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THE DIFFUSION OF $^4$HE IN LIQUID $^3$HE IN THE FERMI LIQUID REGION

DISSERTATION

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

By

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***

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ABSTRACT

This thesis describes measurements of the diffusion coefficient $D$ of $^4$He in liquid $^3$He below 0.1 K, where Fermi liquid theory applies. Theoretically dissolved $^4$He has a unique property: its transport properties, e.g. $D$, are predicted using only thermodynamic properties, e.g. the solubility $X_{^4He}^{sat}$ of liquid $^4$He in $^3$He and known properties of $^3$He.

Theoretically, we first derive $D$ in the low temperature limit and then consider finite temperatures by expanding the scattering amplitude in the $^4$He and $^3$He momenta. Experimentally, a helium mixture is confined in a bellows with so little $^4$He that a bulk $^4$He-rich phase cannot form. Instead a superfluid $^4$He-rich film coats the cell walls and is monitored by two parallel-plate capacitors. A sudden change in pressure changes the amount of dissolved $^4$He and the film changes as $^4$He enters or leaves the bulk. The time dependence of the thickness gives $D$ and the equilibrium thickness gives $X_{^4He}^{sat}$.

The apparatus is also described, including a new dilution refrigerator, a hydraulic cold valve, and the cell. Then difficulties in the analysis are discussed, including the nature of a solid-like layer of $^4$He adsorbed on the cell walls, the possibility of convective $^4$He transport, frictional heating and a non-linear boundary condition of the diffusion equation. Finally the data is analyzed to show that the measured value of $D$ agrees with the theoretical prediction.
Dedicated to the memory of Hanan Baddar
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I want to thank my advisor, Prof David Edwards, for his patience and help throughout the numerous crises and difficulties that came along with this project. I also must thank him for teaching me a great deal about physics and sharing his unbounded enthusiasm for the field.

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CHAPTER 1

INTRODUCTION

This thesis describes experiments that measured the diffusion coefficient $D$ of $^4\text{He}$ in liquid $^3\text{He}$ below 0.1 K, where Fermi liquid theory is applicable. This work was part of a close collaboration between myself and Jizhong He that involved measuring both $D$ and the solubility $X_{^4\text{He}}^{\text{sat}}$ of $^4\text{He}$ in $^3\text{He}$ in order to test a prediction based on Fermi liquid theory. My thesis emphasizes the diffusion coefficient, while He's thesis[14] emphasizes the solubility. Additional work on this project, especially in the analysis of the data, was done by Hans Hjort and will be described in his thesis[15].

Figure 1.1 is the phase diagram of liquid mixtures of the two stable isotopes of helium, $^4\text{He}$ and $^3\text{He}$. Below the lambda line, $^4\text{He}$-rich solutions are superfluid; they can flow with no viscosity. In the two phase region, mixtures of the isotopes separate into two distinct phases. The $^3\text{He}$-rich phase is non-superfluid and is called the upper phase since it is less dense than the $^4\text{He}$-rich phase and therefore floats on top of it. The $^4\text{He}$-rich phase is called the lower phase and is superfluid. At $T = 0$, the concentration of $^3\text{He}$ in the lower phase is 6.4% at $P = 0[16]$, however, the amount of $^4\text{He}$ that can dissolve in the upper phase decreases exponentially with temperature. In fact, we find that at $T = 0.05$ K and $P = 0$, the saturated solubility is only 2 ppm. This has made experimental work in this region difficult; excepting publications from

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Figure 1.1: The phase diagram of liquid $^3\text{He}-^4\text{He}$ mixtures. The horizontal axis is the $^3\text{He}$ concentration. This thesis describes diffusion of $^4\text{He}$ in the $^3\text{He}$-rich phase below 0.1 K. in the lower left hand corner of this diagram.

our group, the only other experimental work on the $^4\text{He}$ in the upper phase in the Fermi liquid region is by Nakamura's group, who measured the solubility at $P = 0[7]$. Solutions of $^4\text{He}$ in liquid $^3\text{He}$ are predicted to have a unique property: the transport properties of the dissolved $^4\text{He}$ can be predicted from the thermodynamic properties of the $^4\text{He}$ and host $^3\text{He}$. In particular, the diffusion coefficient $D$ and thermal diffusion ratio $k_T$ can be predicted from $m_4^*$ and $v_4^*$, the effective mass and partial volume of the dissolved $^4\text{He}$. These values are found by measuring $X_4^{\text{sat}}(P, T)$ as a
function of pressure $P$ and temperature $T$. Thus, measurements of $D$ and $X_{4}^{stat}(P, T)$ provide a test of Fermi liquid theory in this system.

The reason this is possible is that the scattering between the $^4$He and $^3$He quasi-particles, at low temperatures, is "forward scattering." As seen in the center of mass frame, they are deflected very little. This is because the $^4$He obey Bose statistics and have energies near $k_B T$, while the $^3$He obey Fermi statistics and have energies near the Fermi energy, $k_B T_F$. Here $k_B$ is Boltzmann constant and $T_F$ is the Fermi temperature of pure $^3$He, which is about 1.77 K at $P = 0$[17]. In the lab frame, the $^3$He have momenta near the Fermi momentum $(2m_3^* k_B T_F)^{1/2}$, while the $^4$He momentum is much smaller, about $(m_4^* k_B T)^{1/2}$. In addition, the Pauli principle allows the $^3$He to scatter only into states with energies within about $k_B T$ of the Fermi energy. This makes the momentum transfer small compared to the $^3$He momentum.

Saam and Laheurte[18, 19] derived an expression for the forward scattering amplitude $a_0^{34}$ between $^4$He and $^3$He that contains only thermodynamic properties. If the scattering were not forward scattering, then Saam's expression would not provide enough information to calculate a transport property. Higher order terms that could not be expressed in terms of thermodynamic properties would then be necessary, as is the case for pure $^3$He, where two colliding quasiparticles can scatter through a large angle.

The $l = 0$ forward scattering amplitude found[18, 19] by Saam and Laheurte is

$$a_0^{34} = \frac{1}{\nu(0) v_3} = \frac{2}{3} v_4^* k_B T_F,$$  \hfill (1.1)

where $\nu(0)$ is the density of states at $T = 0$ and $v_3$ is the molar volume of pure $^3$He. The scattering amplitude is also related[18, 9] to the $s = 0$ part of the Landau function $f_0^{34}$ by $a_0^{34} = f_0^{34} / (1 + F_0^s)$. The Landau function $f_0^{34}$ is the functional derivative of
the energy with respect to the $^3\text{He}$ and $^4\text{He}$ distributions and $F_0^*$ is the symmetric Fermi liquid factor.

In section 2.1, which is a transcription of a Physical Review Letter that I co-wrote[20], we make use of eq. (1.1) and solve the Boltzmann equation in the $T \to 0$ limit to find $D$ and the thermal diffusion ratio $k_T$. The result is

$$D = (0.4461 \ldots) \frac{\hbar}{m_*^4} \frac{T_F}{T} \frac{m_3^*}{m_4^*} \left(\frac{v_3^*}{v_4^*}\right)^2. \quad (1.2)$$

where $m_3^*$ is the effective mass of the $^3\text{He}$ quasiparticles. Finite temperature corrections to this expression are in section 2.2 and are derived in Hans Hjort's thesis[15] and ref. [21].

To determine $m_4^*$ and $v_4^*$ we study the solubility $X_4^{\text{sat}}$ of $^4\text{He}$ in $^3\text{He}$. By equating the chemical potentials in the upper and lower phases, one finds the expression [19, 9],

$$X_4^{\text{sat}} = \left(\frac{T}{T^*}\right)^{3/2} \exp\left[-\left(\frac{\Delta + \pi_3^{\text{sat}} v_4^*}{T}\right)\right]. \quad (1.3)$$

where

$$T^* = \frac{2 \pi \hbar^2}{m_4^* v_3^{3/2} k_B}. \quad (1.4)$$

and $\Delta$ is the difference in binding energy of a $^4\text{He}$ atom in the upper phase $E_4$ and the binding energy in pure $^4\text{He}$ $L_4$.

$$\Delta = (L_4 - E_4)/k_B. \quad (1.5)$$

Here $\pi_3^{\text{sat}}$ is the osmotic pressure of the $^3\text{He}$ in the lower phase. The product $\pi_3^{\text{sat}}$ times $v_4$, the pure $^4\text{He}$ molar volume, corrects $L_4$ for the $^3\text{He}$ dissolved in the lower phase. The partial volume of a dissolved $^4\text{He}$ atom in $^3\text{He}$ is $v_4^* = -dE_4/dP$. Since $L_4$ and the osmotic pressure are known, by fitting measurements of $X_4^{\text{sat}}(P, T)$ one can determine the values of $m_4^*$ and $v_4^*$ that go into the expression for $D$. We also
measure $D$ directly. Comparing the predicted and measured values of $D$ provide the test of Fermi liquid theory.

The concentration is so small that it is impractical to measure it directly. Instead we confine an unsaturated mixture in the cell and use two parallel plate capacitors to measure the thickness of a superfluid $^4\text{He}$-rich film that forms on the walls of the container. The thickness of this film changes when the solubility in the bulk $^3\text{He}$-rich mixture changes. The solubility is measured by observing the equilibrium change in film thickness as the temperature is changed at constant pressure. The diffusion coefficient is measured by observing the time dependence of the film thickness after sudden change in pressure.

Chapter 3 provides details of the apparatus used to measure both $D$ and $X_i^{\text{sat}}$. It describes the installation of a new dilution refrigerator along with its new wiring and gas handling system. The design of a hydraulic cold valve and the experimental cell are also in chapter 3 and it concludes by talking about the hardware and software used to collect the data.

Much of the information needed to analyze the data is in chapter 4. This includes a discussion of the behavior of the immobile $^4\text{He}$ adsorbed on a substrate, in our case the walls of the cell. Next I describe the characterization the cell's surface, and then the behavior of the capacitance ratio when the $^4\text{He}$ was removed from the cell. Finally the possibility of convective transport as opposed to diffusive transport is considered.

Chapter 5 explains the experimental method and data analysis. It also summarizes the solubility measurements, which are needed to help analyze the data and compare it to theory. Finally, in chapter 6 the results for $D$ are presented. Although the uncertainties in both the experimental data and the parameters derived from $X_i^{\text{sat}}$
are larger than we would have liked, we found good agreement with the predictions from Fermi liquid theory.

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The diffusion coefficient $D$ and thermal diffusion ratio $k_T$ for $^4$He in liquid $^3$He at low $T$ have been calculated by solving the Boltzmann equation derived from Fermi-liquid theory. As $T \to 0$, $k_T$ approaches a constant and $D$ varies as $1/T$: and both are determined solely by $a_0^{34}$, the $\ell = 0$ forward scattering amplitude. Since $a_0^{34}$ can be found from thermodynamics, a strict test of the theory can be made without any assumptions about the dependence of the scattering amplitude on the momentum transfer, making $^4$He in $^3$He unique among applications of Fermi-liquid theory.

PACS numbers: 67.60.Dm, 51.10.+y, 67.55.Lf, 67.55.Hc.

In this Letter we present the solution of the Boltzmann transport equation of Zharkov and Silin [22] for the diffusion of dilute $^4$He in non-superfluid liquid $^3$He at low temperature. As the temperature $T$ and the $^4$He mass concentration $c$ tend to zero, the diffusion coefficient $D$ is proportional to $1/T$. We have discovered that the product $DT$ tends to a value determined solely by $a_0^{34}$, the $\ell = 0$ forward scattering
amplitude [19, 18] which can be obtained from thermodynamic data. Moreover the thermal diffusion ratio $k_T$ is simply given by $(0.3823\ldots c)$ and independent of the value of $a_0^{34}$. Thus a measurement of $k_T/c$, or $DT$ and some thermodynamic data, permit a strict quantitative test of the underlying Fermi-liquid theory. In this respect, $^4$He in $^3$He is qualitatively different from other applications of Fermi-liquid theory. In pure $^3$He or $^3$He in liquid $^4$He for example, simplifying assumptions about the dependence of the scattering amplitude on the momentum transfer are needed to relate the kinetic coefficients to the thermodynamic properties [23]. In $^4$He in $^3$He, the momentum dependence appears only in terms of order $T/T_F$. The $^3$He Fermi temperature $T_F$ is about 1.77 K at zero pressure.

To test the theory, the experiments must be performed in the temperature range where the solvent liquid $^3$He obeys Fermi-liquid theory, i.e. below 0.1 K and above the $^3$He superfluid transition. In this range the solubility of $^4$He in $^3$He, $c^{\text{sat}}(T)$ is very small. However recent measurements of $c^{\text{sat}}$ at zero pressure and below 0.1 K by Nakamura et al. [7] indicate that such experiments are, in fact, feasible. At higher pressure, near 10 atm., $c^{\text{sat}}$ may become substantially larger [24]. A way to measure $D$ and $k_T$ is to modify the technique used by Nakamura et al. By measuring the thickness of the superfluid $^4$He film on the walls of the container, one could find the rate at which $^4$He diffuses in or out of the liquid after a sudden change of pressure or temperature.

Fermi-liquid theory was first applied to solutions of $^4$He in liquid $^3$He by Zharkov and Silin [22] who estimated $D$ and $k_T$ and set up the $^4$He and $^3$He Boltzmann equations. They considered two situations: (A) the dilute limit where $^4$He quasiparticles are mainly scattered by $^3$He, and (B) the concentrated limit where $^4$He-$^4$He scattering
is dominant. In ordinary circumstances $c_{\text{sat}}$ decreases so rapidly with decreasing $T$ that case (B) cannot be observed. However, recent experiments [25] indicate that $^4\text{He}-^3\text{He}$ mixtures in a confining medium, aerogel, may enter region (B). In (B), the diffusing $^4\text{He}$ quasiparticle distribution is close to internal equilibrium but it drifts with respect to the $^3\text{He}$. According to Zharkov and Silin, $D$ in this region is still determined by the $^3\text{He}-^4\text{He}$ scattering amplitude, and it is likely that the low temperature limit of $D$ is determined solely by $a_{34}^2$, just as in region (A). In this Letter we consider only the dilute limit, region (A).

Zharkov and Silin also considered two possibilities for the $^4\text{He}$ quasiparticle energy: the conventional, particle-like spectrum, $E = -E_{43} + q^2/2m_4^*$, and a roton-like, "bubble spectrum", $E = -E_{43} + (q - q_0)^2/2m_4^*$. Here $E_{43}$ is the binding energy of one $^4\text{He}$ atom in liquid $^3\text{He}$ in the ground state. A recent microscopic calculation [10] indicates that the conventional spectrum is the appropriate one, with an effective mass ratio $m_4^*/m_4 \approx 1.21$ at zero pressure. This agrees with a fit [26] to the data of Nakamura et al. [7] which gives $m_4^*/m_4 = 1.1^{+0.4}_{-0.1}$. We consider only the conventional spectrum.

The Zharkov-Silin result for $D$ in the dilute limit with the conventional spectrum was independent of $T$ due to an incorrect approximation in the collision integral. This was pointed out by Leggett and ter Haar [27] who showed that, as $T \to 0$, $D$ should vary as $1/T$. Calculations of the product $DT$ were made by Dandache et al. [28] and Geilikman and Chechetkin [29], again with simplifying assumptions [30] about the collision integral. Their results and ours may be expressed as

$$D = D_0 \frac{\hbar}{m_4^*} \frac{T_F}{m_4} \frac{m_3^*}{m_4} \frac{1}{(1 + a_0^2)^2}$$  \hspace{1cm} (2.1)
where $D_0$ is some dimensionless number and $m_3^*$ is the $^3\text{He}$ quasiparticle effective mass. The $^3\text{He}$ Fermi temperature is given by $k_B T_F = \frac{p_F^2}{2m_3^*}$ and $(1 + \alpha_4^0) v_3$ is the partial volume of a $^4\text{He}$ atom in liquid $^3\text{He}$ of atomic volume $v_3$.

Equation (2.1) makes use of the relation between $a_0^{34}$ and the thermodynamic properties derived by Saam and Laheurte [19, 18]:

$$a_0^{34} = \frac{(1 + \alpha_4^0)}{\nu(0)}. \tag{2.2}$$

Here, $\nu(0) = \frac{3}{(2 v_3 k_B T_F)}$ is the $^3\text{He}$ density of states. The forward scattering amplitude $a_0^{34}$ is equal to $|\nu_0|$ used in previous work [22, 28, 29].

Estimates of $D_0$ and $k_T$ vary widely; they are shown in Table 2.1. Our $D_0$ leads to a diffusion coefficient $D \approx 5.0 \times 10^{-3} \, \text{cm}^2/\text{s}$ at 0.1 K and zero pressure. In Eq. (2.1) we assumed $\alpha_4^0 \approx -0.34$ at zero pressure, as estimated in Ref. [26] from the data of Laheurte [31]. The predicted $D$ and, for comparison, the thermal diffusivity of liquid $^3\text{He}$ are shown in Fig. 2.1. The measurements of $D$ in the figure are those by Vvedenskii and Peshkov [1] at the freezing pressure with $c$ between $\sim 10$ and $\sim 40\%$. Equation (2.1) predicts only a slight decrease in $D$ with increasing pressure. Since the theory breaks down above 0.1 K, the disagreement between theory and experiment is not significant.

For Fermi-liquid theory to be valid for $^4\text{He}$ in $^3\text{He}$, the $^4\text{He}$ quasiparticle states (as well as the $^3\text{He}$ states) must be well defined. This condition can be written as $\hbar / t \ll k_B T$ where $t$ is the $^4\text{He}$ quasiparticle lifetime or collision time. Following Zharkov and Silin, we define $t$ by $D = 2 k_B T t/(3 m_4)$, which gives $t \propto T^{-2}$. The criterion for validity of the theory becomes $(3/2) m_4 D / \hbar \gg 1$. We find this ratio to be about 50 at 0.1 K.
Table 2.1: Values of $D_0$ and $k_T$ from various authors. The values of $D_0$ from Refs. [11] and [12] were found by replacing $|v_0|$ by $a_0^{34}$ and using Eqs. (2.1) and (2.2). Refs. [11] and [12] did not use Eq. (2.6) for the $^4$He mass current, resulting in the factors of $m_4^*/m_4$ in $D_0$.

<table>
<thead>
<tr>
<th>Source</th>
<th>$D_0$</th>
<th>$k_T$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref. [28]</td>
<td>$\frac{60}{\pi} \frac{m_4^*}{m_4} \approx 21$</td>
<td>2c</td>
</tr>
<tr>
<td>Ref. [29]</td>
<td>$\sim \frac{2}{\pi^4} \frac{m_4^*}{m_4} \frac{m_4}{m_3} \approx 0.01$</td>
<td>–</td>
</tr>
<tr>
<td>Present Work</td>
<td>0.4461...</td>
<td>(0.3823...)c</td>
</tr>
</tbody>
</table>

Figure 2.1: Logarithmic plot of $D$, the diffusion coefficient of $^4$He in liquid $^3$He, versus the temperature $T$. The full line is the $1/T$ prediction for zero pressure; the dashed line is for 25 atm. The lines are dotted where the Fermi-liquid theory is no longer valid, above 0.1 K. The data points were measured by Vvedenskii and Peshkov Ref. [1], at the freezing pressure. The curves are the thermal diffusivity of liquid $^3$He at zero pressure and 25 atm. from Ref. [2].
While a momentum-independent interaction $\nu_{\alpha} \delta(\mathbf{r})$ between the $^4$He and $^3$He quasiparticles was assumed in earlier work [22, 28, 29], we consider the effect of a more general momentum-dependent scattering amplitude $a^{34}(\mathbf{p}, \mathbf{q}, \mathbf{k})$. Here $\mathbf{p}$ and $\mathbf{q}$ are the momenta of the incoming $^3$He and $^4$He quasiparticles, and $\mathbf{k}$ is the momentum transfer $\mathbf{k} = \mathbf{p}' - \mathbf{p} = \mathbf{q} - \mathbf{q}'$, where $\mathbf{p}'$ and $\mathbf{q}'$ are the final $^3$He and $^4$He momenta. The scattering amplitude $a^{34}(\mathbf{p}, \mathbf{q}, \mathbf{k})$ may be expanded as a constant $a_0^{34}$ plus terms proportional to scalar combinations of the momenta, such as $k^2$, $q^2$, $p^2 - p_F^2$, $\mathbf{p} \cdot \mathbf{q}$, etc. The momentum dependent terms appear only in the temperature dependence of $DT$ and $k_T$ and not in the $T \to 0$ limits, which depend only on the constant term $a_0^{34}$. The reason for this is clear. Since the $^4$He quasiparticles obey Boltzmann statistics, the average values of $q$, $q'$ and $k$ vary as $T^{\frac{1}{2}}$. Thus terms in the interaction proportional to $q$ and $k$ vanish as $T \to 0$. In addition, the variation in $p$ becomes very small at low $T$. A detailed analysis [21] shows that $DT$ and $k_T$ should vary as $1 + O(T/T_F)$. The terms in $a^{34}$ that depend on $\mathbf{p}$, $\mathbf{q}$, and $\mathbf{k}$ appear only in the $T/T_F$ part. Therefore as $T \to 0$ we need to include only $a_0^{34}$ in the Boltzmann equation.

In setting up and solving the Boltzmann equation, we follow the pioneering work of Refs. [22], [27] and [28], using nearly the same notation. The $^4$He Boltzmann equation in the dilute limit (no $^4$He-$^4$He scattering) for small temperature and concentration gradients and with zero total mass current, $\rho v = 0$, is [22]

$$n_{i0} \frac{\mathbf{q}}{m_4^*} \cdot \left[ \frac{\mathbf{c}}{c} + \left( \frac{q^2}{2 m_4^* k_B T} - \frac{3}{2} \right) \frac{\mathbf{c}}{T} \right] = J_{if}. $$

Here $n_{i0}(q)$ is the equilibrium Maxwell-Boltzmann $^4$He quasiparticle occupation number. The collision integral is [22, 28]
\[ J_{ij} = \frac{(2\pi/\hbar)}{\int |a_0^{34}|^2 n_{f0} (1 - n_{f0}^{'}) n_{io} (\psi_i - \psi_i^{'}) \times \delta(\epsilon + E - \epsilon' - E') 2\, dp\, dq'/\hbar^6. \]

In agreement with Ref. [22], we have a factor of 2 multiplying \( dp \) for the sum over the initial \(^3\)He spin states. Refs. [28] and [29] have a factor of 4. This implies that a \(^3\)He quasiparticle may change its spin orientation during a \(^4\)He-\(^3\)He collision, which we think is incorrect.

In the collision integral, \( p' \) is related to the other momenta by conservation of momentum. The Fermi functions \( n_{f0} \) and \( n_{f0}^{'} \) are the equilibrium occupation numbers for the initial and final \(^3\)He states, with energies \( \epsilon \) and \( \epsilon' \). The initial and final \(^4\)He energies are \( E \) and \( E' \), so that the delta function \( \delta(\epsilon + E - \epsilon' - E') \) enforces conservation of energy. The quantities \( \psi_i = -\delta n_i/n_{io} \) and \( \psi_i^{'} = -\delta n_i^{'}/n_{io}^{'} \) are the small fractional deviations from the equilibrium \(^4\)He distribution function. Because they are linear in the concentration and temperature gradients, \( \psi_i \) and \( \psi_i^{'} \) depend on \( q \) and \( q^{'} \) according to the equation [22],

\[ \psi_i = a_c(q) \mathbf{q} \cdot \mathbf{\nabla} c + a_T(q) \mathbf{q} \cdot \mathbf{\nabla} T. \]

The functions \( a_c(q) \) and \( a_T(q) \) depend only on the magnitude of \( q \) and are found by solving the Boltzmann equation.

The deviations from equilibrium of the \(^3\)He occupation number are assumed to be negligible in \( J_{ij} \) because, in the dilute limit, the number of \(^3\)He at the Fermi surface and available for scattering is much larger than the number of \(^4\)He.

To find \( J_{ij} \), we first integrate over \( p \) and then over \( q' \). The integral over \( p \) depends only on the momentum transfer \( k = q - q' \) and the energy transfer \( E - E' \).
We choose an axis along \( k \) and write \( dp = 2\pi p^2 dp d\cos \theta \). Integrating over \( \cos \theta \), the delta function requires that the collision takes place with the angle \( \theta \) between \( p \) and \( k \) given by:

\[
\cos \theta = \frac{m_3^*(E - E') - k^2/2}{pk}.
\] (2.3)

The right hand side of this equation is of the order of \( q/p \) or \((T/T_F)^{1/2}\). This means that, in most cases, the momentum transfer \( k \) is very nearly perpendicular to \( p \).

From (2.3), the condition \(-1 \leq \cos \theta \leq 1\) gives the lower limit for the integral over \( p \), namely \( p \geq |m_3^*(E - E')/k - k/2| \). At sufficiently low temperature, this limit may be replaced by zero. The integral with respect to \( p \) is then found using

\[
\int_{-\infty}^{\infty} n(s)[1 - n(s + \Delta)] ds = \frac{\Delta}{(1 - e^{-\Delta})}
\] (2.4)

where \( \Delta = (E - E')/k_B T \), \( n(s) = (e^s + 1)^{-1} \) and \( s = (p^2 - p_F^2)/(2 m_3^* k_B T) \).

Since the integrand in (2.4) is always positive, using \( p = 0 \) or \( s = -\infty \) instead of the true lower limit for \( p \) overestimates the effect of collisions with large \( k \). Large \( k \) corresponds to large negative values of \( \Delta \), because \( q \) is small at low \( T \) (due to the Boltzmann distribution) and therefore \( q' \) must be large if \( k \) is large. Because the collision rate, which is proportional to (2.4), is very small when \( \Delta \ll -1 \), the approximation is self consistent. The true lower limit for \( p \) is of the order of \((2 m_3^* k_B T)^{1/2}\). hence it has an exponentially small effect on the integral. When calculated, the error is found to be \( \sim O(1) \exp(-T_F/T) \). This is negligible compared to \( \sim T/T_F \), the error due to the momentum dependence of the scattering amplitude.

After integrating over \( p \) using (2.4), the integration over the direction of \( q' \) is elementary and \( J_{ij} \) reduces to the one-dimensional form

\[
J_{ij} = C_0 \int F(q, q') \frac{\Delta}{(1 - e^{-\Delta})^2} q'dq'
\] (2.5)
where, when $\nabla T = 0$,
\[
F(q, q') = q' \left[ a_c(q) - \frac{1}{3} (q'/q)^2 a_c(q') \right] \quad q' \leq q,
\]
\[
= q \left[ a_c(q) - \frac{1}{3} a_c(q') \right] \quad q' \geq q.
\]
and
\[
C_0 = 2 n_0 |m_3^* a_0^{24}|^2 k_B T \hat{q} \cdot \nabla c/(h^4 h^2).
\]

Using (2.5), the Boltzmann equations for the two situations, $\nabla T = 0$ or $\nabla c = 0$, were simplified by writing them in the dimensionless variables $x \equiv q^2/(2 m_4^* k_B T)$ and $y \equiv q'^2/(2 m_4^* k_B T)$. The resulting integral equations were solved in terms of the variables $t$ and $t'$ defined by $t = \exp(-x/g)$ and $t' = \exp(-y/g)$, where $g$ is a dimensionless scale factor. The equations were discretized over a one-dimensional lattice of $N$ points evenly spaced in $t$. As a result they each became $N$ algebraic equations which were solved by iteration. An initial approximate form was used for $a_c(y)$, giving a new estimate for $a_c(x)$ and so on. Accurate solutions were obtained with $N = 100$ or 200, iterating up to 15 times. The solutions $a_c(x)$ and $a_T(x)$ are shown in Fig. 2.2.

After solving the Boltzmann equation, $D$ and $k_T$ were found from the $^4$He mass current
\[
i = -\rho D \left[ \nabla c + (k_T/T) \nabla T \right]
\]
\[
= m_4 \int (q/m_4^*) \delta n, dq/h^3. \tag{2.6}
\]

Here $q/m_4^* = \nabla_q E$ is the $^4$He quasiparticle velocity. Previous authors [22, 28] used $\int q \delta n, dq$, the momentum density associated with the $^4$He, to obtain $i$. However this includes a contribution from entrained $^3$He.

In summary, dilute $^4$He in liquid $^3$He has a simple and unambiguous relation between the low temperature kinetic coefficients, $D$ and $k_T$, and the thermodynamic
Figure 2.2: The solutions of the Boltzmann equation for an isothermal concentration gradient (full curve), and for a temperature gradient at constant concentration (dashed). The fractional deviations from the equilibrium $^4\text{He}$ distribution function, $a_c(x)$ and $a_T(x)$ in arbitrary units, are plotted against the reduced $^4\text{He}$ quasiparticle momentum, $x^{1/2} = q/(2m^*_T k_BT)^{1/2}$. The equilibrium Maxwell-Boltzmann distribution is proportional to $xe^{-x}$, shown as the dotted curve.

properties. The relation for $D$ is Eq. (2.1) with $D_0 = 0.4461$ while $k_T$ is simply $0.3823c$. If the experimental difficulties in making measurements at low temperatures, where the solubility of $^4\text{He}$ approaches a few ppm [7, 24, 26], can be overcome, one has a unique opportunity to test Fermi-liquid theory. One needs to measure $D(T)$, $m^*_T$, and the ratio $1 + \alpha^0_T$, which is related to the pressure variation of the binding energy $dE_{43}/dP$. Measurement of $k_T/c$ provides a further strict test of the theory.

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2.2 Finite Temperature Effects in the Theory of Diffusion

The theory of the effects of finite temperature on $D$ and the thermal diffusion ratio $k_T$ has been published by Hjort, Culman, Edwards and He[21] and will be discussed in Hans Hjort's thesis[15]. A summary of the results are given here. The scattering amplitude of a collision between $^3$He and $^4$He quasiparticles is expanded in the mean momenta of the quasiparticles. The $^3$He and $^4$He quasiparticles have momenta $p$ and $q$ respectively before the collision and they scatter into states with momenta $p'$ and $q'$. The momentum transfer is $k = p' - p = q - q'$. The mean momenta are $q_m = q - k/2$ and $p_m = p + k/2$. The inverse collision must have the same amplitude as the forward collision. This requires the scattering amplitude to have the following symmetry.

$$|a^{34}(p_m, q_m, k)| = |a^{34}(p_m, q_m, -k)|. \quad (2.7)$$

With this in mind, the scattering amplitude is expanded to second order in the small quantities $q_m$ and $k$.

$$|a^{34}| = a_0^{34} \left[ 1 + \alpha_1 \left( \frac{p_m^2}{p_F^2} - 1 \right) + \alpha_2 \frac{p_m \cdot q_m}{p_F^2} + \alpha_3 \frac{q_m^2}{p_F^2} + \alpha_4 \frac{(p_m \cdot q_m)^2}{p_F^4} + \ldots \right. \nonumber$$

$$\left. + \alpha_5 \frac{k^2}{p_F^2} + \alpha_6 \frac{(p_m \cdot k)^2}{p_F^4} + \ldots \right]. \quad (2.8)$$

Some of the coefficients of the expansion can be determined using Galilean invariance and invariance with respect to the reference liquid.

$$a_0^{34} = \frac{2}{3} k_B T_F v_4^*,$$

$$\alpha_2 = 3 \frac{v_3}{v_4} \left( 1 - \frac{m_4}{m_4^*} \right),$$

$$\alpha_3 = \frac{3}{2} \frac{v_3}{v_4^*} \frac{m_4^*}{m_4^*} \frac{1}{(1 + F_0^*)} \frac{d \log m_4^*}{d \log v_3}. \quad (2.9)$$

The rest of the $\alpha_i$'s are undetermined. Inserting eq. (2.8) for the scattering amplitude into the collision integral and solving the Boltzmann equation using a method similar
that in ref. [21], gives,

\[
D = \frac{\hbar}{m_4 T} \left( \frac{v_3}{v_1} \right)^2 \left[ 0.4461 \frac{m_3^2}{m_4^2} - \left( \frac{T}{T_F} \right) (2.127 \gamma_1 + 1.479 \gamma_2 + 0.337 \gamma_3) \right].
\]  

(2.10)

The parameters \( \gamma_i \) are given by,

\[
\gamma_1 = 2\alpha_5 - \frac{\alpha_1}{2},
\gamma_2 = \frac{\alpha_3}{2} + \frac{\alpha_4}{4} + \frac{\alpha_2^2}{8},
\gamma_3 = \frac{\alpha_2 m_3^2}{2 m_4^2} - \frac{\alpha_4}{4} - \frac{\alpha_2^2}{8}.
\]  

(2.11)

While \( \alpha_2 \) and \( \alpha_3 \) are both less than one, the value of \( \alpha_5 \), which has the largest coefficient in \( \gamma_1 \), may be large. It might give a significant contribution to \( D \) in the region where the experiments were performed. It may be so large that higher order terms are needed in \( a^{34} \).

An oversight in the calculation of \( k_T \) in chapter 2.1 was corrected in ref. [21]. In section 2.1, the effect of the temperature gradients on the \( ^3 \)He quasiparticle distribution was ignored. When this effect is included in the calculation of the collision integral, the result to lowest order in \( T/T_F \) is

\[
\left( \frac{k_T}{c} \right)_{T=0} = 0.3823 - c_3(P) \left[ 0.1045 + \frac{0.391 \lambda_K}{3 - \lambda_K} \right].
\]  

(2.12)

Here \( c_3(P) \) is a small quantity, equal to about 0.03 at \( P = 0 \). The quantity \( \lambda_K \) varies from 1.0 to about 1.4 as the pressure increases from 0 to 35 atm. Both of these quantities are explained in detail in ref. [21]. When the momentum dependent terms are added to the scattering amplitude, the expression for the thermal diffusion ratio becomes,

\[
\frac{k_T}{c} = \left( \frac{k_T}{c} \right)_{T=0} - \frac{m_3^2 T}{m_4^2 T_F} (2.194 \gamma_1 + 1.057 \gamma_2 + 0.055 \gamma_3).
\]  

(2.13)
Because $k_T$ is proportional to the $^4$He concentration, it is very small. Therefore it is unlikely that the effect of thermal diffusion can be measured in our apparatus.
CHAPTER 3

APPARATUS

3.1 Dilution Refrigerator

An Oxford model 200 dilution refrigerator cools the experiment to the temperatures needed to perform experiments in the Fermi liquid region of $^3$He - $^4$He mixtures. This unit is designed to give $200 \mu W$ of cooling power at 0.1 K. Although this unit can circulate over $600 \mu \text{mol/s}$ of $^3$He, it typically runs at $125 \mu \text{mol/s}$. This is sufficient to cool the experimental cell from 0.1 K to 0.02 K overnight. The refrigerator reaches a base temperature of $< 0.006$ K. Lounasmaa describes in detail the workings of a dilution refrigerator in his monograph[32].

The dilution unit is attached to a $^4$He pot, which holds 0.7 liters of liquid helium. It is continuously pumped by two Stokes mechanical pumps connected in parallel. The pot runs at $\sim 1.2$ K. To keep the Pot filled continuously, a 0.127 mm inner diameter capillary 27 cm long, runs from the pot to the liquid $^4$He bath at 4.2 K. The pot is mounted on the 4 K flange holding the vacuum jacket that encloses the pot and dilution unit. The vacuum jacket is suspended in a superinsulated dewar fill with liquid $^4$He.
3.1.1 Wiring and Fill Tubes

The wiring of the cryostat was carefully planned to prevent excessive heat conduction. The cryostat was designed to be able to cool the bundle of a nuclear demagnetization cooling stage. Since radio frequency electromagnetic fields can easily be picked up by the wiring in the cryostat and cause heating in the thermometers and heaters, the cryostat was built inside an electrically shielded screened room. As few as possible pieces of digital electronics are allowed inside the screened room, since they emit radio frequency radiation.

The maximum heat leaks allowed into each stage of the refrigerator were the similar to those that Jan Feder allowed when designing a similar refrigerator[33]. Since our dilution refrigerator is more powerful than his, we can accept more heat into some of our stages. The wiring is summarized in Table 3.1.

The heat sinks for the electrical leads consist of grooved pieces of copper to which formvar insulated copper fingers were glued with Stycast 2850 epoxy. The leads were then soft soldered to the fingers. The heat sinks were attached to the cryostat with nylon-copper squeeze connectors[34].

**Single Leads**

The leads to various devices in the refrigerator, such as thermometers and some heaters are unshielded inside the refrigerator. They are exposed to the liquid helium bath space from room temperature at the top of the cryostat to the 4 K flange. The wires are made of manganin, an alloy with a large thermal resistance and electrical resistance that decreases only 10% from 300 K to 4 K[35]. Below the pot, the
manganin is coated with a thin layer of Pb-Sn solder. The solder is superconducting below 7 K and prevents resistive heating.

**Coaxial Leads**

Properly heat sinking the coaxial cables is particularly important. Commercially available microcoaxial cable (UT-20-SS, Superior Tubes) run through a tube from a vacuum feedthrough at room temperature to the 4 K flange. This cable has a copper-plated stainless steel inner conductor, teflon dielectric and stainless steel outer conductor. Because the teflon is a poor heat conductor and not necessarily in good thermal contact with the outer and inner conductors, both conductors were heat sunk individually on a special mount on the 4 K flange.

Below the 4 K flange, special homemade coaxial cables were used. The outer conductor is a 0.610 mm outer, 0.305 mm inner diameter cupro-nickel tube coated with Pb-Sn solder. Cupro-nickel plated Nb-Ti wire, 0.076 mm in diameter, was strung through the tube to form the inner conductor. A jig was made to force Stycast 1266 epoxy through the tubes to form the dielectric. The jig was a cylindrical container with holes in the bottom. The cupro-nickel tubes were soldered into the holes with Wood's metal, an alloy that melts at ~ 70°C. The epoxy was poured into the jig and a lid with a gasket was bolted to the top. The jig was pressurized with 2 atm of compressed air. The epoxy typically flowed through a 2 m tube in about 10 minutes. This epoxy ensured that the inner and outer conductors were in good thermal contact with each other. The cables were then desoldered from the jig. After the epoxy set, the coaxial cables were then glued into the heat sinks below the pot using GE varnish.
Table 3.1: Wiring and Fill Lines in the Cryostat. Heat sinks are designated by “X”. The row “heat leak” represents the heat input to each stage from an individual item, whereas “total” is the total estimated heat current from all of the fill lines and leads. The abbreviations for the materials are: Mang = manganin, Sld-Mang = Pb-Sn solder coated manganin. UT20SS = commercially available microcoaxial cable, special = homemade cable described in the text. CuNi-SC = plated multifilament niobium-titanium. Nb-Ti = monofilament niobium titanium. SS = stainless steel and CuNi = cupro-nickel.
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<th>Material</th>
<th>Single Leads</th>
<th>Coaxial Leads</th>
<th>Hi-current Leads</th>
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<th>$^4$He Fill Lines</th>
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Table 3.1
High-CURRENT LEADS

The high current leads include current taps, for 4-terminal measurements to check thermal links in the refrigerator, heaters and magnets, such as the heat switch. To prevent resistive heating, these are made of superconductors. It is difficult, if not impossible, to solder to most superconducting wires. Above the still, cupro-nickel clad Nb-Ti wires were used. These were heat sunk at the still. The posts on the heat sink were copper clad Nb-Ti that extended 5 cm beyond the body of the heat sink so that the leads could be soldered over a large area, minimizing resistive heating at the joint. Below the still, copper-clad Nb-Ti wire was used, but the copper was removed with acid over all but a few centimeters at the end where solder joints could be made.

FILL LINES

Four fill lines run to the cell. They were designed so that each fill line could carry either $^3$He or $^4$He. This presents a difficulty since the thermal conductivity of pure $^4$He diverges at the $\lambda$-point (2.2 K at 0 atm) whereas below 0.05 K the thermal conductivity of $^3$He increases as the temperature decreases.

Typically heat sinks are made of sintered copper, however, concern that pieces of sinter might break off and clog the lines led to a different design. A heat sink consists of 95 layers of 100 mesh copper screen pressed into a cylindrical copper container 0.653 cm in diameter. The container is attached to the refrigerator with a squeeze connector[34].

Osheroff Connector

Items attached to the dilution refrigerator, including the cell, are mounted on the mixing chamber plate. A “squeeze” connector based on the design of Doug
Osheroff[36] links the mixing chamber plate to the mixing chamber. It exploits the different thermal expansion coefficients of copper, beryllium copper and tungsten to make a tight press fit between between a post on the mixing chamber plate and a sleeve on the mixing chamber. The connector is shown in fig. 3.1.

The Wiedemann-Franz law (see, for instance, ref. [32], pg 247), relates the thermal conductivity to the electrical resistance, \( \kappa = LT/R \), where \( \kappa \) is the thermal conductance, \( L = 25 \) nW \( \Omega/K^2 \) is the Lorenz constant, and \( R \) is the electrical resistance. Four-terminal electrical resistance measurements taken at about 1 K show that the connector has a resistance of 0.25 \( \mu \Omega \). The thermal resistance is therefore \( \kappa = 100T \) mW/K\(^2\). Unfortunately this conductivity is not significantly better than that found for the nylon “squeeze” connectors described in ref. [34]. This is attributed to the fact that the connector was not machined to tolerances, however, it worked well enough that it was not changed.

3.1.2 \(^3\)He Circulation System

A new circulating system, featuring a turbomolecular pump was installed for the dilution refrigerator. Unlike diffusion pumps, which were used on other dilution refrigerators in the laboratory, a turbomolecular pump does not use oil. The poor performance of the previous dilution refrigerator was attributed to the accumulation of diffusion pump oil in the dilution unit from backstreaming and accidents.

A Balzers TPH-450H turbomolecular pump, backed by an Edwards ED660 rotary mechanical pump, was installed. The turbomolecular pump circulates gas at rates up to 440 \( \mu \)mol/s. Since the backing pump contains oil, the dilution unit must be protected from backstreaming or cryopumped oil in the case of an accident or power
Figure 3.1: The "squeeze" connector between the mixing chamber and mixing chamber plate. Upon cooling, the beryllium copper and the copper fingers contract more than the tungsten. This produces a large pressure on the surfaces of the fingers, hence there is good thermal contact.
failure. Pneumatically operated solenoid valves close off the front and back of the turbomolecular pump when the power is off or when the backing pressure is too high.

There are a number of filters and traps to remove any oil or other impurities carried along with the circulating helium. The primary filter is a Balstons type BX filter placed in the circulating system past the backing pump. In addition there is a zeolite filter, a cold trap at liquid nitrogen temperatures and a cold trap in the bath space at 4.2 K.

Since the blades of the turbomolecular pump rotate at ~700 Hz and the controller uses digital components, the pump was mounted outside of the screened room. The pump had to be electrically isolated outside of the screened room so that it would not pick up radio frequency electromagnetic radiation. To do this, a thick ring of Stycast 1266 epoxy was cast and mounted to flanges in the pumping line. The ring was 4 cm thick with an inner diameter of 10 cm and an outer diameter of 20 cm. To prevent excessive heating while curing, the epoxy was cast in six separate layers. To remove any bubbles and trapped air from the epoxy, after mixing the resin and hardener, the epoxy was placed in a vacuum chamber and evacuated for about 10 minutes. It was then poured in the cast and evacuated again, this time for about 20 minutes. It was then allowed to set overnight in a refrigerator.

To minimize the transfer of mechanical vibrations from the pump to the cryostat, a double-gimbal system, very similar to the one described by Movshovich in ref.[37], was installed on the pumping line. In addition the turbomolecular pump was mounted on a metal plate with lots of rubber padding. The metal plate was attached to a wooden frame that was bolted to the floor.
3.2 Cell

The experimental cell was designed to measure both the solubility and the diffusion coefficient of $^4$He in liquid $^3$He. The helium mixture in the cell is confined by a cold valve, so that the overall concentration and number of helium atoms is constant throughout an experiment. A $^4$He-rich superfluid film forms on the cell walls at sufficiently low temperature. Two parallel plate capacitors are used to find the coverage of the $^4$He film. From the coverage as a function of temperature and pressure, the solubility is determined. When the pressure is quickly changed, the dynamic response of the film thickness is used to find the diffusion coefficient.

The overall setup of the experiment is shown in fig. 3.2. The cell consists of two volumes separated by a bellows. The mixture and capacitors are inside the bellows. The space outside the bellows is called the “compressor.” It is filled with $^4$He that controls the pressure by pressing on the bellows and mixture. A capacitive gauge, based on the design of Straty and Adams[38][14], measures the pressure. The mixture is confined to the cell by a hydraulically operated valve at low temperatures, which is described later in this thesis. An indium heat switch[14] allows the cell to be either thermally linked or isolated from the Mixing Chamber Plate of the dilution refrigerator. The temperature of the cell is measured by Ruthenium Oxide[39] and Germanium resistance thermometers, which were calibrated against a $^3$He melting curve thermometer[40].

A more detailed cross-section of the cell is shown in fig. 3.3 and cut-away view is shown in fig. 3.4.
Figure 3.2: Schematic representation of the experimental set up.
EXPERIMENTAL CELL

Figure 3.3: Cross-section of the cell, drawn at scale 1:5.1.
Figure 3.4: A cut-away view of the cell. For simplicity, the safety stop is not shown in this drawing.
3.2.1 Bellows

The bellows was selected to meet a number of criteria. First, it had to reduce its volume by at least 1/3 upon compression. This is the relative decrease in the molar volume of liquid $^3$He when the pressure increases from saturated vapor pressure to 36 atmospheres, where it solidifies. Second it had to be strong enough to withstand high pressures without bursting. Finally, its dimensions had to be such that a reasonable cell could be designed around it. No off-the-shelf commercially available bellows was found that met these criteria. Instead a beryllium copper bellows from Mini-Flex Corporation (model BC-1000-40-12-1) was altered. The bellows has a nominal outer diameter of 3.58 cm and an inner diameter of 2.50 cm with a wall thickness of 0.010 cm. The original bellows had 13 convolutions, however, four of these were cut off. The total length of the convolutions is 2.7 cm.

At room temperature, the spring rate of the bellows is 1660 dynes/cm, however for small displacements, the rate was measured to be 2830 dynes/cm at 1 K. By measuring the pressure of the compressing $^4$He and the mixture simultaneously, and knowing the initial volume and pressure of the mixture, we found the volume inside the cell as a function of the differential pressure, as shown in fig. 3.5. It would take about one atmosphere of differential pressure across the bellows to compress the bellows to 2/3 of its original length.

3.2.2 Capacitors

The two parallel-plate capacitors were made by evaporating gold onto sapphire substrates. The substrates were cylindrical, with a diameter of 19.05 mm and thickness of 2.54 mm. One side was optically flat and the other roughly ground. Spacers
Figure 3.5: The relative volume of the mixture as a function of the pressure difference between the compressing $^4$He and the mixture. The volume of the cell with no differential pressure is $\approx 20 \text{ cm}^3$. The curve is a second order least-squares regression. There is slight hysteresis between data taken when the pressure was increasing (solid squares) and when decreasing (open squares). This is evidence that the stop is scraping on the cell body, as discussed in subsection 3.2.4.

were sputtered onto the narrow gap capacitor and photoresist was used as the spacer for the wide gap capacitor. The sapphire disks were then placed on top of each other and glued together with silver epoxy. The gaps are 6.7 $\mu$m and 16.7 $\mu$m. Details about the capacitors can be found in Jizhong He’s thesis[14].

The capacitors were mounted vertically in the cell, with the narrow one sitting 8.6 mm above the wide. This was to facilitate a proposed experiment to study the interfacial tension of a phase-separated helium mixture. The capacitors sit in wells in the cell, with matching wells in a spacer placed over the capacitors (see section 3.2.3).
Strips of 0.013 mm thick mylar insulates the leads on the sides of the capacitors from the cell.

The wells were made by cutting into the metal with an electric discharge machine (EDM), a type of spark cutter. The electrodes for the EDM were made in the shape of the material to be removed from the cell. The bottom of the cell and the electrode were mounted in the EDM and submerged in oil. The machine kept the distance between the electrode and the cell such that a constant current sparked between them. Since this process not only cuts into the cell, but also removes part of the electrode, it was necessary to replace the electrodes regularly. At first, rough cuts were made using large currents. As the wells approached their final depths, the electrodes were again replaced and a smaller current was used to make finer cuts.

### 3.2.3 Spacer

The way that the capacitors are mounted breaks the azimuthal symmetry of the cell. This would make the analysis of the diffusion experiments very difficult, therefore, a spacer was placed over the capacitors to re-establish the symmetry as shown in figs. 3.3 and 3.4.

The spacer was once suspected of causing some serious problems in the apparatus. First, screws hold the spacer to the cell. The screws pass through the spacer into a tapped hole in the bottom of the cell. This creates a volume between the bottom of the screw and the bottom of the hole. This volume is nearly isolated from the rest of the cell. It may be that the ${^3}$He in this volume takes a long time to reach mechanical equilibrium with the rest of the cell. However, the volume is so small in the screw holes, about 0.2% of the cell volume, that it is unlikely that they could cause much of
a problem. Since the $^4$He film is superfluid, it is unlikely that $^4$He could be trapped in this volume.

Another problem attributed to the spacer is the capillary condensation observed in the cell[14]. Capillary condensation occurs when a substance condenses in a pore or capillary at a larger chemical potential than it would in bulk. The spacer sits flush on the cell bottom. This creates a crevice in which the $^4$He may capillary condense. It may also condense in the cracks created by the screws for the spacer. Analysis of the capillary condensation based on solubility experiments was done by Jizhong He[14]. He found that much more $^4$He was condensed than can be accounted for by the spacer, as discussed in section 4.2.3.

### 3.2.4 Safety Stop

The safety stop, shown in fig. 3.3, was added to the cell after a bellows was accidentally crushed by a few atmospheres of $^4$He in the compressor when there was no $^3$He in the cell. It is a brass cylinder attached to the top of the bellows with screws. The stop hits the bottom of the cell when the travel of the bellows exceeds 1 cm, preventing the bellows from being crushed. Since it was added after the cell had been designed and constructed, there was not much clearance for it between the cell wall and the walls of the bellows. In fact, there was only 0.25 mm clearance per side between the stop and the walls. It seems likely that if top of the bellows were not exactly perpendicular to the cell walls, or if the bellows did not compress symmetrically that the stop could scrape against the wall.

Three pieces of evidence indicate friction in the bellows. First, when doing diffusion experiments, we attempted to adiabatically increase the cell pressure in a number
of steps. We then similarly decreased the pressure back to its original value, however the temperature returned to a higher value. In section 6.1.2, we analyze these experiments and show that a frictional force of \( \approx 0.55 \text{ N (0.12 lbs)} \) could be responsible for the heating. This effect can also be seen as a "temperature spike" when the pressure changes, shown in fig. 3.6(b). Another indication of heating is that there is the hysteresis in the volume of the cell versus pressure difference across the cell (see fig. 3.5). This could be explained by a force of about 0.45 N (0.10 lbs) that opposes motion of the bellows. Finally, before assembling the cell, Jizhong He measured the position of the bellows and stop and found that it was slightly out of line. We made a mistake by cooling down without realizing the consequences of this.

Another problem with the stop is the piece of brass soft soldered to the top bellows convolution. It was designed to be big enough for the stop to be attached with screws. However, the shape of this piece is such that inside the bellows, it nearly separates the space in the top convolution from the rest of the cell. This can be seen in fig. 3.3. It also is possible that this piece rubbed against the inner diameter of the next convolution creating friction when the bellows moved.

3.3 Cold Valve

To keep the amount of helium and concentration in the cell constant and to prevent thermomechanical pressure gradients, \(^4\text{He} \) refluxing, or concentration gradients caused by thermal gradients, it was necessary to seal the cell at low temperatures. This required a superfluid-leak tight valve that opens and closes easily. We constructed a hydraulically operated valve based on the design of Roach, Ketterson, and Kuchnir[41]. The design of Roach \textit{et al.} has been improved by a number of groups
Figure 3.6: A typical attempted diffusion experiment. The pressure step (a) initiates diffusion. The temperature spike (b) is probably caused by the safety stop scraping against the cell wall. The capacitance ratio changes (c) as the solubility changes with pressure. More details are in chapter 4.
and we modeled our valve on the one used by Professor Balibar's group at the Ecole Normale Superieure.

The valve is pictured in fig. 3.7. Its stem is made of a strong plastic called Kel-F and the seat is stainless steel. The hydraulic liquid is $^4\text{He}$, whose pressure is controlled though a capillary that runs to room temperature. This $^4\text{He}$ fills the actuator, a nickel bellows (Servometer, model FC-16), that pushes the valve stem into the seat. A smaller phosphor bronze bellows (Mini-flex, model PBA-125-50-68) is soldered to the valve stem holder and valve stem guide to contain the incoming helium mixture for the cell.

Since the cross-sectional area of the actuating bellows is about 20 times larger than the bronze one, the valve can hold pressures in the cell much larger than the actuating pressure. At first, 3.5 atmospheres is needed to close the valve, however the pressure needed increased with use. At the end of the experiments, after about fifteen cycles, the valve did not seal, even with the actuator pressurized to 25 atm, the freezing pressure of $^4\text{He}$.

When the valve was first constructed, it would not open after closing, even with no pressure in the actuator. To force it open, a spring was placed between the stem guide and adaptor.

The valve seat was cut to have an angle of $60^\circ$ while the stem tip has an angle of $52^\circ$ so that it fits into the seat. In order to get a good seal, both the stem and seat were polished on a lathe with 1 μm alumina powder. The polishing powder was suspended in liquid nitrogen when polishing the kel-F tip, cooling it and making it much harder and easier to polish. The tip and stem were inspected under a microscope to make sure that they were polished well.
Figure 3.7: The hydraulic cold valve, at scale 2:1.
3.4 $^4$He Gas Handling System

The $^4$He gas handling system is used to control the cell pressure and to open and close the cold valve. Most of the lines are 0.16 cm stainless steel tubing from Hi-P, Inc. The system is shown schematically in figure 3.8.

The main pressure gauge in the system is a Paroscientific Digiquartz gauge. It works by measuring the frequency shift of a quartz crystal oscillator when the pressure changes. Because it uses RF frequencies, it had to be mounted outside of the screened room to prevent unwanted pickup and heating in the cryostat. As in the case of the dilution refrigerator's circulation system, we had to connect the metal plumbing outside the screened room to that inside with a non-conducting material. The pressure in the $^4$He system can be much higher than that in the circulating system. Because many insulating materials are permeable to helium and the flow rate through these materials is proportional to the pressure difference[42], it was more difficult to find a suitable isolator for the $^4$He system than for the circulation system. Plastics, quartz, and most glasses were too permeable. The material chosen was Macor. It is a machinable glass-ceramic consisting of an interlocked mica crystal array dispersed in a glassy matrix[43]. A cylindrical Macor tube, 2.5 cm long by 4.2 cm diameter, with a 0.16 cm diameter hole through it served as the isolator. It mated, using an indium 0-ring seal, with flanges hard soldered to the tubing in the rest of the system.

The 250 cm$^3$ bottle serves as a ballast when changing pressure in either the cell compressor or valve actuator. The liquid nitrogen trap ensures that the helium put into the cryostat is clean of contaminants that could freeze and block the lines.

Three flow impedance with ratios 1:5.8:28 were installed in the system to further control the rate at which the pressure could be changed in the cell compressor. This
Figure 3.8: Room temperature gas handling system.
system was added because of the temperature spikes seen in the diffusion measurements. One hypothesis for the cause of these spikes was that the helium rushed into the compressor so quickly that its velocity exceeded the superfluid critical velocity. This would cause frictional heating. We had hoped that changing the pressure slowly would prevent that. Unfortunately, it did not and we now think that the friction between the safety stop and the cell walls caused this heating, as described in section 3.2.4.

The impedances were made by inserting 0.08 mm diameter beryllium copper wire into 0.13 mm diameter cupro nickel or stainless steel tubes. The wire was inserted into the tube as far as possible, usually between 16 and 60 cm, before it kinked and would go in no farther. The wire went in more easily when about 2 atmospheres of air was blown through the tube against the inserted wire. To measure the value of the impedances, the 250 cm³ bottle was filled with about 40 atm of helium and connected through the impedance to a vacuum pump. By recording the pressure in the bottle as a function of time, the impedance was found. The values of the impedances, labeled "Z=1", "Z=5.8" and "Z=28", are $2.7 \times 10^{11}$ cm⁻³, $1.6 \times 10^{12}$ cm⁻³ and $7.7 \times 10^{12}$ cm⁻³ respectively. By opening and closing the four Whitey Ball valves in different configurations, impedances between $2.3 \times 10^{11}$ and $9.6 \times 10^{12}$ cm⁻³ could be made.

There is valve in the cell compressor line on the 4 K flange. The valve assembly runs the length of the bath space to the top of the cryostat were it can be opened or closed. When measuring the solubility, this valve is kept closed to prevent the cell compressor pressure (and hence the cell pressure) from changing as the level in the bath changes.
3.5 Data Acquisition

3.5.1 Hardware

In a typical measurement, the following devices are monitored: (1) the capacitance ratio, (2) the cell temperature with a ruthenium oxide (RuO) thermometer, (3) the cell pressure, (4) the mixing chamber temperature, usually with a germanium resistance thermometer (GeRT). In addition, we sometimes measure the $^3$He melting pressure thermometer (MPT), the $^4$He compressing pressure, and the wide and the narrow capacitances individually.

All capacitances, including the MPT and cell pressure are measured with General Radio 1615 capacitance bridges. The bridge circuit balances the capacitor with stable internal capacitors and ratio transformers. To prevent drifts due to changes in room temperature, a heater and two thermocouples were installed in each bridge and insulation was placed around the capacitors, ratio arms and casing. Temperature controllers were built to keep the capacitance bridges at 36±1°C for room temperature between 12 and 36°C.

A General Radio 1493 precision decade transformer, with the same type of temperature control described above, is used to measure the ratio. The RuO thermometer on the cell body is also part of a bridge circuit that has a reference resistor mounted on the cell and a Singer-Gertsch ratio transformer.

An audio signal excites the bridges and the null signal is detected with a lock-in amplifier. The lock-in amplifier output is a DC signal whose output is proportional to the null signal. We used three Princeton Applied Research 124 lock-ins and one Stanford Research Systems SR830 DSP lock-in.
The DC signal from the lock-in is measured with a digital multimeter, that communicates with the computer using GPIB (General Purpose Interface Bus). We used one Hewlett Packard model 3455A digital voltmeter, two Hewlett Packard 34401A digital multimeters, and one Keithley model 181 nanovoltmeter. The voltmeters and the computer were kept out of the screened room because they are digital and emit radio frequency radiation, which could be picked up by devices in the cryostat and cause heating. The signal from the lock-in was carried by coaxial cable and brought through the screened room wall via a feedthrough.

The conductance of the GeRT, mounted on the mixing chamber was measured with a SHE model PCB conductance bridge. It outputs a voltage proportional to the conductance, which was measured by a voltmeter and fed to the computer.

The pressure of the $^4$He that compresses the cell is measured with a Paroscientific Digiquartz pressure gauge. The output is monitored by the computer via an RS-232 cable to a serial port.

### 3.5.2 Software

Six very similar programs were written using QUICKBASIC to collect data. All versions of the software monitor the four digital voltmeters and some versions also measure the pressure gauge. The versions also differ in that some save the data to file after reading each point. Others save the data only after all the points have been collected. This allows more rapid data collection rates, but risks losing data in the event of a computer crash or power outage. The only other variation of the software is in the scaling used to display the data on the monitor.
The program allows the user to choose the time interval between points, the multimeter function (voltmeter, ammeter, etc.), the range and sensitivity of the multimeters, and the sensitivity of the Paroscientific gauge. The program then estimates the amount of time it will take to collect each data point. The user then chooses the time interval between points and the number of data points to be collected, as long as it is an integer multiple of 256. The program then collects the multimeter and pressure readings and plots them on the computer screen. The QUICKBASIC code for one of the programs is given in Appendix A.
CHAPTER 4

EXPERIMENTAL CONSIDERATIONS

4.1 The Immobile \(^4\text{He}\) Layer

In pure liquid \(^4\text{He}\) and in pure liquid \(^3\text{He}\), the first few layers next to the wall are solid-like and locked to the surface\(^3\). Because the \(^4\text{He}\) atoms are smaller than \(^3\text{He}\) atoms, they can sit deeper in the van der Waals potential of the wall. Hence, in mixtures of the liquids, the walls are preferentially coated by \(^4\text{He}\)\(^44.\, 45.\, 46.\, 47.\, 48.\, 49.\, 50.\, 51.\, 52\). While films of both isotopes individually and of helium mixtures have been extensively studied under their vapor, there are fewer studies of the immobile layer of the \(^4\text{He}\) film under bulk \(^3\text{He}\). Because we must keep track of the \(^4\text{He}\) atoms in the solubility and diffusion experiments, we need to know how much \(^4\text{He}\) is adsorbed on the surface and how the adsorption changes with pressure. In addition, the van der Waals potential at the film-bulk interface must be known to compare the concentration in the bulk to that at the surface of the film, as described in chapter 5. If the thickness of the immobile layer changes, then the potential will change as well. Based on the experiments reviewed below, we assume that the areal density of the immobile layer can increase with pressure.
The van der Waals attraction of a surface causes the pressure to increase as the distance to the wall decreases. Mechanical equilibrium requires that

\[ v(P) \, dP(D) = -dV(D). \] (4.1)

where \( v(P) \) is the molar volume at pressure \( P(D) \) and \( V(D) \) is the van der Waals potential at distance \( D \) from the wall.

One could construct a simple model of the liquid and film system in which a crystalline layer exists from the container wall up to the distance where the pressure equals the bulk freezing pressure. However, experiments on pure \(^4\)He in confined media [46][47], such as Vycor glass and sintered silver powder, show that superfluid \(^4\)He is still present in the pores, even at pressures above where bulk \(^4\)He crystallizes. This is not surprising since helium crystals do not wet most substrates, such as copper[53] or air adsorbed on different substrates[54]. An older, discredited explanation is that many small crystallites nucleate with different orientations in different sections of surface. Where these meet, the additional surface tension was supposed to increase solidification pressure[48]. Neither model predicts the thickness of the immobile \(^4\)He layer under liquid \(^3\)He, so empirical data must be used.

In the descriptions of the following experiments, the \(^4\)He coverage is often expressed as surface density (nmol/cm\(^2\)) and/or atomic layers. The Cornell group considers the first atomic layer of \(^4\)He to have surface density 1.8 nmol/cm\(^2\), while liquid layers at saturated vapor pressure have densities of 1.3 nmol/cm\(^2\)[44]. Some papers only give coverages as atomic layers. In these, I assume that the first layer has density 1.8 nmol/cm\(^2\) and layers above that have density 1.3 nmol/cm\(^2\).

One way to study the immobile layer is to observe the Kosterlitz-Thouless (KT) transition[55]. There is a universal transition in two-dimensional \(^4\)He films in which
the film develops a superfluid component of mass surface density $\rho_s$. The transition temperature[56] is given by

$$T_c = \rho_s \frac{\pi}{2} \frac{\hbar^2}{m_4 k_B}. \quad (4.2)$$

Here $m_4$ is the mass of a $^4$He atom, $k_B$ is Boltzmann's constant and $\hbar$ is Planck's constant divided by $2\pi$.

The KT transition has been observed with torsional oscillators by the Cornell group[57][3]. Changes in the oscillator's period and amplitude measure the superfluid mass and dissipation in the film. In pure $^4$He films, no KT transition was observed below 2.9 nmol/cm$^2$[3]. Figure 4.1 shows the period shift of the oscillator $\Delta P \propto \rho_s$, versus the $^4$He coverage. When $\rho_s$ vanishes, there is a finite coverage. This $^4$He is referred to as the immobile layer. When 13.6 nmol/cm$^2$, or about 12 layers, of $^3$He is added to the system, the thickness of the immobile layer increases by 0.6 nmol/cm$^2$[3], as shown in fig. 4.1. Assuming that 12 layers of $^3$He is thick enough to approximate the bulk, this shows that the immobile layer has a density of 3.5 nmol/cm$^2$ under $^3$He at zero pressure. This point is represented by the upright triangle in fig. 4.2.

Other experiments that give information on the immobile layer were designed to investigate $^3$He quasiparticle scattering at surfaces. $^3$He quasiparticles scatter diffusely from most surfaces, even highly polished ones. The addition of a $^4$He surface layer increases the specularity of the scattering. There are two complementary mechanisms for the enhanced specularity: (1) the replacement of the magnetic $^3$He immobile layer with nonmagnetic $^4$He[44], and (2) the superfluidity of the $^4$He layer[50].

(1) In pure liquid $^3$He an immobile layer coats all surfaces. This layer is magnetic and has a large Curie-Weiss magnetic susceptibility[44][58]. $^3$He quasiparticles scattering off this immobile layer can exchange spin. In fact, in superfluid $^3$He Cooper
Figure 4.1: Period shifts extrapolated to $T = 0$ for pure $^4\text{He}$ (open circles) and $^4\text{He}$ with $\sim 12$ layers of $^3\text{He}$ (closed circles) plotted against coverage. This figure is copied from ref. [3].

Pairs are often broken at the wall[44]. When $^4\text{He}$ is present, it preferentially coats the wall and displaces the magnetic immobile layer of $^3\text{He}$. This greatly reduces the magnetism at the liquid-boundary layer interface[58] and reduces the efficiency of pair breaking[44], even when the $^4\text{He}$ film shows no superfluidity. Measurements of the NMR relaxation times $T_1$ of liquid $^3\text{He}$ and spin-polarized liquid $^3\text{He}$ corroborate the effect of $^4\text{He}$ on the surface. When the surfaces are coated with a few layers of $^4\text{He}$, $T_1$ increases by two orders of magnitude compared to pure $^3\text{He}$[59, and references within]. In pure $^3\text{He}$, most of the relaxation is caused by paramagnetism at the boundary and $^4\text{He}$ layers reduce this relaxation[59].

A recent paper by Sawkey et al. [60] reviews the experiments described above and below. It also reports that the thermomechanical effect, $\Delta P/\Delta T$ across a porous
Figure 4.2: Estimates of the coverage of the inert $^4$He layer under bulk liquid $^3$He. The arrows indicate those points which are limits of the coverage. The lower points are probably the most relevant because they indicate the thinnest film to exhibit any type of superfluid behavior.
plug, in $^3$He with four surface layers of $^4$He is much smaller than in pure $^3$He. The size of the thermomechanical effect disagrees with theory\cite{61}. The reason it changes with the addition of $^4$He is not understood, but it is probably related to magnetic scattering at the immobile helium.

(2) $^3$He quasiparticle reflection at a $^4$He superfluid surface is more specular than at a non-superfluid surface\cite{50}. This is demonstrated by measurements of the effective viscosity of normal and superfluid $^3$He in cells coated with different amounts of $^4$He.

Richie, Saunders and Brewer\cite{49} looked at the effective viscosity of normal fluid $^3$He-$^4$He mixtures in a torsional oscillator made of Stycast 1266. The effective viscosity depends on the slip length which in turn depends on the degree of specularity. Starting with pure $^3$He, they added 1.7 layers (~ 2.7 nmol/cm$^2$) of $^4$He to form their first mixture. This greatly reduced the effective viscosity compared to pure $^3$He. The effective viscosity continued to decrease as more $^4$He was added, saturating at about 10 layers. Hence, 1.7 layers is an upper limit on the amount of $^4$He needed to increase specularity. A similar experiment was done by Tholen and Parpia\cite{62}. They made a torsional oscillator of silicon, polished to ~ 2 nm. The effective viscosity of normal fluid $^3$He decreased with the addition of $^4$He. At 3 mK, they showed that the degree of specularity increased from about 20% to about 92% as the coverage increased from 2.08 nmol/cm$^2$ to 5.77 nmol/cm$^2$. The data of Richie et al. is represented by the open diamond in fig. 4.2 while those of Tholen and Parpia are the solid squares. The square at the highest coverage corresponds to the 92% specular point while the square at low coverage is where the specularity began to increase.
Another experiment, by Steel et al., looked at the flow rate of superfluid $^3$He films out of a copper beaker[51]. The flow rate was much larger when the surface was plated with 1.2 monolayers ($\sim 2.5$ nmol/cm$^2$) of $^4$He. Other coverages were not explored.

The remainder of the experiments observed superfluid $^3$He in confined geometries. The $^3$He superfluid order parameter $A$ is a complex $3 \times 3$ matrix[44]. This matrix corresponds to the three momentum-space projections and three spin projections for the spin one Cooper pairs. The Ginsburg-Landau expansion of the free energy[63] of $^3$He in zero magnetic field is[44, 64, 65]

$$F = a \text{Tr} AA^\dagger + F_4 + F_{g2}, \quad (4.3)$$

where $A^\dagger$ is the Hermitian conjugate of $A$. The term $F_4$ contains quadratics in $A$ and can be found in refs. [44] and [64]. Spatial variations of the order parameter contribute the term $F_{g2}$[65, 44]

$$F_{g2} = \sum_{i=-1,0,1} K_L |\nabla \cdot A_i|^2 + K_T |\nabla \times A_i|^2. \quad (4.4)$$

The three vectors $A_i = (A_{x,i}, A_{y,i}, A_{z,i})$ are constructed from components of the order parameter $A_{ji}$ and $K_L$ and $K_T$ are positive constants. The characteristic length over which the order parameter changes is known as the correlation length. For longitudinal gradients, $A_i$, changing along the direction parallel to the wall, the correlation length is $\xi_L = \sqrt{K_L/a}$ and for transverse gradients, $\xi_T = \sqrt{K_T/a}$[65]. At a wall where the $^3$He quasiparticles reflect specularly, all vector components $A_i$ of the order parameter vanish. At a wall with specular reflection, however, only the component of the order parameter perpendicular to the wall (longitudinal component) vanishes[65, 44]. In confined geometries, much of the $^3$He is close to the complicated,
non-planar walls. It is difficult to separate the longitudinal and transverse components. In this case, the order parameter varies over a distance equal to the shortest correlation length $\xi_T[44]$. It is written as

$$\xi(T) = \frac{\xi(0)}{\sqrt{1 - T/T_c}},$$

where

$$\xi(0) = \left(\frac{7 \zeta(3)}{80}\right)^{1/2} \frac{h v_F}{\pi k_B T_c}.$$  \hspace{1cm} (4.5)

$T_c$ is the superfluid transition temperature, $\zeta(3) \approx 1.202$ is the Riemann zeta function of 3 and $v_F$ is the Fermi velocity; $\xi(0)$ varies from 50.1 nm at zero pressure to 10.9 nm at 29 atm[44]. In geometries where the pore size is comparable to the correlation length, the $^3$He superfluid transition temperature and superfluid density $\rho_s$ are suppressed compared to the bulk. Plating the surfaces with $^4$He increases the specularity, so that the transverse component of the order parameter no longer vanishes at the surface. This partially restores $T_c$ and $\rho_s[44]$.

Freeman, Richardson et al.[44][45] used a torsional oscillator and NMR to observe $^3$He in a stack of Mylar sheets separated by 300 nm diameter alumina spheres. In their first report[45], they found that at 1.5 and 5 atmospheres, the $^3$He superfluid fraction, as a function of $T/T_c$, was suppressed in pure $^3$He, but partially restored when the surface was coated with $2 \pm 1$ layers of $^4$He (3.1 nmol/cm$^2$). In addition, the transverse NMR lineshapes, as a function of normal fluid density, were different from the bulk lineshapes for pure $^3$He, but were similar to bulk after adding the $^4$He.

In a more detailed report[44], samples with no $^4$He, 3.2, 7.0 and 9.0 nmol/cm$^2$ were compared at different pressures. Measured with a torsional oscillator at 8.4 atm, the $^3$He superfluid fraction increased when 3.2 nmol/cm$^2$ of $^4$He was added, even though
the $^4\text{He}$ was not superfluid, see fig. 4.1. At 9.0 nmol/cm$^2$, the superfluid fraction was almost identical to that of bulk $^3\text{He}$. No torsional oscillator data were reported for the 7.0 nmol/cm$^2$ sample, however the NMR lineshifts were similar to those in bulk $^3\text{He}$. A plot of the NMR lineshifts against $T/T_c$ shows the 9.0 nmol/cm$^2$ sample nearly identical to the bulk value, while the lineshifts in the 3.2 nmol/cm$^2$ sample fall between that of the pure $^3\text{He}$ confined in the mylar and that of bulk $^3\text{He}$.

Another torsional oscillator experiment was performed by Tholen and Parpia[4], this time in a packed 70 nm diameter powder of silver. They plot $\rho_s/\rho$ versus pressure at $T/T_c = 0.4$ for pure $^3\text{He}$ and $^3\text{He}$ with 5 nmol/cm$^2$ and 6.5 nmol/cm$^2$ of $^4\text{He}$. The data for 5 nmol/cm$^2$ follow one curve and the pure $^3\text{He}$ data follow a lower one, as shown in fig. 4.3. At about 17 atm, the $^4\text{He}$ data deviate from the upper curve and reach the lower curve at 28 atm. When the pressure is decreased, the data hysteretically return along the lower curve until about 20 atm, where $\rho_s/\rho$ begins to make its way back to the upper curve at 18 atm. This indicates that the $^4\text{He}$ layer began to become immobile at 18 atm and finished “freezing” at 28 atm. It did not begin to melt again, however, until the pressure was lowered to 20 atm. Similarly, the 6.5 nmol/cm$^2$ sample began to “freeze” at 24 atm, but had not completed “freezing” at 30 atm, the maximum pressure that could be obtained. These limits are shown as the solid circles in fig. 4.2.

Kim et al.[52] use fourth sound resonances to find $\rho_s/\rho$ and the specularity of superfluid $^3\text{He}$ in alumina powders with grain sizes of 3000, 1000, and 300 nm, as well as, sintered silver powder of nominal diameter 70 nm. Fourth sound is a pressure wave in a superfluid when the normal fluid component is immobilized by the walls of the pores. At $T/T_c = 0.47$ they plot $\rho_s/\rho$ against $^4\text{He}$ coverage for the four different
powders at 5 atm. From these data they conclude that the specularity begins to increase at 2.45 nmol/cm$^2$ and would become completely specular at 3.88 nmol/cm$^2$. They also plotted $\rho_s/\rho$ versus pressure for the pure $^3$He and two of the coverages. We consider the $^4$He to be completely immobile when the curves for the $^4$He coated samples meet the curve for the pure $^3$He. This occurs at 21 atm for the 2.97 nmol/cm$^2$ sample and 24 atm for the 3.88 nmol/cm$^2$ sample. These results are given by the solid downward triangles in fig. 4.2.

One final point (open triangle) is included in fig. 4.2. Using NMR, Kawae et al.[66] suggest that a 6.35 nmol/cm$^2$ layer of $^4$He becomes immobile at the bulk solidification pressure, 25 atm. Unfortunately, ref. [66] is only a preliminary report.
We use fig. 4.2 to insure that $^4$He is conserved when measuring the solubility. The amount of $^4$He locked to the surface appears to increase with pressure, so we assume that the amount of $^4$He that can dissolve in the bulk decreases with pressure. Since it is difficult, based our limited understanding of the inert layer, to estimate how much the inert layer coverage increases with pressure, the coverage was fitted during the analysis of the solubility data. The results of these fits are consistent with fig. 4.2.

4.2 Surface of the Cell

4.2.1 Gold Plating

To interpret the data, the $^4$He-rich film should have the same thickness on all surfaces throughout the cell. This means that the van der Waals potential from all surfaces inside the cell should be the same. This is not completely possible, since the capacitors are made of sapphire, the feedthroughs are epoxy and there are mylar insulators around the capacitors' leads. Nevertheless, to match the potential of the gold electrodes of the capacitors, all of the metal parts inside the cell were gold plated. First, the metal was cleaned with a Scotch-Brite scouring pad. soap and water. Next it was rinsed in deionized water and submerged in Atomex Immersion Gold solution for five minutes. The plating solution was kept between $45^\circ$ and $75^\circ$ C, usually near $60^\circ$ and its pH was kept between 7-8. According to the product instructions, this should have deposited a gold layer 56 nm thick. After removing the cell parts from the solution, they were rinsed with tap water, followed by deionized water. Although not dull, they were not as shiny as expected. The instructions indicated that this was caused by uneven plating on the surface. After polishing with a paper towel, the surface looked better.
4.2.2 Surface Potential

If we assume that there is only gold on the surface of the cell, we know the potential at any distance from the surface. It is found by integrating the Lennard-Jones potential and taking into account the effects of retardation, which are significant at film thicknesses greater than about 15 nm. Without retardation, the potential is given by $V = -\alpha/d^3$, where $\alpha$ is the van der Waals constant between an atom and a surface, and $d$ is the distance from the wall. When corrected for retardation[67], the potential is:

$$V = \left( \frac{\alpha_{Au}}{d^3} \right) \left[ 1 + 1.64(d^*)^n \right]^{-1/n},$$  \hspace{1cm} (4.7)

where $n = 7/5$ and $d^* = d/d_{1/2}$. For helium on gold, $d_{1/2} = 16$ nm[67], and $\alpha_{Au} = 3.18$ K/nm$^3$[68].

Before starting the experiments, the cell was flushed with helium to remove the air from the cell, however, some air remained in the cell. Analysis of experiments in which the adsorption of $^3$He in equilibrium with its vapor was measured as a function of temperature were fitted. They show that the residual air layer is $1.01 \pm 0.06$ nm thick[14] and that the immobile $^4$He layer was about $0.51 \pm 0.07$ nm thick. These values were rounded to 1 and $0.5$ nm when analysing the diffusion data. The thickness of the air layer is consistent with estimates of the amount of air that remained in the cell. These measurements also gave us the surface area of the cell, $598 \pm 24$ cm$^2$.

The thickness of the immobile layer was assumed not change with pressure. This seems to contradict fig 4.2 where the solid layer density increases with pressure. While fitting the solubility, we tried fitting both the coverage and the thickness independently. The results were that the coverage and thickness decreased with pressure. Again this seemed to contradict the data in fig. 4.2. In another fit, we assumed the
molar volume of the immobile layer was constant, but that its thickness varied linearly with pressure. This fit had large uncertainties. As a compromise, we modeled the thickness of the immobile layer as a constant in pressure. However, at each pressure, the coverage was fitted and was found to increase with pressure in a way consistent with fig. 4.2. It is reasonable that the coverage increases with pressure even though the thickness remains the same. This implies that the compressibility of the immobile layer is such that the higher pressure compresses the film so much that the additional $^4$He does not increase the thickness.

With these assumptions, the potential $V$ at the surface of a $^4$He film $D^*$ thick, where $D^*$ is measured from the top of the gold is,

$$
V(D^*)/k_B = \frac{-\alpha_{Au} + \alpha_{air}}{(D^*)^3\left[1 + 1.64\left(\frac{D^*}{d_{1/2}}\right)^{7/5}\right]^{5/7}} + \frac{-\alpha_{Au}}{(D^* - D^a)^3\left[1 + 1.64\left(\frac{D^* - D^a}{d_{1/2}}\right)^{7/5}\right]^{5/7}}.
$$

(4.8)

Here $\alpha_{Au} = 3.11 \text{ K nm}^3$ and $\alpha_{air} = 0.836 \text{ K nm}^3$ are the van der Waals constants of gold and air respectively. The thickness of the immobile layer is $D^i = 0.5 \text{ nm}$ and the thickness of the air is $D^a = 1.0 \text{ nm}$. The bracketed terms in the denominator of eq. (4.8) are the corrections due to retardation. The constant $d_{1/2} = 16 \text{ nm}$ is the distance where the retarded potential is half the value of the simple van der Waals potential. More details on the potential can be found in Jizhong He's thesis[14].

4.2.3 Surface Area and Capillary Condensation

The geometric area of the cell is 201 cm$^2$, based on machine drawings of all of the parts in the cell. However, to a helium film a few nanometers thick, the area is larger
due to the microscopic roughness of the surface. One way to measure the surface area is to measure the adsorption isotherm[69], however our cell's surface area is so small that this method is not practical.

Parts of the cell were examined with an atomic force microscope (AFM). The AFM drags a sharp tip attached to a cantilever across the surface of the sample. A laser reflects off of a mirror connected to the cantilever and the location of the scattered beam is measured. It is used to determine the position of the cantilever and produces a map of the surface height. The AFM scanned square regions of linear size varying from 200 nm to 50 μm. The AFM digitized 512 points per line, creating a 512 × 512 array of points to represent the surface. To compensate for any macroscopic slope or bowing of the surface, the image was “flattened” using a second order polynomial in two directions of the plane.

The surface of a material is often fractal in nature. This means that the area is inversely proportional to the size of the measuring device raised to the fractal dimension. \( A \propto d^{-f} \). Here \( A \) is the area, \( d \) is the size of the measuring device and \( 2 \leq f < 3 \) is the fractal dimension of the surface[70]. In the diffusion and solubility experiments, the surface area is needed when the measuring device, the \(^4\)He-rich film, varies from a few tenths of a nanometer to about 20 nanometers thick.

To compute the fractal dimension, the AFM tiles the scan with triangles. Using the measurement of the height at the corners of the triangles, the area of each triangle is computed. The area of all of the triangles is summed to find the total area. This process is repeated for triangular tiles of many different sizes. A plot of the log of the total area versus the log of the linear size of the triangle produces a plot of slope \(-f\). The AFM software found typical fractal dimensions of \( f \approx 2.15 \), however the plots
were not close to being linear. One of the problems is that AFM cannot probe deep cracks because of the finite size of the tip. It also does not see overhangs or caves in the surface.

Three different surfaces were examined with the AFM, and are shown at a scale of 10 $\mu$m in fig. 4.4, 1 $\mu$m in fig. 4.5 and 0.1 $\mu$m in fig. 4.6. On a piece of the gold-plated copper cell body from an old cell, there are macroscopic features, such as machine marks, on all scans larger than 5 $\mu$m. Another surface examined was the back of the sapphire capacitor. The surface had been roughly ground by the manufacturer and the images suggest that the surface had been polished with an abrasive approximately one micron in size. Finally, the evaporated gold surface on the sapphire was examined. This was difficult to image because the tip tended to scratch the surface, however, this surface appeared much smoother than the others.

When fitting the solubility measurements, the area was assumed to be both flat and fractal. The results of the fitting did not change much with either assumption. As a result, the surface area was assumed to be a constant $598 \pm 24$ cm$^2$ regardless of film thickness. Nevertheless, measurements of the solubility on samples with a large amount of $^4$He present suggested that there was capillary condensation.

Consider the $^3$He-rich phase with a $^4$He-rich phase capillary condensed between two parallel plates separated by a distance $2h$. By equating the chemical potentials in the $^3$He-rich (upper phase, indicated with superscript $u$) and $^4$He-rich phase (lower phase, superscript $l$), one finds,

$$V \left( \frac{1}{v^l} - \frac{1}{v^u} \right) + \left( \frac{\bar{m}^l}{v^l} - \frac{\bar{m}^u}{v^u} \right) g z = \sigma \Gamma. \quad (4.9)$$

Here $v^i$ is the molar volume, $\bar{m}^i = X_3m_3 + X_4m_4$ is the average molar mass, $g$ is the acceleration of gravity, $z$ is the height above flat, uniform interface, $\sigma$ is the
Figure 4.4: AFM images of parts of the cell at 10 μm horizontal scale and 3.2 μm vertical scale. Clockwise from top left they are: the back of the alumina capacitor substrates, the gold-plated copper surfaces of the cell body, the gold electrode surfaces evaporated onto the front of the alumina substrate, and the gold electrodes at a more sensitive vertical range of 25 nm.
Figure 4.5: AFM images of parts of the cell at 1 μm horizontal scale and 0.3 μm vertical scale. Clockwise from top left they are: the back of the alumina capacitor substrates, the gold-plated copper surfaces of the cell body, the gold electrode surfaces evaporated onto the front of the alumina substrate, and the gold electrodes at a more sensitive vertical range of 25 nm.
Figure 4.6: AFM images of parts of the cell at 100 nm horizontal scale and different vertical scales. Clockwise from top right they are: the gold-plated copper surfaces of the cell body, the gold electrode surfaces evaporated onto the front of the alumina substrate, and the gold electrodes at a vertical range of 25 nm. We were unable to successfully image the back of the capacitor substrate at this small scale.
interfacial tension between the upper and lower phases, and $\Gamma$ is the curvature of the interface. The van der Waals potential $V$ is caused by the walls at the interface and the contribution from both walls must be included. In the limit that the gap $2h$ is much larger than the equilibrium film thickness $D^*$ outside of the capillary, we find,

$$h = D^* \left[ 1 + \left( \frac{D^*}{\lambda} \right)^2 \right], \quad (4.10)$$

where $\lambda^2 = \alpha_{Au}(1/v^l - 1/v^u)/\sigma = (3.16\text{nm})^2$. When the $2h$ is closer to $D^*$, eq. (4.9) was solved numerically and $h$ was fitted to a cubic function in $D^*$. To make the solubility measurements of different samples consistent[14], we assumed that as $D^*$ increased, more and more $^4\text{He}$ condensed in capillaries. We fitted the volume of capillary condensed $^4\text{He}$ to a cubic function in $h^2$. For small $h$, where the dominant term is linear in $h^2$, this corresponds to narrow wedge-shaped cracks. For the sample with the highest $^4\text{He}$ concentration, the value of $h$ was 160 nm at highest $D^* = 11.3\text{ nm}$; this corresponded to $2.2 \times 10^{-4} \text{ cm}^3$ of capillary condensed $^4\text{He}$ or 3.7 nm of film thickness. The physical origins of this much capillary condensation is that the surface of the cell is very rough. We can make a simple estimate based on the upper right image in fig. 4.5. Assuming that the total length of crevices of width $2h = 0.2 \mu\text{m}$ is about 2 $\mu\text{m}$ and that they have rectangular cross sections with an average depth of 0.1 $\mu\text{m}$, then the total amount of $^4\text{He}$ that would capillary condense in the area of the scan, $1 \times 1 \mu\text{m}$, is 0.04 $\mu\text{m}^3$. Multiplying that volume by the ratio of the geometric area to the area of the AFM scan, gives a total volume of $8 \times 10^{-4} \text{ cm}^3$. This is about eight times larger than indicated by the fit. Because the calculation is very simple and we did not examine the bellows, which makes up much of the surface area, with the AFM, the fact that the fits and calculation agree to the same order of
magnitude is reassuring. Other places that capillary condensation may occur include the threads of the screws, the cell spacer, and the indium O-ring seals.

4.3 Background Ratio ($R_0$)

Ideally, the ratio $R$ of the capacitors would only change as the superfluid $^4$He-rich film changes. It should be independent of the temperature, pressure and concentration of the bulk liquid $^3$He. This section deals with $R_0$, the changes in $R$ that are caused by effects other than the film changing.

In the capacitor, the dielectric constant is a function of distance from the electrode, $z$. This is because the capacitor contains an air layer, immobile layer and superfluid layer on the surface and the remaining space between the electrodes is filled with bulk liquid. The pressure in the film increases as one gets closer to the gold electrodes and this causes the dielectric constants of each layer to vary with distance as well. The capacitance can be written as,

$$\frac{1}{C} = \frac{1}{4\pi A} \int_0^d \frac{dz}{\epsilon(z)},$$

where $d$ is the size of the gap and $A$ is the area of the electrodes. The difference between the capacitance of the capacitor with the films on the surface and filled only with a uniform dielectric $\epsilon_0$ can then be written as

$$\frac{1}{C} - \frac{1}{\epsilon_0 C_0} = -\frac{2\delta}{d}. \quad (4.12)$$

Here $C_0 = 4\pi A/d$ and

$$\delta \equiv \int_0^{d/2} \left(1 - \frac{\epsilon_0}{\epsilon_z}\right) dz. \quad (4.13)$$
Except near the walls, $\epsilon_z = \epsilon_0$, therefore we replace the upper limit of the integral by infinity. The quantity $\delta$ therefore describes the part of the capacitance that is due to the films on the surface.

Hans Hjort calculated $\delta$ as a function of the profile of the superfluid film and used this to calculate $R - R_0[15]$. In his model of the profile, the density of the film increases as the distance to the gold decreases due to the pressure increase causes by the van der Walls potential. In thin films, $D^s$ and $N_4^s$ vary rapidly and nonlinearly with $R - R_0$ but for thick films, both $D^s$ and $N_4^s$ vary linearly with $R - R_0$. This is the region where the van der Waals potential is small and the pressure in the film is very near that of the bulk.

When analyzing the solubility and diffusion data, Hjort's model is used, however, to show the approximate dependence of the capacitances and $R$ on the film thickness. I have assumed here that the film has uniform density and have ignored the air layer.

The ratio bridge measures the ratio of the narrow capacitance to the sum of the capacitances.

$$R = \frac{C_N}{C_N + C_W},$$

where $C_N$ and $C_W$ are the capacitances of the narrow and wide capacitors respectively. Each capacitor is treated as an ideal parallel plate capacitor with a gap of thickness $d$ and area $A$. There are three dielectrics in the capacitor: (1) the mixture of dielectric constant $\epsilon$, (2) the immobile layer of thickness $D^i$ at each electrode and dielectric constant $\epsilon^i$, and (3) the superfluid film that extends a distance $D^s$ above the electrode and dielectric constant $\epsilon^s$. The total capacitance is equivalent to these capacitors in series:

$$\frac{1}{C} = \frac{d - 2D^s}{\epsilon A} + \frac{2D^s - 2D^i}{\epsilon^s A} + \frac{2D^i}{\epsilon^i A}.$$
The parts of this equation that do not depend on \( D^* \) are grouped into a piece called \( C_0 \), so that
\[
\frac{1}{C} = \frac{1}{C_0} - \frac{2D^* \Delta \epsilon}{A \epsilon \epsilon^s},
\]
where
\[
\Delta \epsilon = \epsilon^* - \epsilon \approx 0.015
\]
and
\[
\frac{1}{C_0} = \frac{d}{\epsilon A} \left[ 1 - \frac{2D^i (\epsilon^i - \epsilon)}{d \epsilon^i} \right].
\]

The \( R_0 \) is defined as
\[
R_0 = \frac{C_{0N}}{C_{0N} + C_{0W}},
\]
where the subscripts \( N \) and \( W \) here and throughout this thesis refer to properties of the narrow and wide capacitors respectively. Substituting equation (4.16) into (4.14), using the definitions (4.18) and (4.19), and doing some algebra gives the ratio:
\[
R = R_0 \left[ 1 + \frac{2D^* - 2D^i}{d_W} \frac{\Delta \epsilon}{\epsilon^s} \left( 1 - R_0 \left( \frac{d_W}{d_N} - 1 \right) \right) \right].
\]

Because the gold electrodes of both capacitors were deposited using the same mask, the areas of both capacitors are assumed to be the same. Then this expression reduces to
\[
R = R_0 \left[ 1 + \frac{(2D^* - 2D^i) \Delta \epsilon}{d_W \epsilon^s} (2R_0 - 1) \right].
\]

The small quantity in eqs. (4.20) and (4.21) is
\[
\frac{2(D^* - D^i) \Delta \epsilon}{d_W \epsilon^s} \approx 10^{-5}.
\]

This is what the \( \delta \) discussed above would be assuming each layer had constant density.
It was evaluated numerically by assuming \( D^* \approx 15 \text{ nm}, d_W \approx 16.7 \mu\text{m}, R_0 \approx 0.714 \).
and eqn. (4.17). This is the term that we want to measure. It is large enough to be detected since the ratio bridge circuit is sensitive to changes in $R$ of $< 5$ parts per billion, however, an accurate value of $R_0$ must be subtracted from eq. (4.21).

By inserting eq. (4.18) into eq. (4.19), $R_0$ can be expanded to show its dependence on $D^i$ the immobile layer thickness,

$$R_0 = R_{oo} \left[ 1 + \frac{2D^i \Delta \epsilon^i}{\epsilon^i} (1 - R_{oo}) \left( \frac{d_W}{d_N} - 1 \right) \right]. \quad (4.23)$$

Here $\Delta \epsilon^i = \epsilon^i - \epsilon$ and $R_{oo}$ is the ratio when the capacitors are empty, or filled with identical dielectrics and no adsorbed boundary layer,

$$R_{oo} = \left( 1 + \frac{A_W}{A_N} \frac{d_N}{d_W} \right)^{-1} \approx \left( 1 + \frac{d_N}{d_W} \right). \quad (4.24)$$

Assuming $A_N = A_N^*$, eq. (4.23) can be written to look like eq. (4.21),

$$R_0 = R_{oo} \left[ 1 + \frac{2D^i \Delta \epsilon^i}{\epsilon^i} \left( 2R_{oo} - 1 \right) \right]. \quad (4.25)$$

The second bracketed term in eqs. (4.23) and (4.25) is about $2 \times 10^{-6}$. The expected change in this term with pressure are expected to be at least a factor of 10 smaller. As the pressure changes from 0 to 24 atm, $R_0$ was found experimentally to vary by $\sim 7$ ppm. as shown in fig. 4.7. This change is much larger than and at high pressures of opposite sign from the effect of the small increase in the immobile layer density with pressure, discussed in section 4.1.

To measure $R_0$, the superfluid film was removed from the cell at $P=0$ using a method similar to that used by Keystone and Laheurte[71]. They connected the cold helium mixture through a capillary to a still at a temperature above the tricritical point. The $^4$He was transported through the superfluid film to the still where it was pumped away. In our case, we cooled the cell to $\sim 20$ mK and opened the cold
Figure 4.7: Measurements of $R_0$ as a function of pressure. There is hysteresis when the pressure is increased and then decreased. After the cell had warmed to 4 K, $R_0$ did not return to its previous value when the cell was cooled again. This is referred to as the jump.
valve. The superfluid film flowed out of the cell, presumably into a thick film on the surface of the heat exchangers. This should drain the $^4$He film until it is thinner than the thickness of the KT transition [see eqn. (4.2)]. This is shown by equating the chemical potential of the superfluid film $\mu_s$ of thickness $D^s$ in the cell to the chemical potential of the bulk $^4$He-rich phase at $T_x$, the temperature of the heat exchanger. Figure 4.8 shows the superfluid film thickness in the cell as a function of temperature in equilibrium with a phase-separated mixture in the still heat exchanger at different temperatures. The straight line is the thickness of the KT transition in the cell; below this line the film is no longer superfluid and is considered part of the immobile layer.

In practice, the draining of the $^4$He from the cell did not go as smoothly as described above. The first attempt to drain the cell occurred at 24 atm. We first lowered the cold valve actuating pressure. The ratio began to decrease. When it stopped decreasing we closed the valve, but the ratio began to increase again. The ratio decreased when we raised the temperature, indicating that there was still $^4$He present. The pressure in the cell was then lowered to about $P = 0$ and the cold valve was opened again. Again the ratio decreased and we closed the valve when it stopped decreasing. This time the ratio did not increase upon warming and we were sure that the $^4$He had left the cell.

The cell was then brought to 35 mK and the heat switch was opened. The ratio $R_0$ was measured at different pressures $P$. After each pressure step, we waited a few hours to make sure that the system reached equilibrium.

There was hysteresis in the ratio between increasing and decreasing $P$. This can be seen in fig. 4.7. Look at the data points at 5, 10 and 15 atm. The hollow symbols, taken as $P$ was increased, have larger values of $R_0$ than the filled symbols, taken as
Figure 4.8: The film thickness in the cell as a function of cell temperature at $P = 0$, when in equilibrium with a phase-separated mixture at different temperatures. The straight line is the Kosterlitz-Thouless line. Below this line, the film is not superfluid. When the cold valve is opened, the superfluid film should come into equilibrium with the phase-separated mixture in the still heat exchanger at about 0.7 K. This should drain the film to the thickness of the KT transition.
$P$ decreased. At the maximum of the curve near 10 atm, the hysteresis is $\sim 0.1$ parts per million (ppm).

Similar pressure sweeps were made at 85 and 150 mK. While taking data at 85 mK, the utilities to the lab were cut and the cell warmed to 4 K. After cooling to 85 mK again, $R_0$ was about 0.35 ppm larger than previously. This can be seen as the difference between the upright and inverted hollow triangles at 4 atm in fig. 4.7. This change is referred to as a jump, a term used to describe any unexplained change in the value of $R_0$.

To fit the data, we started with a simple formula that had been used to fit some preliminary data,

$$R_0 = R_{00} \left[ 1 + A(\epsilon - \epsilon_0) - BP \right].$$  \hspace{1cm} (4.26)

Here $\epsilon_0$ is dielectric constant of $^3$He and at $T = 0$ and $P = 0$ and $R_{00}$ is the ratio of the empty capacitors. This formula has a simple physical interpretation. The term $A(\epsilon - \epsilon_0)$ can be shown to correspond to fringing fields and electric field lines that go through other dielectrics as well as the helium mixture. For example, field lines could go from the back of the capacitor electrode, through the sapphire, across the helium gap, and through sapphire to the back of the other electrode. The term $BP$ is attributed to the capacitor plate gaps changing with pressure. The spacers of the wide and narrow capacitors are made of different materials, so the compressibility of the gap materials differ.

While this simple fit had the advantage of being simple to interpret, there were a few problems with it. First, there was the jump. During the course of the experiments there were a number of jumps. Often these occurred after the cell warmed above 1 K, but at other times they appeared spontaneously, without an obvious cause.
Sometimes it was difficult to tell when if jump occurred. For instance, when increasing the cell pressure, the system would warm and be out of equilibrium. When it returned to equilibrium, the cell temperature was higher than before. The solubility had changed and the pressure was different. All these effects caused the ratio to change. If a small jump had occurred, it would have been difficult to see compared to the ratio change. Therefore each set of solubility data and each set of diffusion data had its own fitted constant added to $R_0$ to account for the jump. The jump in the $R_0$ data was fitted as well.

To improve the fit, terms in $P^2$, $(\epsilon - \epsilon_0)^2$ and $P(\epsilon - \epsilon_0)$ were added. These terms do not have a clear physical interpretation. However, their contribution to $R_0$ is fairly small, at most 30% that of the linear terms.

Since the fit to $R_0$ is based on a physical model, it was expected to fit additional data taken at higher temperatures, $T > 0.3$ K. Consistency with high temperature data is not terribly important because the solubility and diffusion data are at $T < 200$ mK. Nevertheless, the fit was not consistent with high temperature data. We added some data that had been taken as the cell cooled from 1 K. Still the high temperature data would not fit. Therefore an explicit temperature dependence was added to the fit. The only explanation that we could think of for the temperature dependence was that it was an intrinsic function of the capacitors. We therefore looked at data for $R_{00}$ when the cell was completely empty and cooled. This did not occur during the cooldown during which all of the diffusion and solubility data was taken, but on the previous cooldown. We fitted this data to a fourth order polynomial in the temperature, omitting the linear term so that the there is no additional slope as $T \to 0$, in agreement with the data.
Figure 4.9: The residues of the fit of $R_0$ as function of $P$ and $T$. (a) The hysteresis is evident around 15 atm. (b) The residues for $P > 1$ atm are the solid circles while the open circles are for $P \approx 0$. The curve is the additional temperature dependence added to the $P \approx 0$ data.
The data with pure $^3$He were then refitted with the explicit temperature dependence from the empty cell data. This fit seemed good for all pressures, except $P = 0$, where analysis of the solubility suggested that a small correction to the temperature dependence was needed. The residues for the fit are shown in fig. 4.9. The scatter in the data is $< 0.1$ ppm. The only systematic trend in the residues is in the pressure, where the effect of the hysteresis is evident in fig. 4.9(a) between 5 atm and 20 atm.

The hysteresis was not included in the fit because the history of the cell when doing the diffusion and solubility measurements was not the same as the history during measurements of $R_0$. This is responsible for some of the uncertainty in $R_0$ that is included when fitting our data.

### 4.4 Convective Transport of $^4$He and Heat

When designing the experiment, we assumed that $^4$He would be transmitted only by diffusive processes; no $^4$He would be transmitted by net fluid currents in the mixture. In other words, we assumed that the fluid velocity $u$ would be zero everywhere in the cell. Obviously, as the cell is being compressed, there are bulk velocities, but these do not contribute to the transport of $^4$He out of (or into) the film. Convection, however, causes $u$ to be nonzero. There are body forces in the liquid caused by temperature gradients and concentration gradients in the mixture. If these gradients are not vertical, mechanical equilibrium is not possible and there will be currents[72]. Temperature gradients were not expected in the mixture, but friction between the safety stop and the cell wall (see section 3.2.4) probably created nonuniform temperatures. In this section, we show that it is unlikely that convection significantly transported $^4$He.
Situations where the fluid velocity is caused only by temperature or concentration gradients are referred to as free convection. To completely solve the problem, six coupled, nonlinear differential equations must be solved: the continuity of mass, conservation of energy, conservation of \(^{4}\text{He}\) particles, and conservation of the three components of momentum (Navier-Stokes equations)[72],

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0, \tag{4.27}
\]

\[
\rho c_p \left( \frac{\partial T}{\partial t} + \mathbf{u} \cdot \nabla T \right) = \kappa \nabla^2 T + \sigma'_{ik} \frac{\partial u_i}{\partial x_k}, \tag{4.28}
\]

\[
\frac{\partial X_4}{\partial t} + \mathbf{u} \cdot \nabla X_4 = D \nabla^2 X_4, \tag{4.29}
\]

\[
\rho \left[ \frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right] = -\nabla P + \eta \nabla^2 \mathbf{u} + \left( \zeta + \frac{1}{3} \eta \right) \nabla (\nabla \cdot \mathbf{u}) + \rho g. \tag{4.30}
\]

In these equations, \(\rho\) is the mass density, \(c_p\) is the specific heat at constant pressure \(P\), \(D\) is the \(^{4}\text{He}\) diffusion constant, \(\kappa\) is the thermal conductivity, \(\eta\) and \(\zeta\) are the first and second viscosities respectively, and \(g\) is the acceleration of gravity. The viscous stress tensor \(\sigma'\) is derived in section 15 of Landau and Lifshitz's Fluid Mechanics[72].

\[
\sigma'_{ik} = \eta \left( \frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} - \frac{2}{3} \delta_{ik} \frac{\partial u_l}{\partial x_l} \right) + \zeta \delta_{ik} \frac{\partial u_l}{\partial x_l}. \tag{4.31}
\]

The thermal diffusion ratio is ignored and \(D, \kappa, \eta\) and \(\zeta\) are all considered constant over the small changes in temperature and density in this problem.

One could devote an entire thesis (or more) to solving the problem of convection in our cell, however, we will look at some approximate solutions and some empirical data in somewhat similar geometries to show that convection is negligible in our experiment. The first simplification made is the Boussinesq approximation (see, for example ref.[73]). The density is assumed to be constant everywhere, except in the gravitational term of the Navier-Stokes equation. Here we expand the density as
\( \rho = \rho_0 + \Delta \rho \). The pressure gradient is simply the hydrostatic value \( \rho_0 g \). The density change \( \Delta \rho \) consists of two parts, that due to thermal expansion \( \rho_0 \alpha \Delta T \), and the change due to concentration changes \( -\rho_0 \alpha_X \Delta X_4 \). Here \( \alpha = -1/\rho \partial \rho / \partial T \) is the thermal expansion coefficient, \( \Delta T \) is the difference between the temperature and average temperature, \( \alpha_X = 1/\rho \partial \rho / \partial X_4 \) which in our case is \( X_4 (m_4/m_3 - v_4^*/v_3) \), and \( \Delta X_4 \) is the difference between the local \(^4\)He concentration and average concentration. In addition, the viscous heating term in the energy conservation equation is considered negligible. The equations simplify to:

\[
\nabla \cdot \mathbf{u} = 0, \tag{4.32}
\]
\[
\frac{\partial T}{\partial t} + \mathbf{u} \cdot \nabla T = k \nabla^2 T, \tag{4.33}
\]
\[
\frac{\partial X_4}{\partial t} + \mathbf{u} \cdot \nabla X_4 = D \nabla^2 X_4, \tag{4.34}
\]
\[
\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = \nu \nabla^2 \mathbf{u} + \mathbf{g} \left( \alpha \Delta T - \alpha_X \Delta X_4 \right). \tag{4.35}
\]

In these equations we use the thermal diffusivity, \( k = \kappa / (\rho c_p) \) and the kinematic viscosity, \( \nu = \eta / \rho \). Tritton[73] gives the justification and criteria for the Boussinesq approximation in the appendix to chapter 14. For our system, it is an excellent approximation. We will assume temperature differences \(< 0.4 \) mK and concentration differences \(< 5 \) ppm. In this case, the Boussinesq approximation is valid in our geometry.

In the diffusion experiments, typical temperature transients are about 5 mK. Most of the temperature difference occurs at the metal-helium interface. To see this, consider a heat flux \( Q \) that passes through a copper-\(^3\)He interface and through \( l = 1.0 \) cm of liquid \(^3\)He (roughly the distance between the wall and the center of cell). Then
\[ Q = \Delta T_K / R_K = \kappa \Delta T_{He} / l, \]
where \( \Delta T_K \) is the temperature drop across the interface, \( R_K \approx 2 \times 10^{-5} T^{-3} \text{ K}^4 \text{ cm}^2 \text{s}/\text{erg}[32] \) is the Kapitza resistance, \( \Delta T_{He} \) is the temperature drop across the liquid \(^3\text{He} \) and \( \kappa \approx 70 / T \text{ erg/(s cm)} \). Then the ratio of the temperature difference in the helium and at the boundary is

\[
\frac{\Delta T_{He}}{\Delta T_K} = \frac{l}{R_K \kappa} \approx \left( \frac{T}{0.2 \text{ K}} \right)^4.
\]  

(4.36)

At \( T = 0.1 \text{ K} \), this is 0.062. Therefore, we assumed that a temperature spike of 5 mK measured on the copper cell corresponds to less than a 0.4 mK drop in the liquid.

Before going farther, consider the acceleration due to the body forces when the temperature and concentration gradients are first established, but the convective currents have not been established. That is, set \( \mathbf{u} \) equal to zero in the Navier-Stokes equation. Assume in the temperature range \( 0.04 < T < 0.1 \text{ K} \) that \( \alpha = -0.0013/T[74] \) and that \( \alpha_X = 0.67 \). At \( T = 0.1 \text{ K} \), the accelerations \( a \) are 0.005 and 0.003 cm/s\(^2\) due to thermal and concentration gradients, respectively. If the displacements are \( \frac{1}{2} a t^2 \). one minute after the establishment of the gradients, the fluid elements would have "moved" 9 and 5 cm. Displacements this large would cause significant convective mixing, however, this is a gross overestimate because viscous forces become important as the velocity increases. Also, because the cell is closed and rather small, a complicated flow involving the development of rolls would need to form. Additionally, it takes time for the gradients to establish, yet as time passes, diffusive processes cause the gradients to reduce in magnitude. Therefore, 0.4 mK and 5 ppm are overestimates of the maximum temperature and concentration differences in the cell at any one time.
As is typical in fluid dynamics, we look at the non-dimensional numbers used to characterize the flow\[75\]. The first parameter is known as the Prandtl number,

\[ \text{Pr} = \frac{\nu}{k}. \]  

(4.37)

This is the ratio of the molecular diffusivity of momentum (kinematic viscosity) to the molecular diffusivity of heat and is a property of the fluid. For normal liquid $^3\text{He}$ it is $\sim 6$ and nearly independent of temperature. This value is similar to some liquids and most gases\[73\]. The analogous number for problems involving concentration gradients is the Schmidt number,

\[ \text{Sc} = \frac{\nu}{\sqrt{D}}. \]  

(4.38)

Assuming that $D \propto 1/T$, the Schmidt number for $^3\text{He}$ varies as $1/T$. The values of these parameters, and the ones described below, are plotted against temperature in fig.4.10.

Another important number for thermal convection is the Grashof number. For a system with characteristic length $L$, the Grashof number is

\[ \text{Gr} = \frac{g \alpha \Delta T L^3}{\nu^2}. \]  

(4.39)

When the Grashof number is large, the viscous force is negligible compared to the buoyancy and inertial forces, however, when the Grashof number is small, the inertial forces are negligible compared to the viscous forces\[73\]. For concentration induced convection, the Grashof number is given by

\[ \text{Gr}_X = \frac{g \alpha_X \Delta X_4 L^3}{\nu^2}. \]  

(4.40)

Sometimes the Rayleigh number is also used. It is the product of the Grashof and Prandtl numbers,

\[ \text{Ra} = \text{Gr} \cdot \text{Pr} = \frac{g \alpha \Delta T L^3}{\nu k}. \]  

(4.41)
The final two numbers of interest are the Nusselt number $Nu$ and its concentration analog, the Sherwood number $Sh$. These are the ratios of the total heat or particle flux crossing the wall of the vessel, to the heat or particle flux that is conducted or diffused. When the Nusselt number is 1, all of the heat is conducted away from the walls and there is no convection.

There have been a number of studies of convection in rectangular enclosures, see refs. [5] and [76] and references therein. Most of these studies used air as the fluid. Vertical walls a distance $L$ apart were kept at different temperatures and had aspect ratios (height to length) between 3 and 40. When $Gr < 1500$, the Nusselt number was less than 1.1. This implied that convection played a very small role in heat or particle transport. The standard heat transfer text by Incropera and DeWitt[77] states that when the container is much more narrow than wide that there is absolutely no convection when $Gr < 1708$.

In our experiments the cell is cylindrical, not rectangular, but $Gr < 500.$ as shown in fig. 4.10. In a cylinder, the limiting value of the Grashof number for no convection is probably larger than in a rectangular geometry of similar size. Because the walls are supposedly isothermal, the flow should have azimuthal symmetry. Since $^3$He has a negative expansion coefficient, if the walls are suddenly heated enough to cause convection, the fluid at the side walls would flow downward. Continuity of mass requires all of this fluid to return up the middle of the cell and the velocity in the center would have to be larger than in a rectangular container of similar size. This would increase the viscous forces. It is therefore very unlikely that there is any appreciable convection in our cell during a diffusion measurement.
Figure 4.10: Dimensionless numbers used to characterize free convection. The maximum value of the Grashof number is 500. Convection is negligible in enclosed containers when $\text{Gr} < 1500[5]$. 

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4.5 Time Constants

The thermal time constants in the cell for both mass diffusion and thermal diffusion have been calculated and are shown in fig. 4.11. Our observed time constants, measured by adding a pulse of heat and observing how long it takes the system to approach equilibrium is plotted as the solid symbols in the figure. In the Fermi liquid region, $T < 0.1$ K, the observed time constants were much longer than predicted. This remains a mystery, especially since the data and theory have opposite temperature dependencies. The heat pulses used in the experiments plotted in fig. 4.11 probably caused much larger thermal gradients in the cell than occur in a diffusion measurement. It is possible these would induce convection in the cell and therefore, at high temperatures, where convection is greatest, the time constant is smaller than that from diffusive processes. On the other hand, one could also argue that convection could make the time constants longer. The convection might eliminate horizontal gradients but cause a vertical gradient over the entire length of the cell, breaking the symmetry about the center of the cell. This would effectively double the diffusion length and quadruple the time constant. These large time constants greatly reduced the rate at which we could take data, especially when measuring the solubility.
Figure 4.11: The time constants in the cell. The filled circles are the observed time constants. The solid and dashed curves are the longest mass diffusion and thermal time constants respectively, found by solving the diffusion equation in orthogonal functions. For mass diffusion we assume the concentration at the boundaries was constant and for thermal diffusion we assumed that the temperature gradient vanished at the boundary.
CHAPTER 5

EXPERIMENTAL METHOD AND DATA ANALYSIS

5.1 Experimental Method

The experiments were designed to measure the diffusion constant $D$ by quickly changing the pressure in the cell. This causes the solubility to change, hence the amount of $^4$He in the bulk changes. If this, say, increases the solubility, some of the $^4$He in the superfluid film diffuses into the bulk. As this occurs, the film thickness decreases. The time dependence of the film thickness is analyzed to determine $D$.

The diffusion measurements were made with the heat switch open, so that ideally they would be adiabatic. The helium mixture should then change temperature uniformly as the pressure changes. However, the cell body temperature would not change with temperature. Because the specific heat of the cell is four orders of magnitude smaller than that of the helium mixture, this will be a small effect. Estimating the thermal resistance between the cell and the liquid shows that the cell body should reach thermal equilibrium with the mixture in about two seconds.

5.1.1 Conditions Needed to Measure Diffusion

To successfully measure $D$, the system must be at a temperature and pressure where the bulk concentration changes appreciably with pressure. For example, if the
temperature were too low, almost all of the $^4$He will be in the film, and very little dissolved in the bulk. Similarly, if the temperature is too high, nearly all of the $^4$He will be in the bulk and there will be little $^4$He in the film. If changing the pressure were to increase the solubility, there would not be much $^4$He that could leave the film and the thickness would not change much.

Another difficulty is that when the cell is heated too much, too quickly the capacitance ratio drops drastically. The "temperature spike", discussed in section 3.2.4, was often large enough to cause the capacitance ratio $R$ to decrease so much that diffusion could not be observed, as shown in fig. 5.1. The cause of the drop in $R$ and the time that it takes for the system to return to equilibrium is not completely understood. One possibility is that the safety stop scraping heats the cell, but because the capacitors are not thermally anchored to the cell walls, they may have not been at the same temperature as the walls. Some of the $^4$He in the film may have left the capacitors and went to the walls, causing $R$ to decrease.

There are some experiments where there is no apparent problem with temperature spikes and where the temperature and pressure are such that diffusion can be observed. One of these measurements is shown in fig. 5.2. Details of the analysis will be explained later. In this experiment there is no temperature spike. Perhaps we were lucky and the safety stop did not scrape, or perhaps during this compression, the stop was stuck against the wall. Also notice that the ratio approaches equilibrium more slowly than the pressure. This is an indication of diffusion.
Figure 5.1: An attempted diffusion experiment where the ratio decreased due to the temperature spike. The sharp decrease in ratio is not caused by diffusion, but by the temperature.
Figure 5.2: A successful diffusion experiment. The fitting of this data will be discussed later.
5.1.2 Concentration of the Samples

Diffusion measurements were attempted on three different samples. Each sample contained progressively less $^4$He. The amount of $^4$He in the cell at the beginning of the experiments is rather uncertain. Using a leak detector as a mass spectrometer, the concentration was measured to be $250 \pm 50$ ppm at room temperature. It is very unlikely that this was the concentration in the cell at low temperatures. Some of the $^4$He probably coated the surfaces of the heat exchangers on the still, cold plate and mixing chamber. For each solubility measurement, the amount of $^4$He in the cell was deduced by observing the total change in ratio from high temperatures ($T > 0.2$ K), where nearly all of the $^4$He was dissolved in the bulk to low temperatures ($T < 0.03$ K) where almost all of $^4$He is in the film. The first diffusion sample had a concentration of about 100 ppm while the second and third samples had concentrations of $62 \pm 25$ and $40 \pm 9$ ppm. Attempts to measure diffusion in the third sample were unsuccessful.

5.1.3 Method of Applying a Pressure Step

The method of compressing or decompressing the cell for the first sample was that the $250 \text{ cm}^3$ bottle on the $^4$He gas handling system (see fig 3.8) was filled with $^4$He to about 2 atm above or below the pressure in the sample. The bottle was then quickly opened to the compressor. The compressor pressure would reach equilibrium in about 30 seconds and the total change in pressure of the compressor would be about an atmosphere.

The method for the second sample was similar, except that we tried to change the pressure more slowly. At that time the cause of the heating was unclear. We thought that during the pressure change, the $^4$He superfluid velocity may have been exceeded,
causing viscous heating of the actuating $^4$He. We also thought that the $^4$He entering into compressor may have rushed through the heat exchangers so quickly that it was not adequately cooled. To change the pressure more slowly, the valves between the bottle and compressor were cracked slowly, instead of opened quickly. With this method, the pressure change took about 60 seconds.

There was still heating during these experiments, so the impedance system, described in section 3.4 was installed. For the third sample, different paths through the impedance allowed the time constant to change from about 30 seconds to a few minutes. Of course, this did not prevent the temperature spike. Unfortunately, we were not able to measure any diffusion in this sample.

5.1.4 Dissipation and Noise in the Capacitance Ratio

The noise when measuring diffusion was larger than when measuring solubility[14] because the time constants on the lock-in amplifiers were shorter in order to observe the dynamic change. In the solubility measurements, the time constants were 30 seconds, while for the diffusion measurements, they were 0.3 seconds. Also, the noise was larger in the measurements of the first sample than the other two samples. The noise for the first sample was about 0.2 ppm, while for the second sample it was 0.02 ppm and for the third sample it was about 0.01 ppm. The first sample was particularly noisy because the dissipation in the capacitance ratio was large and increased exponentially with excitation voltage. Also, other aspects of the circuit had not yet been optimized, for instance, there were ground loops in the system.

The large dissipation was probably caused by some type of debris between the narrow capacitor plates. We decided to vaporize the debris by discharging a 0.56 $\mu$F
capacitor across the narrow capacitor. We charged the source capacitor to different voltages. After each discharge, the dissipation was checked. It did not decrease when the source capacitor was charged to 30 and then 50 volts, corresponding to an energy of 0.25 and 0.7 mJ respectively. The dissipation was reduced when the voltage was increased to 70 V, which corresponds to 1.4 mJ.

After the dissipation was reduced, the excitation voltage to the ratio bridge was increased. Also the electronics were optimized so that the measurements on the second and third samples had much lower noise.

5.1.5 Data Collection

In all of the experiments, the cell pressure, cell temperature and ratio were monitored. The data were taken as rapidly as possible. Because the multimeters are very accurate, the time to collect each data point was quite long, about 0.5 seconds. For the first sample, each pressure step was collected into an individual file, containing 256 data points. The analysis of these data files suggested that there may have been a small drift in the ratio during these experiments. For the other two samples, we collected many steps in one data file and allowed a few minutes of run-in time between each step. That way a drift would be obvious and could be accounted for more easily in the analysis.

5.2 Analysis of the Data

This section describes the analysis of the data. This was done in close collaboration with Hans Hjort who wrote much of the software used to fit $D$. Many of the details will be in Hans Hjort's thesis[15].
The first step of the analysis is to translate the ratio $R$ into the film thickness $D^*$ and total amount of superfluid $^4$He on the surfaces of the cell $AN_4^*$, where $A = 598 \pm 24$ cm$^2$ (see section 4.2.3) is the surface area of the cell and $N_4^*$ is the areal density of the superfluid film. Note that we are not interested in the amount of immobile $^4$He since we assume that its thickness does not change with pressure. The value of $R_0(P, T)$, described in section 4.3, was subtracted from the measurements of the ratio $R$. It was then corrected for the “jumps”, which were estimated from high temperature data taken within a few days of the diffusion measurements. A table of $D^*$ and $N_4^*$ as a function of $R - R_0$ was calculated by Hans Hjort[15], using the measured van der Waals potential of the surface (see section 4.2.2 and ref. [14]) and known properties of bulk liquid $^3$He-$^4$He mixtures. When $D^*$ is fairly large ($> 4$ nm), which it was in our experiments, both $D^*$ and $N_4^*$ vary almost linearly with $R - R_0$. The slope $d(R - R_0)/dD^*$ in Hjort’s model is asymptotically the same as that found by differentiating eq. (4.21), however his model takes into account the compressibility of the film close to the wall where the local pressure is large and changes rapidly. In the following discussion of the diffusion, we will use the three independent variables: $P$, $T$ and $D^*$.

5.2.1 Equilibrium Concentration

The saturated solubility $X_4^{\text{sat}}$, given by eq. (1.3), describes the equilibrium concentration of $^4$He in an upper phase in contact with a bulk lower phase at $T$ and $P$. In our cell, however, we do not have bulk lower phase, instead there is a $^4$He-rich superfluid film of thickness $D^*$ on the cell walls. To find the equilibrium value of the concentration $X_4^b(P, T, D^*)$ in the bulk we equate the chemical potential of the $^4$He in
the film \( \mu_4^s \) to that in the bulk \( \mu_4^b \). To maintain mechanical equilibrium, the pressure at the surface of the film \( P^s \) is larger than in the bulk, and given implicitly by

\[
V(D^s) = -\int_P^{P^s} v_3(P') dP',
\]

where the van der Waals potential is given by eq. (4.8). We assume that \( \mu_4^s \) is that of the lower phase at pressure \( P^s \) with the contribution of the van der Waals potential added,

\[
\mu_4^s = -L_4(P^s) - \pi_3^{sat}(P^s) v_4(P^s) + V(D^s).
\]

Here \( L_4 \) is the binding energy of pure \(^4\)He and because \( v_4 = -dL_4/dP \), the value of \( L_4(P^s) \) can be related to \( L_4(P) \).

\[
L_4(P^s) = L_4(P) - \int_P^{P^s} v_4(P') dP'.
\]

The osmotic pressure is \( \pi_3^{sat} \) and the term \( \pi_3^{sat} v_4 \) in eq. (5.2) corrects the binding energy for the dissolved \(^3\)He in the lower phase. Substituting eqs. (5.1) and (5.3) into eq.(5.2) gives

\[
\mu_4^s = -L_4(P) + \int_P^{P^s} (v_4 - v_3) dP' - \pi_3^{sat}(P^s) v_4(P^s).
\]

In Fermi liquid theory, the chemical potential of the \(^4\)He dissolved in the \(^3\)He is similar to that of an ideal gas of particles with mass \( m_4^b \).

\[
\mu_4^b = -E_4(P) + k_B T \ln \left[ X_4^b \left( \frac{T^*(P)}{T} \right)^{3/2} \right].
\]

This is the value in the bulk, when the distance from the walls is large enough (> 50 nm) that the van der Waals potential is negligible. The value of \( T^* \) is

\[
T^* = \frac{2\pi \hbar^2}{m_4^b v_3^{2/3} k_B}.
\]
Equating eqs. (5.2) and (5.5) and rearranging gives,

\[
X_4^b = \left[ \frac{T}{T^*(P)} \right]^{3/2} \exp \left[ \frac{-\Delta(P) - \pi_3^{sat}(P^*) \nu_4(P^*) + \int_{P^*}^{P} (\nu_4 - \nu_3) dP'}{k_B T} \right],
\]

(5.7)

where the difference in binding energy is \( \Delta = L_4 - E_4 \).

For large \( D^s \), \( V(D^s) \) and \( (P^* - P) \) are small. By making the approximation \( \pi_3^{sat}(P^*) \nu_4(P^*) = \pi_3^{sat}(P) \nu_4(P) \) and assuming that the molar volumes in the integrals are constant, eq. (5.7) may be written as,

\[
X_4^b(P, T, D^s) \approx \left[ \frac{T}{T^*(P)} \right]^{3/2} \exp \left[ \frac{-\Delta(P) + (1 - \nu_4/\nu_3) V(D^s) - \pi_3^{sat}(P) \nu_4(P)}{k_B T} \right].
\]

(5.8)

This equation has the advantage that it does not contain \( P^* \).

Equations (5.7) and (5.8) show that the equilibrium bulk concentration changes with \( D^s \) as well as with \( P \) and \( T \). This sets up feedback in a diffusion experiment.

For example, consider a pressure change that would increase the concentration in the bulk. As \(^4\)He goes from the film to the bulk, \( D^s \) decreases, making \( V(D^s) \) in eq. (5.8) become more negative, thus decreasing the equilibrium value of \( X_4^b \).

### 5.2.2 Boundary Condition

The next step is to solve the diffusion equation.

\[
\frac{\partial X_4(r, t)}{\partial t} = D \nabla^2 X_4(r, t).
\]

(5.9)

This is a second order partial differential equation, which requires an initial condition and a boundary condition in order to be solved. To simplify the analysis, we will measure all concentrations relative the initial concentration, \( \delta X_4 = X_4 - X_4(t = 0) \).

The initial condition is easy, we assume that the system is in equilibrium when each
diffusion measurement is started. Then, excepting a few nanometers close to the walls, \( \delta X_4(\mathbf{r}) \) is zero and uniform at all positions \( \mathbf{r} \).

The boundary condition is a little more difficult. The diameter and height of the cell are both about 2.5 cm, but the distance over which the van der Waals potential is important is a only few nanometers. Diffusion over a length of a few nanometers occurs much more quickly than over the length of the cell. When solving the diffusion equation, we therefore assume that the boundary concentration is the equilibrium concentration of the bulk, at pressure \( P \) in contact with a film of thickness \( D^* \), given by eq. (5.7).

Since \( D^* \) changes as the diffusion progresses, the boundary condition changes as well. For simplicity the boundary condition has been written.

\[
\delta X_4^b = (a + 1) \frac{dX_{Eq}}{dP} \delta P - a \delta X_4. \tag{5.10}
\]

Here \( \bar{X}_4 \) is the average concentration in the cell. I explain how to find this from \( D^* \) and \( N^* \) below. The term \( dX_{Eq}/dP \) is the equilibrium change in concentration with pressure. It is \( (X_4^f - X_4^i)/(P^f - P^i) \), where the superscripts \( 'i' \) and \( 'f' \) refer to the initial and final values. Notice that this linearization has the proper limits: when \( t = 0 \), \( \delta X_4^b = 0 \) and when \( t = \infty \), the system has reached equilibrium so that \( \delta X_4^b = \delta \bar{X}_4 = (dX_{Eq}/dP)\delta P \). There is no explicit temperature term in eq. (5.10) because the temperature changes are determined by the pressure changes, that is \( \delta T = \delta T(P) \). The quantity \( a \) is called the \textquotedblleft feedback parameter.\textquotedblright From eq. (5.10), notice that

\[
a = -\left( \frac{\partial X_4^b}{\partial \bar{X}_4} \right)_{P,T} = -\left( \frac{\partial X_4^b}{\partial D^*} \right)_{P,T} \left( \frac{\partial D^*}{\partial \bar{X}_4} \right)_{P,T}. \tag{5.11}
\]
To find this derivative and to relate $\dot{X}_4$ to $D^s$ and $N^s$, we write the conservation equation for $^4$He.

$$n_4^0 = n_3 \left[ (1 - f)X_4 + fX_4^b \right] + n_4^{cap} + AN_4^s. \quad (5.12)$$

Here $n_4^0$ is the total amount of $^4$He in the cell, excluding the immobile layer. The amount of $^4$He condensed in cracks and capillaries is $n_4^{cap}$, as discussed in section 4.2.3, and the amount of $^4$He in the film is $AN_4^s$. During a diffusion experiment, the small changes in $P$ have negligible effect on $n_4^{cap}$ and $N_4^s$. In the model of the capillary condensation, the volume of liquid condensed is only a function of the $D^s$. Since the molar volume of $^4$He changes with pressure, $n_4^{cap}$ does depend on $P$, but this effect is very small compared to the effect of $D^s$, since $n_4^{cap} \propto (D^s)^6$. Similarly, $N_4^s$ is nearly proportional to $D^s$, but the pressure dependence is much smaller, depending on the molar volume. Therefore in analyzing a diffusion experiment, $n_4^{cap}$ and $N_4^s$ only vary with $D^s$.

The term $n_3 \left[ (1 - f)X_4 + fX_4^b \right]$ in eq. (5.12) is the amount of $^4$He dissolved in the $n_3 = 0.55 \pm 0.02$ moles of $^3$He in the cell. The parameter $f$ is the fraction of the cell space that is "dead volume." This represents the spaces between the convolutions of the bellows. Since these spaces are small and close to at least two boundaries, diffusion here takes place so rapidly that the concentration is assumed to be that on the boundaries always. We have numerically solved the diffusion equation in the convolutions and found that the concentration there lags that of the boundary by about 1 second. This is small compared to diffusion times of about 60 seconds measured by the capacitors. The value of $f$ is 38% in our cell. This approximation makes the diffusion equation easier to solve because the complicated geometry of the bellows is replaced by the simple geometry of a cylinder.

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To find the term \((\partial D^s/\partial X_4)_{P,T}\) in eq. (5.11), we differentiate eq. (5.12) with respect to \(D^s\) at constant \(P\) and \(T\),

\[
0 = n_3(1-f)\left(\frac{\partial X_4}{\partial D^s}\right)_{P,T} + n_3f\left(\frac{\partial X_4^b}{\partial D^s}\right)_{P,T} + \frac{dn_{4\text{cap}}}{dD^s} + A\frac{dN_4^s}{dD^s}. \tag{5.13}
\]

In the limit of thick films, \(dN_4^s/dD^s = X_4^l/v^l\) where \(X_4^l\) is the \(^4\)He concentration and \(v^l\) is the molar volume of the lower phase. Solving eq. (5.13) and inserting it into (5.11) gives,

\[
a = -n_3(1-f)\left(\frac{\partial X_4^b}{\partial D^s}\right)_{P,T} \left[n_3f\left(\frac{\partial X_4^b}{\partial D^s}\right)_{P,T} + A\frac{X_4^l}{v^l} + \frac{dn_{4\text{cap}}}{dD^s}\right]^{-1}. \tag{5.14}
\]

From this equation, when \((\partial X_4^b/\partial D^s)_{P,T}\) is large, \(a \to (1-f)/f = 2.33\). This is the upper limit on the feedback parameter.

The term \((\partial X_4^b/\partial D^s)_{P,T}\) can be written as

\[
\left(\frac{\partial X_4^b}{\partial D^s}\right)_{P,T} = \left(\frac{\partial X_4^b}{\partial P^s}\right)_{P,T} \left(\frac{\partial P^s}{\partial D^s}\right)_{P,T}.
\tag{5.15}
\]

Differentiating eq. (5.1) gives

\[
\left(\frac{\partial P^s}{\partial D^s}\right)_{P} = -\frac{1}{v_3(P^s)} \frac{dV}{dD^s}. \tag{5.16}
\]

The value of \(dV/dD^s\) is found by the straightforward, but tedious differentiation of eq. (4.8). We also need to differentiate equation (5.7) to find,

\[
\left(\frac{\partial X_4^b}{\partial P^s}\right)_{P,T} = \frac{X_4^b}{k_BT} \left\{-v_3(P^s) + v_4(P^s) - \frac{\partial [\pi_3^{\text{sat}}(P^s)v_4(P^s)]}{\partial P^s}\right\}. \tag{5.17}
\]

The result of substituting eqs. (5.16) and (5.17) into eq. (5.15) is

\[
\left(\frac{\partial X_4^b}{\partial D^s}\right)_{P,T} = -\frac{1}{v_3(P^s)} \left(\frac{dV}{dD^s}\right) \frac{X_4^b}{k_BT} \left\{-v_3(P^s) + v_4(P^s) - \frac{\partial [\pi_3^{\text{sat}}(P^s)v_4(P^s)]}{\partial P^s}\right\}. \tag{5.18}
\]
This is substituted into eq. (5.14) to find the feedback parameter \( a \), used to determine the boundary condition, eq. (5.10). In the program that analyses the data, we assume that \( P^s \approx P \) and that the derivative of the osmotic pressure term is negligible. The first approximation is valid for thick films, while the second has been shown to be negligible based on the data in ref. [78].

5.2.3 Fitting the Data to find \( D \)

The software used to analyze the diffusion experiments was written by Hans Hjort and is described in more details in his thesis[15]. The program uses the Levenberg-Marquardt method[79, 80] to fit for \( D \), and six other parameters described below. The program takes trial values of \( D \) and solves the diffusion equation.

\[
\frac{\partial X_4(r,t)}{\partial t} = D \nabla^2 X_4(r,t).
\] (5.19)

To find the concentration \( X_4(r,t) \). The alternating Crank-Nicholson algorithm[15, 79] is used and the boundary value is given by eq. (5.10). The concentration \( X_4(r,t) \) is integrated over the volume of the cell to get \( \dot{X}_4 \), which is then translated into \( R \) and compared to the actual data to determine \( \chi^2 \). The Levenberg-Marquardt method looks at the variation of \( \chi^2 \) with \( D \) (and the other fitted parameters) to find the values of the parameters that minimize \( \chi^2 \).

Besides \( D \), there are seven other variables that are fitted in the program. Three of these are fitted to the initial and final data where we think that the system is in equilibrium. These three are the equilibrium change in ratio with pressure \([R' - R^i]/[P' - P^i]\), the initial ratio \( R^i \) and an overall linear drift of the ratio with time. These variables have very small errors. Two other variables are (1) a small constant correction to \( R_0 \) and (2) a small correction to \( dR_0/dP \). These allow for
the uncertainty in $R_0$ and $dR_0/dP$. The data set includes points that represent the probable values of these quantities (zero) with approximate uncertainties of 100 ppb in $R_0$ and 190 ppb/atm in $dR_0/dP$.

The frictional heating of the cell causes the temperature of the walls to be higher than that of the $^3$He. This causes two effects; first it decreases the chemical potential of the film. This lowers the amount of $^4$He in the dead volume, making $X_4^h$ decrease. Also it takes $^4$He-film out of the capacitor, causing $\tilde{X}_4$ to be overestimated. This is equivalent to adding a term in the temperature difference to both $\delta \tilde{X}_4$ on the left-hand side of eq. (5.10) and another term to $X_4^h$ on the right-hand side. These two terms are accounted for by fitting an addition to the ratio that is proportional to the temperature difference between the cell walls, measured with the RuO thermometer, and the liquid. The liquid temperature is deduced from the pressure. It is assumed that at the beginning and end of each experiment the temperature is in equilibrium. The pressure is scaled to the temperature at the beginning and end of the experiment and this is assumed to be the temperature of the bulk liquid.

The difference in temperature between the cell walls and the liquid is due to the Kapitza resistance (see, for instance ref. [32]). The Kapitza resistance varies as $T^{-3}$ and is therefore about 3 times larger at 0.065 K than at 0.095 K. The frictional heating causes about 1 $\mu$W of power to be dissipated while changing the pressure over about 30 seconds during each step. This heat current could cause a temperature difference across the cell walls of about 1 mK at 65 mK.

Finally the value of the feedback parameter $\alpha$ at the beginning of the experiment is fitted. It is given a value as a data point, based on the value of $D^a$ and the fits of the solubility and its uncertainty is 20%. The value of $\alpha$ is updated after each time
step in the Crank-Nicholson algorithm. To see how much the fit for $D$ depended on $a$, we tried many different initial values of $a$ on a particular sample and found that the resulting $D$ did not change by more than about 20%.
CHAPTER 6

RESULTS AND CONCLUSIONS

6.1 The Change in Temperature with Pressure

6.1.1 \( (\partial T/\partial P)_S \)

While the purpose of these experiments was to measure \( D \), our experimental also measured \( (\partial T/\partial P)_S \), the change in temperature with pressure at constant entropy \( S \), of \(^3\)He. The concentration of \(^4\)He is so small that it has no measurable effect on the thermodynamic properties of the \(^3\)He. The experiments were performed with the heat switch open. This was supposed to make them adiabatic, although heating often occurred at the walls because the stop scraped. Ignoring this effect, the cell temperature changes uniformly with the pressure. Figure 6.1 shows the temperature versus the pressure for the attempted diffusion measurements. Each symbol represents a set of measurements that should be isentropic. The lower branch of each curve was taken as the pressure increased and the upper branch as the pressure decreased. In the 100 ppm sample (open circles), there was unexplained heating at 12 atm while compressing the sample. To compensate for this, the data below this pressure were moved up 2.4 mK, as shown by the dotted line. The branches of each curve differ because of the heating. The actual value of \( (\partial T/\partial P)_S \) would be an interpolation of
the two branches. The solid curves in fig. 6.1 are lines of constant entropy. They were found by integrating Greywall’s formulas for the specific heat[6]. These curves originate at the lowest temperature point for each run. They do not run between the two branches of the curves. There are two possible explanations for this: (1) the lowest temperature point is not in the “correct” place compared to the rest of the data, or (2) there is systematic disagreement between our results and Greywall’s. It seems unlikely that Greywall’s work is in error. The error in it is quoted as < 0.5% and is recognized as being very good. Greywall’s data has been used to establish the \(^3\)He-melting curve thermometer temperature scale[40, 81]. It is most likely that the systematic disagreement is due to some type of small systematic error in our data.

6.1.2 Amount of Frictional Heating in the Cell

The data in fig. 6.1 can be used to estimate the amount of heating caused by friction between the safety stop and the cell walls. The experiments started at 6 atm and 98 mK and the pressure was increased in steps of about an atmosphere up to 24 atm. The pressure was then decreased in steps of about 2 atm. We estimate the amount of heat added to the system by comparing the temperature at 6 atm after decreasing pressure to that when we started. The difference in temperature is about 4 mK, shown as the distance between the arrows in fig 6.1. We multiply this temperature difference by the specific heat[6] at 100 mK and 15 atm to find that there was about 46000 ergs of nonreversible heating. The distance that the bellows traveled is known from the dimensions of the bellows and \(v_3(P)\) to be 0.7 cm. The average force was calculated and found to be about 0.65 N (~ 0.15 lbs). A similar calculation with the other two samples give forces of 0.56 and 0.49 N (0.12
Figure 6.1: The cell temperature versus pressure during diffusion experiments. Open and hollow circles are experiment on the 100 ppm sample, open and closed triangles 62 ppm, and open squares 40 ppm. The lower branches are compressing the cell and the upper decompressing, as indicated by the arrows. The dotted line on the 100 ppm sample corrects for the unexplained heating evident from the kink. The double-headed arrow shows the temperature difference used to estimate the frictional heating of the cell, as explained in the text. The heavy solid curves are lines of constant entropy, calculated from Greywall's specific heat[6].
and 0.11 lbs.). These agree with the value of 0.45 N, which was estimated from the hysteresis in the displacement of the bellows versus the pressure difference across the bellows, as discussed in section 3.2.4.

The specific heat of the cell body is calculated to be 16000 erg/K and the 46000 ergs was spread out over 31 steps, so the average amount of heating per step was 1500 ergs. The cell body would then be heated an average of 0.09 K per step. This is much larger than the observed heating, which was less than 0.005 K per step. This discrepancy may have two different explanations. First, the cell body and liquid helium are in such good thermal contact that the heat is adsorbed in the $^3$He before the cell body is heated. This is likely because the thermal time constant between the cell body and the liquid $^3$He is predicted to be about 2 seconds at 0.1 K and $P = 0$, while typically the pressure changed over a time of 30 seconds. Another possibility is that the cell thermometer was not in good thermal contact with the cell, so that the temperature spike would be smoothed.

6.2 Relationship between $D$ and $X^s_{4}^{sat}$

The purpose of the experiments described in this thesis is to compare the diffusion coefficient $D$ to the solubility $X^s_{4}^{sat}$ to test Fermi liquid theory. The theoretical calculation of the diffusion constant $D$ as $T \to 0$ was given in section 2.1. It can be written as:

$$D = (0.4461 \ldots) \frac{\hbar}{m_4^* T} \frac{T_F}{m_3^* T} \left( \frac{v_3}{v_4^*} \right)^2.$$  \hspace{1cm} (6.1)

The solubility of $^4$He in $^3$He is given by[19, 9],

$$X^s_{4}^{sat} = \left( \frac{T}{T^*} \right)^{3/2} \exp \left( -\Delta - \frac{\pi^{sat}_3 v_4}{T} \right),$$  \hspace{1cm} (6.2)
where
\[ T^* = \frac{2\pi \hbar^2}{m^*_4 v_3^{2/3} k_B} = \frac{4}{(9\pi)^{1/3} m_3^3} T_F. \] (6.3)

and,
\[ \Delta = (L_4 - E_4)/k_B. \] (6.4)

At \( P = 0 \), \( T^* \approx 4.9 \) K and \( T_F \) is the Fermi temperature of pure liquid \(^3\text{He}\). In these expressions, \( v_4^* = (1 + \alpha_4) v_3 \) is the partial volume of a \(^4\text{He}\) atom in liquid \(^3\text{He}\). The term \( \alpha_4 \) is called the BBP parameter and is often used in the literature instead of \( v_4^* \).

The binding energy of a \(^4\text{He}\) atom in \(^3\text{He}\) is \( E_4 \) and the binding energy of a \(^4\text{He}\) atom in pure \(^4\text{He}\) is \( L_4 \). At saturated vapor pressure, \( L_4 \) is the latent heat. The term \( \pi_3^{sat} v_4 \), product of the osmotic pressure in the lower phase and the molar volume \( v_4 \) of pure \(^4\text{He}\) is a few milliKelvin[78]. The value of \( v_4^* \) is found by differentiating \( E_4 \) with respect to pressure.
\[ v_4^* = -dE_4/dP \] (6.5)

and similarly, \( v_4 = -dL_4/dP \). The values of the \(^4\text{He}\) effective mass \( m_4^* \) and partial volume \( v_4^* \) are obtained from measurements of the solubility \( X_4^{sat} \) as a function of temperature and pressure.

### 6.3 The Solubility

The solubility of \(^4\text{He}\) in liquid \(^3\text{He}\) is the subject of Jizhong He's thesis[14]. Fits to \( X_4^{sat}(P, T) \) provide the effective mass \( m_4^*(P) \), the binding energy \( E_4(P) \) and the partial volume \( v_4^*(P) \). These values are used to find \( X_4^b \), which is used to calculate the feedback parameter in the analysis of diffusion data. More importantly, these are the thermodynamic variables that are in the formula for the diffusion coefficient \( D \).
The solubility was measured, with the heat switch open, in the same cell as $D$. It was carefully measured at 0 and 10 atm on the 40 ppm sample and at 0, 5, 10, 15, 20 and 24 atm on the 20 ppm sample. At each pressure, the temperature was changed in steps of a few milliKelvin. Data was taken with the temperature both increasing and decreasing. There was no hysteresis in the data for increasing versus decreasing temperatures, as long as $T < 0.12$ K. The cell was held at each temperature for a few hours to make sure that for each datum the system was in equilibrium. Figure 6.2 shows $R - R_0$ versus temperature at different pressures for the 20 ppm sample. The curves are the fits of the data, described in Jizhong He's thesis[14].

At about 0.08 K, there is a step at 24 atm, shown in the inset of fig. 6.2. We speculate that this is the pre-wetting transition, predicted by Pettersen and Saam[82]. It is not clear why we see this transition only at high pressures. In light of this, the fits of $\Delta(P)$ and $m_1^*$ that are used in the diffusion analysis do not include the data for this pressure. However, below the transition, the data at this pressure were fitted and we show the results in the plots of $\Delta$ and $m_1^*$.

The solubility as a function of temperature for the 0.03 atm data is shown in fig. 6.3. The circles are our data, the solid ones for a 20 ppm sample and the open circles for a 40 ppm sample. The solid line is the fit to our data. The open squares and diamonds are the data of Nakamura[7]. Our values of the solubility are about two times smaller than those of Nakamura. We do not have an explanation for this discrepancy.

Our data is very uncertain at both high and low temperature. At high temperatures, the film is very thin and the corrections due to the difference in pressure at the film surface $P^*$ from that in the bulk $P$ are important. Also, at high temperature, the
Figure 6.2: The measured $R - R_0$ versus temperature for the 20 ppm sample. As the temperature increases, the solubility in the bulk increases. The value of $R - R_0$ decreases as the $^4$He leaves the film and dissolves in the bulk. The solid circles are at 0.03 atm, solid circles 5 atm, solid upright triangles 10 atm, open upright triangles 15 atm, solid diamonds 20 atm, and hollow diamonds 24 atm. The curves are the fits to the data. The inset shows the step in $R - R_0$ seen only in the 24 atm data that may be a prewetting transition.
Solubility is very sensitive to the value of $R_0$. A small change in $R_0$ will cause a large change in $X_4^\text{sat}$. At low temperatures the data is also uncertain. The noise becomes important because the concentration in the bulk is very small. Even if the concentration were to change by, say, 50%, the actual number of $^4$He atoms condensing into the film would still be small.

The data at each pressure were fitted to eq. (6.2) to find the binding energy $\Delta$ and the effective mass $m_4^*$ at each pressure. The results are shown in fig. 6.4. The solid circles are our data. The data for both the 20 ppm and 40 ppm samples were fitted together, to the same $m_4^*$ and $\Delta$, at 0 atm and 10 atm. We had tried fitting them separately, but the uncertainty in the capillary condensation caused them to disagree with each other. The inverted triangle is a fit that we made[9] to the data of Nakamura[7]. The dashed line in the plots of $\Delta$ and $m_4^*$ are predictions from the microscopic theory of Boronat et al.[10, 11].

Because there is not a theoretical prediction for the functional form of $\Delta(P)$, it was rather difficult to decide how to fit it. The choice of the fitting function is important because the slope of $\Delta$ determines the partial volume, $v_4^* = v_4 + d\Delta/dP$. Based on the data of Laheurte[12], shown as the dashed line in the inset of fig. 6.4, we decided that $\alpha_4$, the BBP parameter defined by $v_4^* = (1 + \alpha_4)v_3$, should be linear in $v_3$. The fit of $\Delta(P)$ to such a function is the solid curve on the plot of $\Delta$. The resulting $\alpha_4$ is shown as the solid line in the inset. The square point in the inset is from Sato and Suzuki[13]. The partial volume $v_4^*$ that corresponds to our $\Delta(P)$ and $\alpha_4$ is shown as the solid line in fig. 6.5. Also the $v_4^*$ inferred from Laheurte's $\alpha_4$ [12] is shown as well as the prediction of Boronat et al.[11]. For comparison the molar volume of pure $^4$He $v_4$ is also plotted.
Figure 6.3: The saturated solubility $X_{4}^{sat}$ as a function of temperature at 0.03 atm. Our data for the 20 ppm sample are the closed circles while the open circles are for 40 ppm. The data of Nakamura[7] are open diamonds and squares, while the open triangles are from Laheurte[8]. The curve is the fit to the data below 0.1 K, it is extended as a dashed curve above 0.1 K were Fermi liquid theory is not applicable.
Figure 6.4: The difference in binding energy and effective mass versus the pressure. The closed circles are fits to our data and the triangles are a fit[9] to Nakamura’s data[7]. The dashed lines are a microscopic theory[10, 11]. The inset shows $-\alpha_4$, with the solid line being our result, the dotted line that of Laheurte[12] and the square from ref. [13]. See the text for more details. The solid curves were used in the analysis of $D$. 

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Figure 6.5: The $v_4^*$ calculated from the $\alpha_4$ in fig. 6.4 (solid line). For comparison, $v_4$, the molar volume of pure $^4$He, is the dot-dash line. Results inferred from Lahuerte[12] and Boronat's[11] $\alpha_4$ are the short and long dashed lines respectively. The square is from Sato and Suzuki[13].
We decided that the most reasonable fit to the $m_4^*$ data is a straight line. The weighted average of the points (excluding the 24 atm point) is $m_4^*/m_4 = 1.35 \pm 0.23$.

Additional information about $m_4^*/m_4$ has been obtained from the diffusion data. By looking at the initial and final value of $R - R_0$ in each diffusion measurement, we were able to estimate the total change in concentration $\delta X_4^\delta$. We also inserted the initial and final values of $P$, $T$, and $D^\delta$ in eq. (5.8) to calculate the theoretical $\delta X_4^\delta$. These two values of $\delta X_4^\delta$ often differed by a large amount and sometimes had opposite signs. We first assumed that $m_4^*$ did not change with pressure when using eq. 5.8, but then calculated how $m_4^*$ must change with pressure in order to get the same $\delta X_4^\delta$ observed in the experiments. This variation, $d \ln m_4^*/dP$, is plotted in fig. 6.6(a). The line is a linear regression through the data. That line is integrated in fig. 6.6(b) and is consistent with values of $m_4^*$ found from the solubility measurements. The curve in fig. 6.6(b) should be taken cautiously. There are a number of things that make a diffusion experiments less reliable than a solubility experiment in determining $m_4^*$. First the ratio in a diffusion experiment is less accurate and less precise than in a solubility measurement. Also the error bars on $d \ln m_4^*/dP$ are too large to be shown on the plot and there is a data point that is an outlier and not included in the linear regression and is not plotted. Finally there were drifts in $R$ during the diffusion experiments that were not present in the solubility experiments.

### 6.4 Diffusion Coefficient

The results of a fit to a typical diffusion experiment is shown in fig. 6.7. The pressure, shown as the inverted triangles, increased by about 0.2 atm in about 30 seconds. The change in temperature is shown in fig. 6.7(b). The solid curve through the data
Figure 6.6: The dependence of $m^*_4$ on $P$ needed to make the observed and calculated values of $\delta X^*_4$ consistent. (a) Solid symbols are the 100 ppm sample and open ones 62 ppm. The different symbols at each pressure are the same as in fig. 6.8. The solid line is a linear regression through the data. (b) The curve is the integration of the line in (a). The diamonds are the results from the solubility experiments shown in fig. 6.4.
is the pressure scaled to the total change in temperature. This is the temperature that the liquid would have if it changed linearly with pressure. The ratio $R - R_0$ is plotted in fig. 6.7(c). The solid curve going through the data is the fit. Going back to fig. 6.7(a), the solid curve is the average concentration $\bar{X}_4$ in the cell, while the dashed curve is the value at the boundaries $X_4^\circ$. Both have been scaled to vary from 0 to 1. The difference between these two curves drives the diffusion. This difference is shown in terms of the size of the actual signal $R - R_0$ in fig. 6.7(c). The difference of the solid line and dashed line is the amount that the ratio is out of equilibrium.

Figure 6.8 shows our experimental results. The points taken with the 62 ppm sample are hollow and those on the 100 ppm sample are solid. The circles are at about 1 atm, the triangles are near 8 atm, and the squares are near 22 atm. The lines are the predictions of eq. (6.1) at 1 atm (solid) and 22 atm (short dashed) using the results shown in fig. 6.4. These lines are broken above 0.1 K where Fermi liquid theory is not applicable. The bands are the uncertainty in the predictions based on the uncertainties of $v_i^*$ and $m_i^*$ from the fits of the solubility. The diamonds between 0.5 and 1 K are the data of Vvedenskii and Peshkov[1] taken at the melting pressure of $^4$He. about 25 atm. It would be interesting to measure $D$ between 0.1 K and 0.5 K.

Figure 6.9 shows the product $DT$ versus pressure. To order $T/T_F$, this should be independent of temperature according to the theory, eq. (6.1). In this plot, the solid symbols are the 100 ppm sample and the hollow ones are the 62 ppm sample. As discussed in section 5.2.3, we wanted to see if there are any systematic differences between experiments when $R - R_0$ increases and when it decreases. The circles are for $R - R_0$ decreasing while the squares are for $R - R_0$ increasing. The figure shows that there are no systematic effects in the direction of change of $R - R_0$. 

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Figure 6.7: The pressure (a), temperature (b), and $R - R_0$ (c) in a successful diffusion data, with the fits. In (a) the solid line is the scaled value of $X_4$ and the dashed line is $X_4'$. In (c) the difference between the solid curve and dotted line is the size of $R - R_0$ that is out of equilibrium.
Figure 6.8: The diffusion coefficient versus temperature. The hollow circles are for the 62 ppm sample, and the solid symbols 100 ppm. Circles are at 1 atm, triangles at 8 atm and squares at 22 atm. The diamonds are at 25 atm from ref. [1]. The lines are the predictions of eq. (6.1). The solid line is for 1 atm and the short dashed line for 22 atm. The lines are broken above 0.1 K where Fermi liquid theory is not valid. The bands are the uncertainty in the prediction, based on the uncertainty in the fits of $m^*_4$ and $\alpha_4$. 
Figure 6.9: The product of the diffusion coefficient and temperature $DT$ versus pressure. The hollow symbols are on the 62 ppm sample and the solid symbols 100 ppm. Circles are for experiments when $R - R_0$ decreases and squares when $R - R_0$ increases. The diamonds at 25 atm are measurements of Vvedenskii and Peshkov[1]. The solid lines is the prediction of eq. (6.1) and the shaded band indicates the uncertainty in $DT$ due to the uncertainty in $m_4^*$ and $\alpha_4$ from the solubility.
The predicted $D$ and measurements agree well. Although there is a large amount of scatter in the data, it is consistent with the error bars. As shown in fig. 6.7, the size of the signal that we are fitting to find $D$ is very small, often less than 50 parts per billion. The signal to noise ratio is about 2 for this experiment. In other experiments it varied from 1/2 to 4. The error bars shown in figs. 6.8 and 6.9 are the results of the uncertainty in the fitting. Because there are seven fitted parameters, described in section 5.2.3, and the signal to noise is small, it is not surprising that the error bars are large.

6.5 Conclusions

We have successfully measured both the solubility $X_{4}^{sat}$ and diffusion coefficient $D$ of $^4$He in liquid $^3$He below 0.1 K. From the solubility we found the thermodynamic parameters $m_4^*$ and $v_4^*$. We substituted these into the formula, eq. (6.1), that we derived for the diffusion coefficient. This theoretical prediction is consistent with measured values of $D$. We have found no inconsistencies in the Fermi liquid theory.
APPENDIX A

DATA ACQUISITION PROGRAM

Program: SAFE?. BAS
Date: May 1993
Written by: Todd G. Culman
revised: March 1994 by Jizhong He
revised: June 1994 by Todd Culman
Purpose: This program reads the Keithley Model 181
Nanovoltmeter, the Hewlett-Packard
model 3455A Digital Voltmeter and the
Hewlett-Packard 34401 digital multimeter
using the GPIB. This also records the pressure
reading from the Paroscientific Digiquartz
gauge on an RS-232 cable attached to COM1.
This also plots the results on the CRT while
taking data. This may prove useful in
collecting data for the diffusion coefficient
and solubility experiments.

Note: this must be linked with qmor.lib

DECLARE SUB findtime (hpcom$, kcom$, hptime, rr, mintime!, lmt)
DECLARE SUB statanal (e$(), s$(), erro%) 
DECLARE SUB setup (dt!, npts%, knvm%, hpdmv%, hp3%, hp4%, e$(), s$(), erro%, lmt)
DECLARE SUB errstat (e$(), s$())
DECLARE SUB fileinit (name$, i%, comment$( ))
DECLARE SUB bitfind (byte%, b%())
DECLARE SUB hpchk (erro%, hp3%)
COMMON SHARED ibsta%, iberr%, ibcnt%
DIM bit%(16), e$(16), s$(17), comment$(17)

init$ = "INIT": fet$ = "FETC?": red$ = "READ?"
knmvout$ = SPACE$(18): 'reserves proper amount of space for data
hpdmvout$ = SPACE$(15): 'strings from the voltmeters
hp34401$ = SPACE$(16)
hpdmm$ = SPACE$(16)

kxpoint = 0: kypoint = 0: hxpoint = 0: hypoint = 0: 'initialize plot origins
pxpoint = 0: pypoint = -20: dxpoint = 0: dypoint = 0: 'on display screen
mxpoint = 0: mypoint = 0

ON ERROR GOTO Handler

CALL errstat(e$(), s$())
CALL fileinit(filenames, comcnt%, comment$())

CALL setup(deltat, nrnds%, knvm%, hpdlm%, hp3%, hp4%, e$(), s$(), erro%, lmt)

OPEN filenames FOR APPEND AS #1
FOR i = 1 TO comcnt%: PRINT #1, comments(i): NEXT i
PRINT #1, "# "; DATE$; " "; TIME$ 
PRINT #1, "# data pt. ", "time (sec) ", "hp 3455A V "; "Keithley "; "hp 34401 a "; "hp 34401 b "; "paro psi ";
CLOSE #1

begtime! = TIMER

FOR i = 1 TO nrnds%:
  FOR j = 1 TO 256
    tt = (begtime + ((i - 1) * 256 + j) * deltat): 'find time to take
    timeup = (1000 * tt) MOD 86400000:
    timeup! = timeup / 1000
    kk = tt \ 86400
    IF timeup! < deltat THEN
      IF timeup! < TIMER THEN 'waits for proper time to take data
        DO WHILE TIMER > timeup!
          LOOP
        END IF
      END IF
    END IF
    DO WHILE timeup! > TIMER
      LOOP
    CALL ibtrg(knvm%) 'trigger Keithley voltmeter
    IF ibsta% < 0 THEN CALL statanal(e$(), s$(), erro%)
    CALL ibtrg(hpdvm%) 'trigger HP 3455A voltmeter
    IF ibsta% < 0 THEN CALL statanal(e$(), s$(), erro%)
    CALL ibwrt(hp3%, red$) 'trigger HP 34401 a multimeter
  NEXT j
IF ibsta% < 0 THEN CALL statanal(e$( ), s$( ), erro%)

CALL ibwrt(hp4*/, red$) ' trigger HP 34401 b multimeter
IF ibsta% < 0 THEN CALL statanal(e$( ), s$( ), erro%)

rdtime! = (TIMER + (kk * 86400)) - begtime!

CALL ibrd(hpdvm*/, hpdvmout$) ' read HP 3455A voltmeter
IF ibsta*/, < 0 THEN CALL statanal(e$( ), s$( ), erro*/,)

CALL ibrd(hp3%, hp34401$) ' read HP 34401 a multimeter
CALL hpchk(erro%, hp3%)

CALL ibrd(hp4%, hpdmm$) ' read HP 34401 b multimeter
IF ibsta*/, < 0 THEN CALL statanal(e$( ), s$( ), erro*/,)
CALL hpchk(erro%, hp4%)

CALL ibrd(knvm%, knvmout$) ' read Keithley voltmeter
IF ibsta*/, < 0 THEN CALL statanal(e$( ), s$( ), erro*/,)

OPEN "coml:9600,n,8,1,cs,ds,lf" FOR RANDOM AS #2
PRINT #2, "*0100P3" ' trigger pressure gauge
INPUT #2, A$ ' read pressure gage
CLOSE #2

pout = VAL(MID$(A$, 6)) ' convert data to numeric form
hp3out = VAL(LEFT$(hp34401$, 15))
hp4out = VAL(LEFT$(hpdmm$, 15))
kout = VAL(RIGHT$(knvmout$, 14))
IF LEFT$(knvmout$, 1) = "0" THEN kout = 0
hpout = VAL(hpdvmout$)

nn% = 1
LINE (kxpoint, kypoint)-(rdtime!, kout), 10 ' plot data
kxpoint = POINT(2): kypoint = POINT(3) ' on screen

302 nn% = 2
LINE (hxpoint, hypoint)-(rdtime!, hpout), 13
hxpoint = POINT(2): hypoint = POINT(3)

312 nn% = 3
LINE (pxpoint, pypoint)-(rdtime!, pout / 10 - 20), 14
pxpoint = POINT(2): pypoint = POINT(3)

322 nn% = 4
LINE (dxpoint, dypoint)-(rdtime!, hp3out), 12
dxpoint = POINT(2): dypoint = POINT(3)

332 nn% = 5
LINE (mxpoint, mypoint)-(rdtime!, hp4out), 11
mxpoint = POINT(2): mypoint = POINT(3)

342 OPEN filenames FOR APPEND AS #1
PRINT #1, (i - 1) * 256 + j, rdtime!,
PRINT #1, USING "+.#####" " hpout, kout, hp3out, hp4out, pout
CLOSE #1
NEXT j
NEXT i

BEEP: BEEP: BEEP
DO: LOOP WHILE INKEY$ = ""
SCREEN 0: WIDTH 80

IF erro7 = -1 THEN
  PRINT "There was an error during execution."; CD; CAD \ ACA
  PRINT #1, "# there was an error during execution."
END IF

CLOSE
END

Handler:
SELECT CASE nn%
CASE 1
  kypoint = lmt: kxpoint = POINT(2)
  RESUME 302
CASE 2
  hypoint = lmt: hxpoint = POINT(2)
  RESUME 312
CASE 3
  pypoint = lmt: pxpoint = POINT(2)
  RESUME 322
CASE 4
  dypoint = lmt: dxpoint = POINT(2)
  RESUME 332
CASE 5
  mypoint = lmt: mxpoint = POINT(2)
  RESUME 342
END SELECT
RESUME NEXT

SUB bitfind (byte%, b%())
  ' This routine converts a decimal integer to binary
  ' in two's compliment representation, where b%(i) is
  ' the ith bit of the binary number. -1 = TRUE, 0 = FALSE.

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IF byte% < 0 THEN
    b%(15) = -1
    byte% = -byte% - 1
    FOR i% = 14 TO 0 STEP -1
        b%(i%) = NOT (-byte% \ (2 ^ i%))
        byte% = byte% MOD (2 ^ i%)
        NEXT i%
ELSE
    FOR i% = 15 TO 0 STEP -1
        b%(i%) = -byte% \ (2 ^ i%)
        byte% = byte% MOD (2 ^ i%)
        NEXT i%
END IF

END SUB

SUB errstat (e$(), s$())
    ' This subroutine sets the error codes and status codes
    e$(0) = "EDVR": e$(1) = "ECIC": e$(2) = "ENOL": e$(3) = "EADR"
    e$(4) = "EARG": e$(5) = "ESAC": e$(6) = "EABO": e$(7) = "ENEB"
    e$(10) = "ECIP": e$(11) = "ECAP": e$(12) = "EFSO": e$(14) = "EBUS"
    e$(15) = "ESTB": e$(16) = "ESQR"
    s$(15) = "ERR": s$(14) = "TIMO": s$(13) = "END": s$(12) = "SRQI"
    s$(11) = "RQS": s$(8) = "CMPL": s$(7) = "LOK": s$(6) = "REM"
    s$(5) = "CIC": s$(4) = "ATN": s$(3) = "TACS": s$(2) = "LACS"
    s$(1) = "DTAS": s$(0) = "DCAS"
END SUB

SUB fileinit (name$, i%, comments$())
    ' This subroutine gets the data file name & any comments
    CLS
    PRINT "Today is "; DATES; " if it is not, please enter"  'get file name
    INPUT "the proper date here (mm-dd-yyyy) .  ", d$  
    IF d$ <> "" THEN DATES = d$
    PRINT
    name$ = LEFT$(DATES, 2) + MIDS(DATES, 4, 2) + RIGHTS(DATES, 2)
    PRINT "The data file will be called "; name$; ".dat;"
    INPUT "enter additional characters to this name here ", A$
    name$ = name$ + A$ + ".dat"
    PRINT
    PRINT "If you do not want the file to be called "; name$; " ",
    123
INPUT "enter a complete different name with an extension here ", A$
IF A$ <> "" THEN name$ = A$
PRINT
PRINT "You may enter comments to be written at the beginning of the data"
PRINT "file now if you would like. Each line may contain up to 254"
PRINT "characters and you may enter up to 16 lines of comments."
PRINT "Please enter a blank line when you are finished."
PRINT
i% = 1: pound$ = CHR$(35) + CHR$(32)
DO
  IF i% = 17 THEN PRINT "No more comments for now! Hit <Enter> to continue"
  INPUT "", comment!(i'/.)
  IF comment!(i*/.) <> "" THEN comment!(i*/,) = pound! + comment!(i*/,)
  i% = i% + 1
LOOP UNTIL i% = 18 OR comment!(i*/. - 1) = ""

END SUB

SUB findtime (hpcom$, kcom$, hptim3, rr, mintime, lmt)

  ' this subroutine finds the minimum time between data points
  ' and the maximum voltmeter reading

  IF INSTR(kcom$, "TO") > 0 THEN ktime = .178 ELSE ktime = .269 'check set up to
  IF INSTR(hpcom$, "T1") > 0 AND INSTR(hpcom$, "HO") > 0 THEN ' find time
    hptime = .195
  ELSEIF INSTR(hpcom$, "T2") > 0 AND INSTR(hpcom$, "HO") > 0 THEN
    hptime = .057
  ELSEIF INSTR(hpcom$, "T2") > 0 AND INSTR(hpcom$, "HI") > 0 THEN
    hptime = .173
  ELSE hptime = .308
  END IF

  mintime = .299 + hptim3 + ktime + hptime: A% = 0

  k = INSTR(kcom$, "R"): kcode% = VAL(MID$(kcom$, k + 1, 1)) 'check set up
  hp = INSTR(hpcom$, "R"): hpcode% = VAL(MID$(hpcom$, hp + 1, 1)) ' for voltage

  SELECT CASE kcode%
    CASE 1
      krange = .002: mintime = 2 * mintime
    CASE 2
      krange = .02: mintime = 2 * mintime
    CASE 3

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krange = .2: mintime = 2 * mintime
CASE 4
    krange = 2
CASE 5
    krange = 20
CASE 6
    krange = 200
CASE 7
    krange = 1000
CASE ELSE
    krange = 1000
END SELECT

SELECT CASE hpcode%
CASE 1
    hprange = .1
CASE 2
    hprange = 1
CASE 3
    hprange = 10
CASE 4
    hprange = 100
CASE 5
    hprange = 1000
CASE 6
    hprange = 10000
CASE ELSE
    hprange = 100
END SELECT

IF krange > hprange THEN lmt = krange ELSE lmt = hprange
IF rr > lmt THEN lmt = rr

END SUB

SUB hpchk (erro%, hp%)
    
    This subroutine polls the hp34401 to check for errors

    hperr$ = "SYST:ERR?"
    hpexp$ = SPACE$(80)

    50 CALL ibwrt(hp%, hperr$)
    CALL ibrdChp%, hpexp$) IF MID$(hpexp$, 2, 1) <> "0" THEN
        PRINT hpexp$
        PRINT
        erro% = -1
END SUB

SUB setup (dt, n%, knvm%, hpdvms%, hp3%, hp4%, e$(), s$(), erro%, lmt)

' This subroutine gets relative data taking parameters and
' initializes the GPIB board & the voltmeters

DIM y(9), xx(9)
CLS

gpibO$="GPIB0":k$ ="KNVM":hp$ ="HPDVMS":hpdmms$ ="HP34401":hpdmms4$="HPDMM4":

'find GPIB board and voltmeters
CALL ibfind(gpibO$, gpib%)
PRINT "Finding GPIBO board: ";
CALL statanal(e$(), s$(), erro%): PRINT
CALL ibfind(k$, knvm%)
PRINT "Finding Keithley Nanovoltmeter: ";
CALL statanal(e$(), s$(), erro%): PRINT
CALL ibfind(hp$, hpdvms%)
PRINT "Finding Hewlett Packard Digital Voltmeter: ";
CALL statanal(e$(), s$(), erro%): PRINT
CALL ibfind(hpdmm$, hp3%)
PRINT "Finding Hewlett-Packard 34401 Digital Multimeter a:";
CALL statanal(e$(), s$(), erro%)
CALL hpchk(erro%, hp3%)
CALL ibfind(hpdmm4$, hp4%)
PRINT "Finding Hewlett-Packard 34401 Digital Multimeter b:";
CALL statanal(e$(), s$(), erro%)
CALL hpchk(erro%, hp4%)
PRINT

OPEN "COM1:9600,N,8,1,CS,DS,LF" FOR RANDOM AS #2
PRINT #2, "*0100VR": PRINT "Clearing ParoScientific Digiquartz Gauge"
CLOSE #2

CALL ibclr(knvm%)
'clear the voltmeters
PRINT "Clearing Keithley Nanovoltmeter: ";
CALL statanal(e$(), s$(), erro%)
CALL ibclr(hpdvm%)
PRINT "Clearing HP digital Voltmeter: ";
CALL statanal(e$(), s$(), erro%)
CALL ibclr(hp3%)
PRINT "Clearing HP 34401 digital multimeter a: \\
CALL statanal(e$(), s$(), erro%)
CALL hpchk(erro%, hp3%)
CALL ibclr(hp4%)
PRINT "Clearing HP 34401 digital multimeter b: \\
CALL statanal(e$(), s$(), erro%)
CALL hpchk(erro%, hp4%)
PRINT \\
kcom$ = "R5M0T1B0K0X"
hpcom$ = "F1R3T2A0H0"
hpcom3$ = "CONF:VOLT:DC 15,0.0001"
hpcom4$ = "CONF:VOLT:DC 15,0.0001"

' get commands to run Keithley
PRINT "The current commands to set up the Keithley Nanovoltmeter are"
PRINT " \\
PRINT "If you would like to send different commands, enter them now."
PRINT \\
PRINT "TO— cont. on talk (.178 sec) \nPRINT "T1—1 shot on talk (.269 sec) \nPRINT "T2— cont. on GET (.269 sec) \nPRINT "T3—1 shot on GET (.269 sec) \nPRINT "PO— disable FILTER \nPRINT "P1— FILTER off \nPRINT "P2— FILTER on \nPRINT "Y — Terminator \nPRINT : INPUT A$
IF A$ <> "" THEN kcom$ = A$
PRINT \\
'get commands to run HP 3455A
PRINT "The current commands to set up the HP Digital Voltmeter are:"
PRINT " \\
PRINT "If you would like to send different commands, enter them now."
PRINT \\
PRINT "T1—internal trigger \nPRINT "T1—internal trigger \nPRINT "T2—external trigger \nPRINT "T2—external trigger \nPRINT "A0— AUTO CAL off \nPRINT "A1— AUTO CAL on \nPRINT "D0— data ready RQS off \nPRINT "D1— data ready RQS on \nPRINT "B — binary program \\
PRINT : INPUT "", A$
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PRINT "  F6--test                M3--math off"
PRINT
INPUT "", A$
IF A$ <> "" THEN hpcom$ = A$
PRINT
'get commands for HP 344401a
PRINT"The current commands to set up the HP34401 Digital Multimeter a are:"
PRINT            "      "; hpcom3$
PRINT "If you would like a different settings, please choose appropriate number"
PRINT
PRINT "1. DC Volts"
PRINT "2. DC Volts Ratio"
PRINT "3. AC Volts"
PRINT "4. DC Current"
PRINT "5. AC Current"
PRINT "6. Resistance (2-terminal)"
PRINT "7. Resistance (4-terminal)"
PRINT "8. Frequency"
PRINT "9. Period"
INPUT "", b$
SELECT CASE b$
CASE "2"
  c$ = "VOLT:DC:RAT ">
CASE "3"
  c$ = "VOLT:AC "
CASE "4"
  c$ = "CURR:DC "
CASE "5"
  c$ = "CURR:AC "
CASE "6"
  c$ = "RES ">
CASE "7"
  c$ = "FRES "
CASE "8"
  c$ = "FREQ ">
CASE "9"
  c$ = "PER ">
CASE ELSE
  c$ = "VOLT:DC "
END SELECT
PRINT
INPUT "Please enter sensitivity (default = 15)", r$
IF r$ = "" THEN r$ = "15"
PRINT
PRINT "Please choose the # of digits and autozero (default 5 1/2 OFF)"
PRINT
PRINT " # of Digits AutoZero Measurement Time"
PRINT
PRINT "1. 4 1/2 OFF 0.0346"
PRINT "2. 4 1/2 ON 0.0352"
PRINT "3. 5 1/2 OFF 0.0346"
PRINT "4. 5 1/2 ON 0.0357"
PRINT "5. 6 1/2 OFF 0.1971"
PRINT "6. 6 1/2 ON 0.3625"
INPUT "", d$
SELECT CASE d$
    CASE "1"
        e = 4: zero$ = "ONCE": hptime = .0346
    CASE "2"
        e = 4: zero$ = "ON": hptime = .0346
    CASE "3"
        e = 5: zero$ = "ONCE": hptime = .0346
    CASE "4"
        e = 5: zero$ = "ON": hptime = .0357
    CASE "5"
        e = 6: zero$ = "ONCE": hptime = .1971
    CASE "6"
        e = 6: zero$ = "ON": hptime = .3625
    CASE ELSE
        e = 5: zero$ = "ONCE": hptime = .0346
END SELECT
rr! = VAL(r$): mr = rr!: mt = hptime
dig! = rr / (10! ^ e)
digs$ = STR$(dig!)
hp3com$ = "CONF:" + c$ + r$ + ",", + digs$
PRINT
PRINT "The setup for the HP34401a will be ": hp3com$
PRINT "with AutoZero set to ": zero$
PRINT
PRINT "Setting up HP34401a"
CALL ibwrt(hp3%, hp3com$)
CALL hpchk(erro 7 , , hp3%)
CALL statanal(e$(), s$(), erro%)
z$ = "ZERO:AUTO " + zero$
CALL ibwrt(hp3%, z$)

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CALL statanal(e$, s$, erro$)
CALL hpchk(erro$, hp3%)
PRINT
PRINT "If you wish to send other commands to the HP34401a, enter them now."
400 INPUT "", nhp$
IF nhp$ <> "" THEN
    CALL ibwrt(hp3%, nhp$)
    CALL hpchk(erro$, hp3%)
    CALL statanal(e$( ), s$( ), erro$)
    PRINT "Enter next command or <Enter> to continue"
    GOTO 400
END IF

'get commands to set up the HP34401b
PRINT"The current commands to set up the HP34401 Digital Multimeter b are:"
PRINT "                                "; hpcom4$
PRINT "If you would like a different settings,
        please choose appropriate number"
PRINT
PRINT "1. DC Volts"
PRINT "2. DC Volts Ratio"
PRINT "3. AC Volts"
PRINT "4. DC Current"
PRINT "5. AC Current"
PRINT "6. Resistance (2-terminal)"
PRINT "7. Resistance (4-terminal)"
PRINT "8. Frequency"
PRINT "9. Period"
INPUT "", b$
SELECT CASE b$
  CASE "2"
    c$ = "VOLT:DC:RAT "
  CASE "3"
    c$ = "VOLT:AC "
  CASE "4"
    c$ = "CURR:DC "
  CASE "5"
    c$ = "CURR:AC "
  CASE "6"
    c$ = "RES ">
  CASE "7"
    c$ = "FRES "
  CASE "8"
    c$ = "FREQ "
  CASE "9"
c$ = "PER "
CASE ELSE
c$ = "VOLT:DC"
END SELECT

PRINT
INPUT "Please enter sensitivity (default = 15)", r$
IF r$ = "" THEN r$ = "15"
PRINT
PRINT "Please choose the # of digits and autozero (default 5 1/2 OFF)"
PRINT
PRINT "# of Digits  AutoZero    Measurement Time"
PRINT
PRINT "1. 4 1/2  OFF  0.0346"
PRINT "2. 4 1/2    ON  0.0352"
PRINT "3. 5 1/2    OFF  0.0346"
PRINT "4. 5 1/2    ON  0.0357"
PRINT "5. 6 1/2    OFF  0.1971"
PRINT "6. 6 1/2    ON  0.3625"
INPUT "", d$
SELECT CASE d$
CASE "1"
  e = 4: zero$ = "ONCE": hptime = .0346
CASE "2"
  e = 4: zero$ = "ON": hptime = .0346
CASE "3"
  e = 5: zero$ = "ONCE": hptime = .0346
CASE "4"
  e = 5: zero$ = "ON": hptime = .0357
CASE "5"
  e = 6: zero$ = "ONCE": hptime = .1971
CASE "6"
  e = 6: zero$ = "ON": hptime = .3625
CASE ELSE
  e = 5: zero$ = "ONCE": hptime = .0346
END SELECT

rr! = VAL(r$)
IF rr! > mr THEN mr = rr!
mt = mt + hptime
dig! = rr / (10! * e)
digs$ = STR$(dig!)
hp4com$ = "CONF:" + c$ + r$ + "," + digs$
PRINT
PRINT "The setup for the HP34401b will be "; hp4com$
PRINT "with AutoZero set to " ; zero$
PRINT
PRINT "Setting up HP34401b"
CALL ibwrt(hp4%, hp4com$)
CALL hpchk(erro%, hp4%)
CALL statanal(e$(), s$(), erro%)
z$ = "ZERO: AUTO " + zero$
CALL ibwrt(hp4%, z$)
CALL statanal(e$(), s$(), erro%)
CALL hpchk(erro%, hp4%)
PRINT
PRINT "If you wish to send other commands to the HP34401b, enter them now."
425 INPUT nhp$
IF nhp$ <> "" THEN
    CALL ibwrt(hp4%, nhp$
    CALL hpchk(erro%, hp4%)
    CALL statanal(e$(), s$(), erro%)
    PRINT "Enter next command or <Enter> to continue"
    GOTO 425
END IF

trigs$ = "TRIG:SOUR IMM" 'set triggering for HP 34401a
CALL ibwrt(hp3%, trigs$)
CALL hpchk(erro%, hp3%)
PRINT "setting up triggering for HP34401a"
trigd$ = "TRIG:DEL 0"
CALL ibwrt(hp3%, trigd$)
CALL hpchk(erro%, hp3%)
PRINT "setting up trigger delay for HP34401a": PRINT

CALL ibwrt(hp4%, trigs$) 'set triggering for HP34401b
CALL hpchk(erro%, hp4%)
PRINT "setting up triggering for HP34401b"
CALL ibwrt(hp4%, trigd$)
CALL hpchk(erro%, hp4%)
PRINT "setting up trigger delay for HP34401b": PRINT

'get settings for Paroscientific Digiquartz Gauge
pr$ = "12"
PRINT "Please enter the pressure resolution factor (PR), an integer."
PRINT "The resolution of the digiquartz pressure gauge is 0.22/PR psi."
PRINT "The sampling time is 0.0053*PR + 0.553 seconds."
INPUT "The acceptable range is 1 < PR < 10000 (default = 12).", ppr$
450 IF ppr$ <> "" THEN ppr$ = ppr$
pp$ = "*0100EW*0100PR=" + pr$:
OPEN "COM1:9600,N,8,1,CS,DS,LF" FOR RANDOM AS #2
PRINT #2, pp$: INPUT #2, junk$
CLOSE #2
p! = VAL(pr$)
res! = .22 / p!; pgtimel = .553 + .0053 * p!
PRINT "The sample time is "; pgtimel; " seconds and"
PRINT "the resolution is "; res!; " psi."
PRINT
INPUT "If you want a different PR, enter it now", ppr$
IF ppr$ <> "" THEN GOTO 450
PRINT
mt = mt + pgtimel
CALL findtime(hpcom$, kcom$, mt, mr, mintime, lmt)

'find number of data points to take
PRINT "I will take an integer multiple of 256 data points: 256 N"
INPUT "Please enter your choice for N: ", n% ' get number of data points
500 PRINT : PRINT "The number of data points will be "; n% * 256
INPUT "If this is incorrect, please enter the proper N: ", A%
IF A% <> 0 THEN
   n% = A%
   GOTO 500
END IF
PRINT

'find time delay between data points
PRINT"The minimum time between data points that is safe is";mintime;"sec."
INPUT "Please enter the time between data points (in seconds): ", dt
IF dt < mintime THEN
   PRINT "This time is less than that to transfer data."
   INPUT "Please enter new time or <Enter> to risk it: ", x
   IF x > 0! THEN dt = x
END IF
IF dt > mintime THEN
   ddt = dt
ELSE ddt = mintime
END IF
totsec = n% * ddt * 256: PRINT
nhours = totsec \ 3600: resec = totsec MOD 3600
nmin = resec \ 60: resec = resec MOD 60
PRINT "Total running time is "; nhours; " hours "; nmin; " minutes ";
                   resec; " seconds."
INPUT "Is this correct (default is YES)"; A$
IF LEFT$(UCASE$(A$), 1) = "N" GOTO 500
PRINT
CALL ibwrt(knvm%, kcom$)  'write commands to set up voltmeter
PRINT "Setting up Keithley Nanovoltmeter: ";
CALL statanal(e$( ), s$( ), erro%)
PRINT
CALL ibwrt(hpdvm%, hpcom$)
PRINT "Setting up Hewlett Packard Digital Voltmeter: ";
CALL statanal(e$( ), s$( ), erro%)
PRINT
INPUT "Press <Enter> to begin data collection", A$

y(0) = 1: y(1) = 4: y(2) = 8: y(3) = 12: y(4) = 16
y(5) = 19: y(6) = 23: y(7) = 27: y(8) = 30
xx(1) = 12: xx(2) = 22: xx(3) = 31: xx(4) = 41
xx(5) = 51: xx(6) = 60: xx(7) = 70: xx(8) = 76

SCREEN 12  'set up graphics screen
VIEW (30, 0)-(639, 479)
WINDOW (0, -lmt)-(n% * ddt * 256, lmt)
LOCATE 1,28,0:PRINT "VOLTAGE (volts) and PRESSURE (*10 psi) vs TIME (sec)";
LOCATE 2, 32, 0: PRINT "Note: -20 equals 0 psi!"
LOCATE 3, 38, 0: PRINT "Keithley: Green";
LOCATE 4, 38, 0: PRINT "HP dvm: Magenta";
LOCATE 5, 38, 0: PRINT "PARO: Yellow";
LOCATE 6, 38, 0: PRINT "HP 34401a: Red";
LOCATE 7, 38, 0: PRINT "HP 34401b: Cyan";
FOR i = 0 TO 8
  LINE (0, lmt - i * lmt / 4)-(n% * ddt * 2, lmt - i * lmt / 4)
  LINE (i * ddt * n% * 32, lmt / 75)-(i * ddt * n% * 32, -lmt / 75)
  LOCATE y(i), 1, 0: PRINT lmt - i * lmt / 4;
  IF i > 0 THEN LOCATE 17, xx(i), 0: PRINT INT(i * totsec / 8 + .5);
NEXT i
LINE (0, -lmt)-(0, lmt)
LINE (n% * ddt * 256, 0)-(0, 0)
END SUB

SUB statanal (e$( ), s$( ), erro%)
  ' This subroutine reports the status codes and error codes
DIM bit%(16)
CALL bitfind(ibsta%, bit%())
FOR i = 0 TO 14
  IF bit%(i) THEN PRINT s$(i); " ";
NEXT i
IF bit%(15) THEN
    PRINT s$(15)
    PRINT e$(iberr%)
    erro% = -1
END IF
END SUB
BIBLIOGRAPHY


[30] In Ref. [28], an approximate solution of the Boltzmann equation was found by assuming $a_c(q)$ to be independent of $q$. After integrating the collision integral approximately, $a_c(q)$ was found to be proportional to $q^2$. Fig. 2.2 shows the actual behavior of this function. Reference [29] points out the error first noticed by Leggett and ter Haar and explores some of the consequences of the decreased scattering at low temperatures. One is that a small amount of dissolved $^4$He might not affect the $^3$He superfluid transition into the B-phase. The derivation of $D$ is not shown and the resulting formula is written with a "~" instead of an "=". This might imply that it was derived partly by dimensional analysis.


