys. Rev. 76, 864 (1949) and 74, 605 (1948).
<sup>11</sup> J. de Boer, Phys. Rev. 76, 852 (1949).
<sup>12</sup> J. de Boer and C. J. Gorter (to be published in Physica); Phys. Rev. 77, 569 (1950).

<sup>&</sup>lt;sup>12a</sup> We are grateful to Professor J. de Boer for kindly sending us the manuscript of this paper before publication.

Abraham, Weinstock, and Osborne,13 whereas our investigations reported here extend to 89 percent He<sup>3</sup> concentration, which involved measurements down to 0.2°K.

The general method for finding the  $\lambda$ -temperatures adopted by us depended on the anomalously high heat influx into vessels partially filled with liquid helium below the  $\lambda$ -point, first observed for pure liquid He<sup>4</sup> by Rollin<sup>14</sup> and by Rollin and Simon.<sup>15</sup> This anomalously high heat influx is due to the motion of the helium film formed from helium II up the walls of the tube connecting the helium vessel to its surroundings, such motion being toward the warmer end where it is partially or completely evaporated, as has been investigated in detail by Daunt and Mendelssohn.<sup>16</sup> This upward flow of matter in the film is compensated by a downward flow in the vapor phase, bringing down to the helium vessel the large heat of condensation of the vapor. The  $\lambda$ -point can be detected, therefore, as the temperature at which this large heat influx first occurs, due to the two phase convection process. In the experiments described below we investigated the heat influx into vessels partially filled with solutions of He<sup>3</sup> in He<sup>4</sup> below  $1^{\circ}$ K in order to determine their  $\lambda$ -points.<sup>16a</sup> In



FIG. 1. Experimental apparatus.

<sup>13</sup> Abraham, Weinstock, and Osborne, Phys. Rev. 76, 864 (1949). <sup>14</sup> B. V. Rollin, Proc. VII Int. Congress of Refrigeration 1, 187

 <sup>14</sup> B. V. Kollin, Proc. VII Int. Congress of Retrigeration 1, 187 (1936). See also Kurti, Rollin, and Simon, Physica 3, 266 (1936).
 <sup>15</sup> R. V. Rollin and F. Simon, Physica 6, 219 (1939).
 <sup>16</sup> J. G. Daunt and K. Mendelssohn, Nature 141, 911 (1938) and Proc. Roy. Soc. A170, 423, 439 (1939).
 <sup>16a</sup> Another possibility of making thermal observations of λ-points would be by measuring the heat conductivity of the solutions themselves and noting the temperature at which such seat to enductivity first begins to get anomalously large. such as heat conductivity first begins to get anomalously large, such as one could observe in pure liquid He<sup>4</sup>. Such a procedure, however, was thought to be less readily interpretable than the method outlined above using the two phase convection process since (a) over a long column of the solution, such as would be necessary from technical considerations, quite appreciable concentration gradients might be set up owing to internal convection of the superfluid and normal constituents within the liquid itself, thus



FIG. 2. Observed warm-up curves for the 79 percent He<sup>3</sup> solution.

adopting this technique it was assumed that even in the high concentration ranges a mobile helium film would be formed from the liquid phase II, an assumption which appears to be valid in view of the observation of the superfluidity of the He<sup>4</sup> component in concentrations up to 28 percent He3 by Abraham, Weinstock, and Osborne.<sup>13</sup> One advantage of our technique was that concentration of the solution remained essentially constant (see section on "Results") during the course of any one experiment.

### 2. THE EXPERIMENTAL METHOD

A volume of 3.2 cm<sup>3</sup> of gas at N.T.P. of a  $He^3 + He^4$ mixture of concentration 89 percent He<sup>3</sup> was kindly supplied by the Isotopes Division of the AEC. In order to cool the liquid formed from this mixture to very low temperatures the gas was pumped by a Toepler pump, "D" (see diagram given in Fig. 1) via a fine bore stainless steel capillary, "E" (inside diameter 0.24 mm) into a copper reservoir "C" inside the cryostat. This reservoir, which was of 6-mm<sup>3</sup> volume, was pressed inside a cylindrical pill, "A," made up of 5 grams of chromium potassium alum powder compressed under 200-atmospheres pressure, a technique which was found to give good thermal contact between the reservoir and the salt. This arrangement of reservoir and salt was mounted in a vacuum jacket on which was wound the mutual inductance for measurement of the susceptibility of the salt.<sup>16b</sup> It was cooled to helium temperatures by immersing it in a Dewar filled with liquid helium from a small Simon-type expansion liquefier17 and temperatures below 1°K were attained by equipment identical with that previously described.<sup>18-19</sup>

In order to reduce the heat influx to the reservoir "C" at low temperatures, a second salt cylinder, "B," of mass 5 grams, also of pressed chromium potassium alum powder, was mounted higher up the stainless steel

possibly falsifying the measurements, and (b) the heat influx to the lower temperature end might be so large that accuracy of temperature measurement of the temperatures below 1°K would be impaired.

<sup>&</sup>lt;sup>16b</sup> See Daunt and Heer (Phys. Rev. 76, 1324 (1949)) for further details of such arrangements. <sup>17</sup> Daunt, Heer, and Silvidi, Phys. Rev. **75**, 1113 (1949). <sup>18</sup> J. G. Daunt and C. V. Heer, Phys. Rev. **76**, 715 (1949). <sup>19</sup> J. G. Daunt and C. V. Heer, Phys. Rev. **76**, 1324 (1949).

TABLE I.  $\lambda$ -temperatures,  $T_{\lambda}$ , of solutions of He<sup>3</sup> in He<sup>4</sup> for various concentrations, X, of He<sup>3</sup>. Column 5 gives the fractional value of the  $\lambda$ -temperature of the solutions compared with the  $\lambda$ -temperature, (2.18°K), of pure He<sup>4</sup>.

Date	Run	X (percent)	Tλ°K	<i>T</i> <sub>λ</sub> °K/2.18
Aug. 30	#1 #2	$89\pm 2 \\ 89\pm 2$	$0.37 \pm 0.03$ $0.38 \pm 0.03$	$0.17 \pm 0.015$ $0.175 \pm 0.015$
Oct. 20	#1 #2 #3	$81\pm 2 \\ 81\pm 2 \\ 81\pm 2 \\ 81\pm 2$	$0.54 {\pm} 0.03$ $0.57 {\pm} 0.03$ $0.58 {\pm} 0.03$	$0.25 \pm 0.015$ $0.26 \pm 0.015$ $0.27 \pm 0.015$
Oct. 27	#1 #2	$78\pm 2 \\ 78\pm 2$	$0.65 \pm 0.03$ $0.64 \pm 0.03$	$0.30 \pm 0.015$ $0.30 \pm 0.015$
Nov. 1	#1 #2 #3	$71\pm 2$ $71\pm 2$ $71\pm 2$	$0.69 \pm 0.03$ $0.71 \pm 0.03$ $0.68 \pm 0.03$	$0.32 \pm 0.015$ $0.32 \pm 0.015$ $0.31 \pm 0.015$
Nov. 3	#1 #2 #3	$61\pm 2$ $61\pm 2$ $42\pm 2$	$0.81 \pm 0.03$ $0.83 \pm 0.03$ $1.15 \pm 0.05$	$\begin{array}{c} 0.37 {\pm} 0.015 \\ 0.38 {\pm} 0.015 \\ 0.52 {\pm} 0.025 \end{array}$

capillary (see Fig. 1), so as to serve as a heat barrier to the flow of heat along the capillary from the helium bath maintained at about 1.1°K to the reservoir. This barrier salt, "B," was also cooled magnetically simultaneously with the lower pill, "A," it being situated in the fringing field of the magnet which served to magnetize the latter. Magnetic measurements of the temperatures of the barrier salt, "B," could be made alternately with those of the lower salt, "A" surrounding the reservoir, by means of a second mutual inductance.

In order to test the apparatus, and in particular to make sure of the efficacy of the thermal contact between the reservoir and the salt "A," an experiment was carried out using pure He<sup>4</sup> in the reservoir space. Sufficient He<sup>4</sup> was liquefied to fill the reservoir about half-full at  $1.2^{\circ}$ K. The salts were then cooled, salt "A" to  $0.15^{\circ}$  curie and salt "B" to  $0.3^{\circ}$  curie and the warm-up time observed. This was very fast (2 minutes) corresponding to an average heat influx of 600 ergs/sec. This high heat influx<sup>19a</sup> was due, as described earlier, to the motion of the helium film up the walls of the capillary tube to the warmer end where it was evaporated. This upward flow of matter was compensated by a downward flow in the vapor phase, bringing down to the lower temperature the large heat of condensation of the vapor. From this experiment it was concluded that the thermal contact and time for establishment of thermal equilibrium between the reservoir, "C," and the salt

"A" were satisfactory for measurements down to 0.2° curie.

An identical experiment using pure He<sup>4</sup> in the reservoir space was carried out at the end of the long series of runs on He<sup>3</sup>+He<sup>4</sup> mixtures, in order to check for possible changes in the apparatus with time, which yielded a result identical with the first after a period of over four months.

The experiments on solutions of He<sup>3</sup> in He<sup>4</sup> were carried out in the following manner. First, the salt "B" and the salt "A" together with its reservoir, "C," almost filled with solution were demagnetized simultaneously to low temperatures and then the rise of temperature of both salts with time observed magnetically. The temperature on the  $T^*$  or "Curie scale" was calculated in the usual manner,<sup>20-21</sup> by assuming Curie's law for the paramagnetic susceptibility of the chromium potassium alum in the temperature range involved. The calculation of the temperature of the helium bath, necessary for calibrating the apparent Curie constant for our arrangement, was made from vapor pressure measurements, using the tables given by Van Dijk and Shoenberg.22

Diagrams giving typical warm-up curves for both the salts "A" and "B" are given in Figs. 2 and 3, which give the results of one measurement on the 79 percent He<sup>3</sup> solution. Figure 3 is an enlarged diagram of the results shown in Fig. 2 near the  $\lambda$ -point at 0.65°K. The curves show the galvanometer deflections plotted against the time, where the galvanometer deflections are proportional to  $1/T^*$ . For the temperature range used the specific heat of the salt is proportional to  $1/T^2$ and hence the slopes of the curves plotted in Figs. 2 and 3 are directly proportional to the heat influx.

From Fig. 2 it will be seen that the salt "B" which acted as a heat barrier warmed up to the temperature of the bath relatively quickly. This was due mainly to the fact that it absorbed the heat of condensation of the



<sup>20</sup> N. Kurti and F. Simon, Proc. Roy. Soc. A149, 152 (1935) and W. J. de Haas and E. C. Wiersma, Physica 2, 335 (1935).
 <sup>21</sup> N. Kurti and F. Simon, Phil. Mag. 26, 849 (1938).

<sup>22</sup> H. Van Dijk and D. Shoenberg, Nature 164, 151 (1949).

<sup>&</sup>lt;sup>19a</sup> Much smaller average heat influxes have been observed in a somewhat similar arrangement employed by Hudson, Hunt, and Kurti [Proc. Phys. Soc. London 62, 392 (1949)] for other purposes. In comparing this work, however, it must be remembered that the pressure head establishing the vapor flow is given by the helium vapor pressure at the temperature of the bath, which for the temperature of 0.9°K reported in Hudson, Hunt, and Kurti's work was smaller by more than a factor of 10 than in our experiments. Our calculations of the considerable effect that the bath temperature has on the net heat influx to the salts in our arrangement were checked by making several demagnetizations with pure He4 in the reservoir starting from different bath temperatures between 1.05°K and 1.3°K

gas in the capillary which flowed down immediately after demagnetization.<sup>22a</sup>

The salt "A" initially warmed up slowly, so long as salt "B" was well below  $1^{\circ}$ K, due to the small pressure difference in the capillary between them. When salt "B" reached the bath temperature, however, salt "A" commenced to warm up rapidly, at the well-marked point "b" in the curve (see Fig. 2). This rapid warming up of "B" was due, as described previously, to the two phase convective process of heat influx operative below the  $\lambda$ -point of the solution, and continued until the  $\lambda$ -point was reached (point "a" of Figs. 2 and 3). Above the  $\lambda$ -point (point "a") the heat influx to salt "A" became much smaller, owing to the breakdown of the two phase convection. These  $\lambda$ -points, observed by the change in slope of the (1/T) versus time curves, were always clearly observable, as is shown in Fig. 3, where the heat influx to salt "A" changed from 140 ergs/sec. above point "a" to 50 ergs/sec. below point "a."

Curves similar to that of Fig. 2 were observed for the other concentrations of He<sup>3</sup>, the lower concentrations being obtained by adding measured volumes of pure He<sup>4</sup>.

## 3. RESULTS

The observed values of the  $\lambda$ -temperature, as interpreted by the technique reviewed in the previous section, for various concentrations of He<sup>3</sup> are given in Table I and are shown graphically by the circled points of Figs. 4 and 5. The four points marked  $\Box$  in Figs. 4 and 5 are the results obtained by a different method in the liquid helium temperature range of Abraham, Weinstock, and Osborne.<sup>13</sup>

The possible errors in assessing the concentration of He<sup>3</sup> in the solutions are due to (a) possible inaccuracy in measurement of the concentration in the unrefrigerated gas, and, (b) possible differences in the concentration in the solution and in the unrefrigerated gas. Under (a), the initial concentration of the gas mixture was measured before dispatch by the AEC with an optical spectrograph and stated to be 88.7 percent He<sup>3</sup> with He<sup>4</sup> as diluent. In diminishing the concentration, measured volumes of pure He<sup>4</sup> were added, the accuracy of estimation of any one concentration value thus produced being  $\pm 1.0$  percent, this error being due to possible inaccuracy of assessment of the original volume. As a final check, the concentration of the most diluted mixture was measured mass-spectroscopically giving a result in agreement with our calculation. Under item (b), two possible causes of discrepancies between the solution concentration and the unrefrigerated gas concentration need to be considered. First the solution concentration may be diminished owing to the relatively large concentration in the vapor.<sup>9</sup> This effect was negligible, however, owing to the small vapor volume available and to the small vapor pressures below 1°K.

Secondly the heat influx to the solution in the reservoir would cause internal convection of the superfluid and normal constituents within the liquid and thus tend, since the He<sup>3</sup> does not partake in superfluid flow,<sup>1</sup> to produce a vertical concentration gradient in the solution. At such high average concentrations and with such small liquid depths (6 mm) as used here, however, the vapor pressure gradients which would accompany such concentration gradients would provide adequate stirring (see reference 9a). It was concluded that the effective solution concentration would not differ from the unrefrigerated concentration by more than 2 percent.

The possible errors in assessment of the  $\lambda$ -temperature on the absolute scale could arise from, (a) differences between the Curie scale for a spherical specimen,<sup>21</sup>  $T_s^*$ , and the absolute scale,  $T^{\circ}K$ , (b) effects due the shape of salt "A" on the evaluation of  $T_s^*$ , and (c) difficulties in interpretation of the exact transition point from the measured warm-up curves (see Figs. 2 and 3). Under (a), it is estimated that in the temperature range involved, 0.35 degrees and upward,  $T^{\circ}K - T_s^*$ for chromium potassium alum is less than 0.01 degrees.<sup>23</sup>



FIG. 4. Plot of the  $\lambda$ -temperatures,  $T_{\lambda}$ , of the solutions (as a fraction of the  $\lambda$ -temperature, 2.18°K, of pure liquid He<sup>4</sup>) against He<sup>3</sup> concentration. The points marked  $\Box$  are the results reported herewith. The points marked  $\Box$  are the results obtained by Abraham, Weinstock, and Osborne, reference 13. Curve A is the theoretical result of Stout, reference 10. Curve B is the theoretical result of Boer and Gorter, reference 12. Curves C and D are for first-order transitions calculated by de Boer, reference 11 (see text).



FIG. 5. Plot of the  $\lambda$ -temperatures,  $T_{\lambda}$ , of the solutions (as a fraction of the  $\lambda$ -temperature, 2.18°, of pure liquid He<sup>4</sup>) against He<sup>3</sup> concentration, X. Points marked  $\bigcirc$  are the results reported herewith. Points marked  $\bigcirc$  are the results of Abraham, Weinstock, and Osborne, reference 13. Curve B is the theoretical result of de Boer and Gorter, reference 12. Curves E and F are theoretical results reported herewith (see text).

<sup>23</sup> J. H. Van Vleck, J. Chem. Phys. **5**, 320 (1937); M. H. Hebb and E. M. Purcell, J. Chem. Phys. **5**, 388 (1937).

<sup>&</sup>lt;sup>22a</sup> This was checked (a) by the fact that the observed vapor pressure of the solution fell with time along a curve identical with that for the susceptibility of salt "B" and (b) by demagnetizing from various initial temperatures.

Under (b), it should be stated that the temperatures quoted in this paper are uncorrected for the shape factor of the salt "A". Its interior contained the irregularly shaped reservoir which rendered corrections difficult to assess. It was estimated, however, from the geometry of our arrangement that the temperatures quoted here are not smaller than  $T_s^*$  by more than 0.03 degree. Finally under (c), we estimated  $T_{\lambda}$  from the tangents drawn to the curves at either side of the  $\lambda$ -point. This results in a possible error estimated to be  $\pm 0.02$  degrees. The  $\lambda$ -point at the 42 percent concentration was, owing to the fast warm-up encountered at the high temperature involved, in more doubt that the  $\lambda$ -points observed below 1°K. The results of these considerations in temperature and concentration assessment are collected in Table I.

## 4. DISCUSSION

## (A) The Superfluidity

The lowest  $\lambda$ -temperature observed in these experiments was 0.38°K for the 89 percent He<sup>3</sup> solution. If pure He<sup>3</sup> were to show superfluidity, its  $\lambda$ -temperature would therefore be below 0.38°K, and from extrapolation of the experimental results it would certainly be below 0.25°K. Moreover from the agreement between the results and the thermodynamical theory,<sup>12</sup> (to be discussed), which is based on the assumption of nonsuperfluidity of He<sup>3</sup> at all temperatures, it is concluded that liquid He<sup>3</sup> is a non-superfluid liquid. This is in agreement with the theory of F. London,<sup>5</sup> in which superfluidity of the liquid phase would occur only for the isotope obeying the Bose-Einstein statistics.<sup>23a</sup> It is of interest to note, however, that many of the detailed properties of the condensed state of pure He<sup>3</sup> were predicted exactly by de Boer and Lunbeck,<sup>24</sup> independently of consideration of the statistics involved.

#### (B) Statistical and Thermodynamic Considerations

Two basic properties of pure liquid He<sup>3</sup> and of solutions of He<sup>3</sup> and He<sup>4</sup>, based on experimental evidence, have enabled statistical thermodynamic formulations to be evolved for the variation of the  $\lambda$ -temperature with change in concentration of He<sup>3</sup>. These properties are (1) the non-superfluidity of pure liquid He<sup>3</sup>, as discussed previously, and (2) the unusual solubility law for He<sup>3</sup> in liquid helium II. Under item (2), it was found by Taconis and co-workers<sup>9</sup> that the results of their measurements of the vapor pressures of such solutions could be explained by assuming that He<sup>3</sup> does not dissolve in the superfluid part of liquid helium II. Assuming these two properties, the variation of the  $\lambda$ -temperature with He<sup>3</sup> concentration has been calculated by Stout,<sup>10</sup> de Boer,<sup>11</sup> and by de Boer and Gorter.<sup>12</sup> The results of these calculations are given in Fig. 4, where curve A is due to Stout and curve B to de Boer and Gorter. Both curves A and B represent the  $\lambda$ -temperatures for transitions of second order. Curves C and D give the phase diagram for transitions of first order as calculated by de Boer<sup>11</sup> and by de Boer and Gorter.<sup>12</sup> A first-order transition would result in the solution splitting into two separate phases, helium I and helium II, such that the helium II phase contains only a small fraction of the He<sup>3</sup> atoms. (In Fig. 4, curve C is for the helium I phase and curve D for the helium II phase.)

The large differences between curves A and B, both of which refer to transitions of second order, are due to differences in arbitrary assumptions made regarding the free energy of liquid helium II, (pure He<sup>4</sup>). It would appear that the choice made by de Boer and Gorter (curve B) for this free energy is in closer agreement with experiment.<sup>24a</sup>

The points obtained by Abraham, Weinstock, and Osborne,<sup>13</sup> as shown in Fig. 4, together with our results for the higher concentration range, form a moderately continuous curve. The slight discrepancies may be due partly to the difference in the methods of observation of the  $\lambda$ -temperatures and partly to the difficulties that arise at higher temperatures in assessing the concentration of He<sup>3</sup> in the solution, due to the high concentrations in the vapor phase.9 Unfortunately, it is not possible from these results to decide whether the transition is of first or second order, although the observed  $\lambda$ -temperatures lie close to those calculated for a first-order transition (curve C). This uncertainty is due to the fact that small differences in the assumptions made regarding the free-energy of helium II make large differences in the computed  $T_{\lambda}$  versus concentration curves. It can only be concluded that, if the transition is of first order, the free energy expression adopted by de Boer and Gorter,<sup>12</sup> yielding curve C, is satisfactory.

On the other hand, if the transition is of second order, as seems to be indicated by recent vapor pressure measurements of Weinstock, Osborne, and Abraham,<sup>25</sup> Fig. 5 shows some of the  $T_{\lambda}$  versus concentration curves that can be computed. In Fig. 5 curve *B* is due to de Boer and Gorter;<sup>12</sup> curves *E* and *F* have been computed using similar methods, adopting the assumption that the "normal" density  $\rho_n$  of helium II is proportional to  $T^7$ , rather than  $T^6$  as assumed by de Boer and Gorter. This seventh-power variation of  $\rho_n$  seems probable at the lower temperatures from the recent measurements of Andronikashvilli.<sup>26</sup> Curve *E* is for a linear variation with temperature of the free energy, *G* at  $\rho_n = \rho$  (total)

<sup>&</sup>lt;sup>23a</sup> As a further check it would be of interest to investigate the properties of He<sup>6</sup> in liquid He<sup>4</sup>, perhaps by a method analogous to that used first by Daunt *et al.* (see reference 1) for dilute solutions of He<sup>3</sup> in He<sup>4</sup>.

<sup>&</sup>lt;sup>24</sup> J. de Boer and R. J. Lunbeck, Physica 14, 510 (1948).

<sup>&</sup>lt;sup>24a</sup> For further detail regarding this phenomenological approximation for the free-energy of helium I, see C. J. Gorter, Physica 15, 523 (1949).

Weinstock, Osborne, and Abraham, Phys. Rev. 77, 400 (1950).
 Andronikashvilli, J. Exp. Theor. Phys. U.S.S.R. 18, 429 (1948).

whereas curve F is for quadratic variation<sup>26a</sup> (see de Boer, reference 11). It will be seen from Fig. 5 that the experimental points lie somewhere in between the various theoretical curves, all of which differ only slightly in the phenomenological basis for their computation.

It is concluded, therefore, (a) that the experimental results do not provide evidence for establishing whether the  $\lambda$ -transition of the solutions is of first or second order; (b) from the general agreement of the results with the theories of de Boer and Gorter, that the basic assumptions of non-superfluidity of pure liquid He<sup>3</sup> and of the Taconis solubility law receive welcome confirmation, and (c) that the general type of free-energy function for liquid helium II (pure He<sup>4</sup>) which forms the basis of the computations shown in Fig. 5 is satisfactory for both types of transition, even down to the lowest  $\lambda$ -temperature measured (0.38°K).

As is well known the "condensation" temperature,  $T_{\lambda}$ , of a perfect Bose-Einstein gas is proportional to the number density of particles to the two-thirds power. The observed decrease in the  $\lambda$ -temperature of liquid He<sup>4</sup>+He<sup>3</sup> solutions with decreasing He<sup>4</sup> concentration, as reported herewith, might be ascribed therefore to this decrease in the number density of Bose-Einstein particles. Although the conditions obtaining in a perfect gas cannot adequately represent those of liquid helium, it was considered to be of interest to calculate the expected variation of  $T_{\lambda}$  for a perfect He<sup>3</sup>+He<sup>4</sup> gas mixture with concentration, x, of He<sup>3</sup>, assuming the liquid mixture densities. The liquid mixture densities were calculated by assuming the law for perfect solutions using the known values of the molar volumes of pure liquid He<sup>4</sup> (27.6 cc) and pure liquid He<sup>3</sup> (37.5 cc) at the lowest temperatures. The result of this calculation is given by curve H of Fig. 4,  $T_{\lambda}$  at x=0 being normalized to 2.18°K. In view of the range of experimental error in measurement of  $T_{\lambda}$  (especially, as noted, that for the point at x=42 percent) it would appear that curve H represents the results as adequately as curves B and E which are the closest fit to the experimental results on the basis of the theory of de Boer and Gorter.<sup>12</sup> It is hoped to present later the detailed relationship between these various methods of calculation of  $T_{\lambda}$ .

Note added in proof.—O. G. Engel and O. K. Rice [Phys. Rev. 78, 55 (1950)] have recently also proposed a thermodynamic model to allow calculation of  $T_{\lambda}$  as a function of x for high concentrations of He<sup>3</sup>. In it, as in the models discussed above, the observed results can only be calculated after making *ad hoc* assumptions regarding the free energy of pure liquid He<sup>4</sup> in the normal state. A detailed discussion must, however, be postponed for a subsequent communication.

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## On the Azimuthal Asymmetry of Cosmic-Ray Intensity above the Atmosphere at the Geomagnetic Equator\*

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By means of Geiger-Mueller tube telescopes in an Aerobee sounding rocket fired to high altitude at the geomagnetic equator, information on the azimuthal asymmetry of the cosmic-ray intensity above the atmosphere has been obtained. These results, in conjunction with previously reported vertical intensities and specific ionizations at  $\lambda = 0^{\circ}$  and  $\lambda = 41^{\circ}$ N, are consistent with the hypothesis that most of the primaries are positively charged protons with differential number spectrum of the form  $dN = KE^{-1.9}dE$  in the energy region 5 to 23 Bev.

#### 1. INTRODUCTION

A PREVIOUS paper<sup>1</sup> reports cosmic-ray intensities above the atmosphere at the geomagnetic equator as obtained by means of Aerobee sounding rocket A10 fired from the USS Norton Sound on March 17, 1949. From the same set of data, information on the azimuthal asymmetry<sup>2</sup> has been derived by a detailed analysis of the variation of the counting rate of telescopes AOB, XOY (axes at 45° to the rocket axis) as the rocket rotated about its longitudinal axis. This information is presented herein.

#### 2. EXPERIMENTAL RESULTS

It is essential in measurements of this type to know the angular motion of a system of axes fixed in the rocket, and hence of the telescope axes, during the flight of the rocket above the appreciable atmosphere. Two devices were used in the A10 flight for this purpose. The first was a system of sixteen photoelectric cells, whose apertures were systematically arranged to cover

<sup>&</sup>lt;sup>26a</sup> The quadratic temperature function is indicated by extrapolation of the entropy *versus* temperature curve below  $T_{\lambda}$  (see Daunt and Mendelssohn, Proc. Roy. Soc. **A185**, 237 (1946)) and is the same as that obtained for the roton spectrum postulated by Landau (J. Phys. U.S.S.R. **11**, 91 (1947)).

<sup>\*</sup> Supported by the Navy Bureau of Ordnance under Contract NOrd 7386.

<sup>&</sup>lt;sup>1</sup> J. A. Van Allen and A. V. Gangnes, Phys. Rev. 78, 50 (1950).

<sup>&</sup>lt;sup>2</sup> A preliminary and partially incorrect report was given at the Echo Lake Conference (June, 1949). A corrected manuscript was subsequently submitted for inclusion in the report of the conference.