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Vapor Pressure of He³-He⁴ Solutions*

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Measurements have been made of the vapor pressures of solutions of He³ in He⁴ having molar He³ concentrations 1.91, 3.34, 6.51, and 12.04% in the temperature range 1.4°K to 2.6°K. The measurements involved the observation of the excess pressure of the solution over that of pure liquid He⁴ which was carefully maintained at the same temperature as the solution using an apparatus which ensured high precision of pressure transmission between the low- and high-temperature parts of the apparatus, especially for liquid He⁴ temperatures above the lambda temperature. The results are compared with previous evaluations and comparisons made with recent theories of He³-He⁴ solutions.

I. INTRODUCTION

IN the past several years there have been many measurements¹⁻⁸ of the vapor pressures of solutions of He³ in He⁴. Much of the work has been of an exploratory character or has confined its attention to a small range of either He³ concentration or temperature.

Three investigations, reported by Sommers,⁹ by Eselson and Bereznyak,¹⁰ and by Roberts and Sydoriak,¹¹ however, covered a wide range of temperature and He³ concentration and represent the most significant complete work in the field.

* Supported by a Grant from the National Science Foundation.

¹ Weinstock, Osborne, and Abraham, *Phys. Rev.* **77**, 400 (1950).

² Eselson, Lazarew, and Alekseevski, *J. Exptl. Theoret. Phys. U.S.S.R.* **20**, 1055 (1950).

³ J. G. Daunt and C. V. Heer, *Phys. Rev.* **86**, 205 (1952).

⁴ B. N. Eselson, *J. Exptl. Theoret. Phys. U.S.S.R.* **26**, 744 (1954).

⁵ J. G. Daunt and T. P. Tseng, *Suppl. Bull. inst. intern. froid Annexe 1955*, **3**, 22 (1955).

⁶ Wansink, Taconis, and Staas, *Physica* **22**, 449 (1956).

⁷ V. P. Peshkov and V. N. Kachinskii, *J. Exptl. Theoret. Phys. U.S.S.R.* **31**, 70 (1956).

⁸ A. K. Sreedhar and J. G. Daunt, *Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry, Madison, Wisconsin, August, 1957*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 168. See also Ohio State Symposium on Liquid and Solid He³, Ohio State University, Columbus, 1957, p. 141.

⁹ H. S. Sommers, Jr., *Phys. Rev.* **88**, 113 (1952).

¹⁰ B. N. Eselson and N. G. Bereznyak, *J. Exptl. Theoret. Phys. U.S.S.R.* **30**, 628 (1956) [translation: *Soviet Phys. (JETP)* **3**, 568 (1956)].

¹¹ T. R. Roberts and S. G. Sydoriak, *Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry, Madison, Wisconsin, August, 1957*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 170.

The experiments reported here were a continuation of previous work,^{5,8} and are especially concerned with the vapor pressures at and above the lambda temperatures of the solutions, T_λ^α , for He³ concentrations, α , of from 1 to 12%, a region not stressed in the work of others. Our previous work^{5,8} had shown that even above T_λ^α the solutions showed nonideal behavior. It showed moreover a vapor pressure anomaly at T_λ^0 , the lambda temperature of pure liquid He⁴, which was thought to be an experimental artifact. This disconcerting anomaly at T_λ^0 is also evident in the work of Eselson and Bereznyak. The present work therefore was undertaken, using completely different apparatus than heretofore, in order to investigate the reality of this apparent anomaly at T_λ^0 and to provide satisfactory data on the vapor pressures above T_λ^α .

It is considered that the results reported here, which show no anomalies at T_λ^0 , can be regarded as free from consistent error. Together with the other data quoted,⁹ suitably weighted, there now is a satisfactorily complete tabulation of vapor pressures of He³-He⁴ solutions. This allows one to suggest that it is an appropriate time now to consider undertaking the computation of detailed thermodynamic potentials of the solutions, particularly in view of the fact that other vital data, such as volume changes on mixing^{12,13} and virial coefficients,¹⁴ have already been established. Appro-

¹² E. C. Kerr, reference 11, p. 158.

¹³ T. P. Ptukha, *J. Exptl. Theoret. Phys. U.S.S.R.* **34**, 33 (1958) [translation: *Soviet Phys. JETP* **7**, 22 (1958)].

¹⁴ W. E. Keller, *Phys. Rev.* **98**, 1571 (1955); and Kilpatrick, Keller, and Hammel, *Phys. Rev.* **97**, 9 (1955).

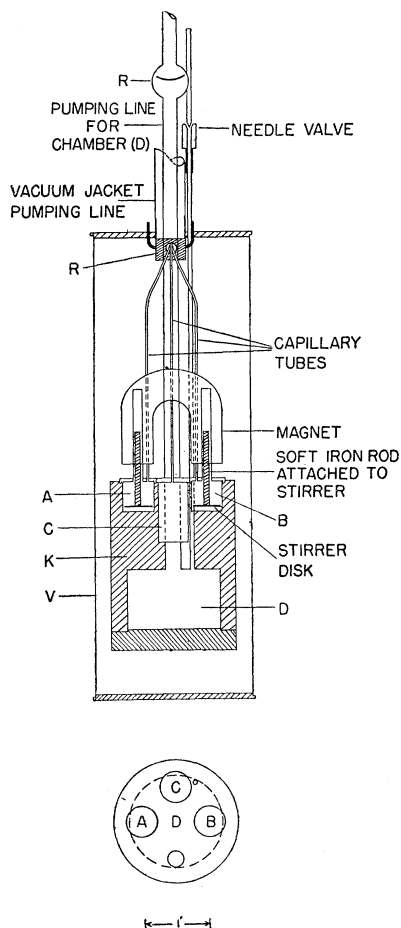


FIG. 1. Scale drawing of low-temperature part of apparatus, which is immersed in liquid helium contained in a Dewar (not shown).

priately these computations should go forward together with detailed numerical comparison with the various theoretical models,¹⁵ as has been outlined by Daunt.¹⁶ A very elegant start in this direction has been made by Wansink¹⁷ and some preliminary computations are reported in the present paper and elsewhere by Sreedhar.¹⁸ It is hoped that these will be extended in a future publication.

II. APPARATUS

The vapor pressure measurements were made by measuring the differential pressure, Δp , between a vessel or "cell" containing the He^3 - He^4 solution and a cell containing pure He^4 , the two cells being maintained at the same temperature in the range 1.4°K to 2.6°K. To ensure temperature uniformity the two

identical cells, *A* and *B* (see Fig. 1), were hollowed out of one copper block, *K*, of dimensions: length=7.5 cm and diameter=5.0 cm. The inside dimensions of each of the vapor pressure cells were length=1.11 cm and diameter=1.27 cm.

As is shown in the scale drawing of the low-temperature part of the apparatus given in Fig. 1, the copper block was located inside a vacuum vessel *V*, which was immersed in a bath of liquid helium (not shown in Fig. 1). The initial cooling of the block to the temperature of the bath was done by admitting exchange gas into the vacuum jacket *V*; whereas during an experiment the vacuum jacket was pumped out to a high vacuum via a 12-mm diameter german silver pumping tube, and the block temperature maintained by a subsidiary liquid helium reservoir, *D*, of volume approximately 30 cc within the block. Liquid helium from the main bath could be let into this chamber, *D*, by means of a needle valve controlled from the top of the cryostat. The liquid in this chamber served to maintain constancy of temperature of the copper block and of the condensed liquids in the two vapor pressure cells, the temperature being regulated by pumping on the liquid helium in the chamber *D*, through a 6-mm diameter Inconel tubing. The pressure could be maintained at any desired value by means of regulating valves in the pumping line. The temperature of the copper block could thus be thermostated at any value between 2.6°K and 1.4°K.

Two capillary tubes of Inconel of internal diameter 1.0 mm connected the tops of the vapor pressure cells, *A* and *B*, to a manometer manifold at room temperature. The manifold consisted of two butyl-phthalate oil manometers to measure the absolute pressures, p_4^0 and p , of pure liquid He^4 and of the solution, respectively, and a differential manometer for measuring the pressure difference, $\Delta p = p - p_4^0$. These manometers were of glass of i.d. 0.50 cm. The total dead volume of the lines leading from the copper block and of the manometers was of the order of 25 cc.

In order to avoid possible cold spots in the tubes leading from the cells to the manometers, the capillary tubes, as well as the entire copper block, were surrounded by the vacuum jacket, *V*. The capillary tubes were brought up to room temperature through the 12-mm diameter tube used for pumping the vacuum jacket, care being taken to avoid thermal contacts along their lengths. Radiation shields, *R*, were installed in the pumping lines to avoid excessive heat influx to the copper block.

One of the main sources of error in these types of experiments can be inhomogeneity of the He^3 - He^4 solution in the vapor pressure cell. To minimize this a stirrer was placed in the cell. This stirrer consisted of a circular aluminum disk of diameter just less than the i.d. of the cell and having about 100 holes of 0.4-mm diameter drilled through it. It could be moved vertically up and down through the liquid in the cell by means of

¹⁵ See reference 16, also J. G. Daunt, *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1952), Vol. 1, p. 209, and Sec. VI of this paper for references to the theoretical work.

¹⁶ J. G. Daunt, reference 11, p. 151.

¹⁷ D. H. N. Wansink, *Physica* 23, 140 (1957).

¹⁸ A. K. Sreedhar, Ph.D. dissertation, Ohio State University, 1958 (unpublished).

a permanent magnet outside the cell, the magnet actuating a soft iron rod attached to the aluminum disk (see Fig. 1). A similar stirrer was installed in the pure He⁴ cell. The magnets were moved by means of a nylon string connected to the plunger of a solenoid, outside the Dewar. The entire system worked in such a way that, by operating the solenoid, the liquids in the cells were forced to flow through the small holes in the stirrers.

Provision was made for a constant volume He³ gas thermometer. The low temperature part of this consisted of a cylindrical thermometer bulb, *C*, of volume 3.24 cc, hollowed out of the copper block. A capillary tube led from this bulb to a butyl-phthalate constant volume oil manometer.

III. PROCEDURE

The copper block was cooled to 4.2°K by admitting exchange gas into the vacuum jacket. The main helium bath was then pumped down to 2°K. At this temperature pure He⁴ gas from the storage was condensed into the vapor pressure cell until the cell was 9/10ths full. The He³-He⁴ mixture gas was then admitted in stages into the solution vapor pressure cell, the amount of mixture condensed being measured in each experiment. The entire gaseous mixture at each stage was pumped into the vapor pressure cell by means of a Toepler pump. This procedure ensured the avoidance of any depletion of He³ in the condensation process. At the end of each stage of condensation a reading, $\Delta p = p - p^0$, of the differential pressure was taken. The process was continued until the solution cell was also 9/10ths full. During all this process both the stirrers were operated continuously. A plot of Δp against the amount of liquid mixture in the cell was then made. A typical plot is shown in Fig. 2. It was found that the differential pressure was independent of the volume of the liquid in the solution cell when the cell was filled by the solution to more than 7/10ths of its volume. After that Δp remained independent of the amount of filling until the cell was completely full as is shown in Fig. 2. At this stage Δp increased suddenly, indicating the condensation of the liquid in the capillary tube. The variation of Δp with volume of solution in the cell for small filling was due to the gradual enrichment of the He³ concentration of the solution as the vapor volume was reduced.

After the two cells had been filled to 9/10ths their volumes, and after the chamber *D* was filled with liquid He⁴, the vacuum jacket was pumped out and then the temperature of the copper block was reduced in stages down to 1.4°K by pumping on the helium in the chamber *D*. At each stage sufficient time elapsed to allow the attainment of equilibrium and then the temperature was held constant to within a millidegree for about five minutes, and readings of the vapor pressures were taken.

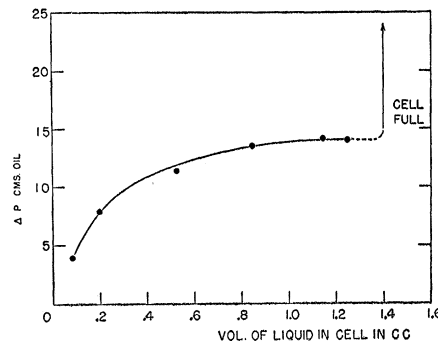


FIG. 2. Filling curve showing Δp as a function of the volume of liquid in the solution cell for a gas mixture of He³ concentration 3.34%.

IV. ERRORS AND EVALUATION OF ACCURACY

The possible sources of error that arise in an experiment of this nature are discussed below.

a. Concentration of the Mixture

The original gas mixture used in the experiments was supplied by the Atomic Energy Commission and upon mass spectroscopic examination was found to contain 6.51 ± 0.1 mole percent He³. This concentration was cross checked by diluting the original gas mixture with known amounts of pure He⁴ gas and mass spectroscopically analyzing these diluted mixtures.¹⁹ For experiments on the 3.34% and 1.91% mixtures, the original 6.51% gas mixture was diluted by adding requisite amounts of pure He⁴ to it. For experiments on the 12.04% mixture, the 6.51% mixture was enriched by adding requisite amounts of pure He³ gas to it.

It is assumed that the concentration of the solution is the same as that of the bulk gas before condensation. This is true if the amount of vapor is negligible in comparison with the amount of liquid in the vapor pressure cell. It has already been pointed out in the previous section that it was found experimentally that Δp becomes independent of the amount of liquid condensed in the cell when the liquid fills more than 7/10ths of the volume of the cell. Above this filling therefore, the vapor pressure of the liquid is independent of the amount of vapor in the cell. An estimation of the mass of vapor in the cell and in the manometer lines when the cell was 7/10ths full of liquid and at 2°K shows that the vapor mass is only about 1/2% of the total mass of liquid in the cell. Since all measurements were made with the cell 9/10ths full, we are justified in assuming that the liquid concentration is equal to that of the bulk gas before condensation with a maximum error of 1.5%. This maximum possible error was estimated by assuming a maximum value of 35 for

¹⁹ We are grateful to Dr. E. C. Kerr of Los Alamos Scientific Laboratories and Dr. F. L. Mohler of the National Bureau of Standards for kindly performing many of these mass-spectrographic measurements.

the distribution coefficient for the 6.51% solution, this value being the highest observed value¹⁸ (observed at 1.4°K) for the distribution coefficient for that solution.

b. Manometry

The vapor pressures were measured by butyl-phthalate oil manometers. These manometers were calibrated against a mercury manometer at room temperature. The oil levels in the two absolute manometers could be read to 0.1 mm by a vernier arrangement and the oil level in the differential manometer could be read to 0.2 mm causing a maximum error of 1% in Δp for the 1.91% He³ solution at 1.4°K and causing correspondingly smaller errors at all other temperatures and He³ concentrations.

It was estimated that at 2°K the thermomolecular correction for a capillary tube of 1-mm internal diameter was of the order of 5×10^{-3} mm mercury. This leads to an error of much less than 0.25% in the pressure readings when account is taken of the partial cancellation of these thermomolecular effects by the differential Δp measurement.

The pressure difference arising out of the film flow of the pure liquid He⁴ below its lambda point was estimated to be of the order of 10^{-2} mm of mercury,

leading to an error of not more than $\frac{1}{2}$ % in the pressure readings.

c. Temperature

The temperature was determined from the vapor pressure of pure He⁴ in the vapor pressure cell. The vapor pressure was measured by the butyl-phthalate oil manometer with a precision of 0.01 mm of mercury. The 1958 vapor pressure temperature scale²⁰ was used in converting the pressure readings to temperature. It was not found necessary to use the He³ gas thermometer, though provision was made for it.

d. Thermal Equilibrium and Demixing Effects

Demixing effects in the solution arising from heat leaks²¹ into the vapor pressure cells were minimized, as described in the previous section, by employing stirrers in each of the vapor pressure cells. These stirrers were actuated about twenty times a minute, thus ensuring thorough mixing and consequent absence of concentration gradients in the mixture. Moreover, the data taken on different days with different temperature cycles agreed among themselves to better than 1.0%.

V. RESULTS

The observed results giving Δp as a function of T for different He³ concentrations are shown in Fig. 3. The upward pointing arrows in Fig. 3 represent the lambda temperatures of the solutions, as computed from the data of Dash and Taylor.²²

Figure 4 shows the variation of $(p_{\text{obs}} - p_{\text{pes}})$ with temperature, where p_{obs} is observed total vapor pressure of the solution and p_{pes} the theoretical perfect classical solution value of the pressure calculated using the results of Roberts and Sydoriak²³ on the vapor pressure of pure He³ and the 1958 vapor pressure temperature scale²⁰ for pure He⁴. In evaluating p_{pes} corrections have been made for the nonideality of the vapor phase using the known values of the virial coefficients.¹⁴ Smoothed values of p_{obs} were taken from Fig. 4 at 0.1°K temperature intervals and these are tabulated in Table I.

VI. DISCUSSION

Our results are in good agreement with the results of Roberts and Sydoriak¹¹ over the entire temperature range, and with the results of Eselson and Bereznyak¹⁰ and with our previously reported results⁸ for temperatures below the lambda temperature of He⁴. Above this temperature the two latter references reported solution

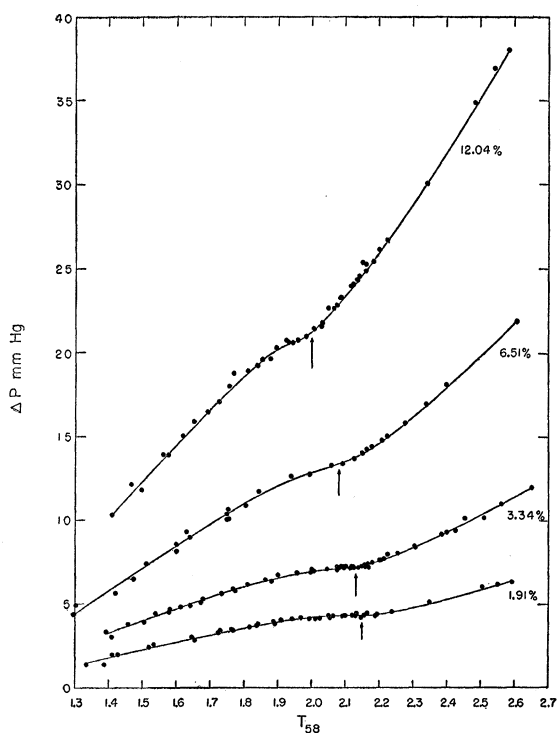


FIG. 3. Plot of the observed values of Δp ($\Delta p = p - p^0$) in mm Hg as a function of absolute temperature, T (1958 temperature scale) for the four concentrations used. The He³ solution concentrations are indicated, in percent, by the number attached to each curve. The vertical arrows mark the lambda points of the solutions.

²⁰ Van Dijk, Durieux, Clement, and Logan, *Physica* **24**, S129 (1958).

²¹ For a detailed discussion of this demixing mechanism, peculiar to superfluid helium, see J. G. Daunt and C. V. Heer, National Bureau of Standards Cryogenic Conference Report, 1951 (unpublished), p. 155.

²² J. G. Dash and R. Dean Taylor, *Phys. Rev.* **99**, 598 (1955).

²³ S. G. Sydoriak and T. R. Roberts, *Phys. Rev.* **106**, 175 (1957).

pressures higher than the present results. The origin of this difference is discussed below.

The vapor pressures of all the solutions at all temperatures show positive deviations from the perfect classical solution (pcs) values. This is clearly brought out in Fig. 4. It is seen that all solutions show changes in the slope of the vapor pressure at their lambda points, T_λ^s , in agreement with the results of other workers.⁹ Using the data on the variation of p with temperature near T_λ^s , one could calculate the jump in the specific heat, ΔC_p , on the assumption that the transition at T_λ^s is of second order, as has been pointed out by de Boer and Gorter.²⁴ There is no evidence of any anomaly at T_λ^0 , the lambda point of pure He⁴, as had been found in our earlier investigations with a simpler cryostat⁸ and as is also implicit in the results of Eselson and Bereznyak.¹⁰ The present results with a more elaborate apparatus, where the vapor pressure cells and the capillary tubes are thermally isolated from the helium bath, show that there are no anomalies

TABLE I. Smoothed values of the total vapor pressure, p_{obs} (mm Hg), versus T for various values of the molar He³ solution concentration, X .

$T^\circ\text{K} \setminus X$	0.0191	0.0334	0.0651	0.1204
1.4	3.84	5.29	7.94	12.13
1.5	5.90	7.61	10.89	15.84
1.6	8.49	10.47	14.38	20.20
1.7	11.84	14.04	18.62	25.29
1.8	16.10	18.55	23.64	31.12
1.9	21.43	24.11	29.66	37.66
2.0	27.96	30.80	36.60	45.08
2.1	35.78	38.68	44.93	54.81
2.2	44.83	48.08	55.05	66.33
2.3	55.85	59.42	67.17	79.67
2.4	68.66	72.63	81.27	95.13
2.5	83.39	87.82	97.39	112.80
2.6	100.22	105.11	115.71	132.84

at T_λ^0 . The origin of the anomalies in our preliminary investigations and in results of Eselson and Bereznyak may thus be due to the possible existence of cold spots in the capillary tubes leading to the vapor pressure cells, arising out of thermal gradients in the helium bath for temperature $T > T_\lambda^0$. These thermal gradients vanish at $T \leq T_\lambda^0$ due to the high heat conductivity of HeII in the bath.

It should be emphasized here that apart from the peculiar influence of the lambda transition of the solution on the vapor pressure, the observed results show nonideal behavior even at temperatures T greater than T_λ^s . Various attempts have been made to explain the strong positive deviations of the vapor pressure of the solutions. The theory of de Boer and Gorter²⁵ and its later modification by Nanda²⁶ assumed that below the lambda temperature of the solution He³ is only

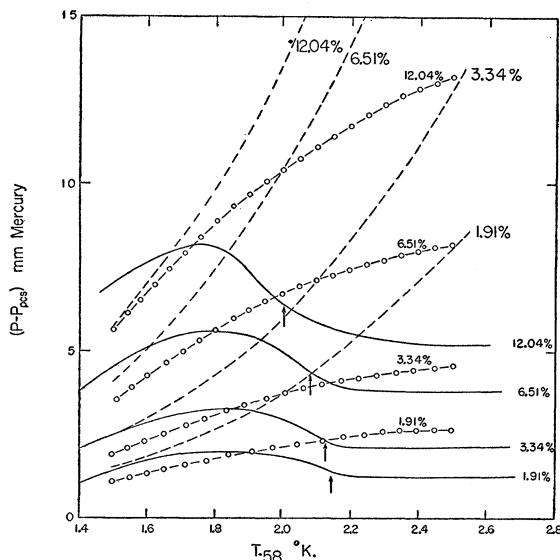


FIG. 4. The full curves plot $(p - p_{pcs})$ in mm Hg versus T (1958) for the four concentrations used. p is the total observed vapor pressure of the solution and p_{pcs} is the calculated theoretical vapor pressure for a perfect classical solution of the same concentration. The He³ solution concentrations are indicated by the number attached to each curve. The vertical arrows mark the lambda points of the solutions.

The dashed curves are computed from the theory of Prigogine *et al.*²⁹ using an approximate formulation only (see text) and the curves with circles are computed from the theory of Heer and Daunt.^{27,28}

soluble in the normal component of He⁴. This assumption introduced too great a positive deviation at lower temperatures. The theory is also open to question because of the empirical nature of the assumed expression for the Gibbs' function of pure liquid He⁴. The theory of Heer and Daunt²⁷ assumed that the solution is a mixture of Bose-Einstein and Fermi-Dirac systems in a smoothed potential well and yields values of p shown in Fig. 4. In evaluating these theoretical values of p , corrections were made for the nonideality of the vapor phase. A complete comparison of the experimental results for temperatures below the lambda temperature of the solution has been made by Sommers,⁹ Daunt, Tseng, and Heer,²⁸ and Sreedhar.¹⁸ A discussion of the results in the light of existing theories has been given by Daunt,¹⁶ where a complete reference to the literature on the theories of these mixtures can be found.

Among the more recent theories the theory of Prigogine,^{29,30} who assumed that the deviation of the observed results arises from the large difference in the molar volumes of liquid He³ and of liquid He⁴, is of particular interest because, as in the theory of Heer and Daunt, the properties of the solutions can be explicitly

²⁷ C. V. Heer and J. G. Daunt, *Phys. Rev.* **81**, 447 (1951).

²⁸ Daunt, Tseng, and Heer, *Phys. Rev.* **86**, 911 (1952).

²⁹ Prigogine, Bingen, and Bellemans, *Physica* **20**, 633 (1954).

³⁰ I. Prigogine, *Molecular Theory of Solutions* (North-Holland Publishing Company, Amsterdam, 1957).

²⁴ J. de Boer and C. J. Gorter, *Physica* **18**, 565 (1952).

²⁵ J. de Boer and C. J. Gorter, *Physica* **16**, 228, 667 (1950).

²⁶ V. S. Nanda, *Phys. Rev.* **97**, 571 (1955).

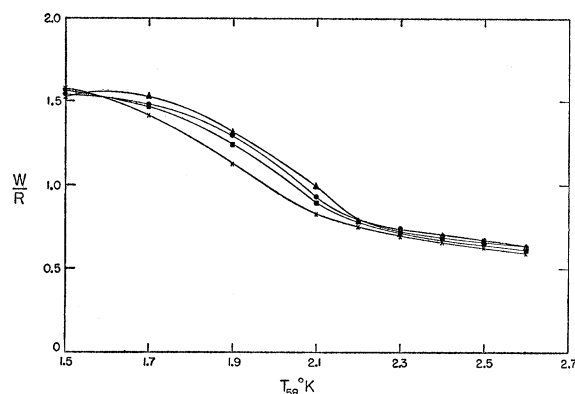


FIG. 5. Plot of the nonideality parameter, W , in units of the universal gas constant, R , versus T (1958), computed from the observed vapor pressures for the four He^3 solution concentrations, X , used. Δ for $X=1.91\%$; \odot for $X=3.34\%$; \square for $X=6.51\%$; \times for $X=12.04\%$.

described in terms of the properties of the pure components only. The necessary data, namely the compressibilities³¹ and the molar volumes,^{32,33} are now known. Using these data, a numerical evaluation of the vapor pressure on the basis of Prigogine's theory has been made for the four solutions under investigation. The results are shown in Fig. 4 by the broken curves. In this evaluation correction has been made for the nonideality of the vapor using the experimental data of Keller¹⁴ on the virial coefficients of He^3 and the data of Kilpatrick, Keller, and Hammel¹⁴ on the virial coefficients of He^4 . This evaluation of p was made using the approximate expansion for the free energy given by Prigogine [Eq. (19.3.7), reference 30], and it is probable that the more exact expression [Eq. (19.4.1), reference 30] would yield modified results.³⁴ However,

³¹ Laquer, Sydoriak, and Roberts, *Phys. Rev.* **113**, 417 (1959).

³² E. C. Kerr, *Phys. Rev.* **96**, 551 (1954).

³³ H. Van Dijk and M. Durieux, *Physica* **24**, 1 (1958).

³⁴ For example, the approximate expression for the free energy leads to values of the volume changes on mixing at 1.6°K which change sign as the concentration varies, a behavior very different from observation (see references 12 and 13). We would like to

numerical evaluation of the latter equation is not possible without a detailed knowledge of the p , v , T data over a wide range of temperature and pressure.

In order to investigate the nature of the deviation from ideality, a nonideality parameter W , assumed to be concentration independent and defined by the Gibbs' function of the solution

$$G = XG_0^3 + (1-X)G_0^4 + RT[X \log X + (1-X) \log(1-X)] + X(1-X)W$$

has been calculated from our results by a method of successive approximations, which is similar to the method of Morrow³⁵ and which takes full account of vapor imperfections. Figure 5 shows a plot of this parameter as a function of temperature for various concentrations. It is seen that W is a function of temperature showing, thereby, that a temperature independent nonideality parameter is not able to account for the observed results. It is also seen that W decreases with the concentration, but for temperatures above 2.2°K the changes are within the uncertainty in the concentration determination of the mixtures. In this higher temperature range, therefore, for the range of He^3 concentrations studied, it may be possible to describe our results by the generalized regular solution theory of Chester³⁶ which contains a temperature dependent, concentration independent, nonideality parameter, W .

VII. ACKNOWLEDGMENTS

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thank Professor Prigogine and Dr. Bellemans for valuable discussions of this matter and for pointing out the approximate nature of Eq. (19.3.7), reference 30.

³⁵ J. C. Morrow, *Phys. Rev.* **84**, 502 (1951); **89**, 1034 (1952).

³⁶ G. V. Chester, *Phys. Rev.* **100**, 446 (1955).