70-23,488

CHANG, Donald Choy, 1942-
THE DIFFUSION COEFFICIENT OF He\(^3\) IN He\(^3\) - He\(^4\)
SOLUTIONS.

Rice University, Ph.D., 1970
Physics, solid state

University Microfilms, A XEROX Company, Ann Arbor, Michigan
RICE UNIVERSITY

THE DIFFUSION COEFFICIENT OF $^{3}\text{He}$

IN $^{3}\text{He} - ^{4}\text{He}$ SOLUTIONS

by

Donald Choy Chang

A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY

Thesis Director's signature:

Houston, Texas

May 1970
ACKNOWLEDGEMENT

I wish to express my sincere gratitude to Professor Harold E. Rorschach, Jr., for his direction and encouragement throughout this work, and to Professor Paul L. Donoho for his technical advice. I would like to thank Professor Riki Kobayashi for serving on my oral committee. The cooperation from my colleagues in the Low Temperature Laboratory of Rice University is also deeply appreciated.

I am indebted to Rice University and the National Science Foundation for their financial support. I would also like to thank the U.S. Naval Office for their supply of helium gas.
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acknowledgement</td>
<td>I.1</td>
</tr>
<tr>
<td>I. INTRODUCTION</td>
<td>I.1</td>
</tr>
<tr>
<td>II. METHOD OF MEASUREMENT</td>
<td></td>
</tr>
<tr>
<td>II.1 Spin-echo</td>
<td>II.1</td>
</tr>
<tr>
<td>II.2 Relaxation Time $T_1$</td>
<td>II.4</td>
</tr>
<tr>
<td>II.3 Diffusion</td>
<td>II.5</td>
</tr>
<tr>
<td>II.4 Convection</td>
<td>II.8</td>
</tr>
<tr>
<td>III. APPARATUS</td>
<td></td>
</tr>
<tr>
<td>III.1 Pulsed NMR Spectrometer</td>
<td>III.1</td>
</tr>
<tr>
<td>III.2 Sample Gas Handling System</td>
<td>III.11</td>
</tr>
<tr>
<td>III.3 Cryostat</td>
<td>III.12</td>
</tr>
<tr>
<td>III.4 Cryogenics</td>
<td>III.13</td>
</tr>
<tr>
<td>IV. EXPERIMENT PROTOCOLS PROCEDURES</td>
<td></td>
</tr>
<tr>
<td>IV.1 Sample Preparation</td>
<td>IV.1</td>
</tr>
<tr>
<td>IV.2 Sample Condensation</td>
<td>IV.2</td>
</tr>
<tr>
<td>IV.3 Diffusion Measurement</td>
<td>IV.3</td>
</tr>
<tr>
<td>IV.4 Measurement of $T_1$</td>
<td>IV.7</td>
</tr>
</tbody>
</table>
V. DATA PROCESSING AND RESULT

V.1 Diffusion

V.2 Longitudinal Relaxation Time

VI. DISCUSSION

APPENDIX A

APPENDIX B

APPENDIX C

APPENDIX D

References
I. INTRODUCTION:

Because of the exceptionally weak interaction between the He atoms, liquid helium has been of particular interest to the physicist. It provides a unique, simple system in which one can study the effects of quantum statistics on macroscopic physical properties. All three types of statistics, namely, the Bose-Einstein statistics, the Fermi-Dirac statistics and the Boltzmann statistics are represented in the liquid helium system of different isotopes (He$^3$ and He$^4$) and at different temperatures. Among these systems, pure liquid He$^4$ has been studied most expensively, and its superfluid properites are rather well understood at present.\textsuperscript{1,2} Liquid He$^3$ is somewhat less thoroughly understood. The ideal Fermi gas model is inadequate for this system, because there are contradictions between the theoretical predictions and the experimental results.\textsuperscript{2} The Fermi liquid theory, a phenomenological theory developed by Landau, has removed some of these contradictions.\textsuperscript{3,4} In this model a new phenomenon called "zero sound"\textsuperscript{5} was also predicted and was verified by experiment.\textsuperscript{6} But no theory based on first principles was attempted until Brueckner and Gammel\textsuperscript{7} published their paper in 1958. Recently, with the help of the development
of many-body techniques, theoretical study in this direction has received great attention. For example, the binding energy due to two-body and three-body correlations in liquid He$^3$ has been calculated by Østgaard. In the same theory he also had some success in calculating the parameters employed in Landau's Fermi-liquid model.

On the other hand, dilute solutions of He$^3$ in He II (superfluid helium) represent another class of systems, and has also received a great amount of attention. The earliest interest in this system was to study the effect of the foreign particles (He$^3$) in superfluid He$^4$. A useful model was proposed by Landau and Pomeranchuk. They suggested that the He$^3$ atoms dissolved in the He II can be regarded as weakly interacting quasi-particles with effective mass $m^*$; the energy-momentum relation of this quasi-particle is similar to that of an elementary excitation, the roton. Then the dilute solution can be regarded as a mixture of two components: (1) The background superfluid which does not interact (2) The normal fluid which consists of a gas of elementary excitations, namely, phonons, rotons and He$^3$ quasi-particles. Based on this model, Khalatinkov and Zharkov solved the Boltzmann equation and calculated the transport coefficients of He$^3$ in He II on the basis of certain simpli-
fying assumptions concerning the character of the He$^3$-roton, 
He$^3$-phonon and He$^3$-He$^3$ scattering. Later, after the phase 
separation phenomenon was discovered,$^{17}$ a new interest in 
dilute He$^3$-He$^4$ solutions at low temperature arose. As pointed 
out by Bardeen et al,$^{18}$ dilute solutions of He$^3$ in He$^4$ are 
of special interest for at least three reasons:

1. They represent a new set of Fermi liquids, whose density, 
and hence degeneracy temperature, may be varied at will.

2. The effective interaction between the He$^3$ quasi-particles 
is sufficiently weak that perturbation theory may be 
used to calculate the thermodynamic parameters and trans-
port properties.

3. The effective interaction is attractive, so that at 
sufficiently low temperatures there might be a super-
fluid transition associated with the He$^3$.

Extensive studies of this system using slightly different 
effective interaction potentials have been carried out by 
Bardeen, Baym and Pines,$^{18}$ Emery,$^{19}$ Baym and Ebner,$^{20}$ etc.
A theory of Fermi-Bose quantum liquids was also developed 
by Khalatnikov$^{21}$ in the same spirit as was done by Landau 
for Fermi liquids.$^{3,4}$ Experimental work on Helium 3 solutions 
has been done mostly at low concentrations, Anderson et al.$^{22}$
has measured the diffusion coefficient of 1.3% and 5% $^{3}$He solutions at temperatures above 10 m K. Garwin and Reich, who were the first to apply spin-echo techniques to the study of liquid helium, measured the diffusion coefficient and relaxation time of pressurized 1% and 2% $^{3}$He solutions above 0.5 K. Opfer et al measured the diffusion coefficient and nuclear magnetic susceptibility of 0.03% and 3% $^{3}$He-$^{4}$He II solutions above 0.4 K. Some other measurements of the transport coefficients of dilute $^{3}$He-$^{4}$He solutions have also been reported.

As we review the overall situation of the liquid helium problem, we see that the only area which is relatively little explored is the non-dilute $^{3}$He-$^{4}$He solutions. This is not surprising, because the complexity of this system makes it very hard to propose any treatable theoretical model, since many approximation and perturbation techniques can not be applied. Because of this lack of theoretical prediction and support, only a few experimental measurements have been made in this concentration range.

However, it is important to understand the physics of the transition from one system (the dilute solution) to another (the pure liquid $^{3}$He). It is the purpose of this work to
provide some useful information to bridge this gap. In particular, we try to answer the following questions:

(1) Up to what concentration limit of $\text{He}^3$ can the dilute solution model be applied? Ptukha measured the diffusion coefficient of 0.0139% and 0.132% $\text{He}^3$ solutions above 1.5 K and found the results in satisfactory agreement with the Khalatnikov-Zharkov theory. Horvitz and Rorschach, by use of a simple wall relaxation argument, were able to deduce the diffusion coefficient from longitudinal relaxation time measurements on 5% and 14% $\text{He}^3$ solutions. They found their 5% data agreed with the K.Z. theory but the 14% data did not. We wished to determine the transport coefficient (i.e. diffusion coefficient in this case) precisely for a variety of $\text{He}^3$ concentrations and hoped to find out in what region the K.Z. theory, and hence the Landau Pomeranchuk model, holds.

(2) What is the behavior of the transport properties of $\text{He}^3$ in superfluid He II for a concentration that is too high to apply the dilute solution model? Knowing this behavior, some physical mechanism can perhaps be suggested.

(3) Is there a discontinuity in the transport coefficients
when the He\textsuperscript{3}–He\textsuperscript{4} solution undergoes the lambda transition\textsuperscript{29}.

Although several measurements have been made in the low temperature region, no one has yet measured the diffusion coefficient at a temperature high enough to show the behavior of the diffusion near the lambda point. We would like to investigate if there is any abnormal behavior in the transport coefficient when T approaches the lambda point.

(4) What are the behavior of the transport coefficients for the He\textsuperscript{3}–He\textsuperscript{4} solution in the normal fluid phase? Does the solution behave exactly like a classical fluid?

In this study, the diffusion coefficient and relaxation times of 5\%, 9\%, 14\% and 24\% He\textsuperscript{3} in He\textsuperscript{4} solutions were measured. Some useful information has been obtained from our results. The method of measurement used in this work is the "Method B" of the spin-echo technique, the detail of which will be given in the next chapter.
II. METHOD OF MEASUREMENT:

1. Spin-echo:

The spin-echo method was first discovered by Hahn\(^{30}\) and then extended by Carr and Purcell.\(^{31}\) The principle is very simple. Consider a spin system sitting in a slightly inhomogeneous static magnetic field

\[
H_Z = H_0 + \vec{G} \cdot \vec{r}
\]  

(II-1)

The spins will precess with different Larmor frequencies. Suppose at time \(t < 0\), the system is thermalized, and all spins are parallel to the static field. At \(t = 0\), a 90° pulse, which is a r.f. pulse with pulsewidth \(\Delta T_{90°}\) such that

\[
\int_{0}^{\Delta T_{90°}} \gamma H_1 \, dt = \frac{\pi}{2}
\]

(II-2)

is applied (\(\gamma\) is the gyromagnetic ratio, \(H_1\) is the applied r.f. field which rotates around the static field at the Larmor frequency \(\gamma H_0\)). This 90° pulse will tilt the magnetization (the parallel spins) into a plane perpendicular to \(H_Z\). (We will call this the \(x-y\) plane). Now since each spin precesses at a different frequency, the spins will soon spread out like a fan, and the magnetization in this \(x-y\) plane then decays. Actually it can be shown\(^{31}\)
\[ M(t) = \int_{\text{sample volume}} \rho(r) \cos(\mathbf{G} \cdot \mathbf{r} t) d^3r \]  

(II-3)

where \( \rho(r) \) is the spin density and \( M(t) \) is the transverse magnetization. As the fan spreads wider and wider, the spins eventually become completely out of phase. Then suppose at a time \( t = \tau \), a 180° pulse is applied. (The 180° pulse is similar to a 90° pulse except

\[ \int_0^{\Delta T_{180}} \gamma H_1 dt = \pi \]  

(II-4)

This pulse will reverse the order of the spins in the x-y plane. This means that the spin which precesses faster now is behind the spin which precesses more slowly, and the phase difference between the two (spins) is equal to the precession frequency difference multiplied by \( \tau \). It is obvious that, after a time period \( \tau \) (i.e. \( t = 2\tau \)), the faster spin will catch up with the slower one. This argument holds true for any two spins. Therefore, all spins will become in phase at the moment \( t = 2\tau \). The transverse magnetization is thus reconstructed. (Or, we can say, the spins are refocused by the 180° pulse).

If a pickup coil oriented in the x-y plane surrounds the sample, we will see an induction signal as the magnetization
is tilted by the $90^\circ$ pulse. This induction signal quickly decays because of the spin fan-out. (For this reason it is often called an "induction tail"). After the $180^\circ$ pulse is applied at $t = \tau$, we will see another induction signal at $t = 2\tau$ due to the reconstruction of the transverse magnetization. This second induction signal is called an "echo" or "spin-echo". From the preceding argument it is clear that the echo actually is two induction tails back-to-back.

It should be also mentioned, if more than one $180^\circ$ pulse is applied, each of them will refocus the spin system once and generates an echo (provided the pulse separation is long enough for refocusing). For example, if r.f. pulses in a $90^\circ - \tau - 180^\circ - 2\tau - 180^\circ - \cdots$ sequence are applied to a sample, echoes will arise at $t = 2\tau, 4\tau, 6\tau$, etc..

The foregoing discussion considers a rather "free" spin system. In an actual physical system, the order of spins is not always completely reversible. There are mechanisms which couple the spins with their neighbors or other systems. These couplings will destroy the spin order irreversibly, and therefore make the reconstructed transverse magnetization smaller than the initial one. Since the number of spins being irreversibly spread-out increases with time, the echo amplitude
declines as the echo gets farther from the 90° pulse. The amplitude decay follows an exponential form and can be characterized by a time constant $T_2$, which is called the "Spin-spin relaxation time" or "Transverse relaxation time". The echo amplitude then is described by

$$m(t) = m_o e^{-t/T_2} \quad \text{(II-5)}$$

where $t$ is the time between the 90° pulse and the echo (e.g. $t$ equals to $2 \tau$ for the first echo). Actually this expression can be obtained by solving the Bloch equation

$$\frac{d \hat{M}}{dt} = \gamma \hat{M} \times \hat{H} - \frac{M_x \hat{X} + M_y \hat{Y}}{T_2} - \frac{M_x - M_o}{T_1} \quad \text{(II-6)}$$

for the magnetization component in the $x$-$y$ plane. An additional factor $e^{i\omega t}$ will appear in the solution, but since $m(t)$ concerns only the amplitude of the echo, this factor can be dropped.

2. Relaxation Time $T_1$:

Now let us consider what happens to the longitudinal magnetization after it is completely destroyed at $t = 0$. Due to the interactions between the spins and the lattice, the spins and the wall, etc., the spins will be gradually thermalized to occupy lower energy levels by alignment parallel to the
static field. The growth of the longitudinal magnetization can be calculated from the Bloch equation by considering only the $z$ component. The solution is

$$M_z(t) = M_0(1 - e^{-t/T_1})$$  \hspace{1cm} (II-8)

The growth rate is therefore characterized by a time constant $T_1$, which is called the "Spin - lattice relaxation time" or the "Longitudinal relaxation time".

Then suppose at a later moment $t$, we apply a $90^\circ - \tau - 180^\circ$ pulse sequence to excite the sample. An echo will arise, and the amplitude of this echo is

$$m = m_0 e^{-\tau/T_2}$$

$$= M_0(1 - e^{-t/T_1}) e^{-\tau/T_2}$$  \hspace{1cm} (II-9)

because $m_0$ is essentially equal to $M_z$ (Remember $m_0$ was generated by tilting $M_z$ into the x-y plane). So if we keep $\tau$ constant and change $t$, then

$$m(t) = m(\infty) \cdot (1 - e^{-t/T_1})$$  \hspace{1cm} (II-10)

Using this relation $T_1$ can be easily measured.

3. Diffusion:
When we consider the spin system of a fluid sample, an additional type of irreversible spread-out other than those already discussed will occur. It is due to the diffusion of spins inside the sample. Recall in our earlier discussion, the result that the 180° pulse is able to refocus the spins at \( t = 2 \tau \), is based on the assumption that the Larmor frequency of a spin is kept constant, so that the phase difference between any two spins developed in the time between \( t = 0 \) and \( t = \tau \), can be exactly compensated by their precession difference in the time between \( t = \tau \) to \( t = 2 \tau \). Then it is clear, as a spin drifts to a different region of the sample by diffusion, it will experience a different local field and so change its precession frequency. Now the phase compensation would no longer be exact, and the echo amplitude is damped. This damping had been investigated by Carr and Purcell\(^{31}\) in their well known paper in 1954. They were able to derive an expression for this damping by employing a random-walk model. Later it was shown by Torry\(^{37}\) that the diffusion damping can be evaluated rather easily by adding a diffusion term\(^{35}\)

\[ \nabla \cdot D \nabla M \]

to the Bloch equation (\( D \) is the diffusion coefficient and
M is the magnetization. The expression for the total echo decay (relaxation and diffusion) can be found by solving this generalized Bloch equation

\[
\frac{\text{d}}{\text{d}t} \mathbf{M} = \gamma \mathbf{M} \times \mathbf{H} - \frac{M_x \hat{x} + M_y \hat{y}}{T_2} - \frac{M_z - M_0}{T_1} + D \nabla^2 M \quad (II-11)
\]

For the \(n\)th echo arising at \(t\), the echo amplitude is given as

\[
m(t) = m_0 \exp\left[-\frac{t}{T_2} - D\gamma^2 G^2 t^3 / 12n^2 \right] \quad (II-12)
\]

where \(G\) is the magnetic field gradient and \(T_2\), \(D\), \(\gamma\) are as previously defined. This expression turns out to be exactly the same as that derived by Carr and Purcell. A detailed discussion of the diffusion term and the solution of the generalized Bloch equation is given in Appendix A of this thesis.

Two kinds of pulse sequences are commonly used in the diffusion measurement experiments. The first kind in principle consists of only one 90° pulse and one 180° pulse \(31\) (i.e. 90°-\(\tau\)-180°). \(n\) in equation (II-12) is then equal to one. \(D\) is measured by comparing the echo decay for different values of \(\tau\) (\(t = 2\tau\)). This method is called "Method A". This method is time consuming, because a time interval larger than the order of \(T_1\) must elapse before each measurement. In another method, called "Method B",
more than one 180° pulse is used in a pulse train. All the echoes needed will be obtained in a single measurement. The pulse sequence being used in this case is \( 90° - \tau - 180° - 2\tau - 180° - 2\tau - 180° - \cdots \). Echoes arise at

\[
t = 2n\tau
\]

with \( n = 1, 2, 3, \) etc.

Substituting (II-13) into (II-12), we get

\[
m(t) = m_0 \exp \left[ -\frac{t}{T_2} - \frac{1}{3} DT^2G^2\tau^2t \right]
\]

This is the relation that we want to use to measure \( D \).

4. Convection:

It should be pointed out that diffusion is not the only type of motion of molecules inside a sample that would make the spins drift so that they would experience different local fields at different times. Convection can also cause a similar effect. However, there is a great difference between these two types of motion. Diffusion represents the statistical net effect as a result of the individual Brownian motion; the directions in which the molecules move are completely random and independent of each other. Convection is a collective motion, in which a group of molecules moves in the same direction at roughly the same speed. We therefore would an-
ticipate that these two different types of motion would have
different effects on the spin-echo. The effect of convection
has been investigated by Carr and Purcell\textsuperscript{31} in a simplified
situation. Their result indicated that convection would not
irreversibly destroy the order of spins, (this is not hard
to imagine, since convection lacks the sense of randomness),
and it only contributes to a modulation effect and does not
cause an absolute decay of the echoes. To illustrate these
points, Carr and Purcell considered a convection current which
carries a group of nuclei containing spins in a direction such
that the magnetic field increases with time linearly. It can
be shown that the phase retardation of this group of spin
when the $n^{\text{th}}$ echo arises is

$$\begin{cases} 
\gamma h t^2 & \text{for odd } n \\
0 & \text{for even } n 
\end{cases}$$

(where $h$ is the field increment in unit time).

So we see the even numbered echoes are not affected by the
convection, while the odd numbered echoes are damped by a
constant factor.

We have extended the theory of Carr and Purcell to consider
the convection effect in general conditions. Our calcula-
tion (which will be given in Appendix B) shows that the con-
vection current may cause a very complicated interference
pattern. Nevertheless, it does not contribute any long-term effect to the echo decay. Therefore the convection current will not hinder the spin-echo measurement of diffusion.
III. APPARATUS:

1. Pulsed NMR Spectrometer:

The spectrometer components are the pulse sequence generator, transmitter, switch and impedance matching unit, tank circuit, receiver, electromagnet and the magnetic field gradient generator. A block diagram of the spectrometer is shown in Fig. III-1.

A special pulse train like that of Fig. III-2 is generated by the pulse sequence generator. These pulses are used to gate the transmitter which amplifies the 30 MHz r.f. wave generated from a General Radio type 1112-B Unit Oscillator. The output signal of the transmitter is shown in Fig. III-3. These transmitting pulses (90° pulses and 180° pulses) are sent to the tank circuit through the switch unit to excite the sample. The received signal is also picked up by the coil in the tank circuit. After being amplified by the receiver, the echoes are displayed on a Hewlett Packard 175 A oscilloscope and recorded by a HP 196 B scope camera. A more detailed description of the various components of the spectrometer is given in the following paragraphs:

Pulse Sequence Generator:
Fig. III-1
Pulsed NMR Spectrometer
Fig. III-2

Pulse train generated by the pulse sequence generator

Fig. III-3

Transmitting pulse
This generator is designed to generate a sequence of pulses of two different pulsewidths and variable pulse separation. The parameters which can be independently controlled in the experiment are summarized in the following table (Table III-1). This table also gives the range of these parameters in the actual experimental setup.

<table>
<thead>
<tr>
<th>Independently controllable parameters</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T_{90^\circ}$: Pulsewidth of the $90^\circ$ pulse</td>
<td>$10 \text{ sec} \sim 10 \mu\text{sec}$</td>
</tr>
<tr>
<td>$\Delta T_{180^\circ}$: Pulsewidth of the $180^\circ$ pulse</td>
<td>$10 \text{ sec} \sim 10 \mu\text{sec}$</td>
</tr>
<tr>
<td>$\tau$: Time lag between the $90^\circ$ pulse and the first $180^\circ$ pulse</td>
<td>$0 \sim T$</td>
</tr>
<tr>
<td>$t'$: Pulse separation between the $180^\circ$ pulses</td>
<td>$0.1 \text{ msec} \sim 10 \text{ sec}$</td>
</tr>
<tr>
<td>$N$: Number of $180^\circ$ pulses in a single pulse train</td>
<td>$0 \sim [1 \text{ sec}/t']$ Step function</td>
</tr>
<tr>
<td>$T$: Repetition period of the whole pulse train</td>
<td>$0.1 \text{ sec} \sim \infty$</td>
</tr>
</tbody>
</table>

Table III-1
This pulse sequence generator is built with a set of pulse generators (Tektronix type 161 pulse generator and type 162 waveform generator). These building-block generators can be arranged in various ways to generate a variety of pulse sequences. In the experiment to measure the diffusion coefficients these generators are programmed to generate the kind of pulse sequence shown in Fig.III-2. The arrangement in this case is illustrated in the block diagram in Fig.III-4.

The pulse output of the waveform generator 162(A) is used to trigger the pulse generator 161(A), which generates a 90° DC pulse (i.e. a DC pulse with the same pulsewidth as a 90° pulse). At the same time the negative sawtooth output from the 162(A) is sent to trigger the 161(B) and generate another pulse, which is used to gate the 162(B). The 90°-180° time lag $\tau$ is controlled by varying the triggering level of the 161(B); the pulsewidth of the output of the 161(B) also determines the number of 180° pulses; $t'$ is controlled by the repetition rate of the 162(B). The pulse output of 162(B) is sent to trigger the 161(C), which generates the 180° DC pulses. Finally the 90° DC pulse and the 180° DC pulses are mixed by an electronic mixer.

The gating signal of the FET gate is also provided by this
Transmitter Scope
Gate in trigger

Fig. III-4

Pulse Sequence Generator
pulse sequence generator. Since the gating pulse has to be wider than $\Delta T_{90^\circ}$ or $\Delta T_{180^\circ}$, an additional type 161 pulse generator (161(D)) is needed to generate these pulses. The differential circuit triggering is a means of providing a time lead for the gating pulses over the $90^\circ/180^\circ$ pulses.

Transmitter:

The transmitter is a gated power amplifier. It is tuned to operate at 30 MHz. The transmitter circuit diagram is shown in Fig.III-5. R.f. wave from the General Radio Unit Oscillator are coupled to the grid of a pentode 7788/E810F. This tube is normally cut off by a negative bias on the grid. When a positive $90^\circ/180^\circ$ DC pulse is sent in from the pulse sequence generator, the grid bias drops almost to zero and so turns on the pentode. The output of this pentode is amplified by two stages of push-pull power amplification. The isolation of this transmitter is very good. The r.f. leakage between pulses is always negligibly small ($<1 \mu$volt). The maximum peak-to-peak r.f. pulse height is on the order of 300 volts. The pulse height is controlled by the $B^+$ voltage of the output stage.

Tank Circuit:
The tank circuit is a simple solenoid 11/16" long and 3/8" I.D.. It is wound on a nylon support with #18 enamel copper wire. The total number of turns is 16. The inductance is about one microhenry when the sample chamber is not filled.

Switch and Impedance Matching Unit:

This spectrometer uses a common transmission line for the transmitter and the receiver. Since the transmitting pulse is on the order of hundred volts while the smallest echo is less than ten microvolts, these requirements must be satisfied:

(1) The amount of r.f. power transmitted to the sample is maximized.

(2) The echoes from the sample must be received by the receiver with the least loss.

(3) The leakage of the transmitting pulses into the receiver must be small enough not to damage or seriously saturate the receiver.

Condition (1) requires a strong coupling between the transmitter and the tank circuit. Condition (2) requires exactly the opposite. It requires the tank circuit to be strongly coupled with the receiver but separated from the transmitter. Condition (3) means that the receiver must be very well isolated
from the transmitter even though they have to share a common transmission line. If a magic Tee were available in this radio frequency range, the problem would be easily solved. But unfortunately it is not. So we must make some kind of fast automatic switch which connects the tank circuit and switches between the transmitter and the receiver. During the transmitting phase, the tank circuit - transmitter loop is shorted and the tank circuit - receiver loop is open; during the receiving phase, the switch is turned to the opposite position, i.e. tank circuit - transmitter open and tank circuit - receiver shorted. In this way all three requirements listed above could be satisfied. We utilize the non-linear property of a semiconductor diode to make this switch. Consider a pair of parallel diodes connected back to back with opposite polarity. For a large voltage signal (larger than one volt) this diode pair would look like a short circuit. But for a small voltage signal (say, less than 0.05 volt), this same diode pair would look like a very large resistor. Taking advantage of this property, we put a diode pair in series with the transmitter output, and another pair in parallel with the receiver input. (Please see Fig.III-6 for the actual arrangement). When the r.f. power is transmitted from the transmitter, the parallel diode pair protects the receiver
Transmitter

1N914B

12 PF 22 .01

Tank circuit

100K

5K .01

2N4252

-20v

Receiver

Fig.III-6

Switch and Impedance Match Unit
from serious saturation. The series diode pair isolates the transmitter from the received pulse. The parallel diode pair also becomes an open circuit in the receiving phase and does not cause any loss. Both condition (2) and (3) can thus be well satisfied. Condition (1) is not very well satisfied. But since the transmitter is more powerful than is needed, we can afford to sacrifice some of the r.f. power. The diode used in this switch unit is a fast silicon diode - type 1N914B. It can stand a reverse voltage larger than 100 volts. The switching time is about four nanoseconds.

Since the echoes are weak signals, to minimize the losses is not sufficient to get a good signal-to-noise ratio. We must also optimize the impedance match with the tank circuit so that the received signal transmitted to the receiver is maximized. A variety of ways have been tried. We finally decided to use an emitter-follower to separate the tank circuit and the receiver, and then match the output impedance of the tank circuit by a tunable inductor. The circuit diagram of this unit is shown in Fig.III-6.

A FET (Field Effect Transistor) gate was also constructed. The circuit diagram of the gate is shown in Fig.III-7. This gate is placed in the input of the preamplifier. Normally this
Fig. III-7

FET Gate
gate is on. When the 90°/180° pulses are transmitted, this gate will be turned off by the positive rectangular pulses generated in the pulse sequence generator. The receiver is then well protected.

Receiver:

Our receiver includes a low noise preamplifier (LEL Type IF-31-30-08-5) and a transistorized post-amplifier (LEL Type ITA-34-30-3-50). It is tuned to operate at 30 MHz with a bandwidth @ 3 db equal to 3 MHz. This rather wide bandwidth guarantees that the echo shape will not be distorted by the amplifiers. The maximum noise figure of the preamplifier is 1.5 db. The actual noise input level is about 1 microvolt. The highest combined voltage gain is 150 db. (The preamplifier was modified to give a higher gain and a narrower bandwidth than originally provided). The linearity of this receiver is good. (See Fig.III-8). The recovery time is a few microseconds. When the received signal is too large, we insert an attenuator (Shallcross r.f. attenuator) between the preamplifier and the post-amplifier to insure the linearity of the echoes. The power source of the preamplifier is a modified Hamner type H 105 powersupply. A Harrison Model 865C constant voltage/current powersupply provides the operating voltage
Preamp bias = -4.27 v
Post-amp bias = -1.2 v
The slope of the line is exactly 45°
for the post-amplifier. The power cable and connections are carefully shielded and grounded. The preamplifier, postamplifier and powersupplys all sit on a flat aluminum plate. This provides an effective common ground as well as a good heat sink.

Electromagnet:

The static magnetic field required in this experiment is provided by a Varian Model V 4012A Twelve-Inch Electromagnet System. This magnet provides a stable magnetic field up to 13.8 kilogauss. The air gap is 1.75 inches. The magnetic field is uniform within one tenth of a gauss in a region two inches in diameter in the center of the pole faces. This magnet is water-cooled. The power source is a Varian Model V 2100A Regulated Magnet Power Supply. The regulation is very good. The field is stable within 0.005% after a warmup period of two hours.

Magnetic Field Gradient Generator:

Fig.III-9 shows the magnetic field gradient generator. The central part is a field gradient coil pair. The operational principle of this pair is similar to an air-core Maxwell gradient pair, except that instead of having the two coils
Fig. III-9

Applied Field Gradient Generator
on parallel planes, our coils are bent to a cylindrical surface. This design is required mainly because of the limited space available. These coils are manufactured in the following way: We wind the #30 enamel copper wire in a mold to make a flat ten-turn single-layer coil. The dimension of this coil are 30 mm I.D. and 37 mm O.D.. The shape of this coil is fixed by applying several layers of Q C Polystyrene Q-Dope low loss coil coating. Before the coating is completely dry, the coil is compressed by another mold and bent to fit the surface of a cylinder which is 1 3/8 inches in diameter. Then we attach two of these identical coils on the pyrex glass outside wall of the liquid helium dewar. These coils are carefully aligned so that their center axis intersects with the dewar symmetry axis, and the intersection is also coincident with the center of the sample chamber.

In our experiment, the field gradient must be uniform throughout the whole sample volume. So the size and shape of the coil pair are rather critical. The theoretical value of the field gradient is very hard to calculate because the coil pair is placed inside a 1.75 inches electromagnet gap. We thus first make a simple calculation ignoring the electromagnet to find the approximate dimensions of the coil pair. The actual field gradient is then measured and the coil pair optimized by trial
and error. The result was very successful. Fig.III-10 shows the relative magnetic field versus distance along the center axis. The dependence is exceptional linear. The gradient therefore is very uniform. This measurement is made with a Bell Model 240 Incremental Gaussmeter. The probe used is a Bell ModelHFT 2401 Hall effect probe. The measurement of field two millimeters off the center axis gives a similar field gradient as that in Fig.III-10. Based on the data of this figure, the magnetic field gradient is

\[ \frac{G}{I} = 3.70 \text{ gauss/cm\cdotampere} \]

The current source of this field gradient generator is a Harrison 6265A DC powersupply. The maximum current output is three amperes.

2. Sample Gas Handling System:

The sample gas handling system is built with stainless steel components. 304 stainless steel tubing and 316 stainless steel Swagelok tube fittings was used to construct this system. The valves used are Nupro Type SS-4H bellow valves, which are bakable valves that are also made with 316 stainless steel. This system should work well in both pressure and vacuum systems. In the latter case the out-gassing time
I = 500 ma

G = 1.85 gauss/cm

Fig.III-10
required for purification should also be reduced to a minimum amount.

Fig.III-11 shows the schematic diagram of the sample gas handling system. The sample gas is stored in a half-litre stainless steel cylinder. A pyrex glass manometer is employed in the system so that the pressure of sample gas can be measured. The manometer also functions as a Toepler pump when the sample gas must be restored to the storage tank.

Other than the manometer, the only non-stainless-steel part of this system is the liquid helium cold trap. This trap is an H-shape copper structure filled with fine copper cotton. It is submerged in liquid helium, which is contained in a double jacket dewar. This cold trap can remove all the impurity gases in the sample gas system.

3. Cryostat :

The sample cell is made of nylon. The chamber is 4 mm long and 5 mm in diameter. The sample cell is sealed to a metal plug via a brass-nylon differential contraction seal. The detailed structure is shown in Fig.III-12. When the nylon cell is screwed tightly onto the brass plug, the wedge-shaped structure near the tip of the brass plug forms a good vacuum
Fig. III-11

Sample Gas Handling System
A : Brass Plug
B : Nylon Chamber
C : Nylon Piston
D : Sample Chamber

Fig.III-12
Sample Chamber
seal. We make the slope of the nylon cell at that point at a slightly steeper angle than the brass wedge, so that a good surface contact is always insured and the tightness becomes less critical. The seal is leakproof at room temperature. It becomes even tighter at lower temperature since nylon contracts more than brass. A nylon piston separates the brass plug from the sample chamber so that the r.f. field seen by the sample will not be distorted.

A copper shell surrounding the sample cell shields out external noise. The brass plug is soldered to a 304 stainless steel capillary (0.026" I.D.), through which the sample gas flows into the chamber. The capillary is copper coated. It is fixed in the center of a half inch O.D. thin wall 304 stainless steel tubing by a series of teflon separators. This coaxial tubing is used as a r.f. transmission line for the electronic signal (i.e. the tank circuit transmission line). This tubing also supports the weight of the sample cell.

4. Cryogenics:

The temperature range of interest in this experiment is obtained by reducing the vapor pressure of a liquid He\(^4\) bath.
The liquid helium dewar is made of pyrex glass. It has an extended tail section 1 3/8 inches O.D.. This tail section is placed inside the gap of the electromagnet. The helium dewar is surrounded by a metal liquid nitrogen dewar. This nitrogen dewar also has an extended tail section, which is about 1 3/4 inches O.D. and also sits inside the magnet gap. The jacket of the nitrogen dewar was evacuated with a diffusion pump and then sealed. The He dewar jacket is evacuated and flushed with air several times before it is cooled. A small amount of dry air (pressure about 1 torr) is allowed in the jacket when it is sealed. This gas helps to cool down the helium dewar to liquid nitrogen temperature. As liquid helium is transferred, the air in the jacket freezes and a high vacuum is created.

Fig. III-13 shows the cryogenic system. Temperatures down to 1.2 K are reached by pumping the liquid helium bath with a Kinney Model KD-30 mechanical pump. Below 1.2 K, a Consolidate Model KS-200 diffusion pump is used. The lowest temperature that can be reached is about 0.9 K. Three types of gauges are used to monitor the pressure of the liquid helium bath. A mercury manometer gives convenient and direct readings of the pressure. A Stokes McLeod gauge gives an accurate
reading in the low pressure region (below 5 mm Hg). A Texas Instrument Fused Quartz Precision Gage measures the relative pressure against a vacuum reference. The quartz gauge also compares the actual pressure with a reference setting. The electrical output resulting from this comparison is used to drive a servo motor, which in turn regulates the helium bath pressure. By this means a stable temperature is maintained. The variation of temperature is controlled by changing the reference setting in the quartz gauge. Below 1.2 K, the servo control does not operate. The temperature then is determined by the setting of a powerstat autotransformer, which controls the power fed into the diffusion pump heater.

This cryogenic system holds two and half litres of liquid helium and is sufficient to provide a twelve hour run.
IV. EXPERIMENTAL PROCEDURES:

1. Sample Preparation:

Four solution samples of 5%, 9%, 14% and 24% He\textsuperscript{3} concentration have been studied in this experiment. These samples were prepared in a sample preparation system and then sealed and put onto the sample gas handling system. Fig.IV-1 shows schematically the sample preparation apparatus.

A thick pyrex glass manometer 1.8 cm I.D. is used as a Toepppler pump. The whole system was carefully cleaned and evacuated with a diffusion pump. A small amount of He\textsuperscript{4} gas was admitted, and the stopcock and the storage tank valve were closed. The dead volume of the capillary and the upper part of the Toepppler pump was measured by observing the volume and pressure change as the trapped He\textsuperscript{4} gas was being compressed. A cathetometer was used to measure the mercury level. After knowing the dead volume, the storage tank valve was opened and the volume of the storage tank was measured with the same method.

Pure He\textsuperscript{4} gas (99.995% He\textsuperscript{4}, supplied by the U.S. Bureau of Mines) was admitted into the storage tank and the Toepppler
Fig. IV-1

Sample Preparation System
pump. After the equilibrium pressure was measured, we closed the storage tank valve and pumped out the $\text{He}^4$ gas in the rest of the system. Since the volume and pressure of $\text{He}^4$ gas in the storage tank were known, the amount of $\text{He}^3$ gas needed to prepare the desired concentration solution sample could be easily calculated. This amount of $\text{He}^3$ gas ($> 99.9\% \text{He}^3$, 0.01% $\text{He}^4$; supplied by Monsanto Research Corp.) was admitted to the Toepler pump and the stopcock closed. The storage tank valve was then opened, and the $\text{He}^3$ gas was compressed into the tank. In order to ensure that the $\text{He}^3$ gas was well mixed with the $\text{He}^4$ gas, the gas was pumped back and forth between the Toepler pump and the storage tank several times. Finally the gas mixture was compressed into the storage tank and sealed off by closing the tank valve. This sample tank was then ready for use in the experiment.

2. Sample Condensation:

On each run, the cryogenic system is first cooled with liquid nitrogen for about three hours. Then liquid helium is transferred into the liquid helium cold trap and the experimental L $\text{He}$ dewar. The liquid helium dewar system is sealed from the atmosphere after the transfer, and the pressure is gradually
reduced until the desired temperature is reached.

Since He\(^3\) gas condenses at a lower temperature than He\(^4\) gas does, the concentration of He\(^3\) in the liquid sample may be lower than that in the gas sample, if the sample gas is allowed to condense gradually starting at 4.2 K. To avoid this fractionation, we established the following guidelines:

1. Allow the sample gas to condense at a temperature below the condensation temperature of He\(^3\) and He\(^4\).
2. Isolate the storage tank from the sample chamber and thereby reduce the gas phase volume. The manometer was utilized as an intermediate carrier. First the manometer is isolated from the chamber, and sample gas is admitted from the storage tank. Then the storage tank is isolated and the sample gas is pumped into the sample chamber through the L He trap. These procedures are repeated until the sample chamber is filled. At the end the LHe trap is isolated. A by-pass loop sustains the link between the sample chamber and the manometer.

3. Diffusion Measurement:

All electronics are first turned on and allowed to warm up for about half an hour. \(\Delta T_{90^\circ}\) is set to be about 40 \(\mu\) sec. \(\Delta T_{180^\circ}\) is twice \(\Delta T_{90^\circ}\). The transmitting pulses are carefully
adjusted until their envelopes look rectangular. The pulse amplitude is adjustable and is controlled by the $B^+$ voltage of the transmitter. For test purpose we first employ a simple $90^\circ-\tau-180^\circ$ pulse train.

Since the relaxation time $T_1$ of our $^3\text{He}$ sample is on the order of several minutes, it is very inconvenient to hunt for the echo directly. There are at least half a dozen parameters that can be adjusted in this experiment. It is unpractical to turn a knob a little bit and then wait several minutes to see what happens. A proton probe was therefore used to test and set the electronics. This probe is placed in the magnet gap near the $^3\text{He}^-\text{He}^4$ solution sample. The tank circuit and transmission line of this proton probe are duplicates of those actually used in the experiment. The sample content of this probe is dilute $\text{Fe(NO}_3)_3$ water solution, the $T_1$ of which is about 10 m sec. This proton probe is connected to the pulsed NMR spectrometer and the proton echo observed. This signal is used to optimize the settings of the spectrometer (such as matching the tank circuit output impedance etc.). The spectrometer is then switched to the $^3\text{He}^-\text{He}^4$ solution sample. Since the operational frequency is kept constant, most of the settings
are still valid after the switching. Only the static field must be changed, together with the 90°/180° pulse amplitude. The field is calculated to be 9.25 kilogausses. The exact setting is determined by the shape of the echo. The amplitude of the 90°/180° pulses is also adjusted to give: (1) maximum signal amplitude of the 90° induction tail and echo, (2) no induction tail following the 180° pulse. Fig.IV-2a shows a good echo with proper settings. In our experiment we always observe the echoes directly in the r.f. output, thus avoiding any possible distortion caused by the demodulation process and eliminating the baseline shift problem. Fig.IV-2b shows a good echo when a field gradient of 1.85 gauss/cm is applied.

Now the 90°-τ 180°-t'-180°-t'- ····· pulse sequence can be applied. In the Carr and Purcell method B, t' is exactly 2τ. In our experiment we set τ equal to 10 m sec. The echoes of this pulse sequence are again checked. If the 90°(180°) pulses are not exactly 90°(180°), the echoes would have structure like those shown in Fig.IV-3a. When the 90°/180° pulses are properly adjusted, the echoes will look like those in Fig.IV-3b. We verify there that the 180° pulses are really 180°, because they reflect the echoes without a jump. As a magnetic field gradient is applied, the width
Fig.IV-2a

Sweep: 20 m sec/cm  Applied G = 0

Fig.IV-2b

Sweep: 20 m sec/cm  Applied G = 1.85 gauss/cm
Fig.IV-3a

Sweep: 20 m sec/cm  Applied G = 0

Fig.IV-3b

Sweep: 20 m sec/cm  Applied G = 0
of the echo is greatly reduced and the amplitude decays rapidly. This is because the magnetic field gradient enhances the diffusion effect. (See equation (II-14) of Chapter II). It is by observing this echo decay that the diffusion coefficient can be measured. Fig.IV-4a shows a good echo sequence for an applied field gradient of 1.85 gauss/cm. When the 180° pulse differs slightly from its correct value, induction tails will appear following the 180° pulses. Fig.IV-4b illustrates the received signal in this case.

In the course of making measurements, the sample is always kept in thermal equilibrium at a stable temperature. To maintain a constant He³ concentration, evaporation of the sample mixture is prevented by following the procedure of starting a run by filling the cell at a higher temperature, and going monotonically to lower temperature during the run. Before a spin-echo measurement is made, the magnetization of the sample is first destroyed by applying a series of fast 90°-180°-90°-180°-··· pulses, and then the sample is permitted to relax for a time longer than T₁. This precaution prevents the interference of any possible spurious echoes with our measurement.
Fig.IV-4a
Sweep: 20 m sec/cm    Applied G = 1.85 gauss/cm

Fig.IV-4b
Sweep: 20 m sec/cm    Applied G = 1.85 gauss/cm
4. Measurement of Longitudinal Relaxation Time:

In measuring $T_1$, the transmitter and receiver are adjusted in the same manner as in the diffusion measurement, except a different pulse sequence $90^\circ - \tau' - 180^\circ - \tau'' - 90^\circ - \tau' - 180^\circ - \cdots - 90^\circ - \tau' - 180^\circ - t - 90^\circ - \tau - 180^\circ$ is applied. Here $\tau'$ and $\tau''$ are on the order of 10 m sec and 100 m sec respectively; $t$ varies from seconds to minutes. The pulses before $t$ are used to destroy the initial magnetic ordering of the sample; the pulses $90^\circ - \tau - 180^\circ$ are used to produce the echo. Since the magnitude of this echo is directly proportional to the magnetization of the sample (at time $t$), the evolution of the magnetization with time can be traced easily by observing the echoes with increasing settings of $t$. In our experiment, pictures of echoes corresponding to six different values of $t$ are taken. Usually they are recorded in one photograph by a multiple exposure technique.
V. DATA PROCESSING AND RESULT:

1. Diffusion:

As mentioned before, we have used the Carr and Purcell method B to measure the diffusion coefficient. All the echoes obtained in one measurement are recorded on a Polaroid picture. These echoes generally resemble those shown in Fig.IV-4a. The amplitude of these echoes is measured with a precision vernier caliper. The total background (noise plus CRT linewidth) is regarded as the width of the baseline, and is subtracted from the echo height. Then the reduced echo amplitude is plotted semilogarithmically versus time. In our measurements, this plot is always closely linear, as expected theoretically. (See Chapter II).

A typical example of our semilog plot is shown in Fig.V-1.

According to equation (II-14), the slope of the line (i.e. the time constant of the exponential decay of the echoes) is given as

\[ \frac{1}{T_2^*} = \frac{1}{T_2} + \left( \frac{1}{3} D \gamma^2 G^2 c^2 \right) \]  

(V-1)

For liquid He\(^3\), the transverse relaxation time \(T_2\) is very long (larger than seconds; see Appendix C), compared to
\( T = 1.70 \text{ K} \)
\( 24\% \text{ He}^3 \)
\( \frac{1}{T_2^*} = 15.42 \text{ sec}^{-1} \)
\( G = 2.96 \text{ gauss/cm} \)

\( T = 1.70 \text{ K} \)
\( 24\% \text{ He}^3 \)
\( \frac{1}{T_2^*} = 14.39 \text{ sec}^{-1} \)
\( G = -2.96 \text{ gauss/cm} \)
the total decay time $T^*_2$, which is on the order of 0.05 sec. Therefore, under our experimental conditions, $T^*_2$ can be neglected.

$$\frac{1}{T^*_2} = \frac{1}{3} \, D \, \gamma^2 G^2 \tau^2$$ (V-2)

Since $\gamma$ is known ( $\gamma = 2.038 \times 10^4$ gauss$^{-1}$ sec$^{-1}$), $G$ can be calculated from the current by equation (III-1), and $\tau$ is set in the experiment, $D$ can then be directly deduced from the slope of the exponential decay constant $T^*_2$.

One may suppose that, due to the imperfection of the magnetic pole faces and the presence of other material in the gap, there is a background field gradient present in the sample other than that produced by the field gradient coils. This background field gradient in fact is very small. Before the cryostat was constructed, all materials which were placed in the gap were carefully tested under high field to make sure that they were non-magnetic. (The stainless steel parts were slightly magnetic, but they are kept away from the gap by using brass extensions). The actual background field gradient can be estimated from the echoes in Fig.IV-2 by comparing the widths of the echoes with and without an applied field gradient. This comparison shows
that the background field gradient is less than 0.1 gauss/cm.
Compared to a typical value of applied $G$ (on the order of 2 gauss/cm), the background gradient is really small.

However, since $G$ appears as a square term in equation (V-2), the error due to $G_o$ (background field gradient) is doubled, and therefore cannot be neglected. To avoid this error, we always do the measurement twice; once with the applied $G$ in the positive direction and the other with $G$ negative. Then $1/T^*_2$ is averaged from these two measurements. From equation (V-2), we see

\[
\left( \frac{1}{T^*_2} \right)_{av} = \frac{1}{2} \left[ \frac{1}{3} D\gamma^2 (G+G_o)^2 \tau^2 + \frac{1}{3} D\gamma^2 (-G+G_o)^2 \tau^2 \right] \\
= \frac{1}{6} D\gamma^2 \tau^2 [(G+G_o)^2 + (G-G_o)^2] \\
= \frac{1}{3} D\gamma^2 (G^2 + G_o^2) \tau^2
\]

Since $G_o \sim 1/20 G$, $G_o^2 \sim 1/400 G^2$

Therefore, the error in $\left( \frac{1}{T^*_2} \right)_{av}$ due to $G_o$ is less than one percent.

In fact, we always make four measurements to find one data point. We first set $G$ equal to a certain value, and make two measurements with $\pm G$. Then we change $G$ to another value, and make two more measurements. In this way the accuracy of
the measured diffusion coefficient can be checked. $D$ is found to be independent of $G$. The diffusion coefficient measured with different $G$ values usually agrees within a few percent. For illustrative purposes, Fig.V-1 to V-4 shows the spin-echo measurements of a 24% $\text{He}^3$ solution sample at a temperature of 1.70 K. The average $1/T_2^*$ of the first two pictures gives a diffusion coefficient

$$D = 1.229 \times 10^{-4} \text{ cm}^2/\text{sec}$$

while the calculated value from the last two pictures gives

$$D = 1.240 \times 10^{-4} \text{ cm}^2/\text{sec}$$

The experimental result is then taken as the average of these two, i.e.

$$D = 1.23 \times 10^{-4} \text{ cm}^2/\text{sec}$$

This data averaging procedure should reduce the random error introduced in the line-fitting. The values of $D$ at other temperatures are determined in the same way. Fig.V-5 shows the result of our measurement of $D$ for a 24% $\text{He}^3\text{He}^3\text{He}^4$ solution sample in the temperature range between 0.9 K and 2.5 K. The data obtained in different runs are indicated by different symbols. The reproducibility is excellent. The smoothness of the curve suggests that our experimental error
T = 1.70 K
24% He\(^3\)

\[ \frac{1}{T_2} = 24.23 \text{ sec}^{-1} \]

G = 3.70 gauss/cm

---

T = 1.70 K
24% He\(^3\)

\[ \frac{1}{T^*} = 22.78 \text{ sec}^{-1} \]

G = -3.70 gauss/cm
24% He$^3$

- : First run
- : Second run
- : Third run

Fig. V-5
is very small. (It is estimated to be 3%).

Fig.V-6, Fig.V-7 and Fig.V-8 show the measured D of 14%, 9% and 5% He\textsuperscript{3} solution samples, respectively. The reproducibility of the 9% He\textsuperscript{3} data in the lowest temperature region is not very good. This is attributed to the fact that the sample is accidentally overfilled; the sample gas thus condenses in the capillary as well as the chamber. Since the He\textsuperscript{4} bath has to be pumped for a long time in order to reduce the temperature, the refrigerant liquid helium level is greatly reduced as the low temperature is reached. Some part of the filling capillary that was originally submerged in liquid helium now gradually warms up. The overfilled sample contained in this part then vaporizes. As the He\textsuperscript{3} has a much higher vapor pressure than He\textsuperscript{4}, most of the vaporized gas is He\textsuperscript{3}. Then the concentration of the whole sample will become more dilute than when it was originally condensed. In the low temperature region, as shown by our data, the diffusion is very sensitive to He\textsuperscript{3} concentration. Then the change of concentration due to the warming of the capillary would introduce a sizable error.

The reproducibility of the 14% He\textsuperscript{3} data in the low temperature
14% He$^3$

- : First run
- : Second run
× : Third run
△ : Fourth run

Fig. V-6
\begin{figure}
\centering
\includegraphics[width=\textwidth]{Fig.V-7}
\caption{Plot showing the relationship between temperature (Kelvin) and the variable \( D \) in the unit \( \text{cm}^2/\text{sec} \).}
\end{figure}

- \( \bullet \) : First run
- \( \circ \) : Second run
- \( \times \) : Third run
- \( \Delta \) : Fourth run

9\% He\textsuperscript{3}
5% He\textsuperscript{3}

- : First run
- : Second run
\triangle: Third run

Fig.V-8
region is also somewhat poor. Since the first three runs on this sample are consecutive runs while the fourth one is not, we doubt the error may be due to the variation of sample concentration when the sample was repurified for the last run.

The 5% and 9% He$^3$ concentration samples are especially prepared for this experiment. The uncertainty of the He$^3$ concentration is less than 0.1%. The 14% and 24% sample were originally prepared for other experiments. Their concentration was checked by measuring the vapor pressure. The uncertainty of these samples is about 1%.

2. Longitudinal Relaxation Time:

In the $T_1$ measurement, the echo height is governed by the relation (II-10), i.e.

$$ h(t) = h_o (1 - e^{-t/T_1}) $$

(V-3)

Since both $h_o$ and $T_1$ are unknown, we cannot make semilog plots to deduce $T_1$ as we did for D. There are some complex ways in which one can plot the data and determine $T_1$, but the error involved in those kinds of processing is usually large. Finally we decided to use the computer to do a least squares fit. Since a library program which can handle the
least squares fit of an exponential function is not available, we wrote a special program to do the job. (This program is rather simple - see Appendix D). All the computations are done on a remote terminal of a Burroughs B5500 Time Sharing System. Fig.V-9 shows the actual print-out from the computer. The print-out tells us the $T_1$, $h_0$ and the sum of the square errors

$$ERS = \sum_{i=1}^{n} [h_{\text{compute}}(t_i) - h_{\text{measured}}(t_i)]^2$$

Twenty points of the theoretical curve (i.e. equation(V-3)) are also given by the computer. Fig.V-10 shows how the computer-selected curve fits our data. The result is quite good.

The resulting longitudinal relaxation time of a 5% He$^3$ solution sample is plotted against temperature in Fig.V-11. The curve is not so smooth as the diffusion measurement. This is because (1) only one measurement is made at each temperature, (2) the relaxation time is highly sensitive to paramagnetic impurities. This makes the run-to-run reproducibility rather poor. The measured $T_1$ of the 14% and 24% He$^3$ solution samples are shown in Fig.V-12 & Fig.V-13.
1 TEMPERATURE = 1.850 DEG K
T1 = 88.8 SEC
HZERO = 2.028
ERS = 0.0140

<table>
<thead>
<tr>
<th>H (CM)</th>
<th>T (SEC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.315</td>
<td>15.0</td>
</tr>
<tr>
<td>0.581</td>
<td>30.0</td>
</tr>
<tr>
<td>0.806</td>
<td>45.0</td>
</tr>
<tr>
<td>0.996</td>
<td>60.0</td>
</tr>
<tr>
<td>1.157</td>
<td>75.0</td>
</tr>
<tr>
<td>1.292</td>
<td>90.0</td>
</tr>
<tr>
<td>1.406</td>
<td>105.0</td>
</tr>
<tr>
<td>1.503</td>
<td>120.0</td>
</tr>
<tr>
<td>1.585</td>
<td>135.0</td>
</tr>
<tr>
<td>1.653</td>
<td>150.0</td>
</tr>
<tr>
<td>1.712</td>
<td>165.0</td>
</tr>
<tr>
<td>1.761</td>
<td>180.0</td>
</tr>
<tr>
<td>1.802</td>
<td>195.0</td>
</tr>
<tr>
<td>1.837</td>
<td>210.0</td>
</tr>
<tr>
<td>1.867</td>
<td>225.0</td>
</tr>
<tr>
<td>1.892</td>
<td>240.0</td>
</tr>
<tr>
<td>1.913</td>
<td>255.0</td>
</tr>
<tr>
<td>1.931</td>
<td>270.0</td>
</tr>
<tr>
<td>1.946</td>
<td>285.0</td>
</tr>
<tr>
<td>1.959</td>
<td>300.0</td>
</tr>
</tbody>
</table>

Fig. V-9

Example of computer print-out in the least squares fit for $T_1$ data.
5\% \text{He}^3

○ : First run
△ : Second run
● : Third run

Fig. V-11
14% He\(^3\)

- : First run
- : Second run

Fig.V-12
24% He$^3$

- $E_1$ (sec)
- $T$ (Kelvin)

- ○: First run
- △: Second run
- ●: Third run

Fig.V-13
VI. DISCUSSION:

Let us look first at the general behavior of our results. Fig.VI-1 summerizes our data for the diffusion measurements. The diffusion coefficient of pure liquid He$^3$ as measured by Hart and Wheatley$^{41}$ is also given in the same figure for comparison purpose. From the plotting of this figure, some very useful information can be immediately deduced:

(1) Our data clearly show that there is no discontinuity for the spin diffusion when the solution undergoes the lambda phase transition.

(2) The spin diffusion above the lambda temperature is independent of He$^3$ concentration. The order of magnitude and temperature dependence is also somewhat similar to the diffusion of pure liquid He$^3$. In fact, it is reasonable to expect that the normal fluid solution would behave as a pure liquid, since the isotope should have very little effect on the atomic interaction in a temperature region where quantum statistical effects do not arise. (The mass difference in general is not important and can be regarded as a multiplicative factor in the strength of interaction). The considerable difference between the diffusion of the
Fig. VI-1
normal fluid solutions and pure liquid $^3$He shown in our data is not easy to explain. It might be due to the fact that our measurements are sufficiently close to the lambda temperature so that some quantum statistical effects might still affect the solution system. It will be useful to carry the diffusion measurements to higher temperatures so that this question can be resolved.

(3) The results of our measurements clearly indicate that the diffusion is highly sensitive to the $^3$He concentration in the superfluid solution (below $T_\lambda$). This is in direct contradiction to the prediction of Ebner, who argued that above 1 K, the spin diffusion coefficients of different concentration solutions (of $^3$He) become equal, indicating that the predominant limiting mechanism is the scattering of $^3$He from the thermal excitations of the background $^4$He.

In fact, it seems to be necessary to include the $^3$He-$^3$He scattering in order to understand our diffusion data. From Fig.VI-1, we see that in the He II region, the spin diffusion always goes up with decreasing temperature. The rate of increase varies with $^3$He concentration. The 5% data seems to increase with no indication of leveling-off at a lower temperature. For higher concentrations, such as
the 9% and 14% He\textsuperscript{3} solutions, some indication of leveling-off starts to appear. At even higher concentrations (the 24% solution), a maximum is shown at low temperatures.

The behavior of our data can be more or less understood on the basis of a two-fluid model of He II. Suppose the He\textsuperscript{3} particle does not interact with the superfluid background. The processes determining the transport properties of He\textsuperscript{3} then are the He\textsuperscript{3}–He\textsuperscript{3} scattering and the He\textsuperscript{3}–roton scattering. (At a temperature above 1 K, the roton is the important elementary excitation in the normal fluid).\textsuperscript{16}

Since the density of normal fluid in He II is a fast decaying function with decreasing T (see Fig.VI-2), the number of rotons decreases with temperature, and the mean-free-path of the He\textsuperscript{3}–roton scattering becomes longer. On the other hand, from the indication of the data above the lambda temperature, we expect that the He\textsuperscript{3}–He\textsuperscript{3} scattering in He II solutions would be similar to that in pure liquid He\textsuperscript{3}, except that the density is modified.* Then according to the liquid He\textsuperscript{3} diffusion measurements, the He\textsuperscript{3}–He\textsuperscript{3} scattering in our temperature region would cause the

* Further analysis of our data shows that this is not exactly correct. However, the overall argument still holds. Please see the latter part of this chapter.
Fig. VI-2

Normal fluid density $\rho_n$ as a function of temperature:
- ○ derived from oscillating disk experiments,
- ● from the velocity of second sound.
VI.4

diffusion coefficient to decrease when T decreases. Therefore we actually have two competing mechanisms which give different behavior for the temperature dependence of the diffusion coefficient. If the He$^3$ concentration is low, the mean-free-path of the He$^3$-He$^3$ scattering is very long. Then in our experimental temperature region the He$^3$-roton scattering dominates the transport phenomena. The diffusion coefficient then continues to increase as the temperature decreases. If the He$^3$ concentration is rather high, the He$^3$-roton scattering mean-free-path is still shorter than that of the He$^3$-He$^3$ scattering at high temperature (near $T_\lambda$). But as the temperature decreases, so does the roton density, and the mean-free-path of the He$^3$-roton scattering eventually becomes comparable to the He$^3$-He$^3$ scattering mean-free-path. The He$^3$-He$^3$ scattering then becomes more important, and we begin to see a leveling-off or a maximum.

A comparison between the calculation of the Khalatnikov-Zharkov (K.Z.) theory$^{16}$ and our experimental results is attempted. The K.Z. theory includes very cumbersome computations. In the temperature region of our experiment, the K.Z. theory predicts

$$D = 2.8 \times 10^{-6} \ e^{\Delta/kT} \left( \frac{p_{He}}{p_\alpha} \right)^2$$

(VI-1)
(where $\rho_{n0}$ is the density of the normal fluid component associated with thermal excitation, $\rho_n$ is the total density of the normal component of the solution, $\Delta$ is the roton activation energy). There are some difficulties involved in trying to fit this equation to our data. There are no reliable expressions for $\rho_n$ and $\rho_{n0}$ for $^3$He-$^4$He solutions in the range of concentration and temperature reported in this work. Pellam,\textsuperscript{44} Berezni an and Esel'son,\textsuperscript{45} and Dash and Taylor\textsuperscript{46} measured $\rho_n$ of several dilute solutions. They found that in general the normal density is a linear superposition of contributions from the $^3$He and $^4$He components. It was also theoretically suggested by Pomeranchuk\textsuperscript{48} that

$$\rho_n = \rho_{4n} + n_3 m_3^*$$

(VI-2)

(where $\rho_{4n}$ is the density of normal component in superfluid $^4$He; $n_3$ is the density of $^3$He atoms; $m_3^*$ is the effective mass of $^3$He in solution). However, it has been pointed out by Price\textsuperscript{49} that Pomeranchuk's formula fails to describe the solution density correctly at $T_a$. Neverthe less, (VI-2) is still a good working formula for $\rho_n$ not too close to $T_a$.\textsuperscript{2} This approximation is sufficient when a calculation like the K.Z. theory is used, which already
VI.6

involves some rather crude approximations.

In making the theory fit the data, we use the relation

\[ \rho_n = n_3 m_3^* = \frac{X N_a m_3}{V_{\text{mol}}} \cdot \frac{m_3^*}{m_3} = 3 \frac{X}{V_{\text{mol}}} \cdot \frac{m_3^*}{m_3} \]  \hspace{1cm} (VI-3)

where \( N_a \) is Avogadro's number, \( X = n_3/(n_3 + n_4) \) is the molar concentration; \( V_{\text{mol}} \) is the molar volume of solution, the value of which was given by Kerr and Taylor. We choose \( \Delta/k = 8.6 \text{ K} \). Fig.VI-3 shows how the K.Z. calculation compares with our results. The curve with "o" is a plot of equation (VI-1) with \( m_3^* = 2.8 \ m_3 \) (this value is used by Khalatnikov, see Ref. 15). The curve with "•" is also a plot of (VI-1) with a minimum value of \( m_3^* (m_3^* = m_3) \). It seems neither of these two curves fits our data. We therefore conclude that the Khalatnikov-Zharlov theory can not be applied for solutions with \( \text{He}^3 \) concentration equal or higher than 5%.

One may suspect that the disagreement between the K.Z. theory and our data may be related to the inaccuracy of the approximation (VI-2). This does not seem to be the case. We also use the interpolated values of \( \rho_n \) measured in \( \text{He}^3-\text{He}^4 \) solutions by Dash and Taylor. The resultant plot of equation (VI-1) is similar to the curves given in
$5\% \text{ He}^3$

- : Result of this work
- : K.Z. calculation ($m^*_3 = m_3$)
- : K.Z. calculation ($m^*_3 = 2.8 \, m_3$)

Fig. VI-3
We have also compared our results with the data on more dilute solutions obtained by other investigators. Fig.VI-4 shows the comparison between our data and the data obtained by Ptukha\textsuperscript{26} and Opfer et al.\textsuperscript{24} All these data seem to agree in behavior. The only non-dilute solution data available in the literature are those obtained by Husa et al.\textsuperscript{27} on a 12% $\text{He}^3$ solution sample. The comparison between their data and our experimental results is shown in Fig.VI-5. In the same figure, the diffusion coefficient of 5% and 14% $\text{He}^3$ solutions derived by Horvitz and Rorschach from measurements of $T_1$\textsuperscript{28} is also shown. Despite the fact that these were indirect measurements, the results are in reasonably good agreement with our direct measurement.

In the earlier part of this discussion, we argued that our data can be understood by considering two competing mechanisms due to $\text{He}^3$ - roton scattering and $\text{He}^3$ - $\text{He}^3$ scattering. This interpretation is also strongly supported by the data in Fig.VI-6. In this figure, the inverse of the diffusion coefficient at a certain temperature is plotted as a function of $\text{He}^3$ concentration. It is clearly indicated that these data can be fitted linearly within
Fig. VI-4

- \(0.132\% \text{ He}^3\) (by Ptukha)
- \(3\% \text{ He}^3\) (by Opfer et al)
- \(5\% \text{ He}^3\) (This work)
Fig. VI-5

- • : 5% $\text{He}^3$ (from $T_1$ measurement)
- ○ : 14% $\text{He}^3$
- △ : 12% $\text{He}^3$ (by Husa et al.)

Graph showing $D$ (in $10^{-4}$ cm$^2$/sec) as a function of temperature $T$ (in Kelvin) with data points and curves representing different isotopes of helium.
Fig.VI-6
experimental error. i.e.

\[ \frac{1}{D} = C_1 + C_2 X \]

(where \( X \) is the \( \text{He}^3 \) concentration, \( C_1 \) and \( C_2 \) are constants). This kind of plot suggests that the scattering mechanisms that determine \( D \) can be divided into two components:

\[ \frac{1}{D} = \frac{1}{D_1} + \frac{1}{D_2} \] \hspace{1cm} (VI-4)

where \( D_1 \) is the component which does not depend on the \( \text{He}^3 \) concentration, while \( D_2 \) is the component which is inversely proportional to it. Since the probability of a \( \text{He}^3 \) atom being scattered by a thermal excitation is independent of the concentration of \( \text{He}^3 \), we identify \( D_1 \) as the part of diffusion which arises from the \( \text{He}^3 - \text{roton scattering} \). Then it naturally follows that the concentration-dependent \( D_2 \) is that part of the diffusion due to \( \text{He}^3 - \text{He}^3 \) scattering. As we recall in the statistical theory that \( 1/D \propto X \) is a characteristic showing that the system obeys Boltzmann statistics, we see that the equilibrium state of the \( \text{He}^3 \) atoms dissolved in \( \text{He} \ II \) solutions in our experimental temperature region is governed by the Boltzmann distribution. Since the \( \text{He}^3 \) degeneracy temperature is known to be quite low (on the order of 0.5 K), this result is theoreti-
cally expected and agrees with the experimental results on nuclear susceptibility measurements. However, the temperature dependence of \( D_2 \) is very different. Fig.VI-7 shows \( D_2X \) as a function of temperature. Husa et al.\(^{51} \) have measured the nuclear susceptibility of He\(^3\) in He II solutions with a concentration varying from 2% to 15% He\(^3\). They concluded that the temperature dependence of the susceptibility data agree with the hard-sphere model calculation. This agreement is not seen in our data. The hard-sphere model predicts \( DX \propto T^{1/2} \), while our data show \( D_2X \propto T^{-2} \) (illustrated in Fig.VI-8).

Emery\(^{52} \) has calculated the spin diffusion coefficient of He\(^3\) in He\(^4\) solutions due to He\(^3\)-He\(^3\) scattering in a semi-classical region. His result is

\[
D = \frac{3}{8m^*n^3} \frac{kT}{\Omega_{12}} \tag{VI-5}
\]

where \( \Omega_{12}^{(\nu)} \) is a special case of

\[
\Omega_{12}^{(n,t)} = \left( \frac{kT}{\pi m^*} \right)^{\frac{1}{2}} \int_0^\infty d\gamma \ e^{-\gamma^2} \gamma^{2t+3} \ A^{(n)}(\gamma)
\]

and

\[
A^{(\nu)}(\gamma) = \frac{4\pi \hbar^2}{m^* kT} \sum_{k} (\ell+1) \sin^2 (\delta_{\ell+1} - \delta_{\ell})
\]

The density and temperature dependence of \( D \) then is
Fig. VI-7
\[ D \propto T^{3/2} n_3^{-1} \]

We see that although the \( n_3 \) dependence agrees with our result, the temperature dependence does not. Fukuda and Kubo\textsuperscript{53} have also considered the spin diffusion of \( \text{He}^3 \) in liquid \( \text{He} \ II \). Their calculation indicated that when the solute-solute interaction becomes appreciable, the spin diffusion \( D_s \) will be described by

\[
\frac{1}{D_s} = \frac{1}{D_{FB}} + \frac{1}{D_{FF}} \tag{VI-6}
\]

Where \( D_{FB} \) is the diffusion component due to \( \text{He}^3 - \text{He}^4 \) interaction, \( D_{FF} \) is that due to \( \text{He}^3 - \text{He}^3 \) interaction. This expression agrees with our fitting formula (VI-4).

In the nondegenerate region,

\[
D_{FF} \propto \frac{1}{n_3 T^2} \tag{VI-7}
\]

We see that the \( n_3 \) dependence agrees exactly with our result. The \( T \) dependence also fits our \( D_2 \) data at 1.2 K \( \leq T \leq 1.7 \) K. However, this theory also predicts \( D_{FB} \propto T^{-3} \) which does not seem to fit our data for \( D_1 \).

(The temperature dependence of \( D_1^{-1} \) is shown in Fig.VI-9).

Ebner\textsuperscript{43} extended the \( \text{He}^3 \) quasi-particle effective interaction proposed by Bardeen, Baym and Pines\textsuperscript{18} and calculated
Fig. VI-9
the spin diffusion of $^3$He in liquid He II at a temperature between 1 K and $2T_F$ ($T_F$ is the Fermi degeneracy temperature).

By the propor choice of a few parameters he was able to fit the experimental data. We would like to see this calculation extended above 1 K so that our data can be compared.

Finally we would like to mention that although the diffusion coefficient measured in the spin-echo method is generally called spin diffusion, it is essentially the diffusion associated with atomic motion. The diffusion coefficient due to spin exchange is very small; it was estimated to be on the order of $10^{-12}$ cm$^2$/sec.\textsuperscript{23} Comparing to our measured $D_s$ (on the order of $10^{-4}$ cm$^2$/sec) the spin exchange effect is negligible.

Our data on longitudinal relaxation times agree with the experimental results obtained by other investigators.\textsuperscript{23,24} The behavior of $T_1$ can be roughly described by

$$\frac{1}{T_1} \propto D \quad \text{(VI-8)}$$

This does not agree with the BPP theory\textsuperscript{34} which predicts

$$\frac{1}{T_1} \propto \frac{1}{D} \quad \text{(VI-9)}$$

But as pointed out by Low and Rorschach\textsuperscript{39} and Horvitz and Rorschach,\textsuperscript{28} the relaxation in general consists of two
components: the bulk relaxation and the wall relaxation. i.e.

\[
\frac{1}{T_1} \text{net} = \left( \frac{1}{T_1} \right)_{\text{bulk}} + \left( \frac{1}{T_1} \right)_{\text{wall}} \quad \text{(VI-10)}
\]

in which

\[
\left( \frac{1}{T_1} \right)_{\text{bulk}} \propto \frac{1}{D}
\]

and

\[
\left( \frac{1}{T_1} \right)_{\text{wall}} \propto D
\]

At low temperature, D is large, and the wall relaxation is the predominant mechanism. Therefore \( T_1 \) is inversely proportional to D.

In conclusion, our study shows that the diffusion coefficient, D, of \( \text{He}^3 \) in \( \text{He}^3 - \text{He}^4 \) solutions behaves remarkably differently at temperatures above and below \( T \). In the He I region, the behavior of the diffusion coefficient is independent of \( \text{He}^3 \) concentration and is similar to that of pure liquid \( \text{He}^3 \). When the solution undergoes the lambda transition, no discontinuity in D is found. Below the lambda temperature, the diffusion coefficient is highly sensitive to the concentration of \( \text{He}^3 \), suggesting that the \( \text{He}^3 - \text{He}^3 \) scattering, which has been neglected by some theoreticians above 1 K, is important for solutions whose
He$^3$ concentration is 5% or more. Two different contributions to the diffusion by different mechanisms are also clearly indicated. The diffusion due to He$^3$-roton scattering is independent of He$^3$ concentration, while the diffusion due to He$^3$-He$^3$ scattering is inversely proportional to $n_3$. The He$^3$ atoms are found to obey Boltzmann statistics as theoretically expected. Yet the He$^3$-He$^3$ interaction in He II solutions deduced from the diffusion measurements cannot be explained in a hard-sphere model. The dilute solution calculations are also shown to be inadequate. The interaction mechanism seems to vary smoothly between the dilute and non-dilute solutions. No basic difference is detected up to 24% of He$^3$ concentration.
APPENDIX A

The Effect of Diffusion on Spin-echo

Consider the component of the magnetic moment in one direction, say, x. Let us quantize the spins along this coordinate. For simplicity, we assume \( s = \frac{1}{2} \). (The final result is independent of the spin). Then we denote

\[
\begin{align*}
n_+ &= \text{density of spin with } s_x = \frac{1}{2} \\
n_- &= \text{density of spin with } s_x = -\frac{1}{2}
\end{align*}
\]

From Fick's law, the diffusion current of spin then is

\[
\vec{J}_\pm = -D \nabla n_\pm \tag{A-1}
\]

where the subscripts \( \pm \) labels the different current with \( s_x = \frac{1}{2} \) or \( s_x = -\frac{1}{2} \). (D is the diffusion coefficient). The net current of magnetic moment is

\[
(\vec{J}_+ - \vec{J}_-) = -\mu D \nabla (n_+ - n_-)

= -D \nabla [\mu (n_+ - n_-)]

= -D \nabla M_x \tag{A-2}
\]

The change of magnetization in a unit volume due to diffusion is then

\[
\int_{\text{Unit vol.}} \left( \frac{\partial M_x}{\partial t} \right) \text{diff} \,^3 r = -\int_{\text{surface}} \mu (\vec{J}_+ - \vec{J}_-) \cdot \hat{n} \, ds
\]
\[
\begin{align*}
&= \int_S \nabla M_x \cdot \hat{A} \, ds \\
&= \int_{\text{vol}} \nabla \cdot (D \nabla M_x) \, d^3r
\end{align*}
\]

\[
\therefore \quad \left( \frac{\partial M_x}{\partial t} \right)_{\text{diff}} = \nabla \cdot D \nabla M_x \tag{A-3}
\]

Similarly,
\[
\begin{align*}
\left( \frac{\partial M_y}{\partial t} \right)_{\text{diff}} &= \nabla \cdot D \nabla M_y \\
\left( \frac{\partial M_z}{\partial t} \right)_{\text{diff}} &= \nabla \cdot D \nabla M_z
\end{align*}
\]

\[
\therefore \quad \left( \frac{\partial \vec{M}}{\partial t} \right)_{\text{diff}} = \nabla \cdot D \nabla \vec{M} \tag{A-4}
\]

Therefore, if we want to take the diffusion effect into consideration, we must add this term to the phenomenological equation of Bloch, i.e.

\[
\frac{\partial \vec{M}}{\partial t} = \gamma \vec{M} \times \vec{H} - \frac{M_x \vec{M} + M_y \vec{J}}{T_2} - \frac{M_z - M_0}{T_1} A + \nabla \cdot D \nabla \vec{M} \tag{A-5}
\]

This is the so called "Generalized Bloch equation". In most case, D is a constant coefficient and so can be taken outside the divergence operator. To evaluate the spin-echo damping due to diffusion, we have to solve (A-5). Suppose the sample is small enough so that the slightly inhomogeneous magnetic field can be approximated as a homogeneous
field plus a uniform field gradient, i.e.

$$\vec{H} = (H_0 + \vec{G} \cdot \vec{r}) \hat{z}$$  \hspace{1cm} \text{(A-6)}$$

(Here we choose \( \hat{z} \) as the static field direction).

Denote the transverse magnetization

$$m = M_x + i M_y$$  \hspace{1cm} \text{(A-7)}$$

From (A-5) and (A-6),

$$\frac{\partial m}{\partial t} = -i m \gamma (H_0 + \vec{G} \cdot \vec{r}) - \frac{m}{T_2} + D \nabla^2 m$$  \hspace{1cm} \text{(A-8)}$$

If both \( G \) and \( D \) are negligible, the solution will be the ordinary Bloch equation solution

$$m = m_0 \exp \left\{ -i \omega_s t - t/T_2 \right\}$$

(\( \omega_s = \gamma H_0 \)). Since \( G, D \neq 0 \), we try a solution

$$m(\vec{r},t) = m_0 \varphi(\vec{r},t) \exp \left\{ -i \omega_s t - \frac{t}{T_2} \right\}$$  \hspace{1cm} \text{(A-9)}$$

Substituting this into (A-8), we get

$$\frac{\partial \varphi}{\partial t} = -i \gamma (\vec{G} \cdot \vec{r}) \varphi + D \nabla^2 \varphi$$  \hspace{1cm} \text{(A-10)}$$

Since the solution for \( D = 0 \) is

$$\varphi = \exp \left\{ -i \gamma (\vec{G} \cdot \vec{r}) t \right\}$$  \hspace{1cm} \text{(A-11)}$$
let us try a solution

$$\varphi(r,t) = A(t) \exp \left\{ -i \gamma (\vec{\alpha} \cdot \vec{r}) t \right\}$$  \hspace{1cm} (A-12)

Substitute in (A-10):

$$\frac{\partial A}{\partial t} = -A D \gamma^2 \vec{r}^2 t^2$$  \hspace{1cm} (A-13)

Then

$$A(t) = \exp \left\{ -\frac{1}{2} D \gamma^2 \vec{r}^2 t^2 \right\}$$  \hspace{1cm} (A-14)

This is the expression of a 90° induction tail decay.

However, if a pulse sequence like

$$90°-t-180°-2t-180°-2t-180°-\ldots\ldots$$

is applied, the relation (A-12) is no longer a proper trial solution. Since the phase changes whenever a 180° pulse is applied, (A-11) has to be modified. Let us assume that all 180° pulses are applied in such a way that the r.f. field is along the $y'$ axis of the rotating frame.

(The rotating frame rotates around the z axis of the lab frame with frequency $\omega_0$).

At $0 < t < t$, the phase of the spins at $r$ is

$$\phi(t) = -\gamma (\vec{\alpha} \cdot \vec{r}) t$$

At $t = t$, a 180° pulse is applied. The phase is flipped
from

\[ \phi(\tau-) = -\gamma (\vec{G} \cdot \vec{r}) \tau \]

to

\[ \phi(\tau+) = \pi + \gamma (\vec{G} \cdot \vec{r}) \tau \]

Then for \( \tau < t < 3\tau \)

\[ \phi(t) = \pi + \gamma (\vec{G} \cdot \vec{r}) \tau - \gamma (\vec{G} \cdot \vec{r})(t - 2\tau) \]

\[ = \pi - \gamma (\vec{G} \cdot \vec{r})(t - 2\tau) \]

At \( t = 3\tau \), another 180° pulse is applied, the phase is again flipped and to an angle

\[ \phi(3\tau+) = \pi - \phi(3\tau-) \]

\[ = \gamma (\vec{G} \cdot \vec{r}) \tau \]

For \( 3\tau < t < 5\tau \)

\[ \phi(t) = \gamma (\vec{G} \cdot \vec{r}) \tau - \gamma (\vec{G} \cdot \vec{r})(t - 3\tau) \]

\[ = -\gamma (\vec{G} \cdot \vec{r})(t - 4\tau) \]
In summary, for \((2n-1) \tau < t < (2n+1) \tau\)

\[
\phi(t) = -\gamma (\mathbf{q} \cdot \mathbf{v}) (t - 2n \tau) + \Theta(n) \tag{A-15}
\]

where

\[
\Theta(n) = \begin{cases} 
1 & \text{for odd } n \\
0 & \text{for even } n 
\end{cases}
\]

From this argument it is clear that the proper trial solution for (A-10) is

\[
\psi(\mathbf{r}, t) = A(t) \exp \left\{ -i \left[ \gamma (\mathbf{q} \cdot \mathbf{v}) (t - 2n \tau) + \Theta(n) \right] \right\} \tag{A-16}
\]

Substitute this in (A-10), then for \((2n-1) \tau < t < (2n+1) \tau\),

we get

\[
\frac{\partial A}{\partial t} = -A D \mathbf{q} \cdot \mathbf{v} (t - 2n \tau)^2 \tag{A-17}
\]

Integrate between \((2n-1) \tau\) and \((2n+1) \tau\):

\[
A(t) = A((2n-1) \tau) \exp \left\{ -\frac{1}{2} D \mathbf{q} \cdot \mathbf{v} (t - 2n \tau)^2 + \mathbf{r}^2 \right\} \tag{A-18}
\]

When an echo arises, \(t = 2n \tau\)

\[
A(2n \tau) = A((2n-1) \tau) \exp \left\{ \frac{1}{2} D \mathbf{q} \cdot \mathbf{v} \tau^2 \right\} \tag{A-19}
\]

Also from (A-18)

\[
A((2n+1) \tau) = A((2n-1) \tau) \exp \left\{ -\frac{3}{2} D \mathbf{q} \cdot \mathbf{v} \tau^2 \right\}
\]
Substitute this into (A-19) iteratively

\[ A(2n\tau) = A(t) \exp \left\{ -\frac{1}{2} D \mathbf{r}^2 \mathbf{g}^2 \tau^2 (2n-1)^2 \right\} \]

From (A-14)

\[ A(t) = \exp \left\{ -\frac{1}{2} D \mathbf{r}^2 \mathbf{g}^2 \tau^3 \right\} \]

\[ \therefore \quad A(2n\tau) = \exp \left\{ -\frac{1}{2} D \mathbf{r}^2 \mathbf{g}^2 \tau^3 (2n)^2 \right\} \]

Or, recall \( t = 2n\tau \),

\[ A(t) = \exp \left\{ -\frac{1}{2} D \mathbf{r}^2 \mathbf{g}^2 \tau^3 t \right\} \quad (A-20) \]

Substitute this in (A-16) and (A-9), the amplitude of the transverse magnetization when an echo arises then is

\[ m(t) = m_0 \exp \left\{ -\frac{t}{\tau_2} -\frac{1}{2} D \mathbf{r}^2 \mathbf{g}^2 \tau^2 t^2 \right\} \quad (A-21) \]

This is the result given in Chapter II.
APPENDIX B

The Effect of Convection Current on Spin-echo

Consider a group of nuclei containing nuclear spins that move uniformly in a region in which the magnetic field is inhomogeneous. The field seen by this group of nuclear spins can be written as

\[ H(t) = H_0 + \int_0^t \vec{G}(\vec{r}) \cdot \vec{dr} = H_0 + \int_0^t \vec{G}(t) \cdot \vec{v}(t) dt \quad (B-1) \]

where \( \vec{G} \) is the field gradient and \( \vec{v} \) is the velocity. For simplicity let us first assume both \( G \) and \( v \) are constant, then

\[ H = H_0 + ht \quad (B-2) \]

where \( h = \vec{G} \cdot \vec{v} \).

Since the echo amplitude is determined by the spin fan-out, let us consider this problem in a frame rotating with angular frequency \( \omega H_0 \). The phase angle of the group of spins in this rotating frame is then

\[ \phi_c(t) = \int_0^t \omega h t dt = \frac{1}{2} \omega h t^2 \quad (B-3) \]

After a 180° pulse applied at \( t = \tau \), the phase angle starts to decrease and becomes
\[
\phi_c(t) = \int_0^t \gamma h t \, dt - \frac{1}{2} \gamma h t^2 \\
= \frac{1}{2} \gamma h (t^2 - 2t^2)
\]  
(B-4)

When the echo arises at \( t = 2\tau \), this group of spins have a non-zero phase angle

\[
\phi_c(2\tau) = \gamma h \tau^2
\]

This means that the convection current forces a group of spins out of phase by \( \gamma h \tau^2 \) radians compared to those stationary spins. Therefore the first echo amplitude is damped.

Then at \( t = 3\tau \), another 180° pulse is applied. Now

\[
\phi_c(t) = \int_0^t \gamma h t \, dt - \phi_c(3\tau) \\
= \frac{1}{2} \gamma h (t^2 - 9\tau^2) - \frac{1}{2} \gamma h \tau^2 \\
= \frac{1}{2} \gamma h (t^2 - 16\tau^2)
\]  
(B-5)

The second echo arises at \( t = 4\tau \). At this moment

\[
\phi_c(4\tau) = 0
\]

This means the phase angle due to convection is exactly
cancelled out. The second echo therefore experiences no damping from the convection current!

This argument can be extended to the $n^{th}$ echo, since

$$
\phi_c(2n\tau) = \int_{(2n-1)\tau}^{2n\tau} y \dot{t} dt - \phi_c((2n-1)\tau)
$$

$$
= \int_{(2n-1)\tau}^{2n\tau} y \dot{t} dt - \left[ \int_{(2n-2)\tau}^{(2n-1)\tau} y \dot{t} dt + \phi_c((2n-2)\tau) \right]
$$

$$
= y h \tau^2 - \phi_c((2n-1)\tau) \tag{B-6}
$$

Substitute this equation into itself by putting $n \rightarrow n-1$

$$
\phi_c(2n\tau) = y h \tau^2 - \left[ y h \tau^2 - \phi_c(2(n-2)\tau) \right]
$$

$$
= \phi_c(2(n-2)\tau) \tag{B-7}
$$

Therefore, if $n$ is even

$$
\phi_c(2n\tau) = \phi_c(0) = 0 \tag{B-8}
$$

if $n$ is odd

$$
\phi_c(2n\tau) = \phi_c(2\tau) = y h \tau^2 \tag{B-9}
$$

So we see that the net effect of the convection is simply to reduce the amplitude of the odd numbered echoes, the over-all decay of the echoes is not affected.
The foregoing argument is somewhat idealistic. In a real physical system, \( \vec{G} \cdot \vec{v} \) in equation (B-1) would not be a constant. Due to the constrain of the wall, the convection current usually follows a curvilinear path instead of a straight one. In that case the increment of field should be expressed as

\[
\Delta H(t) = H(t) - H_0
= \int \vec{G}(t) \cdot \vec{v}(t) \, dt \tag{B-10}
\]

The phase retardation of an echo which arises at \( t = 2n\tau \) will be

\[
\phi_c(2n\tau) = \int_{(2n-1)\tau}^{2n\tau} \gamma \Delta H(t) \, dt - \phi_c((2n-1)\tau) \\
= \int_{(2n-1)\tau}^{2n\tau} \gamma \Delta H(t) \, dt - \left[ \int_{(2n-1)\tau}^{(2n-2)\tau} \gamma \Delta H(t) \, dt + \phi_c((2n-2)\tau) \right] \\
= \Delta \phi_c(2n\tau) - \phi_c((2n-2)\tau) \tag{B-11}
\]

Here

\[
\Delta \phi_c(n) = \int_{(2n-1)\tau}^{2n\tau} \gamma \Delta H(t) \, dt - \int_{(2n-2)\tau}^{(2n-1)\tau} \gamma \Delta H(t) \, dt \tag{B-12}
\]

is the phase retardation developed between the \((n-1)\)th echo and the \(n\)th echo due to the convection current.

Substitute (B-11) into itself, and recall \( \phi_c(0) = 0 \), then
\[ \phi_c(2n\tau) = \Delta \phi_c(n) - \Delta \phi_c(n-1) + \Delta \phi_c(n-2) - \cdots \]

\[ = \sum_{m=1}^{n} (-1)^{n-m} \Delta \phi_c(m) \quad (B-13) \]

To evaluate \( \Delta \phi_c(n) \), let us first expand \( \Delta H \) in a Taylor series:

\[ \Delta H(t) = h_o + h_1 t + h_2 t^2 + \cdots \quad (B-14) \]

where

\[ h_k = \frac{1}{k!} \left. \frac{d^k (\Delta H)}{dt^k} \right|_{t=0} \]

Substitute (B-14) into (B-12):

\[ \Delta \phi_c(m) = \int_{n(2m-1)\tau}^{2m\tau} \varphi(\sum_k h_k t^k) dt - \int_{n(2m-2)\tau}^{(2m-1)\tau} \varphi(\sum_k h_k t^k) dt \]

\[ = \varphi \left[ \sum_{k=1}^{(2m-1)\tau} h_k t^k \right]_{(2m-1)\tau}^{2m\tau} - \varphi \left[ \sum_{k=1}^{(2m-2)\tau} h_k t^k \right]_{(2m-2)\tau}^{(2m-1)\tau} \]

\[ = \varphi \sum_{k=1}^{(2m-1)\tau} h_k t^k \left[ 2m^k - 2(2m-1)^k + (2m-2)^k \right] \]

\[ = \sum_{k=1}^{(2m-1)\tau} \varphi_k(m) h_{k-1} \quad (B-15) \]

where

\[ \varphi_k(m) \equiv \frac{\varphi}{k} t^k \left[ 2m^k - 2(2m-1)^k + (2m-2)^k \right] \quad (B-16) \]
To evaluate the number inside the bracket, let us consider a function

\[ S_k(x) = (x+1)^k - 2x^k + (x-1)^k \]

\[ = \sum_{i=0}^{k} \binom{k}{i} x^i - 2x^k + \sum_{i=0}^{k} \binom{k}{i} (-x)^i x^{k-i} \]

\[ = \sum_{j=1}^{[k/2]} \binom{k}{2j} x^{k-2j} \]  \hspace{1cm} (B-17)

Comparing (B-17) with the bracket in (B-16), we see they are identical if we put \( x = 2m-1 \)

\[ \therefore \varphi_k(m) = \frac{2^{2k}}{k^k} \sum_{j=1}^{[k/2]} \binom{k}{2j} (2m-1)^{k-2j} \] \hspace{1cm} (B-18)

From this expression two properties of \( \varphi_k(m) \) are immediately recognized for any positive \( k \) and \( m \):

(1) \( \varphi_k(m) \equiv 0 \) \hspace{1cm} (B-19)

(2) \( \varphi_k(m) \equiv \varphi_k(m-1) \)

Then if we substitute (B-15) into (B-13)

\[ \Phi_c(2n \tau) = \sum_{m=1}^{n} (-1)^{n-m} [ \sum_{k=1}^{m} \varphi_k(m) h_{k-1} ] \]

\[ = \sum_{k=1}^{n} h_{k-1} [ \sum_{m=1}^{n} (-1)^{n-m} \varphi_k(m) ] \] \hspace{1cm} (B-20)
and denote

$$A_k(n) \equiv \sum_{m=1}^{n} (-1)^{n-m} \varphi_k(m)$$  \hspace{1cm} (B-21)$$

then

$$A_k(n) = [\varphi_k(n) - \varphi_k(n-1)] + [\varphi_k(n-2) - \varphi_k(n-3)] + \cdots \geq 0$$  \hspace{1cm} (B-22)$$

Similarly

$$A_k(n+1) = \sum_{m=1}^{n+1} (-1)^{n+1-m} \varphi_k(m) \geq 0$$

But since

$$A_k(n+1) = \varphi_k(n+1) - \sum_{m=1}^{n} (-1)^{n-m} \varphi_k(m)$$

$$= \varphi_k(n+1) - A_k(n)$$

then

$$A_k(n) \leq \varphi_k(n+1)$$  \hspace{1cm} (B-23)$$

or, combining (B-22) and (B-23)

$$0 \leq A_k(n) \leq \varphi_k(n+1)$$  \hspace{1cm} (B-24)$$

Substitute (B-21) into (B-20)

$$\varphi_c(2\pi \tau) = \sum_{k=1} h_k A_k(n)$$  \hspace{1cm} (B-25)$$
and compare it with (B-15)

$$\Delta \phi_c(m) = \sum_{k=1} h_k \phi_k(m)$$

We see that if all $h_k$'s are of the same sign, then (B-24) implies

$$0 \leq |\phi_c(2n\tau)| \leq |\Delta \phi_c(n+1)|$$  \hspace{1cm} (B-26)

$\Delta \phi_c(n+1)$ is the phase retardation developed between the $n^\text{th}$ echo and the $(n+1)^\text{th}$ echo as the convection current carries the group of spins from a region $\vec{r}(2n\tau)$ to a region $\vec{r}((2n+1)\tau)$. Since the field gradient and speed of the spin motion are roughly constant, $\Delta \phi_c(n+1)$ is always finite and insensitive to $n$. Then (B-26) shows: The accumulated phase retardation of the $n^\text{th}$ echo does not exceed the maximum phase retardation developed between two successive echoes.

In general, it is unlikely that all $h_k$'s will happen to have the same sign. But since in our discussion the time concerned is very short (it is the time interval between the 90° pulse and the echo, generally on the order of 0.05 sec), an approximation taking only the first few terms in the Taylor's series will be good enough for equation (B-14). This means that (B-26) will hold if only the first
few \( h_k \)'s are of the same sign.

Actually, the requirement that the \( h_k \)'s have to be of the same sign is not so strict. Suppose we neglect the cubic and higher terms in (B-14). Both (B-15) and (B-25) will terminate at \( k = 3 \). From (B-16)

\[
\phi_1(m) = 0
\]

(B-15) and (B-25) can be reduced to

\[
\Delta \phi_c(m) = h_1 \phi_2(m) + h_2 \phi_3(m) \quad \text{(B-15')}
\]

\[
\phi_c(\Delta n \tau) = h_1 A_2(n) + h_2 A_3(n) \quad \text{(B-25')}
\]

There are two possible cases:

Case (A) : \( h_1 \) and \( h_2 \) are of the same sign

Case (B) : \( h_1 \) and \( h_2 \) are of opposite sign

We do not need to consider case (A) because it already satisfies the condition for (B-26). In case (B), we assume
\( h_1 > 0 \). (If \( h_1 \) is negative, same conclusion can be obtained by using a similar argument). Then

\[
\phi_c(2n \tau) = h_1 A_2(n) - |h_2| A_3(n)
\]

Suppose \( \phi_c(2n \tau) < 0 \) for some \( n \), as shown in the following diagram

\[
\begin{align*}
|\phi_c(2n \tau)| &= h_1 A_2(n) - |h_2| A_3(n) \\
&\leq h_1 A_2(n) - |h_2| A_3(1) \\
\therefore A_2(n) &\leq \phi_2(n+1) = \phi_2(1) \\
\text{and} \quad A_3(1) &= \phi_3(1)
\end{align*}
\]

\[
\therefore |\phi_c(2n \tau)| \leq h_1 \phi_2(1) - |h_2| \phi_3(1) \\
&= |\Delta \phi_c(1)| \quad \text{(B-27)}
\]

So in either case (A) or case (B), the underlined statement in page 8 holds. Therefore, the convection current only
causes a finite fluctuation on the echo amplitude; it will not damp the echoes with time. The relations (B-18) and (B-20) also indicate that this fluctuation does not necessarily follow a regular pattern as shown by Carr and Purcell (i.e. equation (B-8) and (B-9)). It can be very complicated. The exact behavior will depend on the function $\Delta H(t)$. 
APPENDIX C

The Estimation of $T_2$

In the spin-echo measurement, the time constant of the echo decay is given in Chapter II as

$$\frac{1}{T_2^*} = \frac{1}{T_2} + \frac{1}{3} D \tau^2 G^2 \gamma^2$$  \hspace{1cm} (C-1)

In order to measure $D$, it is important to know $T_2$. In most experiments, including ours, $T_2$ is very difficult to measure, because the diffusion term always dominates the total relaxation time, and $T_2$ is buried in its random error. Theoretically, $T_2$ should be very large. According to the classical theory of Bloembergen, Purcell and Pound\(^{34}\) in the region that the correlation time $\tau_c \ll 1/\omega_0$, (this is true for liquid helium solutions),

$$\frac{1}{T_1} = \frac{\gamma^4 n^2 N_0}{a D} = \frac{1}{T_2}$$  \hspace{1cm} (C-2)

$T_1$ of the dilute solution as well as of pure $\text{He}_3$ has been measured by several groups\(^{23,24,39}\) and found to be quite long, generally on the order of one hundred seconds between 1 K and 2 K. We also measured the longitudinal relaxation time for non-dilute solutions and find that it agrees with
the results obtained from the other samples. Then, based on equation (C-2) one would expect $T_2$ to be on the order of one hundred seconds. However, this seems not the case, since people doing spin-echo experiments always find that $T_2$ is actually much shorter. Husa et al.\textsuperscript{27} reported that $T_2 \approx 0.08$ sec in their measurement for 2\% and 12\% He\textsuperscript{3}–He\textsuperscript{4} solution samples at temperature between 0.4 K and 1.26 K. Garwin and Reich\textsuperscript{23} found that $T_2$ is much larger than 0.2 sec in their study of the 1\% and 2\% solution samples. Later in the work of Opfer et al.\textsuperscript{24} it was reported that no transverse relaxation effect was evident in the diffusion data, this means that $T_2$ is much longer than 0.5 sec. In our work, $T_2$ has been checked for different samples and at different temperatures. We observe the echo decay without applying any field gradient. Then from equation (C-1) the lower limit of $T_2$ is given by the echo decay constant $T_2^*$. From data like those shown in Fig.IV-2b, $T_2^*$ is about 1.56 sec. Throughout our measurements we always find $T_2$ larger than one and half seconds.

The difference in the reported $T_2$'s can be understood by considering the following two factors: (1) For that part of the inhomogeneity of the static field which can not be
approximated as a uniform field gradient, there will be a
damping effect other than those treated by the diffusion
term in the equation (II-12). This effect can very easily
affect the measurement of $T_2$. We noticed that there is a
large difference in the field homogeneity for those magnets
employed in the foregoing mentioned experiments, and it
seems in most cases that the reported $T_2$ is correlated with
the field homogeneity. (2) Paramagnetic impurities usually
can not be completely avoided in the sample system (e.g.
gas absorbed on the chamber wall). The exchange between
the sample nuclear spins and these impurities will greatly
shorten $T_2$. Low, Rorschach and Schwettman$^{55}$ measured the
$T_2$ of pure liquid $\text{He}^3$ and found it to be about one order
of magnitude smaller than $T_1$. They concluded that this can
be attributed to the possible presence of paramagnetic
impurities. This impurity effect was also examined by
Bloembergen.$^{56}$ We believe that the anomalous $T_2$ shortening
seen in the spin-echo measurement is mainly due to this
effect.
APPENDIX D

A Simple Least Squares Fit Program For the Data Processing
of $T_1$

FILE: CHANG  TYPE: FORTRAN  --04/23/70  9:46 PM

101 DIMENSION H(9), T(9), HC(9)
110 READ 1, MT
120 1 FORMAT(I10)
130 DO 50 M=1, MT
140 READ 11, J, TMIN
150 11 FORMAT(I10, F10.1)
160 READ 12, (H(I), I=1, J)
170 12 FORMAT(9F8.3)
180 READ 13, (T(I), I=1, J)
190 13 FORMAT(9F8.1)
210 HZERO=H(J)*0.8
220 DELH=H(J)*0.03
230 DO 40 K=1, 20
240 HZERO=HZERO+DELH
250 T1=TMIN
251 DELT=TMIN*0.04
260 DO 40 L=1, 50
270 T1=T1+DELT
280 TDHS=0
290 DO 10 I=1, J
300 RT=-T(I)/T1
310 X=EXP(RT)
320 HC(I)=HZERO*(1-X)
330 DHS=(HC(I)-H(I))*S2
340 TDHS=TDHS+DHS
350 10 CONTINUE
360 IF(K.GT.1) GO TO 20
370 IF(L.EQ.1) GO TO 30
380 20 CONTINUE
390 IF(TDHS-ERS) 30, 40, 40
400 30 TIC=T1
410 HGC=HZERO
420 ERS=TDHS
430 40 CONTINUE
440 READ 14, TEMP, HE3
450 14 FORMAT(F10.3, F10.1)
460 PRINT 15, TEMP, HE3
470 15 FORMAT(15H1 TEMPERATURE =, F7.3, 6H DEG K,
     EX, F5.1, 6H % HE3)
480 PRINT 16, T1C, H0C, ERS
490 16 FORMAT(5H T1 =,F6.1,4H SEC,10X,7HHZERO =,F8.3,5X,SHERS =,F8.4/)
500 PRINT 17
510 17 FORMAT(1OH H (CM),10X,8H T (SEC))
520 TD=T1MIN*0.25
521 TP=0
530 DO 50 I=1,20
540 TP=TP+TD
550 RTP=-TP/T1C
560 XP=EXP(RTP)
570 HP=H0C*(1-XP)
580 PRINT 18, HP, TP
590 18 FORMAT(1H F7.3,10X,F8.1)
600 50 CONTINUE
610 STOP
620 END
REFERENCES:

1. R.P. Feynman, Progr. Low Temp. 1, 17, (1955)
15. I.M.Khalatnikov, THEOTY OF SUPERFLUIDITY, Benjamin, (1965)


25. J.J.M. Beenakker et al., Physica XVIII, 433 (1952)


30. E.L. Hahn, Phys.Rev. 80, 580 (1950)


33. F. Bloch, Phys.Rev. 70, 460 (1946)

34. N. Bloembergen, E.M. Purcell, and P.V. Pound, Phys.Rev. 73, 679 (1948)
35. N. Bloembergen, Physica XV, 386 (1949)
42. N.Bernardes and D.F. Brewer, Rev.Mod.Phys. 34, 190 (1962)
43. C. Ebner, Phys.Rev. 156, 222 (1967)
48. I. Pomeranchuk, Zh. eksp. teor. Fiz. 19, 42 (1949)
49. P.J. Price, Phys.Rev. 89, 1209 (1953)
52. V.J. Emery, Phs.Rev. 161, 194 (1967)