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THE PHASE SEPARATION CURVE AND MOLAR DENSITY
OF DILUTE $\text{He}^3$ - $\text{He}^4$ MIXTURES

DISSERTATION

Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in the Graduate
School of The Ohio State University

By

Edward Milton Ifft, B.S.

* * * * * * *

The Ohio State University

1967

Approved by

[Signature]
Adviser
Department of Physics
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VITA

July 19, 1937 . . Born - Grove City, Pennsylvania

1955 . . . . . Graduated, Butler High School, Butler, Pennsylvania

1955-1960 . . . General Motors Scholar, Antioch College, Yellow Springs, Ohio

1960 . . . . . B.S. (Physics), Antioch College, Yellow Springs, Ohio

1960-1963 . . . National Science Foundation Cooperative Fellow, Physics Department, Ohio State University, Columbus, Ohio

1963-1964 . . . Research Assistant, Physics Department, Ohio State University, Columbus, Ohio

1964-1965 . . . Participant, Annual Exchange of Graduate Students and Professors between the USA and USSR (assigned to Moscow State University and the Institute of Physical Problems, Moscow)

1965-1967 . . . Research Associate, Physics Department, Ohio State University, Columbus, Ohio

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CHAPTER I

INTRODUCTION

In recent years, an increasing amount of research, both experimental and theoretical, has been carried out on liquid mixtures of He\(^3\) and He\(^4\). In some respects, these mixtures are proving even more interesting than the pure helium isotopes themselves. Mixtures offer the possibility of studying a simple quantum liquid in which the temperature and density may be varied independently, and in which the two components vary in mass, magnetic properties and statistics. Unfortunately, these mixtures yield readily to theoretical analysis only when the concentration of one of the components is small.

1.1 Dilute Solutions of He\(^4\) in He\(^3\)

Zharkov and Silin (1) were the first to treat dilute solutions of He\(^4\) dissolved in He\(^3\). They proposed that the change in the system energy caused by placing a He\(^4\) atom into pure He\(^3\) be given by

\[ \epsilon_4 = -E_4 + \frac{p^2}{2m^*_4} + \ldots \ p^4 \ldots \]  \hspace{1cm} (I-1)

where \(E_4\) is the energy needed to place a He\(^4\) atom at rest into the He\(^3\), \(p\) is its momentum and \(m^*_4\) is an effective mass. De Bruyn Ouboter et al. (2) used
specific heat measurements to obtain a value of 53.6 joules/mole for $E_4$.

Edwards and Daunt (3) estimated $m_4^*$ to be $5.3m_4$. Both $E_4$ and $m_4^*$ were assumed to be independent of concentration and temperature. The solvent energy spectrum was assumed to be the Fermi quasi-particle spectrum of pure He$^3$.

If the He$^4$ concentration, $1 - X$, is so small the Maxwell-Boltzmann statistics apply, it follows from Eq. (I-1) that the He$^4$ chemical potential, or Gibbs function per molecule, is given by

$$\mu_4 = -NE_4 + RT \ln \left[ \frac{N(1 - X)h^3}{v(k_BT)^{3/2}(2\pi m_4^*)^{3/2}} \right]$$

where $N$ is Avogadro's number, $v$ is the molar volume and $k_B$ is Boltzmann's constant.

1.2 Dilute Solutions of He$^3$ in He$^4$

The case that was actually treated first historically was a dilute solution of He$^3$ dissolved in He$^4$. Landau and Pomeranchuk (4, 5) assumed that the He$^3$ could be viewed as a small impurity which modifies the phonon-roton excitation spectrum of superfluid He$^4$ by adding a branch of Fermi quasi-particles. These quasi-particles are equal in number to the number of He$^3$ atoms and have an energy given by

$$\epsilon_3 = -E_3 + \frac{p^2}{2m_3^*} + \ldots + p^4 \ldots$$

where $E_3(X)$ is known from vapor pressure measurements (2) to be nearly equal to the latent heat of
vaporization per atom of pure liquid He\textsuperscript{3} at 0°K. m* has been found to be approximately 2.34m\textsubscript{3}(1 + X) \textsuperscript{6}. [It should be noted that a value of 2.5m\textsubscript{3} has also been reported (7,8) and the question is not fully resolved.]

The theory predicted that, when the He\textsuperscript{4} solvent was below the \( \lambda \)-point, the He\textsuperscript{3} would move with the normal component only. Landau and Pomeranchuk (4) pointed out that this was not due to the fact that He\textsuperscript{3} obeys Fermi-Dirac statistics, but that it would be true of any small impurity dissolved in the He\textsuperscript{4}.

The Landau-Pomeranchuk theory, while a highly successful one, was limited to solutions in which X, the He\textsuperscript{3} concentration, was so small that He\textsuperscript{3} - He\textsuperscript{3} interactions could be neglected. It also did not predict any of the transport properties of the solutions. Recent theoretical work has centered on developing a more realistic approach to these solutions. In this connection, one should mention the contributions of Bardeen, Baym and Pines (9,10), Ebner (11,12), Emery (13,14) and Campbell (15).

A. The microscopic theory of Bardeen, Baym and Pines

Bardeen, Baym and Pines (BBP) have developed a theoretical view of solutions with small X which takes into account the weakly attractive potential between He\textsuperscript{3} atoms. Their theory, which reduces to the Landau-Pomeranchuk theory in the limit of weak interactions, does not attempt to calculate this effective interaction from first principles. Rather, it is assumed at the outset that the potential is independent of the spin, velocity and concentration of the He\textsuperscript{3} particles and depends only upon the distance between the He\textsuperscript{3} particles.
A further crucial assumption is made that the potential is weak. The theory confines itself to the Fermi liquid regime of the He$^3$ system.

Because the problem to be solved was essentially a scattering problem, it was convenient to use the Fourier transform of the potential $V(r)$:

$$V(k) = \int e^{-i\mathbf{k} \cdot \mathbf{r}} V(r) d^3r = V_0 \cos \left( \frac{\beta k}{\hbar} \right)$$  \hspace{1cm} (I-4)

where $k$ is the momentum transfer in the scattering process and $V_0$ and $\beta$ are parameters to be fitted to experiment. A two-parameter form was chosen for the potential because spin diffusion data were available for concentrations of 1.32% and 5.0%. It was also known that $V(k)$ was negative at $k = 0$ from the phase separation curve measurements of Edwards (see Section 1.2C).

Fitting their potential to the data, BBP found that

$$V_0 = -0.0754 \frac{m_4 s^2}{n_4} \quad \text{and} \quad \beta = 3.16 \Lambda$$  \hspace{1cm} (I-5)

where $m_4$ is the mass of a He$^4$ atom and $s$ and $n_4$ are the first sound velocity and number density of pure He$^4$ at 0°K.

Some brief mention should be made of the physical nature of the He$^3$ - He$^3$ interaction. BBP visualize the interaction as made up of two large contributions, nearly equal in magnitude, but opposite in sign. The direct interaction between the He$^3$ particles is calculated at constant He$^4$ density. The calculation considers the change in chemical potential which results when two He$^4$ atoms are replaced by two He$^3$ atoms of opposite spin. A He$^3$ atom occupies a volume in the liquid $(1 + \alpha)$ times that occupied by the displaced
He\(^4\) atom. \(\alpha\) was determined by Kerr (16) to be about 0.28 at 1.2\(^\circ\)K and relatively independent of temperature below the \(\lambda\)-point. Since the force fields of the He\(^3\) and He\(^4\) atoms are identical, it is this volume difference, due to the larger He\(^3\) zero-point energy, which plays the major role in the interaction. Using the fact that the energy needed to replace a He\(^4\) atom with a He\(^3\) with spin up in the presence of a He\(^4\) is the same as that needed to replace a He\(^4\) with a He\(^3\) with spin down in the presence of a He\(^3\) with spin down, BBP found that the direct interaction at long wavelengths is repulsive and given by:

\[
V_0^{\text{dir}} = (1 + 2\alpha) \frac{m_4 s^2}{n_4}.
\]  

(I-6)

The second major contribution to the interaction arises from the fact that, as the He\(^3\) atom moves through the He\(^4\) background, it excites density fluctuations in the He\(^4\) due to the disparity in He\(^3\) and He\(^4\) volumes. At low temperatures, these density fluctuations may be viewed as long-wavelength virtual He\(^4\) phonons, which are being exchanged between He\(^3\) particles. As in the case of phonon exchange between electrons in metals, the interaction resulting from this exchange is attractive, and in the long wavelength limit is given by:

\[
V_0^{\text{ph}} = -(1 + \alpha)^2 \frac{m_4 s^2}{n_4}.
\]  

(I-7)

A further velocity-dependent term resulting from the overlap between the phonon screening clouds surrounding the two He\(^3\) atoms is shown by BBP to be very small compared to \(V_0^{\text{ph}}\).
Adding the two contributions to the effective interaction [Eqs. (1-6) and (1-7)] gives:

\[ V_0 = -\alpha^2 \frac{m_4 s^2}{n_4} \]  \hspace{1cm} (I-8)

Using \( \alpha = 0.28 \),

\[ V_0 = -0.078 \frac{m_4 s^2}{n_4} \]  \hspace{1cm} (I-9)

in excellent agreement with Eq. (1-5).

In the next section, we outline how a knowledge of the effective interaction leads to predictions of the physical properties of the dilute solution.

B. Ebner’s extension of the BBP theory

The original BBP theory was valid for concentrations up to about 5% and temperatures below \( T_F \). In order to extend the range of applicability of the theory, Ebner (11,12) undertook an extension of the BBP potential using spin diffusion measurements on a 5% solution between 0.5°K and 1.0°K (6). In this region, \( \text{He}^3 - \text{He}^3 \) scattering is the limiting mechanism. In addition, Maxwell-Boltzmann statistics are valid to within a few per cent for the \( \text{He}^3 \) system and the mean free paths of the quasi-particles are long enough that Born approximation may be used. Ebner’s five-parameter \( V(k) \), which will not be quoted here, has \( V(0) = 1.08V_0 \) at \( k = 0 \). Using Hartree-Fock techniques, Ebner obtained this expression for the energy acquired by the quasi-particle through its interactions with other \( \text{He}^3 \) quasi-particles:

\[ E^I_0(X, T) = n_3 V(0) - \frac{1}{(2\pi\hbar)^3} \int n_3 q^* V(k) d^3q \]  \hspace{1cm} (I-10)
where $\sigma$ is the spin index, $n_3$ is the He$^3$ number density, $n_{\Omega q}$ is the occupation number of the single particle state of momentum $\vec{q}$ and $\vec{r} = \vec{p} - \vec{q}$ is the momentum transfer. When the system is in thermal equilibrium,

$$n_{\Omega q} = \frac{1}{\exp[\beta(q^2/2m^* + E_0 - \mu_3)] + 1}$$  \hspace{1cm} (I-11)

Recalling that the total number of quasi-particles is just the number of He$^3$ atoms, one may solve for $\mu_3 - E_0$.

Ebner solved for the chemical potential by expanding in powers of $(T/T_F)^2$ at low temperatures and of $(T_F/T)^{3/2}$ at high temperatures.

One may obtain $\mu_4$ from $\mu_3$ by using the Gibbs-Duhem relation:

$$\left( \frac{\partial \mu_4}{\partial x} \right)_{p, T} = -\frac{X}{1 - X} \left( \frac{\partial \mu_3}{\partial x} \right)_{p, T}$$  \hspace{1cm} (I-12)

The thermodynamic properties of the system may then be obtained from the chemical potentials $\mu_3$ and $\mu_4$.

C. **Phase separation**

One of the most interesting aspects of the study of solutions is the phenomenon of phase separation. If the second derivative of the Gibbs function with respect to the concentration of one of the components becomes negative, a binary mixture becomes unstable and separates into two phases. This was observed to occur in He$^3$ - He$^4$ mixtures by Walters and Fairbank in 1956 using NMR techniques (17). They showed that a He$^3$-rich phase forms and floats above a heavier He$^4$-rich phase. Subsequently, a variety of experiments
were performed by Zinov'eva and Peshkov (18,19), de Bruyn Ouboter, Taconis, le Pair and Beenakker (20) and Roberts and Sydoriak (21) to determine the shape of the phase separation curve down to about 0.4°K. The theory predicted, and an experiment by Brewer and Keyston (22) confirmed, that the upper phase is almost pure He\textsuperscript{3} for T ≤ 0.2°K. In 1961, Edwards and Daunt (3) pointed out that, at the other end of the curve, dilute solutions of He\textsuperscript{3} in He\textsuperscript{4} might be stable even at 0°K. Specific heat measurements of the lower phase by Edwards, Brewer, Seligmann, Skertic and Yaqub (23) strongly supported this prediction. They succeeded in mapping the separation curve down to 0.16°K and, extrapolating their data, showed that the limiting solubility of He\textsuperscript{3} in He\textsuperscript{4} at 0°K is about 6%. Their results were confirmed in part by Anderson et al. (24) who showed that a 5% solution does not phase separate down to 0.01°K.

It was desirable to determine the phase separation curve with greater precision and at still lower temperatures, both because of its use in the theory of dilute solutions and because of its importance in the operation of dilution refrigerators (25). The development of very sensitive capacitance bridges made possible a new method of determining X in the lower phase. A tiny capacitor was placed in the solution and the liquid allowed to serve as the dielectric between the plates. The dielectric constant of the solution is a function of the number density, so that a capacitance measurement is a determination of the number density. The number density, of course, is a function of the He\textsuperscript{3} concentration X. This method has an intrinsic advantage over the specific heat method. In a given specific heat experiment, only one
point on the phase separation curve is determined (\(C_P\) has a discontinuity when the curve is crossed). However, the capacitance method, in principle, allows one to trace out a large section of the curve in a single experiment. As a bonus, several other quantities of interest are obtainable from capacitance measurements. As well as \(X_0\), the limiting solubility at \(0^\circ\)K, one can determine the BBP parameter \(\alpha(X,T)\) and the derivative of the \(\text{He}^3\) effective mass \(m^*\) with respect to pressure. This latter quantity comes from the thermal expansion of the solutions in the single phase region.

This dissertation reports on capacitance measurements made on seven different dilute solutions of \(\text{He}^3\) in \(\text{He}^4\) in the temperature range 1.25\(^\circ\)K to 0.024\(^\circ\)K.
2.1 General Considerations in Design

In order to carry out very precise dielectric measurements of $\text{He}^3 - \text{He}^4$ mixtures down to the temperature range $0.02^\circ K - 0.03^\circ K$, certain general requirements had to be met by the cryostat. A fairly large chromium potassium alum "pill" had to be constructed to provide magnetic cooling and it had to be in good thermal contact with the liquid mixture, even at these low temperatures. A $\text{He}^3$ refrigerator was required to cool the CrKAlum before demagnetization and to thermally shield it after demagnetization. Provisions had to be made to effectively shield the leads to the capacitor from each other and from stray fields. The apparatus had to have a very small heat leak into the cell, both to permit it to reach the desired temperature and to insure that the warm-up rate was small enough to permit measurements at virtually constant temperature. Finally, care had to be exercised to insure that no magnetic materials interfered with the magnetic thermometry.

There was available in the laboratory a general purpose cryostat which was capable of being modified to meet the above requirements. The
pumping tubes, thermal shields and He\textsuperscript{3} refrigeration system have been described in detail previously (26).

A general view of the cold part of the cryostat is presented in Figure 1.

2.2 He\textsuperscript{4} Bath and Nitrogen Bath

The temperature of the cryostat can be lowered to about 1.23°K by pumping away the vapor of the liquid He\textsuperscript{4} bath which surrounds it. The He\textsuperscript{4} bath, in turn, is surrounded by a liquid nitrogen bath at 77°K. The He\textsuperscript{4} vapor is pumped away and into a recovery system by a Stokes pump with a displacement of 80 CFM. If the bath is pumped at full speed, a charge of liquid helium will last eight or nine hours. A pair of fiberglass wicks wound around the bath filling tube coil and the He\textsuperscript{3} pumping tube hung down about 15" in the bath space. These wicks caused helium to be drawn up out of the bath to these warmer metal parts where it evaporated. The resultant cooling permitted experiments to continue for a time even after the level of the bath had become quite low.

2.3 Outer and Inner Vacuum Systems

The outer vacuum space isolates the He\textsuperscript{3} refrigerator from the He\textsuperscript{4} bath. The jacket was constructed of 1-1/2" o.d., 347 stainless steel with a 0.01" wall. The concave bottom of the jacket was of the same material and was silver soldered to the wall. The top of the jacket was soft soldered (60/40 solder) to a cylindrical copper sleeve. The sleeve could be attached to the outer vacuum flange with Wood's metal.
Figure 1

Demagnetization Apparatus

Reich seal ................................................ A
Filling tube coil .......................................... B
Outer vacuum jacket .................................. C
Epibond seal through inner flange ................ D
Copper collar on inner flange ....................... E
Copper rods leading to Pb switch .................. F
Pb switch .................................................. G
He\textsuperscript{3} reservoir .............................. H
He\textsuperscript{3} reservoir carbon thermometer .... I
Inner vacuum jacket ..................................... J
Teflon ring ............................................... K
Copper ring connecting coil foil and Pb switch . L
Coil foil ...................................................... M
Nylon spacer ............................................. N
Upper CrKAlum pill .................................... O
Upper carbon thermometer ......................... P
Heater ...................................................... Q
Pb switch .................................................. G
Lower CrKAlum pill .................................... R
CrKAlum mutual inductance coils .................. T
Lower carbon thermometer ......................... U
Press-fit seal for capacitor lead .................... V
Nulling coil for CMN mutual inductance coils .... W
Sample cell ............................................... X
CMN mutual inductance coils ....................... Y
Nylon spacer screwed into bottom of cell ........ Z
The inner vacuum space serves to isolate the Cr pills and the cell from the He³ refrigerator. The jacket was constructed of 1-1/4" o.d., 18% nickel-silver with a 0.013" wall. A concave piece of the same material formed the bottom and a copper lip formed the top. Both pieces were hard soldered to the wall. Thin copper strips 0.01" x 0.04" were glued with GE 7031 varnish to the can and baked at 180°F for 2-1/2 hours. These strips were also soft soldered (60/40 solder) to the jacket on top of the hard solder joint at the top. The thin copper strips were in this form to reduce eddy current heating. A minimum amount of GE varnish was used because it is slightly magnetic. The entire inner jacket could be attached to the inner vacuum flange with Wood's metal.

The inner and outer vacuum pumping lines ran to separate CVC Type PMC 115 oil diffusion pumps, which were backed by a common rotary oil-sealed mechanical pump.

2.4 Filling Tube

The filling tube can be a large source of heat into the cell because of the refluxing He film. Care was therefore exercised to provide a long length of tubing to increase the thermal resistance. The tube was well anchored thermally at several points and isolation coils were provided.

In the bath about 5" above the outer flange the tube entered a coil of five turns (74 cm). The coil was constructed of Cu capillary 0.083" o.d., 0.031" i.d. with a Cu wire of diameter 0.022" inserted to reduce dead volume.
Just above the outer flange was situated another coil of five turns (24 cm) made of stainless steel capillary 0.04" o.d. with a 0.004" wall. Below this coil, all the tubing was copper-nickel 0.0156" o.d. with a 0.003" wall. Cu-Ni was found to be preferable to stainless steel, which tended to crack when coiled.

Between the inner and outer flanges, the filling tube had an isolation coil of 21 turns (277 cm). The tube then had a coil of 4-1/2 turns (50 cm) completely soft soldered to a copper collar, which was in turn soft soldered to the inner flange just above the inner vacuum jacket. This served to thermally anchor the tube to the He³ reservoir.

Another isolation coil of 14 turns (97 cm) was located just below the inner flange. Just below the upper CrKAlum pill was a nylon cylinder 7/8" in diameter which screwed into a threaded hole in the nylon support for the pill. Coil foil (27) from the interior of the upper CrKAlum pill went around this nylon cylinder and another filling tube coil of 12 turns (97 cm) was wound over this coil foil. Another piece of coil foil from the pill ran over one side of these 12 turns and a separate piece of coil foil with wires running horizontally was wrapped around the entire bundle. Apiezon N grease was spread liberally over the coils and foil and the whole assembly bound tightly with cotton thread. This served to thermally anchor the filling tube to the upper CrKAlum pill.

The tube ran straight down the side of the lower CrKAlum pill. It was greased and bound to the pill with a large piece of transverse coil foil which completely surrounded the lower pill, the filling tube and the two shielding tubes for the capacitor leads.
Between the lower CrKAlum pill and the cell was a coil of two turns (30 cm). Originally there was a coil of ten turns in this position. However, eight turns were removed because it was suspected that this coil had a low spot which allowed the liquid sample to form a pool. During the demagnetization, when the CrKAlum pills may be colder than the cell, He\textsuperscript{3} can be lost from the cell due to evaporation. If the filling tube has a low spot, the He\textsuperscript{3} may recondense and not all drain back into the cell when temperature equilibrium is reached. In spite of the fact that the filling tube was designed to be sloping downward at every point, there was evidence that this effect was still present. This will be discussed further later.

All the coils, with the exception of the bath coil, the coil at the He\textsuperscript{3} reservoir and the two turns above the cell, were held together with Epibond 100A (28). The coils were tightly wound over a teflon form, sprinkled with Epibond 100A and baked for six hours at 280\textdegree F. When the teflon form was removed, a strong coil with many turns in a small space was obtained.

The approximate length of the filling tube was as follows: from room temperature to outer flange, 169 cm; between outer flange and inner flange, 328 cm; from inner flange to top of cell, 348 cm; total length, 845 cm. These lengths and the number of turns per coil are, of course, somewhat arbitrary. The general principle followed was to maximize the length of the tube, consistent with space limitations and the avoidance of low spots. The filling tube coils are summarized in Table 1.
### TABLE 1

**FILLING TUBE COILS**

<table>
<thead>
<tr>
<th>Location</th>
<th>Purpose</th>
<th>Material</th>
<th>Number of turns</th>
<th>Length (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He(^4) bath</td>
<td>Thermal contact with bath</td>
<td>Copper</td>
<td>5</td>
<td>74</td>
</tr>
<tr>
<td>He(^4) bath</td>
<td>Thermal contact with bath</td>
<td>Stainless steel</td>
<td>5</td>
<td>24</td>
</tr>
<tr>
<td>Between outer and inner flanges</td>
<td>Isolation</td>
<td>Copper-nickel</td>
<td>21</td>
<td>277</td>
</tr>
<tr>
<td>On inner flange</td>
<td>Thermal contact with He(^3) refrigerator</td>
<td>Copper-nickel</td>
<td>4 (\frac{1}{2})</td>
<td>50</td>
</tr>
<tr>
<td>Just below inner flange</td>
<td>Isolation</td>
<td></td>
<td>14</td>
<td>97</td>
</tr>
<tr>
<td>Between CrKAlum pills</td>
<td>Thermal contact with guard pill</td>
<td>Copper-nickel</td>
<td>12</td>
<td>97</td>
</tr>
<tr>
<td>Between lower CrKAlum pill and cell</td>
<td>Isolation</td>
<td>Copper-nickel</td>
<td>2</td>
<td>30</td>
</tr>
</tbody>
</table>

#### 2.5 Carbon Thermometers and Heater

Four carbon thermometers were available in the cryostat. One was soldered to the upper filling tube coil in the He\(^4\) bath. Another was soldered to the He\(^3\) reservoir. The third was soldered to a piece of coil foil which was greased and bound with the foils from the upper CrKAlum pill on the nylon
cylinder below the pill. It thus measured the temperature of the upper pill. The fourth was soldered to a coil foil tab which was greased and bound to the foil leading from the lower pill to the cell. All these thermometers were 220 $\Omega$, 1/2 watt Speer resistors. All the resistors were prepared as follows:

The end of a piece of coil foil was soldered with indium to an ordinary resistor lead. The lead was soldered with indium to the Speer resistor lead and the foil greased and wrapped about the resistor, whose outer insulation was left intact. A layer of mylar was wrapped about the foil. Another identical piece of coil foil was greased, wrapped on top and soldered to the other Speer resistor lead. The entire cylinder was then tightly bound with cotton thread. This technique was developed by Black, Roach and Wheatley (29).

A heater was prepared by wrapping 0.001" enameled Evanohm wire (835 $\Omega$/ft) over a 0.080" copper wire 1-1/4" in length. The Evanohm wire was held down by GE 7031 varnish. The copper wire had a small 0.020" thick copper tab on the end and this was soldered with indium onto the coil foil which led into the interior of the lower pill. Thus the lower pill was between the heater and the cell. The total resistance of the heater was 1000 $\Omega$. The circuits used for the heater and the carbon thermometers have been described earlier (26).

2.6 Magnetic Thermometry

The temperature of the cell was determined by measuring the relative magnetic susceptibility of a small quantity of cerium magnesium nitrate just
above the capacitor. This, as well as a measurement of the temperature of the CrKAlum pills, could be done by means of an electronic bridge. The bridge, which has been fully described elsewhere (30,31), was operated at 200 Hz. In practice, the mutual inductance coils for the CrKAlum, which were identical to those used earlier (26), were not required. The CMN coils consisted of a main primary coil 3-3/4" in length wound of 4100 turns of #40 formex-coated copper wire in four layers. Two oppositely wound secondaries were placed symmetrically over the primary and separated by 1/2". These secondaries were each 1-5/16" long and consisted of 1432 turns in four layers. The new feature of this apparatus was that two nulling coils were added to the main primary. These were 1/8" wide and consisted of 115 turns in four layers. They were situated 3-3/4" on either side of the center of the main primary. These nulling coils, which opposed the main primary coil, created a quadrature coil which had a symmetrical field less sensitive to the stainless steel vacuum jacket, CrKAlum and other magnetic materials which could have interfered with measurements of the CMN.

All these coils were wound over 0.005" fish paper. Each layer was sprayed with Krylon and covered with lens paper. The diameter of the coils was such that coils and fish paper would slide snugly onto the outer vacuum jacket.
2.7 Internal Wiring

The leads from the top of the cryostat to the outer flange were of #36 formex-coated copper wire. Capacitors (0.05 μF) were placed from each heater and carbon resistor lead to ground at the terminal box at the top of the cryostat. Capacitors (100 pF) were placed between these lines and ground at the top of the outer flange. The purpose of these capacitors was to reduce high frequency pickup.

Copper to nylon pressure seals (Reich seals) (32) were used to bring the leads through the outer vacuum flange. Between outer and inner flanges, the leads were 4" pieces of spring-coiled 36.7% lead-on-constantan wire (0.002") supplied by Sigmund Cohn Corporation. In passing through the inner flange, the leads took the form of manganin wire embedded in Epibond 100A (33). The manganin wires were soldered to tiny tinned screws held in a teflon ring just below the inner flange. To the other end of each screw were soldered the lead-constantan wires which ran directly to the heater and carbon thermometers.

A copper ground wire running from the top of the cryostat was soldered to a pumping line just above the outer flange. One side of each carbon thermometer and one side of the heater were grounded to the filling tube. The coil foil running from the lower CrKAlum pill to the cell was also grounded to the filling tube, as was the lead switch between the CrKAlum pills.
2.8  CrKAlum Salt Pills and Lead Switches

In order to reach the temperature region desired, it was decided to construct two CrKAlum pills, the lower to cool the cell and the upper to act as a guard. Single crystals of CrKAlum were grown from the saturated solution. The best crystals obtained were about 1-1/2" across. In order to obtain a high density of crystals and good thermal contact in a small space, it was decided to slice the crystals up into slabs 3 mm in thickness. A problem arises in the handling and cutting of CrKAlum crystals because of the rapidity with which they lose their water of hydration and are reduced to a useless white powder. The crystals must be completely protected with grease and refrigerated whenever possible.

The cutting was done with a diamond thin section cut-off saw in the laboratory of M. P. Weiss of the Geology Department. This saw had a vacuum chuck. Hence the crystals were mounted on glass microscope slides with Dow Corning Glass and Ceramic Adhesive. It was found that when the saw blade was cooled with a mixture of ethylene glycol and water, there was no apparent degeneration of the crystals during the cutting.

A. Upper pill

The construction of the pills is illustrated in Figures 2 and 3 (34). A nylon form 0.1" thick and 1" wide had a thin sheet of mylar wrapped around it. In the upper pill, the 3 mm slabs, covered thickly with N grease, were fitted closely together in six layers. Each layer was 3-1/2" in length. The two
Figure 2

Upper (Guard) CrKAlum Salt Pill
Transverse foil To Pb switch and He$^3$ pot

CrKAlum

Nylon form

To lower pill

1″

3 1/2″
Figure 3

Lower CrKAlum Salt Pill
Transverse foils connecting both halves of pill.
bottom layers were 1" wide, the middle layers 3/4" wide, and the outer layers 1/2" wide. Coil foil was arranged between the layers as shown. All foils had the copper wires running vertically, except those labeled "transverse," which had horizontal wires. The entire pill was very tightly bound with many turns of cotton thread. The total mass of CrKAlum in the upper pill was about 48 grams and the area of contact between crystals and coil foil was about 200 cm^2.

B. Lower pill

The lower pill was similar except that there were four layers of crystals rather than six. In addition, there was a piece of transverse coil foil surrounding the entire pill. The filling tube and leads to the capacitor were well greased and placed under this outer foil. The mass of crystals in the lower pill was 41 grams and the area of contact to coil foil was about 160 cm^2.

The magnet used in these experiments had a maximum field of 18,000 gauss with a maximum current of 200 amperes at 300 volts. The pole faces had a gap width of 3-1/16" and diameter of 9-1/2". The field was at 95% of its maximum over the central 8" of this diameter. The apparatus was arranged so that the center of the pole faces came just between the CrKAlum pills. Thus the maximum field at the extreme ends of the pills was about 14,500 gauss.

C. Lead switches

Three lead switches were used for thermal contact. Two switches in series connected the He^3 reservoir and the upper CrKAlum pill. A copper tube 1/4" in diameter and 4-1/8" long was inserted to its full length into the inner
vacuum pumping line. At the bottom, the tube was clamped tightly around the He³ reservoir by a bracket. Two screws insured a tight fit. The area of contact was 6.9 cm² and was covered with N grease. Inside the copper tube was a bundle of four 99.99% pure copper rods held away from the tube by nylon spacers. At the top of the tube, a lead switch connected the rods to the tube. A/\ell_1 for this switch was 9 \times 10^{-3} \text{ cm}. At the bottom of the tube, another lead switch with A/\ell_2 = 4.3 \times 10^{-3} \text{ cm} connected the copper rods to a copper post 1/4'' x 1/16'' x 1-1/2''. This post was silver soldered to a copper ring 3/8'' in diameter and 1/2'' high. Two coil foils leading to the interior of the upper pill were greased and bound tightly around the ring. About 950 wires were in these foils and the area of contact with the ring was about 4.5 cm². The reason for this elaborate two-switch arrangement was to get the upper lead switch into a region of weaker field where it would go from the normal to the superconducting state early in the demagnetization process. The maximum field at the upper switch was about 1500 gauss. The first step in the demagnetization lowered the voltage to 125 volts, which corresponded to a field of less than 500 gauss at the upper switch. Since the critical field for lead is about 800 gauss below 1°K, it is clear the switch went superconducting during this first step, thus isolating the He³ refrigerator from the lower parts of the apparatus.

Two coil foils running downward from the interior of the upper pill had their ends silver soldered. The solder was then filed down to form a smooth, flat tab. A foil running upward from the interior of the lower pill was given a
similar tab. The third lead switch connected the two pills through these tabs. This switch had an A/\zeta of 4.5 \times 10^{-3} \text{ cm}, and was positioned as nearly as possible to be parallel to the field.

All the lead switches were constructed from 99.9999\% pure Johnson-Matthey lead. All soldering of the switches was done with 99.999\% pure indium (Indium Corporation of America). The dimensions of the switches were the result of a compromise. One desires the A/\zeta to be large when removing the heat of magnetization from the CrKAlum pills. However, one wants A/\zeta to be small after the demagnetization has been completed in order to have a small heat leak into the colder parts of the apparatus. The dimensions chosen proved to be satisfactory on both counts. The temperature of the upper CrKAlum pill typically dropped from 2.4°K to 0.8°K in three hours.

D. Cooling wires

Three foils running downward from the interior of the lower pill were silver soldered into a press fit seal of copper. This seal was pressed into the top of the nylon cell case, forming a vacuum seal. The wires, about 1370 in number, ran on through the nylon and into the CMN chamber, where they came into contact with the He^3 - He^4 sample.

2.9 The Capacitor

Since the dielectric constant of the mixture would be approximately 1.057 (35) and since the molar volume of the mixture would vary roughly as (27.5 + 7.8X) cm^3/mole (16), the change expected for a 1\% change in X would
be $1.5 \times 10^{-16}$ farad if the capacitor had a value of $1 \times 10^{-12}$ farad. The electronics was capable of reading $1 \times 10^{-17}$ farad. Thus it was decided to construct a capacitor with a nominal value of $5 \times 10^{-12}$ farad or 5 pF.

A. Old capacitor

It was very important that the capacitor be as free as possible from magnetic materials. Six plates, 3 mm by 7 mm, were machined from 22K gold. The impurities were silver and copper. The plates were held in a rigid package with Epibond 100A which extended in 1 mm from each end of the plates. The gap width between plates was 0.25 mm. The capacitor is depicted in Figure 4. The leads to the capacitor were 0.01" diameter platinum wires, which were spot-welded to alternate plates.

This capacitor had one deficiency which led finally to its being abandoned. It turned out that, when the capacitor was filled with pure He$^4$, the capacitance was temperature dependent, decreasing with temperature to a minimum at about 0.05°K and increasing slightly at lower temperatures. This effect is not fully understood, but almost certainly is due to some electrical interaction in the Epibond epoxy between the ends of the plates. This temperature dependence is shown in Figure 9. In addition, at temperatures below about 0.1°K, the capacitance had a voltage dependence. Rather than attempt to correct for these unexpected effects, it was decided to construct a new capacitor without any epoxy between the plates.
Figure 4

Old Capacitor
B. **New capacitor**

This new capacitor is illustrated in Figure 5. Three pieces of Epibond 100A were machined as shown. They fit together to form a disk 0.810" in diameter and 0.157" thick with two parallel slits 0.008" in thickness near the center. Four pieces of gold foil (99.99% purity from Sigmund Cohn Company) 0.0005" in thickness were bonded to the sides of the slits with Epibond 121 (36). After immersion in liquid nitrogen, the foil showed no wrinkles or signs of pulling away from its substrate. Each foil was approximately 0.157" by 0.450". The two inside plates were set in 0.05" from one end and the outside plates 0.05" from the other end. Also the Epibond was machined away from the ends of the plates as shown. This was to minimize end effects and insure that the capacitor would "see" as little epoxy as possible. The gap width between the plates was 0.006".

The inner plates had tiny tabs which lay in grooves in the Epibond perpendicular to the plates. These tabs were soldered with minute amounts of indium to 0.005" copper wires which were soldered to the H line leading to the capacitance bridge (see below). The outer plates also had fine gold tabs soldered with indium to copper wires leading to the L line of the bridge. All the indium solder joints were covered with Epibond 121 for additional strength.

The amount of indium used in these joints has been estimated at four milligrams. Assuming it to be an ideal superconductor, one can quickly estimate the magnetic effect of this indium.
Figure 5

Top View of the New Capacitor
\[ \rho \text{ for indium} = 7.3 \text{ grams/cm}^3 \]
\[ 0.004 \text{ g gives } 5.48 \times 10^{-4} \text{ cm}^3 \]

For an ideal superconductor, \( \chi = \frac{1}{4\pi} \text{ per cm}^3 \)
\[ \therefore \chi = \frac{5.48 \times 10^{-4}}{4\pi} = 4.36 \times 10^{-5} \]

This may be compared with \( \chi \) for CMN. There were 2.56 grams of CMN in the cell. At 1°K, this gives \( \chi = 1 \times 10^{-3} \).

C. Capacitance measurement

Capacitance was measured by means of a General Radio 1615A capacitance bridge. A General Radio 1311A audio oscillator and General Radio 1232A tuned amplifier were accessories. A Princeton Applied Research JB-4 lock-in amplifier was used to give increased sensitivity. Measurements were made with the oscillator at 2 kHz and an output of eight volts. These were compromise figures chosen to give good sensitivity without excessive heating of the cell.

The bridge was capable of resolving easily changes in capacitance of \( 10^{-5} \) pF. All measurements were made in the three-terminal mode. In this mode, all capacitances to ground from either of the unknown connectors are excluded from the measurement. One terminal, designated the "H" terminal, is connected to the low-impedance output of the transformer. The other terminal, designated the "L" terminal, is connected to the high-impedance side of the detector. Thus the L line must always be shielded, while shielding is optional for the H line.
D. The "H" line

Identical 15 foot sections of Belden 8259 shielded coaxial cable carried the H and L lines from the bridge to the top of the cryostat. There the H line entered the cryostat through a BNC connector and black wax joint. In the He<sup>4</sup> bath space, the H line was shielded by a grounded stainless steel capillary which was open at the bottom. The lead passed through the outer flange by means of a Reich seal, was spring-coiled between the flanges and passed through the inner flange in the Epibond seal. Thus, from just above the outer flange to just below the inner flange, the H line was identical to the heater and carbon thermometer leads. Just below the inner flange, the H line entered a 1 mm grounded copper-nickel capillary open at the top. This capillary was thermally anchored to the lower CrKAlum pill as described earlier. At the top of the cell, the capillary terminated at a small nylon insulator. The wire was soldered with indium into a small press-fit copper seal, which was pressed into the top of the cell. From the top of the cryostat to just below this seal, the wire used was 0.002" lead-on-constantan. To avoid possible magnetic effects, this was soldered just below the seal to a 0.005" copper wire which ran straight down to the capacitor. Except for the short distance between inner and outer flanges, the wire was protected with fine teflon spaghetti.

E. The "L" line

The L line, at the top of the cryostat, passed through a BNC connector and black wax joint into a stainless steel vacuum-tight tube which had a
thermocouple gauge and pumping line. During experiments, this line was evacuated. In the He\(^4\) bath, about 8" above the outer flange, this tube was interrupted by a copper box. This box contained a central cylinder around which many turns of the lead-constantan wire were wound, covered with N grease and bound with cotton thread. The two halves of the box were screwed together and were kept vacuum-tight with an indium O-ring seal. The purpose of this box, of course, was to insure that the wire was really at 1.2\(^\circ\)K before it entered colder parts of the apparatus. The vacuum-tight shielding tube continued on through both inner and outer flanges (copper capillary from the box to the outer flange and copper-nickel thereafter). Just below the inner flange, the tube was terminated by a vacuum-tight Stycast plug. The wire continued through the plug and immediately entered another 1 mm grounded copper-nickel capillary. Shielding was made continuous by a grounded copper-nickel sleeve which overlapped both capillaries. Thermal contact between the two tubes was made poor, however, by teflon washers. From this point on to the capacitor, the L line was identical to the H line. Except in the copper box in the bath and at the press fit seal at the top of the cell, the lead-constantan wire of the L line was protected throughout with fine teflon spaghetti.

The capacitor leads were thus shielded from each other everywhere except for about 4" in the cell itself (see Figure 6). There was some concern that the leads might move about with changes in temperature thus causing significant and uncontrollable changes in capacitance. An experiment was done with all the leads in place but not connected to the capacitor. The capacitance
Figure 6

Sample Cell
at 1.2°K and below due to the unshielded portions of the leads was about 0.06750 pF. From this experiment, it was concluded that changes in capacitance due to the leads were negligible in the temperature region of interest.

2.10 The Sample Cell

The cell is shown in Figure 6. The 3" section of solid nylon at the top was designed to keep the magnetic materials at the top of the cell well removed from the CMN. The CMN chamber, originally a cylinder 0.750" in diameter and 0.625" high, was made smaller by the addition of a nylon plug 0.215" thick with appropriate slots and holes to allow the leads, cooling wires, and liquid mixtures to pass through. The CMN was in the form of 1 mm grains (passed by a #10 sieve but stopped by a #20) and 2.56 grams were used. The 1370 cooling wires extended about 1" into the CMN chamber. The wires and CMN were arranged as uniformly as possible and the packing was loose.

A filter was necessary to prevent particles of CMN from falling down onto or into the capacitor. For this purpose, a nylon ring was prepared with o.d. 0.805", i.d. 0.715" and height 0.040". A circular piece of woman's nylon stocking was bonded to one side of this ring with a small amount of Epibond 121. This formed a very effective filter. The CMN particles were thus directly above the nylon mesh, while the top of the capacitor was 0.040" below it.

The bottom of the cell consisted of a nylon plug, which extended 0.502" up into the cell. A vacuum seal between this plug and the cell walls was obtained by using liquid phenol. Between the top of the plug and the bottom of
the capacitor was a loose nylon spacer disk 0.279" in height. The purpose of
the disk was to allow the bottom plug to be drilled out, should the need arise,
without destroying the capacitor. This operation was carried out successfully
once when the old temperature-dependent capacitor was replaced.

2.11 Dead Volumes

Based on the room temperature dimensions, the dead volume of the
cell and fill line to the top of the cryostat was 3.72 cm$^3$. The volume of the fill
line to the top of the cell was 0.68 cm$^3$. These two quantities were measured
experimentally by filling up the respective volumes with liquid He$^4$ at 1.2°K.
The measured volumes were 4.04 cm$^3$ and 0.65 cm$^3$. Part of the discrepancy
could be accounted for because the bottom plug was pushed out about 1 mm from
the cell while the phenol was drying. This problem of the dead volumes will be
dealt with in more detail in the discussion of the data.

2.12 Spacers and Heat Leaks

Because of the length of the apparatus and the small clearance between
it and the inner vacuum jacket, spacers were needed to prevent an excessive
heat leak due to a "touch". All spacers were of nylon and were filed to sharp
points to minimize contact area. A three-pronged spacer screwed into the
bottom of the sample cell. A four-pronged spacer was located at the top of the
cell. Ten spacers about 1/2" long and 1/8" wide were ground down until they
were very thin except for a sharp ridge in the center. These were then bound
tightly to the CrKAlum pills. Four were bound symmetrically around the top
of the lower pill. Six were bound symmetrically around the upper pill, three at the top and three at the bottom. Only a few of the total of 17 nylon points should have been touching the inner jacket at any one time. A nylon spacer was also fastened to the inner jacket to prevent its touching the outer jacket.

Thermal resistances between the He$^3$ reservoir and the cell have been tabulated in Table 2. The measured heat leak into the guard pill was about 7.5 ergs/sec at 0.09°K. The heat leak into the lower pill was about 3 ergs/sec at 0.05°K. The heat leak into the cell at these temperatures appeared to be too small to be readily measured.
### TABLE 2

**THERMAL RESISTANCES BETWEEN He$^3$ RESERVOIR AND CELL**

<table>
<thead>
<tr>
<th>Material or Boundary</th>
<th>Thermal Resistance (cgs units)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>He$^3$ reservoir - copper collar</td>
<td>$\frac{3 \times 10^{-6}}{AT^3} = \frac{4.4 \times 10^{-7}}{T^3}$</td>
<td>27</td>
</tr>
<tr>
<td>Copper tube</td>
<td>$\frac{0.568 \times 10^{-7}}{AT} = \frac{5.20 \times 10^{-6}}{T}$</td>
<td>37</td>
</tr>
<tr>
<td>Pb switch</td>
<td>$\frac{2.5 \times 10^{-8}}{AT} = \frac{2.78 \times 10^{-6}}{T}$ (normal)</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>$\frac{1.25 \times 10^{-6}}{AT^3} = \frac{1.39 \times 10^{-4}}{T^3}$ (superconducting)</td>
<td>38</td>
</tr>
<tr>
<td>Copper rods</td>
<td>$\frac{0.568 \times 10^{-7}}{AT} = \frac{2.84 \times 10^{-5}}{T}$</td>
<td>37</td>
</tr>
<tr>
<td>Pb switch</td>
<td>$\frac{2.5 \times 10^{-8}}{AT} = \frac{5.81 \times 10^{-6}}{T}$ (normal)</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>$\frac{1.25 \times 10^{-6}}{AT^3} = \frac{2.91 \times 10^{-4}}{T^3}$ (superconducting)</td>
<td>38</td>
</tr>
<tr>
<td>Copper ring</td>
<td>$\frac{0.568 \times 10^{-7}}{AT} = \frac{3.50 \times 10^{-6}}{T}$</td>
<td>37</td>
</tr>
</tbody>
</table>
### TABLE 2 - (Continued)

<table>
<thead>
<tr>
<th>Material or Boundary</th>
<th>Thermal Resistance (cgs units)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Copper ring - coil foil</td>
<td>( \frac{3 \times 10^{-6}}{\text{AT}^3} = \frac{6.7 \times 10^{-7}}{T^3} )</td>
<td>27</td>
</tr>
<tr>
<td>Coil foil</td>
<td>( \frac{0.568 \times 10^{-7} \ell}{\text{AT}} = \frac{2.43 \times 10^{-5}}{T} )</td>
<td>37</td>
</tr>
<tr>
<td>Coil foil - CrKAlum</td>
<td>( \frac{3 \times 10^{-6}}{\text{AT}^3} = \frac{6.7 \times 10^{-8}}{T^3} )</td>
<td>27</td>
</tr>
<tr>
<td>CrKAlum - coil foil</td>
<td>( \frac{3 \times 10^{-6}}{\text{AT}^3} = \frac{2.2 \times 10^{-8}}{T^3} )</td>
<td>27</td>
</tr>
<tr>
<td>Coil foil</td>
<td>( \frac{0.568 \times 10^{-7} \ell}{\text{AT}} = \frac{1.44 \times 10^{-5}}{T} )</td>
<td>37</td>
</tr>
<tr>
<td>Pb Switch (normal)</td>
<td>( \frac{2.5 \times 10^{-8} \ell}{\text{AT}} = \frac{5.55 \times 10^{-6}}{T} ) (normal)</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>( \frac{1.25 \times 10^{-6} \ell}{\text{AT}^3} = \frac{2.78 \times 10^{-4}}{T^3} ) (superconducting)</td>
<td>38</td>
</tr>
<tr>
<td>Coil foil</td>
<td>( \frac{0.568 \times 10^{-7} \ell}{\text{AT}} = \frac{4.05 \times 10^{-5}}{T} )</td>
<td>37</td>
</tr>
<tr>
<td>Coil foil - CrKAlum</td>
<td>( \frac{3 \times 10^{-6}}{\text{AT}^3} = \frac{9.7 \times 10^{-8}}{T^3} )</td>
<td>27</td>
</tr>
<tr>
<td>Material or Boundary</td>
<td>Thermal Resistance (cgs units)</td>
<td>Reference</td>
</tr>
<tr>
<td>---------------------------</td>
<td>--------------------------------</td>
<td>-----------</td>
</tr>
<tr>
<td>CrKAlum - coil foil</td>
<td>$\frac{3 \times 10^{-6}}{AT^3} = \frac{2.4 \times 10^{-8}}{T^3}$</td>
<td>27</td>
</tr>
<tr>
<td>Coil foil</td>
<td>$\frac{0.568 \times 10^{-7}t}{AT} = \frac{4.22 \times 10^{-5}}{T}$</td>
<td>37</td>
</tr>
<tr>
<td>Copper wires - helium</td>
<td>$\frac{7.0 \times 10^{-6}}{AT^3} = \frac{1.28 \times 10^{-7}}{T^3}$</td>
<td>39</td>
</tr>
<tr>
<td>Helium - CMN</td>
<td>$\frac{6.0 \times 10^{-6}}{AT^3} = \frac{8.0 \times 10^{-8}}{T^3}$</td>
<td>40</td>
</tr>
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</table>
CHAPTER III

EXPERIMENTAL PROCEDURE AND RESULTS

3.1 Procedure

A. Experiments at constant $X$

The He$^3$ concentrations in the solutions investigated in this research were 3.92%, 6.96%, 8.10%, 9.92%, 12.02%, 15.02% and 15.38%. The 15.02% mixture was prepared by the author. The other mixtures were prepared by Dr. M. Skeric and Dr. D. Husa (26). The concentrations were determined by calculations involving the relevant volumes, pressures and temperatures. Two runs were made with each solution except for the two 15\% solutions, which were each run once.

To insure thermal equilibrium, a very small amount of solution was let into the cell while the He$^4$ bath was being pumped down to 1.2\(^\circ\)K. With the He$^3$ reservoir full and closed off, a CMN calibration point was taken against He$^3$ vapor pressure, after equilibrium had clearly been achieved at 1.2\(^\circ\)K. A separate calibration run of the CMN had been made earlier against He$^3$ and He$^4$ vapor pressures and the slope of the calibration line was presumed to remain constant. The results of this run are shown in Figure 7. The temperature is determined from the equation $T = A/r - B$, where $r$ is the inductance bridge.
Figure 7

CMN Salt Calibration
reading for the CMN, A is the slope of the line which results when r is plotted vs 1/T, and B is a constant. We found A to be 1033 ± 5. B was considered to be determined to ±0.5% in each experiment. Hence temperatures were measured to about ±1%. In Figure 8, an earlier calibration is shown for comparison. Between these two calibration runs, the apparatus was warmed to room temperature and disassembled.

The solution was let into the cell through two Zeolite (41) traps cooled with liquid nitrogen. Filling the cell took from one to five hours, depending upon the pressure in the mixture storage bottle. As soon as the cell was closed off, the magnet was positioned and the field brought quickly up to 18,000 gauss. When the upper CrKAlum pill reached about 1.9°K, we began pumping on the He³ reservoir. When the upper pill reached about 1.2°K, we recondensed He³ back into the reservoir and closed it off. He⁴ was then retransferred into the bath. If the transfer was done carefully, it was found that the He³ reservoir stayed at 1.2°K. We then pumped down the He⁴ bath and resumed pumping on the He³. This technique of recondensing He³ and retransferring He⁴ with the magnet on seemed to yield the maximum time for taking data before the supply of either He³ or He⁴ ran out.

After the retransfer, the magnet was kept on full for approximately two hours. By this time the lower pill was usually at about 0.9°K and the upper pill at 0.8°K (measured by the carbon thermometers). Demagnetization was begun at this point. The magnet voltage was turned down manually very slowly with stops at voltages of 125, 65, 35, 17 and 7 volts. The demagnetization
Figure 8

CMN Salt Calibration
required about 90 minutes. This left 4 to 5 hours for taking capacitance measurements before the He\textsuperscript{4} bath level dropped too low to maintain thermal equilibrium.

After the magnet was rolled away from the cryostat, the apparatus was allowed to come into equilibrium for a data point at the lowest temperature. The heater was then used to bring the capacitor and lower pill up in temperature for successive points. The times required for achieving equilibrium following these heat pulses will be discussed later.

A typical experimental timetable has been included in Appendix A.

B. Evaporation effects

In making measurements on He\textsuperscript{3} - He\textsuperscript{4} mixtures, one must be careful to avoid a depletion of the He\textsuperscript{3} in the cell by evaporation. In our experiments, the cell was generally about two-thirds full of liquid (exact volumes of liquid for each run are given in Appendix C). In certain cases there was less than this amount of liquid because some mixtures were at very low pressures in their storage bottles and a normal filling was impossible. Tests were made which showed that the variations in the amount of liquid in the cell in the experiments made no difference in the thermal equilibrium and made no changes in the results at low temperatures. In any case, we were careful to avoid overfilling the cell, since having liquid in the filling tube could have caused a serious heat leak.
If the cell is at a temperature where the vapor pressure of the solution is appreciable (see Figure 13), some He\textsuperscript{3} will be in the vapor phase in the tubing. If some of the tubing is colder than the cell, He\textsuperscript{3} may recondense and become trapped there. This would not be serious while taking measurements of the lower phase in a phase-separated mixture. However, in the single phase region, a depletion of He\textsuperscript{3} in the cell would lead to incorrect results. In particular, it would lead one to believe that there is a larger thermal contraction in the solution than is actually the case. This effect caused some difficulty in our preliminary measurements and has also been noted by other investigators (42).

During demagnetization it is quite unavoidable that portions of the filling tube in contact with the CrKAlum pills should be colder than the cell. However, if the filling tube has been carefully constructed so that it is sloping downward at every point, any He\textsuperscript{3} that is drawn up out of the cell during demagnetization should drain back down as thermal equilibrium is established. If, however, the tube contains low spots where liquid can collect, some He\textsuperscript{3} could be trapped in pools and never get back to the cell. We attempted to avoid low spots in our tubing. However, it is rather difficult to guarantee the configuration of many turns of fine capillary tubing, so we took an additional precaution. As the cell and lower pill warmed up to the point where the mixture was about to pass from a phase-separated to a non phase-separated state, the upper (guard) pill was warmed up sufficiently to evaporate and drive down any liquid He\textsuperscript{3} that might have collected near it. This was accomplished
by using the upper carbon thermometer as a heater. For each succeeding point, up to 1.2°K, the guard was kept slightly warmer than the cell. Naturally, we did not let the guard get too much warmer than the cell, since this would have spoiled the thermal equilibrium of the cell and lower pill while taking a point. In addition, when the temperature reached 0.3°K, we stopped pumping on the He³ reservoir and heated it as well.

3.2 Data Analysis and Results

A. The capacitor model

In order to analyze the data, we adopted the following model of the capacitor: When the capacitor is empty, let the "ideal capacitance" (that due to the field in the gap between the gold plates) be denoted by $C_1$. Then the capacitance measured by the bridge will be that due to $C_1$ plus another capacitance in parallel with $C_1$. This second capacitance may be viewed as due to the fringing field, whose lines of force travel mainly through the epoxy, rather than directly across the gap between the plates. If we denote the measured value of the empty capacitor by $C_0$, this second capacitance will have the value $C_0 - C_1$. Experimentally $C_0 - C_1$ was found for the run of May 3 to have the value 0.54555 pF at $T = 0.05°K$.

When liquid helium, with dielectric constant $\varepsilon$, is let into the cell, the ideal capacitance will become $C_1\varepsilon$. The change in the second capacitance will be negligible, however, since the dielectric constant of the epoxy is far greater than that of the liquid. Hence the measured capacitance will be just
\[ C = C_0 + C_1(\epsilon - 1) \]  

(III-1)

In general, one would expect that \( C_0 \) and \( C_1 \) might depend upon temperature and pressure and that there might be a zero shift between runs. Thus one could write:

\[ C_0 = C_0(0) + C_0(T) + C_0(P) + C_0(R) \]  

(III-2)

It was found that, in fact, \( C_0 \) did depend upon these three variables. We defined \( C_0(T) = 0 \) at \( 0.05^\circ \text{K} \), \( C_0(P) = 0 \) when the liquid is under very low pressure and \( C_0(R) = 0 \) for the run of May 3 (R denotes the particular run). It will be shown that all three terms are small compared to 5 pF, the nominal value of \( C \). In Eq. (III-1), \( C_1 \) is multiplied by \( (\epsilon - 1) \), which has a value of approximately 0.05. We are only concerned with the variations in \( \epsilon - 1 \), which, of course, are smaller still. Therefore it is permissible to neglect the effects of \( P, T \) and \( R \) upon \( C_1 \). Hence we can write:

\[ C = C_0(0) + C_0(T) + C_0(P) + C_0(R) + C_1(\epsilon - 1) \]  

(III-3)

B. The capacitance equation

The dielectric constant of the solution is related to the density by the Clausius-Mosotti equation, which is taken to be applicable to both the \( \text{He}^3 \) and the \( \text{He}^4 \). It has been shown recently that this is not true rigorously (43). However, the deviations will be extremely small in an experiment where density variations are small.

The Clausius-Mosotti equation may be written:

\[ \epsilon - 1 = \frac{4\pi N\alpha}{1 - (4\pi/3)N\alpha} \]  

(III-4)
where \( N = N_0 / v \) is the number density and \( \alpha \) is the electric polarizability per atom.

Equation (III-4) reduces to

\[
\epsilon - 1 = \frac{1}{v} \left( \frac{1}{4\pi N_0 \alpha} - \frac{1}{3} \right)
\]

(III-5)

where \( v = v(X, T) \) is the molar volume of the solution.

\( N_0 \alpha = 0.1234 \) at zero frequency according to Atkins (44).

\[
\therefore \quad \epsilon - 1 = \frac{1.551 \text{ cm}^3}{v(X, T) - 0.5169 \text{ cm}^3}
\]

(III-6)

Substituting this expression into Eq. (III-3),

\[
C(X, T) = C_0(T, P, R) + \frac{1.551 \text{pF cm}^3}{v(X, T) - 0.5169 \text{ cm}^3}
\]

\[
= C_0(T, P, R) + \frac{\Gamma}{v(X, T) - 0.5169 \text{ cm}^3}
\]

(III-7)

where \( \Gamma \) is now a constant with the dimensions of a capacitance times a volume.

By measuring the capacitance before and after filling the capacitor at 1.25°K with pure He\(^4\), whose molar volume is accurately known (45), we could solve Eq. (III-7) for \( \Gamma \). Four trials produced values of \( \Gamma \) of 7.121, 7.076, 7.084 and 7.076 pF·cm\(^3\). We took the average value to be \((7.09 \pm 0.02)\) pF·cm\(^3\).

1. Temperature and pressure corrections

The term \( C_0(T) \) was obtained by varying the temperature with pure He\(^4\) in the cell. The results are plotted in Figure 9. The data have been corrected for the expansion of He\(^4\) with increasing temperature. It will be recalled that
Figure 9

Temperature Dependence of the Capacitor
\[ C_0(T) \times 10^{-5} \text{ pF} \]

- **OLD CAPACITOR**
- **NEW CAPACITOR**

Graph showing the variation of \( C_0(T) \times 10^{-5} \text{ pF} \) with temperature \( T(\text{°K}) \).
the new capacitor with no epoxy directly between the plates was constructed to reduce the size of $C_q(T)$. It is seen that $C_q(T)$ is taken to be 0 at 0.05°K and increases to $36 \times 10^{-5}$ pF at 1.25°K. For comparison, $C_q(T)$ for the old capacitor is also shown.

The term $C_0(P)$ was obtained by filling the cell and tubing to the top of the cryostat with pure He$^4$. At a constant temperature of 1.25°K, pressures of up to 1.6 atmospheres were then applied to the surface of the liquid. The results are shown in Figure 10. The data have been corrected for the change in density of liquid He$^4$ with pressure. It is seen that the data fall along a straight line of slope $-1.16 \times 10^{-5}$ pF per mm Hg pressure.

2. The filling experiment

The term $C_0(R)$ represents zero shifts which the capacitor underwent from run to run. In order to accurately find this term, another experiment was devised. Successive small amounts of He$^3$ were added to pure He$^4$ in the cell, changing $X$ from 0.0 to about 0.15. The capacitance was measured for each $X$. This will hereafter be referred to as the "filling experiment." The experiment was carried out at a constant temperature of 1.27°K (the temperature of the He$^4$ bath) or 0.69°K (obtained by regulating the rate of pumping on the He$^3$ reservoir. In the 1.27°K experiment, an uncertainty developed due to evaporation and refluxing effects in the filling tube. Since at 0.69°K the vapor pressures are so small that this uncertainty is very much less, we have considered the 0.69°K
Figure 10

Pressure Dependence of the Capacitor
\[ \frac{[C(P=0) - C(P=P)]}{pF} \]

\( P \) (cm Hg)
data to be more reliable. The evaporation correction will be discussed in
more detail later.

From Eq. (III-7), we can find the molar volume of a solution from the
measured value of the capacitance if we know $C_0$:

$$v(X, T) = 0.5169 \text{ cm}^3 + \frac{7.09 \text{ pF-cm}^3}{C - C_0}$$  \hspace{1cm} (III-8)

The molar volume of pure He$^4$ at 0.69°K is 27.5810 cm$^3$/mole (45). Thus,
using this value and the value of $C$ obtained when the cell contained only He$^4$,
Eq. (III-8) gave $C_0 = 5.10183 \text{ pF}$ for $P = 0$, $T = 0.69°K$ and this particular run.
Now the molar volumes for each concentration between 0.0 and 0.15 could be
found. $C_0(R)$ was found from this data as explained in Section 4.

3. The BBP parameter $\alpha$

The molar volumes, determined in the filling experiment explained
above, lead to a determination of the BBP parameter $\alpha$ (10). We recall that
$1 + \alpha$ represents the ratio of the volumes per atom which He$^3$ and He$^4$ carve out
of the liquid:

$$v(X, T) = v_4(T) [1 + \alpha(X, T)X].$$  \hspace{1cm} (III-9)

Thus, knowing $v$ for various values of $X$, we can solve for $\alpha$. Values of $\alpha$ are
plotted vs $X$ in Figures 11 and 12. The 0.69°K data lie along a straight line
with the equation

$$\alpha = 0.267 + 0.018X.$$  \hspace{1cm} (III-10)
Figure 11

The BBP Parameter $\alpha(X)$ at 0.69°K
Figure 12

The BBP Parameter $\alpha(X)$ at 1.27°K
Therefore with the aid of Eq. (III-9), we can find the molar volume at 0.69°K for any X:

\[ v = 27.5810[1 + 0.267X + 0.018X^2] \text{ cm}^3/\text{mole}. \quad \text{(III-11)} \]

4. The zero shift

Now we return to the question of the zero shift [or \( C_0(R) \)]. The molar volumes of the 6.96, 8.10, 9.92, 12.02, 15.02 and 15.38% mixtures must, of course, coincide at any given temperature in the phase-separated region. Clearly, \( C_0(R) \) must be chosen so that this is true. Therefore, we arbitrarily defined \( C_0(R) \) to be 0 for the first 9.92% run. Then for the other nine runs, \( C_0(R) \) was chosen so that the molar volumes coincided at 0.070°K. This temperature was chosen because the curve is relatively flat in this region and because there were good data points at or near this temperature for all runs.

Values of \( C_0(R) \) are shown in Table 3.

**TABLE 3**

<table>
<thead>
<tr>
<th>Date</th>
<th>( X(%) )</th>
<th>( C_0(R) ) (pF x 10^{-5})</th>
</tr>
</thead>
<tbody>
<tr>
<td>May 3, 1967</td>
<td>9.92</td>
<td>0</td>
</tr>
<tr>
<td>May 5, 1967</td>
<td>6.96</td>
<td>-124</td>
</tr>
<tr>
<td>May 8, 1967</td>
<td>8.10</td>
<td>+956</td>
</tr>
<tr>
<td>May 12, 1967</td>
<td>12.02</td>
<td>+682</td>
</tr>
<tr>
<td>May 15, 1967</td>
<td>15.38</td>
<td>+443</td>
</tr>
<tr>
<td>May 17, 1967</td>
<td>3.92</td>
<td>+463</td>
</tr>
<tr>
<td>May 27, 1967</td>
<td>3.92</td>
<td>+982</td>
</tr>
<tr>
<td>May 29, 1967</td>
<td>6.96</td>
<td>+891</td>
</tr>
<tr>
<td>May 31, 1967</td>
<td>12.02</td>
<td>+916</td>
</tr>
<tr>
<td>June 2, 1967</td>
<td>9.92</td>
<td>+1487</td>
</tr>
<tr>
<td>June 5, 1967</td>
<td>8.10</td>
<td>+1462</td>
</tr>
<tr>
<td>June 23, 1967</td>
<td>15.02</td>
<td>+1742</td>
</tr>
</tbody>
</table>
C_0(R) could not be determined in the same way for the two 3.92% runs because they do not phase-separate. Instead, C_0(R) was found for these runs by requiring that the data give the correct value of \( v(X, T) \) at 0.69°K as determined by the filling experiment. The mixtures which do phase-separate must, of course, also give molar volumes consistent with the 0.69°K filling experiment, and this allows us to solve for the constant C_0(0). We did this by making the 12.02% run of May 31 give \( v(X, T) = 28.4730 \text{ cm}^3/\text{mole} \) at 0.69°K. [This value follows from Eq. (III-11).] This gave a value for C_0(0) of 5.11679 pF. Molar volumes from the other runs proved to be consistent to within the equivalent of about \( \pm 2 \times 10^{-5} \) pF.

5. The evaporation correction

As has been mentioned earlier, for data at temperatures above about 0.7°K, a complication arises due to evaporation and refluxing effects in the filling tube. The vapor pressure of He^3 is much larger than that of He^4 and this results in a decrease in X for the liquid in the cell at these higher temperatures. A correction must be made for this change in concentration, but a precise calculation is difficult. The vapor pressures of He^3 - He^4 solutions are known from the work of Sydoriak and Roberts (46). We have plotted their data as it pertains to our solutions in Figure 13. The volume of liquid used in each run is known and is listed in Appendix C. The volume available for the vapor is also known. However, the high temperature data were taken near the end of the run when the He^4 bath level was low. Hence there were probably
Figure 13

Vapor Pressures of He$^3$ - He$^4$ Mixtures and Pure He$^4$
temperature gradients along the tubing. Moreover, the vapor did not fill the volume completely, since the refluxing film of superfluid He\textsuperscript{4} would have tended to keep flushing the vapor back down into the cell. The presence of the 1370 cooling wires in the volume above the liquid further complicated matters. Attempts were made to determine how much of the filling system the vapor effectively occupied by taking measurements with the liquid level at various points in the cell and up in the tubing. The results, however, were inconclusive. Therefore, it was decided to estimate the correction at slightly over one-half the correction one would make if the vapor occupied the full volume available to it. This was tantamount to assuming that the volume available for liquid and vapor was 2.88 cm\textsuperscript{3} instead of the actual physical volume of 4.04 cm\textsuperscript{3}. The correction amounted to adding to \( v(X, T) \) a small quantity equal to the decrease in \( v(X, T) \) due to the decrease in \( X \) in the liquid at a given temperature. The largest correction proved to be 0.0130 cm\textsuperscript{3} (equivalent to 12 \times 10\textsuperscript{-5} pF) at 1.25°K for the second 9.92% run which had an unusually small volume of liquid.

C. The molar volumes

We are now able to compute the molar volumes for each data point of each mixture. From Eq. (III-8),

\[
v(X, T) = 0.5169 \text{ cm}^3 + \frac{7.09 \text{ pF cm}^3}{C - C_0}
\]  

(III-12)

These molar volumes are listed in Appendix C and plotted vs \( T \) in Figure 14. In the single phase region, it is seen that there is a pronounced thermal contraction in the solutions which is approximately proportional to \( X \) and \( T \).
Figure 14

The Molar Volume of Dilute Solutions of He³ in He⁴ at the Saturated Vapor Pressure
It is shown in Appendix B that the change in molar volume with temperature can be expressed as:

\[ v(X, T) - v(X, 0) = X_4 \left[ U_F(T) - U_F(0) \right] \left[ \frac{2}{3} (1 + \alpha X) + \beta^* \right] + v_4(T) - v_4(0) - \chi P_{\text{sat}} \]  

(III-13)

where \( \chi_4 \) is the isothermal compressibility of pure He\(^4\), \( U_F(T) \) is the molar energy of an ideal Fermi gas of the same number density, \( \alpha_\chi = (1/\chi_4) \left[ \partial \chi(X)/\partial X \right] \)

\[ = \left[ v_4(0) \right] \left[ \partial \alpha_0/\partial v_4(0) \right] \] and \( \beta^* = \left[ v_4(0)/m^*(X) \right] \left[ \partial m^*(X)/\partial v_4(0) \right] \). Stoner (47) has conveniently tabulated the internal energy of an ideal Fermi gas.

The last term in the expression gives the very small correction that must be applied to the molar volumes because the solutions are slightly compressed by their own vapor pressure. To evaluate this expression at zero pressure then, we use an average value for \( v \) in each experiment, the \( P_{\text{sat}} \) data of Sydoriak and Roberts (46) and an average value for the compressibility. We choose a value for \( \chi \) of \( 1.2 \times 10^{-8} \) cm\(^2\)/dyne from the recent measurements of the velocity of sound in pure He\(^4\) by Whitney and Chase (48) and from the data of Boghosian and Meyer (49) on the variation of \( \chi \) with temperature. We assume that the variation of \( \chi \) with \( X \) can be neglected. Now we have a molar volume, \( v_A \) which differs from the previous \( v \) at the higher temperatures:

\[ v_A(X, T) = v - \delta v_4 + \chi P_{\text{sat}} = X_4 \left[ U_F(T) - U_F(0) \right] \left[ \frac{2}{3} (1 + \alpha_\chi) + \beta^* \right] \]  

(III-14)

In Figures 15 and 16, values of \( v_A \) have been plotted against the function \( X[U_F(T) - U_F(0)]/R \). It is seen that the data for each concentration fall along
Figure 15

\( v_A \) vs \( X[U_F(T) - U_F(0)]/R \) for 3.92, 6.96 and 8.10% Solutions
Figure 16

\[ v_A \text{ vs } X[U_F(T) - U_F(0)]/R \text{ for 9.92, 12.02, 15.02 and 15.38\% Solutions} \]
a straight line. The slopes of these lines, from Eq. (III-14), will be just

\[ x_4 \left[ \frac{2}{3} (1 + \alpha_n X) + \beta^* \right]. \]

In Figure 17, we have plotted the slopes of the lines divided by \( x_4 \) against \( X \). The Y-intercept of the resulting line is \(-0.58 \pm 0.05\); its slope is \( 0.36 \pm 0.3 \). Therefore,

\[ \frac{2}{3} (1 + \alpha_n X) + \beta^* = 0.36X - 0.58 \]  

(III-15)

Boghosian and Meyer (50) have found \( \alpha_n \) to be 1.4. Hence we find

\[ \beta^* = -(1.25 \pm 0.05) - (0.58 \pm 0.3)X \]  

(III-16)

in excellent agreement with the recent second sound data of Sandiford and Fairbank (51).

The value of \( \beta^* \) determines the pressure dependence of the effective mass of the He\(^2\) quasi-particles. In Appendix B, Eq. (B-20) gives:

\[ \beta^* = -\frac{1}{x_4} \left( \frac{1}{m^*} \frac{\partial m^*}{\partial P} \right)_T \]

Taking \( x_4 = 1.2 \times 10^{-8} \) cm\(^2\)/dyne, \( m^* = 2.5m_3 \) and the value of \( \beta^* \) given above,

\[ \left( \frac{\partial m^*}{\partial P} \right)_T = [(0.038 \pm 0.004) + (0.018 \pm 0.01)X] m_3/\text{atm}. \]  

(III-17)

Now if we extrapolate the lines in Figure 16 to \( X[U_F(T) - U_F(0)]/R = 0 \), we obtain a molar volume at \( T = 0^\circ \text{K} \) for each of the mixtures. Thus we can find \( \alpha_0(X) \) for each mixture, since, from Eq. (III-9),

\[ \alpha_0(X) = \frac{v(X, 0) - v_4(0)}{Xv_4(0)} \]  

(III-18)
Figure 17

\[ \frac{2}{3} (1 + \alpha \kappa X) + \beta^* \text{ vs } X \]
$\left[ \frac{2}{3} (1 + \alpha_k X) + \beta^* \right]\overline{X}$

-0.7

-0.6

-0.5

0 5 10 15 20 25 30 35

$X(\%)$

$\Delta$ Sandiford & Fairbank

$\square$ Kerr

$\circ$ Present work
The results are shown in Figure 18. There is considerable scatter in the value of $\alpha_0(X)$. If we express $\alpha(X, 0)$ in the form $\alpha(X, 0) = \alpha_0 + \alpha_0'X + \ldots$, the data may be fit either by $\alpha_0 = (0.284 \pm 0.005)$, $\alpha_0' = 0$ or $\alpha_0 = (0.280 \pm 0.005)$, $\alpha_0' = 0.55$. While the first pair of values perhaps fit our experimental points better, the second pair give a good fit with data for $X = 0.273$ and $X = 0.533$, measured by Kerr (52) and with the volume of pure He$^3$ (53). Both values of $\alpha_0$ are considerably below that recently found by Boghosian and Meyer (50).

The right side of Eq. (III-13) is useful in calculating the solubility curve (see D below) and can be called $\delta v(X, T)$, the decrease in molar volume as the solution goes from 0°K to $T$°K. Equation (III-13) was solved for $\delta v(X, T)$ and the results are presented in Figure 19.

D. The solubility curve

With the use of Eq. (III-18), the values of $X(T)$ in the lower phase may be found:

$$ X(T) = \frac{v - \delta v - v_4(0)}{v_4(0)\alpha_0} \quad \text{(III-19)} $$

Since the filling experiment at 0.69°K gave a value of 0.284 for $\alpha_0$, assuming that $\alpha_0$ is independent of $X$,

$$ X(T) = \frac{(v - \delta v) - 27.5793 \text{ cm}^3}{27.5793 \text{ cm}^3 \times 0.284} \quad \text{(III-20)} $$

or

$$ X(T) = 0.12767(v - \delta v) - 3.5211. \quad \text{(III-21)} $$

Since $\delta v$ is a function of $X$ as well as $T$, the equation was solved for $X(T)$ by iteration. The results are given in Appendix C and in Figure 20.
Figure 18

The BBP Parameter $\alpha_0$ as a Function of Concentration, $X$
Figure 19

The Change in Molar Volume with Temperature
Figure 20

The Solubility Curve for $\text{He}^3$ in $\text{He}^4$ at the Saturated Vapor Pressure
Since the low temperature portion of the solubility curve is of special interest, we have plotted $X$ vs $T^2$ in this region (Figure 21). The data lie along a straight line, indicating that the solubility curve comes down toward $0^\circ$K as a parabola. Below $0.15^\circ$K, the curve fits the empirical equation,

$$X = X_0[1 + 10.8(\circ K)^{-2}T^2]$$  

(III-22)

where $X_0$, the solubility at $0^\circ$K is $(6.37 \pm 0.05)\%$.

E. Equilibrium times

Whenever a current pulse was delivered to the heater and also during the cool-down, it was necessary to wait some finite time for thermal equilibrium to be established in the cell. The times one had to wait ranged from immeasurably small at the high temperatures to 30 minutes or more at the lowest temperatures. A typical plot of temperature vs time for a $6.96\%$ solution is shown in Figure 22. A large number of equilibrium times have been displayed in Figure 23. There is considerable scatter in the points, but the general dependence of $\tau$ upon $T$ is clear. Points obtained by Roach (54) using grains of CMN similar in size to ours and a $5.0\%$ solution are included for comparison.

One might view $\tau$ as made up of three relaxation processes which are occurring simultaneously. The first of these, whose time constant is $\tau_1$, is the local relaxation of the helium solution with the CMN. Secondly, there is the relaxation between the lower CrKAAlum pill and the cell ($\tau_2$). Finally there is the conduction of heat through the solution. Its time constant, $\tau_3$, is given by
Figure 21

He$^3$ Concentration, $X$, along the Phase Separation Curve

as a Function of (Temperature)$^2$
Figure 22

Sample Thermal Equilibrium Time for a 6.96% Solution

\( T_{\text{Final}} = 0.079^\circ \text{K} \)
MAY 29, 1967

$X = 6.96\%$

$T = 0.079^\circ$

$\tau = 280.1 \text{ sec}$
Figure 23

Thermal Equilibrium Times for Dilute Solutions of He$^3$ in He$^4$
\( t^2 C_v / 2 \xi v \), where \( t \) is of the order of 2 cm, \( C_v \) is the specific heat, \( \xi \) the compressibility and \( v \) the velocity of sound. Theoretical curves representing these three processes have been drawn in the figure. It is seen that \( \tau_1 \), which varies as \( 1/T^5 \), is negligible in the region of interest. It is assumed that the Kapitza boundary resistance between CMN and the mixture approximates that between CMN and pure liquid \( \text{He}^4 \) (see Table 2 for a list of thermal resistances). \( \tau_2 \) seems to make the major contribution to the relaxation time in this apparatus.
CHAPTER IV

DISCUSSION

4.1 The Chemical Potential

When the two phases are in thermodynamic equilibrium, the temperature, pressure and chemical potential of each component must be equal. The Gibbs-Duhem relation connecting $\mu_3$ and $\mu_4$ holds separately for each phase. Using Eq. (I-2) for the He$^4$ chemical potential in the upper phase, we can find $\mu_3$ for the upper phase from the Gibbs-Duhem relation:

$$\mu_3^U = \mu_3^0 + RT \ln X \quad \text{(IV-1)}$$

where the superscript 0 refers to the pure liquid and X denotes the He$^3$ concentration in the upper phase.

Edwards and Daunt (3) derived an approximate expression for the He$^3$ concentration in the upper phase. Inserting this expression into Eq. (IV-1),

$$\mu_3^U = \mu_3^0 + RT \ln [1 - 1.13T^{3/2} e^{-0.71/T}] \quad \text{(IV-2)}$$

One must not expect this expression to hold rigorously, since Edwards and Daunt had assumed that the lower phase would be pure He$^4$ at very low temperatures, which turned out to be incorrect. However, the existing experimental data indicate that the expression is still reasonably accurate. In Figure 24, we have plotted the data of Zinov'eva and Peshkov (18, 19).
Figure 24

Percentage of $\text{He}^4$ in the Upper Phase
$T(°K)$

- Brewer & Keyston
- Keyston & Laheurte
- Ouboter, et al.
- Zinov'eva & Peshkov

$1.13T^{3/2} e^{-0.71/T}$
Taconis, le Pair and Beenakker (20), Brewer and Keystone (22) and Keystone and 
Laheurte (55). We shall regard Edwards and Daunt's expression, shown by the 
solid curve, as a useful semi-empirical relation accurate to perhaps ± 10%.

Now, \( dG = -SdT + VdP \),

\[ \mu_3^0(T) = \mu_3^0(T=0) - \int_0^T S_3^0 dT \]  

where we have neglected the small term \( \int VdP \).

\[ \mu_3^0(T) = -L_3^0(T=0) - \int_0^T S_3^0 dT \]  

Therefore, Eq. (IV-1) becomes

\[ \frac{\mu_3^0 + L_3^0}{R} = -\int_0^T \frac{S_3^0}{R} dT + T \ln \left[ 1 - 1.13T^{3/2} e^{-0.71/T} \right]. \]  

Recalling that \( S = \int_0^T (C_P/T) dT \), we can evaluate the entropy of pure He\textsuperscript{3} by 
integrating the specific heat. We have chosen to use the data of Anderson,
Reese and Wheatley (56) at 0.12 atmospheres pressure. These data undoubtedly 
contain some errors. However, in view of the considerable scatter in the 
results of various workers (see, for example, reference 57), the ARW data 
seemed to be satisfactory and convenient to use in the temperature region of 
interest.

We recall from section 1.2B that Ebner has derived theoretical 
expressions for the chemical potential of He\textsuperscript{3} in the lower phase by expanding 
in powers of \( (T/T_F)^2 \) at low temperatures and in powers of \( (T_F/T)^{3/2} \) at high 
temperatures. His expressions are given below.
At low $T$ ($T/T_F < 1$):

$$\mu_3 - E_0 = \mu_F^0 + n_3 V(0) - \frac{n_3 |V_0|}{2} \left[ F(n_3) + G(n_3) \frac{\pi^2}{12} \left( \frac{T}{T_F} \right)^2 \right] \quad (IV-6)$$

At high $T$ ($T/T_F > 1$):

$$\mu_3 - E_0 = k_B T \ln \left[ \frac{4}{3\sqrt{\pi}} \left( \frac{T_F}{T} \right)^{3/2} \right] + k_B T \left\{ \frac{\beta}{3/\pi} \left( \frac{T_F}{T} \right)^{3/2} \right. \left. \right. \right.$$  

$$+ \frac{1}{3\pi} \left( 1 - \frac{16}{9\sqrt{3}} \right) \left( \frac{T_F}{T} \right)^3 + \frac{n_3 V(0)}{k_B T} - \frac{n_3 \langle V \rangle}{2k_B T} \left[ 1 + \left( \frac{T_F}{T} \right)^{3/2} \sqrt{2} \left( 1 - \frac{\langle V \rangle}{\langle V' \rangle} \right) \right] \right.$$  

$$ (IV-7)$$

Ebner has provided plots of the functions $F(n_3)$, $G(n_3)$, $\langle V \rangle$ and $\langle V' \rangle$. In Eq. (IV-6), $\mu_F^0$ is the chemical potential at temperature $T$ of an ideal Fermi gas of number density $n_3$ and mass $m^*_0$.

From these equations, one may find the quantity $[\mu_3^L + (0_3^0)/R]$. If we use our experimental values of $X$ and $T$ along the phase separation curve in Ebner's expressions, we can compare his predictions with Eq. (IV-5). This has been done in Figure 25. The error bars on the solid curve refer to the uncertainty in the specific heat measurements. One would expect Ebner's low-temperature formula to give good results over the range of temperatures shown. It is seen that it agrees well with the experimental data up to about 0.2°K. When it is required that the curve go to zero at $T = 0°K$, it yields a value for the binding energy of one He$^3$ atom at 0°K, $E_0$, of $E_0/R = L_3^0/R = (0.284 \pm 0.007)$ deg K, where $L_3^0$ is the latent heat of pure He$^3$ at 0°K, $L_3^0/R = (2.47 \pm 0.01)$ deg K (58). One would not expect Ebner's high-temperature formula to give good results in this range since, even at 0.365°K, $T/T_F$ is only 0.52. To
Figure 25

The $^3$He Chemical Potential and $T/T_F$ as Functions of Temperature
conform to Ebner's calculations, we have computed the Fermi temperatures using \( m^* = 2.34m_3 \).

It is interesting to note that Ebner's low-temperature formula is brought into better agreement with the experimental curve if \( \alpha_0 \) is increased slightly. For example, the agreement is excellent up to about 0.3°K if \( \alpha_0 \) is changed from 0.284 to 0.288. This, in turn, forces a change in \( E_0; E_0/R = L_0^0/R + (0.281 \pm 0.007) \) deg K.

4.2 Conclusion

We have measured the solubility curve and number density relative to pure \( \text{He}^4 \) of dilute solutions of \( \text{He}^3 \) in \( \text{He}^4 \) at saturated vapor pressure. We have also determined the limiting solubility, the BBP parameter \( \alpha_0 \), the pressure dependence of the \( \text{He}^3 \) reduced mass and the binding energy of a \( \text{He}^3 \) atom at 0°K. Our results seem to be in good agreement with those obtained by other workers, with the exception of the discrepancy between our value of \( \alpha_0 \) and that of Boghosian and Meyer. Because of the importance of this quantity, it is hoped that this disagreement can soon be resolved.

With the rapid advance in the design of dilution refrigerators comes the possibility of carrying these capacitance measurements down to even lower temperatures. It would also be instructive to carry out these measurements at pressures other than the saturated vapor pressure. This will undoubtedly be an interesting piece of work for some clever researchers in the near future.
APPENDIX A

EXPERIMENTAL TIMETABLE

Apparatus at 77°C; 200 microns He⁴ exchange gas in the outer vacuum space, 200 microns He³ exchange gas in the inner vacuum space, a few mm pressure of He⁴ in the He⁴ bath space and 100 microns air in the dewar wall.

9:30 p.m. Bring He⁴ bath to atmospheric pressure with He⁴ gas. Transfer liquid He⁴.

10:30 Apparatus at 4.2°C. Pump inner and outer vacuum spaces and dewar wall overnight.

9:45 a.m. Apparatus still at 4.2°C. Retransfer liquid He⁴.

10:15 Pumping He⁴ bath slowly.

10:45 Begin condensing He³ into He³ reservoir. Let a tiny amount of He³ - He⁴ mixture into the cell for thermal contact.

11:15 He³ all in and reservoir closed off. He⁴ bath now at 1.2°C.

11:30 Take a CMN calibration point. Read the mutual inductance bridge, capacitance bridge, He³ and He⁴ vapor pressures and all carbon thermometers.

12:00 Begin condensing mixture into cell.
3:30 p.m.  Cell full and closed off. (Filling time varies with mixtures.)
Read all bridges and thermometers.

4:00  Position magnet and bring it up to full power.

4:20  Begin pumping He$^3$ reservoir.

5:10  Stop pumping He$^3$ and recondense it into the reservoir. Close off reservoir.

5:20  Stop pumping He$^4$ bath. Retransfer liquid He$^4$. Pump bath down to 1.2°K again.

5:30  Resume pumping on He$^3$ reservoir.

7:30  Begin demagnetization. Decrease magnet voltage to 125 volts.

7:50  Magnet at 65 volts.

8:10  Magnet at 35 volts.

8:30  Magnet at 17 volts.

8:50  Magnet at 7 volts.

9:10  Magnet off. Begin taking data.

1:30 a.m.  Let a few microns of exchange gas into inner and outer vacuum spaces for the final data point at 1.2°K.

1:50  Take another CMN calibration point. Read both bridges, He$^3$ and He$^4$ vapor pressures and all carbon thermometers.

Note: This calibration point always agreed well with the previous one. Slight changes were ascribed to the effects of magnetization and this latter point was the one actually used to compute the final temperatures.
2:00 a.m.    Experiment over.

10:00       Mixture back in sample storage bottle. Check to see that filling line has not blocked. Pump out cell and all traps very well in preparation for the next experiment.
APPENDIX B

THE CHANGE IN MOLAR VOLUME WITH TEMPERATURE

One of the Maxwell equations in thermodynamics gives the derivative of entropy with respect to pressure at constant temperature:

\[
\left( \frac{\partial S}{\partial P} \right)_T = -\left( \frac{\partial V}{\partial T} \right)_P \quad (B-1)
\]

Now assume that the molar entropy of a solution with He\textsuperscript{3} concentration \( X \) is given by:

\[
S = XSF + S_4^0
\]

where \( SF \) is the entropy of one mole of an ideal Fermi gas of the same number density as the solution and \( S_4^0 \) is the entropy of pure He\textsuperscript{4}.

\[
SF = SF\left( \frac{T}{T_F} \right) = SF(\tau) \quad (B-2)
\]

\[
\left( \frac{\partial S}{\partial P} \right)_T = X \frac{dS}{dT} \left( \frac{\partial \tau}{\partial P} \right)_T - \left( \frac{\partial V_4}{\partial T} \right)_P
\]

\[
= X \frac{dS}{dT} \left( \frac{\partial \tau}{\partial T_F} \right)_T \left( \frac{\partial T_F}{\partial P} \right)_T - \left( \frac{\partial V_4}{\partial T} \right)_P
\]

\[
= -X \tau \frac{dS}{d\tau} \left( \frac{1}{T_F} \frac{\partial T_F}{\partial P} \right)_T - \left( \frac{\partial V_4}{\partial T} \right)_P
\]

\[
= -XC_F \left( \frac{\partial \ln T_F}{\partial P} \right)_T - \left( \frac{\partial V_4}{\partial T} \right)_P \quad (B-3)
\]
where $C_F = \tau(dS/dr)$.

Now $T_F = \frac{\hbar^2}{8m^*K} \left[ \frac{3}{\pi} \frac{n_3}{v} \right]^{2/3}$, where $m^*$ is the effective mass of a He$^3$ particle.

\[ \ln T_F = \text{constant} - \ln m^* - \frac{2}{3} \ln v \]  \hspace{1cm} (B-4)

\[ \frac{\partial \ln T_F}{\partial P}_T = 0 - \frac{1}{m^*} \left( \frac{\partial m^*}{\partial P}_T \right)_T - \frac{2}{3} \left( \frac{1}{v} \frac{\partial v}{\partial P}_T \right)_T \]  \hspace{1cm} (B-5)

\[ \text{From Eqs. (B-1) and (B-2),} \]

\[ \left( \frac{\partial v}{\partial T} \right)_P = - \left( \frac{\partial S}{\partial P} \right)_T \]

\[ = XCF \left[ - \frac{2}{3} \left( \frac{1}{v} \frac{\partial v}{\partial P}_T \right)_T - \frac{1}{m^*} \left( \frac{\partial m^*}{\partial P}_T \right)_T \right] + \left( \frac{\partial v_4}{\partial T} \right)_P \]  \hspace{1cm} (B-6)

But $-1/v(\partial v/\partial P)_T$ is just the compressibility $\chi$.

\[ \left( \frac{\partial v}{\partial T} \right)_P = XCF \left[ \frac{2}{3} \chi - \frac{1}{m^*} \left( \frac{\partial m^*}{\partial P}_T \right)_T \right] + \left( \frac{\partial v_4}{\partial T} \right)_P \]  \hspace{1cm} (B-7)

Now the experiments are performed under saturated vapor pressure.

\[ v = v(P, T) \]

\[ \frac{dv(P, T)}{dT} = \left( \frac{\partial v}{\partial P} \right)_T \frac{dP}{dT} + \left( \frac{\partial v}{\partial T} \right)_P \]

\[ = -\chi \frac{dP}{dT} + \left( \frac{\partial v}{\partial T} \right)_P \]  \hspace{1cm} (B-8)

Substituting from Eq. (B-7),

\[ \frac{dv}{dT} = -\chi \frac{dP}{dT} + XCF \left[ \frac{2}{3} \chi - \frac{1}{m^*} \left( \frac{\partial m^*}{\partial P}_T \right)_T \right] + \left( \frac{\partial v_4}{\partial T} \right)_P \]  \hspace{1cm} (B-9)
Assuming \( m^* \) to be independent of \( T \) and integrating from 0 to \( T \) at saturated vapor pressure,

\[
v(X, T) - v(X, 0) = -v^*P_{\text{sat}} + X[U_F(T) - U_F(0)]\left[\frac{2}{3} \nu - \frac{1}{m^*} \frac{\partial m^*}{\partial P}\right]
+ v_4(T) - v_4(0)
\]  

(B-10)

Approximations for \( X \) Small

Let \( \chi \) be expressed as a power series:

\[
\chi(P, T) = \chi_4(P, T)(1 + \alpha \nu X + \ldots X^2 + \ldots)
\]  

(B-11)

Now \( \alpha_{\chi} = \lim_{X \to 0} \left[ \frac{1}{\chi_4} \frac{\partial \chi}{\partial X} \right] \), but

\[
\chi = -\frac{1}{v} \left( \frac{\partial v}{\partial P} \right)_T
\]  

(B-12)

and the molar volume \( v \) can be written as

\[
v = v_4(1 + \alpha X)
\]  

(B-13)

where \( \alpha \) is the BBP parameter:

\[
\alpha = \frac{1}{v_4} \frac{\partial v}{\partial X}
\]  

(B-14)

\[
\frac{\partial \chi}{\partial X} = \frac{1}{v^2} \frac{\partial v}{\partial X} \frac{\partial v}{\partial P} - \frac{1}{v} \frac{\partial^2 v}{\partial P \partial X}
\]

\[
= -\frac{\chi v_4 \alpha}{v} - \frac{1}{v} \frac{\partial}{\partial P} (v_4 \alpha)
\]

\[
= -\frac{\chi v_4 \alpha}{v} - \frac{v_4 \alpha}{v} \frac{\partial \alpha}{\partial P} - \frac{\alpha}{v} \frac{\partial v_4}{\partial P}
\]  

(B-15)

For \( X \) very small this reduces to

\[
\frac{\partial \chi}{\partial X} = -\frac{\partial \alpha_0}{\partial P}
\]  

(B-16)
Using this equation and Eq. (B-11),

\[
\frac{\partial \alpha_x}{\partial X} = \kappa_4 \alpha_x \\
= - \frac{\partial v_4(0)}{\partial P} \frac{\partial \alpha_0}{\partial v_4(0)} \\
= + v_4(0) \kappa_4 \frac{\partial \alpha_0}{\partial v_4(0)}
\]  
(B-17)

\[
\therefore \alpha_x = v_4(0) \frac{\partial \alpha_0}{\partial v_4(0)}
\]  
(B-18)

Now substituting Eq. (B-11) into Eq. (B-10),

\[
v(X, T) - v(X, 0) = -v_x P_{sat} + X[U_F(T) - U_F(0)] \frac{2}{3} \kappa_4 (1 + \alpha_x X) \\
- \left(\frac{1}{m^*} \frac{\partial m^*}{\partial P} \right)_T + v_4(T) - v_4(0) \\
= -v_x P_{sat} + X \kappa_4 [U_F(T) - U_F(0)] \frac{2}{3} (1 + \alpha_x X + \beta^*) \\
+ v_4(T) - v_4(0)
\]  
(B-19)

where

\[
\beta^* = -\frac{1}{\kappa_4} \left(\frac{1}{m^*} \frac{\partial m^*}{\partial P} \right)_T \\
= \frac{v_4(0)}{m^*} \left(\frac{\partial m^*}{\partial v_4(0)} \right)_T
\]  
(B-20)
APPENDIX C

TABULATED RESULTS

For each experiment, there are listed the He\(^3\) concentration of the mixture used, the volume of liquid used and the Fermi temperature of the mixture. For each data point, there are listed the temperature, the reading of the capacitance bridge, the molar volume of the solution without the evaporation correction, the molar volume of the solution with the evaporation correction and the He\(^3\) concentration of the liquid at the capacitor.
### May 17, 1967

**Concentration = 3.92%**

Volume of Liquid = 1.77 cm$^3$

$T_F = 0.276^\circ K$

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<th>$^\circ K$</th>
<th>pF</th>
<th>$v$ without evaporation correction cm$^3$</th>
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### May 27, 1967

**Concentration = 3.92%**

Volume of Liquid = 1.77 cm$^3$

$T_F = 0.276^\circ K$

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<th>$^\circ K$</th>
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May 5, 1967

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Concentration = 6.96%

$T_F = 0.402°K$

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May 29, 1967

Volume of Liquid = 1.79 cm$^3$

Concentration = 6.96%

$T_F = 0.402°K$

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<th>$V$ with evaporation correction (cm$^3$)</th>
<th>$X$</th>
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May 8, 1967

Volume of Liquid = 1.78 cm³

Concentration = 8.10%

\( T_F = 0.444^\circ K \)

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<th>T °K</th>
<th>C pF</th>
<th>V without evaporation correction cm³</th>
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June 5, 1967

Volume of Liquid = 1.57 cm³

Concentration = 8.10%

\( T_F = 0.444^\circ K \)

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<th>V without evaporation correction cm³</th>
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<th>X %</th>
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<tbody>
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May 3, 1967

**Volume of Liquid = 1.69 cm³**

**Concentration = 9.92%**  

<table>
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<th>C (pF)</th>
<th>V without evaporation correction cm³</th>
<th>V with evaporation correction cm³</th>
<th>X %</th>
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</thead>
<tbody>
<tr>
<td>0.0385</td>
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June 2, 1967

**Volume of Liquid = 1.52 cm³**

**Concentration = 9.92%**  

<table>
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<th>C (pF)</th>
<th>V without evaporation correction cm³</th>
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May 12, 1967  
Concentration = 12.02%  
Volume of Liquid = 1.80 cm³  
$T_F = 0.574°K$

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<th>X %</th>
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May 31, 1967  
Concentration = 12.02%  
Volume of Liquid = 1.79 cm³  
$T_F = 0.574°K$

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</table>
May 15, 1967

Volume of Liquid = 1.77 cm$^3$

Concentration = 15.38%  
$T_F = 0.672^\circ K$

<table>
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<th>C (pF)</th>
<th>$V$ without evaporation correction (cm$^3$)</th>
<th>$V$ with evaporation correction (cm$^3$)</th>
<th>X</th>
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June 23, 1967  

Volume of Liquid = 2.30 cm$^3$

Concentration = 15.02%  

$T_F = 0.663^\circ K$

<table>
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<tr>
<th>$T$ °K</th>
<th>$C$ pF</th>
<th>$V$ without evaporation correction cm$^3$</th>
<th>$V$ with evaporation correction cm$^3$</th>
<th>$X$ %</th>
</tr>
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<tbody>
<tr>
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<td>5.35652</td>
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<td>3.6798</td>
<td>14.96</td>
</tr>
<tr>
<td>0.930</td>
<td>35.154</td>
<td>3.6641</td>
<td>3.6663</td>
<td>14.93</td>
</tr>
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<td>1.008</td>
<td>35.163</td>
<td>3.6541</td>
<td>3.6574</td>
<td>14.92</td>
</tr>
<tr>
<td>1.112</td>
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<td>3.6418</td>
<td>3.6451</td>
<td>14.90</td>
</tr>
<tr>
<td>1.211</td>
<td>35.186</td>
<td>3.6262</td>
<td>3.6306</td>
<td>14.85</td>
</tr>
<tr>
<td>1.251</td>
<td>35.195</td>
<td>3.6150</td>
<td>3.6195</td>
<td>14.75</td>
</tr>
</tbody>
</table>
APPENDIX D

He$_3$ - He$_4$ MIXTURE STORAGE SYSTEM

In Table 4 are shown the volumes (up to the Hoke valves) and contents of the storage bottles as of November 1967.

**TABLE 4**

MIXTURE STORAGE BOTTLES

<table>
<thead>
<tr>
<th>Storage Bottle</th>
<th>Mixture Concentration</th>
<th>Pressure (cm Hg)</th>
<th>Volume (liters)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8.10%</td>
<td>43.9</td>
<td>2.955</td>
</tr>
<tr>
<td>2</td>
<td>3.92</td>
<td>49.8</td>
<td>2.891</td>
</tr>
<tr>
<td>3</td>
<td>5.97</td>
<td>65.0</td>
<td>2.891</td>
</tr>
<tr>
<td>4</td>
<td>-</td>
<td>-</td>
<td>0.0879</td>
</tr>
<tr>
<td>5</td>
<td>14.85</td>
<td>69.5</td>
<td>2.973</td>
</tr>
<tr>
<td>6</td>
<td>12.02</td>
<td>68.7</td>
<td>2.889</td>
</tr>
<tr>
<td>7</td>
<td>14.85</td>
<td>69.5</td>
<td>2.889</td>
</tr>
<tr>
<td>8</td>
<td>6.96</td>
<td>63.5</td>
<td>2.889</td>
</tr>
<tr>
<td>9</td>
<td>1.99</td>
<td>59.5</td>
<td>2.472</td>
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<td>6.96</td>
<td>63.5</td>
<td>2.473</td>
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<td>11</td>
<td>9.92</td>
<td>43.2</td>
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<tr>
<td>12</td>
<td>-</td>
<td>-</td>
<td>0.5982</td>
</tr>
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</table>
REFERENCES

1. V. N. Zharkov and V. P. Silin, Zh. Eksperim. i Teor. Fiz. 37, 143 (1959); [English Trans: Soviet Physics JETP 10, 102 (1960)].


28. Epibond 100A is a one-component powdered epoxy adhesive available from Furane Plastics, 16 Spielman Road, Fairfield, New Jersey.


34. The construction of the pills was based on a design developed by Anderson, Salinger and Wheatley (see reference 27).


36. Epibond 121 is a two-component room-temperature-setting epoxy available from Furane Plastics.


41. Zeolite is an adsorbing material in the form of 1/16" pellets. It is available from the Linde Division of Union Carbide.

42. P. P. Craig, Brookhaven National Laboratory (private communication).


52. E. C. Kerr, Phys. Rev. Letters 12, 185 (1964) and private communication.