= 0.0065 ± 0.0005°K obtained by Peshkov[4] using a similar technique.

Assuming the validity of Eq. (1) at the temperature of the density maximum and using the measured value[2] of C_p at T_M, yields[6]

$$(dS/dT)_\lambda = 2.74 \pm 0.08 \ \mathrm{J\,g^{-1}\,(°K)^{-2}}. \qquad (4)$$

This value of $(dS/dT)_\lambda$ can be compared to Chase, Maxwell, and Millett's[7] value of 2.88 $\mathrm{J\,g^{-1}\,(°K)^{-2}}$ and Kerr and Taylor's[8] values of 2.94 $\mathrm{J\,g^{-1}\,(°K)^{-2}}$ for $T > T_\lambda$ and 2.46 $\mathrm{J\,g^{-1}\,(°K)^{-2}}$ for $T < T_\lambda$.

Note added in proof. Recently the specific heat C_p above T_λ has been measured with high accuracy [G. Ahlers, Bull. Am. Phys. Soc. 13, 506 (1968) and private communication]. Using this value of C_p gives $(dS/dT)_\lambda = 2.69 \pm 0.04 \ \mathrm{J\,g^{-1}\,(°K)^{-2}}$.

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Phase Separation in He^3-He^4 Mixtures Observed With Slow Neutrons*

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A method of studying phase separation in He^3-He^4 mixtures is described in which thermal neutrons are used to determine the He^3 number density in each phase. Results of measurements made over the temperature range 0.04–0.85°K are presented and compared with those obtained by other methods. Extrapolation of the measurements made below 0.15°K indicates that the limiting solubility of He^3 in He^4 at $T = 0$°K is $6.84 \pm 0.06\%$. The second phase at low temperatures is found to be pure He^3 within the 1% accuracy of the measurement.

I. INTRODUCTION

Below 0.87°K mixtures of liquid He^3 and He^4 separate into two phases, an upper He^3-rich phase and a lower He^4-rich phase. In general, the description of this phase separation is a problem of considerable complexity. At low temperatures, however, the situation becomes somewhat better defined, since each phase is a dilute solution of one isotope in the other, and the description is considerably simplified. Thus, in principle, the most useful measurements for comparison with theory would appear to be those made at low temperatures, i.e., temperatures below 0.1°K. In practice, however, experiments in this temperature range pose enough difficulties so that they were not seriously attempted before 1965. Nevertheless, by 1965 the general characteristics of the phase diagram above 0.1°K were reasonably well established.

A variety of methods of measuring the phase diagram were used, all of them belonging to one or the other of two rather distinctly different categories of experiments.

The first of these consisted of experiments in which mixtures of known concentration were prepared and cooled through the one-phase region. The end of the one-phase region, i.e., the phase separation line, was noted by observing some property which changed abruptly at this point. For example, phase separation has been observed visually,[1] via the sound velocity,[2] the density,[3] and the specific heat.[4]

A second class of experiments comprised measurements on the separate phases *after* phase separation. Among these were the original NMR studies of Walters and Fairbank,[5] in which phase separation was first observed, and also the recent precision measurements of the density of either one or both phases.[6, 7]

For reasons which will be discussed in the next section, experiments with the separated phases offer certain advantages, particularly in terms of control over systematic errors. The current emphasis at both lower and higher temperatures has therefore been on work of this type, although the earliest work below 0.1°K, i.e., the specific-heat measurements of Edwards *et al.*,[8] involved cooling in the one-phase region to the phase boundary.

It is our purpose in this paper to describe measurements in the separated phases which utilized thermal neutrons to determine the He^3 number density. This technique yields a precise determination of the phase separation line over a wide temperature range; furthermore, the limitations and sources of systematic error are very different from those encountered using other methods. The neutron measurements therefore provide a valuable check on the consistency of the other results. By and large, the agreement is excellent. A particular point of interest is that our observations confirm the recent evidence from the other experiments[7, 8] that He^3 remains soluble in He^4 even at the lowest temperatures.

II. CRITIQUE OF THE VARIOUS METHODS OF MEASURING PHASE SEPARATION

Let us now turn to a more detailed examination of each of the classes of experiments mentioned in Sec. I.

Because of the fact that experiments of the first type are essentially performed in the one-phase region, they have in common certain possibilities for systematic error which have already been discussed by Sydoriak and Roberts.[9] Basically the problem is that only the average concentration of the sample is known precisely; its concentration as a function of position in the apparatus is not established. Thus, if there is an appreciable vapor volume above the sample liquid, the concentration of the liquid will only be well known if the mass and composition of this vapor are known. Since the vapor volume usually extends from the low-temperature space to room temperature, it may be difficult to account for this satisfactorily. If, in addition, a portion of the vapor volume changes temperature during the course of the experiment, then the concentration of the liquid will also change as vapor is either condensed or evaporated. This leads to a further complication, since the heat of condensation and mixing of this vapor must be transferred to the thermal sink. It is entirely possible in this circumstance that the liquid temperature can be significantly different from that of whatever thermometer is being used.

If the sample cell is not the coldest point in the apparatus, He^3 will preferentially distill out of the sample and deposit on a colder wall, from which it may or may not return, depending upon geometry, surface tension, etc. In addition, a temperature gradient across the liquid will lead to comparable concentration gradients, particularly in the superfluid region. These last two effects are particularly difficult to avoid in apparatus designed to operate below 0.3°K.

Although it may be possible to minimize these sources of error by appropriate experimental design, it is even better to utilize techniques which are not affected by them. This is the case with measurements in the second class, which are made on the liquid phases after phase separation has occurred. The presence of the second phase reduces the number of degrees of freedom, according to the Gibbs phase rule. Consequently at a given temperature and pressure the concentrations of the two phases are fixed, independent of the average concentration, and only their relative volumes may vary. As a peripheral advantage the entire phase diagram can be traced out, at least in principle, with just one starting concentration. This is more than a convenience advantage over the methods in the first class, for which each point requires a new starting concentration. It also results in a far higher density of experimental points.

There are, of course, certain disadvantages in this approach. For example, none of the methods in the second category measures the He^3 concentration directly. The capacitance densitometer technique described in Refs. 6 and 7 requires a calibration with a mixture of known concentration in the one-phase region. This process is subject to the same difficulties which affect measurements in the first class. Further, it is apparent from the data in Ref. 7 that the thermal expansion of the solutions has to be accurately determined. In the NMR measurements, assumptions must be made as to the density and susceptibility as a function of concentration. For the neutron experiments to be described, the density must be known *a priori*.

It should be emphasized, however, that by combining the results of these various measurements the phase diagram can be determined free of any assumptions. The susceptibility and thermal expansion of the phases along the phase separation line would also follow from these combined results.

III. DESCRIPTION OF THE NEUTRON METHOD

A. Interaction of He^3 with Neutrons

The interaction of slow neutrons with He^3 proceeds almost entirely via the reaction $He^3\,(n,p)T$. At the neutron energy of 0.0754 eV used for this experiment, the cross section for this reaction is 3080 b.[10] Since the cross section varies almost exactly as $1/v$,[11] where v is the neutron velocity, the reaction rate is independent of how the He^3 nucleus is bound in the target. Binding affects the scattering rate, but since the scattering cross section is less than 5 b,[10] it can be neglected. The interaction of slow neutrons with He^4 is entirely elastic scattering, with a cross section of the order of 1 b. Except for very dilute solutions of He^3 in He^4, this can also be neglected.

Thus, if a beam of neutrons is passed through a sample containing He^3, essentially all the neutrons removed from the beam will be removed by the $He^3\,(n,p)T$ reaction, with a cross section that does not depend upon the physical state of the He^3. The number density of He^3 atoms is then directly determined by the degree to which the beam is attenuated. Finally it should be pointed out that the neutron beam causes virtually no disturbance in the sample. The Q value of the $He^3\,(n,p)T$ reaction is 764 keV. With about 10^2 neutrons/sec absorbed by the target, the heat input is less than 0.01 erg/min which is negligible compared to the other sources of target heating.

B. Transmission Measurements

Let a beam of incident intensity I_0 neutrons/cm^2 sec pass through a target containing N_i nuclei/cm^3 in a thickness t_i of the ith nuclear species, with cross section σ_i. The transmitted beam intensity I is given by the well-known law

$$I = I_0 \exp[-\textstyle\sum_i (Nt\sigma)_i]. \qquad (1)$$

We define the transmission τ as I/I_0.

If the neutron beam is polyenergetic, (1) must be integrated over the incident neutron spectrum, remembering that the σ_i are, in general, functions of the neutron energy. This would make the results difficult to analyze. We therefore use a mono-energetic beam of energy 0.0754 eV, provided by Bragg diffraction from a single crystal of $Co_{0.92}Fe_{0.08}$ alloy. Higher-order reflections are removed with a Pu^{239} filter. The beam is also polarized, which is incidental to the phase diagram determination but allows us to measure, if we so desire, the nuclear susceptibility of He^3.[12]

After a solution has been cooled below its phase separation temperature, the He^3-rich phase, being lighter, floats on top of the heavier He^4-rich phase. By using a sharply defined beam, and traversing it along the sample, the transmission of each phase may be measured separately. In principle, an additional measurement is then made with the solution removed from the cell, which gives the terms in (1) due to extraneous materials in the beam, such as the cell walls and supporting structure. The ratio of the two measurements gives the value of $(Nt\sigma)$ for the He^3 alone, from which N follows if t and σ are known.

For optimum statistical accuracy, a target thickness for which $(Nt\sigma)$~2 is generally chosen. For pure liquid He^3 at 1°K this thickness is 0.016 in. Rather than attempt to build a cell of very uniform thickness, we found it easier to calibrate the cell thickness by measuring the transmission τ_3 of pure He^3 liquid at various positions in the cell. Combining this with the transmissions τ_x through the mixture and τ_0 through the empty cell, it follows from (1) that

$$\frac{\ln[\tau_x/\tau_0]}{\ln[\tau_3/\tau_0]} = \frac{(Nt\sigma)_{3x}}{(Nt\sigma)_3} = \frac{N_{3x}}{N_3} = X\left(\frac{v_3}{v_x}\right), \qquad (2)$$

where N_{3x} is the He^3 number density in a mixture of concentration X and molar volume v_x, and N_3 and v_3 are equivalent quantities for pure He^3 under the conditions of temperature and pressure used in the calibration run.

C. Precision of the Measurements

The precision of the various transmission measurements in (2) is determined by the total number of neutron counts collected. If we compare the relative precision of measurements at different X, the precision with which τ_3 and τ_0 are known will not enter. These latter are only important when comparing transmissions taken at different positions in the cell and when judging the over-all accuracy of the data. For the relative precision, then, we find by differentiating (2)

$$\frac{d(N_{3x})}{N_{3x}} = -\frac{1}{(Nt\sigma)_{3x}} \frac{d(\tau_x)}{\tau_x}. \qquad (3)$$

In the concentrated phase at low temperatures, $(Nt\sigma)_{3x}$ approaches 2, but with a count rate of ~10/sec it is not practical to collect more than ~10^4 counts, i.e., $d\tau/\tau = 1.0\%$. A concentrated phase point thus will have a precision on the order of 0.5%. The observed scatter in the data is of this order. In the dilute phase at low temperatures, $(Nt\sigma)_{3x}$ approaches 0.2, but the count rate is higher by the factor $\exp(-0.2)/\exp(-2)$. It is then feasible to collect 10^5 counts at a point for a precision of ~1.5%. The data scatter somewhat more than this, for reasons which will be discussed below. If a number of such points are combined, however, the result is a phase separation curve with a precision of better than 1% in regions in which the points are sufficiently dense. The over-all accuracy is then set by the measurements of τ_3 and τ_0, and is also expected to be of the order of 1%.

D. Experimental Precautions

While the measurements needed to apply (2) are relatively straightforward, a number of experimental precautions must be observed.

1. Ideally, the only neutrons which enter the counter should be those of the desired energy which traverse the sample region under study. To this end we have defined the beam by slits 0.500-in. wide by 0.100-in. high at both the exit and entrance windows of the cryostat, in addition to multiple-slit collimators which limit the angular divergence of the beam to 15 min. of arc. The ideal is not quite realized, however. There is a background count which is measured, as usual, by rotating the monochromating crystal "off Bragg." In view of the low count rates in this experiment, special care was taken to reduce this background to a low value (less than 0.7/sec) and to check it for possible variation with time. This background is measured separately for I and I_0 in Eq. (1), for each of the transmissions τ_x, τ_3, and τ_0.
2. In practice, the incident-beam count rate cannot be measured at the same time as the "sample-in" count rate, which leads to an error in τ if the incident intensity changes between the two measurements. The beam intensity oscillates by 1–2% over the course of many hours, and this will produce similar errors in τ if not properly accounted for. However Eq. (3) shows that in the concentrated phase, with $Nt\sigma$ ~2, the resulting error $d(N_{3x})$ is only ~$\frac{1}{2}$–1%. For the most dilute solution, on the other hand, the error is ~5–10% and is clearly significant. Although we remeasured the incident intensity every 2–4 hours, this source of error was not completely eliminated. The data in the dilute phase thus tend to scatter, due to this effect, ~3% about a mean line, rather than the ~1.5% we would have expected from the raw counting statistics.

IV. DESCRIPTION OF THE EXPERIMENT

The sample cell and beam geometry are indicated schematically in Fig. 1. The upper section of the cell is in the form of a copper picture frame with brass faces which provides a reservoir for the He^3–He^4 solution, 1-in. high by 0.700-in. wide by 0.016-in. thick. The lower section of the cell

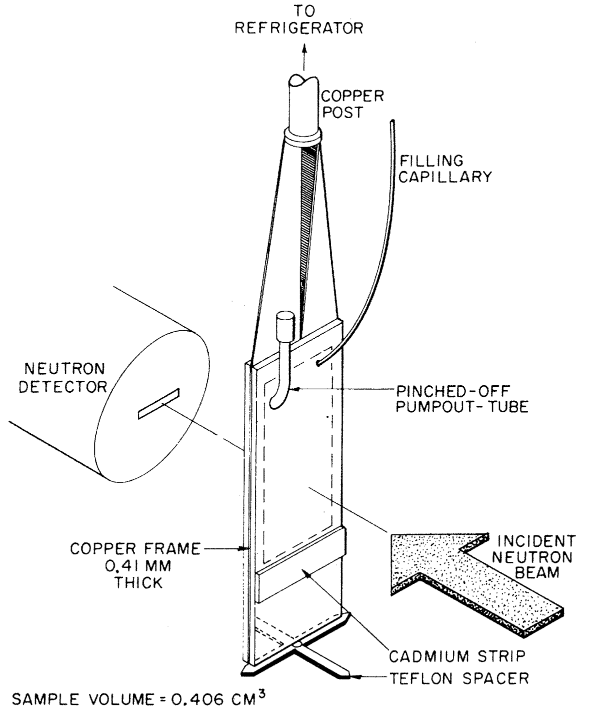

FIG. 1. Sample cell and beam geometry.

contains only the same brass faces as the upper half, with no sample reservoir. The incident beam intensity, I_0 in (1), is measured through this lower section. This is a convenient, standard technique which automatically accounts for most of the empty-cell transmission τ_0, and the statistical accuracy with which τ_0 is known becomes relatively unimportant. The two sections of the cell are divided by a cadmium strip, which is a very strong neutron absorber. This strip enables us to locate the cell precisely in the neutron beam. In addition, the apparent profile of this strip gives a measure of the uniformity and spatial resolution of the beam.

For various reasons it is considerably easier to scan the sample past the beam, rather than vice versa. The cryostat is designed so that the inner vacuum space, containing the sample, paramagnetic salts, He^3 refrigerator, and associated pumping lines, may be translated vertically with respect to the main, 1°K He^4 bath. The sample position is read to 0.001 in. by a dial indicator bearing on a flange which moves with this assembly. If the sample or dial indicator are removed from the cryostat, the Cd strip in Fig. 1 enables us to relocate the sample to an estimated 0.005 in. We have determined on a number of occasions that the vertical motion of the sample causes no detectable thermal effects even after adiabatic demagnetization.

Measurements above 0.45°K were taken with the sample cell attached directly to the He^3 refrigerator with a length of $\frac{1}{2}$-in. o.d. copper tubing. For lower temperatures, the sample was soldered to copper wires projecting from a 250 g, ferric ammonium alum refrigerating salt. This salt was connected by means of a Pb heat switch to a guard salt, made up of 350 g of ferric alum, which in turn was connected to the He^3 refrigerator by means of a second heat switch.

Since a part of the sample is superfluid, the filling capillary must be adequately thermally grounded to the various thermal sinks, with adequate isolation lengths between them. We have used 0.004-in. i.d., stainless-steel capillary in the following lengths:

i. 45 cm thermal ground at 1°K,

ii. 200 cm isolation between 1°K and He^3 refrigerator,

iii. ~40 cm thermal ground on each salt,

iv. ~15 cm between salts and from lower salt to sample.

The thermal ground at the He^3 refrigerator was formed of 50 cm of 0.015-in. i.d. copper capillary, filled with 0.0125-in. nichrome wire to reduce its volume. The upper (1°K) end of the 0.004-in. capillary was soldered into a 75-cm length of 0.014-in. stainless-steel capillary which in turn was joined to a 1/8-in. cupronickel tube at a point somewhere between 4 and 77°K. All the capillary coils were wound, as well as possible, so as to slope continuously downward.

This arrangement appeared to be adequate. Even with the 0.004-in. capillary filled with superfluid up to 1°K, we noticed no increase in the salt warmup rate.

Before the system was sealed, the capillary and cell were purged with dry helium gas for several hours and then pumped for several more hours, usually overnight. The pumping tube was then crimped off, and the system closed up and cooled to 4°K. We introduced the solutions in a variety of ways: either mixing the gases at room temperature or condensing them sequentially, and either at 1°K or while cooling from 4 to 1°K. In all cases, the solution concentrations, as monitored by the neutron transmission, reached steady values within several minutes, although the final concentration depended very slightly upon the sequence followed. As already pointed out, the details of this procedure will not affect the determination of the phase separation line in a "two-phase" technique. On cooling the mixtures below 1° we often observed apparent shifts in concentration, which we attributed to the various effects discussed in Sec. II.

Figure 2 shows a set of typical scans of the sample cell, plotted as neutron counts per minute against vertical cell position. Position "zero" is with the beam passing roughly through the center of the lower, or "open beam," part of the cell. Regions which are covered with Cd, or which are relatively rich in He^3, are seen as sections of low counting rate.

Curve 2a shows the empty-cell scan, equivalent to the measurement of τ_0 in Eq. (2). The Cd strip is seen as a region in which the count drops to the background level, with sharply defined edges of apparent height equal to the beam height (0.100 in.). This indicates that we have succeeded in spatially defining our beam very well.

Curve 2b shows the cell filled with a 54.7% mix-

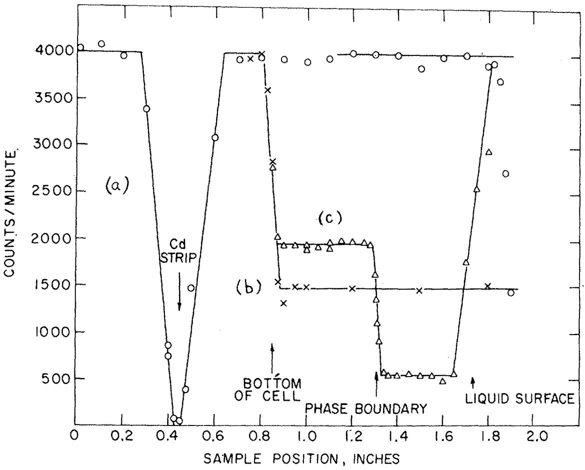

FIG. 2. Typical scans of sample cell. Neutron count rate versus vertical distance along sample in inches: (a) 0 – empty cell; (b) x – 54.7% He^3 solution at 1°K; (c) Δ – 54.7% He^3 solution at 0.65°K, which is below the phase separation temperature.

ture at 1°K. The transmission of the liquid appears uniform as a function of position because the cell thickness was quite uniform. In practice, the cell thickness was measured at several convenient positions with pure He^3 before the mixture was introduced. The scan of curve 2b then served to check against possible distortion of the cell in this operation.

Finally the cell was cooled below the phase separation temperature, resulting in a scan similar to curve 2c. The two phases, phase boundary and, in this case, the liquid surface, are clearly discernible. Transmission measurements in each phase were made at positions lying safely within the boundaries of the phase, whose thickness had previously been measured.

Although curve 2c shows the liquid surface in the sample cell, and thus represents a measurement at saturated vapor pressure, we did not ordinarily run in this condition. Usually the filling capillary was at least partly full of liquid, sometimes up to the 1°K bath level. This latter would correspond to a measurement at a constant pressure of 8 mm. Within our experimental precision we could detect no difference between data taken at these different pressures, and will make no distinction between them. It was necessary to ensure that the pressure did not rise significantly above 8 mm, however, since the resultant density increase in the liquid and mechanical distortion of the cell was very noticeable. For this reason the pressure in the sample line was monitored with a bourdon gauge mounted at the top of the cryostat.

The points above a temperature of 0.45°K were taken chiefly with the sample cell connected directly to the He^3 refrigerator. The temperature was measured by means of a He^3 vapor pressure bulb attached to the refrigerator. In addition a Ge resistance thermometer, previously calibrated against the He^3 vapor pressure, was attached to the sample. It indicated that the sample attained the same temperature as the refrigerator. The reading of this resistor was recorded along with the neutron data, and served to monitor the constancy of the sample temperature. After the temperature of the refrigerator was changed, data were not taken until the resistance thermometer indicated essentially constant sample temperature. Points after either cooling or warming the sample to a given temperature agreed, and are not distinguished in the final data presentation.

Several points above 0.45°K and all points below this temperature were obtained by adiabatic demagnetization. Data were taken as the system warmed up continuously after demagnetization at a rate of $1-2\times10^{-3}$ °K/hour. Measurements were taken sequentially on both phases, interspersed with suitable "open-beam" and background counts. The magnetic temperature of the refrigerating salt was measured and converted to thermodynamic temperature using a $T-T^*$ relation established by studying the nuclear polarization of Re^{185} in Fe-Re alloy, as previously described.[12]

We attempted to prove that the liquid in the cell actually cooled to the salt temperature by performing an auxiliary experiment. A new cell was constructed, essentially identical to that shown in Fig. 1 except that the sample region was deep enough to contain an Fe-Re slab, 0.040-in. thick, and still retain a space approximately 0.016-in. thick for the liquid mixture. The Fe-Re was mounted on Teflon supports so that the heat transfer to the sample case, and hence to the salt, would be basically through the liquid. The temperature of the Fe-Re as a function of time after demagnetization was measured by monitoring the nuclear polarization of the Re^{185} with 2.16-eV neutrons. The results are shown in Fig. 3. With no liquid in the cell (curve 3a) the sample had only reached 0.16°K 10 hours after demagnetization, although the salt was still below 0.11°K at this time. With liquid in the cell (curve 3b), there was an apparent temperature decrease during the first 30–45 min, after which the sample temperature began to increase as the salt warmed at approximately 3×10^{-3} °K/hour. Curve 3c shows the salt temperature corresponding to 3b. The salt temperatures for 3a were 0.015°K higher, and are omitted in Fig. 3 for clarity.

For temperatures greater than 0.08°K, the Fe-Re temperature and salt temperature were identical. At lower temperatures we detected a gradient of several millidegrees between the two. Since all this indicates is that the liquid mixture temperature is intermediate between the two thermometers, there is an uncertainty in the mixture temperature. However, in this temperature range, the concentration changes very slowly with temperature, so that the uncertainty has little effect upon the final results. It is also probable that the temperature gradient is much less in the actual measurement than in this auxiliary one. In the Fe-Re experiment it was necessary to apply a magnetic field to the sample. This field, which was not present in the original phase diagram determination, caused eddy-current heating in the Fe-Re slab of about 1 erg/sec, as judged from the increased

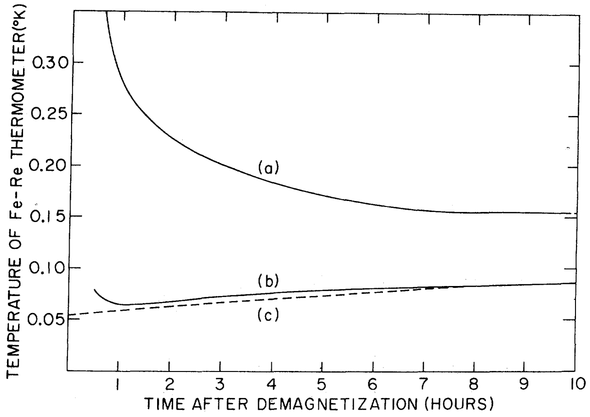

FIG. 3. Temperature of Fe-Re sample versus time after adiabatic demagnetization. (a) No liquid in cell, heat transfer only through Teflon supports; (b) He^3-He^4 solution ($x \sim 0.3$) in cell, serving as an added heat-transfer medium; (c) temperature of refrigerating salt corresponding to curve (b). The temperature of the refrigerating salt corresponding to curve (a) parallels (c) but is displaced upwards from it by 0.015°K.

warmup rate of the salt when the field was on. Of course this heat must be transferred across the boundary from Fe-Re to the liquid and finally into the brass cell wall. If we assume a boundary resistance of the same order of magnitude as that given by Vilches and Wheatley[13] for a copper-He interface, we predict a temperature gradient of several millidegrees between Fe-Re and salt below 0.07°K, in agreement with the observations.

There is therefore good reason for believing that with no field present the liquid temperature is equal to the salt temperature down to the lowest temperatures reached, i.e., ~0.04°K.

V. DATA ANALYSIS

We performed the pure He^3 calibration several times under various conditions of temperature and pressure. The molar volume, needed in Eq. (2), was taken from the results of Kerr and Taylor,[14] combined with the thermal expansion and compressibility results of Boghosian *et al.*[15] interpolated to match our experimental conditions. These calibrations tended to scatter slightly more than would have been expected from statistics, and if they were to be used in the final analysis would lead to an uncertainty of ±1% in the quoted number densities. This scatter, in all likelihood, is a result of systematic errors involved in correcting all runs to the same temperature and pressure.

Using this calibration, for the moment, we find that the upper phase at low temperatures, which is expected[16, 17] to be essentially pure He^3, has too high a number density. It remains high, by an average of 1%, even after accounting for the known thermal contraction of pure He^3 below 1°K. Since this 1% shift is, however, within the accuracy of our calibration, we may say that the experiment supports the conclusion that the upper phase is pure He^3 at low temperatures. We then argue backwards, and use the average of the upper-phase data below 0.1°K as our pure He^3 calibration data. The statistical error introduced in this procedure is very small.

In Table I we present smoothed values of X/v_X for both phases, interpolated to even increments of temperature.

In order to plot our data on the usual scale of temperature versus concentration we must next convert our number density, or X/v_X, results to X. The only set of density data on mixtures convering the entire range $0 \leq X \leq 1$ is that of Kerr,[18] at 1.2°K. There are two problems associated with using these results. First, the pure He^3 volume is systematically shifted by 0.4% from later data,[14] which we have already used in the above calibration procedure. Most important, we have no way of correcting for thermal expansion of the mixtures over most of the concentration range at low temperatures. The only applicable data are those of Ifft *et al.*,[7] which shows that for $X < 0.16$ the volume change between 1.25°K and the phase separation line is less than $\frac{1}{2}$%. This correction may be neglected, since for the low He^3 concentrations the experimental precision is not that good.

The region of the peak ($0.50 < X < 0.75$) has been investigated by Graf *et al.*[6] and Kerr.[19] The former authors saw only small changes in molar volume on cooling their mixtures (all prepared with $0.6 < X < 0.7$) from 1.1° to the phase boundary, as might be expected from the relatively small change in temperature involved in this concentration region. They give no quantitative results for the thermal expansion, and apparently make no correction for it. From Kerr's results on $X = 0.533$

TABLE I. Smoothed values of (X/v) for the equilibrium phases along the phase separation line, interpolated to equal temperature increments. The lowest-temperature, concentrated phase data have been used in normalizing these results and thus do not appear in this table.

T (°K)	10^2 (X/v) (cm^{-3}) Dilute phase	 Concentrated phase
0.05	0.2485	...
0.10	0.2686	...
0.15	0.2996	...
0.20	0.3417	...
0.25	0.3938	...
0.30	0.4532	2.720
0.35	0.5200	2.685
0.40	0.5878	2.645
0.45	0.6653	2.600
0.50	0.7458	2.549
0.55	0.8540	2.493
0.60	0.9797	2.432
0.65	1.123	2.366
0.70	1.286	2.294
0.75	1.465	2.217
0.80	1.668	2.134
0.85	1.884	...

and $X = 0.745$, it appears that the correction will again be ~$\frac{1}{2}$% or less, and thus not important on our scale. In any event we have very few data points in this region.

For $X > 0.75$, the phase separation temperature again decreases, so that the molar volume change can become significant. Further, an apparent 1% shift in X, due to a 1% shift in v_x, is most obvious at large X. For lack of anything better, we have assumed that the molar volume change of mixtures with $X > 0.75$ is given simply by

$$v(X, T_1) - v(X, T_2) = X[v(1, T_1) - v(1, T_2)]. \quad (4)$$

This is equivalent to assuming that the expansion coefficient of the mixture is simply an average of that for the He^3 and He^4 components separately, and then ignoring the small expansion of the He^4. While Eq. (4) should be quite accurate for $X \rightarrow 1$, it is an overestimate by a factor of 3 for $X = 0.745$.[19]

Figure 4 shows the phase separation curve resulting from the above corrections. We have not smoothed our data points, so that the scatter indicates the precision of the measurement. For the sake of clarity, however, we have deleted over one-half of the data points for $X < 0.2$ and $X > 0.95$.

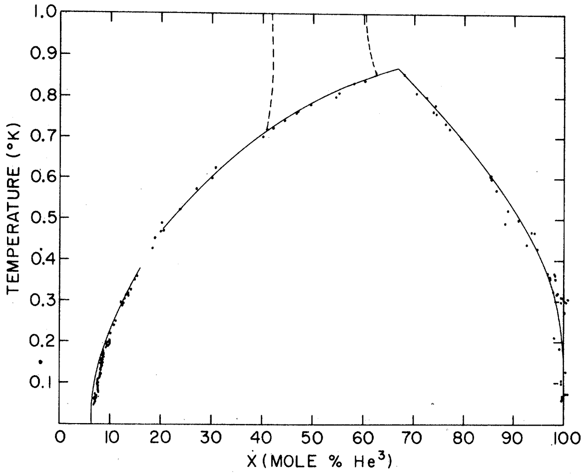

FIG. 4. Phase separation curve of He^3-He^4 solutions. Solid points are from neutron transmission. Vertical dotted lines show paths followed in one-phase region by systems of nominal starting concentration 0.420 and 0.605. Solid line for $x < 0.16$, data from Ref. 7. Solid line for $0.20 < x < 0.90$, data from Ref. 2 (corrected to T_{62}) and Ref. 6. Solid line for $x > 0.90$, theory of Ref. 16.

VI. COMPARISON WITH OTHER WORK AND WITH THEORY

The measurements prior to 1964 have been reviewed and compared by Taconis and De Bruyn Ouboter.[20] In general, the sound velocity[2] and specific heat[4] measurements agree very well with each other, while the visual observations[1] tend to lie somewhat off the consensus curve. This early data was based on the 1957 E scale,[21] which gives temperatures 7.5–8.5 mdeg higher than[22] T_{62} in the range $0.9 > T > 0.5$.

After correction to T_{62}, we find that these earlier data are in extremely good agreement with ours, as can be seen on Fig. 4 in which the smoothed data of Roberts and Sydoriak[2] are given by a solid line. The fractional deviation, $\Delta X/X$, averages less than 1% over the entire concentration range $0.20 < X < 0.95$ in which the measurements overlap. In the vicinity of the consolute point, we are in agreement within $\frac{1}{2}$% with the high-precision results of Graf *et al.*[6]

For concentrations $X \leq 0.20$, however, the agreement with other work is not as good. Although it is difficult to judge the phase separation temperature with any precision from the published heat-capacity curves,[8] our data appear to agree fairly well at $X = 0.15$, 0.12, and 0.10, but not at $X = 0.08$ where we get a considerably lower temperature.

These heat capacity data are presumably superseded by the density data of Ref. 7, which in general agree with them. These results are presented for comparison on Fig. 4. For $X < 0.10$, at a given temperature, our data give a concentration ~ 7% higher than this work.

It is difficult to account for such a discrepancy. It cannot be thermometry differences; the necessary differences are much too large to be plausible, i.e., 0.050°K at 0.1°K and 0.02°K at 0.2°K. The density data are extremely precise, but their real accuracy may in fact be determined by the systematic errors discussed in Sec. II. As was mentioned, variations in our incident beam intensity with time led to the undulations in the data points visible in Fig. 4. This effect could be minimized by repeating the measurement with a thicker target. But even if we take the observed scatter, rather than the counting statistics, as a precision index, the discrepancy with the density data is well outside expectations. However, it does not seem to be enough to seriously affect any thermodynamic quantities derived from the data.

For $X > 0.75$, Edwards and Daunt[16] have developed a semi-empirical formula for the separation curve based on the earlier measurements[2,4] and the Zharkov-Silin[23] model for He^4 impurities in He^3. They find the following formula for the He^4 concentration in the upper He^3-rich phase in the low-temperature limit

$$(1 - X_u) = 1.13T^{3/2}e^{-0.71/T}. \quad (5)$$

A more exact expression, indistinguishable from (5) for $T < 0.3$°K, fits the older data very well, and therefore also fits our data. In principle, we could fit our measurements for $T < 0.4$°K to obtain new values for the constants in (5), which involve the effective mass and binding energy of a He^4 atom in He^3. In practice, the small uncertainty (~ $\frac{1}{2}$%) in X_u due to counting statistics, calibration precision, and uncertainty in the thermal expansion correction, produces a very large uncertainty in $(1 - X_u)$. This in turn leads to such large uncertainties in the constants in (5) that it is not worth pursuing. We simply plot (5) as it stands on Fig. 4 where it is seen to agree well enough with our data.

It is clear that in order to improve the constants in (5) it is necessary to measure the He^4 concentration directly. The present technique, which is sensitive to the presence of He^3 rather than He^4, is eminently unsuitable for this.

To derive the equation of the phase separation curve at low concentrations, the He^3 in each phase is considered as a Fermi gas well below its respective degeneracy temperature. The equation of the solubility curve for He^3 in He^4 at low temperatures may be shown to be[7] $X = X_0 (1 + \beta T^2)$. Upon fitting our data for $T < 0.15\,°K$ we find

$$X_0 = (6.84 \pm 0.06)\% \text{ and } \beta = (11.1 \pm 0.7)(°K)^{-2} .$$

The value of X_0 is shifted significantly from the value $(6.37 \pm 0.05)\%$ reported by Ifft *et al.*[7]; the shift is 7%, as noted above. The β value agrees very well with that reported by Ifft, i.e., 10.8 $(°K)^{-2}$.

VII. CONCLUSIONS

We have shown that neutron transmission can yield an accurate determination of the He^3 -He phase diagram. For He^3 concentrations > 0.20, the results are in excellent agreement with those obtained by other techniques. Since the systematic errors tend to be very different for the various techniques, this agreement is heartening. It indicates that, at least in this range of concentration and temperature ($T > 0.35\,°K$), it is possible to minimize many of the experimental pitfalls.

At low concentrations, and correspondingly low separation temperatures, there appears to be a discrepancy of ~ 7% between our measurements and those of Ifft *et al.*[7] This may indicate some systematic error in either, or both, of the measurements in this region.

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