1379 (1967). Also R. Hakim and A. Mangeney, J. Math. Phys. 9, 116 (1968).

<sup>6</sup>J. Krizan and P. Havas, Phys. Rev. <u>128</u>, 2916 (1962) See also P. Havas, in <u>Statistical Mechanics of Equili-</u> brium and Nonequilibrium, edited by J. Meixner (North-

Holland Publishing Company, Amsterdam, 1965), p. 1. <sup>7</sup>J. Krizan, Phys. Rev. 140, A1155 (1965).

<sup>8</sup>J. Krizan, Phys. Rev. <u>152</u>, 136 (1966).

<sup>9</sup>B. A. Trubnikov, Nucl. Fusion 8, 51, 59 (1968).

<sup>10</sup>V. P. Silin, Zh. Eksperim. i Teor. Fiz. 40, 1768 (1961). [English transl.: Soviet Physics - JETP 13, 1244 (1961)]; Y. L. Klimontovich <u>Statistical Theory</u> of Non-equilibrium Processes in a Plasma, (Moscow University Press, 1964); V. O. Shafranov, Elektromagnithye Volny v Plazme 3, Voprosy Teorii Plazmy, (Gosatomizdat, Moscow, 1963). [English transl.: Reviews of Plasma Physics, edited by M. A. Leontovich (Consultants Bureau, New York, 1967), Vol. 3.]

<sup>11</sup>However, as is known, the relativistic Landau equation does not by itself take collective effects into

account properly.  $^{12}$ E. L. Woolet, Phys. Fluids <u>9</u>, 118 (1966). This

author uses the Darwin interaction, but a reduction to a longitudinal-transverse form is not apparent. <sup>13</sup>L. D. Landau and E. M. Lifshitz, Electrodynamics

of Continuous Media, (Pergamon Press, New York, 1960) p. 251.

<sup>14</sup>R. Balescu, <u>Statistical Mechanics of Charged Par-</u>

ticles, (Interscience Publishers, New York, 1963) p. 223. [For a treatment of nonrelativistic-plasma kinetic theory from the standpoint of the hierarchy-equation theory, see D. C. Montgomery and D. A. Tidman, <u>Plasma</u> <u>Kinetic Theory</u> (Mc Graw-Hill Book Co., <u>New York</u>, 1964)].

<sup>15</sup>See Ref. 14, p. 101.

<sup>16</sup>D. Tidman and T. Dupree, Phys. Fluids <u>8</u>, 1860

(1965). See also T. J. Birmingham, J. M. Dawson and R. M. Kulsrud, Phys. Fluids 9, 2014 (1966).

<sup>17</sup>See Ref. 14, p. 242.

<sup>18</sup>H. Ikegami, H. Ikezi, M. Hosokawa, S. Tanaka, and

K. Takayama, Phys. Rev. Letters <u>19</u>, 778 (1967). Similar structure appears to have been observed by H. Herold, Bull. Amer. Phys. Soc. <u>13</u>, 1489 (1968).

<sup>19</sup>C. C. Lin, SIAM J. Appl. Math. <u>14</u>, 876 (1966).

<sup>20</sup>C. S. Wu, Phys. Fluids 11, 316 (1968).

<sup>21</sup>T. Y. Wu, Kinetic Equations of Gases and Plasmas

(Addison-Wesley Publishing Co., Reading, Mass.

 $1966). \ \ \, This work examines some recent work on the convergence problem.$ 

 $^{22}\mathrm{H.}$  A. Gould and H. E. DeWitt, Phys. Rev. <u>155</u>, 68 (1967).

<sup>23</sup>J. Krizan, Phys. Rev. Letters <u>21</u>, 1162 (1968). Misprints in this paper: a minus sign should be in the last exponent in (1); arguments in (2) and (3) should be labelled with  $\vec{p}_{3}$ , not  $\vec{P}_{13}$ ; Ref. 7 should refer to Ref. 6.

PHYSICAL REVIEW

#### VOLUME 177, NUMBER 1

**5 JANUARY 1969** 

## Number Density and Phase Diagram of Dilute He<sup>3</sup>-He<sup>4</sup> Mixtures at Low Temperatures\*

D. O. Edwards, E. M. Ifft, and R. E. Sarwinski Physics Department, Ohio State University, Columbus, Ohio 43210 (Received 15 July 1968)

The molar volume at the saturated vapor pressure has been determined by a dielectric method for solutions containing up to 15 atomic percent of He<sup>3</sup> in He<sup>4</sup> between 0.025 and 1.2°K. In the single-phase region there is a pronounced thermal expansion from which it is deduced that the derivative of the He<sup>3</sup> effective mass with respect to pressure is for low concentrations  $[(0.0151 \pm 0.0006)m^*]$  atm<sup>-1</sup>. The single-phase measurements also show that at 0°K and for low concentrations, He<sup>3</sup> atoms occupy  $(1.284 \pm 0.005)$  times the volume occupied by He<sup>4</sup>. In the two-phase region, the concentration of the lower phase at 0°K is found to be  $(6.40 \pm 0.07)\%$ He<sup>3</sup>. The He<sup>3</sup> chemical potential along the solubility curve has been obtained and compared with the predictions of Bardeen, Baym, and Pines (BBP). Assuming the effective interaction originally proposed by BBP, the binding energy at 0°K of a single He<sup>3</sup> atom in He<sup>4</sup> is found to be  $L_3^{0} + R(0.284 \pm 0.010)$ °K, where  $L_3^{0}$  is the latent heat of pure He<sup>3</sup> at 0°K.

#### 1. INTRODUCTION

The experiment described here uses measurements of the capacitance of a parallel-plate capacitor immersed in liquid  $He^3$ - $He^4$  solutions to determine their atomic or molar number density under the saturated vapor pressure. The principal results of the experiment are (a) The determination of the limiting solubility of  $He^3$  in  $He^4$ 

at  $0^{\circ}$ K, (b) The determination of the volume occupied by a He<sup>3</sup> atom in liquid He<sup>4</sup> at finite temperatures and at  $0^{\circ}$ K, (c) The determination of the derivative of the He<sup>3</sup> effective mass in solution with respect to pressure. An earlier and less accurate form of the experiment was reported some time ago<sup>1</sup> while a brief account of the present measurements has been published in a Letter.<sup>2</sup> Most of the properties of dilute solutions can be easily understood in terms of the theory of Landau and Pomeranchuk, <sup>3</sup>, <sup>4</sup> which states that  $He^3$  atoms dissolved in superfluid  $He^4$  behave like independent particles with an energy-momentum relation of the form

$$\epsilon = -E_3 + q^2/2m^*. \tag{1}$$

This equation can be regarded as a series expansion in the square of the momentum, q, and it has been amply borne out by numerous experiments. The effective mass  $m^*$  is found from specific heat<sup>5</sup> and second sound<sup>6,7</sup> experiments at the saturated vapor pressure to be 2.4 or 2.5 times the mass of a bare He<sup>3</sup> atom  $m_3$ . The fact that the He<sup>3</sup> "quasiparticles" are fixed in number, as contrasted with the phonons and rotons which disappear at T = 0, means that they form the whole of the normal component at low temperatures and they play a dominant role in the properties of the liquid. Since the He<sup>3</sup> quasi-particles obey Fermi-Dirac statistics and have spin  $\frac{1}{2}$ , they behave like a Fermi gas with degeneracy temperature  $T_F$  given by

$$kT_{F} = (\hbar^{2}/2m^{*})(3\pi^{2}n_{3})^{2/3}, \qquad (2)$$

where  $n_3(P, T, X)$  is the He<sup>3</sup> number density of the solution at P and T with atomic He<sup>3</sup> concentration X. The molar chemical potential of He<sup>3</sup> is given by

$$\mu_{3} = -N_{A}E_{3} + \mu_{F}(T_{F}, T), \qquad (3)$$

where  $N_A$  is Avogadro's number,  $E_3$  is (in the original form of the theory) a constant, and  $\mu_F$  is the chemical potential of an ideal Fermi gas (tabulated by Stoner<sup>8</sup>).

The chemical potential of the solvent  $He^4$  can be obtained from Eq. (3) using the Gibbs-Duhem relation

$$(1-X)\left(\frac{\partial\mu_4}{\partial X}\right)_{P, T} = -X\left(\frac{\partial\mu_3}{\partial X}\right)_{P, T}, \qquad (4)$$

and the fact that at X=0,  $\mu_4$  is just the molar Gibbs function of pure He<sup>4</sup>. The specific heat and entropy of the solution are close to those of an ideal gas below about  $0.5^{\circ}$ K where the effect of the phonons and rotons is small.

The binding energy for one He<sup>3</sup> atom in He<sup>4</sup> at 0°K,  $E_3$  in Eq. (1), can most easily be measured by studying the equilibrium of the liquid mixture with some other phase, for instance, the saturated vapor. The conditions for thermodynamic equilibrium require that the chemical potentials of both isotopes be equal in the two phases. Using the vapor pressure data of Roberts and Sydoriak,<sup>9</sup> Ouboter, Taconis, Le Pair, and Beenakker<sup>10</sup> found values of  $E_3$  which increased slightly with X, and which are close to and a little larger than the latent heat per atom of pure He<sup>3</sup>, denoted by  $L_3^{\circ}$ . They also measured the specific heat down to 0.5°K and found it to correspond to roughly  $\frac{3}{2}R$  per mole of He<sup>3</sup> as predicted by the Pomeranchuk theory.

Edwards and Daunt<sup>11</sup> pointed out that, on the basis of the Landau-Pomeranchuk theory and the

values of  $E_3$  calculated by De Bruyn Ouboter *et al.*, He<sup>3</sup> might still be soluble in He<sup>4</sup> even at  $0^{\circ}$ K, at least up to a certain limiting concentration. In other words, the phase-separation first observed by Walters and Fairbank<sup>12</sup> would not be complete at  $0^{\circ}$ K. At the same time, Cohen and Van Leeuwen<sup>13</sup> calculated the properties of a dilute gaseous mixture of Bose and Fermi hard spheres with properties similar to liquid helium and showed that at 0°K the fermions were soluble up to a concentration of 15%. Measurements of the specific heat and the phase-separation line of helium mixtures by Edwards et al.<sup>14</sup> gave more precise measurements of  $E_3$  and showed the limiting concentration of He<sup>3</sup> at 0°K, namely  $X_0$ , to be about 6%. The specific heat was confirmed to be like that of an ideal Fermi gas with the appropriate degeneracy temperature  $T_F$  (Eq. 2) corresponding to  $m^*$ = 2.5 $m_3$ . Anderson *et al.*<sup>15,5</sup> made accurate measurements of the specific heat, spin susceptibility and spin diffusion coefficient down to very much lower temperatures, of the order of a few millidegrees, and determined the effective mass from the specific heat to be  $(2.38 \pm 0.04)$   $m_3$  at X = 1.3%and  $(2.46 \pm 0.04) m_3$  at X = 5.0%.

The quasi-particle theory of He<sup>3</sup> in solution in superfluid He<sup>4</sup> has been greatly refined and extended by the work of Emery<sup>16</sup> and Bardeen, Baym and Pines (BBP).<sup>17,18</sup> Both Emery and BBP have taken into account the effect of the dissolved He<sup>3</sup> atoms on one another by means of an effective interaction which is fitted to experiment. In these theories, the binding energy  $E_3$  and the mass  $m^*$ in Eq. (1) are independent of concentration and temperature, but dependent on pressure or total density. The apparent small variation of  $E_3$  and  $m^*$  with concentration deduced from phase-separation and specific heat experiments is caused by the use of formulas such as Eq. (3) which neglect the effective interaction. The true values of  $m^*$ and  $E_3$  are therefore those found experimentally as  $X \rightarrow 0$ , namely  $m_0^*$  and  $E_0$ . Bardeen, Baym and Pines have determined the Fourier transform of their effective interaction, defined by

$$V(k) = \int V(r) \exp(-i\vec{k}\cdot\vec{r})d\vec{r}$$

for values of the wave vector k up to about 0.6 Å<sup>-1</sup> by fitting to the low temperature values of the spin diffusion coefficient measured by Anderson *et al.*<sup>5</sup> Ebner<sup>19</sup> has extended their V(k) to higher values of k by fitting to the high temperature  $(T > T_F)$  spin diffusion data. Recently, Baym and Ebner<sup>20</sup> have given another version of V(k) for low k based on a more accurate solution of the Boltzmann equation for spin diffusion and thermal conductivity data.

An important parameter in the BBP theory is the number  $\alpha_0 = \alpha(X=0, T=0)$ , defined so that the molar volume of a solution of concentration X is

$$v(X, T) = [1 + X\alpha(X, T)] v_4(T), \qquad (5)$$

 $v_4$  being the molar volume of pure He<sup>4</sup>. Since

$$\left(\frac{\partial \mu_3}{\partial P}\right)_{X, T} = v + (1 - X) \left(\frac{\partial v}{\partial X}\right)_{P, T}$$

and  $\mu_3 = -N_A E_0$  at 0°K and X = 0, we find that

$$\alpha_0 = -(N_A/v_4)(dE_0/dP)^{-1}$$

According to a theorem of Baym,<sup>21,18</sup>  $\int V(r) d\vec{r} = V(k=0)$  is related to  $\alpha_0$  by the equation

$$-\alpha_0^2 m_4 s^2 v_4(0) = N_A V(k=0), \qquad (6)$$

where  $m_4$  is the mass of He<sup>4</sup> and s is the velocity of sound in pure He<sup>4</sup>. The present experiment provides fairly accurate values of  $\alpha_0$  as well as values of  $\mu_3$  along the phase-separation curve, which may be compared to the BBP theory.

## 2. APPARATUS

The cryostat was designed to cool a cell containing a small quantity of helium of known He<sup>3</sup> concentration by adiabatic demagnetization, and then to allow precise measurements of the capacitance (as a function of temperature) of a parallel plate capacitor immersed in the liquid. From the known atomic polarizability of helium and calibration measurements we could then obtain the number density and molar volume and, when phase-separation had occurred, the concentration of the lower phase.

The cryostat contained two paramagnetic salt pills enclosed in a pair of vacuum jackets. The outer jacket was immersed in a Dewar of liquid He<sup>4</sup> pumped to 1.25°K. The inner vacuum jacket was thermally connected to a pumped reservoir of liquid He<sup>3</sup>, which absorbed the heat of magnetization from the two paramagnetic salt refrigerators and which acted as a thermal guard. The upper salt pill, which also acted as a guard, and the lower salt pill, which was the main refrigerator, were made like those described by Anderson, Salinger, and Wheatley.<sup>22</sup> Each contained about 45 g of chromium potassium alum in the form of 3-mm slabs greased to coil-foil<sup>22</sup> and each having a total area of contact of about 200 cm<sup>2</sup>. Lead thermal switches were inserted between the guard and the  $He^3$  reservoir and between the guard and cooling salt pills. Coil-foil ending in a loose brush of 1300 copper wires connected the cooling salt pill to the liquid in the cell. A general view of the cryostat with some further details of its construction are given in Fig. 1.

The nylon cell, shown in Fig. 2, had a free volume of  $3.4 \text{ cm}^3$  and contained 2.5 g of 1 - mm grains of cerium magnesium nitrate (CMN) in a cylindrical chamber of diameter 1.90 cm and height 1.04 cm. The susceptibility of the CMN, calibrated between 0.65°K and the  $\lambda$  point and measured with an electronic mutual inductance bridge, <sup>23</sup> was used to determine temperature. A filter made from a piece of nylon stocking prevented the CMN from falling into the capacitor which was directly below. Hermetic seals for the brush of cooling wires, the filling tube and the leads to the capacitor were made at the top of the cell by pushing smooth, slightly oversize brass plugs into smooth holes in the nylon. The wires and filling tube were either hard-soldered or soldered with a very small quantity of indium into







FIG. 2. Sample cell.

the plugs. The nylon cap at the bottom of the cell was sealed using liquid phenol.

The filling tube between the cell and the outside of the cryostat had to make contact to the various thermal guards and also to preserve isolation between them and the cell. It was mostly constructed from cupro-nickel tube, 0.0396 cm outside diameter with 0.0076-cm walls made up into spiral coils containing between 30 and 300 cm of tube. Where thermal contact was needed the spirals were bound to coil-foils greased with Apiezon N grease. Where isolation was needed the spirals did not touch the apparatus, and the shape of the coil was maintained by winding the spirals over three strips of cloth and then impregnating the arrangement with Epibond 100A, a high temperature setting epoxy.<sup>24</sup> To minimize the possibility of liquid remaining in the line, thus changing the concentration in the cell. an attempt was made to keep the spirals of the filling tube sloping downwards. The dead volume of the tubing (about 0.65  $cm^3$ ) was fairly small compared to that of the cell.

The parallel-plate capacitor was designed to be as stable and as compact as possible. In addition, since it was placed just below the CMN crystals, it had to be made of non-magnetic material and to produce very little eddy current heating in the field of the magnetic thermometer coils. Two versions were constructed, both with about 5 pF capacitance.

The first version of the capacitor<sup>1</sup> was made of six parallel plates of gold about 3 mm by 7 mm and 0.25 mm apart. The plates were separated and held in place by Epibond 100A which extended in about 1 mm from the short sides of the plates. When cooled to  $0.03^{\circ}$ K with He<sup>4</sup> in the cell for thermal contact, we found this capacitor to have interesting but inconvenient properties. The

capacitance was strongly temperature-dependent. mainly decreasing with temperature but with a minimum at about 0.05°K. In addition, below 0.1°K, the capacitance was voltage dependent. This effect is not fully understood but it was certainly caused by the dielectric properties of the epoxy between the plates. A second version, with two pairs of thin gold plates 0.15 mm apart and with no epoxy between the plates, was constructed as shown in Fig. 3. The epoxy near the ends of the plates was machined away so that the fringing field passed through as little of it as possible. The 0.001-cm thick plates were bonded to their machined Epibond 100A support with Epibond 121, which cures at room temperature.<sup>24</sup> After a number of rapid immersions in liquid nitrogen, the plates showed no sign of wrinkling or pulling away from the form. Each capacitor plate was about 4 mm wide and 11.4 mm long. Electrical contact was made by soldering the plates to 0.13-mmdiameter copper wire with minute amounts of indium. At the top of the cell the leads were soldered to 0.05-mm diameter lead-coated Constantan wire. Except for those inside the cell, the leads were shielded by cupro-nickel tubing inside the cryostat and by coaxial cable outside the cryostat.

The capacitance measurements were made using the three-terminal mode of a General Radio 1615A bridge, so that all capacitances to ground were excluded from the measurement. The bridge was driven by a General Radio 1311A oscillator, operating at 2 kHz with an output of eight volts. At this level and frequency, the measuring apparatus introduced negligible heating and provided sufficient sensitivity. With a General Radio 1232A tuned amplifier and a Princeton Applied Research JB4 phase-sensitive detector, we could easily resolve  $10^{-5}$  pF.

#### 3. PROCEDURE AND DATA ANALYSIS

The concentrations of He<sup>3</sup> in He<sup>4</sup> investigated were 3.92, 6.96, 8.10, 9.92, 12.02, 15.02, and 15.38 at % of He<sup>3</sup>. The samples were made up by mixing the pure gases in accurately measured proportions. Two runs were made with each solution except for the two 15% solutions which were





each run once. Each solution was first cooled by adiabatic demagnetization and then the apparatus was allowed to come into equilibrium for the first point. A heater was used to bring the cell and chrome-alum refrigerator salt up in temperature for successive points. At each point the cell was allowed to come into equilibrium with the cooling salt. The equilibrium time varied from about 10 min at the lowest temperature to 2 min or less about  $0.1^{\circ}$ K.

During the measurements precautions were taken to avoid errors due to liquid He<sup>3</sup> trapped in low spots in the filling tube. Of course, as long as two phases were present the reading of the capacitor did not depend on the amount of He<sup>3</sup> in the cell. As soon as the cell was heated above the phase-separation temperature, the guard salt, He<sup>3</sup> reservoir, and fill line were heated sufficiently to evaporate any trapped liquid and to recondense it in the cell. Sometimes, but not always, a small amount of He<sup>3</sup> was observed to return to the cell, shown by a small decrease in the capacitance reading. The section of the filling tube in contact with the cooling salt and which could not be heated without the cell sloped steeply and was checked to have no low spots. During all measurements in the singlephase region the filling line was kept slightly warmer than the cell. Data were normally taken up to about  $1.2^{\circ}$ K, the temperature of the outer He<sup>4</sup> bath.

For measurements above about 0.7°K a correction is necessary for He<sup>3</sup> vapor in the filling line and the unfilled part of the cell. Since the vapor pressure of He<sup>3</sup> is so much larger than that of He<sup>4</sup> even for very dilute solutions, evaporation results in a depletion of the He<sup>3</sup> in the liquid. Since the volume of liquid used in each run was known and the volume of the cell and filling tube were measured by filling with liquid He<sup>4</sup> at  $1.2^{\circ}$ K, the correction could be calculated, at least in principle, using the vapor pressure data of Sydroiak and Roberts.<sup>9</sup> Unfortunately, the situation was further complicated by temperature gradients in the filling tube and by the refluxing He<sup>4</sup> film which tended to keep flushing He<sup>3</sup> vapor back into the cell. We therefore applied a correction of only one-half that which would be necessary if He<sup>3</sup> vapor occupied the full volume available to it. We assume that this procedure results in an additional uncertainty equal to the applied correction: in other words we assume the true correction to be somewhere between zero and the theoretical equilibrium value. The uncertainty is roughly proportional to the vapor pressure and is appreciable only above  $0.7^{\circ}$ K. At  $1.2^{\circ}$ K it is equivalent to an error of about 1% of X, which for X = 0.15 is equivalent to about 0.01  $\text{cm}^3/\text{mole}$  in the molar volume.

In analyzing the data so as to obtain the molar volume from the measured capacitance, we had to allow for several complicating effects: (a) The capacitance of the empty cell varied from one helium run to the next by amounts varying between  $10^{-3}$  and  $10^{-2}$  pF. Presumably this was due to small movements in the apparatus caused by thermal expansion and contraction, and possibly to small amounts of air and other contaminants.

(b) There was a slight linear dependence on pressure, presumably caused by differential compression of the body of the capacitor and of the leads. (c) Some temperature dependence remained even in the second version of the capacitor due to the fringing field passing through epoxy and other materials. After a fairly lengthy analysis it was shown that the capacitance could, with sufficient accuracy, be represented by

$$C = C_0(T, P, R) + C_1(\epsilon - 1) , \qquad (7)$$

where  $\epsilon$  is the dielectric constant of the liquid helium in the cell;  $C_0$  is a function of the pressure P, the temperature T, and the run R; and  $C_1$  is a constant. The dependence of the dielectric constant of the solution on its density is given by the Clausius-Mossotti equation which is accurate enough for our purpose. The Clausius-Mossotti equation is

$$(\epsilon - 1) = 4\pi n\gamma / (1 - \frac{4}{3}\pi n\gamma) , \qquad (8)$$

where *n* is the number density and  $\gamma$  is the atomic polarizability. For low frequencies<sup>25</sup> we may use  $N_A \gamma = 0.1234 \text{ cm}^3/\text{mole}$  ( $N_A$  is Avogardo's number). So we can now write Eq. (7) in terms of the molar volume  $v(X, T) = N_A/n$  as

$$C = C_0 + \Gamma / [v(X, T) - 0.5169 \text{ cm}^3/\text{mole}]$$
, (9)

where  $\Gamma$  is a constant with dimensions of capacitance times molar volume. By measuring *C* before and after filling the capacitor with pure He<sup>4</sup> at 1.25°K, we determined  $\Gamma$  since the molar volume for pure He<sup>4</sup> is accurately known.<sup>26</sup> Four trials resulted in a value for  $\Gamma$  of 7.09 ±0.02 pF cm<sup>3</sup>/mole.

In using Eq. (7), and hence Eq. (9), we have made two approximations: We have treated  $C_1$  as a constant independent of temperature T, pressure P, and run R; and we have neglected a third term in  $(\epsilon - 1)^2$  which arises because some of the fringing field passes through both liquid helium and epoxy. Both approximations are found to be amply justified on the basis of both theoretical and experimental estimates of the contribution to Cfrom the fringing field and the leads. As will become clearer from the further description of the procedure given below, in all the experiments we are only concerned with small differences or changes in  $(\epsilon - 1)$  corresponding at most to the difference between pure  $He^4$  and the solution. For the highest concentration used in the present experiments,  $X \simeq 0.15$ , this difference in  $\epsilon$  is ~ 0.0025 corresponding to a change in molar volume of ~1.1 cm<sup>3</sup>/mole. An error of 0.3% in the constant  $C_1$  in Eq. (7) or  $\Gamma$  in Eq. (9) then results in an error of ~ 0.003  $\text{cm}^3/\text{mole}$  in the molar volume of the solution relative to that of pure He<sup>4</sup>.

Since the changes in  $C_0$  due to pressure and temperature and from one experiment to another are all small, we can write

$$C_0 = C_0' + C_0(T) + C_0(P) + C_0(R) , \qquad (10)$$

 $C_0(T)$  is arbitrarily defined to be zero at 0.05°K,

 $C_0(P)$  to be zero at P = 0 and  $C_0(R)$  to be zero for an arbitrarily chosen run. The temperature dependence  $C_0(T)$  was determined when the cell was filled with pure He<sup>4</sup>. After correcting for the known<sup>26</sup> thermal expansion of He<sup>4</sup> the results are shown in Fig. 4. The term  $C_0(P)$  was obtained by filling the cell with pure  $He^4$  at 1.25°K and applying pressure up to 1.6 atmospheres. After taking into account the change in the density of He<sup>4</sup> with pressure, the data fell on a line of slope - 1.16×10<sup>-5</sup> pF/mm Hg. The term  $C_0(R)$  represents zero shifts of the capacitor from run to run. For all but the two runs with X = 0.0392,  $C_0(R)$  was determined by adjusting the data to agree at  $0.07^{\circ}$ K. Here all the samples except those with X = 0.0392 were in the two-phase state, and would have given the same capacitance if the shift from one run to another had not occurred. The values of  $C_0(R)$  for the X = 0.0392 runs were determined by requiring their data to give the correct molar volume at 0.69°K, as explained below.

To establish the value of  $C_0'$ , a constant for all experiments, and the value of  $C_0(R)$  for the two low-concentration runs, a different set of measurements was performed which may be described as the "filling experiment." Successive small amounts of He<sup>3</sup> were added to a measured amount of pure  $He^4$  in the cell, changing X from zero to about 0.15. The capacitance was measured for each concentration at 0.69°K where the vapor pressure of the mixture was small and the equilibrium times were reasonably short. Using the known<sup>26</sup> molar volume of pure He<sup>4</sup> at 0.69°K (27.581 cm<sup>3</sup>/mole) and the value of  $\Gamma$  determined above, the molar volume for each concentration was found from Eq. (9). Since the temperature was fixed and the vapor pressure quite low,  $C_0$ was treated as a constant which was adjusted to fit the molar volume of pure He<sup>4</sup>. The results are best displayed in terms of the BBP parameter of Eq. (5),  $\alpha(X, T)$  at  $T = 0.69^{\circ}$ K, which is shown in Fig. 5. The error bars in Fig. 5 are calculated from the uncertainty in the amount of He<sup>3</sup> added. The temperature in this experiment was maintained by the He<sup>3</sup> refrigerator with He<sup>3</sup> exchange gas in the inner jacket. It was therefore impossible to heat the parts of the filling tube in



FIG. 4. Temperature dependence of the first (old) and second (new) versions of the capacitor  $C_0(T)$ .



FIG. 5. Experimental values of  $\alpha(X, T)$  versus X at 0.69°K from the "filling experiment."

contact with the guard salt and  $He^3$  reservoir above the temperature of the cell, as was done in the demagnetization experiments. On the other hand it is improbable that there was any error caused by liquid, rich in  $He^3$ , being caught at low points in the filling tube. If this had occurred one would expect that the first point in Fig. 5 would be much more seriously affected than the others since during this first point  $He^3$  was admitted to the filling tube for the first time. Secondly, sufficient time was allowed for each point (up to one hour) so that any liquid in the tube had time to reach the same concentration as that in the cell by the action of the fillm and of the vapor.

The straight line in Fig. 5 corresponds to

v(T = 0.69, X)

 $= 27.581(1 + 0.267X + 0.018X^2) \text{ cm}^3/\text{mole.}$  (11)

With the aid of Eq. (11) we were able to fix the value of  $C_0'$  by fitting to one of the 12% runs at 0.69°K. All the other runs in which phase-separation occurred then gave values of v(T = 0.69, X) which agreed with Eq. (11) to within  $\pm 0.002 \text{ cm}^2/\text{mole}$ . The values of  $C_0(R)$  for the two 4% runs were determined similarly. A filling experiment was also performed at 1.27°K which agreed fairly well with the other data, but in view of the uncertainties in the vapor correction it was decided not to use this experiment in the data analysis.

#### 4. RESULTS

The results of the experiment with the vapor corrections applied are shown in Fig. 6 as a graph of molar volume versus temperature. The over-all accuracy, below  $0.7^{\circ}$ K, is estimated to be about  $\pm 0.005$  cm<sup>3</sup>/mole.

As shown in Fig. 6 there is a pronounced dependence of the volume on temperature in the single-phase region. This thermal contraction, which is approximately proportional to X and T, may be easily understood<sup>1</sup> in terms of the Pomeranchuk theory discussed in the Introduction. The molar entropy of a solution with He<sup>3</sup> concentration X is quite accurately given by



FIG. 6. The molar volume of dilute solutions of  $He^3$  in  $He^4$  at the saturated vapor pressure.

$$S = XS_{F} + S_{A} , \qquad (12)$$

where  $S_F$  is the entropy of one mole of an ideal Fermi gas of the same number density as the solution and with an effective mass,  $m^*(X)$ , which depends on concentration, and where  $S_4$  is the entropy of pure He<sup>4</sup>. Then from

$$\left(\frac{dv}{dT}\right)_{\text{sat}} = -v\kappa \left(\frac{dP}{dT}\right)_{\text{sat}} + \left(\frac{\partial v}{\partial T}\right)_{P},$$

and the Maxwell equation

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

we obtain

$$\left(\frac{dv}{dT}\right)_{sat} = -v\kappa(X)\left(\frac{dP}{dT}\right)_{sat} + XC_F\left(\frac{2}{3}\kappa(X)\right)$$
$$-\frac{1}{m^*(X)} \frac{\partial m^*(X)}{\partial P} + \left(\frac{\partial v_4}{\partial T}\right)_P, \qquad (13)$$

where we have assumed  $m^*(X)$  to be independent of temperature but not of concentration. Integrating and neglecting the partial pressure of He<sup>4</sup> and the rather small dependence of  $\kappa$  on T,

$$\begin{aligned} v(X, T) &- v(X, 0) = -v\kappa(X)P_{\text{sat}} \\ &+ X[U_F(T) - U_F(0)] \left(\frac{2}{3}\kappa(X) - \frac{1}{m^*} \frac{\partial m^*}{\partial P}\right) + v_4(T) - v_4(0), \end{aligned} \tag{14}$$

where  $U_F(T)$  is the molar internal energy of an ideal Fermi gas, tabulated by Stoner.<sup>8</sup> To sufficient accuracy we may write

$$\kappa(X) = \kappa_A (1 + \alpha_{\nu} X) \ .$$

Here  $\kappa_4$  is the compressibility of pure He<sup>4</sup> and

$$\alpha_{\kappa} = \frac{1}{\kappa_4} \frac{\partial \kappa}{\partial X} = \frac{-1}{\kappa_4} \frac{\partial \alpha_0}{\partial P} = v_4(0) \frac{\partial \alpha_0}{\partial v_4(0)} , \qquad (15)$$

where  $v_4(0)$  is the molar volume of liquid He<sup>4</sup> at 0°K (a function of the pressure) and  $\alpha_0$  was defined in the Introduction in connection with Eq. (5). Numerically  $\alpha_{\kappa} = 1.4$  (Refs. 27 and 28). If we also define the number

$$\beta^*(X) = -\frac{1}{\kappa_4 m^*(X)} \frac{\partial m^*(X)}{\partial P} , \qquad (16)$$

Eq. (14) can be put in the form

$$v(X, T) - v(X, 0) = X\kappa_{4} [U_{F}(T) - U_{F}(0)]$$
$$\times [\frac{2}{3}(1 + \alpha_{\kappa}X) + \beta^{*}(X)] + v_{4}(T) - v_{4}(0) - v\kappa P_{sat}.(17)$$

Part of the effect predicted by Eq. (17) has a simple physical explanation: The leading term is, omitting the contribution proportional to  $\beta^*$ , simply the expansion of the liquid helium due to the increase in the pressure of the quasiparticle gas  $\frac{2}{3}[U_F(T) - U_F(0)]/v$ . The contribution involving  $\beta^*$ , which is larger and of opposite sign arises because the quasiparticle mass  $m^*$  depends on the density of the solution. The solvent He<sup>4</sup> caused by phonon and roton excitation and the third term, which is very small, is the compression due to the saturated vapor pressure.

To compare Eq. (17) with experiment we have calculated a volume  $v_A$  given by

$$v_A(X, T) = v(X, T) - v_4(T) + v_4(0) + v\kappa P_{\text{sat}}$$
 (18)

The expansion of He<sup>4</sup> was taken from Kerr and Taylor, <sup>26</sup>  $v\kappa$  is an average value of the volume times the compressibility<sup>29</sup> and  $P_{\text{sat}}$  was taken from Sydoriak and Roberts.<sup>9</sup> In Fig. 7 we have plotted  $v_A$  versus  $X[U_F(T) - U_F(0)]/R$ , which according to Eq. (17) should be a straight line for a given concentration with a slope  $\kappa_4[\frac{2}{3}(1 + \alpha_K X) + \beta^*(X)]$  depending only slightly on X. As the figure shows this is well satisfied. In plotting the graph we have used the values of  $m^*(X)$  calculated by Radebaugh<sup>30</sup> from the BBP theory, and the tables of  $U_F$  given by Stoner.<sup>8</sup> The values of  $\frac{2}{3}(1 + \alpha_K X) + \beta^*(X)$  from the slopes and the compressibility<sup>29</sup> of pure He<sup>4</sup>,  $\kappa_4 = 1.212 \times 10^{-8} \text{ cm}^2/$ 



FIG. 7. The thermal expansion of dilute solutions compared with Eq. (17). The quantity  $v_A$  is defined by Eq. (18).

dyne, are plotted in Fig. 8. From the straight line drawn on this graph, using  $\alpha_{\kappa} = 1.4$ , <sup>27,28</sup> we obtain at the saturated vapor pressure

$$\beta^* = -(1.25 \pm 0.05) + (0.6 \pm 0.3)X.$$
 (19)

Figure 8 also includes a point from molar volume



FIG. 8. The dimensionless number  $[\frac{2}{3}(1 + \alpha_{K}X) + \beta^{*}]$  versus concentration. Present data  $\bigcirc$ , Kerr (Ref. 31)  $\square$ , Sandiford and Fairbank (Ref. 6)  $\triangle$ .

measurements by Kerr<sup>31</sup> on a 27% solution, treated in the same way as the present data, and a point estimated from the variation of  $m^*$  with pressure from the second sound measurements by Sandiford and Fairbank.<sup>6</sup> From  $\beta^*$  and Eq. (16) we can determine the pressure dependence of the effective mass,

$$\frac{1}{m^*} \frac{\partial m^*}{\partial P} = 10^{-2} [(1.51 \pm 0.06) - (0.7 \pm 0.4)X] \text{ atm}^{-1}.$$
 (20)

Extrapolation of the straight lines of Fig. 7 to zero gives the molar volume of the mixture at  $0^{\circ}$ K, namely v(X, 0). Thus we can find  $\alpha(X, 0)$  for each mixture from

 $\alpha(X, 0) = [v(X, 0) - v_4(0)] / X v_4(0),$ 

where  $v_4(0) = 27.5793$  cm<sup>3</sup>/mole according to Kerr and Taylor.<sup>26</sup> These results are shown in Fig. 9. If we express  $\alpha(X, 0)$  as

$$\alpha(X, 0) = \alpha_0 + \alpha_0' X + \cdots, \qquad (21)$$



FIG. 9. The "BBP parameter" at 0°K,  $\alpha_0$ , versus He<sup>3</sup> concentration. Present work  $\bigcirc$ , Kerr and Taylor (Ref. 32) **a**, Kerr (Ref. 31)  $\Box$ , Boghosian and Meyer (Ref. 28)  $\triangle$ .

2.0

the data may be fitted by either  $\alpha_0 = (0.284 \pm 0.005)$ ,  $\alpha'_0 = 0$  or by  $\alpha_0 = (0.280 \pm 0.005)$ ,  $\alpha'_0 = 0.55$ . While the first pair of values fit our experimental points a little better, the second pair fitted Kerr's data<sup>31</sup> for X = 0.273 and 0.533 and the value<sup>32</sup> for pure He<sup>3</sup> very well. We may therefore write

$$v(X, 0) = 27.5793 (1 + 0.280X + 0.55X^2)$$
  
cm<sup>3</sup>/mole,  $0 \le X \le 1$ . (22)

Unfortunately our values of  $\alpha(X, 0)$  are well below the one obtained by Boghosian and Meyer<sup>28</sup> from measurements above 1°K on a 6% solution. At present there is no explanation for this discrepancy, which is just outside the combined experimental error of the two experiments.

Finally, we would like to point out that the theoretical and empirical Eqs. (17), (18), and (22), together with the numerical values we have quoted, provide a method of calculating the empirical molar volume for any temperature and concentration in the range we have discussed. More simply, Eq. (22) may be used with the values of v(X, T) - v(X, 0) calculated from Eqs. (17) and (19) and given in Table I. A useful equation, valid for small X, is

$$\alpha(X \rightarrow 0, T) = (0.284 \pm 0.005)$$
  
- [(0.032 ± 0.003) deg<sup>-1</sup>]T

This is obtained from Eqs. (5), (17) and (19) using  $U_F(T) - U_F(0) = \frac{3}{2}RT$  and neglecting the effect of the He<sup>3</sup> vapor pressure and other small terms.

To determine the solubility curve, that is the value of X in the lower phase  $X^{l}(T)$  in the two phase region, the values of v below the phase-separation temperature  $T_{s}$  were corrected to 0°K by means of Eq. (17), using a first approximation to  $X^{l}$ . We then used

$$X = [v(X, 0) - v_4(0)] / \alpha_0 v_4(0)$$

with  $\alpha_0 = 0.284$  to obtain a second approximation to  $X^I$ , and so on. The same procedure was also used on the data above  $T_S$  to check the consistency of the method. The results are, shown in the phase diagram of Fig. 10. It may be seen that in the single-phase region the points reproduce the known concentrations of the samples very well. The accuracy of the phase diagram is really independent of the accuracy of the filling experiment which is only necessary for determining  $\alpha_0$ . The calculation of  $X^I$  via the molar volume can be regarded merely as a way of fitting concentrations



FIG. 10. The solubility curve for  $He^3$  in  $He^4$  at the saturated vapor pressure.

to the capacitance readings. Since the low temperature portion of the solubility curve is of special interest, we have plotted  $X^{l}$  versus  $T^{2}$  for this region. Below 0.15°K, the data fit the empirical equation,

$$X^{l} = X_{0} [1 + 10.8(^{\circ} \text{K})^{-2} T^{2}], \qquad (23)$$

where  $X_0$ , the solubility at 0°K is  $(6.40 \pm 0.07)\%$ (slightly different from our original estimate in Ref. 2). This is in excellent agreement with the value  $X_0 = (6.35 \pm 0.30)\%$  deduced by Vilches and Wheatley<sup>33</sup> from the performance of a dilution refrigerator. A set of smoothed values for the concentration  $X^{\bar{l}}$  and the molar volume  $v^{\bar{l}}$  along the solubility curve is given in Table II.

In Fig. 11 we compare the present measurements with other low temperature determinations of the solubility curve. As the figure shows we are in excellent agreement with the points from the specific heat<sup>14</sup> except for the specific heat point at X=0.0992 which is below the present

TABLE I. Change of molar volume with temperature, calculated from Eq. (17), in cm<sup>3</sup>/mole.

<b>Т</b> (°К) <u>Х</u>	0.2	0.4	0.6	0.8	1.0	1.2
0.00	0.000	0.000	+0.001	+0.003	+0.006	+0.007
0.04	-0.004	-0.011	-0.017	-0.022	-0.026	-0.032
0.08	-0.007	-0.019	-0.031	-0.042	-0.053	-0.066
0.12	- (0.009)	-0.025	-0.043	-0.060	-0.077	-0.097
0.16	- (0.010)	-0.031	-0.053	-0.076	-0.098	-0.124

Т (° К)	x <sup>l</sup> (%)	$v(x^{l}, T)$ (cm <sup>3</sup> /mole)	$(\mu_3 + L_3^{0})/R$ (° K)
0.00	6.40	28.079	$0.000 \pm 0.000$
0.03	6.45	$28.08_{3}$	-0.001
0.05	6.55	$28.09_2$	-0.003
0.07	6.71	$28.10_4$	-0.006
0.09	6.92	$28.12_{1}$	-0.010
0.10	7.05	$28.13_2$	$-0.013 \pm 0.001$
0.128	7.5	$28.16_{6}$	-0.021
0.153	8.0	28.201	$-0.030 \pm 0.002$
0.175	8.5	$28.24_4$	-0.038
0.195	9.0	28.281	$-0.046 \pm 0.003$
0.213	9.5	$28.31_4$	-0.055
0.230	10.0	$28.35_{0}$	-0.064
0.246	10.5	$28.39_{5}$	-0.073
0.262	11.0	$28.43_4$	-0.082
0.278	11.5	$28.47_{6}$	-0.091
0.291	12.0	$28.50_8$	-0.099
0.304	12.5	$28.54_{6}$	$-0.108 \pm 0.004$
0.317	13.0	$28.58_3$	-0.117
0.329	13.5	$28.62_0$	-0.126
0.341	14.0	$28.65_{5}$	-0.135
0.353	14.5	28.695	$-0.144 \pm 0.005$
0.365	15.0	28.73 <sub>0</sub>	-0.154

curve. There is some reason to believe that this last point is incorrect due to errors in temperature measurement; it was taken with a somewhat different experimental arrangement from the other points.<sup>34</sup> We also agree with the original NMR measurements of Walters and Fairbank,<sup>12</sup> recalculated using the appropriate Fermi-Dirac expression for the spin susceptibility, verified experimentally by Husa *et al.*<sup>35</sup> and Anderson *et al.*<sup>5</sup> On the other hand, our values of  $X^{l}$  are all below those recently measured by Schermer, Passell



FIG. 11. Comparison between the present solubility curve (full line) and other measurements. Neutron absorption (Ref. 36), dashed line; Walters and Fairbank (Ref. 12)  $\triangle$ ; specific heat data (Ref. 14)  $\Box$ .

and Rorer<sup>36</sup> using neutron absorption (the dashed line) by an amount which is almost constant at 0.0050. This must reflect a systematic error in either or both of the experiments.

In the two-phase system the chemical potential of the He<sup>3</sup> in the upper concentrated phase must equal that of the He<sup>3</sup> in the lower dilute phase,

$$\mu_3^l = \mu_3^u.$$

The upper phase is almost pure He<sup>3</sup> so that  $\mu_3^{u}$  is the Gibbs function of pure He<sup>3</sup>  $G_3(T)$ , with a small correction for the dissolved He<sup>4</sup>:

$$\mu_3^{\ u} = G_3(T) + RT \ln X^{\mathcal{U}}, \quad T \leq 0.4^{\circ} \mathrm{K},$$

from which

...

$$\mu_{3}^{u} + L_{3}^{0} = -\int_{0}^{T} S_{3} dT + RT \ln X^{u}, \quad T \leq 0.4^{\circ} \text{K}, \quad (24)$$

where  $L_3^0$  is the latent heat of pure He<sup>3</sup> at 0°K and  $S_3$  is the entropy of pure He<sup>3</sup>. The correction term has been discussed by Edwards and Daunt,<sup>11</sup> who have also given a semi-empirical formula for the concentration  $X^{\mathcal{U}}(T)$  in the upper phase:

$$1 - X^{u} = 1.13T^{3/2} \exp(-0.71^{\circ} K/T), \quad T \le 0.4^{\circ} K.$$
 (25)

Although in the derivation of Eq. (25) it was assumed that  $X^{\tilde{l}} \rightarrow 0$  at 0°K, it is nevertheless in good agreement with the available experimental data,<sup>10,37-39</sup> shown in Fig. 12. The largest uncertainty in  $\mu_3^{u} + L_3^{0}$  occurs in  $S_3$  for which we have used the empirical equation of Anderson, Reese, and Wheatley,<sup>40</sup> which is in fair agreement with later experiments.<sup>41</sup> This uncertainty is shown in Table II and in Fig. 13 which give  $\mu_3^{u} + L_3^{0}$  calculated from Eqs. (24) and (25).

Figure 13 also shows values of  $\mu_3^{l}(T)$  calculated from the present measurements of  $X^{l}(T)$  and the appropriate equation of the BBP theory as extended by Ebner.<sup>42</sup>



FIG. 12. Comparison between the equation of Edwards and Daunt (Ref. 11) Eq. (25), and measurements of the concentration in the upper phase  $X^{\mathcal{U}}(T)$ . Zinov'eva and Peshkov (Ref. 37)  $\diamond$ , De Bruyn Ouboter *et al.* (Ref. 10)  $\nabla$ , Brewer and Keyston (Ref. 38)  $\Box$ , Keyston and LaHeurte (Ref. 39)  $\triangle$ .



FIG. 13. The  $He^3$  chemical potential in the two-phase region as a function of temperature. The dotted curve shows the results calculated from the theory of Eq. (26). At the top of the graph is shown the degree of degeneracy of the dilute phase along the solubility curve,  $T/T_F$ versus T.

$$\begin{split} & \mu_3/N_A = -\,E_0 + \mu_{F0}/N_A + n_3 V(0) + \frac{1}{2} n_3 V(0) \\ & \times \big[F(n_3) + \frac{1}{12} \pi^2 (T/T_{F0})^2 G(n_3)\big] \;, \; T \leq \frac{1}{2} \, T_F \;, \; (26) \end{split}$$

\*Work supported by the National Science Foundation. <sup>1</sup>E. M. Ifft, M. M. Skertic, and D. O. Edwards, Bull. Am. Phys. Soc. 12, 96 (1967).

- <sup>2</sup>E. M. Ifft, D. O. Edwards, R. E. Sarwinski, and
- M. M. Skertic, Phys. Rev. Letters 19, 831 (1967).
- <sup>3</sup>L. D. Landau and I. Pomeranchuk, Dokl. Akad. Nauk. SSSR 59, 669 (1948).
- <sup>4</sup>I. Pomeranchuk, Zh. Eksperim. i Teor. Fiz. <u>19</u>, 42 (1949).
- <sup>5</sup>A. C. Anderson, D. O. Edwards, W. R. Roach,
- R. E. Sarwinski, and J. C. Wheatley, Phys. Rev. Letters <u>17</u>, 367 (1966). <sup>6</sup>D. J. Sandiford and H. A. Fairbank, Phys. Rev. <u>162</u>,
- 192 (1967).

<sup>'</sup>C. G. Niels-Hakkenberg, L. Meermans, and H. C. Kramers, in Proceedings of the Eighth International Conference on Low Temperature Physics, London, 1962, edited by R. O. Davies (Butterworths Scientific Publications Ltd., London, 1963), p. 45.

- <sup>8</sup>E. C. Stoner, Phil. Mag. <u>28</u>, 257 (1939)

where  $\mu_{F0}(T, T_{F0})$  is the chemical potential at temperature *T* for an ideal Fermi gas with number density  $n_3$  and concentration-independent mass<sup>18</sup>  $m_3^* = 2.34 m_3^5$ . To evaluate Eq. (26) we have used the present molar volumes to calculate the number density  $n_{2}(X, T)$ , and the value of V(0) given by Eq. (6) using our value of  $\alpha_0 = 0.284$ . The dimensionless functions F and G have been taken from Ebner's thesis, the value of  $E_0$  has been adjusted to agree with experiment at T = 0.

The agreement between the experimental values and theory shown in Fig. 13 is good and could be made better by a slightly higher value of V(0). Using Ebner's or the original BBP V(k),  $(E_0$  $-L_3^{0}/R$  is determined to be 0.284 ± 0.010° K, where  $L_3^{0}$  is the latent heat of pure He<sup>3</sup> at 0°K,  $L_3^{0}/R$  $= (2.47 \pm 0.01)^{\circ} K$  (Ref. 43). On the other hand there is a good deal of uncertainty as to the real values of V(0), F, and G. The value of  $N_A V(0)$ that we have used is  $-0.0807 m_4 s^2 v_4$ , as compared with the original choice of BBP  $-0.0754 m_4 s^2 v_4$ , and  $-0.0815m_4s^2v_4$  as used by Ebner. The function F and G depend on the dependence of V(k) on k which Ebner obtained from a fit to the low and high temperature spin diffusion coefficient D. Unfortunately the validity of the equation used for the low temperature D has been put in doubt by Baym and Ebner<sup>20</sup> who have proposed an alternative V(k) which is on the average weaker than the original one. Baym and Ebner's V(k) does not obey Eq. (6) with experimental values of  $\alpha_0$ . In view of these difficulties the present agreement or disagreement between theory and experiment must not be taken too seriously.

#### ACKNOWLEDGMENTS

The authors gratefully acknowledge the considerable experimental help given by Dr. M. M. Skertic in the early part of this investigation. We are grateful to W. E. Baker, L. Wilkes, and R. L. Kindler for their technical assistance.

<sup>9</sup>S. G. Sydoriak and T. R. Roberts, Phys. Rev. 118, 901 (1960)

- <sup>10</sup>R. De Bruyn Ouboter, K. W. Taconis, C. Le Pair,
- and J. J. M. Beenakker, Physica 26, 853 (1960).
- <sup>11</sup>D. O. Edwards and J. G. Daunt, Phys. Rev. <u>124</u>, 640 (1961).
- $^{12}\mathrm{G.}$  K. Walters and W. M. Fairbank, Phys. Rev. 103, 262 (1956).
- $^{13}\mathrm{E.}$  G. D. Cohen and J. M. J. Van Leeuwen, Physica
- 27, 1157 (1961). See also J. M. J. Van Leeuwen and
- E. G. D. Cohen, Phys. Letters <u>26A</u>, 89 (1967).
- <sup>14</sup>D. O. Edwards, D. F. Brewer, P. Seligmann, M. Skertic, and M. Yaqub, Phys. Rev. Letters 15, 773 (1965)
- <sup>15</sup>A. C. Anderson, W. R. Roach, R. E. Sarwinski, and
- J. C. Wheatley, Phys. Rev. Letters 16, 263 (1966). <sup>16</sup>V. J. Emery, Phys. Rev. <u>148</u>, 138 (1966); and <u>161</u>,
- 194 (1967).
- <sup>17</sup>J. Bardeen, G. Baym, and D. Pines, Phys. Rev. Letters 17, 372 (1966).

<sup>18</sup>J. Bardeen, G. Baym, and D. Pines, Phys. Rev. <u>156</u>, 207 (1967)

<sup>19</sup>C. Ebner, Phys. Rev. 156, 222 (1967).

<sup>20</sup>G. Baym and C. Ebner, Phys. Rev. <u>170</u>, 346 (1968).

"G. Baym and C. Ebner, Phys. Rev. 170, 346 (1968).

<sup>22</sup>A. C. Anderson, G. L. Salinger, and J. C. Wheatley, Rev. Sci. Instr. 32, 1110 (1961).

<sup>23</sup>W. L. Pillinger, P. S. Jastram, and J. G. Daunt, Rev. Sci. Instr. 29, 159 (1958).

<sup>24</sup>Furane Plastics, 16 Spielman Road, Fairfield, New Jersey.

<sup>25</sup>See, for instance, K. R. Atkins, Liquid Helium (Cambridge University Press, Cambridge, England, 1959), p. 40.

<sup>26</sup>E. C. Kerr and R. D. Taylor, Ann. Phys. (N. Y.) 26, 292 (1964).

<sup>27</sup>W. E. Massey and C. W. Woo, Phys. Rev. Letters 19, 301 (1967). <sup>28</sup>C. Boghosian and H. Meyer, Phys. Letters <u>25A</u>, 352

(1967).

<sup>29</sup>W. M. Whitney and C. E. Chase, Phys. Rev. 158, 200 (1967).

<sup>30</sup>R. Radebaugh, National Bureau of Standards Technical Note No. 362 (U.S. Government Printing Office,

Washington, D. C., 1967).

<sup>31</sup>E. C. Kerr, Phys. Rev. Letters <u>12</u>, 185 (1964), and private communication. We are very grateful to Dr.

Kerr for this data in advance of publication.

<sup>32</sup>E. C. Kerr and R. D. Taylor, Ann. Phys. (N. Y.) 20, 450 (1962).

<sup>33</sup>O. E. Vilches and J. C. Wheatley, Phys. Letters <u>24A</u>, 440 (1967); and 25A, 344 (1967).

<sup>34</sup>M. M. Skertic, Ph. D. dissertation, Ohio State University, 1966 (unpublished).

<sup>35</sup>D. L. Husa, D. O. Edwards, and J. R. Gaines, Phys. Letters 21, 28 (1966).

<sup>36</sup>R. I. Schermer, L. Passell, and D. C. Rorer, Phys. Rev. 173, 277 (1968). We are very grateful to these

authors for a copy of their paper before publication. <sup>37</sup>K. N. Zinov'eva and V. P. Peshkov, Zh. Eksperim. i Teor. Fiz. 37, 33 (1959) [English Transl.: Soviet

Phys. --JETP 10, 22 (1960)].

<sup>38</sup>D. F. Brewer and J. R. G. Keyston, Phys. Letters 1, 5 (1962). <sup>39</sup>J. R. G. Keyston and J. P. Laheurte, Phys. Letters

24A, 132 (1967).

<sup>40</sup>A. C. Anderson, W. Reese, and J. C. Wheatley, Phys. Rev. 130, 495 (1963).

<sup>41</sup>W. R. Abel, A. C. Anderson, W. C. Black, and

J. C. Wheatley, Phys. Rev. <u>147</u>, 111 (1966).

- <sup>42</sup>C. Ebner, Ph.D. dissertation, University of Illinois, 1967 (unpublished).
- <sup>43</sup>T. R. Roberts, R. H. Sherman, and S. G. Sydoriak, J. Res. Nat. Bur. Std. 68A, 567 (1964).

PHYSICAL REVIEW

#### VOLUME 177, NUMBER 1

5 JANUARY 1969

# Finite-Temperature Corrections to the Transport Coefficients of a Normal Fermi Liquid\*

C. J. Pethick<sup>†</sup>

Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois

(Received 29 July 1968)

Finite-temperature corrections to the limiting low-temperature behavior of the thermal conductivity and spin-diffusion coefficient of a normal Fermi liquid are calculated using a quasiparticle Boltzmann equation which is solved by the variational method. For liquid He<sup>3</sup> the most important corrections come from small momentum transfer processes, the amplitude for which can be determined exactly by using Landau theory. The expressions obtained are evaluated in detail, assuming that all Landau parameters vanish for  $l \ge 2$ , and comparison of theory with experiment yields an estimate for the previously undetermined Landau parameter  $F_1^{a}$ . The calculations are compared with calculations based on paramagnon theory.

## 1. INTRODUCTION

Although the limiting low-temperature behavior of the specific heat, staticresponse functions, and transport coefficients were predicted over 10 years ago on the basis of Landau's theory of normal Fermi liquids, <sup>1</sup> it is only recently that the properties of a Fermi liquid at finite temperatures have been studied theoretically. The main stimulus to recent theoretical work was the experimental observation<sup>2</sup> that even at 50  $m^{\circ}K$  the specific heat of liquid He<sup>3</sup> showed appreciable deviations from the linear temperature dependence predicted by

Landau theory. Doniach and Engelsberg<sup>3</sup> were able to account for the observed specific-heat data by using a model in which persistent spin fluctuations play an important role. They found that the specific heat calculated using their model had a contribution of order  $T^3 \ln T$  as well as the term linear in T predicted by Landau theory. Other calculations of the  $T^3 \ln T$  term in the specific heat have been performed by Brenig and Mikeska<sup>4</sup> and Amit, Kane, and Wagner<sup>5</sup> using Landau theory, by Brenig, Mikeska, and Riedel<sup>6</sup> and Brinkman and Engelsberg<sup>7</sup> using the random-phase approximation, and by Riedel<sup>8</sup> using the shielded potential