low energy γ -transitions in Tm¹⁶⁹. These authors state that the 8.42 and 20.75 kev γ -transitions are magnetic dipole (M1),* whereas according to our data the first transition is M1 + E2 (< 3%), while the second is M1. In the same paper it is shown that the rotational band (for K = $\frac{1}{2}$) has levels with spins of $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$, while the level with spin $\frac{9}{2}$ is absent. Our measurements are in accord with these data.

Computations by Zaretskii based on the coupling scheme treated in Reference 13 show that the 8.32 and 20.65 kev γ -transitions in Tm¹⁶⁹ should be mainly magnetic dipole, with a possible small admixture of E2' (a few percent). As one can see, the experimental data are not in contradiction with this result.

In conclusion, we express our gratitude to D. V. Timoshuk for much valuable advice.

¹S. A. E. Johansson, Phys. Rev. **100**, 835 (1955).

² B. R. Mottelson and S. G. Nilsson, Z. Physik **141**, 217 (1955).

³Cork, Brice, Martin, Schmid and Helmer, Phys. Rev. 101, 1042 (1956).

⁴S. D. Koički and A. M. Koički, Bull. Inst. Nucl. Sci. "Boris Kidrich" **6**, 1 (1956). ⁵ Mihelich, Ward and Jacob, Phys. Rev. **103**, 1285 (1956).

⁶A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab. Mat.-fys. Medd. 27, No. 16 (1953); <u>Beta and Gamma Ray Spectroscopy</u>, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955).

⁷ Martin, Jensen, Hughes and Nichols, Phys. Rev. 82, 579 (1951).

⁸Cork, Brice et al., Phys. Rev. 101, 1042 (1956). ⁹G. N. Kruzhilin, <u>Reactor for Physical and</u> Technical Investigations, Proceedings of the In-

ternational Conference on Peaceful Uses of Atomic Energy, Geneva, 1955; vol. 2, p. 435.

¹⁰ Hatch, Boehm, Marmier and DuMond, Phys. Rev. 104, 745 (1956).

¹¹ E. N. Hatch and F. Boehm, Bull. Am. Phys. Soc. Ser. 2, 1, 390 (1956).

¹² E. L. Church and J. E. Monahan, Phys. Rev. 98, 718 (1955).

¹³ D. F. Zaretskii and A. V. Shut'ko, J. Exptl. Theoret. Phys. (U.S.S.R.) **32**, 370 (1957), Soviet Phys. JETP **5**, 323 (1957).

Translated by M. Hamermesh 5

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DENSITY OF He³ - He⁴ SOLUTIONS

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The density of He³ – He⁴ solutions at saturated-vapor pressure was determined as a function of the temperature at He³ concentrations of 10, 20.1, 30.3, 41.2, 49.9, 68.5 and 85.4%. The break in the $\rho(T)$ curve affords an estimate of the λ point temperatures for concentrations of 10, 20.1, 30.3, and 41.2%.

HE usual pycnometer method was used to determine the density of $He^3 - He^4$ solutions. The pycnometer (see Fig. 1) comprised a small glass bulb 1, which narrows down into a capillary 2 of volume 6.5×10^{-3} cm³/mm and length 29 mm. A

platimun "cup" 3 is fused into the lower half of the bulb. This cup is needed to accelerate the equilibrium between the solution in the bulb and the helium in the bath. The cup has a volume of 0.44 cm^3 . Marker 4, etched on the capillary, was used as the reference.

Capillary 2 was checked with mercury for absence of taper, after which the volume of the pyc-

^{*}A supplementary communication¹¹ states that the 8.42 kev γ -transition has multipolarity M1 + E2.

nometer was carefully calibrated (accurate to $1 \times 10^{-4} \text{ cm}^3$) by weighing it empty and filled with water to various levels in the capillary. A correction for the expansion of the glass (~ 0.6%) was calculated from the data by van Agt,¹ extrapolated to the region of helium temperatures. An analogous calculation for platinum (data by Henning²) yields an equal correction.

When working with solutions there is great danger of a change in the concentration of the liquid due to enrichment of the gas phase by the lighter isotope. To reduce to a minimum the harmful volume occupied by the vapor over the solution, a small valve 5 was provided to close off the pycnometer directly at the inlet. The device makes it also easy to correct for the mass of the vapor, which is difficult to determine under conditions when the vapor is contained in the entire filling tube. The pycnometer is connected with the valve through a thin capillary 6, 0.9 mm in diameter and 12 mm long. The valve is fused into a tube of stainless steel, which serves to fill the instrument with a mixture, The valve is operated from the outside with the aid of a rod. The pyc-

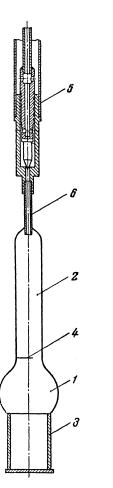


FIG. 1. Pycnometer.

nometer volume, not including the value, is $1.2785 \pm 0.0001 \text{ cm}^3$.

The apparatus used to measure the mass of the gas consisted of a small mercury gas tank 5 liters in volume, a mechanical mercury pump,³ three manometers to measure the pressures in different parts of the system, and a connecting line. The gas-tank volume was calibrated and was known to within 20 cm³. The helium-bath temperature was lowered by pumping. The temperature was determined from the vapor tension. The height of the mercury column in the manometer was measured with a cathetometer accurate to 0.1 mm. At low pressures, check readings were made with an oil-filled manometer.

The mixture was made with He³ of purity not less than 99.98%, as determined with a mass spectrometer.* The solution was prepared for each concentration by mixing known amounts of He³ and He⁴. The concentration was expressed in molar fractions He³/(He³ + He⁴) and could be determined from the ratio of the He³ and mixture pressures with a relative error of 0.3%.

The density ρ was measured as follows. Gas was fed to the bulb in small batches from system of total volume V, comprising the mercury pump, gas holder, and manometer. As soon as enough liquid was condensed, the valve was immediately closed and the gas in the line leading from the pycnometer valve to the system (of constant volume V) was pumped back into the system. A KM-5 cathetometer was used to measure the amount of gas remaining after the condensation. In the calculation of the vapor mass, the manometer readings were referred to 0°C and to standard gravity (980.665 cm/sec²).

During the measurement of the mass of the condensed gas, the room temperature remained constant within 0.5° C. An especially prepared small manometer monitored the absence of vapor in the cold tube over the pycnometer valve during the experiment. The volume of the liquid was measured with the same KM-5 cathetometer, sensitive to 0.01 mm of helium. Actually, since the walls of the helium and air Dewar flasks introduced distortions due to the inhomogeneity of the glass, the accuracy, as determined by control measurements, was somewhat less, but did not drop below 0.05 mm.

The table lists the experimental results for the density ρ of He³ – He⁴ solutions, of various concentrations, as functions of the temperature. The

^{*}The mass-spectrometric analysis of the helium was performed by the laboratory of N. E. Alekseevskii.

Densities of Solutions He³-He⁴

$ \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$	c, %	<i>т</i> , °К	ρ , g/cm ³	c, %	<i>t</i> , °K	ρ , g/cm ³	c, %	<i>Т</i> , ⁰Қ	ρ , g/cm ³
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	10	$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	41.2	$\begin{array}{c} \textbf{1.661}\\ \textbf{1.764}\\ \textbf{1.824}\\ \textbf{1.962}\\ \textbf{2.134}\\ \textbf{2.345}\\ \textbf{2.682}\\ \textbf{3.060}\\ \textbf{3.257}\\ \textbf{3.399}\\ \textbf{3.767}\\ \textbf{1.374}\\ \textbf{1.480}\\ \textbf{1.577}\\ \textbf{1.673}\\ \textbf{1.765}\\ \textbf{2.038}\\ \textbf{2.03}\\ \textbf{2.203}\\ \textbf{2.338}\\ \textbf{2.630}\\ \textbf{2.779}\\ \textbf{2.948}\\ \textbf{3.122}\\ \textbf{3.202}\\ \textbf{3.432}\\ \textbf{3.572}\\ \textbf{1.533}\\ \textbf{1.447}\\ \textbf{1.534}\\ \textbf{1.669}\\ \textbf{1.778}\\ \textbf{1.883}\\ \textbf{2.067}\\ \textbf{2.334}\\ \end{array}$	$\begin{matrix} 0, 12508\\ 0, 12494\\ 0, 12492\\ 0, 12440\\ 0, 12388\\ 0, 12259\\ 0, 12013\\ 0, 11703\\ 0, 11703\\ 0, 11703\\ 0, 11703\\ 0, 11703\\ 0, 11771\\ 0, 10766\\ 0, 11773\\ 0, 10766\\ 0, 11773\\ 0, 10766\\ 0, 11773\\ 0, 10766\\ 0, 11773\\ 0, 10766\\ 0, 11773\\ 0, 10766\\ 0, 11751\\ 0, 11751\\ 0, 11751\\ 0, 11751\\ 0, 11752\\ 0, 11628\\ 0, 11628\\ 0, 11628\\ 0, 11628\\ 0, 10698\\ 0, 10698\\ 0, 10433\\ 0, 10212\\ 0, 11186\\ 0, 11180\\ 0, 11186\\ 0, 11186\\ 0, 11186\\ 0, 11186\\ 0, 11186\\ 0, 11186\\ 0, 10983\\ 0, 10983\\ 0, 10821\\ 0,$	85,4	$\begin{array}{c} 3.215\\ 3.333\\ 3.404\\ 1.309\\ 1.447\\ 1.606\\ 1.651\\ 1.914\\ 2.284\\ 2.577\\ 2.936\\ 3.064\\ 3.278\\ 3.329\\ 3.443\\ 1.323\\ 1.355\\ 1.401\\ 1.503\\ 1.646\\ 1.777\\ 2.252\\ 2.486\\ 2.693\\ 2.758\\ 2.787\\ 2.871\\ 2.933\\ 3.069\\ 1.315\\ 1.401\\ 1.600\\ 1.888\\ 2.162\\ 2.360\\ \end{array}$	\$\rho\$, g/cm³ 0,10043 0.09855 0,09770 0.09966 0.09948 0.09898 0.09887* 0.09887* 0.09887* 0.09887* 0.09887* 0.09887* 0.09887* 0.09887* 0.09898 0.09887* 0.08464 0.08846 0.08846 0.08846 0.08847 0.08914 0.08846 0.08846 0.08846 0.08846 0.08846 0.08846 0.08464 0.07930 0.07791 0.07667 0.077413 0.088111 0.08811 0.08811 0.08811 0.07686 0.07648 0.07648 0.07443 0.07443

table contains also the values of the density of pure He^3 , measured by the same procedure as used for the solutions. The values obtained are in good agreement with the data of Ref. 4. The temperatures are given in the van Dyke and Duryea 1955 scale. The densities of the solutions were calculated from the formula

$$\rho = \frac{[c\mu_{3} + (1 - c)\mu_{4}] \, pVT_{0}}{vT \, p_{0} V_{0}} \, ,$$

where c is the concentration of He³ in the solution, v the volume of the liquid in the pycnometer, and p and v the pressure and volume of the gas in the system, taken at room temperature T. The subscript 0 denotes the same quantities at standard conditions. The atomic weights of He³ and He⁴ were taken to be $\mu_3 = 3.017$ and $\mu_4 = 4.0039$. Since the volume occupied by the saturated vapor was small, the He³ content of the vapor did not exceed 0.5 - 0.7% of the condensed He³ even under the most unfavorable conditions, and the correction for the change in the liquid concentration was therefore disregarded.

The corrections for the mass of the vapor were based on the data on the vapor tensions of solutions,

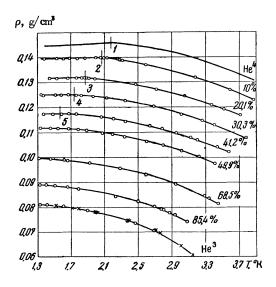
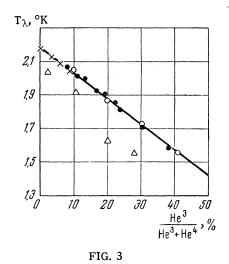


FIG. 2. Dependence of the density of the solutions on the temperature: $\times -$ Kerr's data, $\bigcirc -$ results of the present work. The curve for He⁴ was plotted from data of Box and Kammerling-Onnes.⁷ The vertical bars denote the temperatures of the λ transitions: $1 - 2.17^{\circ}$ K (from Ref. 9), $2 - 2.06^{\circ}$ K, $3 - 1.87^{\circ}$ K, $4 - 1.73^{\circ}$ K, $5 - 1.56^{\circ}$ K.



 ρ , g/cm³

0,133

0,131

0,129

0,127

0,125

0,123

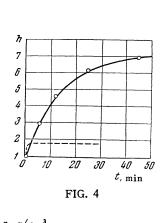
1,3

1,5

1,7

FIG. 5

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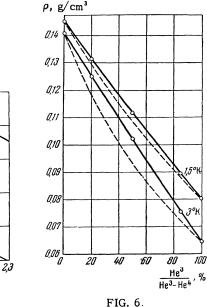


FIG. 3. Dependence of T_λ on the concentration:
× - data of Ref. 9, ● - Esel'son et al.,⁸ Δ Abraham et al.,¹⁰ ○ - results of the present work.
FIG. 4. Curves showing establishment of equi-

librium. h – height of liquid column in the bulb (in arbitrary units), t – time in minutes. Solid curve – T = 1.852° K, dotted curve – T = 1.885° K.

FIG. 5. Dependence of ρ (T) in the vicinity of the λ point for concentrations of 20.1 (a) and 30.3% (b).

FIG. 6. Dependence of the density of the solutions on the concentration. Solid curve – experimental, dotted curve – calculated under the assumption that molar volumes are additive.

obtained by Esel' son and Berezniak.⁵ The vapor pressure of He³ was obtained from Ref. 6. The experimental results are plotted in Fig. 2.

2,1 Т,°К

Three to five experiments were performed for each concentration, to avoid random errors due to incorrect determination of the mass of the condensed gas. The spread of the points relative to each of the curves for $\rho(T)$ does not exceed 0.2%, although the estimated possible systematic error given is on the order of 1%. It is interesting to note that as the concentration increases, the fairly sharp break at the λ point, characteristic of the $\rho(T)$ curve of He⁴, becomes smoothed out. The densities below T_{λ} become constant within 0.1% for concentrations of 30.3 and 41.2%.

The break in the $\rho(T)$ curve made it possible to determine the temperatures of the λ transition for the 10, 20.1, 30.3, and 41.2% concentrations. The λ temperatures were determined for the last two concentrations from the intersection of the tangents at the bend. The points determined in this manner (Fig. 3) are in good agreement with the results of Ref. 8 to within an experimental accuracy of $\pm 0.05^{\circ}$ K.

Let us note that it takes much longer to establish equilibrium in the solutions at temperatures below T_{λ} (50-60 minutes) than at temperatures above the λ transition, when the measurements can be made in practice within 3-5 minutes after the helium-bath temperature is established. Since control measurements with pure He⁴ have shown that the equilibrium is reached here very rapidly (3-5 minutes) over the entire investigated temperature range from 1.3 to 4°K, we can conclude that as the bath temperature is lowered, the pycnometer becomes unevenly cooled and the normal liquid carries the He³ towards the colder portion of the apparatus; then, as the concentration becomes equalized, the system acquires equilibrium through a slow diffusion process. Figure 4 shows typical settling curves for one of the experiments. The solid curve was plotted for the point 1 of Fig. 5, and the dotted one for point 2. Comparison of the two curves shows that point 2 lies above T_{λ} , while point 1 is in the He II region. Thus, the temperature of the λ transition can also be determined from the character of the equilibrium curves.

To verify that the measurements made at temperatures above T_{λ} were really performed in the equilibrium state, several points (marked in the table by asterisks) were obtained under conditions, in which the required temperature was obtained by placing a heater under the pycnometer bulb. After the bath temperature was fixed, the heater was turned off and the dependence of the position of the level in the capillary on the time was plotted. The points corresponding to equilibrium under these conditions fit the $\rho(T)$ curves quite well.

The experimental values of the densities of the solutions differ somewhat from the theoretical ones, calculated under the assumption that the molar volumes are additive (Fig. 6). For a given concentration, this difference increases with the temperature, reaching, for example, 8.5% at c =

50% and T = 3°K. Such a behavior of the density corresponds to a reduction in the molar volumes of He³ and He⁴ upon dissolution.

In conclusion, I express deep gratitude to V. P. Peshkov for valuable advice and help in this work.

¹ F. P. G. A. J. van Agt, H. Kammerling Onnes, Proc. Amsterdam **28**, 667 (1925).

² F. Henning, Ann. Physik **22**, 631 (1907).

³ I. B. Danilov, Приборы и техника эксперимента (<u>Instruments and Measurement Engineering</u>) 1, 93 (1956).

⁴E. C. Kerr, Phys. Rev. 96, 551 (1954).

⁵ B. N. Esel' son and N. G. Berezniak, J. Exptl. Theoret. Phys. (U.S.S.R.) **30**, 628 (1956), Soviet Phys. JETP **3**, 568 (1956).

⁶ Abraham, Osborne, and Weinstock, Phys. Rev. 80, 366 (1950).

⁷ P. H. Keeson, <u>Helium</u>, Amsterdam 1942, p 207. ⁸ Esel' son, Berezniak, and Kaganov, Dokl. Akad.

Nauk SSSR 111, 568 (1956), Soviet Phys. "Doklady" 1, 683 (1956).

⁹J. G. Dash and K. D. Taylor, Phys. Rev. 99, 598 (1955).

¹⁰ Abraham, Weinstock, and Osborne, Phys. Rev. **76**, 864 (1949).

Translated by J. G. Adashko 6

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