ABSTRACT

SWANK, CHRISTOPHER MARK. An Investigation in the Dynamics of Polarized Helium-3 in Superfluid Helium-4 for the Spallation Neutron Source (SNS) neutron-electric-dipole-moment (nEDM) experiment. (Under the direction of Robert Golub and Paul R. Huffman.)

To conserve CPT, charge conjugation-parity-time reversal symmetry, the last discrete symmetry believed to be conserved, there must exist T violation. A measurement of the neutron-electric-dipole-moment (nEDM) would be a direct observation of T violation. The measurement of a nEDM requires the detection of a small shift in the Larmor precession frequency due to an applied electric field. The most recent experiment trying to measure the nEDM has been restricted by a systematic effect termed the geometric phase, a frequency shift linear in the electric field of the experiment. A new nEDM experiment is being planned for the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory. It will use spin-polarized $^3$He as a comagnetometer and detector, and is susceptible to the geometric phase. To investigate, quantify and find ways to reduce this systematic effect is the main goal of this work.

The dynamics of polarized $^3$He in phonon dominated superfluid $^4$He is investigated. A theory that predicts relaxations and frequency shifts, and hence the geometric phase frequency shift, via correlation functions valid from the ballistic through to the diffuse regime for restricted geometries is presented. For the first time, the lower dimensional correlation functions are shown to be projections of the higher dimensional correlation functions with the proper velocity weighting. The correlation function was measured by gradient induced longitudinal relaxation. The presented theory is shown to adequately predict the data. A previous theory, termed the high frequency theory due to its accurate description of relaxation with spins precessing at approximately the same rate as gas collisions, is also valid in this region. The presented theory is shown to agree with the high frequency theory. The two theories diverge with slightly more ballistic motion than the experiment was able to produce. Diffusion theory is shown to be inadequate to describe the data for the longer scattering times measured. All theories agree for the shorter scattering times. Furthermore, the expected operating range in the nEDM apparatus of the parameter $\lambda/L \approx 0.029$ for $^3$He was measured and shown to agree with both valid theories in that region.

The heat flush method of $^3$He is proposed as a transport method for the nEDM apparatus. It is modeled by a combination of the Navier-Stokes and convection-diffusion equations. Solved with the use of the finite element method (FEM), the parameters are optimized to minimize the amount of heat flux and time required for flush, while maximizing the polarization in the measurement cells. The method is shown to meet the strict requirement for maintaining 99% polarization of $^3$He in the measurement cells at the start of a measurement. Optimized
parameters for removing the unpolarized $^3$He are presented for an acceptable amount of heat flux and time.
An Investigation in the Dynamics of Polarized Helium-3 in Superfluid Helium-4 for the Spallation Neutron Source (SNS) neutron-electric-dipole-moment (nEDM) experiment

by
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DEDICATION

To my parents, and grand parents.
BIOGRAPHY

The author was born in Rochester NY in 1980 October 14th, to parents Michael and Nancy Swank. He grew up in Rochester until he moved to Webster NY at the age of 7 where he finished high school. During that time his interests included swimming and science. Three state championship victories in swimming lead him to a scholarship at NCSU where he chose to major in physics. After three years of swimming at the varsity level he chose to pursue physics due to time constraints. He entered Graduate School in 2004, joined the nEDM Collaboration in 2006, and still enjoys working on the subject. In 2009 he joined with fellow graduate student Robert Pattie to form a band. Currently they are still active in the music scene.
ACKNOWLEDGEMENTS

My Mom and Dad for the physical mental and spiritual support, questioning my pessimistic thoughts and guiding me toward being a responsible adult despite my best efforts to the contrary. My sister Bethany for inflating my ego and enabling too much fun during breaks at home, and a living example that it only counts what you think about yourself. I would like to thank my grandmother Morris for her prayers without them I’d surely be dead in a ditch. My Grandfather Morris for his wise guidance and unquestioning confidence in me, without which I would have none of my own. I would like to thank my grandmother Swank for encouraging my interest in math and science and displaying superior intellect and humor, maybe a little remains in my genes, if so, I’ll be alright. My uncle Gary, My uncle Mark and aunt Terri for doing what family does. I would like to thank Bob Golub for pushing me beyond what I thought were my limits, and then enjoying a beer and stimulating conversation afterwards. Paul Huffman for his consistent work behind the scenes, and always keeping his cool with administration details when I was not. Charles Hughes for his help and insight during the hard times and for being a great friend. Robert Pattie for his awesome guitar riffs that I can play drums to, and a good ear to talk serious with when the right amount of ethanol is involved. Randy Tyner II for his humor and keeping things in perspective. Grant Palmquist, always the first and generally the last guy to talk to if you want learn about a topic. Colin Anderson-Evans, Matthew Hamilton and Phil Thompson for getting me through undergrad, working with you guys wasn’t really work.
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CHAPTER 1

SYMMETRY VIOLATION

Symmetry surrounds the human experience. It is found countless in nature and in our own creations including paintings, sculptures, music and movies. However, if we consider any symmetry in detail it is almost never perfect. Small differences can be seen or measured, and in the present author’s opinion, these asymmetries can give rise to beauty; asymmetry is an ancient principle of architecture and is currently popular in modern art. It may well be that symmetry violations give rise to existence [1]. Discrete symmetries, namely parity, P, spatial symmetry and time reversal, T, historically were believed to be conserved, handed down by God himself. It wasn’t until Dirac questioned the idea implicitly in his work on the magnetic monopole, and then explicitly in writing in 1949 that the first mainstream questions to the validity conservation of discrete symmetry arise. In 1956 Lee and Yang [2], suggested that parity violation may explain why the $\tau^+$ and $\theta^+$, the names at that time, despite having the same masses, production rates and lifetimes, decayed into states of opposite parity. They suggested one test for parity violation using the angular distribution from $\beta$ decays of polarized nuclei. In 1957 Wu and collaborators measured the parity-violating $\beta$ asymmetry in the $^{60}$Co atom [3]. The parity violation was explained at that time by the belief in the conservation of CP, the charge conjugation-parity symmetry. However in 1964, Cronin and Fitch measured CP violation in the kaon sector [4]. The last bastion of hope for symmetry conservation is the CPT symmetry charge conjugation-parity-time reversal symmetry. A new source of T violation must be found for this symmetry to be conserved. A measurement of the neutron-electric-dipole-moment (nEDM) would be a measurement of the magnitude of T violation. This chapter briefly summarizes symmetry violation and the current state of the nEDM limits and the nEDM measurement planned at the Spallation-Neutron-Source (SNS).
1.1 Magnetic Monopole

In the first half of the twentieth century, the discrete symmetries party, P, and time symmetry, T, were believed to be conserved. The first suggestion of P violation was in the formulation of the magnetic charge by Dirac, first suggested in 1931 [5], then a more complete theory in 1948 [6], and then a direct suggestion in 1949 [7]. In this he writes “I do not believe there is any need for physical laws to be invariant under reflections in space and time...”. Dirac showed that quantum mechanics does not preclude the possibility of a magnetic charge, and if one existed then the quantization of both electric and magnetic charge would arise naturally. Furthermore he proposed that Maxwell’s equations would appear more symmetric with interchangeable combinations [5,6].

\[
\begin{align*}
\nabla \cdot \mathbf{B} &= \rho_M \\
\nabla \cdot \mathbf{E} &= \rho_E \\
\nabla \times \mathbf{B} - \frac{\partial \mathbf{E}}{\partial t} &= \mathbf{J}_E \\
\nabla \times \mathbf{E} - \frac{\partial \mathbf{B}}{\partial t} &= \mathbf{J}_M
\end{align*}
\]

One can apply the symmetry operation to the individual parameters above to understand how they behave under both party and time reversal, shown in Table 1.1. Using Equations 1.4 and 1.2 one finds that the magnetic current, \( \mathbf{J}_M \) is + in parity, while the magnetic monopole density \( \rho_m \) is − in parity. The magnetic monopole is not a vector and thus should not display any spatial properties yet, it violates parity. This was the first suggestion of parity and time reversal violation. The magnetic charge has not been found to this date.

1.2 Neutron Electric Dipole Moment

The neutron has spin \( I = 1/2 \) as described by the quantum number \( m_I = \pm 1/2 \). It obeys Fermi statistics and can have no other internal quantum number. Thus a dipole moment must be described by the same quantum number \( m_I \). The interaction of the spin with an electric, \( d_n \), and magnetic dipole, \( \mu_n \), is determined by the Hamiltonian,

\[
H = -\frac{1}{2} (d_n \mathbf{I} \cdot \mathbf{E} + \mu_n \mathbf{I} \cdot \mathbf{B}),
\]

where the \( d_n \mathbf{I} \cdot \mathbf{E} \) term does not commute with parity or time reversal operations.
Table 1.1: The transformation of the parameters in Maxwell’s equation under parity (P) and time reversal (T), including angular momentum

<table>
<thead>
<tr>
<th>Variable</th>
<th>P</th>
<th>T</th>
</tr>
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<tbody>
<tr>
<td>E</td>
<td>−</td>
<td>+</td>
</tr>
<tr>
<td>B</td>
<td>+</td>
<td>−</td>
</tr>
<tr>
<td>(\nabla)</td>
<td>−</td>
<td>+</td>
</tr>
<tr>
<td>(J_M)</td>
<td>+</td>
<td>−</td>
</tr>
<tr>
<td>(J_E)</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>(\rho_E)</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>(\rho_M)</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>I</td>
<td>+</td>
<td>−</td>
</tr>
</tbody>
</table>

More explicitly, the time violating interaction is described by the following argument, also shown in Figure 1.1. Start with the spin and the EDM along the same direction and apply time reversal. The spin will change direction but the charge remains in the same orientation, the pictures before and after are different, thus violating the time reversal symmetry.

To conserve CPT, a new source of CP violation must exist. If an EDM is measured that is larger than is predicted by the CP violating phase in the CKM matrix of the Standard Model, then this represents a new source of CP violation outside the standard model. The necessity of an additional CP violating source will be discussed in Section 1.4.

1.3 Ammonia Electric Dipole Moment

Molecular EDM’s exist that seem to be a T violating process according to the Hamiltonian in Equation 1.5. A non-zero EDM in the ammonia molecule for example is best described using Figure 1.2 taken from [8]. This is not a T-violating process. The ammonia molecule differs from the neutron because the Hamiltonian in Equation 1.5 of the ground state neutron can be determined by a single quantum number, however in a zero field, the ammonia time reversed state exists and is thus degenerate. Furthermore by placing the nitrogen atom underneath the hydrogen atoms, the EDM reverses direction and the T-violating process is lifted. This upside-down state is degenerate at high electric fields. Therefore the ammonia EDM is not a T-violating process even according to the Hamiltonian in Equation 1.5. The neutron on the other hand does not have a degenerate ground state because if it did, it would not obey the Pauli exclusion principle or Fermi Statistics.
1.4 Baryon Asymmetry of the Universe

Matter, rather than antimatter, is dominant in the observable universe. It is expected that at the very start of the universe, there would be equal amounts of matter and anti-matter. The abundance of matter can be explained through CP violating processes that would have favored reactions that result in matter. However the only known source of CP violation in kaon decay is too small to account for the asymmetry. The necessary conditions to reach the observable matter-dominated universe is given by the Sakharov criteria; baryon number violation, CP symmetry violation, and interactions out of thermal equilibrium [1]. The following section focuses on the CP violation aspect.

1.4.1 Electroweak CP

In the Standard Model, the nEDM arises from the CP violating phase in the CKM matrix at the level $10^{-31}$ e cm. First order loop corrections cannot pick up the CP violating phase, and second order loops cancel with themselves and do not contribute. The largest contributions arise from third order loops with gluons. Third order loops of quarks only contribute at the level of $10^{-34}$ e cm [9].
Figure 1.2: Sketch of an ammonia molecule, $CH_3$, under the time reversal operation. Before the time reversal operator is applied, the EDM is parallel with the angular momentum, $I$. Afterwards it is anti-parallel. The angular momentum changes direction, but the arrangements of the hydrogen and nitrogen remain in the same position, and thus the EDM is unchanged. However, in the absence of an electric field both states can exist and are degenerate.
CP violation is found directly and indirectly in kaons, D-mesons, B-mesons and the strange B-mesons, $B_s$. While the most precise direct CP-violation measurement is the reaction $K \rightarrow \pi\pi$ with an asymmetry of [10]

$$\mathcal{R}(\epsilon' / \epsilon) = (1.65 \pm 0.26) \times 10^{-3},$$

this amount of CP violation however is not large enough to explain the observed matter asymmetry [1,11].

1.4.2 Strong CP Problem

There are naturally arising terms in the QCD Lagrangian that break CP-symmetry [12]. The most suspicious term contains an arbitrary constant $\theta$. The so-called $\theta$ term is given by

$$\mathcal{L} = -\frac{g^2 \theta}{32 \pi^2} F_{\mu\nu} \tilde{F},$$

(1.6)

where $F$ is the gluonic field strength tensor, and $\tilde{F}$ is its dual. The gluonic field strength tensor is analogous to the electromagnetic strength tensor. The term $F_{\mu\nu} \tilde{F}^{\mu\nu}$ expressed in its analogous form is $E \cdot B$ which conserves C but violates both P and T. $\theta$ is an arbitrary constant. If it was order unity as one might expect of a constant scaled by $1/2\pi$, then EDM’s would arise at the $10^{-18}$ e cm level. Current EDM measurements however limit $\theta \lesssim 10^{-8}$. An arbitrary constant of such a small value is unexpected and highly unusual. The Peccei-Quinn theory [13], treats $\theta$ as a field rather than a constant. They consider that at least one fermion flavor acquires its mass from a Yukawa coupling to a color-singlet scalar field with a nonzero vacuum expectation value, the axion. They go on to show that when the fermion masses are made real by rotations of the fermion fields the resulting $\theta$ is zero. The axion has not been observed to this date, the best published limits are calculated from measurements of transverse relaxation of spin-polarized $^3\text{He}$ [14].

1.5 Measuring the nEDM

In the assumption that at the beginning of the universe there was an equal amount of matter and anti-matter, the currently observed CP violation is not enough to account for the dominance of the baryon matter observed. It is expected that a new source of CP violation will be found. There are many proposed theories that are extensions to the Standard Model that increase the predicted EDM by up to 7 orders of magnitude from the Standard Model prediction. The most predominant of these being the supersymmetric models and L-R symmetric models. Neutron
EDM measurements provide a sensitive probe for these Standard Model extensions.

Purcell and Ramsey started the search for the nEDM with a magnetic resonant measurement. The measurement was made in 1950, but was not published until 1957 due to the controversial nature of the measurement at that time. There have been many attempts to measure the nEDM continuing to the present. The time-line of past measurements is plotted in Figure 1.3 [15]. The sensitivity of most nEDM measurements was limited by the sensitivity of the frequency measurement [16]. In the most precise experiment an effect that was predicted by Commins [17] in 1990 denoted as the “geometric phase,” limited the sensitivity of this nEDM measurement. The main focus of this work is to study this effect and find ways to reduce its influence.

### 1.5.1 Measuring the nEDM in Superfluid Helium

The nEDM experiment at the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL) is planned for a sensitivity of $d_n < 3 \times 10^{-28}$ e cm [18] a factor of 100 lower than the current limit. It will achieve this based on the use of $^3$He as both a comagnetometer and
Figure 1.4: The dispersion curve for helium-II and the free neutron. The intersection of the curves allows production of UCN by single phonon emission, leaving the neutron at rest as described in the text.

detector. $^3$He is transported from an atomic beam source (ABS) that selects the desired spin state through the use of permanent magnets. The polarized $^3$He beam is directed at a bath of liquid $^4$He in the injection volume (INJ). The atoms collect in INJ at a rate of about $10^{14}$ s$^{-1}$. Once collected, the $^3$He is transported to the measurement cell with the expectation that 99% polarization remains upon entering the measurement cell. The transport is achieved through the use of the heat flush, a phenomenon which concentrates $^3$He toward the cold section of a temperature gradient in superfluid helium [19]. A second goal of this work is to study the operation of the heat flush and establish the optimum parameters, described in Chapter 6.

The neutrons will enter the experiment from a cold beam line and down-scatter by phonon emission in liquid helium-II. Single phonon emission is achieved due to the intersection of the dispersion curves seen in Figure 1.4, allowing for a single interaction at that energy that conserves energy and momentum, resulting in a neutron that is essentially at rest.

The down-scattered neutrons that have energies lower than the Fermi potential of the dTPB-dPS (deuterated tetraphenyl butadiene - deuterated polystyrene) coated walls are called ultracold
neutrons (UCN) and are trapped by the material walls of the experimental measurement cell. The UCN will remain in the cell for the measurement with a lifetime, $\tau$, given by

$$\frac{1}{\tau} = \frac{1}{\tau_{\text{bottle}}} + \frac{1}{\tau_n} + \frac{1}{\tau_{^{3}\text{He}}},$$

(1.7)

where $\tau_n$ is the neutron lifetime, $\tau_{^{3}\text{He}}$ the absorption from $^{3}\text{He}$, and $\tau_{\text{bottle}}$ is the lifetime due to interactions with the walls of the bottle. The main source of loss, apart from neutron decay, is up-scattering from the walls, absorption on the walls, and up-scattering from phonons present in the helium-II. However at low temperature the phonon in helium-II density is negligible.

The reaction describing neutron capture on $^{3}\text{He}$ is $^{3}\text{He}(n,p)^{3}\text{H}$ with a $Q$-value of 764 keV, where the majority of the 764 keV of kinetic energy will be deposited into the superfluid helium bath producing scintillations in the extreme ultra violet (EUV $\sim 80$ nm.) The EUV scintillation light is down converted to blue light by the dTPB in the coating on the cell walls. The blue light is guided through the walls to light guides and then guided to an array of photo multiplier tubes (PMTs) operating at 4K.

The absorption of neutrons on $^{3}\text{He}$ is dependent on the relative angle between their nuclear spins. Only the $J = 0$ state leads to absorption. The rate of absorption is described by the equation

$$S(t) = \frac{\rho_{\text{UCN}} V}{\tau_{^{3}\text{He}}} (1 - P_n P_{^{3}\text{He}} \cos \theta_n^{3}(t)),\quad (1.8)$$

where $P_n, P_{^{3}\text{He}}$ are the polarizations, $\rho_{\text{UCN}}$ is the UCN density, $V$ is the volume, and $1/\tau_{^{3}\text{He}}$ is the rate of absorption in the unpolarized configuration.

The neutron and $^{3}\text{He}$ polarization are initially prepared in the same spin state so the capture rate is minimized. The nEDM measurement induces a $\pi/2$ rotation so that both species precess in the plane perpendicular to the $\text{B}$ field as given in Equation 1.5. The volume average of the holding field of both the $^{3}\text{He}$ and the neutron are the same. If they precess at the same rate in the magnetic field the frequencies for measurements with $\text{E}$ directed either parallel or antiparallel to $\text{B}$ will differ by $d_n \cdot \text{E}$. Therefore $\theta_n^{3}$ will increase in proportion to $d_n \cdot \text{E}$, a measurement of $d_n$. The EDM of the $^{3}\text{He}$ has no effect on its precession due to cancellations from the Schiff moment, essentially the shielding of the $^{3}\text{He}$ EDM by the electrons.

The $^{3}\text{He}$ and neutron’s gyromagnetic ratios are related by $\gamma_{^{3}\text{He}} \approx 1.112 \times \gamma_n$. The frequencies are not equal in the same magnetic field and the two species will precess at different rates. However an ingenious method to correct for this is described in reference [8] and is called critical spin dressing. Spin dressing is the process of altering the precession frequency of a spin by applying a non-resonant, specially tuned oscillating magnetic field. The spin will wiggle up and down orthogonal to its precession, slowing down the precession to the desired frequency. Critical
spin dressing is applying this technique to two spins simultaneously such that the Larmor frequencies of both species become equal. For spin dressing, the time average $z$ component is given as \[ 1 \]

\[
\langle \cos(\theta) \rangle_T = \frac{1}{T} \int_T dt \cos \left( \frac{\gamma B_{rf}}{\omega_{rf}} \right) \cos(\omega_{rf}t) = J_0 \left( \frac{\gamma B_{rf}}{\omega_{rf}} \right), \tag{1.9}
\]

where $T$ is the total time of the integration, $B_{RF}$ and $\omega_{RF}$ are the magnitude and frequency of the applied RF magnetic field, respectively. $J_0$ is the zero order Bessel Function. The effective precession frequency is the original precession frequency times the Bessel function of $\gamma B_{rf}/\omega_{rf}$. To achieve critical dressing, one finds the parameters where the precession rates are equal for the two species. Substitute $x_i = \gamma_i B_{rf}/\omega_{rf}$

\[
\gamma_n J_0(x_n) - \gamma_{3\text{He}} J_0(x_{\text{He}}) = 0, \quad \gamma_{3\text{He}} \approx 1.112 \gamma_n, \tag{1.10}
\]

\[
1.1 J_0(1.1x_c) - J_0(x_c) = 0. \tag{1.11}
\]

The solution to the critical dressing is thus

\[
x_c \approx 1.19, \quad J_0(x_c) = 0.65. \tag{1.12}
\]

With the application of an RF field that satisfies the condition of $x_c = \gamma_{3\text{He}} B_{rf}/\omega_{rf} \approx 1.19$, the effective Larmor precession of the neutrons and the $\text{He}$ will be the same. The measurement of $\theta_{n3}$ will result in the measurement of the neutron EDM according to

\[
\theta_{n3}(t) = \frac{2e\bar{d}_n E t}{\hbar}, \quad \bar{d}_n = d_n J_0(x_c). \tag{1.13}
\]

There are other shifts linear in $E$ that must be minimized and accounted for in order for a measurement of maximum sensitivity. The largest of these is the geometric phase, the systematic effect that limited the sensitivity at the nEDM experiment at the ILL [16].

1.5.2 The Geometric Phase Systematic

The immediate motivation for this work is to better understand and predict the geometric phase. The geometric phase is responsible for limiting the sensitivity of the most recent experiment at the ILL [16], and it is expected to be the largest systematic for the SNS nEDM experiment.
Integrated over time, the geometric phase manifests itself as a frequency shift linear in the electric field.

Following the treatment of the Bloch Siegert shift considered in [20], we model the case of a bottle with radius \( R \) with neutrons and/or \(^3\)He in roughly circular orbits in a holding field with a symmetric gradient. From \( \nabla \cdot \mathbf{B} = 0 \) the radial field varies as \( B_r(r) = (r/2)\partial B_z/\partial z = ar \), where \( a = (1/2)\partial B_z/\partial z \). There is also a radial magnetic field originating from the circular motion of the neutron or \(^3\)He spin in an electric field, \( \mathbf{B}_v = \mathbf{v} \times \mathbf{E} = r\omega_r E/c \), where \( \omega_r = 2\alpha/\tau_c \) is the orbital frequency, \( \alpha \) is the angle of incidence on the wall, and \( \tau_c \) is the time between wall collisions. The total radial field the neutrons and \(^3\)He see is thus

\[
B_R = B_r \pm B_v,
\]

where the \( \pm \) refers to the direction of rotation. In this rotating frame, the total field is

\[
B^2 = \left(B_0 - \frac{\omega_r}{\gamma}\right)^2 + (B_R)^2,
\]

where \( B_R \ll B_0 \). Expanding in this limit, and transforming back to the lab frame

\[
B = B_0 + \frac{1}{2} \frac{(aR - \omega_r RE/c)^2}{B_0 - \omega_r/\gamma} = B_0 - \frac{\gamma aR^2 \omega_r E}{c(\omega_0 - \omega_r)}.
\]

Averaging over the rotation directions yields a frequency shift \( \delta \omega \) linear in \( E \),

\[
\delta \omega = \gamma \delta_B = -\frac{\gamma^2 a v^2 E}{c(\omega_0^2 - \omega_r^2)}.
\]

The fact that the frequency shift is linear in \( E \) arises from the interaction of \( B_v \) and \( B_r \). Conceptually this can be understood with the following argument. The magnetic field experienced for a moving spin in an electric field, \( \mathbf{B}_v = \mathbf{v} \times \mathbf{E} \), changes sign with direction of the velocity, while magnetic field gradients change the holding field with position. Considering the magnetic interaction with fields linear in \( E \), neutrons or \(^3\)He traversing the cell in one direction will rotate their spins a given angle. Then upon wall reflection, the \( \mathbf{B}_v = \mathbf{v} \times \mathbf{E} \) term is equal but opposite. However an inevitable gradient causes the magnetic field to be slightly different than at the start. The phase accumulated upon traversing the cell again does not cancel with the first pass because the starting magnetic fields are different and rotations do not commute. The phase shift
will manifest itself as a frequency shift over time. The shift will be linear in $E$ and appear as a false EDM. This is the geometric phase effect.

An important design feature of the nEDM measurement in superfluid helium is the $^3$He comagnetometer. The comagnetometer is necessary for high sensitivity frequency measurements in order to account for the inevitably changing $B_0$ field. Although both neutrons and $^3$He experience this phase shift, one can actually control the size of the effect on $^3$He. A property of the $^3$He is the strong temperature dependence of the average $^3$He phonon scattering time in superfluid helium, where the diffusion coefficient is given as $D = 1.6T^{-7}$ cm$^2$/s. Thus by changing the temperature of the helium bath one can change the diffusion rate which allows the possibility to tune the frequency shift to zero as was discovered by theoretical analysis of the effect [21]. The theory proposed that describes the frequency shift due to the geometric phase is described in Chapter 2.
The geometric phase is a frequency shift linear in the applied electric field due to interactions of the neutron or $^3$He with the electric field via the motional magnetic field ($B_v = \mathbf{v} \times \mathbf{E}$) coupled with deviations in the holding field. Without deviations in the holding field, the phases accumulated from the motional magnetic field would average to zero because the average time spent traversing one direction is equal to the average time spent traversing the opposite direction. In reality however, the holding field is not completely uniform. The holding field experienced along a path is reversed in time for opposite path directions. Since rotations do not commute, the phases accumulated along these opposite paths will not cancel, resulting in a non-zero phase shift.

This phase shift, termed the geometric phase shift or geometric frequency shift, is the largest systematic effect in the most recent nEDM measurement at the ILL and sets the limit on the experiment’s sensitivity. It is also the primary motivation for this work. It is treated analytically by a density matrix formalism. Similar treatments can be found in references [22, 23] and [20].

First the case of spin relaxation is examined. This is important because it is the method for estimating the geometric frequency shift without an electric field. The relaxation is shown to be dependent on the spectrum of correlation functions. The geometric frequency shift is treated in the same manner and is shown to depend on the same spectrum of correlation functions. The spectrum of velocity weighted correlation functions for random walks in restricted geometries for arbitrary mean free paths is derived according to references [24] and [25]. Finally, the geometric phase is modeled through simulation of the correlation function. Results are in good agreement with the analytic model.
2.1 Spin Dynamics

In the regime of the magnetic gradient induced spin relaxation in magnetic fields that are small enough such that the magnetic-spin interaction does not affect the Brownian motion of the particle, the theory derived by Redfield [22] can be implemented to show that phase shifts and relaxations depend on the field correlations experienced by the ensemble average of the spins [22]. In a static field these are represented by field-position correlation functions, [14,26] or in the special case of linear gradients, the position-position correlation function weighted by the strength of that gradient [23].

A formulation specific to our goals, the holding field in the \( z \)-direction using the theory given by Redfield will be shown to relate the relaxation matrix to the correlation functions of the ensemble. Nearly the same derivation is given in reference [20], except a changing field in the same direction as the holding field axis will be allowed. This is shown to give rise to the dominant term in the transverse decay rate (\( T_2 \)). A similar result is shown by [27] and [23], however in our case, arbitrary field shapes are considered and are not limited to constant gradients across the cell.

We start by defining the magnetic field orientation so that the volume average of the field components not along the holding field axis are zero; the time average experienced by the moving particles averages to zero. This is the same formulation found in reference [20], except we include a changing field in the \( z \) direction,

\[
\begin{align*}
B'_x(t) &= B_x(t) - \langle B_x(t) \rangle, \\
B'_y(t) &= B_y(t) - \langle B_y(t) \rangle, \\
B'_z(t) &= B_z(t) - \langle B_z(t) \rangle, \\
\langle B_z(t) \rangle &= B_0.
\end{align*}
\]

(2.1)

Here, \( \langle \Box \rangle \) denotes the time average. For this purpose, the volume average is equivalent to the time average for the fields because only static fields are considered and the particles are uniformly distributed.

This is also valid for fields that are changing slowly compared to the Larmor precession. In this case, an average must be taken by integrating over the changing field strengths. We define the holding field as

\[
B'_0 = \sqrt{B_0^2 + \langle B_x(t) \rangle^2 + \langle B_y(t) \rangle^2},
\]

(2.2)

where the \( z \)-direction is redefined so that all the perturbing fields have averages of zero, and \( B'_0 \)
is the value of the field along the new $z$-direction. This is the same as in the case of the Redfield theory and also utilized in reference [20]. The fields are written in terms of an angular frequency,

$$\omega_0 = \gamma B_0,$$

(2.3)

$$\omega_{x,y,z}(t) = \gamma B_{x,y,z}(t).$$

(2.4)

The Hamiltonian can be written as a static diagonalized part and a time dependent perturbing part,

$$H = H_0 + H_1(t),$$

(2.5)

$$H_0 = -\frac{\omega_0}{2}\sigma_z,$$

(2.6)

$$H_1(t) = \sum_{x,y,z} \frac{\omega_{x,y,z}(t)}{2}\sigma_{x,y,z}.$$  

(2.7)

The density matrix, $\rho$, contains the information of the orientation of the spin,

$$\rho = \begin{pmatrix} 1 + \rho_z & \frac{\rho_x}{2} + i\frac{\rho_y}{2} \\ \frac{\rho_x}{2} - i\frac{\rho_y}{2} & 1 - \rho_z \end{pmatrix}.$$  

(2.8)

The time evolution is given by

$$\frac{d\rho}{dt} = -i[H_0 + H_1(t), \rho].$$  

(2.9)

Transforming into the rotating frame, the Hamiltonian becomes

$$H'_1(t) = e^{iH_0t}H_1(t)e^{-iH_0t}, \rho' = e^{iH_0t}\rho e^{-iH_0t}.$$  

(2.10)

So, the density matrix and perturbing Hamiltonian become

$$\rho = \begin{pmatrix} 1 + \rho_z & e^{-i\omega_0t}\left(\frac{\rho_x}{2} + i\frac{\rho_y}{2}\right) \\ e^{i\omega_0t}\left(\frac{\rho_x}{2} - i\frac{\rho_y}{2}\right) & 1 - \rho_z \end{pmatrix}.$$  

(2.11)

$$H_1 = \begin{pmatrix} \frac{\omega_x}{2} & e^{-i\omega_0t}\left(\frac{\omega_x}{2} + i\frac{\omega_y}{2}\right) \\ e^{i\omega_0t}\left(\frac{\omega_x}{2} - i\frac{\omega_y}{2}\right) & -\frac{\omega_x}{2} \end{pmatrix}.$$  

(2.12)

The time evolution in the interaction picture is

$$\frac{d\rho}{dt} = -i[H_1(t), \rho].$$  

(2.13)
To second order, with the introduction of $\tau = t - t'$

$$\frac{d\rho}{dt} = -i[H_1(t), \rho(0)] - \int_0^t d\tau [H_1(t), [H_1(t - \tau), \rho(0)]].$$ \hspace{1cm} (2.14)

In our case, the first term will average to zero since the initial $\rho$ is not correlated with the perturbing Hamiltonian. The initial positions and rotation angles of the spin should not be correlated with the perturbing field. This can be dictated through the fabrication of the RF spin flipping coils. We require an initial flipping pulse that is uniform over the region where the spins are located. This way, the interaction with the perturbing fields will average to zero since $H_1$ averages to zero over volume and time.

The second term in Equation 2.14 can be transformed from $\rho(0)$ to $\rho(t)$, which introduces errors on the order of $\tau_c/t$. We require $t >> \tau_c$, where $\tau_c$ is the time it takes for the position autocorrelation function to become small. From this assertion, it is allowable to replace the integration of $0 \to t$ with $0 \to \infty$. This formulation is valid as long as either $\rho(\tau_c) = \rho(0)$ or the time required for a change in the density matrix is much longer than $\tau_c$.

Further simplification is achieved by considering special cases. For now, consider relaxation and restrict this relaxation to non-uniformities in static magnetic fields. Assume the field that the particle experiences is a function of its position, and the position autocorrelation function of differing dimensions is uncorrelated. Therefore, cross terms containing $\omega_i \omega_j$ will average to zero for $i \neq j$. The correlation function is an even function of $\tau$ and terms rotating with $2\omega_0$ can be ignored. The sum and the difference of the off-diagonal elements gives $\rho_x$ and $\rho_y$ respectively. To further simplify the equation we can set $\exp(i\frac{\pi}{4} \sigma_z)\rho_x \exp(-i\frac{\pi}{4} \sigma_z) \rightarrow \rho_y$ and $\exp(i\frac{\pi}{4} \sigma_z)\rho_y \exp(-i\frac{\pi}{4} \sigma_z) \rightarrow -\rho_x$. This is seen by setting $\omega_0 \tau = \pi/2$ in Equation 2.11, and it is equivalent to a $\pi/2$ step in the rotating frame. The difference in the diagonal elements gives $\rho_z$.

For relaxation in a static field it is found that

$$\frac{d\rho_x}{dt} = i\omega_0 \rho_x$$ \hspace{1cm} (2.15)

$$-\frac{1}{2} \left( \int_{-\infty}^{\infty} \omega_y(t)\omega_y(t + \tau)e^{-i\omega_0 \tau} d\tau - \int_{-\infty}^{\infty} \omega_z(t)\omega_z(t + \tau)d\tau \right) \rho_x$$

$$\frac{d\rho_y}{dt} = i\omega_0 \rho_y$$ \hspace{1cm} (2.16)

$$-\frac{1}{2} \left( \int_{-\infty}^{\infty} \omega_x(t)\omega_x(t + \tau)e^{-i\omega_0 \tau} d\tau - \int_{-\infty}^{\infty} \omega_z(t)\omega_z(t + \tau)d\tau \right) \rho_y$$

$$\frac{d\rho_z}{dt} = -\frac{1}{2} \left( \int_{-\infty}^{\infty} \omega_x(t)\omega_x(t + \tau)e^{-i\omega_0 \tau} d\tau \rho_z ight)$$ \hspace{1cm} (2.17)

$$+ \int_{-\infty}^{\infty} \omega_y(t)\omega_y(t + \tau)e^{-i\omega_0 \tau} d\tau \rho_z.$$
This result is precisely what is given in Slichter [27] and McGregor [23] for the same assumptions of \( B_0 \) and \( B_1 \). It follows from Equations 2.15-2.17 that the longitudinal decay rate is

\[
\frac{1}{T_1} = \frac{\gamma^2}{2} \left( \int_{-\infty}^{\infty} B_x(t)B_x(t + \tau)e^{-i\omega_0 \tau}d\tau + \int_{-\infty}^{\infty} B_y(t)B_y(t + \tau)e^{-i\omega_0 \tau}d\tau \right). \tag{2.18}
\]

The decay has been reduced to the spectrum of a correlation function. Equation 2.18 is used to calculate the gradient-induced decay rate of the \(^3\text{He}\), the quantity measured in the experiment described in Chapters 3 and 5. For the transverse decay (\( T_2 \)), the rate that the phase angle spreads irreversibly over the plane transverse to the longitudinal direction, thereby losing the magnetization in the transverse direction, is given by

\[
\frac{1}{T_2} = \frac{1}{2T_1} + \frac{\gamma^2}{2} \int_{-\infty}^{\infty} B_z(t)B_z(t + \tau)d\tau. \tag{2.19}
\]

### 2.1.1 Phase Shifts Linear in Electric Field

To find the phase shifts linear in electric field, we first assume linear magnetic field gradients. Assume a symmetric field and \( \nabla \cdot \mathbf{B} = 0 \) so that \( \partial B_z/\partial z = -2\partial B_x/\partial x \) = \(-2\partial B_y/\partial y \). In the case of an electric field, not considered in the derivation of Equations 2.15-2.17, the \( \mathbf{v} \times \mathbf{E} \) field must also be included. The frequencies thus become,

\[
\omega_x = \frac{\gamma}{2} \frac{\partial B_z}{\partial z} x + \frac{\gamma}{c} v_y
\]

\[
\omega_y = \frac{\gamma}{2} \frac{\partial B_z}{\partial z} y - \frac{\gamma}{c} v_x. \tag{2.20}
\]

We are only interested in terms linear in \( E \) and we will not consider other terms. The phase shift will only occur in the transverse plane, therefore to find the phase shift it is sufficient to find how \( \rho_x \) evolves, or the sum of the off-diagonal elements of the density matrix. We start from Equation 2.14 keeping only terms linear in \( E \) with correlation functions that are not zero. It is found that
$$\delta \omega = -\frac{\gamma^2 G_z E}{4c} \int_0^t d\tau \cos \omega_0 \tau \langle \omega_x(t + \tau) \omega_y(t) - \omega_z(t) \omega_y(t + \tau) \rangle + \frac{\gamma^2 G_z E}{4c} \int_0^t d\tau \sin \omega_0 \tau \langle \omega_x(t) \omega_x(t + \tau) + \omega_x(t + \tau) \omega_x(t) \rangle$$

$$\delta \omega = -\frac{\gamma^2 G_z E}{4c} \int_0^t d\tau \cos \omega_0 \tau \times \langle y(t + \tau) v_y(t) + x(t + \tau) v_x(t) - y(t) v_y(t + \tau) - x(t) v_x(t + \tau) \rangle,$$

(2.21)

where \(G_z\) is the uniform gradient strength. The terms linear in \(E\) in the sine integral are uncorrelated and will not contribute. This is the same as is derived in reference [20]. To continue the approach found in reference [26] also seen in reference [28] is used. The velocity position function can be written in terms of the position-position correlation function as given in reference [29], equation 15.42. Also the definition that \(t' = t + \tau\), and that correlation functions are even in \(\tau\), enable the simplification

$$\langle x(t) v_x(t + \tau) \rangle = \frac{\partial}{\partial \tau} \langle x(t) x(t + \tau) \rangle,$$

(2.22)

$$\langle x(t + \tau) v_x(t) \rangle = \langle x(t) v_x(t - \tau) \rangle = -\frac{\partial}{\partial \tau} \langle x(t) x(t + \tau) \rangle.$$

(2.23)

With these definitions, equation 2.20 does not need a linear gradient in the magnetic field. The frequency shift from an arbitrary field can be calculated with the time derivative of the correlation function for any arbitrary function, we leave this for later. We extend the integral to infinity so the correlation will become arbitrarily small during a measurement. We therefore have

$$\delta \omega = \frac{\gamma^2 G_z E}{4c} \int_0^\infty d\tau \cos \omega_0 \tau \frac{\partial}{\partial \tau} \langle 2x(t)x(t + \tau) + 2y(t)y(t + \tau) \rangle,$$

(2.24)

where equation 2.24 is the Fourier cosine transform. The derivative can be expressed in terms of the sine transform, and the value of the function at \(\tau = 0\) [30],

$$\delta \omega = \omega_0 \frac{\gamma^2 G_z E}{4c} \int_0^\infty d\tau \sin \omega_0 \tau \langle 2x(t)x(t + \tau) + 2y(t)y(t + \tau) \rangle - \frac{\gamma^2 G_z E}{4c} \langle 2x(t)x(t) + 2y(t)y(t) \rangle.$$

(2.25)
Note that the last term depends only on the field shape and not the conditional density. The first term can be written in terms of the imaginary part of the Fourier transform,\[\delta \omega = \omega_0 \frac{\gamma^2 G_z E}{4c} \Im \left\{ \int_{-\infty}^{\infty} d\tau e^{i\omega_0 \tau} \langle x(t)x(t+\tau) + y(t)y(t+\tau) \rangle \right\} \]
\[\quad - \frac{\gamma^2 G_z E}{2c} \left( \frac{1}{12} L_x^2 + \frac{1}{12} L_y^2 \right) \]
(2.26)

For an arbitrary magnetic field the frequency shift is linear in \(E\), and given by
\[\delta \omega = \omega_0 \frac{\gamma^2 E}{2c} \Im \left\{ \int_{-\infty}^{\infty} d\tau e^{-i\omega_0 \tau} \langle B_x(t)x(t+\tau) + B_y(t)y(t+\tau) \rangle \right\} \]
\[\quad - \frac{\gamma^2 E}{c} \langle B_x x + B_y y \rangle . \]
(2.27)

### 2.2 Conditional Densities Beyond Diffusion

For the application of the theory to the experimental parameters, the diffusion model is not sufficient to adequately describe the spin transport problem. The mean free path is comparable to the restrictive dimensions, so the diffusion approximation will break down, becoming inaccurate. Therefore it is necessary to describe the Brownian motion of the spin polarized particles through a continuous time random walk. This approach is shown to accurately describe the ballistic regime, the diffusive regime and the transition in-between.

A key ingredient to the continuous time random walk is the probability of scattering per unit time,
\[\psi(t) = \frac{1}{\tau_c} e^{-\frac{t}{\tau_c}} , \]
(2.28)

where \(\tau_c\) is the average time between collisions. To continue, the number of dimensions in the problem must be specified. For one dimension it is known that the continuous time random walk satisfies the telegrapher’s equation [24,25,31], given by
\[\frac{\partial^2 p}{\partial t^2} + \frac{1}{\tau_c} \frac{\partial p}{\partial t} = v^2 \frac{\partial^2 p}{\partial x^2} \]
(2.29)

where \(v\) is the velocity of the particles, and \(p\) is the conditional probability density, the probability of being at position \(x\) at time \(t\), after starting from position \(x_0\) at time \(t = 0\). For reflecting
boundaries spaced a distance $L$, with $x = 0$ defined as the center point between the two boundaries, the solution is [32]

$$p(x, t|x_0, 0) = \frac{1}{L} \left\{ \sum_{n=\text{even}} \cos \frac{n\pi x}{L} \cos \frac{n\pi x_0}{L} + \sum_{n=\text{odd}} \sin \frac{n\pi x}{L} \sin \frac{n\pi x_0}{L} \right\} \times \left\{ \cosh \frac{s_n t}{2\tau_c} + \frac{1}{s_n} \sinh \frac{s_n t}{2\tau_c} \right\} e^{-\frac{t}{2\tau_c}} \tag{2.30}$$

where $s_n = \sqrt{1 - 4\omega_n^2\tau_c^2}$, $\omega_n = n\pi v/L$.

The natural assumption one would expect in extending to higher dimensions is that the telegrapher’s equation would be the solution. This is readily shown to be incorrect since the solution to the 2D telegrapher’s equation can go negative and thus cannot be the solution to a probability. The validity of the telegrapher’s equations to describe transport phenomena is further discussed in reference [33].

To solve the restricted problem in 2-dimensions we follow the solution given in Masoliver [24] for the free space solution, and then apply rectangular boundaries [25]. Starting with the scattering density, $\rho$, where $\rho$ is the probability density of particles scattering at a position $r$ and time $t$ traveling at an angle $\theta$ with respect to the $x$-axis,

$$\rho(r, t; \theta) = \frac{1}{2\pi} f(r, t; \theta) \psi(t) \tag{2.31}$$

$$+ \int d^2r' \int_{-\pi}^{\pi} d\theta' \int_0^t d\tau \rho(r', \tau; \theta') \beta(\theta|\theta') f(r - r', t - \tau, \theta) \psi(t - \tau).$$

The first term is the probability of scattering at point $(r, t)$ and traveling at an angle $\theta$ with respect to the $x$-axis of unscattered particles. The second term is the probability of scattering at the same point having previously scattered at a point $(r', \tau)$ and traveling with angle $\theta'$ previous to the scattering. $\beta$ is the scattering distribution. In this work only isotropic scattering is considered. In reference [24] they have extended the unrestricted random walk to scattering with angles of weighted probability. The same technique discussed below could be applied to find the non-isotropic scattering in the restricted walk. For this work only isotropic scattering is considered, so $\beta(\theta|\theta') = 1/2\pi$. $f$ is the function that propagates the unscattered particles, or particles between scattering. $f$ is the same at the starting point or at a scattering point and is a circle expanding with velocity $v$,

$$f(r, t; \theta) = \delta(x - vt \cos \theta) \delta(y - vt \sin \theta). \tag{2.32}$$
Because only isotropic scattering is considered and the distribution \( f \) is a circle expanding outward, a probability density of scattering at position \((r, t)\) starting at the origin will not depend on \( \theta \) or \( \theta' \),

\[
\rho(r, t) = \frac{1}{2\pi} \psi(t)f(r, t) + \frac{1}{2\pi} \int d^2r' \int_0^t d\tau \rho(r', \tau)f(r - r', t - \tau)\psi(t - \tau).
\] (2.33)

Similarly the probability of arriving at position \((r, t)\) starting at the origin, will also not depend on \( \theta \) or \( \theta' \)

\[
p(r, t) = \frac{1}{2\pi} \Psi(t)f(r, t) + \frac{1}{2\pi} \int d^2r' \int_0^t d\tau \rho(r', \tau)f(r - r', t - \tau)\Psi(t - \tau),
\] (2.34)

where \( \Psi(t) \) is the probability of having made it to time \( t \) without scattering,

\[
\Psi(t) = \int_0^\infty \psi(\tau)d\tau = e^{-\frac{t}{\tau_c}} = \tau_c\psi(t).
\] (2.35)

Equations 2.33 and 2.34 contain convolutions of \( \rho \) and can be linearized using a Laplace Transform, defined as,

\[
f(Q, s) = L(f(r, t)) = \int d^2r \int_0^\infty f(r, t)e^{iQr-st}dt, \quad s = \sigma + i\omega.
\] (2.36)

Substituting,

\[
g(r, t) = f(r, t)\psi(t)
\] (2.37)

\[
G(r, t) = f(r, t)\Psi(t)
\] (2.38)

Where \( G(r, t) = \tau_c g(r, t) \), equations 2.33 and 2.34 become,
\[ \rho(r, t) = \frac{1}{2\pi} \left( g(r, t) + \int d^2 r' \int_0^t \rho(r', \tau) g(r - r', t - \tau) d\tau \right), \quad (2.39) \]

\[ p(r, t) = \frac{1}{2\pi} \left( G(r, t) + \int d^2 r' \int_0^t \rho(r', \tau) G(r - r', t - \tau) d\tau \right). \quad (2.40) \]

After the transformation the above equation for \( \rho \) and \( p \) can be solved,

\[ \rho(Q, s) = \frac{g(Q, s)}{2\pi - g(Q, s)}, \quad (2.41) \]

\[ p(Q, s) = \frac{G(Q, s)}{2\pi - g(Q, s)}. \quad (2.42) \]

To solve for \( g(Q, s) \) is sufficient to solve the problem, since \( G(r, t) = \tau_c g(r, t) \). The definition of \( g(r, t) \) is an expanding circle decaying with the same rate as the scattering rate of a walker. Its transform is

\[ g(Q, s) = \int d^2 r \int_0^\infty f(r, t) \psi(r, t) e^{iQ \cdot r} dt = \frac{2\pi}{\sqrt{(1 + \frac{s}{\tau_c})^2 + \frac{v^2Q^2}{\tau_c^2}}} \quad (2.43) \]

Thus the spectrum of the 2D random walk is

\[ p(Q, s) = \frac{1}{\sqrt{\left(\frac{1}{\tau_c} + s\right)^2 + v^2Q^2 - \frac{1}{\tau_c}}} \quad (2.44) \]

The same approach can be used to solve the spectrum of the 3D isotropic continuous time random walk without much difference. Simply change the integration to be over the volume so that each \( 2\pi \rightarrow 4\pi^2 \), including \( \beta = 1/4\pi^2 \) and \( f(r, t) \) becomes an expanding sphere instead of a circle. The solution for the spectrum in three dimension is thus

\[ p(Q, s) = \frac{\arctan \left( \frac{vQ}{\frac{1}{\tau_c} + s} \right)}{vQ - \frac{1}{\tau_c} \arctan \left( \frac{vQ}{\frac{1}{\tau_c} + s} \right)}. \quad (2.45) \]

In reference [24] they continue by solving for the conditional density as a function of the time and position. However for the purpose of this work it is sufficient to stop at this point because the relaxations and frequency shifts all depend of the spectrum of the correlation functions. However, there is some applicability for the 2D conditional density in the following section for
other purposes outside the application of this work so it is given below for reference,

\[ p(r, t) = e^{-\frac{t}{\tau_c}} \left[ \frac{\delta(r - vt)}{2\pi r} + \frac{1}{2\pi v \tau_c \sqrt{v^2 t^2 - r^2}} \exp \left( \frac{1}{\tau_c \sqrt{v^2 t^2 - r^2}} \right) \right]. \] (2.46)

There is no solution to date for the conditional probability in 3D.

The free space solution will now be implemented to solve for the continuous time random walk in restricted geometries.

### 2.3 Random Walk in Restricted Geometries

The continuous time random walk can be solved in restricted geometries of specular reflection in more than one dimension using the method of images [25]. The easiest and most applicable geometry for this purpose is the rectangle in 2D or for 3D a rectangular cell also known as a rectangular solid, right rectangular prism, irregular hexahedron, rectangular cuboid, etc. Both the nEDM and the current gradient relaxation experiment are using a rectangular cell. For 2D, the image of the cell must be made in both the \( x \) and the \( y \) plane and then the set containing the four images are then tessellated as shown in Figure 2.1 [25].

As stated in the caption in Figure 2.1, if the points represent the observer, then the source is restricted to the original cell. If the points represent the source then the observer is restricted to the original cell. Both pictures are mathematically equivalent. The present author believes the picture with the observer as the imaged and tessellated points, with the source emanating from the original cell is the most intuitive. The remainder of this chapter will assume this is the picture. It is a simple procedure to transform the free space probability into the tessellated picture. This is done by summing the image points as in Equation 2.50, where the \( p(r, t) \) is from Equation 2.46. Here,

\[ r = \sqrt{(x - x_0)^2 + (y - y_0)^2}, \] (2.47)

\[ x_{l\pm} = 2l_x L_x \pm x, \] (2.48)

\[ y_{l\pm} = 2l_y L_y \pm y, \] (2.49)

\[ p_{\text{rect}}(x, y, t) = \sum_{l_x, y=-\infty}^{\infty} p(x_{l\pm}, y_{l\pm}, t), \] (2.50)

where the sum is a double sum to account for all values of \( x_l \) and \( y_l \). This will completely describe the restricted random walk in a rectangle with specular reflecting walls from the ballistic to the
Figure 2.1: The original cell is the shaded region, with images of the cell, and tessellations of a set of the original cell and images surrounding them. Also shown below in figure B is an arbitrary field in the $x$ dimension. If the points represent the observer, the source is restricted to the original cell. If the points represent the source then the observer is restricted to the original cell. Both pictures are mathematically equivalent.
diffusive regime. If the free conditional probability for the 3 dimensions existed, then it would be restricted to a rectangular cell as in Equation 2.55. In this case,

\[ r = \sqrt{(x - x_0)^2 + (y - y_0)^2 + (z - z_0)^2}, \quad (2.51) \]

\[ x_{l\pm} = 2l_x L_x \pm x, \quad (2.52) \]

\[ y_{l\pm} = 2l_y L_y \pm y, \quad (2.53) \]

\[ z_{l\pm} = 2l_z L_z \pm z, \quad (2.54) \]

\[ p_{rect}(x, y, t) = \sum_{l, y, z=-\infty}^{\infty} p(x_{l\pm}, y_{l\pm}, z_{l\pm}, t). \quad (2.55) \]

where the sum is now a triple sum.

The relaxation and phase shifts however are dependent on the spectrum of the correlation function. To show this we express the correlation function in terms of the conditional probability, leaving it general for any field shape. The spectrum of the fields experienced by the ensemble is given as

\[ S_B(\omega) = \int_{-\infty}^{\infty} \langle B(t)B(t + \tau) \rangle e^{-i\omega\tau} d\tau, \quad (2.56) \]

where in terms of the conditional density and in the picture of the tessellated observer, specifying three dimensions,

\[ S_B(\omega) = \int_V B(x_0) d^3 x_0 \int_{-\infty}^{\infty} \tilde{B}(x) d^3 x \int_{-\infty}^{\infty} d\tau e^{-i\omega\tau} p(x_0) p(x - x_0, \tau) \quad (2.57) \]

where the \( \tilde{B} \) represents a periodic function of 2L. Next inverse transform back into the spectrum of the conditional density, given by Equation 2.45,

\[ S_B(\omega) = \int_V B(x_0) d^3 x_0 \int_{-\infty}^{\infty} \tilde{B}(x) d^3 x... \]

\[ \times \int_{-\infty}^{\infty} d\tau \int d^3 Q \int d\omega' e^{iQ \cdot (x_0 - x)} e^{i\omega'\tau - i\omega\tau} p(Q, \omega'). \quad (2.58) \]

The integration over \( \tau \) is a delta function for \( \omega' \), a constant times \( \delta(\omega' - \omega) \). Setting \( \omega' \to \omega \) gives,
\[ S_B(\omega) = \frac{1}{(2\pi)^3} \int_V \mathbf{B}(\mathbf{x}_0) d^3\mathbf{x}_0 \int_{-\infty}^{\infty} \tilde{\mathbf{B}}(\mathbf{x}) d^3\mathbf{x} \int d\omega \int d^3\mathbf{Q} e^{i\mathbf{Q}(\mathbf{x}_0 - \mathbf{x}) + i\omega \tau} p(\mathbf{Q}, \omega). \]  

(2.60)

Taking advantage of the \(2L\) periodicity of the tessellation, the integration over \(\mathbf{x}\) is simplified by integrating over the cell and its image, and then summed over all cells. This will in turn simplify the integration of \(\mathbf{Q}\),

\[ \int_{-\infty}^{\infty} \tilde{\mathbf{B}}(\mathbf{x}) d^3\mathbf{x} e^{-i\mathbf{Q} \cdot \mathbf{x}} = (2\pi)^3 \sum_{l_x, l_y, l_z} \frac{1}{L_x} \int_{-\frac{L_x}{2}}^{\frac{L_x}{2}} \int_{-\frac{L_y}{2}}^{\frac{L_y}{2}} \int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} d^3\mathbf{x} \mathbf{B}(\mathbf{x}) \times e^{-i\mathbf{Q} \cdot \mathbf{x}} \delta(Q_x - \frac{l_x \pi}{L_x}) \delta(Q_y - \frac{l_y \pi}{L_y}) \delta(Q_z - \frac{l_z \pi}{L_z}) \]  

(2.61)

Substituting Equation 2.61 into Equation 2.60 gives

\[ S_B(\omega) = \sum_{l_x, l_y, l_z} \frac{B_0(l_x, l_y, l_z) B(l_x, l_y, l_z)}{L_x L_y L_z} \left( \frac{l_x \pi}{L_x}, \frac{l_y \pi}{L_y}, \frac{l_z \pi}{L_z} \right), \]  

(2.62)

where \(B_0(l_x, l_y, l_z)\) and \(B(l_x, l_y, l_z)\) are the Fourier components of the field given by,

\[ B_0(l_x, l_y, l_z) = \sum_{l_x, l_y, l_z} \frac{1}{L_x L_y L_z} \int_{-\frac{L_x}{2}}^{\frac{L_x}{2}} \int_{-\frac{L_y}{2}}^{\frac{L_y}{2}} \int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \mathbf{B}(\mathbf{x}_0) e^{i\pi \left( \frac{l_x x_0}{L_x} + \frac{l_y y_0}{L_y} + \frac{l_z z_0}{L_z} \right)} d^3\mathbf{x}_0, \]  

(2.63)

\[ B(l_x, l_y, l_z) = \sum_{l_x, l_y, l_z} \frac{1}{L_x L_y L_z} \int_{-\frac{L_x}{2}}^{\frac{L_x}{2}} \int_{-\frac{L_y}{2}}^{\frac{L_y}{2}} \int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \mathbf{B}(\mathbf{x}) e^{-i\pi \left( \frac{l_x x}{L_x} + \frac{l_y y}{L_y} + \frac{l_z z}{L_z} \right)} d^3\mathbf{x}. \]  

(2.64)

Here, \(\mathbf{B}(\mathbf{x})\) is the original field from \(-L_q/2 \rightarrow L_q/2\) as well as the mirror image of the original field for \(L_q/2 \rightarrow 3L_q/2\). This is not a complete ensemble average; it is only for a single velocity. For realistic applications, as in the case of \(^3\)He, a Maxwell-Boltzmann distribution should be averaged. This will be discussed further in Section 2.4.

For the case of the gradient relaxation experiment, the gradient is constant. We assume a linear gradient along the \(x\) direction only. Computing the Fourier components gives the spectra, plotted in Figure 2.2,
Figure 2.2: The autocorrelation spectra due to a linear gradient for one, two, and three dimensions. Note the difference when higher-order dimensions are included.

\[
S_{1D}(\omega) = \sum_{n=\text{integers}} \frac{2v^2r_c^3}{\left(\frac{1}{2}\pi + n\pi\right)^2 \left(\omega^2 - \omega_n^2\right)^2 r_c^4 + \tau_c^2 \omega^2}, \quad (2.65)
\]
\[
\omega_n = \frac{v\pi (2n + 1)}{L_x}, \quad n = \text{integers}
\]

\[
S_{2D}(\omega) = \sum_{l_x=\text{odd}} \frac{8L_x^2}{\pi^4 l_x^4} \frac{1}{\sqrt{\left(\frac{1}{\tau_c} + i\omega\right)^2 + \frac{v^2\pi^2 l_x^2}{L_x^2} - \frac{1}{\tau_c}}}, \quad (2.66)
\]
\[
l_x = \text{odd integers.}
\]

\[
S_{3D}(\omega) = \sum_{l_x=\text{odd}} \frac{8L_x^2\tau_c}{\pi^4 l_x^4} \frac{1}{q_x \arctan\left(\frac{q_x}{1 + i\omega\tau_c}\right) - 1}, \quad (2.67)
\]
\[
q_x = \frac{\pi l_x v\tau_c}{L_x}, \quad l_x = \text{odd integers.}
\]

Figure 2.2 shows high peaks in the 1D model, smaller peaks in the 2D model, and steps
in the 3D model, this is due to the resonance of the oscillation between the walls, and is an important effect. However, it is clear that 1D is not a projection of 2D, nor is 2D a projection of 3D. This is expected. If we consider the ballistic limit in 1D, we would observe 2 points spreading away from the origin with speed $v$. In the 2D there is a circle expanding with speed $v$. If we project the 2D case onto a 1D axis, we would observe a continuous distribution with the far ends spreading with speed $v$ from the origin. This is shown in Figure 2.3.

A similar effect for the projection of 3D onto 2D exists. If we wish to unify the dimensions, we need to conserve the projections of the positions from the highest dimension. This is achieved by averaging velocity according to the distribution for that dimension, while assuming 3 is the maximum number of dimensions. Furthermore, velocity averaging is necessary to predict a relaxation and phase shift as the realistic systems are always of some distribution.

### 2.4 Velocity Weighted Ensembles

To correctly average velocity, it is required that position is conserved during a projection. This is accomplished by requiring conservation of momentum for the highest dimensional case,

$$
\langle p^2 \rangle = \langle p_x^2 \rangle + \langle p_y^2 \rangle + \langle p_z^2 \rangle .
$$

(2.68)
A symmetric distribution is assumed in the formulation of the conditional density; \( \langle v^2_{1D} \rangle = \frac{1}{2} \langle v^2_{2D} \rangle = \frac{1}{3} \langle v^2_{3D} \rangle \). For the case of \(^3\text{He}\), it obeys a Maxwell-Boltzmann distribution, \( e^{-\frac{mv^2}{2kT}} \) [34]. The gradient relaxation experiment is measuring \(^3\text{He}\) so only the Maxwell-Boltzmann distribution is considered. UCN are not in thermal equilibrium and will not obey this distribution.

We assume the form of a Maxwell-Boltzmann distribution. In one dimension,

\[
\begin{align*}
\frac{1}{\pi} \left( \frac{m}{kT} \right)^{\frac{1}{2}} e^{-\frac{mv^2}{2kT}} dv, \\
v_p &= 0,
\end{align*}
\]  

\( \sqrt{\frac{2kT}{\pi m}}, \)

\( \sqrt{\frac{kT}{m}}, \)

where \( v_p \) is the most probable velocity, \( v_{ave} \) is the average velocity, and \( v_{rms} \) is the root mean squared velocity.

In dimensions higher than one there is no need to account for the angular distribution; this is already accounted for in the conditional probability. The area element is included in the distribution. Using

\[
\begin{align*}
f(v) = \frac{m}{kT} e^{-\frac{mv^2}{2kT}} \\
v_p &= \sqrt{\frac{kT}{m}}, \\
v_{ave} &= \sqrt{\frac{\pi kT}{2m}}, \\
v_{rms} &= \sqrt{\frac{2kT}{m}},
\end{align*}
\]

This satisfies the requirement on the relation of \( v_{rms} \) from 1D to 2D

\[
v_{rms2D}^2 = 2 \times v_{rms1D}^2
\]

In three dimensions, the Maxwell-Boltzmann distribution is given by
\[ f(v) = \sqrt{\frac{2}{\pi}} \left( \frac{m}{kT} \right)^{\frac{3}{2}} v^2 e^{-\frac{mv^2}{2kT}}, \]  
\[ (2.77) \]

\[ (2.78) \]

and the quantities of interest are,

\[ v_p = \sqrt{\frac{2kT}{m}}, \]  
\[ (2.79) \]

\[ v_{ave} = \sqrt{\frac{8kT}{\pi m}}, \]  
\[ (2.80) \]

\[ v_{rms} = \sqrt{\frac{3kT}{m}}. \]  
\[ (2.81) \]

2.4.1 Relation to Diffusion Coefficient

The diffusion coefficient for different dimensions is given as

\[ D_n = \frac{\tau_c v_n^2}{n}, \]  
\[ (2.82) \]

where \( n = (1, 2, 3) \) is the number of spatial dimensions. It will be shown that \( v_n \) in the above equation is \( v_{rms} \) for the respective dimension. This comes from the fact that the conditional density for the diffusion equation is separable,

\[ p(x, y, z, t|x_0, y_0, z_0, t_0) = p(x, t|x_0, t_0)p(y, t|y_0, t_0)p(z, t|z_0, t_0). \]  
\[ (2.83) \]

A projection from a higher dimension to a lower dimension must result in the same description of that dimension. Therefore the diffusion coefficient must remain the same for any number of dimensions. This is most readily seen from analysis of the spectrum due to a linear gradient in \( x \). The 1D diffusion conditional density with a gradient in \( x \) results in the same expression for the power spectrum as the 3D conditional density with a gradient in \( x \). Projections of higher dimensions are already satisfied by lower dimensions in the diffusion equation so the diffusion constant must stay the same regardless of the number of dimensions. Also, the collision time should not be affected by a change in the number of spatial dimensions,
\[ D_1 = D_2 = D_3, \quad (2.84) \]
\[ \tau_c v_1^2 = \frac{\tau_c v_2^2}{2} = \frac{\tau_c v_3^2}{3}, \quad (2.85) \]
again, \( v^2 \) in the equation is referring to \( \langle v_n^2 \rangle \). Thus the diffusion constant becomes

\[ D_n = \frac{\tau_c \langle v_n^2 \rangle}{n}. \quad (2.86) \]

For a check we examine how this effects the mean free path. Because the collision time does not change with the number of dimensions, we can write the mean free path as

\[ \lambda_n = \sqrt{\langle v_n^2 \rangle \tau_c} = v_{rms} \tau_c. \quad (2.87) \]

The diffusion coefficient is preserved for different spatial dimensions while the correct spatial projection of the mean free path is observed \( \langle \lambda^2 \rangle = \langle \lambda_x^2 \rangle + \langle \lambda_y^2 \rangle + \langle \lambda_z^2 \rangle \). This is in agreement with the treatment in reference [21].

The velocity averaged spectra from Equations 2.65-2.67 are,

\[ S_{1D}(\omega) = \sqrt{\frac{2}{\pi}} \left( \frac{m}{kT} \right) \sum_{n=\text{integers}} \int_0^{\infty} dv \ e^{-\frac{mv^2}{kT}} \]
\[ \times \frac{1}{2v^2 \tau_c} \left( \frac{1}{2\pi} + n\pi \right)^2 \left( (\omega^2 - \omega_n^2)^2 \tau_c^4 + \tau_c \omega^2 \right), \]
\[ S_{2D}(\omega) = \frac{m}{kT} \sum_{l_x=\text{odd}} \int_0^{\infty} dv \ v e^{-\frac{mv^2}{kT}} \frac{8L_x^2}{\pi^4 l_x^4} \frac{1}{\sqrt{\left( \frac{1}{\tau_c} + i\omega \right)^2 + \frac{v^2}{L_x^2} - \frac{1}{\tau_c}}}, \]
\[ S_{3D}(\omega) = \sqrt{\frac{2}{\pi}} \left( \frac{m}{kT} \right)^3 \sum_{l_x=\text{odd}} \int_0^{\infty} dv \ v^2 e^{-\frac{mv^2}{kT}} \frac{8L_x^2}{\pi^4 l_x^4} \frac{1}{\arctan \left( \frac{q_x}{q_x + \tau_c} \right) - 1}. \]

Figure 2.4 shows the spectrum of the \( x \)-autocorrelation function; a model of \( ^3 \)He in solution with superfluid \( ^4 \)He at 100 mK. Assuming the \( ^3 \)He concentration is low so that phonon scattering is dominant, then at this low temperature we are in the ballistic limit. The velocity average is done using numerical integration. The upper limit is 4 times the most probable velocity, and the step size is set small enough so that the final plot has converged and is smooth. In the 1-D case step size is restricted by computing time and memory. The parameters for \( ^3 \)He in superfluid
Figure 2.4: The ballistic spectrum of a uniform gradient for all dimensions after velocity weighting, lower dimensions have become accurate projections of higher dimensions.

$^{4}$He and are: [35]

\[
D = 1.6 \times 10^7 \text{ cm}^2 \text{ s}^{-1} \quad (2.89)
\]

\[
\tau_c = 13.3 \text{ s} \quad (2.90)
\]

\[
\langle v_{1D}^2 \rangle = 1095 \text{ cm s}^{-1} \quad (2.91)
\]

\[
L_x = 2.54 \text{ cm} \quad (2.92)
\]

\[
\gamma^{3He} = 20378.9 \text{ radians s}^{-1} \text{ gauss} \quad (2.93)
\]

Figure 2.5 shows the spectrum of the 1D single velocity autocorrelation as compared to the 1D velocity weighted spectrum. Sharp peaks are seen in the single velocity curve. This is due to the neglect of the angular distribution in velocity, or at least its projection. The single velocity autocorrelation spectrum in 1D only considers one velocity in one direction. As a projection this is incorrect. The model neglects trajectories of the same speed but different directions. However, this is fixed in weighting the speed with a 1D distribution. The velocity weighting recovers the
Figure 2.5: The autocorrelation spectrum for the 1 dimensional case of $^3$He in superfluid $^4$He. The ballistic spectrum is for a uniform gradient in 1D. Plotted is both a single velocity and a Maxwell-Boltzmann distribution corresponding to a temperature of 100 mK.
Figure 2.6: The autocorrelation spectrum for the 2-dimensional case of $^3$He in superfluid $^4$He. The ballistic spectrum is for a uniform gradient in 2D. Plotted is both a single velocity and a Maxwell-Boltzmann distribution corresponding to a temperature of 100 mK.

correct projection found from an angular distribution of a higher dimensional theory. The noise in the 1D velocity weighted curve is due to the difficulty of achieving precision when numerically integrating over a series of sharp peaks. In the integration over velocity 200,000 points were used.

Figure 2.6 shows the same autocorrelation spectrum in 2D. As in the 1D, the peaks go away when using velocity weighting. Here the peaks are smaller because the angular distribution is partially accounted for in the 2D single velocity model. There is also less noise in the velocity weighted spectrum due to the smaller peaks in the single velocity model as compared to the 1D model.

Figure 2.7 is the similar spectrum for the 3D model. It is seen to have steps instead of peaks in the single velocity case. The peaks are not seen because the 3D model has a full account of the angular distribution of speeds. The steps are seen because only one velocity is considered, yielding a well-defined enter and exit time for all trajectories in all cell images. In the velocity averaging case the steps are washed away due to the expanded uncertainty in time a trajectory spends in the cell or its image. Very little noise is seen in the 3D spectrum due to the absence of peaks, making the 3D model the easiest to work with in this case.

The velocity averaged spectra shown in Figures 2.4 2.5, 2.6, and 2.7 are shown to be in agreement within the precision of the numerical integration. In a Brownian motion theory valid
for the transition from the ballistic through to the diffusive regime, the trajectories of lower dimensions are not projections of higher dimensions until the correct velocity projection is used.

2.4.2 Transition from Ballistic Through to the Diffusive Regime

Figure 2.8 shows the spectrum of the velocity averaged $x$ autocorrelation function versus concentration of $^3$He in solution with superfluid $^4$He at 300 mK and 40 G holding field. The transition from ballistic, to high frequency, then to the diffusion limit is observed with increasing $^3$He concentration. The diffusion coefficient for this regime is composed of $^3$He:phonon scattering [35],

$$D_{ph} = 1.6 \times T^{-7},$$

and $^3$He:$^3$He collisions [36],

$$D_{3\text{He}} = (1 + \frac{5.6}{T}) \frac{10^{-5}}{X}.$$

Here $X$ is the concentration ratio of $^3$He to $^4$He. At low concentration, the phonon scattering dominates the diffusion coefficient. This is seen by the curves ‘leveling off’ at the lower
concentrations, the temperature is low so that the low concentration limit is in the ballistic region.

The high frequency theory and diffusion theory are plotted from the function given in Equation 2.96 and 2.97

\[ S_{HF} = \frac{2 \langle v_{1D}^2 \rangle \tau_c}{\omega^2 (1 + \omega^2 \tau_c^2)}, \]  \hspace{1cm} (2.96)

\[ S_{Diff} = \sum_n \frac{16D}{\pi^2 n^2} \frac{1}{\left( \frac{n^2 \pi^2 D}{L^2} \right)^2 + \omega^2}, \quad n = \text{positive odd}. \]  \hspace{1cm} (2.97)

### 2.5 Simulation and Comparison to Theory

The goal of the simulations and theory is to both predict and attempt to minimize through design the geometric phase effect in the nEDM experiment. The continuous-time random walk is simulated and the phase shift is found from the position-velocity correlation function given in equation 2.21. Both specular reflecting walls and diffuse reflecting walls with a \( \cos \theta \) distribution are simulated.

The geometry for the simulations is a 10.2 cm × 40 cm × 7.6 cm cell. We take \( y \) as the long direction and \( z \), the polarization direction which is along the short length. For \(^3\text{He}\) the simulation is run at a temperature of 400 mK. The expected operating temperature of the experiment is 450 mK, however due to the exceedingly small phase shift at this temperature and increased velocity of \(^3\text{He}\), it is difficult to accurately perform simulations at this temperature. Only \(^3\text{He}\)-phonon collisions are considered. The neutrons are simulated with a \( v^2 \) velocity distribution, and a maximum velocity corresponding to a normal velocity of 165 neV at the walls, the expected Fermi potential of the walls. Neutrons do not undergo collisions in the bulk, only wall collisions are considered.

The sensitivity is calculated from the phase shift using Equation 2.98. For reference, \( G_z = 10^{-7} \text{ G/cm} \) and \( E = 50 \text{ keV/cm} \). The false EDM due to a phase shift is given by \( d_f = 4.56 \times 10^{-29} S(\omega) \text{ e cm} \), where \( S(\omega) \) is found from equations 2.26 and 2.88. The false EDM due to the phase shift of \(^3\text{He}\) is shown in Figure 2.9 and good agreement is seen with theory,

\[ 2\delta \omega \Delta \omega = \frac{2 \epsilon d_n \Delta E}{h}. \]  \hspace{1cm} (2.98)

For the neutrons, a realistic treatment of wall scattering is included. According to equation (1) of reference [37] given by,
Figure 2.8: Comparison of the different theories used to predict the spectra responsible for gradient relaxation. The diffusion theory is seen to diverge in the high frequency region because it does not consider gas collisions. This forces the spins to oscillating back and forth limiting the actual gradient seen by the individual spin and lowering its spectrum. The high frequency theory only considers gas collisions, so it diverges form the restricted theory in the ballistic region since the spins oscillating between the walls become the dominant part of the spectrum. Due to the long time between gas collisions, there is less oscillation, and hence a lower spectrum for the high frequency theory in this region. The diffusion spectrum is artificially high in this region because it instantly spreads itself uniformly over the whole cell.
Figure 2.9: Comparison of the simulated false EDM for $^3$He versus the 3D theory at 400 mK. 200,000 particles were used in the simulation.
Figure 2.10: Surface roughness of a dPS-dPTB coating on acrylic as determined using AFM. The resolution of this AFM image is 10 nm. Note that the surface appears flat on this scale.

\[ P_{ns}(\cos \theta_i) = \int I_{ns}(\theta_2, \phi_2) d\Omega, \]

\[ I_{ns}(\theta_2, \phi_2) = \frac{3}{2\pi} \frac{4}{3} k^4 w^2 b^2 \cos \theta_i \cos^2 \theta_2 \exp \left( -\frac{w^2 k^2}{2} \right) \times \left( \sin^2 \theta_i + \sin^2 \theta_2 - 2 \sin \theta_i \sin \theta_2 \cos \phi_2 \right), \]

where \( k \) is the neutron wave vector, and \( \theta_i, \theta_2, \phi_2 \) are the incoming and outgoing angles respectively. \( k \) is used for wall scattering by establishing the energy distribution of the neutrons. \( k \) is sampled from a parabolic velocity distribution with a cutoff of 165 neV, the expected Fermi potential of the dPS coating [5]. Upper limits can be set for the RMS roughness \( b \) and correlation length \( w \) based on atomic force microscopy (AFM) images of an actual dPS-dTPB coating as shown in Figure 2.10. The coatings look completely flat with a resolution of 10 nm. We thus set \( b = 10 \text{ nm} \) and \( w = 100 \text{ nm} \).

Figure 2.11 shows the false EDM for neutrons assuming an average gradient of \( 10^{-7} \text{ G/cm} \), the electric field of 50 kV/cm in a 3-D cell with dimensions \( 10.2 \text{ cm} \times 40 \text{ cm} \times 7.6 \text{ cm} \). The red curve corresponds to the case where the gradients are both positive, or both negative. This translates to summing the spectra. If one gradient is positive and one negative, the difference of the spectra are taken. Figure 2.12 is a plot of the same function along operating frequencies.
that result in an acceptable phase shift in regards to the targeted sensitivity.

2.6 Triangle Tessellation

The solution to the restricted geometry can be viewed as a tessellation of a cell and its images. The natural question to ask is do other geometries that tessellate satisfy the restricted random walk. Regular rectangles, triangles, and hexagons tessellate on the Euclidean plane. It is found that triangles do satisfy the requirements for a solution to the random walk in the time domain. Hexagons also satisfy the requirement with an initial position dependence of the images and their tessellations. The solution to the hexagon is still under investigation and will be covered in a later work.

The spectrum for these geometries are not readily found by the presented method because the boundaries are not periodic on a single dimension. However the time domain solution is applicable to the restricted diffusion spin echo problem, where the echo’s magnitude in the time domain is the main quantity of interest.

We start with the original equilateral triangle cell. The origin of coordinates is at the top vertex, so that the triangle cell is negative for all \( y \). We can create the initial hexagon ‘prison’ (a
Figure 2.12: False EDM due to the phase shift of neutrons over the frequency range that results in an acceptable phase shift in regards to the targeted sensitivity.
collection of cells) by rotating the triangle cell 5 times about the top point. The prison contains
all of the rotations and the initial triangle cell. The hexagonal prisons are tessellated by a simple
translation to the appropriate positions.

A $60^\circ$ rotation for every image is required. Furthermore the image should be reflected with
respect to the $x$ axis of the original triangle for each odd rotation, In matrix notation this is
written as

$$
\begin{bmatrix}
x'

\end{bmatrix} =
\begin{bmatrix}
\cos l\frac{\pi}{3} & -\sin l\frac{\pi}{3} \\
\sin l\frac{\pi}{3} & \cos l\frac{\pi}{3}
\end{bmatrix}
\begin{bmatrix}
(1)^l \\
0
\end{bmatrix}
\begin{bmatrix}
x
\end{bmatrix}
(2.101)
$$

$$
\begin{bmatrix}
\frac{\sqrt{3}L}{2} \\
\frac{\sqrt{3}L}{2}
\end{bmatrix}
\begin{bmatrix}
\begin{bmatrix}
x
\end{bmatrix}
\end{bmatrix}
(2.102)
$$

We insert our image points into the sum below,

$$
P_{\text{prison}}(x_0, y_0|x, y) = P_{\text{triangle}}(x_0, y_0|x, y) + \sum_{l=1.5} P_{\text{triangle}}(x_0, y_0|x'_l, y'_l) (2.103)
$$

where $P_{\text{triangle}}(x_0, y_0|x, y)$ is the propagation of probability found in equation 2.46. The triangle
subscript refers to the limits of $x_0, y_0$. They are bounded to the real triangle cell regardless of
$x, y$.

Now we must translate the prison. This is achieved by tessellating a hexagon.

$$
P(x_0, y_0|x, y) = \sum_{j, k=-\infty} P_{\text{prison}}(x + j\frac{3L}{2}, y + \text{odd}(j)\frac{\sqrt{3}L}{2} + k\sqrt{3}L), (2.104)
$$

where $L$ is the triangle side length and odd$(j)$ is given by

$$
\text{odd}(j) \equiv \begin{cases} 
0 & \text{if } j \text{ is even} \\
1 & \text{if } j \text{ is odd} 
\end{cases}.
(2.105)
$$

This is the conditional probability used to predict the spin echo attenuation based on the
average spread of phases with the position-position correlation function. According to equation
9 in reference [38],

\[ S(2\tau) = S_0 e^{-\frac{\gamma^2 g^2}{2} \langle x^2 \rangle} \]  
(2.106)

\[ \langle x^2 \rangle = \left( \int_0^\tau dt \int_0^\tau dt' - 2 \int_0^\tau dt \int_0^{2\tau} dt' + \int_0^{2\tau} dt \int_0^{2\tau} dt' \right) \]  
\times \langle x(t)x(t') \rangle, \]  
(2.107)

where \( 2\tau \) is the time between echoes. This method is only valid when the phase spreads slowly compared to the Larmor precession. If this is not the case, then \( 2\pi \) degeneracies in the phase contribute and the theory predicts a smaller than expected signal.

### 2.7 Diffuse Wall Reflections

Diffuse wall reflections is a difficult subject to approach analytically. There are no good models that predict accurate correlation functions to this date. Presented in this section is a model that approaches what is observed in the simulations and therefore is thought to be important enough to be included.

Considering the same principles as in the one dimensional velocity correlation function with specular scattering discussed in reference [39], we attempt to adjust for diffuse scattering with the inclusion of a damping parameter for lost correlation from a diffuse wall interaction. Upon each wall collision, the particle has probability to either scatter spectrally or diffusely. This can be represented by a parameter \( \alpha < 1 \), where \( \alpha = 1 \) is a completely specular collision. The velocity-velocity correlation function will take the form

\[ \psi(t) = \frac{v^2}{L} \int_0^L f(x,t)dx = \frac{v^2}{L} \left[ \int_{vt-L}^{vt} (-\alpha)^l dx + \int_{vt-L}^{L} (-\alpha)^{l+1} dx \right] \]

\[ = \frac{v^2}{L} \left[ (-\alpha)^{l+1} (L + Ll - tv) - (-\alpha)^l (Ll - tv) \right] \]

\[ = \frac{v^2}{L} \left[ A_l + \frac{t}{tw} B_l \right], \]  
(2.108)

where,

\[ A_l = (l + 1) (-\alpha)^l - (l) (-\alpha)^{l+1}, \quad B_l = (-\alpha)^{l+1} - (-\alpha)^l. \]  
(2.109)

Our task is to find the Fourier cosine transform
\[ \Psi(\omega) = \frac{v^2}{\pi} \sum_{l=0,1,2...} \left( A_l \int_{t_{\tau w}}^{(l+1)\tau_w} \cos(\omega t)dt + B_l \int_{t_{\tau w}}^{(l+1)\tau_w} \frac{t}{\tau_w} \cos(\omega t)dt \right). \] (2.110)

After integration we first consider terms linear in \( \sin(\omega t) \). Combining these we find

\[
\begin{align*}
&\left[ (l+1)(-\alpha)^l - (l)(-\alpha)^{l+1} \right] \sin [(l+1)\omega \tau_w] \\
+ &\left[ (l+1)(-\alpha)^{l+1} - (l)(-\alpha)^l \right] \sin [(l+1)\omega \tau_w] \\
+ &\left[ (l+1)(-\alpha)^l - (l)(-\alpha)^{l+1} \right] \sin [l\omega \tau_w] \\
+ &\left[ -(l+1)(-\alpha)^l + (l)(-\alpha)^{l+1} \right] \sin [l\omega \tau_w] \\
= &\ (-\alpha)^{l+1} \sin [(l+1)\omega \tau_w] - (-\alpha)^l \sin [l\omega \tau_w] \\
\end{align*}
\] (2.111)

From this we find the following

\[
\Psi_{\text{sine}}(\omega) = \frac{v^2}{\pi \omega} \lim_{N \to \infty} \sum_{l=0}^{N} \left[ (-\alpha)^{l+1} \sin [(l+1)\omega \tau_w] - (-\alpha)^l \sin [l\omega \tau_w] \right] \\
= \frac{v^2}{\pi \omega} \lim_{N \to \infty} \sum_{l=0}^{N} [f(l+1) - f(l)] = \frac{v^2}{\pi \omega} \lim_{N \to \infty} [f(N+1)] \\
= \frac{v^2}{\pi \omega} \lim_{N \to \infty} (-\alpha)^{l+1} \sin [(N+1)\omega \tau_w] \\
= 0.
\] (2.112)

Similarly for the cosine terms,

\[
\Psi_{\text{cosine}}(\omega) = \frac{v^2}{\pi \omega^2 \tau_w} \left\{ \lim_{N \to \infty} \sum_{l=0}^{N} B_l \cos [(l+1)\omega \tau_w] - \lim_{N \to \infty} \sum_{l=0}^{N} B_l \cos (l\omega \tau_w) \right\} \\
= \frac{v^2}{\pi \omega^2 \tau_w} \left\{ \lim_{N \to \infty} B_N \cos [(N+1)\omega \tau_w] - B_0 + \lim_{N \to \infty} \sum_{l=1}^{N} (B_{l-1} - B_l) \cos(l\omega \tau_w) \right\} \\
= \frac{v^2}{\pi \omega^2 \tau_w} \left\{ \lim_{N \to \infty} \sum_{l=1}^{N} (B_{l-1} - B_l) \cos(l\omega \tau_w) + (1 + \alpha) \right\}, \\
\] (2.113)

where
\[ B_{l-1} - B_l = (1 + \alpha) \left[ (-\alpha)^l - (-\alpha)^{l-1} \right] = -(1 + \alpha) \left[ 1 - (-\alpha)^{-1} \right] (-\alpha)^l \]
\[ = (1 + \alpha)(1 - \frac{1}{-\alpha})(-\alpha)^l = (1 + \alpha)(1 + \frac{1}{\alpha})(-\alpha)^l \]
\[ = (2 + \alpha + \frac{1}{\alpha})(-\alpha)^l \quad (2.114) \]

Recombining these two terms give
\[
\Psi(\omega) = -\frac{v^2(1 + \alpha)^2}{\pi \omega^2 \tau \omega \alpha} \left[ \left( \sum_{l=1}^{\infty} (-\alpha)^l \cos(l\omega \tau \omega) \right) + \frac{\alpha}{1 + \alpha} \right] \quad (2.115)
\]

We simplify using the identity in reference [40] section 1.447, number 2, for \(|\alpha| < 1\), (note that this identity is correct in the limit \(\alpha \to 1\)),
\[
\sum_{l=1}^{\infty} (-\alpha)^l \cos(l\omega \tau \omega) = 1 - \frac{1 + \alpha \cos \omega \tau \omega}{1 + \alpha^2 + 2\alpha \cos \omega \tau \omega}.
\]

Also, the phase shift according to the cosine-transformed velocity-velocity correlation function is given by equation (40) in reference [20],
\[
\delta \omega = -ab \int_{-\infty}^{\infty} \frac{\Psi(\omega)}{\omega_0^2 - \omega^2} d\omega.
\]

Taking notice that any constant term vanishes in this integration, we have
\[
\delta \omega = -\frac{abv^2(1 + \alpha)^2}{\pi \tau \omega \alpha} \int \frac{1 + \alpha \cos(\omega \tau \omega)}{\omega^2(\omega + \omega_0)(\omega - \omega_0)(1 + \alpha^2 + 2\alpha \cos \omega \tau \omega)} d\omega
\]
\[
= -\frac{abv^2 \tau \omega^2(1 + \alpha)^2}{\pi \alpha} \int \frac{1 + \alpha \cos(x)}{x^2(x + \omega_0 \tau \omega)(x - \omega_0 \tau \omega)(1 + \alpha^2 + 2\alpha \cos x)} dx, \quad (2.116)
\]

where \(x = \omega \tau \omega\).

The following integral can be evaluated using contour integration. First we consider the multi-pole at \(x = 0\). The residue of a second order pole can be found using the following process. First, multiply the integrand by \((x - x_0)^2\). Then take the derivative of the resulting function with respect to \(x\) and apply the limit as \(x \to x_0\). The resulting expression is the residue of the second order pole. For our case we have
\[
\lim_{x \to 0} \frac{d}{dx} \frac{x^2[1 + \alpha \cos(x)]}{(x + \omega_0 \tau_w)(x - \omega_0 \tau_w)(1 + \alpha^2 + 2\alpha \cos x)} = 0
\] (2.117)

\[
\lim_{x \to 0} \left[ \frac{\alpha \sin(\omega \tau_w)}{(x + \omega_0 \tau_w)(x - \omega_0 \tau_w)(1 + \alpha^2 + 2\alpha \cos x)} + \frac{(x + \omega_0 \tau_w)^2(x - \omega_0 \tau_w)(1 + \alpha^2 + 2\alpha \cos x)}{(x + \omega_0 \tau_w)(x - \omega_0 \tau_w)(1 + \alpha^2 + 2\alpha \cos x)} \right] = 0
\] (2.118)

The next two poles occur at \( x = \pm \omega_0 \tau_w \),

\[
R_{\omega_0 \tau_w} + R_{-\omega_0 \tau_w} = \lim_{x \to \omega_0 \tau_w} \frac{(x - \omega_0 \tau_w)[1 + \alpha \cos(x)]}{x^2(x + \omega_0 \tau_w)(x - \omega_0 \tau_w)(1 + \alpha^2 + 2\alpha \cos x)}
\] (2.119)

\[
= \frac{\omega_0 \tau_w^2(2\omega_0 \tau_w)(1 + \alpha^2 + 2\alpha \cos \omega_0 \tau_w)}{\omega_0 \tau_w(1 + \alpha^2 + 2\alpha \cos \omega_0 \tau_w)^2} = 0.
\]

We are left with an infinite sum of residues of imaginary poles at \( x = \arccos \left(-\frac{\alpha^2 + 1}{2\alpha}\right) + 2\pi m \), where \( m \) is any positive or negative integer, or zero. For now we shall consider,

\[
\lim_{x \to x_0} \frac{x - x_0}{1 + \alpha^2 + 2\alpha \cos x} = \frac{1}{2\alpha} \lim_{x \to x_0} \frac{x - x_0}{A + \cos x}
\] (2.120)

where \( x_0 = \arccos (-A) + 2\pi m \) and \( A = \frac{\alpha^2 + 1}{2\alpha} \). Using the Taylor series expansion of cosine as \( x \to x_0 \), we have

\[
\cos x = \cos x_0 + (x - x_0) \sin x_0.
\] (2.121)

The limit becomes,

\[
\frac{1}{2\alpha} \lim_{x \to x_0} \frac{x - x_0}{A + \cos x} = \frac{1}{2\alpha} \frac{1}{\sin x_0},
\] (2.122)
where,

\[ \sin x_0 = \sin (\arccos (-A) + 2\pi m) \]
\[ = \sin (\arccos (-A)) \cos(2\pi m) + \sin(2\pi m)(-A) \]
\[ = \sin(\arccos(-A)) = \sqrt{1 - A^2} = \frac{i}{2\alpha}(\alpha^2 - 1). \] (2.123)

Finally we have

\[ \lim_{x \to x_0} \frac{x - x_0}{A + \cos x} = -\frac{i}{\alpha^2 - 1}, \] (2.124)

which gives a phase shift of

\[ \delta \omega = \frac{-2\pi i av^2 \tau w^2 (1 + \alpha)^2}{\pi \alpha} \sum_{m=-\infty}^{\infty} \frac{i}{x_0^2(x_0 + \omega_0 \tau_w)(x_0 - \omega_0 \tau_w)(\alpha^2 - 1)} \] (2.125)
\[ = abv^2 \tau_w^2 \frac{(1 + \alpha)^2}{\alpha} \sum_{m=-\infty}^{\infty} \frac{1}{x_0^2(x_0^2 - (\omega_0 \tau_w)^2)} \] (2.126)

where \( x_0 = \arccos (-A) + 2\pi m = i \arccosh \left( \frac{\alpha^2 + 1}{2\alpha} \right) + 2\pi (m + \frac{1}{2}) \). \( \alpha \) cannot go to zero due to the probability of a particle diffusely scattering into its original trajectory, so \( 0 < \alpha \leq 1 \), and \( \arccosh \left( \frac{\alpha^2 + 1}{2\alpha} \right) \) is always real. Using equation 2.108, and assuming the form \((-\alpha)^l = (-1)^le^{i\beta} = e^{i(\pi - \beta)}\); the phase shift simplifies to

\[ \delta \omega = ab2L_i^2(1 + \cosh(\beta)) \sum_{m=-\infty}^{\infty} \frac{1}{x_0^2(x_0^2 - \omega_0^2)}, \] (2.127)

where \( x_0 = -i\beta - 2\pi (m + 1/2) \).

Now proceeding to solve for the velocity-velocity correlation function as a damped simple harmonic oscillator with a loss,

\[ \frac{d^2\psi_m}{dt^2} + \omega_m^2 \psi_m = 0, \] (2.128)

where \( \omega_m \) is the previous \( x_0 \) composed of
\[ \omega_m = -i \beta - 2\pi(m + \frac{1}{2}) \]  
(2.129)

\[ \omega_m^2 = -\beta^2 + 4\pi^2(m + \frac{1}{2})^2 + i4\pi\beta(m + \frac{1}{2}). \]  
(2.130)

This will give a real part and an imaginary part to \( \psi_m \), with coupling between the two via the \( i4\pi\beta(m + \frac{1}{2}) \) term in \( \omega_m^2 \). This coupling can be eliminated by solving for the real and imaginary parts within the complex equation. We are left with a square fourth order equation. This derivation follows, separating the wave function into real and imaginary parts

\[ \psi_m = \text{Re} \psi_m + i \text{Im} \psi_m, \]  
(2.131)

and inserting into equation 2.127 we find that the real part satisfies

\[ \text{Im} \psi_m = \frac{1}{4\pi\beta(m + \frac{1}{2})} \left\{ \frac{d^2 \text{Re} \psi_m}{dt^2} + (-\beta^2 + 4\pi^2(m + \frac{1}{2})^2) \text{Re} \psi_m \right\} \]  
(2.132)

and the imaginary part satisfies

\[ \frac{d^2 \text{Im} \psi_m}{dt^2} + (-\beta^2 + 4\pi^2(m + \frac{1}{2})^2) \text{Im} \psi_m + 4\pi\beta(m + \frac{1}{2}) \text{Re} \psi_m = 0. \]  
(2.133)

Substituting equation 2.132 into equation 2.133, we obtain

\[ \frac{d^4 \text{Re} \psi_m}{dt^4} + 2 \left(-\beta^2 + 4\pi^2(m + \frac{1}{2})^2\right) \frac{d^2 \text{Re} \psi_m}{dt^2} \]  
+ \left[ (-\beta^2 + 4\pi^2(m + \frac{1}{2})^2)^2 + (4\pi\beta(m + \frac{1}{2})^2 \right] \text{Re} \psi_m = 0 \]  
(2.135)

This equation has a solution of the form

\[ \text{Re} \psi_m = C1e^{-\frac{1}{2} \sqrt{-2b + 2\sqrt{b^2 - 4c}}} + C2e^{\frac{1}{2} \sqrt{-2b + 2\sqrt{b^2 - 4c}}} \]  
+ \[ C3e^{-\frac{1}{2} \sqrt{-2b - 2\sqrt{b^2 - 4c}}} + C4e^{\frac{1}{2} \sqrt{-2b - 2\sqrt{b^2 - 4c}}} \]  
(2.136)

where \( b = 2(-\beta^2 + 4\pi^2(m + \frac{1}{2})^2) \), and \( c = \left[ (-\beta^2 + 4\pi^2(m + \frac{1}{2})^2)^2 + (4\pi\beta(m + \frac{1}{2})^2 \right] \). Note that \( \sqrt{b^2 - 4c} = 2i(4\pi\beta(m + \frac{1}{2}) \). The exponential thus simplifies
$$\pm \sqrt{-\left(-\beta^2 + 4\pi^2(m + \frac{1}{2})^2\right)} \pm i \left(4\pi\beta(m + \frac{1}{2})\right)$$

$$= \pm \left(\beta \pm i2\pi(m + \frac{1}{2})\right)$$ \hspace{1cm} (2.137)

Substituting this into equation 2.136,

$$\text{Re } \psi_m = C_1e^{-(\beta + i2\pi(m + \frac{1}{2}))t} + C_2e^{-(\beta - i2\pi(m + \frac{1}{2}))t}$$

$$+ C_3e^{(\beta - i2\pi(m + \frac{1}{2}))t} + C_4e^{(\beta + i2\pi(m + \frac{1}{2}))t},$$ \hspace{1cm} (2.138)

where $C_3$ and $C_4$ must be zero for the constraint on $\lim_{t \to \infty} \psi_m(t) = 0$ to hold.

$$\text{Re } \psi_m = C_1e^{-(\beta + i2\pi(m + \frac{1}{2}))t} + C_2e^{-(\beta - i2\pi(m + \frac{1}{2}))t}$$

$$= e^{-\beta t}c_1 \left\{e^{-i2\pi(m + \frac{1}{2})t} + e^{i2\pi(m + \frac{1}{2})t}\right\}, \text{ (assuming symmetry)}$$ \hspace{1cm} (2.139)

$$= e^{-\beta t}c_1' \cos \left(2\pi(m + \frac{1}{2})t\right)$$ \hspace{1cm} (2.140)

$$= e^{-\beta t}\psi_{S.H.O.}$$ \hspace{1cm} (2.141)

Typically the solution of a decaying simple harmonic oscillator (S.H.O.) is for modeling for example the heating loss in a spring or some mechanism whereby energy can be released to outside the system. For the present system, the decaying simple harmonic oscillator models the diffusely scattering wall collisions. Gas collisions can be included in the bulk by writing the simple harmonic oscillator with a decay term similar to reference [21], equation (32).

Using this model of wall interactions, the position velocity correlation function’s spectrum is compared for a 2D simulation with the theory for three values of $\alpha$, $\alpha = 0.95$, 0.8, and 0.5. Shown in Figures 2.13, 2.14 and 2.15 the results resemble the same form but it is not convincing. Additional work on this is needed. The simulation is a 2D square starting with a single velocity in the $\pm x$ direction. For every wall collision, a random diffuse collision weighted by a $\cos \theta$ distribution will occur with probability $1 - \alpha$. Gas collisions are not considered in either theory or simulations.
Figure 2.13: The spectrum of the position-velocity correlation for a 2D simulation with diffuse wall scattering occurring for 5% of bounces as compared to the 1D diffuse wall scattering theory.
Figure 2.14: The spectrum of the position-velocity correlation for a 2D simulation with diffuse wall scattering occurring for 20% of bounces as compared with the 1D diffuse wall scattering theory.
Figure 2.15: The spectrum of the position-velocity correlation for a 2D simulation with diffuse wall scattering occurring for 50% of bounces as compared with the 1D diffuse wall scattering theory.
2.8 Summary

The geometric phase is shown to depend on the spectrum of the position autocorrelation function according to Equation 2.27. The same spectrum can be measured by gradient-induced longitudinal relaxation of spin polarized nuclei, shown in Equation 2.18. The conditional probability of the restricted continuous-time random walk valid for all mean free paths is found from the unrestricted conditional probability and the method of images. Using the restricted conditional probability spectrum the relaxation rate in Equation 2.18 can be determined by using Equation 2.88. After application of the velocity weighting the theory is shown to agree in the limited regions of validity of the previous theories, and diverge in the region where the resonance of the oscillation between the walls is the dominant contribution to the spectrum, shown in Figure 2.8. The theory and simulations agree in this region and it is expected that the theory is more precise than the simulations.

The theory presented to predict the frequency shift due to the geometric phase of $^3$He for the expected parameters of the nEDM experiment, shown in Figure 2.9, is tolerable for the desired sensitivity of $d_n < 3 \times 10^{-28}$ e cm. Furthermore, the geometric phase of the neutrons was simulated, shown in Figure 2.12, and is tolerable for the desired sensitivity. A theory to model the geometric phase including diffuse wall collisions is presented in Equation 2.126. Marginal agreement with simulations is shown in Figures 2.13-2.15.

An transform-tessellation is used with the unrestricted conditional probability to find the conditional probability restricted to a triangle. The theory can be applied to predict spin echo attenuation from diffusion valid for all mean free paths in triangular restrictions. This is presented in Equations 2.104 and 2.107.
Measurements of the relaxation of spin polarized $^3$He were performed at room temperature. This includes $T_2$ measurements in an applied gradient using spin echo technique, and gradient-induced $T_1$ measurements made by measuring the signal from free induction decay (FID) at various points over the time of the relaxation for varying concentrations. The room temperature measurements were performed in order to characterize the low temperature apparatus. The techniques were developed and tested in order to obtain a general understanding of various components. This included diagnosing NMR pickup and gradient coils, developing and constructing a pulse sequence, and measuring values of diffusion coefficient at room temperatures.

3.1 Spin Echo

A measurement of the diffusion coefficient at atmospheric pressure can be used to extrapolate the value to a lower pressure in the assumption that it is inversely proportional to the pressure. Since the diffusion coefficient is well known for $^3$He, we used it as a check of our system. Slight variations on the measured value are expected since we use a mixture with a ratio of 9:1 $^3$He to nitrogen. Diffusion can be measured using the spin echo technique with a known gradient, yielding a measurement of the $T_2$ relaxation. Diffusion can also be measured using $T_1$ relaxation with a known gradient [23]. The $T_2$ dependence on the gradient and diffusion can be found from the Torrey equation in the limit where diffusion is slow,
\[ D \nabla^2 p(x, t) = \frac{dp(x, t)}{dt} - i\gamma G_x x p(x, t), \]  

where \( D \) is the diffusion coefficient, \( p \) is the magnetization, \( \gamma \) is the gyromagnetic ratio, \( i \) is \( \sqrt{-1} \), and \( G_x \) is the linear magnetic field gradient. Assume \( p \) has the form

\[ p(x, t) = p(t)e^{i\gamma G_x xt}. \]

substituting this into the Torrey equation yields

\[
(-D\gamma^2 G_x^2 t^2 + i\gamma G_x x)p(x, t) = \frac{dp(x, t)}{dt} \tag{3.2}
\]

\[ p(x, t) = p_0 \exp \left( -\frac{1}{3}D\gamma^2 G_x^2 t^3 + i\gamma G_x xt \right). \tag{3.3} \]

When the phases have been aligned, there is a maximum of signal, a condition known as spin echo. The decay from diffusion is

\[ \frac{1}{T_2} = -\frac{1}{3}\gamma^2 G^2 D t^2. \tag{3.4} \]

Therefore, with diffusion slow so that boundaries play a negligible role [41], a spin echo in a known gradient can be used to measure the diffusion constant. The Hahn echo was used for the spin echo sequence and is shown in Figure 3.1. [27] The Hahn echo and other echo sequences are discussed in references [27,42].

There is an ambiguity in the value of \( t \) in the \( T_2 \) equation. The time between the original FID and its echo is \( 2\tau \). However the correct value for \( t \) in the \( T_2 \) equation is \( \tau \). Upon application of the \( \pi \) pulse, the random walk of the spins are reset. Any attenuation that has occurred remains but the time dependence for the decay should be considered to start from zero. The decay can be thought of as consisting of two parts, the part between the \( \frac{\pi}{2} \) pulse and the \( \pi \) pulse, and the part between the \( \pi \) pulse and the refocusing. During each part an attenuation occurs as a result of diffusion which has the form \( \exp(-\frac{1}{3}\gamma^2 G_x^2 D \tau^3) \). The attenuation between the original pulse and the spin echo is the same as the attenuation between subsequent echoes. Therefore we expect every echo to be attenuated by a factor,

\[ A_D = e^{\frac{2}{3}\gamma^2 G_x^2 D \tau^3}. \tag{3.5} \]
Figure 3.1: The Hahn echo sequence.

The initial spin echo was performed in a background gradient of unknown value. Very little information can be obtained without a known gradient. A gradient that is large compared to the background gradient can be applied in order to measure the diffusion coefficient.

### 3.2 Gradient Spin Echo

For spins precessing in a gradient a larger flat band pulse is needed to flip the spins, which now cover a larger frequency domain. A simple square pulse will only flip the fraction of spins that are precessing with the pulse frequency. In the approximation where the tipping angle is proportional to the power spectrum at the precessing frequency, a sinc function, which has a square frequency spectrum, can be applied to flip all the spins precessing in a gradient. The time constant of the pulse is correlated with the applied gradient and the length of the cell. For example, we have an applied gradient of $G_z$ and a cell of length $L_z$. For a uniform tip it is best to include at least 3 lobes. The experiment used 5 lobes,

$$B_1(t) = \text{sinc}(t)$$

$$t = \frac{\tau'}{\tau_B} \quad \text{and} \quad \frac{1}{\tau_B} = \gamma G_z \frac{L}{2}. \quad (3.6)$$
Figure 3.2 shows the spectrum of the 5-lobe sinc pulse. Note that the 3 lobe pulse requires integration of $t \in [-2\pi, 2\pi]$, and the 5 lobe $t \in [-3\pi, 3\pi]$. The approximation that the tip angle is proportional to the value of the Fourier transform at the frequency of the precession breaks down for tip angles of $\pi$ and larger [27]. Therefore it becomes important for a pulse’s spectrum to be flat if it is intended to flip the spins by $\pi$ radians. Numerical modeling of the Bloch equation simulate pulses that have a uniform tipping distribution over the applied frequency spectrum [42]. The pulses are complex which requires phase control of the applied pulse. The NMR control module used is the Apollo model by Tecmag. The Apollo’s phase control speed and precision are considerably more accurate than what is required for the measurements.

The pulse that was applied to our experiment is written as

$$B_1(t) = \left[ \text{sech} \beta t \right]^{1+i\Delta f}$$

$$\Delta f = \frac{\beta \mu}{\pi \Delta t}, \quad (3.7)$$

where $\mu = 5$ is a good $\pi$ pulse. In addition $t$ is centered on zero so that the final time $t_f = \Delta t/2$ [27].

As seen in Figure 3.3 this pulse supplies a sufficiently uniform power spectrum, and in turn, a uniform rotation of spins in a linear gradient.

The pulse input of the Apollo does not accept complex numbers as a pulse sequence. Instead
it must be converted into an absolute amplitude and a phase. This is achieved by

\[ B_1 (t) = A(t)e^{i\phi(t)} \]
\[ \phi (t) = \Im \left[ \ln \left( \left| \text{sech}(\beta t)^{1+i\mu} \right| \right) \right] \]
\[ A(t) = \left| \text{sech}(\beta t)^{1+5i} \right| . \]

(3.8)

(3.9)

The pulse sent by the Apollo to the RF coils is shown in Figure 3.4. This pulse consists of \( N_s = 1001 \) time steps. Given \( N_s \) the amount of time spent during one time step determines the frequency band of the power spectrum. The frequency distribution required is found from the gradient \( \Delta \Omega_z = \gamma_3H_eG_zL \). The appropriate time steps (\( \Delta t_N \)) are found from

\[ \Delta t_NN_s = \frac{\mu}{\pi\Delta \Omega_z} . \]

(3.10)

The spins are polarized and cooled to room temperature in the same manner as described in chapter 3.2.1 in reference [43]. The transfer cell is carefully placed in a portable magnetic field while the field is on. The magnetic holding field is tuned manually to a consistent field value. The coils are known to generate a precession frequency within one Hz of the desired value. The magnetic package, consisting of a solid 1.59 mm thick aluminum Faraday cage, an RF coil, a pickup coil and gradient coil, described in a subsequent section, is placed in the
Figure 3.4: The $\pi$ pulse used for spins precessing in an applied gradient. $N_s = 1001$ time steps are shown.

The Spin Echo is analyzed in the time domain. The maximum of the echo is measured and used to find the diffusion coefficient according to Equation 3.5. The noise can be reduced by applying a filter that only accepts signals in the frequency band of the precession of the spins determined from the applied gradient and the length of the cell.

In our case, this is achieved with a 4th order digital Butterworth filter. A linear transport function was constructed using the ‘butter’ function in MATLAB. There is a time lag in the echo maximum because the filtered signal does not contain the re-phasing of the higher harmonics of the echoes. However, the relative time between echoes and the maximum amplitude remains unchanged and the noise and unwanted signal outside the frequency band is suppressed. The noise for this experiment is acceptable with a 40 to 1 signal to noise ratio, although it can be seen in multiple echoes that an unwanted signal is present. The Fourier transform shows it centered at 28.3 kHz with a width of $\pm 1$ kHz. The source of this noise was never determined and could not be suppressed. The filtered time signal of run number 267 is shown in Figure 3.6.
Figure 3.5: A successful run of a spin echo in an applied gradient. The blue curve is the magnitude, and the red curve is the real part. The real part is seen to alternate between positive and negative values. This is due to the pulse sequence spin flip of $+y$, and $-y$ alternating. The measurement is at room temperature and pressure and measured a diffusion coefficient of $D_{^3\text{He}} = 1.52 \pm 0.01 \text{ cm}^2/\text{s}$. 
Figure 3.6: Data comparison of the original signal from run number 267, shown in blue, and the signal after the application of the 4th order Butterworth filter, shown in red. In the Butterworth filter only 30\% of the frequency band scanned by the Apollo is accepted. A cleaner signal is gained at the cost of a time lag due to a phase shift. The maximum signal only depends on the rephasing of the spins when the relative angle is zero, which is not lost in the filter. The band pass is almost three times the size of the frequency band of the precessing spins in the applied gradient.

and the spectrum is shown in Figure 3.7.

3.4 Longitudinal Relaxation Measurements

The correlation function can be measured from gradient-induced spin echo as shown previously in equations 2.18 and 2.88. The high frequency approximation is valid in the regions of concentration and Larmor frequencies used in the room temperature experiment. Other sources of relaxation must be accounted for according to the equation

$$\frac{1}{T_1} = \frac{1}{T_{1,\text{gradient}}} + \frac{1}{T_{1,\text{pulse}}} + \frac{1}{T_{1,\text{wall}}} + \frac{1}{T_{1,\text{spin}}}$$ \hspace{1cm} (3.11)
Figure 3.7: Data comparison of run number (267) after application of the 4th order Butterworth filter, accepting only the 30% of the frequency band scanned by the Apollo. It is shown that the unwanted signal around 28.5 kHz is largely suppressed by the filter while the echo signal remains unchanged.
where $T_{1\text{gradient}}$ is the relaxation that depends on the correlation function and the gradient strength, $T_{1\text{pulse}}$ represents the signal lost from the tipping of the spins and is responsible for the FID signal, $T_{1\text{wall}}$ represents the relaxation due to interactions with the wall including any magnetic impurities in or on the wall, and $T_{1\text{spin}}$ represents the spin-spin interactions which are completely negligible for our case due to the low density [43].

The measurement of the loss of polarization from tipping the spins is known from the calibration of the angle of tip and is given by

$$ \frac{1}{T_{1\text{pulse}}} = -\frac{\ln \left( \cos \left( \frac{\tau(\alpha)}{\tau(\frac{\pi}{2})} \frac{\pi}{2} \right) \right)}{\Delta t}, \quad (3.12) $$

where $\tau(\alpha)$ is the length of the applied pulse and $\tau(\frac{\pi}{2})$ is the length of a $\frac{\pi}{2}$ pulse. $\Delta t$ is the time between pulses and is constant throughout the relaxation measurement.

The spin relaxation of $^3$He on the wall is determined through measurements of relaxation without gradients. This value is independent of the concentration and diffusion coefficient in the regions of measurement available. This is due to the boundary condition of the transport to the walls, $D \frac{dc}{dr} = \alpha c$, where the diffusion constant $D$ is inversely proportional to $c$. Therefore, a maximum pressure is used in the measurement in order to give maximum signal, 0.5 atm. The wall relaxation was measured to be

$$ T_{1\text{wall}} = (7, 283 \pm 219) \text{ s}. \quad (3.13) $$

The gradient-induced relaxation of rarefied gas was performed as a proof of principle before the magnetic package was moved into the low temperature apparatus. The high frequency theory found in reference [23] is suitable to predict all of the relaxation times shown in Table 3.1. The resonance from the wall however cannot be observed without considerably lower concentrations. This would lead to strong relaxation from impurities in the tube and valve with very small initial signal.
Table 3.1: Table of room temperature $^3$He relaxation measurements. The 1 atm measurement was done in the polarization cell which has a relaxation time of $\approx 48$ hr. The diluted measurements were done in the dilution cell with wall relaxation time calculated using equation 3.13. The loss due to read out pulses is corrected for in the derived gradient relaxation, but it is not corrected for in the measured relaxation. The observation that the lowest pressure point is far from the predicted value is attributed to a non-negligible diffusion current into the tube where the surface to volume ratio is larger and thus results in a faster relaxation due to the surface collisions. Furthermore there may have be impurities in the tube or relaxation on the valve at the end of the tube. This effect is also seen in Table 3.2

<table>
<thead>
<tr>
<th></th>
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<th></th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>20.4</td>
<td>$1.053 \times 10^9 \pm 4.482 \times 10^4$</td>
<td>$3.467 \times 10^9 \pm 1.05 \times 10^9$</td>
<td>$3.81 \times 10^9$</td>
</tr>
<tr>
<td>0.00246</td>
<td>20.4</td>
<td>$1119 \pm 279$</td>
<td>$8984 \pm 703$</td>
<td>9278</td>
</tr>
<tr>
<td>0.00094</td>
<td>20.4</td>
<td>$233.6 \pm 212$</td>
<td>$1094 \pm 851$</td>
<td>3545</td>
</tr>
</tbody>
</table>
Table 3.2: The effect of an increasing diffusion current to a highly depolarizing surface.

<table>
<thead>
<tr>
<th>Pressure [atm]</th>
<th>Measured Relaxation [s]</th>
<th>Simulated Relaxation [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.164</td>
<td>412 ± 23</td>
<td>410</td>
</tr>
<tr>
<td>0.111</td>
<td>258 ± 13</td>
<td>273</td>
</tr>
<tr>
<td>0.049</td>
<td>63 ± 7</td>
<td>74</td>
</tr>
</tbody>
</table>

Room temperature measurements were performed using the low temperature apparatus prior to cooling down. The purpose was to examine the relaxation of the dPS-dTPB coating at room temperature while testing the new dilution volumes that were fabricated for the low temperature configurations. The volumes were installed on top of the the dilution refrigerator where the $^3$He transport glass tube enters the cryostat. They are supported on the same supports as the outer magnetic field coils.

The background gradient was measured to be 1.77 mG/cm. This size gradient only affects the spin dephasing and not $T_1$. A strong dependence on the diffusion constant of the relaxation time was observed as shown in Table 3.2.

The pressure dependence of the relaxation can be explained by the presence of a strongly depolarizing surface in the transfer tube. It is known that copper is strongly depolarizing if the spins are in contact. There is a copper section at the end of the transfer tube that attaches to the acrylic cell used for thermal anchoring. Using the predicted $T_1$ we can adjust to find the depolarization probability per bounce on the copper and the wall by the best fit to the simulation. A surface plot of the simulation is shown in Figure 3.8.

The simulation is the diffusion equation with depolarizing boundaries and is same as is used in the heat flush simulations without the convection term. There is an added section of highly depolarizing material in the transfer tube representing the copper in the copper to glass seal. There is a 1 mm diameter entrance hole through the acrylic cell. The depolarization on the cell wall should be on the order of an hour and plays a negligible role in the depolarization of the sample. The data is well fit with the copper section modeled by a depolarization per bounce of $5 \times 10^{-3}$.

3.5 Room Temperature Magnetics

The RF coil is a saddle shaped coil with a diameter of 7.62 cm. The angular aperture is 135 degrees. The length is 8.89 cm. It uses 22 gauge wire and has 10 turns. This coil will be used to generate magnetic fields of order 100 mG. The uniformity plot produced using FEM is shown in Figure 3.9. It is important that the field be as uniform as possible, with the intent that all the
Figure 3.8: Surface plot of the $^3$He polarization after a 500 s FEM simulation of the depolarization of spins dominated by the copper to glass seal. The larger tube on top represents the copper. It is modeled with a depolarization per bounce of $5 \times 10^{-3}$. 

Min: $6.091 \times 10^{-4}$
Max: 0.163
The RF coil magnetic field uniformity within the spherical cell with field lines. The volume average deviation of the field in the cell is 4.0%, while the maximum deviation is 15.2%. The maximum occurs out of the plane of the surface plot shown.

spins are tipped to the same angle. The variance of the field in the volume of the cell in the coil is 4.0%. This is suitable for our purpose, where the deviation in the RF field only affects the signal strength. A deviation from the mean would become more important if we were doing multi-dimensional imaging. This could be compensated with the knowledge of the field strength as a function of position in the cell so that the spin voxel could be flipped by the same angle over the whole volume of interest by scaling the RF field strength at each voxel.

The measurement coils are Helmholtz design with a 7.62 cm radius, they are wound from 30 gauge wires of diameter 0.254 mm and 0.3385 Ω/m. They have a 2.39 henry inductance, 19 pF capacitance and a measured resistance of 580 Ω. They are designed to be uniform in the volume of interest with enough turns so precession of rarefied spins can still be measured. The average variance of the field is 0.115% and the maximum deviation is 2.49%. It was assumed these coils would be the pickup coils in the low temperature apparatus. However they malfunctioned in the low temperature apparatus and were replaced by a superior design described in the low temperature section. They were successful in all the measurements in the room temperature.
Figure 3.10: The measurement coil uniformity within the spherical cell with field lines. The variance of the sensitivity in the region of the cell is 0.115% while the maximum deviation is 2.49%.

The gradient coils are a Maxwell 2-coil gradient configuration with a 25.4 cm diameter. They provide a uniform linear gradient in the volume of interest to fifth order. The gradient coils are controlled by the Apollo, increasing one control point in the Apollo increases the z-gradient by 80 $\mu$G/cm. The Apollo has 10,000 total control points making the largest possible gradient 0.8 G/cm.

The holding field used in both the spin-exchange optical pumping (SEOP) and the measurements themselves are from the same Helmholtz coils. The number of turns is no longer known nor is the information for the wire used. However, this information is irrelevant for our low field purpose. The coils are 76.2 cm in diameter, this large diameter produces a uniform field in the region of the measurement cell. Here 0.3 A produce a field of 27 G.

Another important part of the magnetics was the Q-switch for the pickup coil. The Q-switch was a JFET transistor that opened and closed a circuit, consisting solely of a resistor in parallel with the pickup coil. It worked by switching the the resistor off after the RF pulse for the
acquisition of the signal, then back on. The resistance was chosen by the critical damping of the parallel RLC circuit,

\[ R = \frac{1}{2} \sqrt{\frac{L}{C}}. \]  

(3.14)

The pickup coil is critically damped at 180 kΩ. The Q-switch reduced the ring time from 5 ms to 0.5 ms, enabling the measurement of the diffusion constant with a decay of echoes in an applied gradient with better precision. The shorter ring time enabled the use of a larger gradient for shorter decay times. Shorter decay times reduce the relative attenuation from other sources, for example the walls. The time scale, \( \tau \), of the successful gradient spin echo is 3.8 ms. This signal would be small compared to the ring time without the Q-switch.

The JFET used in the Q-switch added a source of additional ringing after it is switched off due to the release of the charge present across the small capacitance of the switch. This effect is reduced by gradually switching the voltage from the on to the off position. The gradual switching is controlled by the Apollo. A minimum of added noise was achieved with an exponential curve of the switching voltage applied to the JFET with the voltage value at the switch from on to off near time step 95 as seen in Figure 3.11. If the switch was not closed gradually or if the time of the gradual switch was reduced, the charge released from the switch of the JFET would cause the pickup coil to ring. Hence with the Q-switch, the ring time from the RF pulse was damped to much less than a millisecond, however the JFET must be switched gradually and is responsible for the minimum of 0.5 ms delay time to acquisition.

### 3.6 Room Temperature Rarefaction Volumes

The SEOP is used to polarize the spins. Polarization of the spins takes place at one atmosphere of pressure. In order for a viable rarefied signal, the \(^3\text{He}\) concentration must be rarefied with good accuracy, precision, and polarization. Two methods were investigated for this purpose. The first method attempted was a system of two Teflon bags. The second system utilized was a series of three evacuated glass volumes.

The Teflon bag method consisted of a glass tube that connected the \(^3\text{He}\) transfer cell to a series of two tees with a bag after a valve on each of the tees. A valve separated the bags in series from the measurement cell. Polarization friendly Teflon valves were used for all the valves. The Teflon bags were used because they have spin relaxation times of over an hour. The Teflon bags were sealed in an air tight PMMA container. The container could be evacuated or pressurized separately from the bags.
Figure 3.11: The signal sent to the Q-switch to allow the spin precession signal into the Apollo. The signal is sent to the JFET to detach the resistance in parallel with the pickup coils during acquisition of signal. The signal was tuned so that the switch occurs at time step 95. Typically, the 100 steps occur in a time of 0.5 ms.
This transfer method worked but was inadequate. The Teflon bag is first evacuated, then the container was evacuated. The order is important for preventing the bag from exploding. The $^3$He from the transfer cell would be allowed into the first bag. Once the valve was opened it would be sucked in from the vacuum in the container. A second bag would be evacuated, but its container would have a small amount of pressure, greater than the pressure in the first bag with the $^3$He now inside. When the second bag was opened to the first container, a known portion of the helium would go into the second bag. The valve to the first bag is closed and the measurement cell valve is opened, the measurement cell is at high vacuum prior to opening the valve. The helium in the second bag expands into the measurement cell. The container around the second Teflon bag can be pressurized further to squeeze the bag empty. In the end a fixed amount of polarized $^3$He is in the measurement cell, while the first bag still contains a majority of the $^3$He and can be used for subsequent measurements.

The benefit of this method is the the first bag acts as a reservoir for the $^3$He. A measurement can be repeated until the helium in the bag is depleted. Many rarefied measurements could be performed from a single polarized cell by pumping out the measurement cell prior to repeating the filling and measurement procedure.

The method however had several shortcomings. The bags were attached to the glass tubes using a threaded plastic turn knob to tighten an o-ring seal on the tube. It was found that vacuum on the outside while pressure on the inside could force these seals to leak. Also, the bags would tear or explode if the pressures in the bags relative to the containing volumes were not accurate. Furthermore the accuracy and precision of the rarefication was sufficient on the order of $50 \times$ dilution factors, but not on the order of $1000 \times$ dilution factors.

The second and preferred method used three glass dilution volumes connected to the transfer cell and the measurement cell. All dilution volumes are 500 cc. The transfer cell and the measurement cell are both 69 cc, therefore if all the volumes are used consecutively after a transfer to the measurement cell the dilution factor is $9 \times 10^{-4}$.

After dilution, the $^3$He in the dilution volumes is no longer useful, unlike the bags, there is no method to force the $^3$He out of the glass volumes other than the intrinsic pressure. The $^3$He in the dilution volumes are at a low pressure, the highest being around 61 mbar. The depolarization in the dilution volumes is fast due to the large background gradients arising from the holding fields generated from the measurement and transfer coils. There is no holding field for the dilution volumes. The $^3$He is no longer viable for spin measurements once it enters a dilution volume.
3.7 Room Temperature Measurement Summary

Measurements of the relaxation of spin polarized $^3$He were performed at room temperature. This includes $T_2$ measurements in an applied gradient using spin echo technique, and gradient-induced $T_1$ measurements made by measuring the signal from free induction decay (FID) at various points over the time of the relaxation for varying concentrations, shown in Table 3.1. The room temperature measurements were performed in order to characterize the low temperature apparatus. The diffusion coefficient calculated from $T_2$ measurements in an applied gradient using the spin echo technique and is in agreement with the expected value. Viable dilution volumes and pulse sequences were constructed resulting in a successful measurement of the gradient-induced $T_1$ of spin polarized $^3$He after rarefication of a factor of 1000. Although the measured relaxation did not agree with theoretical predictions in that region, the discrepancy can be accounted for by a diffusion current into the transfer tubes, where the spins will relax much faster due to the impurities in the tube and a larger surface to volume ratio.
Work began on the low temperature apparatus after room temperature measurements were completed, the room temperature measurements justified the plausibility of a measurement of the correlation function at low temperatures. Runs at room temperature that observed the spin relaxation of $^3$He rarefied 1000 times provided this justification. The work performed on the low temperature apparatus included operation of the dilution refrigerator (DR), the rarefication of the sample with the dilution volumes, the transport of the diluted sample into the low temperature helium, and the magnetics of the low temperature apparatus. As with most physics experiments, there were significantly more hurdles to overcome than expected.

4.1 Experimental Procedures

The polarizing cell that contains anywhere from 10% to 30% spin-polarized $^3$He, is transported from the polarizing setup to the apparatus using a portable Helmholtz magnetic field coil powered by 3 car batteries in series. The arrangement is moved using a metal wagon. The holding field is raised 1 m above the metal by a wooden table so that there are minimal losses from the metal wagon. The cell is transported from Physics, room 014, to the French Family Science Center, room 1127. There are no measurable losses observed during transport, as measured using the portable Free Induction Decay (FID) system.

Two people are needed to mount the system on top of the DR to the dilution system. Once the transport field is mounted on the DR, the top holding field for the transfer cell is turned on. The transfer cell is carefully brought to the helium sample tubing and attached. The transport coils are removed after they are turned off by removing the jumper cable from the car battery powering the coils. This is done in a smooth motion to minimize sparking the jumper cable to
the battery terminal, which may cause an unwanted broad spectrum pulse in the holding field.

Once tightened on the apparatus, the $^3$He polarization in the cell is measured with the portable FID system. In order for the cell to be attached to the apparatus, a small portion of sample transport tubing, 0.32 cc, must be open to atmosphere. After the transfer cell is connected, the tubing system is pumped until a pressure $1 \times 10^{-6}$ mbar is reached. With a helium cold trap in place, this typically takes only a few seconds. Prior to the installation of the helium cold trap, 25 min. pumping times were required. The dilution procedure starts after the pressure of the sample gauge is below $1 \times 10^{-6}$ mbar.

### 4.1.1 Dilution Volumes

Dilution can only begin if the dilution volumes shown in Figure 4.1 have been previously pumped out. A pressure of $2 \times 10^{-7}$ mbar on the pressure gauge above the sample turbo is typical with the
helium cold trap attached to the system. The helium cold trap is discussed in subsection 4.5.8.

For the fully developed measurement cycle, up to four relevant measurements with good statistics can be made from a single transfer cell, saving precious $^3\text{He}$. The most recent method saved the spin in the transfer cell by keeping the top holding field on. This disrupts the measurement field, but the uniform field can be recovered by tuning the current in each of the measurement coils separately. The $^3\text{He}$ is transferred from the transfer cell to the intermediate cell so that only half of the atoms in the transfer cell are lost per transfer. This results in measurements of four different concentrations per transfer cell, where each measurement is increasing in concentration from the previous measurement. Typical concentrations measured ($^3\text{He}$ concentration, not polarization concentration), are

$$
X_1 = 1.265 \times 10^{-6} \\
X_2 = 5.500 \times 10^{-6} \\
X_3 = 1.516 \times 10^{-5} \\
X_4 = 1.666 \times 10^{-4}
$$

This technique was not completely developed until the last two runs. The holding field had to be tuned in the measurement region to account for the current in the top holding coils. Before that technique was realized, only a single measurement was completed per transfer cell, this was done in multiples of at least $X = 1.041 \times 10^{-6}$. Further combinations can be made by utilizing the dilution volumes in any order possible.

The volumes are:

<table>
<thead>
<tr>
<th>Volume</th>
<th>Volume</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transfer Cell</td>
<td>68.6 cc</td>
</tr>
<tr>
<td>Intermediate Volume</td>
<td>68.6 cc</td>
</tr>
<tr>
<td>Dilution Volume 1</td>
<td>615.8 cc</td>
</tr>
<tr>
<td>Dilution Volume 2</td>
<td>423.3 cc</td>
</tr>
<tr>
<td>Dilution Volume 3</td>
<td>392.5 cc</td>
</tr>
<tr>
<td>Measurement Cell</td>
<td>24.6 cc</td>
</tr>
</tbody>
</table>

The lengths of the tubes between the volumes are much less than one percent of the volume and are neglected in the calculation of concentrations. The transfer tube from high temperature into the low temperature cell is a significant volume, however the pressure is low enough from the cryopumping of the measurement cell to ignore this contribution.
We equalized the transfer cell with the intermediate cell, so that each have half of the initial concentration (0.167 atm of \(^3\)He were in each). The valve between the transfer cell and intermediate cell is then closed.

The procedure that achieves the desired dilution is as follows:

- The intermediate volume is opened and the pressure is allowed to equalize with the first dilution volume for a few seconds. The intermediate volume and first dilution volume are isolated.

- The second dilution volume is connected to the intermediate volume, and allowed to equalize for 10 seconds, the intermediate volume and second dilution volume are isolated.

- The third dilution volume is connected to the intermediate volume and allowed to equalize for 10 seconds. The intermediate volume and third dilution volume are isolated.

When using a dilution volume to dilute the intermediate cell, it is important to open the intermediate volume last and close it first. This prevents migration of the spins around the volumes and minimizes contact with high gradients or strongly relaxing surfaces. There is no data to support this, but it is a worthwhile precaution.

Finally, we can calculate the total pressure in the intermediate volume,

\[
\frac{1}{3} \text{atm} \times \frac{1}{2} \times \frac{68.4}{615.8 + 68.4} \times \frac{68.4}{423.3 + 68.4} \times \frac{68.4}{392.5 + 68.4} = 3 \times 10^{-4} \text{atm} = 0.3 \text{mbar}.
\]

At 21.1\(^\circ\)C and 1 atm, the number density of \(^3\)He gas is \(3.295 \times 10^{19}\) atoms/cc [44]. Therefore \(6.76 \times 10^{14}\) \(^3\)He atoms enter the cell, resulting in a final molar concentration ratio of \(1.26 \times 10^{-6}\).

Multiple transfers can be completed by pumping out the dilution volumes during the relaxation measurement of the sample in the cell. Due to constraints in the signal pickup it is not efficient to measure with less than \(6.76 \times 10^{14}\) atoms in the cell. Therefore it is not useful to do the same rarefication as with the full transfer cell now that the transfer cell has half the atoms it once had. For example, we could use the 615.8 and 423.3 volumes during the same dilution step to achieve a low concentration, but at a level still sufficient to observe signal. This dilution technique provides four concentrations to choose from, with an adequate range to compare to theory spanning different concentration regions.

Once the dilution process is completed, the sample is allowed to expand into the measurement cell for the measurement. There is initially a temperature spike which falls quickly to a new equilibrium temperature within approximately 1 min. When the temperature is steady, the measurement begins.
4.2 Pressurize the Sample

Once the intermediate cell contains the desired amount of $^3\text{He}$, the measurement procedure begins. The transfer coils are turned on manually and the holding field coils are turned on via the LabView program. In addition the heater current in the still of the DR is turned off. The valve to the measurement cell remains closed, and the valve to the transfer cell is closed. However, the valve to the gas handling system is open. Every valve on the gas handling system is closed except for a buffer volume holding isotopically pure gas held around 500 torr. The gas is used to is pressurize the tube up to the transfer cell, then the transfer cell valve is opened very briefly allowing the isotopically pure $^4\text{He}$ to pressurize the transfer cell. The final pressure in the buffer volume and the intermediate cell is about 380 torr or 500 mbar. The gas handling valve is closed and the measurement cell valve is opened, allowing the small about of 500 mbar helium left in the tube to go into the measurement cell. Then, the intermediate volume valve is opened allowing the 500 mbar polarized helium sample to enter the measurement cell.

When the transfer coils are turned off, the heater current in the still of the DR is turned on and the temperature reaches its steady state the measurement begins.

4.3 Measurements

The $^3\text{He}$ spins are in a magnetic field and thus precess at the Larmor frequency, $(\omega_0/B_0 = 20378.9 \text{ G})$. Figure 4.2 shows a screen shot of a pulse sequence used to measure the gradient induced relaxation on the NTNMR program by TecMag Apollo. Gs_Shape and Gs_Amplitude are the signals used to control voltages that operate the relay disconnecting the gradient coils. The gradient coils are disconnected because it is beyond the precision of the power supply (AETechron) to calibrate the zero current as accurately as needed. (The best is $\approx 100 \pm 50 \mu\text{A}$.) This offset current in the coils causes the signal to decay due to reversible dephasing of the $^3\text{He}$ spins ($T_2^*$). $T_2^*$ is different from $T_2$ in that $T_2^*$ decay can be reversed to recover the signal by flipping the spins an angle $\pi$, or by reversing the gradient. The superconducting gradient coils (SCG) are disconnected for 10 s which allows any transients to decay. This decay time was found empirically and is consistent with the expected value for 650 turn superconducting coils with coils wound in opposite directions. It should be noted that these transients will strongly affect $T_2^*$, but have negligible effect on $T_1$ according to the theory. The static gradient relaxation is negligible for such a small gradient and the natural resonant frequency of the ring time ($\approx 0.5 \text{ Hz}$) is far from the Larmor frequency.
Figure 4.2: Screen shot of the pulse sequence used for the gradient-induced relaxation measurement.
After the 10 s delay, there is a period of 500 ms delay for the Apollo to reset the phase of the read out pulse. The pulse is a sine wave with a phase determined by the timing of the pulse sequence and the direction of the axis of flipping. The pulse is sent from the Apollo to the RF amplifier. The amplified signal is passed on to the RF coils in the apparatus. The Apollo has control of the phase. Typical pulse lengths are 85 $\mu$s, corresponding to 33.8% of the total signal and a loss of 5.9% of the polarization or a tip angle of 48.5 degrees. The RF coils are tuned to a field of 2 G and the tipping angle, $\alpha$ is determined by the length of the pulse supplied. From pulse calibration, $\alpha$ is known to better than 0.05%. Many calibration measurements are taken to ensure the frequency of the tipping pulse is as close to the holding frequency of the spins as achievable. Once the spins have been flipped, the Apollo measures the voltage from the free induction decay (FID) supplied by currents in the coils generated from the magnetic flux of the precessing spins. Visible signal may be seen for longer than 3 s depending on non-uniformity. Typically, the majority of the signal has decayed before 1 s. After the acquisition, the Apollo switches the relay connecting the gradient coils and Helmholtz coils to the power supply, followed by a 0.5 s rest period for the oscillations in the superconducting Helmholtz coils (SCH) and SCG due to the hard step up to the offset current in the superconducting coils. The SCH and SCG’s current is ramped to the desired value via 2000 equal increments equally spaced over 2 s. The spins are left in the field configuration for 1 min., then the field for the SCH and SCG are ramped down via 2000 equal increments equally spaced over 2 s. After another 0.5 s waiting period, the cycle starts over with the 10 s delay prior to the second tipping and acquisition of signal by the pickup coils. The total sequence lasts 1:19.54 and is repeated from 50 to 150 times depending on the measurement. The signal typically decays into background between cycle 15 and 25. Measurements after the 25th sequence are used for background and noise estimates.

The gradient-induced relaxation pulse sequence contains the sections of all pulse sequences used to calibrate and characterize the systematics of the experiment. Other modifications to this sequences are for example the gradient strength ($G_{str}$) value is set to zero to measure the wall relaxation. The pulse length/tip angle calibration starts at 500 ms (the time to reset the phase) and ends after the acquisition period. It repeats 10 times. By taking away the 60 s relaxation period, a comparison of the pulse loss and the loss due to the ramping of the gradient coils is seen. It is verified to be a negligible loss.

Spin echo were performed at the low temperatures but due the relation that $T_2^* \approx T_2$, thus yielded information that our applied frequency was fractions of a Hertz from the precession frequency of the spins. No other useful information is seen from the spin echo because the $T_2^*$ is the same as $T_2$. The technique to measure $T_2$ using spin echo in a known/supplied linear gradient was never used for the low temperature experiment. Gradient induced $T_2$ measurements require fast-changing low-power gradient coils, while the SCG was designed to slowly change high power coils.
4.4 Magnetization in the Apparatus

The field inside the measurement cell can become non-uniform if a magnetizable material is in close proximity. Magnetization was deduced by the history of the applied magnetic fields correlated with the observed $T_2^*$ signal. $T_2^*$ becomes shorter when the material is magnetized. Short $T_2^*$ values result in a diminished signal. The magnetization can diminish $T_2^*$ by up to 100 times. In the apparatus, a stainless steel capillary is used to condense the $^4\text{He}$ into the cell. The stainless steel may be responsible for the magnetization that was observed. The other suspect is the superconducting indium loops used in sealing the mixing chamber and buffer volume.

The magnetization can be tuned so that it does not contribute to $T_2^*$. It is tuned by ramping SCH and SCG. The most effective sequence observed is to first ramp the SCH to 60 G, leaving the SCG open; the maximum value of the field of the SCH. Hold this field for 4 seconds then ramp down. Then simultaneously ramp SCH to 60 G and SCG to 3.13 G/cm and hold for 4 s. This cycle works by forcing the magnetization to a known point on the hysteresis curve, which enables us to force it back to zero. This demagnetization sequence is applied several times immediately before the portable $^3\text{He}$ transfer cell has been fixed atop the apparatus.

Magnetization can be avoided during the measurement by maintaining a fixed ratio for the gradient supplied current relative to the Helmholtz supplied current, $G_{str} = 1.89 \ H_{str}$, during the relaxation portion of the pulse sequence. It is assumed that this forces a zero field in the region of the ferrous material. With these currents, the simulations show the zero field occurs in the region of the buffer volume. It would be beneficial to the experiment for this magnetization to be absent because the given ratio does not relax the spins in the most sensitive time frame. If the gradients could be made larger, the spins would relax faster and there would be less wall relaxation relative to gradient relaxation, making background subtraction more precise.

4.5 Prepping the Dilute Sample for Transfer

The $^3\text{He}$ will not transfer if there is not significant pressure to exceed the vapor pressure in the tube. This pressure can be estimated assuming the film flow from the measurement cell into warmer parts of the tube.

4.5.1 Film Flow Investigation

The gas entering the restriction is assumed to be at 0.5 K with a viscosity of $0.0017 \ \text{g cm}^{-1} \text{s}$. An argument as to why we need to overpressure the transfer cell for the dilute $^3\text{He}$ to go into the sample can be made from the following estimate/calculation. The helium in the measurement cell is held at or below 500 mK. Its vapor pressure is negligible at this temperature. The source of vapor in the tube is thus dominated by the vapor from the superfluid helium film.
The helium film will flow up the pipe at a rate dependent on the tube’s surface and geometry. If there is enough mass flow from the film to overcome the gas flow from the pressure gradient it establishes to the bulk, the vapor from the film can accumulate. If we assume that the change in temperature seen when the cell is filled with superfluid helium is due to the film flow, we can estimate the volume rate of flow up the tube. We will then estimate the gas pressure gradient the film flow can support in the molecular flow regime. The heat transport is described by,

\[
\frac{dQ}{dt} = (L + ST) \frac{dV}{dt},
\]

where

\[S_{ph} = \frac{2\pi^2 k_B^4 T^3}{45 \rho \hbar^3 e^3},\]

\[S_r = \frac{2 (k_B \mu)^{\frac{1}{2}} \rho_0^2}{(2\pi)^{\frac{1}{2}} \rho \hbar^3 T^{\frac{1}{2}}} \left( 1 + \frac{3k_B T}{2\Delta} \right) e^{-\frac{\Delta}{k_B T}},\]

\[S = S_{ph} + S_r,\]

Here \(L = 2.56 \times 10^7 \text{erg/cc}, \rho = 0.145 \text{g/cc}\) and \(\frac{dV}{dt}\) is the volume flow. Entropy in the film is not well known, and the equations for entropy of the bulk liquid are only good to 1.5 K [45]. The \(S \times T\) in equation 4.1 is small compared to the latent heat \(L\). Based on the heat load vs. temperature curves for the DR, we know the heat load due to the film is 486 erg/s. If we assume that the majority of the film can flow to a temperature around the lambda transition \((T \approx 2.2 \text{K})\), we find that this is equivalent to a volumetric flow of \(1.9 \times 10^{-5} \text{cc/s}\) of liquid or \(1.3 \times 10^{-4} \text{cc/s}\) of gas at 4.2 K [46]. This volume flow is in agreement with examples in the literature for a restriction of 0.5 mm diameter [47]. The density of helium vapor at boiling point is 0.017 g/cc about seven times the liquid density [46].

In order for the gas to recombine with the bulk, it must travel through a restriction with a diameter of 0.5 mm and 0.5 cm in length,

\[
\dot{N} = C \ n,
\]

where \(\dot{N}\) is the number of gas atoms passing through the capillary per second, \(C\) is the conductance in liters/s, and \(n\) the number density of the incoming gas. In this model, the outgoing gas is absorbed into the bulk. When equilibrium is reached, \(\dot{N}\) down the tube is the same as the film flow up the tube. This number is calculated to be (from film flow)
\[ \dot{N} = 4.7 \times 10^{17}. \]

\[ C \text{ is given as } [48] \]

\[
C = \frac{12.1d^3}{L(1 + \frac{4d}{3L})} \times \sqrt{\frac{5 \cdot 28}{300 \cdot 4}}
\]

\[ = 0.00033 \text{ l/s} = 0.33 \text{ cc/s} \quad (4.4) \]

The number density of the gas that is entering is calculated to be

\[ n = 1.2 \times 10^{18} \text{ atoms/cc.} \]

This number density corresponds to \( \approx 0.05 \text{ mbar at 500 mK} \) or \( \approx 30 \text{ mbar at room temperature} \). In the assumption that the temperature of the sample cools to the temperature of the capillary as it arrives, then the pressure in the transfer volume should be increased to well over 30 mbar to overcome this barrier pressure. The experiment uses 500 mbar to pressurize the sample. Previous to pressurizing the sample no signal after 1000 times rarefication, resulting in a pressure of 1 mbar, was observed when the \(^3\)He was allowed into the cell.

4.5.2 Superconducting Magnets

There are two superconducting magnets, the superconducting Helmholtz (SCH) and superconducting gradient coils (SCG). The SCH consist of two coils each with 250 turns, a diameter of 26.67 cm set by the IVC and separation equal to the radius. It creates a uniform \( B_0 \) field up to 100 G. The non-uniformity of the field produces a \( T_2^* \) of 10 ms at 100 G and has a negligible effect on \( T_1 \). The SCG configuration consist of two coils each with 650 turns, a diameter of 26.67 cm and a separation of \( \sqrt{3} \) times the radius. This Maxwell gradient configuration produces a gradient up to 20 G/cm and linear to fifth order. A 1 \( \Omega \) power resistor is placed in series with the SCG. It is observed that the resistor greatly reduces the decay time of the transient oscillations that are generated when the current in the SCG is changed. The disruptive magnitude of these oscillations is attributed to the large number of turns and opposing winding directions in the coils. We did not observe as drastic oscillations in the SCH coil, due to the smaller number of windings and the same winding direction in both coils.

Both the SCH and SCG are wound around the IVC of our apparatus. The first attempt to wind the coils failed. They were wound with 0.19 mm single filament NbTi wire. The winding procedure was not as sophisticated as the second and final attempt. On occasion, the wire would
loop and if the loops were pulled tight by the rotation of the IVC, the loops became kinks. The coils did not survive the cool down to liquid nitrogen. It was observed that the wire was broken at the kinks.

The second attempt was successful. The superconducting wire is 0.33 mm, 54 filament NbTi wire. The coils are hand wound with assistance from a large lathe in the Duke University machine shop. The wires are covered several times with Stycast 1266 epoxy. A large volume of Stycast was used to ensure the Stycast would seep all the way into the middle of the windings. Also the wires tended to wick the Stycast into the center. The IVC was placed with the coils parallel to the ground, the epoxy was placed on the top side of the coil so that the epoxy would tend to run into the coils due to gravity and capillary action. After a day the IVC was flipped and Stycast was placed on the opposite side from the previous day. After 4 cycles of this process, the Stycast completely filled the volume between the wires minus a number of small bubbles.

Typically the epoxy is applied during the winding procedure. It was sufficient for our low fields however to apply after winding. Applying the Stycast afterward simplified the procedure, the difficulty of doing two precise procedures at once is less forgiving, and the time it takes to wind the coil is significantly longer than the time it takes the Stycast to become rigid. These difficulties were avoided and the coils worked without any problems.

The SCH and SCG are attached to the power supply using 12 gauge copper wire and extend to room temperature through a VCR feed-through on the exhaust outlet at the top of the apparatus. The wires were put into place by pulling them through the exhaust outlet with twine.

The SCH and SCG were used only during the relaxation process. They were not used during the acquisition of the FID signal. The room temperature coils (measurement coils) were used to supply the \( B_0 \) field during the acquisition. It is better to leave the room temperature measurement coils at a constant current throughout the duration of the measurement. This minimizes the inevitable frequency drift; the current will always drift a little, but more when the power supply is ramped away and back to the desired current. The set of superconducting Helmholtz coils are used to adjust the holding field during the relaxation period of the measurement; during the measurement, the SCH and SCG are disconnected by the Apollo through a voltage controlled relay rated for 25 A.

The AETechron has two outputs and is used as the current control for the SCH and SCG. Relaxation periods are not allowed to last longer than one minute. If the relaxation time went for much longer, the AETechron becomes unstable and transitions to a state where it drives a wildly varying current. This state is unacceptable and is avoided by limiting the relaxation periods to less than one minute. It is unknown why this behavior is observed. The author suspects that long pulses are not the intended purpose of the power supply. The AETechrons are designed to supply current for periods of milliseconds to a few seconds, not minutes. After a minute, the current would start to slowly drift away from the intended value and after a minute of drifting,
the current would suddenly become chaotic.

A given ratio of the current in the superconducting gradient coils with respect to the superconducting Helmholtz coils is forced due to undesired magnetization. For the correct ratio, the magnetic field in the region of the buffer volume is near zero, this prevents the measurement from having a short $T_2^*$, thus the signal remains optimized. This ratio was found by trial and error and is $G_{str} = 1.89$ $H_{str}$ on the MagTec NTNMR program.

### 4.5.3 Gradient Calibration

The gradient is calibrated in two ways. The first is through calculations/simulations of the configuration coils and a supplied current. The second is to measure the dephasing of the spins with a known current being sent through the coils. The data is fit to a sinc pulse and the gradient is extracted through the equation

$$\text{sinc} \left( \frac{\gamma G_z L t}{2} \right) = \pi,$$

(4.5)

where the calculation and simulation agree to below 1%, but the measured gradient differs by about 5% from the calculation. This is presumably due to the interaction with materials and fields that are not present in the simulation.

### 4.5.4 Alpha Calibration Method 1

For method one, a chain of medium loss pulses is applied to the spin with minimal time in between the pulses so that longitudinal relaxation is negligible. The signal strength is fit via

$$S(n) = b(1 - a)^{(n-1)},$$

(4.6)

where $n$ is the pulse number and $a, b$ are parameters to fit. This results in a tip angle found by

$$\theta = \arccos(1 - a).$$

(4.7)

In general, this method requires a lot of $^3\text{He}$, typically the full 69 cc of the transfer cell. The sample cell will have a concentration that is too high after the $\alpha$ pulse has been calibrated to be able to perform geometric phase measurements. The sample must be warmed to liquid nitrogen and replaced before a measurement where the newly calibrated pulse is used. It is possible to have systematics from deviations in the pulse angles between cool-downs. This was checked periodically and was found not to vary.
4.5.5 Alpha Calibration Method 2.

We describe the magnetization of a spin echo in order to derive an expression that can be used to calibrate an unknown $\alpha$ pulse. This method is best used if greater precession is required from the already known $\alpha$ from the Alpha Calibration Method 1. Angle calibration is from a single large alpha but $< \frac{\pi}{2}$ spin echo.

Imagine the magnetization after an unknown $\frac{\alpha}{2}$ pulse where $\alpha$ is close to $\pi$ (see for example Figure 4.3). The signal maximum would look like

$$S(t = 0) = S_0 \left( \cos \frac{\beta}{2} \right),$$  \hspace{1cm} (4.8)

where $S_0$ is the signal if $\alpha$ where a perfect $\pi$ pulse and $\beta$ is the difference from a $\pi$ pulse. The spins will decay according to $T_2^*$. If we then apply an $\alpha$ pulse in the $+y$ direction, looking down the $+y$ direction the orientation of the magnetization of the echo is shown in Figure 4.4. After applying this pulse, we observe the echoes signal maximum expressed by
\[ S(t = t_{\text{echo}}) = S_0 e^{-\frac{t}{T_2}} \left( \cos \frac{\beta}{2} \right) \left( 1 + \sin \frac{\beta}{2} (1 - \cos \beta) \right). \] (4.9)

Apply a second \( \alpha \) pulse along \(-y\) and our maximum signal corresponds to,

\[ S(t = t_{\text{echo}}) = S_0 e^{-\frac{t}{T_2}} \left( \cos \frac{\beta}{2} \right). \] (4.10)

Therefore every odd echo will have a maximum of

\[ S_1(t) = S_0 e^{-\frac{t}{T_2}} \left( \cos \frac{\beta}{2} \right) \left( 1 + \sin \frac{\beta}{2} (1 - \cos \beta) \right) \] (4.11)

and every even echo will have maximum of

\[ S_2(t) = S_0 e^{-\frac{t}{T_2}} \left( \cos \frac{\beta}{2} \right). \] (4.12)

We then fit even echoes and odd echoes as separate \( T_2 \) decays. The \( T_2 \)'s should be similar. In practice, they are \( \lesssim 1 \) ms different. However the coefficient multiplying the exponential decay should be noticeably different. The two coefficients can be compared to find \( \beta \), the error in the difference between \( \pi \) and \( \alpha \)

\[
\frac{S_2(0)}{\left( \cos \frac{\beta}{2} \right)} = \frac{S_1(0)}{\left( \cos \frac{\beta}{2} \right) \left( 1 + \sin \frac{\beta}{2} (1 - \cos \beta) \right)}, \quad (4.13)
\]

\[
\frac{S_1}{S_2} = \left( 1 + \sin \frac{\beta}{2} (1 - \cos \beta) \right). \quad (4.14)
\]

This expression has a complicated solution that a computer algebra program can solve. Maple for example, finds a solution, or it can be solved numerically using MATLAB. Here we use the small angle approximation,

\[
\beta = \left( 2 \frac{S_1}{S_2} - 1 \right)^{\frac{1}{2}}. \quad (4.15)
\]

The error of this method is propagated from the error of the fit. This method proved to be superior to method 1 for analyzing the pulse angle. This is attributed to the large angle used
and the number of data points acquired in one echo train. In the low temperature experiment, it was realized that the best way to calibrate an angle is to use the angle planned to be used in the measurement due to non-linearities. This is the only way to effectively calibrate a $\pi$ pulse at low temperature. The non-linearities were not present in the room temperature experiment.

4.5.6 1 K Pot Trouble

The capillary from the helium bath to the 1K pot clogs during the cool-down to liquid nitrogen. The clog was seen by over-pressuring the 1K pot and watching the different rates the pressure decreases at with the needle valve open versus closed. Typical pressure changes for restricted lines are 10 mbar in 1-2 min. starting at 1200 mbar of gas at liquid nitrogen temperature, while an open line has the same pressure change in only 20-50 seconds at the same temperature and starting pressure.

The clogging of the 1K pot could be avoided by attaching the vapor line of a liquid helium dewar to the 1K pot. The pressure was kept at a constant value using a check valve set between 1250-1350 mbar. The setting of the pressure required very fine adjustments to the check valve. The balancing of the pressure in the dewar and the pressure in the 1K pot was more of an art as it was somewhat unpredictable. For example, the check valve would need to be adjusted as the apparatus cooled to account for the change in density of the gas leaving the 1K pot. Liquid helium dewars do not have the same heat load to the liquid and would supply different pressures that varied differently. Every dewar required a new adjustment schedule to the check valve based on the observation of the 1K pot and helium dewar pressures.

4.5.7 Nitrogen in the Sample

After each measurement the lowest temperature that we can cool the sample to increases. This continues until it saturates at a temperature of 560 mK. At this temperature, letting in more of the sample will no longer increase the base temperature. For example, if we introduce 0.5 atm of nitrogen rich gas from the transfer cell into the sample, the temperature of the copper section connecting the acrylic sample to the glass tube increases from 433 mK to 465 mK. Introducing another 0.5 atm of gas will increase the sample temperature from 465 mK to 480 mK. As we continue to add to the sample the base temperature will continue to increase until it saturates at 560 mK.

The effect is independent of the level of the liquid helium increasing in the tube. Once saturated at a temperature at 560 mK, if we pump out the sample and warm to a temperature above 5 K and then refill the isotopically pure helium into the sample the lowest temperature of the sample will remain at 560 mK. However, if we let the apparatus warm to above liquid nitrogen temperature (77 K) and then cool again we reach a minimum of 430 mK.
The effect is likely due to a coating of condensed gas on the lower part of the glass tube. Condensed gases on otherwise clean flat surfaces are known to dramatically increase superfluid film flow [45]. Presumably, the condensed gas is nitrogen. Nitrogen is part of the SEOP as a buffer gas and present with a pressure ratio of 1:9 nitrogen to helium in the glass cell used to polarize the spins.

Prior to obtaining the lowest temperature, we thus had to warm to liquid above nitrogen temperature. This was accomplished by not refilling the helium and nitrogen baths and allow the entire system to warm. Once the cell reached a temperature above 77 K, (resistor R4 ≈ 600 Ω) the nitrogen will be melted and pumped away. One could observe the phase change by the time variation in the temperature rise. The apparatus was again cooled with the DR to get the sample to its lowest value of 430 mK. Multiple-measurement (several measurements with increasing concentration) could be performed. After repeated measurements were performed this warming process had to be repeated.

This process was successful for three times before a cryogenic leak opened up in the IVC. Repeating warming and cooling cycles was likely the culprit. In a future design, it would be beneficial to send the gas through a liquid helium trap prior to the measurement cell to remove the nitrogen. This would require significant engineering however since this part of the system is glass and the transport of the spin polarized $^3$He would be transported through cold to warm to cold sections.

4.5.8 Helium Trap Prevents $N_2$ Containments of Sample Gas

When condensing the liquid helium into the cells, the capillary condensing $^4$He into the capillary clogged. It was proposed that the isotropically pure $^4$He was contaminated with air. Shortly after, it was revealed that it is known to be contaminated with nitrogen. A liquid helium cold trap was designed and fabricated at the machine shop at Duke University. The trap consists of a pair of thin-walled stainless steel concentric tubes. At the top of the trap, the entrance to the tubes is fitted with Swagelock connections. At the lower end of trap, the outer tube is sealed and helium leak tight. The inner tube is left open and is suspended 0.6 cm from the bottom of the closed outer tube. This allows dirty gas to go down the inner tube to the cold part of the trap, and up and out the outer tube, condensing undesired gas on the walls of the tube. It was designed to be 1.22 m long, but in reality was only 1.14 m. This did not have a significant effect in the operation of the trap, but would decrease the time a single helium dewar would be effective since it did not reach the bottom. It was suggested to hard solder a copper braid to the end of one of the tubes to lower the temperature as the helium level in the dewar decreased, but the trap worked well and it was determined the braid is added risk with little benefit. The trap was placed after the needle valve and was monitored using a pressure gauge. The needle valve
gave control over pressure, preventing liquid helium build up in the trap.

The trap worked well. In addition it provided an added benefit in the cryopumping it provided to the system. Previous to installing the trap the apparatus required 30 min. to pump out a small volume of air that was inevitable when switching $^3\text{He}$ transfer cells onto the sample gas system. This delayed any measurement by 30 min. The cryopumping reduced this time to a few seconds. The cryopumping only works for air. If helium was being pumped out it extended the time to steady state because of the added length and decreased conductance in the pumping line.

After 3-4 days of trap use, the pressure in the sample pumping system would begin to slowly increase ($1 \times 10^{-7}$ mbar/hr, reaching a maximum pressure of $1 \times 10^{-6}$ mbar). The effect is attributed to the slow warming of the cold trap. The gas will tend to condense on the highest spot on the tube. As the liquid in the dewar boils off, the helium level decreases, and therefore the highest spot on the tube capable of condensing gas lowers. The gas that is condensed on the upper section boils off. There is still plenty of cold surface for the gas to condense, but the trap must be cleaned out. The trap is warmed to room temperature and pumped out. Raising the temperature causes the pressure in the system to skyrocket beyond the limit of the gauge, hence this process is started with the turbo pump off. Once the trap was pumped out to below $2 \times 10^{-7}$ mbar, it is placed into the helium dewar and works as usual. After about one month the near empty helium dewar must be replaced with a full helium dewar, it is assumed the lifetime of the dewar would increase had the trap been 1.22 m long as specified. The vapor pressure of nitrogen at 30 K is $1 \times 10^{-7}$ mbar, negligible for our purpose.

### 4.5.9 Pickup Coils

The first set of pickup coils were Helmholtz coils with a diameter of 15.24 cm. Each coil consisted of 2000 turns of AWG 30 copper wire. Its inductance was 2.39 H and its resistance is 660 $\Omega$ at room temperature and 20 $\Omega$ at 500 mK. Due to the large number of turns (a parallel circuit with high inductance and capacitance, $\omega_0 = (LC)^{-\frac{1}{2}}$) its resonant frequency is 6 kHz. The coils are driven by a current and the voltage is measured. The oscillating magnetic field from the precession of the spins generates a current in the coils. The voltage across the coils is measured using the Apollo and it its maximum occurs at $\omega_0 = (LC)^{-\frac{1}{2}} = 6$ kHz. The desired resonant frequency is around 36 kHz, near the maximum supplied field of the room temperature coils. The coils are tuned using an inductor circuit in parallel. The inductors used are coiled wire around a ferrous material. As such they produce a lot of noise. Several inductors were tried with varying results. To test the inductors a coil of 2.54 cm diameter with 10 turns was placed inside the Helmholtz coils to induce a signal. The signal to noise (S/N) ratio was reduced by a factor 0.8 with the inductor in place. The signal gained is proportional to the holding field, so in the
end, shifting the frequency up is a S/N gain of a factor 4.8.

In room temperature calibration measurements, it was observed that after a dilution by a factor of 1000, one could obtain a signal to noise ratio of 10. However the coils did not behave the same at low temperature. A signal from a sample with dilution factor of 1000 at low temperature was never observed. This can be explained by the presence of a barrier pressure in the tube, discussed in section 4.5.1, that prevented the rarefied sample from entering the tube. At the time of diagnosing, the effect of the barrier pressure to the sample was not known; it was believed to be a coil issue. There was a coil malfunction. An excessive leakage current was found and assumed to arise from magnetic interactions with the surrounding material. An op-amp circuit was designed to recover the lost leakage current. This had some success. The signal increased greatly, but the op-amp added noise to the system while there was only a slight S/N gain. A strange effect was however found during analysis of the op-amp and pickup coil circuit. Depending on which coil was placed closer to the op-amp in the circuit, the signal varied by a factor of 4, implying that a single coil was malfunctioning. The apparatus was allowed to warm. When the coils were separated and examined in detail, it was found that the impedance curves did not match. One coil observed the expected impedance, while the other was significantly shifted. Figure 4.5 shows the two impedance curves.

Instead of rewiring the faulty coil, a new four-coil design was fabricated as shown in Figure 4.6, with the aim of reducing the noise while shifting the resonant frequency to 36 kHz at the preamp.

Each coil was wound by hand with 500 turns. The windings of two coils on the left and right of the measurement cell in shown Figure 4.6 are reversed, this reduces background RF noise, and acquires additional signal due to the dipole field produced by the spins. The impedance of the pickup coils is using the circuit shown in Figure 4.7. The oscilloscope impedance is a constant 2 MΩ around the resonant frequency range. This leaves

$$Z(\omega) = \frac{V_2(\omega)}{I_{\text{pickup}}(\omega)}, \quad I_{\text{pickup}}(\omega) = \frac{V_1(\omega) - V_2(\omega)}{R} - \frac{V_2(\omega)}{Z_{\text{scope}}(\omega)}$$

$$Z(\omega) = \left[ \frac{V_1(\omega)}{V_2(\omega)} - 1 \right] - \frac{1}{Z_{\text{scope}}(\omega)} = \left[ \frac{V_1(\omega)}{V_2(\omega)} - 1 \right] - \frac{1}{Z_{\text{scope}}(\omega)}$$

After winding, the impedance of the coils were measured individually as shown in Figure 4.8.

The coils were modified to match the four impedances. Since the wire of the coils had already been cut, the coil with the highest frequency was not changed. The other three coils were unwound until all four resonant frequencies matched. The final frequency response of the coils is shown in Figure 4.9.
Figure 4.5: The impedance mismatch of the two Helmholtz coils. This mismatch resulted in a diminished signal.
Figure 4.6: Picture of the four pickup coils and RF coils in the apparatus.

Figure 4.7: Circuit diagram of the circuit used to measure the impedance of the Helmholtz coils and four-coil arrangement as described in the text.
Figure 4.8: Initial impedance of the four pickup coils as measured using the circuit in Figure 4.7.
Figure 4.9: Impedance of the four pickup coils after modification of the windings in order to match the impedances.
4.6 Minimization of Noise

The process to minimize the noise was extensive and occupied more than 6 months prior to cooling the apparatus and taking data. The initial experiment using this setup measured relaxation on the dPS-dTPB coatings that will be used for the nEDM experiment. The signal to noise of the apparatus was increased by a factor of 15,000.

The experimental conditions that enabled us to achieve this signal to noise were found through trial and error. They include double shielded coax cables for any signal-carrying wires and attaching the preamp as close to the feedthrough of the signal carrying wires as physically possible. A platform was built next to the feedthrough for this purpose. Shorter wires also provided a higher natural resonant frequency in the pickup coils. We found that altering the resonant frequency of the pickup coils with inductors or capacitors add a significant noise. Instead, one should wind the coils to the desired frequency. Op-amps used to eliminate leakage capacitance increases the signal but also increases the noise by more than the signal. The positioning of the coils is important and one can try different angles to reduce the noise picked up from the room. Ensure the different coil sets are as perpendicular as possible, minimizing the transfer of the RF signal to the pickup coil. Eliminate every ground loop possible. Some will always exist, for example the supporting structures made of metal. The optimum reduction in noise with the use of grounding wires and braids with contact points and order of contact to the apparatus and electronics devices was found by trial and error. The realization to use the natural resonant frequency of the coils was the largest step in the noise reduction. Proper grounding was the second largest step in the noise reduction.

4.6.1 Depolarization per Bounce Versus Spin Flip per Bounce

We have a system with two states that can be described by the diffusion equation. One system we describe as a combination of spin up, $\rho_+$, and spin down, $\rho_-$. The other as just the polarization density $\rho$, where $\rho = \rho_+ - \rho_-$. Here $\rho_0 = \rho_+ + \rho_-$ is the total density. The boundary condition is written as,

\[
\frac{d\rho_+(x,t)}{dr}|_{r=R} = -D \left( \alpha_+ \frac{d\rho_+(x,t)}{dr} - \alpha_- \frac{d\rho_-(x,t)}{dr} \right), \tag{4.17}
\]

\[
\frac{d\rho_-(x,t)}{dr}|_{r=R} = -D \left( \alpha_- \frac{d\rho_-(x,t)}{dr} - \alpha_+ \frac{d\rho_+(x,t)}{dr} \right), \tag{4.18}
\]

where now $\alpha_+$ and $\alpha_-$ are the transitions from a $\pm$ state to a $\mp$ state, respectively. In the assumption that these quantities are equal, (it would be interesting if they were not, as this would lead to free polarization), we can reduce this to
\[
\frac{d\rho(x,t)}{dr}\bigg|_{r=R} = \frac{d\rho_+(x,t)}{dr}\bigg|_{r=R} - \frac{d\rho_-(x,t)}{dr}\bigg|_{r=R}
\]
\[
\frac{d\rho(x,t)}{dr}\bigg|_{r=R} = -2\alpha D \left( \frac{d\rho_+(x,t)}{dr} - \frac{d\rho_-(x,t)}{dr} \right) = -D\beta \frac{d\rho(x,t)}{dr}\bigg|_{r=R},
\]
where $\beta = 2\alpha$, $\alpha = \frac{v}{4}K$, and $K$ is the flip probability per bounce. Hence the depolarization rate is twice the spin flip rate.

### 4.7 Summary of the Operation of the Experiment

The actual operation was considerably more complicated than the expected operation of the experiment. The cool down of the DR required constant attention due to the consistent formation of a clog in the capillary between the 1K pot and the helium bath. During a cool down if the line leading to the 1 K pot was not held above 1200 mbar with helium gas from a liquid helium dewar the 1 K pot capillary would clog.

The pickup coils used in the room temperature measurements were found to be malfunctioning after considerable characterization at low temperature. A superior four-coil design was constructed and successfully utilized.

The second version of the superconducting coils cooled down without malfunctioning. They provided a suitable magnetic field for the relaxation measurements.

The isotopically pure $^4\text{He}$ had nitrogen contaminants that clogged the transfer lines, prompting the construction of a liquid helium cold trap.

The dilution volumes worked as expected, however measuring the signal from a diluted sample was not achieved until 500 mbar of isotopically pure $^4\text{He}$ was introduced into the intermediate cell prior to transferring the rarefied polarized $^3\text{He}$ into the sample. This can be attributed to a barrier pressure due to film flowing from the sample past the restriction in the glass transfer tube.

Magnetization in the apparatus required extensive characterization. Its effect was minimized by ramping the SCG and SCH to specific values and times in the proper order. It could be neglected in the analysis of the measurements.

The noise in the signal was minimized by tuning the natural resonance of the pickup coils to the maximum suppliable field of the room temperature coils and by effectively grounding the apparatus while minimizing ground loops.
Data was collected and a significant amount was learned from analysis of the data during the operation of the experiment. This resulted in several successful runs that show good agreement with theory. Although the low temperatures required were not achievable in the apparatus to measure the ballistic transition, the measurement of the correlation function scaled by the quantity $\lambda/L \sim 0.03$ was made, the expected scaling parameter of the $^3$He in the nEDM experiment. This chapter considers the successful runs and summarizes how the data was analyzed and error propagated. Finally a comparison with theory is made.

5.1 Collection of Low Temperature Data

The experiment was operating at low temperature from November 2010 to June 2011. Various runs were performed at temperatures between 420 mK and 600 mK. We were not able to cool the apparatus below 430 mK with liquid condensed in the measurement cell due to superfluid film flow and heat loads from the additional magnetic package used to make the RF fields. This prevented measurements of the truly ballistic/transition regions which were desired. The temperature was not constant throughout the measurements due to varying film flow conditions transferring heat from the mixing chamber to the still. It is also believed that impurities were condensed on the wall of the fill tube each time that the valve is opened and helium was let in. This could be due to the nitrogen present in the mixture with the $^3$He necessary for SEOP as explained in the previous section, or it could from other contaminants. The inability to reach the desired temperature of 330 mK was reason to halt the experiment. Film pinners that are currently being fabricated at North Carolina State University (NCSU) and previously been used by NASA in the Astro-H satellite [49], may decrease or even stop the film flow and enable
measurements at lower temperatures.

A considerable amount was learned about the operation of this apparatus and the experiences learned from the operation should be a starting point in the future designs of the nEDM apparatus and the PULSTAR nEDM systematic studies apparatus. Furthermore, a scaled version of the conditions present in the $^3$He for the nEDM apparatus was measured, $\lambda/l \approx 0.03$, the ratio of the mean free path and the length of the dimension with the highest contribution to the geometric phase effect. This allows one to predict the correlation function at the same scaling ratio expected in the nEDM experiment. The mechanism that governs the mean free path for the two experiments is different and the angular dependence of the scattering may be different, resulting in a slightly different phase shift. In the nEDM experiment, the phonon scattering dominates the mean free path, where in this experiment, the scattering is dominated by $^3$He:$^3$He interactions.

Successful runs were obtained where the signal was observed for the duration of the scheduled measurement. They include gradient relaxation runs, wall relaxation runs and pulse calibration runs. Runs are presented in chronological order in Table A.1.

5.2 Systematics and Uncertainty

In this experiment, and in NMR in general, the number of particles that are being measured is enormous; $N \approx 10^{17}$ for the lowest concentration measured in our apparatus. Error due to counting statistics is negligible in this regime. The only uncertainty comes from the noise and precision in the electronics and the systematics of the experiment. The noise in the experiment was minimized with great effort and measured after each run to minimize the effect on the fitted decay.

The two largest systematics cannot be neglected and are present in the experiment. These are the relaxation from the wall and the loss in signal due to the tipping of the spins to measure the Free Induction Decay (FID). Another systematic includes diffusion of $^3$He up the transfer tube to higher relaxing materials. This process is suppressed by the binding energy of the $^3$He to the liquid. The level of the liquid can be well known by condensing a precise amount of $^4$He into the measurement cell. Another source of relaxation are spin-spin interactions. This process is known to be negligible for $^3$He with depleted density.

The tipping angle must be known precisely. In this experiment, a precision of $< 0.1\%$ is achieved and is sufficient. This is done by repeating the same pulse used in the actual experiment several times in a row in a short time period, 20 s, so that relaxation plays a negligible role in the reduction of the signal. The same square pulse is necessary due to the non-linearity observed in the tipping angle for square pulses of different length. The non-linearity is attributed to the
changing temperature due to heat from the current being carried in the coil. This temperature increase is also observed in the thermometers around the measurement cell and could be due to the interaction of the RF pulse and the electronics in the read out. In any case the time spent at this increased temperature is extremely short (< 0.5 s) and will play a negligible role in changing the decay rate if it exists at all. Square pulses of the same length are observed to be consistent over different runs. On the assumption that the signal decay is negligible during the pulse loss calibration measurement, which it is, the uncertainty in the signal loss from the spin tip arises purely from electronic noise.

Once the tipping angle for pulses used in the experiment are known the wall relaxation can be measured by using the spin relaxation in a uniform holding field. An uncertainty of 0.2% for the wall relaxation is achieved in this experiment. This uncertainty is due to the noise in the signal and the propagation of uncertainty in the signal loss measurement. The change in the decay rate from the two systematic errors can be subtracted and the errors propagated to achieve a final uncertainty in the gradient relaxation measurement.

The noise recorded by the Apollo system is shown to be white noise in the range of the observed spectrum. Combining several runs with consistent length and dwell time can increase the statistics of the noise. This exercise was done for 350 data sets with the same width of integration. A histogram is shown in Figure 5.1, the curve fit to a Gaussian distribution is quite good.

The integral of the real part of the signal was done over 50 Hz at the far end of the decay curve so that no signal was present. It was observed that the noise did not vary from day to day. The signal to noise ratio did change from one run to the next based on the quality of the polarization cell being used in the SEOP and the $T_2$ decay which is extremely sensitive to the background gradients. The background gradients varied slightly from run to run due to the small magnetization near the cell, but would typically remain constant during a single run. If the magnetization changed significantly during a run, the data was excluded.

With the assumption that the noise is white, then it follows from reference [29] chapter 9 that

$$\sigma_{total} \propto \frac{1}{\sqrt{n}} \quad (5.1)$$

where $n$ is the number of runs with the same conditions. It is not worth the cost of $^3$He and liquid $^4$He to redo measurements to reduce uncertainty. As discussed earlier, the temperature of the previous experiment of 330 mK was not achieved in this experiment. According to theory, 330 mK and the uncertainty achieved is sufficient to observe the divergence of the exact and high frequency theories. At 420 mK however, the reduction of the uncertainty is on the order of
Figure 5.1: Noise histogram with 25 bins consisting of 350 Fourier transform integrations of around 50 Hz width centered on the observation frequency. The signal for the lowest concentration measured is a factor of 100 times larger than the standard deviation of the noise. The red curve is a fit to a Gaussian distribution with the same standard deviation. Good agreement is seen. All exponential decay fits have reduced $\chi^2$ close to one.
Figure 5.2: Run 693 wall relaxation calibration, $T_1 = (8988.9 \pm 18.6)$ s. The reduced $\chi^2$ for this run is 1.00, as it is with all acceptable runs due to the only decay being exponential and very white noise.

20 times thus 400 measurements would need be needed in order to observe this divergence. This is unfeasible in our present setup.

### 5.3 Fitting the Decay

The transformed real signal is integrated with a consistent bandwidth to determine the signal strength of each FID spectrum per run. Every FID spectrum is phased so that the maximum real signal is achieved for every signal’s spectrum. This is done using the Apollo. Typically the automatic phasing algorithm is suitable, however for low signals it is best done manually.

The fast Fourier transform (FFT) phased real integrated signal is the best method to find the decay curve. Previously the decay was found using the maximum point of the magnitude of the spectrum. The frequency range of the integration was constant throughout the measurement.
This gave the most accurate measurement of the noise of the system. The noise is the uncertainty in the measurement and in the fit. The fitting algorithm minimizes the uncertainty when the fitting expression is simplest. The fitting expression is

\[ f = ae^{-\frac{x}{b}}, \quad (5.2) \]

where \( f \) is the function being fit, \( a \) is the \( t = 0 \) value, \( b \) is the decay time and \( x \) is the time of the data point. The background is white noise which we have shown to average to zero during the independent background measurements done at the end of each run. A previous method of finding \( T_1 \) was to evaluate a single point of the magnitude of the FFT signal. This does require a background constant in the fit and should be the average of the noise magnitude. The average of the magnitude is used in the estimation of the uncertainty with the real integration method however it is incorrect to be included in the fit. In this case

\[ f = ae^{-\frac{x}{b}} + c \quad (5.3) \]

where \( c \) is some constant. Including this constant artificially raises the uncertainty in the relaxation rate because the algorithm “double counts” the noise in the signal and the background. Using the real integration method, the noise in the signal, the only source for background, will average to zero.

In previous fits, the systematic error of the pulse loss was taken into account in the fit via the fitting function

\[ f = a(1 - \text{loss})^n e^{-\frac{x}{b}}. \quad (5.4) \]

However it was observed that this fit has more uncertainty than the simpler fitting equation because it artificially increases the number of parameters needed to fit the decay signal. The time between pulses remains the same, so the signal will obey an exponential with a single time constant. It is incorrect to include unnecessary complexity in the fitting function.

To keep the good statistics of the simple fit, it is best to account for the systematic error of the pulse loss after the fit. Although the loss is discrete in time, we write the pulse loss as its equivalent relaxation rate. Physically for this measurement, it is equivalent as it is required to give a pulse exactly before a measurement and the time between pulses remains constant during the run. It would not be true if a more complicated pulse sequence is required (e.g. an alpha pulse spin echo), or if the time between pulses changed. The pulse rate is given as
where $\tau(\alpha)$ is the length of the applied pulse and $\tau(\frac{\pi}{2})$ is the length a $\frac{\pi}{2}$ pulse. $\Delta t$ is the time between measurements, and must remain constant during a run.

The experiment did not remain at a consistent temperature from run to run. The only effect this will have on the systematics will be to change the velocity of the $^3$He, increasing the relaxation rate. The relaxation rate that is measured can be scaled using the temperature and thus the increase in uncertainty can be neglected,

$$\frac{1}{T_{1\text{wall New}}} = \frac{1}{T_{1\text{wall Old}}} \sqrt{\frac{T_{\text{Old}}}{T_{\text{New}}}}. \quad \text{(5.6)}$$

To calculate the gradient relaxation rate, one combines the relaxation rates from the individual components,

$$\frac{1}{T_{1\text{total}}} = \frac{1}{T_{1\text{wall}}} + \frac{1}{T_{1\text{Pulse}}} + \frac{1}{T_{1\text{Gradient}}}. \quad \text{(5.7)}$$

### 5.4 Propagation of Uncertainty

The uncertainty in the fit to the relaxation time is found from the root mean square deviation $\text{RMSD}$ (or root mean square error) by

$$\sigma_{T_1} = \frac{\text{RMSD}}{f(0)} b = \frac{\text{RMSD}}{a} b, \quad \text{(5.8)}$$

where $a$ and $b$ are from the fitting function in Equation 5.2. To propagate the uncertainty of the wall and the total relaxation time into the gradient relaxation time, we first write the total rate.

Using Equation 5.7,

$$f = e^{-(\frac{t}{T_{1\text{wall}}} + \frac{t}{T_{1\text{Pulse}}} + \frac{t}{T_{1\text{Gradient}}})} \quad \text{(5.9)}$$
where \( f \) can be written as

\[
f = f_A \times f_B \times f_C
\] (5.10)

where \( f_C \) is the unknown decay,

\[
f_C = \frac{f}{f_A \times f_B}. \tag{5.11}
\]

For this case, the uncertainty is propagated as

\[
\left( \frac{\sigma f_C}{f_C} \right)^2 = \left( \frac{\sigma f}{f} \right)^2 + \left( \frac{\sigma f_A}{f_A} \right)^2 + \left( \frac{\sigma f_B}{f_B} \right)^2. \tag{5.12}
\]

To find the uncertainty in the relaxation rate we write the uncertainty in \( f_A, f_B, f_C \) as

\[
f_C = e^{-\frac{t}{T_{1\text{Gradient}}}},
\]

\[
\frac{\sigma f_C}{f_C} = \frac{\sigma T_{1\text{Gradient}}}{T_{1\text{Gradient}}}. \tag{5.13}
\]

Or, a similar expression for all factors,

\[
\left( \frac{\sigma T_{1\text{Gradient}}}{T_{1\text{Gradient}}} \right)^2 = \left( \frac{\sigma T_{1\text{Total}}}{T_{1\text{Total}}} \right)^2 + \left( \frac{\sigma \text{Pulse}}{T_{1\text{Pulse}}} \right)^2 + \left( \frac{\sigma T_{1\text{Wall}}}{T_{1\text{Wall}}} \right)^2. \tag{5.15}
\]

This is the equation used to solve for the uncertainty in the gradient relaxation rate. It should be noted that the pulse loss is no longer a systematic error in this equation, but the statistical error in the systematic correction since it is statistical in nature. It arises from the uncertainty in the calculated \( \alpha \) pulse. However due to the high precision of the \( \alpha \) pulse, this term contributes little to the total uncertainty.

### 5.4.1 Results

Points of varying \(^3\text{He}\) concentrations were measured for two holding fields successfully with confidence of the correct concentration. Holding fields of 40 G and 50 G were used. Agreement is found for the 50 G case for the temperatures given by the thermometers. The data is plotted in Figure 5.3. The 40 G case however does not fit accurately with the temperature given by the thermometer on the cell 568 mK or wire 698 mK. A good fit is found with a temperature of 460mK as seen in Figure 5.4. There is reason to believe that the temperatures for this run are inaccurate due to condensed gases on the tube. The fill capillary leading the to the cell was clogged and the glass tube was nearly clogged during these measurements. This will be discussed further in Section 5.4.2.
Figure 5.3: Measurements of gradient relaxation with varying molar concentration of $^3$He : $^4$He at 50 G and 2.5 G/cm. The purple curve shows the exact 3D theory for a temperature of 430 mK, the temperature given by the thermometer during these runs. The blue curve is the high frequency limit of the same temperature. The fit to the 3D exact theory yields $\chi^2_\nu = 0.88$, the fit to the high frequency theory yields $\chi^2_\nu = 2.46$
Figure 5.4: Measurements of gradient relaxation with varying molar concentration of $^3\text{He} : ^4\text{He}$ at 40 G and 1.89 G/cm. The green curve corresponds to a temperature of 568 mK, the temperature given by the thermometer on the acrylic cell. The temperature on the copper, 698 mK, is not considered in this figure. It is believed that the thermometer was insulated by condensed gases and displayed a temperature warmer than the cell temperature. The magenta curve is calculated for a temperature of 460 mK. It would seem this more accurately represents the actual temperature of the polarized $^3\text{He}$. The measured temperatures were consistent for each data point.
The first data point in Figure 5.3 corresponds to a $\lambda/L = 0.040$. In the nEDM experiment, the scaled length with the largest contribution to the geometric phase is $\lambda/L = 0.029$, which is further toward the diffusive region. In the assumption that the $^3$He-phonon scattering is isotropic, we can claim good predictive power of the theory. This is based on the agreement with theory and data from runs 769 to 771, and that the theories of relaxation and phase shifts are derived from the same correlation function.

5.4.2 Condensed Gases

One cannot be certain why there is a changing temperature throughout the experiment. It is the author’s belief that it is due to condensed gas, either nitrogen from the buffer gas of the SEOP polarization procedure or air getting into the pipes from a leak in the large dilution volumes or connections to those volumes. The large volumes were repeatedly leak tested with no leak present to the sensitivity of a helium leak detector. That source is unlikely, however due to the large impedance from the long pipes to the leak detector, it can’t be completely ruled out.

Condensed gases can considerably increase film flow [45]. This is believed to be responsible for the increased temperature after each run. To estimate the amount of solid arising from the condensed gases in the polarization cell we estimate that there is 100 mbar of $N_2$ gas in a 68 cc SEOP polarization/transfer cell. In run 692 the initial temperature before the gas was introduced was 430 mK. We estimate that approximately 50 mbar of $N_2$ in the 68 cc transfer cell volume, or 3.4 mg, was let into the measurement cell. As a solid this would occupy a volume $3.4 \times 10^{-3}$ cc assuming a density of 1 g/cc. This is enough to produce a rough coating on the surface of the glass, which would increase the film flow and thus increase the temperature to 450 mK as observed. With consecutive runs, the surface roughness would increase until a maximum is achieved and the temperature is stable from run to run, as observed in Table A.1.

The measurement cell can be emptied and be warmed to 10 K. When cooled down again, the empty cell temperature will reach a temperature of 280 mK. When the measurement cell is filled with liquid, its temperature will increase back to the temperature during the previous run. This was observed several times and an example is seen in the Table A.1 between run 726 and 728. The only way the original 430 mK temperature can be achieved again is by raising the temperature of the measurement cell to above 100 K. This again is consistent with a condensed gas such as nitrogen causing increased film flow. The largest known source of condensed gas is coming from the nitrogen in the measurement cell. This is also consistent with the small increases in temperature of the when a smaller amount 3.4$\mu$g is let in. An example is seen in Table A.1 between runs 769 to 771.

The anomalous temperatures between runs 760 and 764 are difficult to account for. The isotopically pure $^4$He from the McClintock purifier was used. This was later confirmed to be
contaminated with nitrogen. The capillary used to fill the measurement cell clogged during the filling process. With much effort, the remaining $^4\text{He}$ was condensed through the glass tube also used to transfer the polarized $^3\text{He}$. The temperatures seen represent a gradient across the cell of 126 mK at a temperature of 500 mK. It is hard to understand a temperature gradient of that magnitude in a measurement cell filled with superfluid helium. It is likely the condensed gas was altering the measurement of the temperature in the sample. Furthermore, the measured relaxation curve as a function of temperature shown in Figure 5.4 appears to disagree with both temperatures of the thermometers. It is likely that the temperature that the thermometers were reading was incorrect, however the actual temperature is unknown. The relaxation data is fit best to a temperature of 460 mK.

The helium in the measurement cell was pumped out after run 764 without trouble. When attempting to condense isotopically pure $^4\text{He}$ to the measurement cell again through the glass tube, it clogged. The apparatus was warmed to 100 K. When it was cooled again, it was seen that the original temperature of 430 mK was achieved with the measurement cell full of isotopically pure $^4\text{He}$. The cell unclogged by warming the helium in the measurement cell and pumping on the clogged tube. It cleared without exploding the tube itself or the measurement cell.

5.4.3 $^3\text{He}$ in Research Grade Helium Bottles

Research grade helium gas is alleged to have isotopic impurities on the order of $1.6 \times 10^{-6}$. In the beginning of the experiment, it was assumed to be true. However, the data fit to the theory with higher concentrations of $^3\text{He}$, consistent with the atmospheric abundance, $X = 1.3 \times 10^{-4}$. In the region where the theory is linearly increasing, this is the motivation for switching to the isotopically $^4\text{He}$. After the switch, the data fit the theory for the expected concentrations. Comparisons with the concentrations expected versus the best fit with the parameters suggest a starting concentration of $X = 1.25 \times 10^{-4}$. These fits are shown in Figures 5.5 and 5.6.

5.5 Summary of the Results

The gradient-induced $T_1$ was measured in the temperature range of $T = 430 \text{ mK} \rightarrow 560 \text{ mK}$, in magnetic fields from $B_0 = 11.5 \text{ G} \rightarrow 50 \text{ G}$, and in concentrations of $X = 1 \times 10^{-6} \rightarrow 3.3 \times 10^{-3}$, requiring a S/N a factor 15,000 larger than the previous iteration of the experiment. The analysis of the data shows that the theory presented in Equations 2.18 and 2.88 adequately predicts the data. Previous theories exist, found in Equation 2.96, that are adequate to describe the data in this region. The presented theory is shown to agree with previous theories limited to this region theory and diverge with slightly more ballistic motion than the experiment was able to achieve. Diffusion theory, found in Equation 2.97, is shown to be inadequate to describe the data for the
Figure 5.5: Measurements of the gradient relaxation with varying concentration at $B_0 = 50$ G and $2.56 \text{ G/cm}$. The blue curve is the theory. The red error bars are the data with the expected concentration. The purple is the data shifted to a concentration of $1.25 \times 10^{-4}$. 
Figure 5.6: Measurements of the gradient relaxation with varying concentration at $B_0 = 30$ G and $1.23 \text{ G/cm}$. The blue curve is the theory. The red error bars are the data with the expected concentration. The purple is the data shifted to a concentration of $1.25 \times 10^{-4}$. 
longer scattering times measured. The measurements and all presented theories agree for the shorter scattering times. Furthermore, the expected operating rage in the nEDM apparatus of the parameter $\lambda/L \approx 0.029$ was measured and shown to agree with both valid theories in that region.

The $^3\text{He}$ concentration in the research grade bottle initially used in the experiment is shown to have an isotopic impurity of $X = 1.25 \times 10^{-4}$. This is higher than expected, although the isotopic impurity is known to vary with the source of the helium.
CHAPTER 6

PIONEERING THE HEAT FLUSH, $^3$HE TRANSPORT IN PHONON DOMINATED HELIUM II

$^3$He transport is a major component of the operation of the nEDM apparatus. $^3$He of very high polarization, $\sim 99\%$, is required for the desired sensitivity. In this chapter, a description of the model used to simulate the heat flush is described. Due to its effectiveness found from this work, the heat flush is the expected method for transporting the $^3$He in the nEDM experiment. The model is used to find the optimum parameters, and ultimately a solution is found to satisfy the $\sim 99\%$ polarization requirement. The problem of quantum vortices is also considered as it may hinder the effectiveness of the flush.

$^3$He transport is modeled from the compilation of two governing equations. The phonon gas density is governed by the temperature. The Navier-Stokes describes the normal fluid, while the convection-diffusion equation describes the $^3$He transport. The Navier-Stokes equation governs the temperature/pressure gradients in the geometry based on boundary conditions which include temperature (via pressure), heat flux (via velocity) and wall velocity (simulating a slip). The convection-diffusion equation utilizes the velocity field from the Navier-Stokes solution. Conceptually this is the phonon wind that drags the $^3$He. Theoretically the diffusion coefficient is derived from the scattering amplitude of the $^3$He on phonons. It has also been experimentally measured to good agreement with theory [35]. The experimental value is used in the model.

6.1 Modeling the Phonon Wind

The equation of motion for the normal fluid is originally derived by Landau and given by [45] on page 56. This is the Navier-Stokes equation for an incompressible liquid,
\( \rho_n \left( \frac{\partial v_n}{\partial t} + v_n \cdot \nabla v_n \right) = -\nabla p_n + \eta_n \nabla^2 v_n. \) (6.1)

where \( \eta \) is the viscosity and is defined from the phonon density \( \rho_n \), thermal speed \( u \), and the phonon-phonon mean free path \( \lambda \). Parameters used in this calculation are

\[
\eta = \frac{\lambda v}{3} \quad (6.2)
\]
\[
\rho_n = \frac{2\pi^2 k_B^4}{45h v_T^5} T^4 \quad (6.3)
\]
\[
u = \frac{24000 \text{ cm}}{s} \quad (6.4)
\]
\[
\lambda = (1.28 \times 10^{-5} T^{-9} + 3.80 \times 10^{-3} T^{-4}) \quad \text{[cm]}. \quad (6.5)
\]

The phonon speed is dependent on the slope of the dispersion curve and is constant in the temperature ranges considered. The mean free path was found in Greywall [50]. Collaborators used different values of mean free path, Maris [51] and Whitworth [52] and found similar results as those shown in Figure 6.1.

The heat flush occurs with constant volume and mass in the system. The dependent variable representing pressure can be related to the temperature for small temperature differences. This effect is known as the fountain pressure [45],

\[
dp = SdT \quad (6.6)
\]
\[
S_n(T) = \frac{2\pi^2 k_B^4}{45h v_T^5} T^3 \quad (6.7)
\]
\[
p(T) = \frac{2\pi^2 k_B^4}{180h v_T^5} T^4, \quad (6.8)
\]

where \( k_B \) is Boltzmann’s constant and \( v_T \) is the thermal average speed of the phonons. The dependent variable representing the velocity field is interpreted as a heat flux. The heat flux is used as a boundary condition and is measured from the model by the dependent variables according to the equation

\[
dQ = ST v_\perp dA = 4pv_\perp dA \quad (6.9)
\]
\[
v_\perp = \frac{dQ}{dt} \frac{1}{STA}. \quad (6.10)
\]
Phonon–Phonon Mean Free Paths from Three Different References

Figure 6.1: A plot of the different expressions that represent the phonon-phonon mean free path as a function of temperature used by different collaborators [50,51,52]. The spread in the values of the mean free path give the final optimum geometry more robustness.
6.1.1 Boundary Conditions

The temperature and heat flux at the boundary are converted to the model dependent variables by the same equations that they are determined by the bulk. Temperature and heat flux boundary conditions are the major contributing boundary conditions in determining the flow of phonons, the temperature is converted to a pressure and the heat flux converted to velocity field by the expressions given. Other boundaries conditions include a velocity slip on the walls and heat exiting the pipes and cell walls submerged in liquid helium.

6.1.2 Velocity Slip on the Walls

For dilute gases, the Naiver-Stokes produces a more accurate result with a velocity slip on the walls [50]. This is found by solving the Boltzmann equation for a boundary layer on the wall and taking the value of the boundary layer as the boundary for the Navier-Stokes equation. The slip is determined by the slope of the curve normal to the boundary. In the first order approximation, it is found that

\[ v_{\text{slip}} = -\eta \lambda \frac{dv}{dn} \]  

(6.11)

where \( \lambda \) is the mean free path of the phonons, and \( \frac{dv}{dn} \) is the slope normal to the boundary. \( \eta \) is the coefficient that determines the magnitude of the slip. Differences in this coefficient are prescribed by the roughness and material of the walls. The value of \( \eta = 8/15 \) was used in the model. This is the measured value given in reference [50]. However in that experiment the tubes were composed of copper that is sand blasted for artificial roughness. The actual value of the slip coefficient of the pipes is expected to be larger, which will increase the efficiency of the flush.

6.1.3 Heat Flux Through Acrylic

The measurement cells and sections of the pipes are submerged in liquid helium. They will leak heat through the acrylic walls. In the measurement cells, this effect is desired and is the heat sink/source used at the respective stages of the flush. However heat leaking through sections of a tube submerged in liquid helium is not desired, but must be included in the model. The boundary condition that describes this effect is given as

\[ \frac{dQ}{dt} = -KL\nabla T \]  

(6.12)

\[ v_{\perp} = -\frac{KL\nabla T}{4p} \]  

(6.13)
where the thermal conductivity of acrylic is given as $K = 4000T^{-2}$ erg/cmK [53]. The temperature difference driving the flow of heat, $\nabla T$, is taken as the temperature from the bath to the temperature inside the pipes on the boundary layer.

### 6.2 Convection-Diffusion

When the model for the Navier-Stokes has converged, the velocity field is used as the independent velocity field in the convection-diffusion equation,

$$\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = \nabla \cdot (D \nabla c),$$

where $c$ is the concentration of $^3$He, $D$ is the diffusion coefficient of $^3$He due to phonons, and $\mathbf{u}$ is the velocity field which drags the concentration of the $^3$He through interactions accounted for in the diffusion coefficient. The diffusion coefficient as a function of temperature was measured to be $D = 1.6T^{-7}$ [35]. In the concentrations that are considered for the experiment, the self diffusion is nearly infinity. Diffusion from phonons is dominant. The self diffusion is irrelevant in mass transport due to detailed balance or conservation of momentum [35]. For spin transport in a magnetic field, the self diffusion becomes relevant if it is the dominant term.

Spin relaxation from the wall depolarization can be simulated in this model with the addition of a dependent concentration that represents the depolarized $^3$He. The bulk equation is the same as equation 6.14. The boundary interacts with the $^3$He by the boundary conditions

$$D \frac{dc_+(\mathbf{x}, t)}{dr}|_{r=R} = -\alpha (c_+ - c_-),$$

$$D \frac{dc_-(\mathbf{x}, t)}{dr}|_{r=R} = -\alpha (c_- - c_+),$$

where $\alpha$ is the depolarization coefficient, the rate per surface area of the polarized atoms striking the wall times the probability per bounce of a spin flip. A previous work [43] measured the depolarization per bounce ($DP$) of the coating planned for use as $DP = 1 \times 10^{-7}$. As seen in Section 4.6.1 and Equation 4.20, $DP$ is related to $\alpha$ by

$$\alpha = \frac{v}{4} 2DP$$

(6.15)

The $2 \times DP$ is due to differences in the models. In the experiment [43], $DP$ was calculated from
a model of the polarization \( P = \frac{c_+ - c_-}{c_+ + c_-} \). The polarization was proportional to the magnetization, and in the model wall relaxation it is equivalent to absorption on the walls. The number density of \(^3\)He is lost in this model. In the FEM model it is necessary to keep track of both the up and the down state simultaneously so that the number density of \(^3\)He is known.

There is one additional boundary condition that represents the transfer from a surface to the vapor of \(^3\)He. The rate of transfer per surface area of the liquid to vapor boundary is,

\[
\frac{dc}{dt} = -\frac{v}{4} \left( \frac{m^*_{^3\text{He}}}{m_{^3\text{He}}} \right)^{\frac{3}{2}} e^{-\frac{E_b}{k_B T}}
\]

where \( v \) is the thermal speed in the gas, \( m^*_{^3\text{He}} \) is the effective mass in the superfluid, \( k_B \) is Boltzmann’s constant, \( T \) is the temperature in Kelvin and \( E_b \) is the binding energy to the liquid and is 2.8 K. It is assumed that a perfectly adsorbing charcoal adsorber is in the vapor above the liquid level. If \(^3\)He is evaporated from the surface it always goes to the charcoal where it is adsorbed. No return flux of \(^3\)He at the evaporation surface is included in the model.

### 6.2.1 Combining the Models

There are two methods of combining the models. The fastest way is to solve the steady state Navier-Stokes equation and use the solution for the velocity field in the time dependent convection-diffusion equation. This is the best way to examine a large parameter space with minimal computation time. The most accurate way is to solve the time dependent Navier-Stokes stokes in conjunction with the convection-diffusion. The convection-diffusion model is dependent on the velocity field found by the Navier-Stokes, so they are both solved for in a time long enough for the \(^3\)He to achieve steady state in the convention-diffusion model. Steady state for the Navier-Stokes occurs in a matter of 30-50 s seen in Figure 6.2, but steady state for the concentration gradient is on the order of 70 to 300 s depending on the phase of the flush. The difference between solving the steady state of the Navier-Stokes versus the time dependent model with the convection-diffusion turns out to be very small.

The Navier-Stokes is solved without consideration for the \(^3\)He. The \(^3\)He will transport entropy during the flush. Due to the low concentrations considered this effect is negligible. The parameter space is varied to find the optimum geometry limiting time and heat flux, the search in the parameter space was done by brute force and controlled by a MATLAB script.
Figure 6.2: Velocity of the phonon wind in the center of the pipe 50 cm from the entrance to the cell. The time lag is due to the time it takes the wind to reach the particular point.
6.3 $^3$He Transport in the nEDM Apparatus

The $^3$He transport consists of three phases, flushing the polarized $^3$He in from the atomic beam source into an intermediate volume, flushing the polarized $^3$He from the intermediate volume into the measurement cells, and flushing the depolarized $^3$He out of the experiment and into a purifier.

The details of the atomic beam source are outside the scope of this document. It is assumed that the $^3$He enter the injection volume (INJ) at a rate of $10^{141}/s$. The INJ is held at a temperature of 350 mK optimized for absorbing $^3$He in the superfluid with low vapor scattering.

In order to keep the INJ at a constant temperature, an intermediate volume (IV1) is placed close to the INJ. This enables the INJ to continue to accumulate $^3$He without interruption from changing temperatures necessary to establish transport. The mechanism which brings the $^3$He into IV1 is diffusion. IV1 is therefore around four times bigger than the INJ such that 4/5 of the $^3$He that is present inside the INJ transfers to IV1. The time it takes to obtain a uniform distribution of $^3$He in the INJ and IV1 is less than 10 s.

IV1 is then heated 130 mK from 350 mK to 480 mK. This requires a valve connecting IV1 to the INJ with very low thermal conductivity as well as a valve from IV1 to the transfer tube. The $^3$He is then flushed from IV1 to the measuring cells (MCELL). After the measurement is made, $^3$He is then flushed to the second intermediate volume (IV2). A schematic diagram is shown in Figure 6.3.

The injection cell is assumed to have a volume of 392.8 cc, while IV1 has a volume of 1570 cc. The initial concentration in the simulated IV1 is 1 mol/cc. If the residual $^3$He from prior flushes left in the pipes and volumes is assumed to have a polarization of zero, then the pipe from the IV1 to the MCELL must be emptied below 0.01% prior to filling the MCELL, and the MCELL must be lowered to at least 0.3% prior to being filled. These strict emptying fractions are necessary to reach a polarization of 99% in the MCELL at the start of the measurement or the remnant depolarized fraction will begin to accumulate and 99% polarization will not be obtainable. With this requirement, a secondary flush of the pipe is unavoidable. This can be achieved by the addition of a valve so a flush from IV1 to IV2 can occur during a measurement.

6.3.1 Maximization of Polarized $^3$He in the Measurement Cells

The first phase of the heat flush can be completed with 5 mW. The parameters are given in Table 6.1 and are determined to be optimum with the restriction of 5 mW of cooling power. The subdomain of the model is shown in Figure 6.4 with the color representing the polarized concentration distribution, more precisely the quantity $N_p/Volume$. A plot of the volume average polarization in the measurement cells is given in Figure 6.5.
Figure 6.3: Schematic diagram of the $^3$He transport in the nEDM apparatus.

Table 6.1: Optimized values for the heat flush from IV1 to the measurement cells.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Optimized Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pipe Length [cm]</td>
<td>450</td>
</tr>
<tr>
<td>Pipe Radius [cm]</td>
<td>1.3</td>
</tr>
<tr>
<td>Dependent Conditions</td>
<td>Dependent Variable’s Value</td>
</tr>
<tr>
<td>$v_{\text{max}}$ [cm/s]</td>
<td>44</td>
</tr>
<tr>
<td>Temperature [K]</td>
<td>Cell 0.463, IV1 0.500</td>
</tr>
<tr>
<td>$\frac{N}{N_{\text{Total}}}$ in MCELL after 35 s</td>
<td>0.760</td>
</tr>
<tr>
<td>Polarization in MCELL after 35 s</td>
<td>99.13%</td>
</tr>
<tr>
<td>$\frac{N}{N_{\text{Total}}}$ in MCELL after 66 s</td>
<td>0.973</td>
</tr>
<tr>
<td>Polarization in MCELL after 66 s</td>
<td>99.01%</td>
</tr>
</tbody>
</table>
Figure 6.4: Distribution of the desired spin state of $^3$He in the model after 50 seconds of flush with the optimum parameters given in Table 6.1. The quantity plotted is $\frac{N_T}{\text{Volume}}$. 
Figure 6.5: The volume average polarization in the measurement cells. The initial increase is due to the new polarized $^3\text{He}$ entering the cell from the IV1. The polarization begins to decrease as the $^3\text{He}$ that has spent more time in the transport tubes begins to accumulate. While the helium is in the tubes, the effective surface to volume ratio is larger, thus decreasing the relaxation time.
Figure 6.6: The volume average concentration for flushing the $^3$He from the measurement cell is shown to decrease to acceptable values after 250 s. Notice that the integral does not start at one, this is due to the inclusion of the pipe void of $^3$He up to the valve. Still, the volume average is below 0.3%, the value used as the remnant $^3$He in the flush from IV1 to the measurement cell, so this process is repeatable without buildup of depolarized $^3$He.
Table 6.2: Optimized values for flush out of measurement cell to IV2

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Optimized Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pipe Length Total, First section, Second Section [cm]</td>
<td>450, 110, 340</td>
</tr>
<tr>
<td>Pipe Radius, First section, Second Section [cm]</td>
<td>1.3, 1.85</td>
</tr>
<tr>
<td>Dependent Conditions</td>
<td>Dependent Variable’s Value</td>
</tr>
<tr>
<td>( v_{\text{max}} ) [cm/s]</td>
<td>63</td>
</tr>
<tr>
<td>Temp [K]</td>
<td>Cell 0.45, IV2 0.432</td>
</tr>
<tr>
<td>( N_{\text{Total}} &lt; 0.3% ) in MCELL [s]</td>
<td>285</td>
</tr>
</tbody>
</table>

6.3.2 Measurement Cells to Intermediate Volume 2

After the measurement in the cell is completed, a portion of the \(^3\text{He}\) will have depolarized. This depolarized \(^3\text{He}\) must be flushed away. However we cannot flush only the depolarized fraction so we flush all of the \(^3\text{He}\). Theoretically it possible to raise the B-field so high that it strongly affects the motion of the helium and force a gradient so that the low field seekers tend to exit the cell. Although this would clean the cell of depolarized \(^3\text{He}\), it would not introduce new \(^3\text{He}\), but this could be flushed in. In reality it is an awful idea to raise a B-Field so high in a measurement designed to detect nano-hertz scale Larmor frequency shifts. A flush of all \(^3\text{He}\) away from the measurement cell is thus required. The measurement cell cannot be made to go much below 450 mK due to the large volume of liquid, a large volume the IV2 must be lowered in temperature so that the helium will be flushed towards that volume.

A volume about twice the size of the measurement cells is required to flush the \(^3\text{He}\) out of the measurement cell in a reasonable amount of time after a measurement cycle. The heat flux required is also larger. A reasonable flush time occurs with a minimum of 8 mW heat flux and a volume of 16 liters. The optimum parameters for this heat flux are given in Table 6.2. A plot of the volume average concentration up to the valve is shown in Figure 6.6.

6.3.3 Injection Volume to the Intermediate Volume 2

There will be left over \(^3\text{He}\) in the injection volume after filling MCELL with \(^3\text{He}\). Before the flush to MCELL from IV1, there is a uniform concentration between the IV1 and INJ. That concentration remains in the INJ. There is also a very small, yet none the less significant, amount of depolarized \(^3\text{He}\) in the IV1 and in the pipes leading to the measurement cells. The concentration is small, but enough so that the 99% polarization required for the measurement cell is no longer possible according to the model, thus dampening the sensitivity. The \(^3\text{He}\) must be flushed from the INJ and the pipes leading from IV1 to the first valve. With superior
cryogenic engineering, this flush ideally could be done during a measurement, without affecting the temperature in the measurement cell. However opening and closing valves may cause a systematic noise in the measurement. The flush may need to be delayed until the measurement is complete. The concentration at the IV2 pipe flush valve, shown in Figure 6.3, is plotted as a function of time shown in Figure 6.7

### 6.3.4 Intermediate Volume 2 to Sequestration Volume or Evaporation

The final step to purify the depolarized $^3$He is a flush from the large IV2 volume to a 50 cc sequestration volume. This is done at a temperature of 500 mK at the sequestration volume. The reason to increase the temperature is to decrease the mean free path of the $^3$He in order to prevent back diffusion and increase the phonon density, overall increasing the effectiveness of the heat flush. Then 8 mW of heat is applied to the tube after the valve connecting to IV2.
The sequestration volume is connected to IV2 by a 1 cm inner diameter, 20 cm long tube. With 8 mW of heat flowing through this tube the phonon velocity will be 250 cm/s. This is required due to the high concentration gradient needed for this segment of the flush.

It is uncertain if a flow of such high normal fluid velocity is achievable without the occurrence of quantum turbulence, which would severely restrict the flow and establish a temperature gradient. This is discussed in detail in the following section. The temperature of the IV2 will reach 532 mK. After the flush of $^3$He to the sequestration volume is complete, the $^3$He concentrated mixture will flow into an evaporation volume that is held at a higher temperature and pumped continuously. The gas can be analyzed after it has been pumped from the apparatus to verify the expected operation of the transport. The small volume will be re-filled with pure $^4$He before the next flush from IV2. The time it takes to empty IV2 is around 890 s and is shown in Figure 6.8.
Another possibility includes the use of a charcoal evaporator. The flush to the evaporator from IV2 and the last 425 cm of the pipe from MCELL was simulated assuming an evaporator with a radius of 5 cm and a height of 12.7 cm, and 1 liter in volume. The IV2 reaches a concentration of 0.5% after a flush of 700 s, and a concentration of 0.2% after 570 s seconds. It is assumed that the charcoal is perfectly absorbing, so if $^3$He is evaporated from the surface it always goes to the charcoal where it is adsorbed. No return flux of $^3$He at the evaporation surface is included in the model. A plot of the concentration at the exit of the evaporator is shown in Figure 6.9.

The evaporator adds complexity to the cryogenic engineering, and operation of the apparatus, but it does not require normal fluid velocities on the order of 250 cm/s.
6.4 Quantum Turbulence

The full effect of turbulence is not solvable in the Navier-Stokes model with the current analytical computation technology. With high enough normal fluid velocity, quantum vortices could become the dominant source of temperature gradients, preventing the phonon wind from flushing the $^3$He in a predictable fashion. It is thus necessary to examine quantum turbulence.

Turbulent flow in the two fluid model of superfluid helium can be described by the balance of the classical Magnus force, a force due to pressure gradients from quantum vortices, and drag forces between normal and superfluid components [54]. The behavior of the turbulent balance in our temperature and counterflow regime is examined.

6.4.1 Quantized Vortices

The superfluid component of He II is described by a single wave function. Microscopic excitations in the superfluid component make up the normal fluid. Other macroscopic excitations for the superfluid exist in the form of rotational flow. Rotational flow in a single valued wave function must be quantized,

$$\oint \frac{P}{m} \, dl = N\kappa$$  \hspace{1cm} (6.17)

$$\kappa = \frac{\hbar}{m},$$  \hspace{1cm} (6.18)

where $N$ is an integer. The existence of these excitations are a threat to slow or stop the flush of $^3$He by impeding the laminar flow of phonons to the heat sink. The motion between the normal fluid and any vortex line results in a drag force per unit length that can be expressed as [54]

$$f_D = -\alpha \rho_s \kappa \hat{s} \times (\hat{s} \times (v_n - v_s)) + \alpha \rho_s \kappa \hat{s} \times (v_n - v_s)$$  \hspace{1cm} (6.19)

where $\alpha$ is a dimensionless drag coefficient, $\hat{s}$ is the unit vector in the direction of rotation and tangent to the vortex line. $v_n$ is the normal fluid velocity, and $v_s$ is the local superfluid velocity. From the drag force and the two fluid equation of motion we can obtain a temperature gradient dependent on only the turbulent line flow [54]

$$\nabla T_{turb} = -\alpha \kappa \frac{G}{S} L^2 v_{ns}.$$  \hspace{1cm} (6.20)
Here $\nabla T_{\text{turb}}$ is the local temperature gradient due to vortices, $S$ is the local entropy, $v_{ns} = v_n - v_s$, $G$ is a geometric flow factor of order unity, and $L$ is a the vortex line density with units of inverse area.

The dynamics of quantum vortices has been accurately described as a two component fluid governed by the balance of the Magnus force, and the drag force, Equation 6.19, arising from the interactions in the fluid. The Magnus force can be written as

$$ f_M = \rho_s \hat{k} \times (v_L - v_S). $$

Solving for $v_L$ by balancing Equation 6.19 with Equation 6.21, we find

$$ v_L = v_s + \alpha \hat{s} \times (v_n - v_s) - \alpha \hat{s} \times [\hat{s} \times (v_n - v_s)]. $$

The $\alpha$ coefficient can be determined using measurements of charged vortex rings as done in reference [55]. If we consider the balance of forces in the Magnus force, the drag force, and a charge in an electric field

$$ 2\pi R (f_M + f_D) + eE = 0, $$

where $R$ is the radius of the vortex ring, $e$ is the total electric charge of the ring, and $E$ is the applied electric field. By changing the electric field the dynamics of the vortex can be controlled; the radius of the vortex can be made to expand, contract or held constant. The governing equations can be solved for special cases of vorticular motion. The force of a constant radius is calculated to be [55,56],

$$ F = eE = \delta \left[ \ln \left( \frac{8R}{a} \right) - \frac{1}{2} \right], $$

where $R$ is the radius of the vortex ring and $a$ is the radius of the vortex. For all cases considered, $a << R$. $\delta$ is a friction coefficient with units of force and is related to the dimensionless alpha by

$$ \delta = \frac{\rho_s \kappa^2}{2} \left( \frac{\alpha}{1 - \alpha} \right) \approx \frac{\rho_s \kappa^2 \alpha}{2}, $$

where $\rho_s$ is the superfluid density. The general form of $\delta$ is derived in reference [55]. For the different excitations, the values of $\delta$ are given below:
Phonons:

\[ \delta_p = \frac{\pi^6 \kappa}{20h^3} \left( \frac{kT}{c} \right)^4 \bar{\sigma}_p \]
\[ \bar{\sigma}_p = 0.3 \times 10^{-8} \text{cm} \]  \hspace{1cm} (6.26)

\[ \bar{\sigma}_p = 0.3 \times 10^{-8} \text{cm} \]  \hspace{1cm} (6.27)

Rotons:

\[ \delta_r = \frac{3\pi^2 \kappa}{8h^3} p_0^4 \pi^{\frac{3}{2}} \bar{\sigma}_r \]
\[ \bar{\sigma}_r = 9.5 \times 10^{-8} \text{cm} \]  \hspace{1cm} (6.28)

\[ \bar{\sigma}_r = 9.5 \times 10^{-8} \text{cm} \]  \hspace{1cm} (6.29)

\[ ^3\text{He} \]

\[ \delta_{^3\text{He}} = \frac{3}{8} \kappa \sqrt{2\pi m^* kT n_{^3\text{He}}} \bar{\sigma}_{^3\text{He}} \]
\[ \bar{\sigma}_{^3\text{He}} = 18.3 \times 10^{-8} \text{cm} \]  \hspace{1cm} (6.30)

All quantities except the vortex line density are defined in order to predict a temperature gradient due to a steady state vortex tangle. For the nEDM case, the only non-negligible contribution is from phonons. The temperature gradient that is established in the presence of a vortex tangle is given a normal fluid velocity on the order of 50 cm/s except in the case of a flush to the sequestration volume where velocities reach up to 250 cm/s. The total temperature gradient across the tube during a heatflush is described by the equation

\[ \nabla T_{\text{total}} = \nabla T_{\text{turb}} + \nabla T_{\text{lam}} \]  \hspace{1cm} (6.32)

where \( T_{\text{lam}} \) is the temperature gradient due to the laminar flow and is responsible for the heat flush. If \( \nabla T_{\text{turb}} \) approaches the same order as \( \nabla T_{\text{lam}} \), the expected time of the flushing will increase significantly and become very difficult to predict due to trapping of \(^3\text{He} \) on vortices and an effective reduction of the phonon wind. The temperature gradient can be computed according to the FEM model, however for this exercise the prediction found from reference [50] is sufficient. Solving for the temperature gradient in terms of the normal fluid velocity and geometry of the pipe,
Contribution of Turbulent Temperature Gradient for 250cm/s flow in 1cm diameter tube.

Figure 6.10: Contribution of the turbulent temperature gradient for 250 cm/s flow in a 1 cm diameter tube as a function of temperature. With a value of $L \leq 10^4$, the parameters are acceptable. If $L \geq 10^5$, it would seriously disrupt the heat flush.

\[ \nabla T_{\text{turb}} / \nabla T_{\text{total}} \]

\[ \nabla T_{\text{turb}} = \frac{v_n T S}{K} \]  

\[ K = \frac{1}{3} u C R \left( \frac{5}{8} R + \frac{4}{3} \right) \]  

\[ C = 2.71 \times 10^{-3} T^3 - 1.50 \times 10^{-3} T^5 + 2.64 \times 10^{-3} T^7 \left[ \frac{J}{\text{cm K}} \right] \]  

\[ \lambda = 1.28 \times 10^{-5} T^{-9} + 3.80 \times 10^{-3} T^{-4} \text{ [cm]} \]  

where $K$ is the thermal conductivity of the pipe, $v_n$ is the normal fluid velocity, $S$ is the entropy defined in Equation 6.6, $C$ is the heat capacity, $u$ is the speed of the phonons (24000 cm/s for this temperature range), $R$ is the radius of the pipe, and $\lambda$ is the mean free path of the phonons. Evaluating the ratio of $\nabla T_{\text{turb}}$ with $\nabla T_{\text{total}}$ under the assumption $G = 2/3$ for an isotropic vortex tangle at a temperature of 450 mK, and leaving $L$ as a parameter, we see the ratio is acceptable for values of $L \leq 10^4$ (see Figure 6.10).
6.4.2 Comment on the Line Density

Determining the effect the vortex line density will have on the temperature gradient is reduced to finding the line density for the flow velocity, pipe geometry and operating temperature. Not much is certain about the line density in the range of the operating temperature and geometry of the pipes. However simulations have been performed in references [57], and [54] for higher temperatures, where they find for \( T = 1.07 \) K and pipe diameter of 1 cm with normal fluid velocity of 190 cm/s, a steady state line density of \( L = 48.1 \ \text{1/cm}^2 \). The line density is proportional to the cube of the normal fluid velocity, so an increase to 250 cm/s would not increase the line density to disruptive levels. The friction coefficients by which the vortices are sustained by the flow go quartic in temperature for phonons. A serious reduction would be expected by decreasing the temperature to around 500 mK. The rotons need not be included at the temperatures of the expected operation. However rotons are major source of the friction coefficient at temperatures around 1 K and would be present in Schwarz’s simulations [54, 57], a serious reduction would be expected by the exclusion of the rotons. The experimental data appears to agree with these assumptions. A pipe of 1 cm and a heat flux of 8.7 W is required to form line densities of order \( 10^3 \) for temperatures above 1 K [54].

It can be concluded that, assuming a small density to begin with, the largest normal fluid velocity of the heat flush will not generate enough vortices to disrupt the heat flow. This does not exclude vortices that are already present in the geometry. With such a low friction coefficient, the decay time of such vortices would be extremely long.

The cooling of the apparatus will create substantial heat flows in parts of the apparatus. If there is substantial heat flow when the \(^3\text{He}\) transport tube is filled with liquid helium during cool down, then this heat flow will create vortices. If the heat flow is substantial enough for the vortices to remain until the apparatus is cooled below 400 mK, they could persist for several hours due to the low friction coefficients at these low temperatures. There may be other sources of vortices in the apparatus due to mechanical motion. For example, if the valves open with a rapid motion due to being stuck until popping out, there may be vortices generated by this rapid motion. The densities of such an event would probably be small, none the less unknown.

The line density can be described to decay according to [57]

\[
\frac{dL}{dt} = \alpha I_1 \left[ v_{ns} L^2 - \frac{1}{c} L^2 \right],
\]

(6.37)

where \( I_1 \) is the anisotropy of the vortex tangle, which is typically around 0.4 for tangles generated by heat flow [57], and \( c \) is a constant of order unity [54].

Assuming an initial line density of \( L = 10^5 \), enough to stop the heat flush, we can show the decay times of these vortices (see Figure 6.11).
Figure 6.11: Decay of vortices with no normal fluid flow at 400 mK. An initial line density of $L = 10^5 \, [1/cm^2]$ quickly decays below acceptable levels, but takes several hours to completely decay.
6.5 $^3$He Transport Summary

The heat flush method of $^3$He is proposed as a transport method for the nEDM apparatus. It is modeled by a combination of the Navier-Stokes and convection-diffusion equations. Solved with FEM, the parameters are optimized to minimize the amount of heat flux and time required for flush, while maximizing the polarization in the measurement cells. The method is shown to meet the strict requirement for maintaining 99% polarization of $^3$He in the measurement cells at the start of a measurement. Optimized parameters for removing the unpolarized $^3$He are presented for an acceptable amount of heat flux and time.

The problem of quantum vortices is considered and shown to have a negligible effect on the $^3$He transport for the parameters presented.
A positive measurement of the nEDM is a direct measurement of T-violation. A positive measurement at the level of uncertainty expected for the SNS nEDM experiment would be a measurement of physics outside the Standard Model. The largest systematic expected in this experiment is the frequency shift due to the geometric phase effect.

The geometric phase is shown to depend on the spectrum of the position autocorrelation function according to Equation 2.27. The same spectrum can be measured by gradient-induced longitudinal relaxation of spin polarized nuclei, shown in Equation 2.18. For the first time, the spectrum of the velocity weighted position autocorrelation function in restricted geometries valid for all mean free paths is presented in Equation 2.88. The velocity weighted theory is shown to agree in the limiting cases of previous theories, and diverge in the region where oscillation between the walls is the dominant contribution to the spectrum, shown in Figure 2.8. The theory agrees with simulations in this region.

Measurements of the relaxation of spin polarized $^3$He were performed at room temperature. This includes $T_2$ measurements in an applied gradient using spin echo technique, and gradient-induced $T_1$ measurements made by measuring the signal from free induction decay (FID) at equally spaced points over the time of the relaxation for varying concentrations, shown in Table 3.1. The room temperature measurements were performed in order to characterize the low temperature apparatus. The diffusion coefficient calculated from $T_2$ measurements in an applied gradient using the spin echo technique is in agreement with the expected value. Viable dilution volumes and pulse sequences were constructed resulting in a successful measurement of the gradient-induced $T_1$ of spin polarized $^3$He after rarefication of a factor of 1000.

Several improvements to the previous iteration of the experiment were made. A superior four-coil design was constructed and successfully implemented to increase the S/N by a factor of
15,000. Superconducting magnets consisting of 1800 turns were constructed, successfully cooled to low temperature and provided a suitable magnetic field for accurate relaxation measurements. A liquid helium cold trap was designed and provided isotopically pure $^4$He free of nitrogen contamination. A system of dilution volumes for the low temperature apparatus were constructed, and the problem of the barrier pressure due to film flow was realized. Prompting the technique to introduce 500 mbar of isotopically pure $^4$He into the intermediate cell prior to transferring the rarefied polarized $^3$He into the sample.

Magnetization in the apparatus was controlled after extensive characterization. Its effect was minimized by ramping the SCG and SCH to specific values and times in the proper order. It could be neglected in the analysis of the measurements.

The gradient-induced $T_1$ was measured in the temperature range of $T = 430$ mK→560 mK, in magnetic fields from $B_0 = 11.5$ G→50 G, and in concentrations of $X = 1 \times 10^{-6} \rightarrow 3.3 \times 10^{-3}$, requiring a S/N a factor 15,000 larger than the previous iteration of the experiment. The analysis of the data shows that the theory presented in Equations 2.18 and 2.88 adequately predicts the data. Previous theories exist, found in Equation 2.96, that are adequate to describe the data in this region. The presented theory is shown to agree with previous theories limited to this region and diverge with slightly more ballistic motion than the experiment was able to achieve. Diffusion theory, found in Equation 2.97, is shown to be inadequate to describe the data for the longer scattering times measured. The measurements and all presented theories agree for the shorter scattering times. Furthermore, the expected operating range in the nEDM apparatus of the parameter $\lambda/L \approx 0.029$ was measured and shown to agree with both valid theories in that region.

The heat flush method of $^3$He is proposed as a transport method for the nEDM apparatus. It is modeled by a combination of the Navier-Stokes and convection-diffusion equations. Solved with FEM, the parameters are optimized to minimize the amount of heat flux and time required for flush, while maximizing the polarization in the measurement cells. The method is shown to meet the strict requirement for maintaining 99% polarization of $^3$He in the measurement cells at the start of a measurement. Optimized parameters for removing the unpolarized $^3$He are presented for an acceptable amount of heat flux and time.

This work presents a method to model the heat flush by the Navier-Stokes and convection-diffusion equations. This model was used to find optimum parameters that enable 99% polarization of polarized $^3$He to remain after the transport and at the start of a measurement. During the measurement, $^3$He will experience a frequency shift linear in the applied electric field due the the geometric phase effect. This effect limited the sensitivity of the most precise nEDM measurements to date. The most accurate method to predict this frequency shift is using the spectrum of the position autocorrelation function for the continuous time random walk undergoing isotropic scattering in a restricted geometry. A theory that satisfies this is
derived for up three spatial dimensions. The application of a velocity weighting to the theories of different dimensions is shown to unify all three theories and previous limited theories in their valid regions. The theory is shown to agree with relaxation measurements for values of the mean free path in the measurement, determined from the concentration of $^{3}$He and temperature of the sample. The frequency shift can be calculated and scaled from relaxation rates measured to the nEDM apparatus. The nEDM is expected to operate in a region around the scaling factor $\lambda/L \approx 0.029$. This region was measured and is presented in this work. According to theory and measurements, the predicted frequency shift due to the geometric phase of $^{3}$He, $d_f \approx 5 \times 10^{-30}$ e cm, is acceptable for the desired sensitivity of $d_n < 3 \times 10^{-28}$ e cm.

7.1 Outlook

Film pinners currently under investigation for the PULSTAR $^{3}$He systematic studies apparatus could potentially solve the problem of film flow up the tube and decrease the base operating temperature, allowing for a measurement in the region where the theories diverge. The final stage for the PULSTAR $^{3}$He systematic studies apparatus will measure the properties of the correlation function in the ballistic regime.
REFERENCES


[53] Design and physical parameters. Internal collaboration note from George Seidel.


Table A.1: Successful measurements in the low temperature apparatus prior to using isotopically pure $^4$He.

<table>
<thead>
<tr>
<th>Date</th>
<th>Run</th>
<th>Type</th>
<th>Result</th>
<th>error and type</th>
<th>T wire</th>
<th>T cell</th>
<th>He3:He4</th>
<th>Notes:</th>
</tr>
</thead>
<tbody>
<tr>
<td>12/14/2010</td>
<td>692</td>
<td>Pulse Loss</td>
<td>120us</td>
<td>pi/2=348.59257 us</td>
<td>0.1%</td>
<td>Noise/ Fun. Gen.</td>
<td>450mK</td>
<td>463mK</td>
</tr>
<tr>
<td>12/14/2010</td>
<td>693</td>
<td>Wall T1</td>
<td>T1=8988.9 s</td>
<td>18.6 s Noise &amp; Pulse</td>
<td>450mK</td>
<td>463mK</td>
<td>3.12E-03</td>
<td></td>
</tr>
<tr>
<td>12/17/2010</td>
<td>Notes: Put in 3 fills of 3.24E-6 concentration each. No data but:</td>
<td>461mK</td>
<td>468mK</td>
<td>3.13E-03</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12/21/2010</td>
<td>701</td>
<td>Gradient T1</td>
<td>T1G=1734.7s</td>
<td>80.5s Noise &amp; Pulse</td>
<td>466mK</td>
<td>475mK</td>
<td>3.12E-06</td>
<td>B=11.5G G=80mG/cm</td>
</tr>
<tr>
<td>12/22/2010</td>
<td>702</td>
<td>Wall T1</td>
<td>T1=4807.2s</td>
<td>133.9s Noise &amp; Pulse</td>
<td>466mK</td>
<td>475mK</td>
<td>6.25E-06</td>
<td>Reduced T1</td>
</tr>
<tr>
<td>12/24-12/28</td>
<td>Notes: Put in 3 fills of 3.12E-3 concentration each. No data but:</td>
<td>520mK</td>
<td>496mK</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>25-Dec</td>
<td>1 k pot pump broke! Merry Christmas</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12/29/2010</td>
<td>721</td>
<td>Gradient T1</td>
<td>T1G=1042.3s</td>
<td>702s A Lot of Noise.</td>
<td>513mK</td>
<td>491mK</td>
<td>3.12E-06</td>
<td>B=50G G=2.56G/cm</td>
</tr>
<tr>
<td>12/30/2010</td>
<td>722</td>
<td>Gradient T1</td>
<td>T1G=824.3</td>
<td>63.2s Noise and Pulse</td>
<td>512mK</td>
<td>493mK</td>
<td>9.37E-06</td>
<td>B=50G G=2.56G/cm</td>
</tr>
<tr>
<td>12/31/2010</td>
<td>723</td>
<td>Gradient T1</td>
<td>T1G=1066.3</td>
<td>88.8s Noise and Pulse</td>
<td>512mK</td>
<td>491mK</td>
<td>2.11E-05</td>
<td>B=50G G=2.56G/cm</td>
</tr>
<tr>
<td>1/2/2011</td>
<td>724</td>
<td>Gradient T1</td>
<td>T1G=1249.3s</td>
<td>56.6s Noise and Pulse</td>
<td>513mK</td>
<td>493mK</td>
<td>3.12E-06</td>
<td>B=30G G=1.23G/cm</td>
</tr>
<tr>
<td>1/3/2011</td>
<td>725</td>
<td>Gradient T1</td>
<td>T1G=1317.5s</td>
<td>55.3s Noise and Pulse</td>
<td>495mK</td>
<td>494mK</td>
<td>9.40E-06</td>
<td>B=30G G=1.23G/cm</td>
</tr>
<tr>
<td>1/4/2011</td>
<td>726</td>
<td>Gradient T1</td>
<td>T1G=2839.3</td>
<td>89.8 Noise and Pulse</td>
<td>513mK</td>
<td>489mK</td>
<td>3.12E-06</td>
<td>B=40G</td>
</tr>
<tr>
<td>1/7/2011</td>
<td>728</td>
<td>Gradient T1</td>
<td>T1G=2452.6</td>
<td>72.6 Noise and Pulse</td>
<td>510mK</td>
<td>486mK</td>
<td>2.11E-05</td>
<td>B=40G G=1.89G/cm</td>
</tr>
<tr>
<td>1/8/2011</td>
<td>732</td>
<td>Wall T1</td>
<td>T1=4095.6s</td>
<td>18.5s Noise and Pulse</td>
<td>512mK</td>
<td>486mK</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table A.2: Successful measurements in the low temperature apparatus after using isotopically pure \(^4\)He.

<table>
<thead>
<tr>
<th>Date</th>
<th>Run</th>
<th>Type</th>
<th>Result</th>
<th>error and type</th>
<th>T wire</th>
<th>T cell</th>
<th>He3:He4</th>
<th>Notes:</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/8-1/20/2011</td>
<td>Notes:Several runs were made to diagnose the fields and concentrations, concentrations seemed inaccurate due to T2. Therefore a switch to isotopically pure helium-4 produced from a Mcintosh purifier</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1/22/2011</td>
<td>760</td>
<td>Gradient T1</td>
<td>T1=759.4</td>
<td>9.5121 Noise and Pulse</td>
<td>692mK</td>
<td>565mK</td>
<td>3.12E-06</td>
<td>B0=40G Gz=1.89G/cm</td>
</tr>
<tr>
<td>1/23/2011</td>
<td>761</td>
<td>Gradient T1</td>
<td>T1=491.9</td>
<td>6.3s</td>
<td>692mK</td>
<td>566mK</td>
<td>6.25E-06</td>
<td>B0=40G Gz=1.89G/cm</td>
</tr>
<tr>
<td>1/24/2011</td>
<td>762</td>
<td>Gradient T1</td>
<td>T1=338.0</td>
<td>5.1s</td>
<td>692mK</td>
<td>566mK</td>
<td>9.37E-06</td>
<td>B0=40G Gz=1.89G/cm</td>
</tr>
<tr>
<td>1/25/2011</td>
<td>763</td>
<td>Gradient T1</td>
<td>T1=248.1</td>
<td>2.3s</td>
<td>692mK</td>
<td>568mK</td>
<td>2.15E-05</td>
<td>B0=40G Gz=1.89G/cm</td>
</tr>
<tr>
<td>1/27/2011</td>
<td>764</td>
<td>Gradient T1</td>
<td>T1=333.2</td>
<td>3.1s</td>
<td>692mK</td>
<td>568mK</td>
<td>4.34E-05</td>
<td>B0=40G Gz=1.89G/cm</td>
</tr>
<tr>
<td>Prior to this set of measurements there was a clog in the capillary and the helium was slowly condensed through the glass tube. The glass tube was clogged on the next attempt to condense the helium. It is believed that the temperatures are accurate. As its hard to believe there is 124mK difference across the cell filled with helium.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3/22/2011</td>
<td>769</td>
<td>Gradient T1</td>
<td>3547.4s</td>
<td>132.9s</td>
<td>430mK</td>
<td>no rec.</td>
<td>1.04E-06</td>
<td>B0=50G Gz=2.50G/cm</td>
</tr>
<tr>
<td>3/23/2011</td>
<td>770</td>
<td>Gradient T1</td>
<td>1927.0s</td>
<td>20.0s</td>
<td>433mK</td>
<td>?</td>
<td>2.08E-06</td>
<td>B0=50G Gz=2.50G/cm</td>
</tr>
<tr>
<td>3/24/2011</td>
<td>771</td>
<td>Gradient T1</td>
<td>1413.6s</td>
<td>88.1s</td>
<td>435mK</td>
<td>?</td>
<td>3.12E-06</td>
<td>B0=50G Gz=2.50G/cm</td>
</tr>
<tr>
<td>First successful implementation of multiple measurements per transfer cell</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3/30/2011</td>
<td>777</td>
<td>Wall T1</td>
<td>T1=8784.3s</td>
<td>222.19s</td>
<td>512mK</td>
<td>523mK</td>
<td>1.04E-06</td>
<td>B0=11.5G</td>
</tr>
<tr>
<td>3/30/2011</td>
<td>778</td>
<td>Wall T1</td>
<td>T1=8828.4</td>
<td>118.8s</td>
<td>512mK</td>
<td>523mK</td>
<td>4.54E-06</td>
<td>B0=11.5G</td>
</tr>
<tr>
<td>3/30/2011</td>
<td>779</td>
<td>Wall T1</td>
<td>T1=8992.1s</td>
<td>79.8s</td>
<td>512mK</td>
<td>523mK</td>
<td>1.25E-05</td>
<td>B0=11.5G</td>
</tr>
<tr>
<td>3/30/2011</td>
<td>782</td>
<td>Gradient Switch Loss</td>
<td>Loss=0.00 (from expected)</td>
<td>516mK</td>
<td>526mK</td>
<td>1.38E-04</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3/30/2011</td>
<td>783</td>
<td>Pulse Loss 85u pulse</td>
<td>pi/2=389.01 us</td>
<td>0.09%</td>
<td>516mK</td>
<td>526mK</td>
<td>1.38E-04</td>
<td></td>
</tr>
<tr>
<td>3/30/2011</td>
<td>784</td>
<td>Pulse Loss 50u Pulse</td>
<td>pi/2=403.18 us</td>
<td>0.08%</td>
<td>516mK</td>
<td>526mK</td>
<td>1.38E-04</td>
<td></td>
</tr>
</tbody>
</table>

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