Abstract

Nuclear Spin-Dependent Parity Nonconservation in Diatomic Molecules
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Nuclear spin-dependent parity nonconserving phenomena produce effects that are relevant to particle, nuclear, and atomic physics. They unite these increasingly disparate disciplines via the subtle effects of the weak force. Currently, weak interactions between nucleons present at best a confusing picture, and neutral weak coupling of leptons to baryons, even at high energy, is one of the most poorly characterized aspects of the Standard Model.

Perhaps surprisingly, one of the most effective laboratories for studying parity violation is the diatomic molecule. The naturally close spacing of molecular hyperfine-rotational levels, which can be Zeeman shifted even closer to degeneracy, allows small parity violating effects to be enhanced dramatically. Here we describe the beginning of our experimental program to study nuclear spin-dependent parity nonconservation (NSD-PNC) using a beam of diatomic free radicals. Specifically, we seek to measure weak matrix elements connecting hyperfine-rotational states in a series of molecules, beginning with barium fluoride. Our experimental technique is based on a Stark interference method: we look for asymmetries in transitions that depend on the sign of an applied electric field.

A key element in the experimental program is the magnetic field used to Zeeman shift the levels in our molecules to near crossing. Carefully controlling both the strength
and uniformity of this field is critical to the success of the experiment. In order to measure the field over the wide range of values required by our experiment, both a completely novel nuclear magnetic resonance probe, as well as a unique field shimming apparatus and algorithm, were developed.

A second important element in the program is the ability to detect the states of our molecules with as high a signal to noise as possible. To that end a new detection method based on a two-photon transition was developed.

Both of these areas, which constitute the bulk of the author’s contribution to the experimental program, are described in detail.
Nuclear Spin-Dependent Parity Nonconservation in Diatomic Molecules

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To my parents
Chapter 1: Theory

1.1 Introduction

In this chapter we discuss the theoretical underpinnings of our experimental program. We describe the physical processes which drive parity violation in atoms and molecules, then explain how they result in an observable signal. We finish by discussing the potential impact of our experimental program. Much of the work in this chapter draws heavily on the treatments in Khriplovich [1] as well as Ginges and Flambaum [2]. A convenient summary of the nuclear spin-dependent contributions can be found in Section 1.4.

1.2 Overview

Parity violation in atomic and molecular systems derives primarily from two distinct causes: neutral weak interactions between electrons and the nucleus, and both charged and neutral weak interactions within the nucleus itself. The first are characterized in terms of what are known as $C_{1,2}$ constants, while the second give rise to the so called nuclear anapole moment. The effects of these interactions can be conveniently divided into those which depend on the nuclear spin (NSD) and those which do not (NSI).

A nuclear anapole moment is a $P$-odd electromagnetic moment which appears due to weak interactions between nucleons. It gives rise to a magnetic field inside the nucleus. Electrons in atoms or molecules interact with this magnetic field while they are inside the
nucleus, and the effect is to mix \(|s\rangle\) and \(|p\rangle\) electronic states. The electron-nucleon NSD-PNC interaction also mixes these states, so any PNC measurement sees both of these effects simultaneously. In order to distinguish between the two we take advantage of their different scaling with atomic number. Since the anapole contribution scales as \(A^{2/3}\) while the \(C_2\) contribution is roughly constant in atomic number, we can measure the anapole dominated PNC amplitude in several heavier nuclei, then subtract it to determine the \(C_2\) coefficients. The nuclear structure calculations necessary to interpret the anapole moment measurements have an uncertainty at the 30% level, which is consistent with our ultimate experimental precision. Other PNC effects, including interference terms between the nuclear spin independent electron-nucleus interaction and the ordinary hyperfine interaction are either small or well understood, so can effectively be ignored.

A summary of the NSD-PNC effects relevant for our experiment is presented in Section 1.4.
1.3 Contributions

1.3.1 Electron-Nucleon Weak Interactions

Consider the following interaction:

\[ H = \bar{N} \left( -\frac{ig_z}{2} \gamma^\mu (C_{V,N} - C_{A,N} \gamma^5) \right) N \frac{ig_{\mu\nu}}{M_z^2} e \left( -\frac{ig_z}{2} \gamma^\nu (C_{V,e} - C_{A,e} \gamma^5) \right) e \]  

where \( N \) is the nucleon field, \( e \) is the electron field, \( g_z \) is the neutral weak coupling constant, \( g_{\mu\nu} \) is the Minkowski metric, \( M_z \) is the mass of the \( Z \) boson, and the \( C_{V,A,N,e} \) are the coupling constants characterizing the current (vector or axial vector) and the associated particle (nucleon or electron). We are using the notation of Khriplovich [1], where the Dirac matrices are given by:

Figure 1-1: \( Z \) exchange between an electron and a nucleon is a fundamental source of parity violation in atoms and molecules.
\[ \gamma_0 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad \gamma_m = \begin{pmatrix} 0 & \sigma_m \\ -\sigma_m & 0 \end{pmatrix}, \quad \gamma_5 = \begin{pmatrix} 0 & -I \\ -I & 0 \end{pmatrix}, \]

with the \( \sigma_m \) representing the Pauli spin matrices. Because we are working at low energy, \( q \) is much lower than \( M_z = 91.2 \) GeV, and our propagator has been reduced to \( \frac{ig_{\mu\nu}}{M_z^2} \).

If we expand (1.1) we are left with four terms, two of which are P-even and two which are P-odd. Considering only the P-odd terms we can rewrite this as

\[ \hat{h} = \frac{g_F}{\sqrt{2}} [C_{1N} \bar{e} \gamma_\mu \gamma_5 e \bar{N} \gamma^\mu N + C_{2N} \bar{e} \gamma_\mu e \bar{N} \gamma^\mu \gamma_5 N]. \]  (1.2)

Or, including the interaction with the entire nucleus,

\[ \hat{h} = \frac{g_F}{\sqrt{2}} \sum_N [C_{1N} \bar{e} \gamma_\mu \gamma_5 e \bar{N} \gamma^\mu N + C_{2N} \bar{e} \gamma_\mu e \bar{N} \gamma^\mu \gamma_5 N] \]  (1.3)

The constant \( C_{1N} \) is proportional to \( C_{V,N} C_{A,e} \) and describes the vector nucleon – axial vector electron \( (V_N A_e) \) contribution, while the constant \( C_{2N} \) is proportional to \( C_{A,N} C_{V,e} \) and describes the vector electron – axial vector nucleon \( (V_e A_N) \) contribution. As we will see shortly the \( C_1 \) term gives rise to nuclear spin-independent parity violation, while the \( C_2 \) term produces parity violation which depends on the nuclear spin. As an aside, note that in (1.3) we are ignoring the anomalous magnetic moment of the proton. The interested reader can see [1] for details.
If we assume that the nucleon is infinitely heavy, with only spin degrees of freedom, we can rewrite this field-theoretic expression (1.2) as a Hamiltonian which operates on four-component Dirac wavefunctions of the electron in an atom or molecule:

$$H = \frac{G_F}{\sqrt{2}} \sum_N \left[ C_{1N} \gamma_5 + C_{2N} (\gamma_0 \cdot \vec{\gamma}_N) \sum \right] \delta(r)$$

(1.4)

Here \( \Sigma \) refers to the nucleon spin. In the literature the \( \gamma_0 \cdot \vec{\gamma} \) term is frequently written as \( \vec{\alpha} \). This is the fundamental Hamiltonian used when describing parity violating effects in atoms and molecules. An important feature is the \( \delta \)-function, which highlights the nuclear-localized character. The \( \delta \)-function appears because we approximate the nucleon as infinitely heavy, and having only spin degrees of freedom. In this limit nucleon currents take the forms compiled in Appendix A. Because of the presence of the \( \delta \)-function, parity violating interactions will clearly have a larger effect on electrons that spend more time inside the nucleus.

We can carry this one step further by again following Khriplovich [1] and taking the limit in which the electrons are non-relativistic. This leads to:

$$H_{NR} = \frac{G_F}{2m_e} \sum_N \left[ C_{1N} (\overline{\sigma} \cdot \overline{p}) + C_{2N} (\overline{\Sigma} \cdot \overline{\sigma})(\overline{\sigma} \cdot \overline{p}) \right] \delta(r)$$

(1.5)

This Hamiltonian operates on electron wavefunctions written in the basis of two-component non-relativistic spinors. The second term is clearly associated with nuclear spin-dependent effects, while the first depends only on the electron spin. While this expression is inappropriate for heavy atoms, where relativistic effects are relevant, for light atoms it is a good approximation.
1.3.1.1 Connection to Standard Model

In the previous section we considered neutral weak currents of a nucleon-electron system. The \( C_{1,2} \) constants for both protons and neutrons can be expressed, to lowest order in the electroweak interaction [3], as

\[
C_{1p} = \frac{1}{2} (1 - 4 \sin^2 \theta_W), \quad C_{1n} = -\frac{1}{2}
\]

\[
C_{2p} = -C_{2n} = -\frac{1}{2} (1 - 4 \sin^2 \theta_W) \lambda
\]

where \( \theta_W \) is the Weinberg angle (\( \sin^2 \theta_W = 0.23 \)) and \( \lambda \approx 1.26 \), where \( \lambda \) is a correction factor due to the strong interaction. In a simple model of nucleons as a sum of three non-interacting quarks, from electroweak theory one expects \( \lambda = 1 \). However, the strong interaction between quarks in the nucleon renormalizes this axial coupling. The renormalized value has been determined by measurements of the charged-current axial vector coupling in the beta decay of free neutrons, and is found to be \( \lambda \approx 1.26 \).

It is important to note that \( C_{1n} \) is approximately an order of magnitude larger than the other coefficients, which are roughly equal; thus interactions which include it will be the dominating source of parity violation.

The constants can also be written in terms of the more fundamental quark couplings \( C_{2u} \) and \( C_{2d} \). See for example [4] and [5]. Including SU(3) and radiative corrections,
\[ C_{1p} = 2C_{1u} + C_{1d} \]
\[ C_{1n} = C_{1u} + 2C_{1d} \]
\[ C_{2p} = 2FC_{2u} + (F - D)C_{2d} \]
\[ C_{2n} = (F - D)C_{2u} + 2FC_{2d} \]

where \( F + D = g_d \approx 1.26 \), \( F = 0.425 \) and \( D = 0.825 \). Again here, the deviation of \( F \) and \( D \) from 0.5 is caused by strong interactions within the nucleon.

The fact that nucleons are composite particles of strongly interacting quarks is an important consideration. This is especially true for the spin-dependent portion, as it derives from a nucleon axial current, and is not protected from strong interaction effects. Khriplovich [1] calls the strong interaction and nuclear structure effects "a serious problem of a principal character". Looking forward, while a priori this is a concern for our experiment, the precision of our measurement is similar to the uncertainty in the theory so it is not a limitation. Seen from another point of view, our measurements could provide one of the few ways to actually determine the quantities \( F \) and \( D \), and hence probe the way the neutral-current weak interaction is renormalized by strong interactions in the nucleus.
1.3.2 Nuclear Spin-Independent Interactions

Consider the first term in (1.4). After summing over all nucleons, this can be written as:

\[ H = \frac{G_F}{2\sqrt{2}} \gamma_5 [Z C_{1p} \rho_p(r) + N C_{1n} \rho_n(r)] \]  
(1.7)

where \( Z \) is the number of protons and \( N = A - Z \) is the number of neutrons. Here \( \rho_{p,n}(r) \) is the nuclear density function for protons and neutrons. The total number of protons and neutrons appears because the nucleons contribute coherently to the interaction. This occurs because the electron wavefunction is nearly constant over the size of the nucleus. If instead of being constant the wavefunction oscillated inside the nucleus, the simple coherent contribution would be replaced by an additional form factor related to the nuclear structure.

If we assume that the densities are the same for both protons and neutrons, (1.7) reduces to

\[ H = \frac{G_F}{2\sqrt{2}} Q_w \rho(r) \gamma_5 \]  
(1.8)

or, in the case of a non-relativistic electron,

\[ H = \frac{G_F}{2m_e \sqrt{2}} Q_w \left( \vec{\sigma} \cdot \vec{p} \right) \delta \left( \vec{r} \right) \]  
(1.9)

The constant \( Q_w \) is known as the nuclear weak charge, and

\[ Q_w = Z(1 - 4 \sin^2 \theta_w) - N \approx -N . \]  
Radiative corrections modify the nuclear weak charge, giving it a value of \( Q_w = -0.9857N + 0.0675Z \) [6].

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Because of the large relative size of $C_{1n}$, as well as the coherent contribution from all nucleons, the spin independent interaction is the largest source of parity violation in atoms. The effect is further enhanced in heavy atoms because it scales as $Z^3$ [7,8,9], where one factor of $Z$ comes from the expectation value of the delta function, one factor comes from the expectation value of the momentum operator, and the final factor comes from $Q_{N}$, with the approximation that $N \approx Z$. The operator is a pseudoscalar, however, so can only connect states of the same angular momentum and of opposite parity. In particular, the spin-independent interaction plays no role in our experiment, as our states of interest have differing angular momenta.

1.3.3 Nuclear Spin-Dependent Interactions

The focus of our experiment is to measure parity violation that depends on the nuclear spin. There are two major contributing processes to the effect: neutral weak currents between electrons and the nucleus, and weak interactions inside the nucleus. The contribution from weak currents between nucleons stems from what is known as the anapole moment and will be discussed in the next section. Here we focus on the spin dependent portion of the vector-electron axial-nucleon coupling. This interaction originates from the second term of (1.4), and is written most transparently by assuming a non-relativistic electron, as in the second term of (1.5):

$$H = \frac{G_F}{2m_e \sqrt{2}} \sum_N C_{2N} \left( \sum_N \vec{\sigma} \left( \sum_N \vec{\sigma} \right) \right) \delta \left( \vec{r} \right)$$

(1.10)

In direct contrast with the spin-independent interaction discussed above, here the nucleons do not contribute coherently. Instead their contributions cancel in pairs. We can then rewrite the Hamiltonian as
\[ H = \frac{G_F}{2m_e \sqrt{2}} C_2 \left( \vec{j} \cdot \vec{\sigma} \right) \left( \vec{\sigma} \cdot \vec{p} \right) \delta \left( \vec{r} \right) \]  

(1.11)

This pair canceling occurs because, unlike the filling of electron shells, nuclear shells almost always fill such that, beginning with an unpaired proton (or neutron), the next proton (or neutron) added will be in a state with opposite spin to the original proton (or neutron). This leaves two unpaired spins for odd proton - odd neutron nuclei, and for others at most a single unpaired spin. This reduces significantly the effect of the spin dependent contribution relative to the spin independent contribution, by a factor of \( \sim N \).

Since \( C_{2p} = -C_{2n} \), odd proton and odd neutron nuclei will see the same magnitude of spin-dependent effect due to this electron-nucleon weak interaction. However, \( C_2 \) is small compared to \( C_{1n} \), so this reduces the effect relative to the NSI interaction discussed earlier by another factor of \( 1 - 4 \sin^2 \theta \) \( \sim 0.08 \).

In order to facilitate future connection of this spin dependent electron-nucleon weak interaction with that of the nucleon-nucleon weak interaction, it is useful to rewrite the second term in (1.4) to explicitly include the sum over nucleons \( N \):

\[ H = \frac{G_F}{\sqrt{2}} \sum_N C_{2N} \bar{\alpha} \cdot \vec{\sigma}_N \rho(r) \]  

(1.12)

Here \( N \) refers to nucleon, and should not be confused with \( n \), which refers specifically to a neutron. \( \bar{\alpha} = \gamma_0 \cdot \vec{r} \) and \( \sigma_N \) are the nucleon Pauli matrices. Averaging over the nucleus (see [1], Chapter 8 or [2]) yields

\[ H = \frac{G_F}{\sqrt{2}} \kappa_2 \bar{\alpha} \cdot \vec{l} \rho(r) \]  

(1.13)
where \( K = (I + 1/2)(-1)^{I+1/2-I} \) and \( \kappa_2 = C_{2\nu} \frac{1/2 - K}{I+1} \). Here \( \nu = n(p) \) for a neutron (proton) with angular momentum \( I \). This expression has the same form as the contribution from the nuclear anapole moment, which is discussed in the next section. Note that in the literature there are various conventions for the definitions of \( K \) and \( \kappa \).

Here we use the convention of [10].

### 1.3.4 Nuclear Anapole Moments

The origin of the nuclear anapole moment can be explored following references [1],[2],[11] and [12]. An informative review is in [13]. The only measurement to date of an anapole moment was performed in \(^{133}\text{Cs}\) by Wieman et al [14]. The relation between nuclear anapole moments and meson exchange models for nuclear weak forces can also be found in [11],[12], [13] and [16]. The "best values" for the experimentally based nucleon-meson couplings are determined in [16].

In the context of this experiment the anapole moment can be thought of as the source of an additional magnetic field, confined strictly to the nucleus, that is caused by weak interactions between nucleons. Electrons can interact with this magnetic field when they are inside the nucleus, and this interaction takes the same form as the spin-dependent parity violating electron-nucleon interaction discussed above. The punch line of all that follows is that weak interactions between nucleons give rise to the following effective Hamiltonian for an electron in an atom or molecule:

\[
H = \frac{G_F}{\sqrt{2}} \kappa \frac{\bar{\alpha} \cdot \vec{l}}{I} \rho(r)
\]  

(1.14)
The variables are the same as above with the addition of $\kappa_A$, which is a dimensionless parameter describing the strength of the anapole moment[15]:

$$\kappa_A = \frac{9}{10} \frac{\alpha \mu}{m r_0} A^{2/3} g_\nu.$$  \hspace{1cm} (1.15)

Here $\alpha$ is the fine structure constant, $\mu$ is the magnetic moment of the valence nucleon in nuclear magnetons, $r_0 = 1.2$ fm is the nuclear radius in the liquid-drop model, and $g_\nu = g_{p,n}$ is a constant which varies depending on whether the valence nucleon is a proton or a neutron. These constants $g_{p,n}$ describe the strength of the weak interaction between nucleons, and thus fall within the theoretical sweep of low energy hadronic PNC. Their values are approximately in the ranges $g_p \approx 4 - 6$ and $g_n \approx -(0.2 - 1)$. These interactions are difficult to analyze theoretically, and historically the most widely accepted attempt is a meson exchange model developed by Desplanques, Donoghue, and Holstein [16]. The basic process consists of one nucleon emitting a meson via a weak interaction, and a second nucleon absorbing the meson through a strong interaction. The theory cannot calculate the $\bar{NNM}$ couplings (where $N$ is a nucleon and $M$ is a meson) from first principles, so they are fit to experimental numbers and then tuned within the context of the model. The so called DDH “best values” for these couplings are found in [16], where DDH are the authors’ initials. In total there are six parameters required to describe all possible low energy hadronic PNC effects. Our constants of interest can be expressed as:

$$g_p = 2.0 \times 10^5 W_p \left[ 176 \frac{W}{W_p} f_x - 19.5 h_\rho^0 - 4.7 h_\rho^1 + 1.3 h_\rho^2 - 11.3 \left( h_\omega^0 + h_\omega^1 \right) \right]$$

$$g_n = 2.0 \times 10^5 W_p \left[ -118 \frac{W}{W_p} f_x - 18.9 h_\rho^0 + 8.4 h_\rho^1 - 1.3 h_\rho^2 - 12.8 \left( h_\omega^0 + h_\omega^1 \right) \right]$$
Each of the \(f, h\) parameters here are weak meson-nucleon coupling constants, with the subscript indicating the meson and the superscript indicating whether the interaction is isoscalar, isovector, or isotensor. Their roles are important here only because our experiment will help constrain their phase space. This explosion in theoretical parameters is indicative of the difficulty in calculating weak nuclear effects. It is worth noting though that the Standard Model itself has 28 free parameters.

One challenge of our experiment is to distinguish the parity violating effects due to the anapole moment from those due to the electron-nucleon interaction. Bouchiat and Bouchiat [8, 9] have shown that the effects of the anapole moment scale as \(A^{2/3}\). As seen in (1.6), the \(C_2\) contribution is constant in atomic number. We can use these scaling effects in conjunction with PNC measurements in several nuclei to extract the portion corresponding to each contributing term.

1.3.4.1 **Origin of the Anapole Moment and Its Effect on Nuclear Structure**

As previously mentioned, a nuclear anapole moment is a \(P\)-odd electromagnetic moment which appears due to weak interactions between nucleons. In the meson exchange picture of nuclear weak forces, a typical interaction might be as in Figure 1-2.
Figure 1-2: Meson exchange picture of low energy hadronic PNC interaction.

If we adopt a shell model of the nucleus, with a valence nucleon orbiting a nuclear core, we can write the effective $P$-odd weak interaction between an unpaired nucleon and the nuclear core as [2]:

$$ H = \frac{G_F}{2\sqrt{2}m_N} g \left[ \vec{\sigma} \cdot \vec{p} \rho(r) + \rho(r) \vec{\sigma} \cdot \vec{p} \right] $$

(1.16)

where $\rho(r)$ is the core nucleon number density. If we assume the nuclear density is constant, we can treat (1.16) as a perturbation to the general Hamiltonian, and solve the Schrödinger equation using perturbation theory. The result is

$$ \psi = e^{i\theta \cdot \vec{r}} \psi_0 $$

(1.17)

where $\theta = -g \frac{G_F \rho}{2}$ and $\vec{r}$ is the position of the valence nucleon. The spin of the unperturbed wave function $\psi_0$ is rotated around $\vec{r}$ by an angle of $2\theta r$. Thus the PNC Hamiltonian gives the valence nucleon wave function a spin helix structure. An illustration of this structure can be found in Figure 1-3. This spin helix produces a toroidal magnetic field localized within the nucleus, depicted in Figure 1-4.
Recall that the ranges for $g_p$ and $g_n$ are $g_p \approx 4 - 6$ and $g_n \approx - (0.2 - 1)$. Because $g_p$ is so much larger than $g_n$, unlike the electron-nucleon weak interaction discussed above, odd proton nuclei have much greater anapole moments than odd neutron nuclei. This has the result of making our experimental measurement of $C_{2n}$ more precise than $C_{2p}$ because of reduced uncertainty in subtracting the anapole contribution from the total PNC signal.

Figure 1-3: Parity violating interactions between a valence nucleon and the nuclear core create a spin helix
1.3.5 Manifestation in Atomic and Molecular Systems

Parity violating interactions manifest themselves in atomic and molecular systems by admixing $|s\rangle$ and $|p\rangle$ electronic states. Formally we can see this by considering (1.5) as a perturbation and solving the Schrödinger equation. The resulting perturbation to the wave function takes the form:

$$
|s_{1/2}\rangle \rightarrow |s_{1/2}\rangle + \frac{\langle s_{1/2}|H_{\text{Weak}}|p_{1/2}\rangle}{(E_{p_{1/2}} - E_{s_{1/2}})} |p_{1/2}\rangle = |s_{1/2}\rangle + i\eta |p_{1/2}\rangle
$$

(1.18)

Qualitatively, we can also understand the effect by considering that because the weak interaction violates parity, parity can no longer be considered a good quantum number.

The stationary states of a system which includes weak interactions must now be mixtures of states with the same total angular momentum but of opposite parity.
1.3.6 Additional Contributions

1.3.6.1 Hyperfine Interference

There is an additional contribution to NSD-PNC due to a combined effect of the spin-independent weak interaction and the usual magnetic hyperfine interaction. This interaction can be seen in Figure 1-5, and is discussed in [17] then more recently in [18]. Its contribution is much smaller than either the anapole term or the $C_2$ term, and its scaling behavior is well understood, so henceforth we ignore it.

1.3.6.2 Electron-Electron Weak Interactions

Neutral weak coupling also occurs between electrons. As discussed in [1],[8] and [19], it can also mix levels of opposite parity. However, the effect is several orders of magnitude smaller than other contributions and does not lead to NSD-PNC effects.

1.3.6.3 Parity Conserving Weak Interactions

Equation (1.1) also contains parity conserving terms. These lead to energy level shifts which in many cases are measurable. For example, $P$-even weak interactions shift the hyperfine splitting of the hydrogen ground state by a factor on the order of $10^{-4}$ MHz. As far back as 1966 this splitting was measured to a precision of $\pm 1.7 \times 10^{-6}$ MHz [20]. However, theoretical uncertainties due to proton polarizability and strong interaction effects are on the order of $10^{-2}$ MHz, making the result uninterpretable. In muonium [21], the ground state hyperfine interval has been measured to 12 ppb, but the theoretical uncertainty in the muon-electron mass ratio is 120 ppb. The contributions from parity conserving weak interactions are about an order of magnitude smaller than the $m_\mu / m_e$ uncertainty.
1.3.6.4 Atomic Anapole Moments

There also exists an atomic anapole moment. In the same way that nucleon-nucleon weak interactions admix $s$ and $p$ nuclear wave functions to give the valence nucleon a spin helix, so too do parity violating electron – nucleon interactions admix $s$ and $p$ electronic wave functions to give the valence electron a spin helix. This creates a magnetic field within the atom with the same shape as that produced by a toroidal current distribution. One interesting effect of this is to produce an energy difference between enantiomers (optical isomers). When the electron spin helix is of the same handedness as the molecular structural helix, the energy is different than when they are opposite. This results in a splitting of vibrational frequencies at a level of approximately $10^{-12}$ for $Z\sim 80$. There have been several attempts to measure this effect, with new proposals as recent as July 2008. See [22] and references therein.

1.4 Summary of Spin-Dependent Contributions

Here we summarize and unify the notation for major contributing factors to nuclear spin-dependent parity non-conservation. Figure 1-5 illustrates the Feynman diagrams of the largest factors, namely the vector-electron axial-nucleon neutral weak current and the nuclear anapole moment. Also included is the coherent contribution of the spin independent weak coupling with the usual magnetic hyperfine coupling. As previously mentioned we note the presence of the interference diagram but ignore it.
Figure 1-5: Feynman diagrams of the 3 processes contributing to NSD-PNC. (a) Tree-level $Z^0$ exchange ($V_eA_n$ term). (b) $Z^0$ and $W^\pm$ exchange within the nucleus gives rise to the nuclear anapole moment (indicated by the blue ellipse) along the nuclear spin $I$, which couples magnetically to the electron spin $\sigma_e$. (c) Coherent combined effect of $Z^0$ exchange (dominated by the $A_eV_n$ "weak charge" term) and ordinary hyperfine structure (magnetic coupling between $I$ and $\sigma_e$). Reprinted from an NSF proposal by D. DeMille.

The relativistic Hamiltonian which describes these interactions can be written as:

$$H = \frac{G_F}{\sqrt{2}} \kappa' \frac{\vec{\alpha} \cdot \vec{l}}{I} \delta^3(\vec{r}),$$

(1.19)

where $\vec{\alpha} = \gamma_0 \cdot \vec{\gamma}$ and $\kappa' = \kappa_2 + \kappa_A + \kappa_{hf}$. $\kappa_2$ is the $V_eA_n$ contribution, $\kappa_A$ is the anapole contribution, and $\kappa_{hf}$ is the NSI-hyperfine contribution. When the Hamiltonian is written as in (1.19), the coefficients are defined as:

$$\kappa_A = 1 \times 10^{-3} g_e \mu_e A^{2/3} \frac{K}{l + 1}$$

(1.20)

and
\[ \kappa_2 = \frac{1/2 - K}{I + 1} C_{2\nu} \]  

(1.21)

Here \( \mu, g, \) and \( C_2 \) depend on whether the valence nucleon is a proton or a neutron, with \( C_{2\nu} \) defined in (1.6) and \( g_\nu \) defined in Section 1.3.4. \( \kappa_2 \) is constant in atomic number, while \( \kappa_A \) scales as \( A^{2/3} \). \( g_p \) is approximately four times larger than \( g_n \).

The strength of nuclear spin-dependent parity nonconserving interactions scales roughly as \( \langle H_{NSD-PNC} \rangle \sim Z^2 \left( Z^{2/3} \kappa_A + \kappa_2 \right) \). Here the \( A^{2/3} \sim Z^{2/3} \) scaling of \( \kappa_A \) has been explicitly factored out. This scaling behavior can be seen by taking the non-relativistic limit of the Hamiltonian, \( H_{NSD} \propto (\kappa_z + \kappa_\sigma + \kappa_\gamma) \left( \vec{I} \cdot \vec{\sigma} \right) \left( \vec{\sigma} \cdot \vec{p} \right) \delta^3(\vec{r}) \). As in the spin-independent interaction, the expectation value of \( \delta^3(\vec{r}) \) contributes one factor if \( Z \) to the scaling, while the expectation value of \( \vec{p} \) contributes a second factor of \( Z \).

### 1.5 Potential Impact of Experimental Program

The potential impact of our experimental program comes from two distinct areas: we will more precisely measure the \( C_2 \) coefficients, and we will add significantly to the meson-nucleon coupling data via new anapole moment measurements. Over the full course of our experimental program we expect to be able to measure several anapole moments at the level of 20-30%. We expect to also be able to measure \( C_{2n} \) to 20% and \( C_{2p} \) to 70%.
Measuring anapole moments is important because purely hadronic weak interactions are very poorly understood; anapole moment measurements can both significantly expand the data landscape as well as improve the potential for interpreting it. The uncertainty in the nuclear theory required to relate the anapole moment measurements to the meson-nucleon couplings appears accurate to \(\sim30\%\) [12, 13, 15, 23, 24]. Thus our measurements should contribute substantially to better understanding the theory.

The \(C_2\) coefficients are the most imprecisely measured constants in the Standard Model (SM), with current errors being 70% and 300% of the SM predicted value. Our expected measurements will thus represent a substantial improvement. Our projected \(C_{2p}\) measurement uncertainty is greater than that of \(C_{2n}\) because of the larger contribution from the anapole moment in odd-proton nuclei. See (1.15) and the discussion shortly thereafter.

1.5.1 Anapole Moment Measurement Impact

The impact of the anapole moment measurements will be a dramatic increase in the hadronic PNC data landscape. A phase plot of the current parameter space with our projected impact overlaid can be found in Figure 1-6. The two largest bands are due to experiments in \(^{133}\)Cs [14] and \(^{205}\)Tl[25]. Also shown are p-p scattering experiments [26], a p-\(\alpha\) scattering experiment [27], a \(\gamma\) -ray polarization experiment in \(^{18}\)F [28], and an experiment which looks for angular asymmetry in \(^{19}\)F decay [29]. As is clearly seen, the data are inconsistent with each other. Our measurements will double the number of input data points available.
Figure 1-6: Current status of hadronic PNC parameters. The x- and y- axes correspond to particular linear combinations of effective meson-nucleon PNC couplings, as described e.g. in Ref. [16]. These two linear combinations can (along with small corrections) be used to describe nearly all data on hadronic PNC, including that from nuclear anapole moments. The range shown in the plot is the theoretical (DDH) “reasonable range”; the red dot corresponds to the theoretical “best values”. The horizontal green line swath indicates the region we expect to cover with odd proton isotopes, while the vertical line blue swath indicates the region we expect to cover with odd neutron isotopes. Figure reproduced from an NSF proposal by D. DeMille.
1.5.2 Neutral Weak Coupling Measurement Impact

The impact of the $C_2$ measurements can be seen in Figure 1-7. The two most important measurements to date are a SLAC deep inelastic scattering measurement [30] performed in 1979, and a more recent measurement by the SAMPLE experiment [31] at MIT’s Bates accelerator. The SLAC experiment measured $2C_{2u} - C_{2d}$ through electron-deuteron scattering, with an uncertainty of 300% of the SM predicted value. The SAMPLE experiment measured $C_{2u} - C_{2d}$ using electron-deuteron and electron-proton scattering with an uncertainty of 70% of the SM predicted value. Both of these uncertainties are significantly larger than the 0.1-5.0% uncertainty typical of other electroweak parameters. By making measurements in both odd-proton and odd-neutron nuclei we can dramatically reduce the uncertainty of the $C_2$ determination. Green and purple bands show anticipated uncertainties that could be achieved with our method.

1.5.3 Physics Beyond the Standard Model

It has also been suggested that precision measurement of the $C_2$ ’s can serve as a probe for physics beyond the Standard Model (BSM). Because the $C_2$ ’s are suppressed in the Standard Model, even relatively imprecise measurements showing deviation from prediction can be indicative of new physics. For example, an article by Mann et al [32] from 1995 suggests that certain types of 4-fermion contact interactions, present at the 12 TeV level, could cause a 30% deviation in $C_{2u} + C_{2d}$. However, it is unclear at this point which of the BSM theories remain not ruled out by experiment. In particular, W. Skiba points out that the impact of the LEP measurements may be significant [33].
Figure 1-7: Current status of $C_2$ parameters. The yellow dot represents the Standard Model prediction [34]. The blue band is from the 1979 SLAC e-D deep inelastic scattering experiment. The magenta band is from the SAMPLE experiment at Bates. The green and purple bands indicate the projected sensitivity of our experimental program for measurements on protons and neutrons, respectively. Figure reproduced from an NSF proposal by D. DeMille.
Appendix A: Proton Currents

As described in [1], under the approximation that the nucleon is infinitely heavy, only spin degrees of freedom remain, and one can neglect neutral weak magnetism. The components of the vector and axial-vector proton currents can then be written as:

\[
\begin{align*}
\bar{p}(x)\gamma_0 p(x) &= \delta(r), & \bar{p}(x)\gamma_1 p(x) &= 0, \\
\bar{p}(x)\gamma_0\gamma_5 p(x) &= 0, & \bar{p}(x)\gamma_5 p(x) &= -\sigma_\rho \delta(r),
\end{align*}
\]

where \(\sigma_\rho\) are the proton Pauli matrices.
Chapter 2: Experiment

2.1 Introduction

Here we explain our experimental method and give an overview of the apparatus. More detailed descriptions of both the method and apparatus as well as derivations of the signal can be found in [35, 36, 37].

Our experiment uses a Stark interference method to measure the ratio of PNC-induced transitions to Stark-induced transitions between two states in a polar molecule. This method was originally developed by D. Budker at Berkeley for use with atomic dysprosium [38], and has been adapted by our group for use with diatomic molecules. The fundamental idea is to Zeeman shift two states of opposite parity to near degeneracy, then apply a time-varying electric field to induce Stark transitions between the states. Because the states are nearly degenerate, the weak mixing between them becomes appreciable. The Stark induced population transfer will go as the square of the electric field, while the Stark-weak interference will lead to a term in the population transfer that is linear in the electric field. Thus for positive electric fields the population transfer will be different than that for negative electric fields. The difference, or asymmetry, is directly proportional to the strength of the weak interaction. The signature figure for our experiment is a mapping of this asymmetry as a function of the detuning between the states.
2.2 Apparatus and Method

Our experimental apparatus consists of four distinct regions: a molecular beam source, located in Chamber 1; a state preparation region, located in Chamber 2; an interaction region, located inside a large superconducting magnet; and a state detection region located in Chamber 3. An overview diagram can be found in Figure 2-1.

The molecules in our beam are diatomic free radicals. They have one unpaired electron, and are thus very reactive. Because of this we have to create them in a beam. We follow a procedure similar to Hinds et al [39], and the overall method is as follows: First, a mixture of argon seeded with SF$_6$ is allowed to expand into vacuum through the nozzle of a pulsed valve. Because of the rapid expansion and the pressure difference between the valve cylinder and the expansion chamber, a supersonic jet is formed and the beam becomes internally cold (T~20K). Immediately outside the pulsed valve nozzle a barium metal target is ablated by an Nd:Yag laser, causing barium atoms to be thrown up into the path of the beam. As the beam exits the nozzle the SF$_6$ interacts with the barium atoms to form our target molecule, BaF. The ablation laser pulse and the gas pulse are carefully timed so that the laser pulse arrives at the proper time within the gas pulse. This beam then proceeds through two chambers and skimmers on to the magnet and the interaction region. The source is typically operated at a repetition rate of between 10-50 Hz, and produces approximately $5 \times 10^7$ molecules in a given low-lying rotational state per pulse.

A key characteristic of the molecular beam is that it be rotationally cold. This is important because it insures that a large fraction of our molecules are in the ground state, thus giving us a larger population in our states of interest.
Figure 2-1: Overview of Experiment. Molecules are created in (a) Chamber 1, then move to (b) Chamber 2, where states are prepared through interaction with a laser. They then interact with electric and magnetic fields in region (c), and finally pass on to (d) Chamber 3, where changes to the prepared states are detected via laser-induced fluorescence.

The preparation and interaction regions of our experiment are best understood by considering two states, labeled $|A\rangle$ and $|B\rangle$. These states are hyperfine/rotational levels of the electronic ground state of the molecule, and are described in detail in 5.2. Note that the $|A\rangle$ state discussed here is distinct from the higher lying electronic state also labeled by A. The two states $|A\rangle$ and $|B\rangle$ appear initially in our beam in roughly equal thermal populations.
Figure 2-2: Experimental Process Diagram. (1): molecules are formed by laser ablation into a pulsed jet; states $|a\rangle$ and $|b\rangle$ have equal thermal populations. (2): a laser beam (green) de-populates state $|a\rangle$ by optical pumping. (3): an oscillating E-field (orange) is applied parallel to the B-field formed by the solenoid. (4): population transferred to state $|a\rangle$ by the oscillating E-field is probed by laser-induced fluorescence.

In Chamber 2 we depopulate one of the levels, $|A\rangle$, by driving transitions to a higher electronic state. Because the higher state can decay to many lower states, we can effectively deplete the entire population of $|A\rangle$. The beam then continues into a large superconducting magnet where the ~5000 G field Zeeman shifts $|A\rangle$ and $|B\rangle$ to near degeneracy. The magnet is described in 3.6. Near the center of the magnet a specially designed electrode structure produces a spatially varying electric field. Because the molecules are moving along the beam they see the spatially varying electric field as a time varying field. This field causes Stark-induced mixing between the two states,
transferring some of the population of $|B\rangle$ back into $|A\rangle$. In Chamber 3 laser induced fluorescence is used to probe the population of state $|A\rangle$. A summary diagram of this process can be found in Figure 2-2.

2.3 Signal

The signature signal of our experiment is an asymmetry in the population transfer between states $|A\rangle$ and $|B\rangle$ which depends on the sign of the electric field. This electric field, which is produced by the electrode structure placed at the center of the magnet, is given by $E = E(z)\hat{z}$, where $E(z) = E_0 \sin(2\pi N z/L)$, $N$ is an integer, $z$ is the coordinate along the beam axis, and $L$ is the length of the electric field interaction region. If the molecules are moving with velocity $v = v\hat{z}$, then in their rest frame the field appears as $E(t = z/v) = E_0 \sin(\omega t)$, where $\omega = 2\pi N v/L$. We can write the Hamiltonian, including the weak interaction, as

$$H = \begin{pmatrix} 0 & iW + dE \\ -iW + dE & \Delta \end{pmatrix}$$

(2.1)

Here $\Delta$ is the detuning from degeneracy, $iW$ is the (pure imaginary) weak matrix element, $d$ is the dipole moment of the molecule, and $E$ is the electric field described above. The wave function is written as

$$|\psi(t)\rangle = c_A(t)|A\rangle + e^{-i\omega t} c_B(t)|B\rangle.$$  

(2.2)

Using this wavefunction with the Schrödinger equation gives two coupled first order differential equations:
\[
\dot{c}_A = -ic_B (dE + iW) e^{-i\Delta t} \\
\dot{c}_B = -ic_A (dE - iW) e^{i\Omega t}
\] (2.3)

By depopulating \( |A \rangle \) in Chamber 2, we prepare the initial conditions of \( c_B (0) = 1 \) and \( c_A (0) = 0 \). By choosing appropriate experimental parameters, we can ensure that \( c_A \) is always small. Thus \( c_B \approx 1 \) and the first equation of (2.3) reduces to:

\[
\dot{c}_A = -i(dE + iW) e^{-i\Delta t}
\] (2.4)

Solving (2.4) under the assumptions that \( W \ll (dE_0, \Omega) \ll \omega \) gives us the amplitude of \( |A \rangle \):

\[
c_A(t) = -2i e^{-\frac{\Delta t}{2}} \left[ \cos \left( \frac{\Delta t}{2} \right) \frac{dE_0}{\omega} \sin^2 \left( \frac{\omega t}{2} \right) + i \sin \left( \frac{\Delta t}{2} \right) \left( \frac{W + dE_0}{\Delta} \right) \cos^2 \left( \frac{\omega t}{2} \right) \right]
\] (2.5)

We are interested in the population after the molecules have passed through the interaction region, namely at time \( t = T \equiv L / v \). It is important to note that the product \( \omega T = 2\pi \) is independent of velocity, thus all of the molecules will see the same number of oscillations of the electric field.

The laser-induced signal we collect in Chamber 3 is given by:

\[
S = N_0 |c_A(T)|^2 = 4N_0 \sin^2 \left( \frac{\Delta T}{2} \right) \left[ \left( \frac{W + dE_0}{\Delta} \right)^2 \right]
\] (2.6)

or, expanding and dropping small terms of \( O \left[ (W / \Delta)^2 \right] \),

\[
S = N_0 |c_A(T)|^2 \approx 4N_0 \sin^2 \left( \frac{\Delta T}{2} \right) \left[ 2 \frac{W}{\Delta} \frac{dE_0}{\omega} \left( \frac{dE_0}{\omega} \right)^2 \right].
\] (2.7)
Here $N_0$ is the number of molecules in the absence of both the optical pumping in Chamber 2 as well as the electric field. Note that the population transfer contains two terms: one which is independent of the weak interaction and goes as the square of the electric field, which we call the Stark-induced term, and one which is linear in both the electric field and the weak interaction, which we call the PNC-induced term. The interference between these terms, which is created by changing the sign of the electric field, is the key to measuring the weak interaction. We characterize this interference as an asymmetry.
Figure 2-3: Asymmetry vs. detuning for homogeneous magnetic field. The dispersive shape is a signature feature of our Stark interference PNC signal. The asymmetry approaches unity because effects due to non-uniform magnetic fields have not been included. The parameters used here are described in the text.

\[ \mathcal{A} = \frac{S(+E_0) - S(-E_0)}{S(+E_0) + S(-E_0)} = 2 \frac{W}{\Delta} \frac{\omega}{dE_0} \]  

(2.8)

The divergence which appears in the asymmetry when \( \Delta \to 0 \) is an artifact of having dropped higher order terms in moving from (2.6) to (2.7). If we include these terms the asymmetry becomes:

\[ \mathcal{A} = \frac{2dE_0W\Delta\omega}{(dE_0\Delta)^2 + W^2\omega^2}, \]  

(2.9)

which goes to zero as \( \Delta \) goes to zero. In practice in our experiment we will map the asymmetry as a function of the detuning. A calculated plot for reasonable experimental
values \[d = -3.0 \text{ kHz cm/V}, \quad E_0 = 1 \text{ V/cm}, \quad W = -5.2 \text{ Hz}, \quad \omega = 26 \text{ kHz}\] appears in Figure 2-3. The dipole moment \(d\) is that of the \(m_F = 5/2\) level of the ground state of \(^{137}\text{BaF}\). \(W\) is the diagonal weak matrix element for the same state. The electric field frequency \(\omega\) assumes that molecules with a velocity of \(v = 650 \text{ m/s}\) encounter \(N = 2\) periods of the electric field over an interaction region length of \(L = 5 \text{ cm}\).

2.3.1 Effects of Inhomogeneous Magnetic Field

An important effect which limits both the size of the asymmetry as well as the precision with which we can extract \(W\) is a non-uniform detuning caused by variations in the magnetic field. Under realistic experimental conditions the magnetic field will not be perfectly uniform. This has the effect of reducing \(\mathcal{A}\). One approach for evaluating this is to model the variation in detuning to be Gaussian, with width \(\Gamma\), as in

\[
f(\Delta) = \frac{e^{-[(\Delta - \Delta_0)/\sqrt{2}\Gamma]^2}}{\Gamma\sqrt{2\pi}}.
\]

(2.10)

This can then be numerically integrated against (2.9). For different effective detuning widths \(\Gamma\), the maximum asymmetry \(\mathcal{A}_{\text{max}}\) will occur at different values of the center of the detuning distribution \(\Delta_0\). The detuning which gives the maximum asymmetry, \(\Delta_0^{(\text{max})}\), is plotted as a function of \(\Gamma\) in Figure 2-4. This is relevant because for a given
Figure 2-4: $\Delta_0^{(\text{max})}$ vs. $\Gamma$. $\Gamma$ is the effective width of the detuning distribution. $\Delta_0^{(\text{max})}$ is the detuning for a given $\Gamma$ which gives the maximum asymmetry. As the field uniformity decreases, the distribution in detunings becomes larger, resulting in a larger $\Gamma$.

magnetic field uniformity, and thus distribution of detunings, it tells us where to expect the maximum asymmetry. The maximum asymmetry itself is plotted as a function of $\Gamma$ in Figure 2-5. The critical point here is that as the effective width $\Gamma$ of the detuning increases, the asymmetry decreases. Thus having as uniform a magnetic field as possible is critical to a successful measurement of $W$.

2.3.2 Uncertainty in Weak Coupling Extraction

The uncertainty in the weak coupling $W$ can be related to the uncertainty in our measurement of the asymmetry $A$. Then
Figure 2-5: $A_{\text{max}}$ vs. $\Gamma$. $\Gamma$ is the effective width of the detuning distribution. $A_{\text{max}}$ is the maximum asymmetry. As the field uniformity decreases, $\Gamma$ becomes larger, resulting in a smaller asymmetry.

$$\frac{\delta W}{W} = \frac{\delta A}{A}$$

If we rewrite $A$ as

$$A = \frac{S(+E_0) - S(-E_0)}{S(+E_0) + S(-E_0)} = \frac{S^+ - S^-}{S^+ + S^-},$$

then the uncertainties in the numerator and denominator are, in the shot noise limit,

$$\delta [S^+ - S^-] = \delta [S^+ + S^-] = \sqrt{(\sqrt{S})^2 + (\sqrt{S})^2} = \sqrt{2S}.$$

The total uncertainty in $A$ is given by
\[
\left( \frac{\delta A}{A} \right)^2 = \left( \frac{\sqrt{2S}}{(S^+ - S^-)} \right)^2 + \left( \frac{\sqrt{2S}}{(S^+ + S^-)} \right)^2 .
\]

The quantity \((S^+ + S^-)\) is large relative to the other terms, so we discard the second term above. Then we have

\[
\delta A = \frac{\sqrt{2S}}{(S^+ + S^-)} = \frac{\sqrt{2S}}{2S} = \frac{1}{\sqrt{2S}} \tag{2.11}
\]

Thus

\[
\delta W = W \frac{i}{\sqrt{2S}} \tag{2.12}
\]

In the limit of no magnetic field broadening

\[
\delta W = \Delta / \left[ 4\sqrt{2N_0} \sin \left( \frac{\Delta T}{2} \right) \right] \tag{2.13}
\]

The limit as \(\Delta \to 0\) is given by

\[
\delta W_{\text{min}} = 1 / \left( 2\sqrt{2N_0T} \right) \tag{2.14}
\]

We can include the effects of an inhomogeneous magnetic field, as described above, by integrating a Gaussian distribution in detuning (2.10) against both the asymmetry (2.9) and the signal (2.6). The results of this can be seen in Figure 2-6.
Figure 2-6: Uncertainty in $\delta W$ vs. effective detuning width. As the field becomes less uniform and the spread in detuning increases, the uncertainty in the extraction of $\delta W$ increases.

2.4 Systematic Effects

As discussed in [35] and [36], our experiment provides strong control over systematics. This is due largely to our ability to reverse several experimental parameters, each of which changes the sign of the parity violating signal. Both the systematic effects and their control are similar to those encountered in [38]. Here the largest effects were found to be due to stray DC electric fields combined with magnetic field gradients. Because of the relatively small dipole moments in diatomic free radicals, our own sensitivity to stray electric fields becomes relevant only for fields greater than about 1 mV/cm, a level which we should be able to control. A further control over systematics which is unique to our
experiment is that the ratio of $d/W$ varies widely in both magnitude and sign over the range of accessible level crossings in the molecule.

Table 2-1 shows the crossing points and their associated matrix elements for $^{137}\text{BaF}$.

**Table 2-1: B-field crossing points for $^{137}\text{BaF}$**

| B (G) | m$_F$ | $\langle A|W|B\rangle$ (Hz) | $W/d$ (mV/cm) |
|-------|-------|-----------------------------|----------------|
| 3250  | 5/2   | +4.0                        | -1.5           |
| 3550  | 3/2   | -3.0                        | +2.0           |
| 3930  | 3/2   | +1.9                        | -0.5           |
| 4330  | -1/2  | -3.5                        | +2.3           |
Chapter 3: The Magnetic Field

3.1 Introduction

The magnetic field performs the crucial role in our experiment of bringing the states of interest in our molecules to near degeneracy. Recall that our goal is to characterize the strength of parity violating interactions by looking for interference between weak mixing and externally driven Stark mixing of two particular molecular states. Specifically the figure of merit is a mapping of the asymmetry as a function of detuning, where detuning refers to the energy separation between the states (see for example Figure 2-3). A critical property of the magnetic field is its uniformity. As seen in Figure 2-4, as the variation in the field grows, the asymmetry we wish to measure is made smaller.

In order to help maintain a homogeneous field, our magnet installation includes, in addition to the main solenoidal coil, a series of correction coils known as shim coils. Each of these coils produces a specific gradient of the magnetic field, and by adjusting the current in each coil we can make the field more homogeneous. This process is known as shimming.

In this chapter we discuss the specific experimental requirements for the magnetic field. We describe the NMR spectrometer which coordinates measurements of the field, the magnet itself, the procedure for shimming the field, and finally the overall performance. A detailed description of the novel NMR probes designed to measure the field follows in
the next chapter. Much of the material in this chapter and the next is covered in [40] and
[41].

3.2 Experimental Requirements and Constraints

Here we summarize the specific experimental requirements for the magnet and shimming
system. Based on the results described in Section 2.3, we seek a target field uniformity
of 0.1 ppm. The central magnetic field value depends on the molecule of choice, and will
range from 0.1 T to 0.6 T. In order to map the PNC-induced asymmetry as a function of
detuning, we must be able to move the magnetic field around a given level crossing by
roughly ± 5 mG while maintaining the target uniformity. Finally, control of systematics
requires us to make the PNC measurement at several sets of level crossings. In order to
allow us to determine the location of these level crossings efficiently, the magnetic field
needs to remain uniform at the few ppm level while being shifted by 5 to 10 G.

Two practical constraints dictated by our experiment are that because our molecular beam
passes through the center of the magnet, we cannot include any real-time monitoring
elements along the axis of the magnet. In addition, because of the necessity to quickly
adapt to changes in field strength, and thus operating frequency, our entire control system
must be extremely broadband in nature.
3.3 Measurement System Overview

3.3.1 Introduction

We have developed an NMR-based system for mapping, monitoring, and shimming the field of our 2 T superconducting magnet. A stationary array of 32 spatially distributed NMR probes measures the field at distributed points in space. Information from these probes is processed and used to drive the magnet's correction coils, with the goal of removing any spatial or temporal inhomogeneity in the field. The process, which is discussed in Section 3.7, converges rapidly and addresses all correction coils simultaneously, thus dramatically reducing the time required to shim the field compared to conventional methods. Our system is similar to a device reported by Ntouteume et al. [42], but includes more than three times as many probes, as well as a more sophisticated shimming algorithm. Additionally, the system includes no frequency dependent components such as quarter wavelength lines, probe tuning, or impedance matching elements, thus making it inherently ultra-broadband.

3.3.2 Measurement Process

The measurement of magnetic field strength via nuclear magnetic resonance (NMR) consists of the following sequence of events. A “transmitter” produces a radiofrequency (RF) pulse known as the tipping pulse. The tipping pulse is sent to the NMR probe which induces, via the magnetic resonance phenomenon [43], a transverse magnetization of the sample. This magnetization precesses around the static magnetic field $B$. The precessing magnetization induces a signal in the probe known as the free induction decay, or FID.
The free induction decay is an exponentially decaying sinusoid, the frequency of which is related to the magnetic field strength via \( \omega = \gamma B \), where \( \gamma \) is the gyromagnetic ratio of the nuclear species being sampled, in our case protons in water. This signal is sent to a receiver where it is amplified and mixed down, then sent to a computer for digitization. The mixing is used in order to bring the RF signal from the probe down to an audio frequency, typically a few kilohertz, so that it can be easily digitized. This signal is then Fourier transformed and the field strength is computed from the fitted peak frequency.

A typical signal for the tipping pulse is tens to hundreds of volts, and a typical signal received from the probe is microvolts. The sensitive receiver electronics must therefore be protected during the transmitter pulse. This is accomplished by way of a duplexer, which blocks the receiver channel during a transmitter pulse. An additional component in our system is a multiplexer, which allows us to individually address any of our 32 probes. This collection of transmitter, receiver, duplexer, multiplexer, and signal analysis computer is referred to as the spectrometer.

We will now discuss the main elements of our measurement system individually, beginning with the array of probes, then following with the spectrometer.

### 3.4 Probe Array

Central to our measurement system is an array of NMR magnetometer probes which are distributed about the main axis of our magnet. The probes themselves are described in detail in the next chapter. Briefly, the probes act electrically as a properly terminated 50
Ohm transmission line with a bandwidth of 100 MHz or greater. They are fabricated on flexible circuit board, with electrical connections made via a U.FL standard miniature coaxial cable.

The flex circuit probes are secured with plastic cable ties around long glass tubes which contain the water samples necessary to produce an FID signal. The tubes are jacketed (inner tube OD = 5 mm, outer tube OD = 15 mm), with the inner tube containing the sample. This arrangement was chosen in order to limit the volume from which our probes could measure a signal, thus more precisely locating the position of our magnetic field measurement. The outer tube diameter remained large in order to match the diameter of our probes. The active region of protons sampled by each probe is cylindrical in shape, having a radius of 2.5 mm and a length of ~1.5 cm. Their spatial arrangement is as seen in Figure 3-1. The locations of the probes are given in Table 3-2. The entire assembly is held in place through a mechanical support structure which is bolted to the end face of the solenoidal magnet. The U.FL cable from each probe is connected to standard BNC cable via a breakout board which is mounted outside the magnet. Other than this short section of U.FL, all cables in the entire assembly are standard BNC.

A diagram showing the spatial layout of the array relative to the beam can be found in Figure 3-2.
Figure 3-1: Representation of spatial layout of probe array. The hollow green tube in the center is the molecular beam vacuum pipe and defines the z-axis. The four columns of maroon colored probes are spaced at 90 degree angles on circles of radius 3.81 cm. Their centers are separated by 3.2 cm in the z-direction, with the middle probe in the column centered at $z = 0$, the center of the magnet. The gold colored probes are placed at the same angular positions, but on a circle of radius 5.72 cm, and are located at $z = 0$. The blue colored probes are located at $z = \pm 3.2$ cm, on circles of radius 4.78 cm, with their angular locations offset from the previously discussed probes by an angle of 45 degrees.

3.5 Spectrometer

The spectrometer consists of five main parts: the computer system, transmitter, receiver, duplexer, and multiplexer. A schematic diagram is shown in Figure 3-3. A timing diagram for the control pulse sequence is shown in Figure 3-4.
3.5.1 Computer System

In the spectrometer, a computer is responsible for selecting a probe, initiating a pulse sequence, and acquiring and processing data. All computer data acquisition and control is done with standard software (LabView) and hardware (National Instruments PCI-6024E card).

Timing for a measurement sequence is initiated by the computer, which triggers a multi-channel pulse and delay generator (Quantum Composers 9518). These pulses control the

![Diagram](image.png)

Figure 3-2: Beamline diagram for magnet and Chamber 3. The center of the RT Z1 shim coil is located at (a), and the SC Z1 shim coil at (b). Note that these may shift over time. As of 11-08 both the probe array and interaction region are centered on (b). The Teflon tube holding the interaction region in place extends 12.5 cm on either side of (b).
switch which makes the RF pulse, the duplexer relay, and a blanking line to the RF power amplifier, as well as provide a trigger to the data acquisition system. Under normal operation the difference signal from the receiver is acquired, Fourier transformed, then fit using the standard Levenberg-Marquardt routine found in the LabView software. The fit parameters are then stored to disk or sent to other software for analysis.

Figure 3-3: Spectrometer schematic diagram. All lines are 50 Ω cables of arbitrary length. Several standard RF attenuators are suppressed for clarity. The reading of a single probe is initiated by software in the computer, which produces a hardware trigger sent to the pulse generator. The pulse generator controls the timing of other required triggering pulses. The sequence proceeds as described in the text.
Figure 3-4: Timing diagram. The pulse sequence is started by an external trigger coming from the computer. The duty cycle is also set by the computer. (a) opens the relay on the duplexer board. When the signal is high the probe is connected to the transmitter, when low the probe is connected to the receiver. (b) is the blanking input to the power amplifier. When this signal is low the amplifier is blocked. (c) controls the RF switch. The RF signal will be present at the amplifier for as long as (c) is high. The width of this signal is the width of the RF pulse, currently optimized for protons. (d) is the DAQ trigger. When it is high the computer acquires data.

3.5.2 Transmitter

The RF signal which comprises the tipping pulse originates in a frequency synthesizer (PTS 160, stabilized to a Stanford Research Systems PRS10 rubidium standard). This continuous wave RF signal is gated by a switch (Mini-Circuits ZYSWA-50DR) to form a
square pulse. The pulse is amplified in a power amplifier (CPC MRI Plus Model 3T1000) and sent to the probe. A typical pulse amplitude is ~60 V_{rms}, and widths are 60-300 μs.

3.5.3 Duplexer

The duplexer consists of a single electromechanical reed relay (Coto 2341) located on the same board as the multiplexer. The relay has a rated switching time of 0.5 ms and the carry current rating is 1.5 A. By using a relay we avoid the frequency dependence of the quarter wavelength lines found in more standard protection circuits. Although we have not tested the bandwidth of this relay, it performs well at 22 MHz. (Relays with similar specifications, specifically designed to act as 50 Ω transmission lines for signals with RF frequencies up to the GHz range, are available if needed.)

3.5.4 Receiver

The receiver consists of two amplifiers (Mini-Circuits ZFL-500LN and Mini-Circuits ZHL-3A) in series, followed by a mixer (Mini-Circuits ZAD-8) and a low pass filter (cutoff frequency = 190 kHz). The mixer LO input is the continuous wave RF signal from which the tipping pulse is derived, and the RF input is the free induction decay signal coming from the probe. The filtered IF output of the mixer will be referred to as the difference frequency. The frequency of the tipping pulse signal is intentionally chosen to be greater than the Larmor frequency ω of the protons in our probe sample, thus removing any sign ambiguity in the difference signal. Note that because we are not interested in the phase of our FID we do not need the quadrature detection schemes found in many NMR spectrometers.
3.5.5 Multiplexer

The multiplexer consists of a cascade of relays (identical to the duplexer relay) controlled by five TTL logic level lines from the computer. Both sides of the duplexer/multiplexer board are covered with a ground plane in order to reduce the effects of stray RF.

3.6 Magnet

Our magnet is a 2 T Oxford 85/310HR horizontal superconducting solenoidal magnet. The main superconducting coil is designed to produce a large and nominally homogeneous field in a region surrounding the center of the magnet. In addition, however, the magnet contains a number of “shim coils”, which are designed deliberately to create field gradients. These coils can be used to shim out residual field inhomogeneities by applying the correct field gradients to cancel imperfections in the main coil. Our magnet contains two sets of shim coils, a superconducting set and a room temperature set. Each coil in a set is designed to produce a correcting gradient corresponding in shape to a spherical harmonic. A list of the coils that each of our shim sets contains can be found in Table 3-1. Our magnet also requires regular filling with liquid helium and liquid nitrogen. The specific procedures can be found in [44].

3.7 Shimming

There are many approaches to shimming, differing both in the way the field is measured and in how the proper correction currents are decided upon. Each has its advantages and disadvantages, and different methods are appropriate for differing situations. A
discussion of a recent approach can be found in Michal [45], and of earlier approaches in references therein.

Once field data is acquired from the probe array we must convert it into an appropriate set of currents to be applied to our shim coils. Here we explain our basic strategy, first giving a conceptual overview then proceeding to the details. Note that in our installation there are both superconducting (SC) and room temperature (RT) shim coils. Although the analysis is similar for both sets of coils, their currents are adjusted independently.

The axial ($z$-) component of our magnetic field, $b(\vec{r}) = B_z(\vec{r})$, can be expressed as an expansion in spherical harmonics: [46]

$$b(\vec{r}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} A_{lm} r^l G_l^m(\theta, \phi)$$

(3.1)

where

$$G_l^m(\theta, \phi) = \frac{Y_l^m + (-1)^m \text{sign}(m)Y_l^{-m}}{2\sqrt{\text{sign}(m)}}$$

(3.2)

is a purely real-valued function. This can be rewritten more compactly as:

$$b(\vec{r}) = \sum_{k=0}^{\infty} c_k f_k(\vec{r})$$

(3.3)

where the $f_k$ are given by

$$f_k = r^l G_l^m(\theta, \phi)$$

(3.4)

where $k = l^2 + l + m$. To obtain $l$ and $m$ from $k$ use $l = \text{IntegerPart}(\sqrt{k})$ and $m = k - l^2 - l$.

We refer to the $f_k$ as "ideal" basis functions.
Note that although the NMR probes measure the total field magnitude $B$ rather than the $z$-component, the field from our magnet is sufficiently homogeneous that the transverse components give negligible contributions to $B$ over the volume of interest. Hence we take $b = B$ throughout our analysis. If all terms except the first, constant, term ($k = 0$) in the expansion are eliminated, then the field will be homogeneous everywhere.

The general goal of shimming then is to create fields with our shim coils which are equal in magnitude but opposite in sign to the non-constant ($k > 0$) terms in the expansion of Eqn. (3.3). Nominally this should be straightforward, because each of our $N_s = 15$ room temperature shim coils is designed to produce a field proportional to a given ideal basis function [47]. That is, ideally each shim coil would produce a field corresponding to a single term in (3.3). In practice, however, the shim coils do not correspond exactly to the ideal basis functions. Instead, each coil $j$ produces a field given by a linear combination of these functions. While one can rewrite the expansion of the magnetic field in terms of these so called “coil functions,” we have chosen to take a more direct approach.

The basic principle of our technique is to first measure offline the change in magnetic field produced at each of our $N_p = 32$ probe locations $\vec{r}_i$ ($i = 1 - N_p$) per unit change in current on each shim coil $j$ ($j = 1 - N_s$). This gives us an $N_s \times N_p$ matrix $\vec{G}$, whose elements are defined by

$$g_{ij} = \frac{b_j^*(\vec{r}_i) - b_j^*(\vec{r}_i)}{2I_0} \quad (3.5)$$

Here $b_j^*(\vec{r}_i)$ is the magnetic field at the probe position $\vec{r}_i$ with the current $I_j$ in coil $j$ set
to $I_j^\pm = \pm I_0$. Typically we use $I_0 = 0.1A$. Measuring this matrix can take as little as ten minutes if the field is already relatively homogeneous and the signal to noise on each probe is high. If the field is not homogeneous, for example in a new installation, and signals from the probes must be averaged for longer times, then this process can take several hours. As described below, there is a more accurate way of measuring the matrix which involves fitting time series data from the magnetic field. In practice this method takes around six hours.

Having measured the $\tilde{g}$ matrix, we shim the magnetic field by measuring the field values at each point in the array $B_i = B(\vec{r}_i)$. We then calculate the desired change in field value at each point in the array $\Delta B_i = B_{\text{goal}} - B_i$, where $B_{\text{goal}}$ is our target magnetic field. These $\Delta B_i$ form a $1 \times N_p$ vector $\vec{d}$ of the desired change in magnetic field at each probe location. We can now write the following matrix equation:

$$\vec{d} = \tilde{g} \vec{I}$$

(3.6)

where $\vec{I}$ is the $1 \times N_s$ vector of required change in shim currents.

The shimming process then reduces to solving equation (3.6) for the shim currents $\vec{I}$. Because $N_s \leq N_p$, our system is overconstrained; we use a least-squares minimization to find the optimal currents. In practice, if the initial inhomogeneities are large, achieving the desired homogeneity requires several iterations of the shimming procedure.
3.8 Performance

3.8.1 Overview

The performance of the magnet system can be broken down into three distinct categories. The first is how uniform a field the shimming system is able to create. The second is how much the uniformity changes when we move the field, and the third is how much the uniformity changes due to sources beyond our control.

3.8.2 Shimming Performance

In our experiment the volume of interest for our magnetic field is a cylinder, centered on the magnet, of radius 2.5 mm and length 5 cm along the magnet axis. This volume corresponds roughly to the volume subtended by the molecular beam, while it is subjected to the electric fields of the interaction region. In order to determine the results of our shimming procedure, we directly map the field along this axis by moving an additional probe, identical to the ones used in our array, along the axis of the magnet. A plot of the field in this region, as measured by this probe, is shown in Figure 3-5 for the magnet both before and after shimming. The evolution from original to shimmed data here is the result of five iterations, which took around fifteen minutes. The time required
Figure 3-5: The magnetic field along the z-axis before and after shimming. The value is relative to the field at $z = -0.5$ cm. Each point is the average of five measurements, with a typical standard error on the mean of 2.3 $\mu$G. At each point, the probe sample averages over a cylindrical volume of 2.5 mm radius and $\sim$15 mm length coaxial with the magnet, and centered on the z-position shown for each data point. For our region of interest between $z = \pm 2.5$ cm, the maximum variation in the proton frequency before shimming is 71 mG, or 14.2 ppm. The maximum variation after shimming is 1.82 mG, or 0.36 ppm. A narrower range plot which shows the variation on axis in greater detail can be found in Figure 3-6.

to shim is dominated by the amount of time we must wait for the field to settle after changing the current in a shim coil. Starting from zero current in the room temperature coils, the first shimming pass often produces shim current changes of several hundred milliamps, corresponding to field shifts on the order of 175 mG, which require a
Figure 3-6: Magnetic field on axis after shimming, May 2008. The field is relative to the value at $z = -0.5$ cm. The maximum variation is 1.81 mG, or 0.36 ppm.

settling time of around five minutes to reach <1 ppm stability. On subsequent passes, where field shifts are smaller, a typical wait time is 90 seconds.

Distinct areas which we considered in attempts to improve these results are both making more precise determinations of the matrix elements required in the shimming process, as well as adjusting the number of constraints used when solving equation (3.6) for the shim currents. In the first case we improved on the method described above by frequently sampling the magnetic field at a given probe location before, during and after applying a
change in current to a given coil. This gave us the magnetic field as a function of time
during a given applied change in current. We then fit this function and extracted values
for \( \left( \frac{dB}{dl} \right) \) which were statistically more precise than those given by a single point
measurement. The \( g \) matrix given by this more painstaking method however produced
little improvement on the final field homogeneity. This method was extended by making
three such \( g \) matrices and averaging them to form a composite \( g \) matrix. During this
process any spurious data points were thrown out. This composite \( g \) matrix also yielded
marginal improvement.

Our second focus for improvement involved varying the number of constraints used in
solving equation (3.6), which yielded no improvement. A final avenue we explored was
employing different weights for the probes used in the fitting routine. Specifically, in
addition to a constant weighting scheme as described above, we tried weighting each
probe’s reported field value by the variance in the individual measurements which made
it up. This also made no quantitative difference in the final field homogeneity. Further
attempts at improvement, along with the results of other studies which may be of interest,
are described in 3.9.
3.8.3 Changes in Uniformity Due to Controlled Field Movement

One of our experimental requirements is that the magnetic field remain substantially homogeneous as we sweep the detuning across a level crossing. We achieve this field sweep by changing the current on the room temperature Z0 coil. In practice we will wish to move the detuning by up to perhaps ±10 kHz, which roughly corresponds to moving the magnetic field by ±5 mG. As can be seen in Figure 3-7, the maximum variation of the field on axis for any given central field value over a range of 2.5 G is 0.33 ppm. In other words, after optimizing the field uniformity for a given central field value, we were able to move that central field value over a range of 2.5 G and only affect the uniformity at the 0.33 ppm level. This easily meets the uniformity required by the experiment over the much smaller ±5 mG tuning range required for sweeping.

3.9 Additional Studies

Here we describe distinct areas which we considered in attempts to better understand our measurement system as well as improve the shimming results in general. It is hoped that these will prove useful to subsequent graduate students. The studies in this section are somewhat of an amalgam, and are presented in no particular order. Many do not depend on others.

3.9.1 Measurement Uncertainties

In order to characterize the measurement uncertainty associated with reading the array of probes, we took ten array readings in a row and evaluated the range of values we obtained. This study was taken using a well shimmed field, and required a total of 3
Figure 3-7: Field uniformity on axis at various central field values. The magnetic field is relative to the value at $z = 0.5$ cm. This central field value was changed by applying 1 V shifts to the RT Z1 coil. In our region of interest, $z = \pm 2.5$ cm, the maximum variation for any central field value is 1.67 mG, or 0.33 ppm. Prior to this series of measurements the uniformity on axis was optimized by hand to a maximum variation of 0.23 mG, or 0.05 ppm, in our region of interest. Varying the central field value over a 2.5 G range only changes the field uniformity by 0.33 ppm.

minutes. The results are seen in Figure 3-8 and Figure 3-9. The average standard deviation of an individual probe’s measurements was 0.32 Hz, or 16 ppb. The average range of an individual probe’s measurements was 1.01 Hz, or 50 ppb. The largest range
observed was a single outlier at 2 Hz, or 0.1 ppm. The probes consistently measure the field with a precision better than our required 0.1 ppm.

Figure 3-8: Standard deviations of ten measurements of the same probe, for each of the 32 probes in the array. The standard error would be $1/\sqrt{10}$ of these values.

Figure 3-9: Maximum range of ten measurements of the same probe, for each of the 32 probes in the array.
3.9.2 Weighted versus Unweighted Shimming

We compared two weighting schemes for the shimming algorithm. Typically when measuring the magnetic field several FID's are acquired, Fourier transformed, and fit. The average of the fitted values is then reported as the measurement. There is of course uncertainty associated with this measurement. Under the unweighted routine, when solving (3.6) for the correction currents we did not account for any uncertainty in the individual probe measurements which make up $\tilde{d}$. Under the weighted routine, each probe measurement $B_i = B(\tilde{r}_i)$ was weighted by $1/\sigma_i^2$, where $\sigma_i$ is the standard deviation of the individual fit values which are averaged to form the measurement. As seen in Figure 3-10, the final shimming results obtained through both of these methods are practically identical. A caveat to this is seen in Figure 3-11, however. Here you see the weighted and unweighted routine results starting from two different sets of initial conditions: zero voltage on the room temperature shim coils, and random voltages on the room temperature shim coils. In this case the unweighted routine produced identical results, while the weighted routine starting from random voltages produced results which differ significantly at the 0.1 ppm level. This may well be a spurious result however, and the author suggests repeating the study.
Figure 3-10: Shimming results on axis using weighted and unweighted routines. The magnetic field is measured relative to the value at $z = -0.5$ cm. Both routines produce essentially the same result. The two data points at every $z$ position in each series are the result of moving the probe first in the direction of decreasing $z$, then returning in the direction of increasing $z$. 

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Figure 3-11: Shimming results on axis using weighted and unweighted routines starting from both zero volts on RT shims and random voltages on RT shims. The unweighted routine produces a magnetic field with a variation of 0.5 ppm. The weighted routine starting from zero volts yields a similar variation to the unweighted routines. The weighted routine starting from random voltages produces a field with uniformity of 0.82 ppm.
3.9.3 Previous Shimming Routine vs. Current Routine

The shimming routine described in this thesis, colloquially known as the dBdV routine, has been found to yield superior results to an earlier more complicated routine (Figure 3-12). An important point to note here is that in this study both routines were compared starting from the same superconducting shim currents. As will be discussed soon, in 2006 superior automated shimming results were obtained despite using the older shimming routine. We suspect that this may be due to the field being more homogeneous.

![Graph showing shimming results](image)

**Figure 3-12:** Shimming results on axis using the older (DIT) method and using the current (dBdV) method. The newer routines outperform the older routine. These data were taken on 4-21-08.
to begin with in 2006.

3.9.4 2006 vs. 2008

Here we compare the difference between the shimming results in December 2006 and in May 2008. Despite having demonstrably improved the shimming algorithm (see 3.9.3), the post-shimming field uniformity on axis was better in 2006. The results of both are shown in Figure 3-13. This is somewhat surprising due to the 2008 shimming technology being both faster as well as being based on a more accurate characterization of how the

![Graph showing magnetic field comparison between 05/2008 and 12/2006](image)

**Figure 3-13:** Magnetic field on axis after shimming in 2006 vs. 2008. The uniformity is about a factor of five worse in 2008.
room temperature shim coils change the field. One critical difference however is that the initial field produced by the main solenoid and the superconducting shim coils was substantially more uniform in 2006 than in 2008. This can be seen in Figure 3-14. In 2006 the SC shims had been carefully tuned by hand during the magnet installation, whereas by 2008 we had moved the main field several times, and while we scaled the SC currents by the same factor as the main current, it is clear that the resulting uniformity was poorer. The ratio of uniformity on axis before and after shimming was ~39 in 2006 and ~35 in 2008, however, so the improvement due to shimming was better in 2006. One reason that less effort has been focused on the SC shims is that they presently cannot be controlled by computer. It is clear from this study however that while the SC shims may not be the sole source of our change in shimming performance, achieving a better uniformity from the SC coils alone is an important target.

A related point is that shimming automatically then optimizing the field on axis manually with the RT coils may prove of comparable time efficiency to shimming manually with the SC coils. In May 2008 we shammed using the automated routine, then made manual adjustments to the currents to optimize the field on axis. Results are seen in Figure 3-15. The manual adjustment process took around 1.5 hrs. Recall that in 3.8.3 we found that once the desired field uniformity is attained it will remain uniform over a broad range of field shifts. In particular, for a given level crossing, once a uniform field is obtained, whether it is obtained with the RT or SC shims, shifting the field through the range of values necessary to map the asymmetry vs. detuning curve should not change the field uniformity at a level relevant to the experiment.
Figure 3-14: Uniformity on axis before shimming in 2006 vs. 2008. The variation is about a factor of six worse in 2008.

Figure 3-15: Results of shimming first automatically and then by hand in 2008. 2006 data are included for reference.
3.9.5 Array-Axis Uniformity Puzzle

One surprising aspect of the shimming algorithm performance is that when the field, as measured at the $N_p$ points of the array, is shinned to a minimum in variation, the variation in the field along the axis is not necessarily minimized. It also appears that the converse is true: when the variation in field along the axis is minimized, the variation in field over the array is not at a minimum. We see evidence for this through the following experiment: starting from zero current in the room temperature shim coils, we iterated five times through the standard shimming routine. We then measured the field on the axis as a function of $z$, the axial coordinate. The average of the four innermost array probes at a given $z$, as well as the field on the axis itself, can be found in Figure 3-16. A clear linear gradient along $z$ can be seen. We next removed this gradient by manually applying extra current to the Z1 room temperature correction coil. As can be seen in Figure 3-17, this was effective in removing the $z$ gradient along the axis. The variation over our 5 cm region of interest along the axis was reduced from $\sim0.5$ ppm to $\sim0.03$ ppm. However, the variation in the average of the innermost set of array probes was increased by nearly a factor of three. As is seen in Figure 3-18 and Figure 3-19, there is not a clear correlation of this change with probes at a given radial location. We thus learn that the shimming algorithm, while appearing to minimize the variation over the array, does not minimize the variation along the axis. Furthermore, we observe that when we manually correct residual gradients along the axis, this moves the field as measured at the array away from a minimum. When performing this experiment we used the most modern
Figure 3-16: Magnetic field after automatic shimming. Field is shown on axis and averaged over four innermost probes of the array. The field is relative to the value on axis at $z = -0.5$ cm. The uniformity on axis is $\sim 0.5$ ppm. Compare with Figure 3-17.

Figure 3-17: Magnetic field after automatic shimming followed by manual optimization. Compare with Figure 3-16. Field is shown on axis and averaged over four innermost probes of the array. The field is relative to the value on axis at $z = -0.5$ cm. After the manual optimization, the uniformity on axis increases to 0.03 ppm, but the uniformity on the array drops by $\sim 3$. 

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Figure 3-18: Magnetic field after automatic shimming. Field is shown on axis as well as the innermost probes in the array. These probes are located at \((-x_0, 0), (+x_0, 0), (0, -y_0)\) and \((0, +y_0)\), where \(x_0, y_0 = \pm 3.81\, \text{cm}\). \(z\) is along the beam. The field is relative to the value on axis at \(z = -0.5\, \text{cm}\).

Figure 3-19: Magnetic field after automatic shimming followed by manual optimization. Probe locations are the same as Figure 3-18.
incarnation of the shimming routine, and were careful to wait long enough for the field to settle while shimming. A simple explanation of this result remains to be discovered

3.9.6 X and Y Magnetic Field Gradients

It is important to quantify the magnetic field gradient along dimensions perpendicular to the molecular beam, as well as along the beam. A complicating factor is that the volume of the proton sample inside the probe is the same diameter (5 mm) as our molecular beam. The most direct approach to measuring these perpendicular gradients would be to make use of the width of the Fourier transformed NMR signal. However, this includes a convolution of the RF excitation field with the field produced by the magnet, which complicates interpretation. Furthermore, a study performed in May 2008 using pure water showed that removing a 0.6 mG/cm x-gradient, in the presence of negligible y-gradient, produced no significant change in the FFT width. A simpler approach, which we employ here, is to look at the gradient in the x- and y- directions over larger distance scales, then infer the gradient across our beam. This relies on there being no magnetic fields which oscillate on a length scale smaller than the one we use to measure the gradient. From Figure 3-20, we see a maximum gradient between probes at $\pm r = 3.81$ cm to be 0.6 mG/cm along the x- direction, and 0.86 mG/cm along the y-direction. For a 5 mm diameter molecular beam this would correspond to variations in field uniformity across the beam at the 0.12 ppm and 0.17 ppm levels. However, if we restrict ourselves to the 5 cm long region centered on $z = 0$ where we expect there to be an appreciable electric field, the variation is 0.1 ppm along x, and 0.02 ppm along y. The main purpose of including this discussion is to illustrate the measurement technique, as
Figure 3-20: Magnetic field profile as measured by probes in the array nearest the molecular beam. Large x- gradients are clearly present. Probes are located at \((-x_0, 0)\), \((+x_0, 0)\), \((0, -y_0)\) and \((0, +y_0)\), where \(x_0, y_0 = \pm 3.81\) cm. \(z\) is along the beam. Fields are relative to the probe at \((-x_0, 0)\), with \(z = -1.0\) cm.

well as to highlight that the gradients along the x- and y- directions can be important at the 0.1 ppm level.

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3.9.7 Automated Tuning Range

Being able to scan the magnetic field in a completely automated fashion is of strong interest to our experiment. While the central field value can be controlled via computer, as of Winter 2008 the ability to control the proton tipping pulse frequency via computer has not yet been implemented. It is important therefore to know how far we can move the magnetic field before losing the proton resonance in our NMR probes. By changing the combination of pulse width and excitation frequency, we maximized the total range of magnetic fields accessible without tuning the tipping pulse frequency. The resulting

![Figure 3-21: FFT amplitude from probe #2 vs. RT Z0 voltage. The RF pulse time is 20 µs, and the excitation frequency is 19.726500 MHz. The 4 V change on Z0 results in a field shift of ~2 G (see Figure 3-22). This combination gave the largest range of magnetic field strengths we could scan without losing the proton resonance in the NMR probes. If the field is shifted by a larger amount, the excitation frequency will need to be adjusted.](image)
maximum range is seen in Figure 3-21, where we plot the amplitude of the Fourier
transform of a probe signal as a function of the voltage applied to the room temperature
Z0 coil. The voltage applied to the coil is the more useful laboratory unit; a
characterization of the magnetic field as a function of this voltage appears in Figure 3-22.

Figure 3-22: Magnetic field vs. RT Z0 voltage. The field was measured at probe
#2, which is located near the center of the magnet (see Table 3-2). A linear fit
gives a slope of $651 \pm 1\text{ mG/V}$
3.9.8 Iterative Shimming

In the shimming scheme described so far, all of the correction currents are both solved for and applied in the same step. This is a dramatic departure from traditional shimming routines, which adjust one coil at a time. To investigate the potential for improvement via eschewing the simultaneous change strategy, we tried several variations on iterative shimming.

Shim coils can be classified in two ways, either by the order of the coordinate in the formula describing their effects (e.g. \( Z2 = 2z^2 - (x^2 + y^2) \) would be a second order coil), or by the degree to which they “interact” with other shims. In this case “interaction” refers to change induced in the optimal setting of one shim coil due to changes in the setting of another. For example, if the current in the Z1 coil has been adjusted to maximize the field uniformity, changing the current in the ZX shim coil will produce a gradient which may require the Z1 coil to be re-optimized. The literature unfortunately often calls the degree to which a shim interacts with other shims its “order”. It is also sometimes called the “interaction type.” However, here we adopt the convention of labeling this the shim coils’ “degree.” A table of shim coils with their equations, orders and degrees can be found in Table 3-1. The purpose of giving shim coils a degree is to characterize to what extent a given coil can be adjusted without expecting to need to readjust other coils. This is important in traditional shimming scenarios where coils are optimized one at a time. Zero degree shim coils can be adjusted independently to maximize uniformity. First degree coils can be adjusted by maximizing one, the readjusting the next first degree coil, then the next, etc. In a traditional setup, the
shimming operator manually loops through all of the first degree coils until the field no longer improves. With second degree coils, the operator adjusts the coil in question, then re-optimizes all of the first degree coils (which may require several iterations). If the result is better than before, the operator again moves the second degree coil, and in the same direction. If the result is worse, the operator moves the second degree coil in the opposite direction. The process is similar in theory to a beam walk in optics, but with multiple mirrors. It is easily understood how second degree coils in particular can be quite time consuming to optimize.

Because adjusting each shim coil individually is such a common practice within the magnet community, and is so distinct from our own approach of adjusting all coils simultaneously, we were interested in testing the results of the more traditional method. Our attempts at automating iterative shimming using our array however met with lackluster success. Unidentified software glitches resulted in frequent crashes. To date a complete optimization of the field moving through the coils by degree has not been successfully performed. However, iterative shimming with the coils by order has been achieved. The sequence followed was three passes of the following: three passes of zero order and first order simultaneously, then three passes of first order and second order simultaneously, then three passes of all higher order shims simultaneously. The entire process took about 1.5 hours, and the resulting uniformity was 0.5 ppm over a 5 cm range on axis. In other words, it took 4.5 times longer than our non-interactive shimming procedure, and produced a field on axis with almost double the variation. While it is
possible that more careful iterative shimming will yield a better uniformity on axis, we do not identify this as an area of future experimental interest.

3.9.9 Probe on Axis Study

One way to study the effects of possible sources of field perturbation on the array as a whole is to use a disconnected probe as a proxy for an unknown source of localized magnetic field. In this study we placed a probe (with the cable terminated) on the axis of the magnet, and compared array readouts both with and without the axis probe. The results are seen in Figure 3-23. Because this is the difference in the average of five readings without the probe and five readings with the probe, it seems reasonable to conclude that the axis probe may produce an effect on the array reading. However, if this effect is real it is at the 25 ppb level, thus not relevant on our experimental scale.

Figure 3-23: Difference between array reading without a dummy probe on axis and with such a probe on axis. Five measurements were taken in each state, alternating states between measurements. Probe positions are given in Table 3-2. For reference 0.5 mG = 0.1 ppm.
3.9.10 Sources of Perturbation to Individual Probe Measurements

We now shift our focus from studies performed with the goal of bettering the uniformity achieved by the shimming algorithm to isolating sources of field shift affecting an individual probe.

The probes in the array have no space between them in the z-direction. In order to ascertain whether or not the presence of one probe was affecting the measurements made by a neighboring probe, we recreated on axis the physical layout found in the array. Measurements taken both with and without a neighboring probe showed no statistical difference at the 50 ppb level.

A second study we performed was to investigate any possible effects due to the small cap which seals the sample holder for the probe we translate down the axis. Measurements were taken at the same location in the magnet, but at differing distances from the end cap. This was achieved by securing the probe at different locations on the sample tube. Again, no statistically relevant effect at the 50 ppb level was seen.

A variety of other possible sources of field perturbation were studied by using the probe on axis, and measuring the field with a perturbative source present and again without it. The most notable unanticipated effect came from the cable ties which secure the probes’ flex circuits to the water sample tubes in the array. These produced shifts in the range of 0.050 – 0.075 ppm, the magnitude and direction of which depended on the location of the tie head. After discovering this, the axial probe’s cable ties were replaced with
symmetrically wrapped electrical tape. This reduced the width of the signal by a factor of three (both measurements were made with pure water). While removing the cable ties did shift the field measurements by a constant amount, it did not make a significant difference in the uniformity of frequency readings along the axis. Worth noting is that the probes in the array are still secured with cable ties. Other noteworthy effects were due to a large wad of electrical tape placed asymmetrically on one side of the probe, which caused shifts at the 0.17 – 0.2 ppm level. This is somewhat surprising because the amount of material present in electrical tape, even a large wad of it, is relatively small.

3.9.11 Effects Due to the Interaction Region

Here we measured the field on axis both with and without the interaction region flex circuit, and with and without the long Teflon tubes which hold it. The difference measured on the array on 3/11/08 can be seen in Figure 3-25, and the difference measured on axis from 07/28/08 can be seen in Figure 3-24. An important point is that in both sets of data, the time between measuring the field with the Teflon (and interaction region) in and measuring it with nothing in was several hours. Variations in the magnetic field due to external unknown sources have been observed on this level over this time scale. This could result in either an artificial enhancement or suppression of any effect due to the interaction region. However, while additional measurements of the possible effects due to the Teflon and interaction region should be performed, it appears that this is not a likely cause of significant uniformity degradation.
Figure 3-25: Difference between precessional frequencies measured on 3-11-08 with and without the Teflon interaction region holding tube in the magnet. The data shown is the difference in the average of three array measurements taken in each state. The three measurements with the Teflon in were taken two hours before the measurements with it out. For reference 0.5 mG = 0.1 ppm.

Figure 3-24: Measurements of precessional frequency on axis with the Teflon holding tube and interaction region both present and absent. Time between data series is approximately five hours. For reference, 2 Hz in the precessional frequency corresponds to 0.1 ppm in the magnetic field.
## Appendix A: Shim and Probe Tables

Table 3-1: Room temperature shim coils. Our superconducting set contains shims Z1, X, Y, ZX, ZY, XY, and X2-Y2. Z2 is notably absent.

<table>
<thead>
<tr>
<th>Shim</th>
<th>Equation</th>
<th>Degree (Interaction Order)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Z0</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>Z1</td>
<td>(z)</td>
<td>1</td>
</tr>
<tr>
<td>Z2</td>
<td>(2z^2 - (x^2 + y^2))</td>
<td>1</td>
</tr>
<tr>
<td>Z3</td>
<td>(z(2z^2 - 3(x^2 + y^2)))</td>
<td>2</td>
</tr>
<tr>
<td>Z4</td>
<td>(8z^2(z^2 - 3(x^2 + y^2)) + 3(x^2 + y^2)^2)</td>
<td>2</td>
</tr>
<tr>
<td>X</td>
<td>(x)</td>
<td>0</td>
</tr>
<tr>
<td>Y</td>
<td>(y)</td>
<td>0</td>
</tr>
<tr>
<td>ZX</td>
<td>(zx)</td>
<td>2</td>
</tr>
<tr>
<td>ZY</td>
<td>(zy)</td>
<td>2</td>
</tr>
<tr>
<td>X2-Y2</td>
<td>(x^2 - y^2)</td>
<td>1</td>
</tr>
<tr>
<td>XY</td>
<td>(xy)</td>
<td>1</td>
</tr>
<tr>
<td>Z2X</td>
<td>(x(4z^2 - (x^2 + y^2)))</td>
<td>2</td>
</tr>
<tr>
<td>Z2Y</td>
<td>(y(4z^2 - (x^2 + y^2)))</td>
<td>2</td>
</tr>
<tr>
<td>ZXY</td>
<td>(zxy)</td>
<td>2</td>
</tr>
<tr>
<td>Z(X2-Y2)</td>
<td>(z(x^2 - y^2))</td>
<td>2</td>
</tr>
</tbody>
</table>
Table 3-2: Probe positions in cylindrical coordinates as of 03-03-08. The array is centered on the superconducting Z1 center, while the coordinate system is centered on the room temperature Z1 shim center, which at present is offset by 1 cm from the SC Z1 shim center. This is why the probes at position 3 are at \( z = 1 \) cm instead of \( z = 0 \) cm. Phi is from the x-axis, theta is from the z-axis. \( z \) increases in the direction of the molecular beam. Angular coordinates are in degrees.

<table>
<thead>
<tr>
<th>Probe #</th>
<th>Rho</th>
<th>Phi</th>
<th>z</th>
</tr>
</thead>
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<tr>
<td>0</td>
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<td>180</td>
<td>5.4</td>
</tr>
<tr>
<td>1</td>
<td>3.81</td>
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</tr>
<tr>
<td>3</td>
<td>3.81</td>
<td>180</td>
<td>-4.2</td>
</tr>
<tr>
<td>4</td>
<td>3.81</td>
<td>180</td>
<td>-7.4</td>
</tr>
<tr>
<td>5</td>
<td>3.81</td>
<td>90</td>
<td>5.4</td>
</tr>
<tr>
<td>6</td>
<td>3.81</td>
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<td>90</td>
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<td>0</td>
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</tr>
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<td>0</td>
<td>-1</td>
</tr>
<tr>
<td>13</td>
<td>3.81</td>
<td>0</td>
<td>-4.2</td>
</tr>
<tr>
<td>14</td>
<td>3.81</td>
<td>0</td>
<td>-7.4</td>
</tr>
<tr>
<td>15</td>
<td>3.81</td>
<td>270</td>
<td>5.4</td>
</tr>
<tr>
<td>16</td>
<td>3.81</td>
<td>270</td>
<td>2.2</td>
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<td>3.81</td>
<td>270</td>
<td>-1</td>
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<td>19</td>
<td>3.81</td>
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<td>-7.4</td>
</tr>
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<td>20</td>
<td>5.72</td>
<td>180</td>
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</tr>
<tr>
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<td>5.72</td>
<td>90</td>
<td>-1</td>
</tr>
<tr>
<td>22</td>
<td>5.72</td>
<td>0</td>
<td>-1</td>
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<tr>
<td>23</td>
<td>5.72</td>
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<td>-1</td>
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</tr>
<tr>
<td>31</td>
<td>4.76</td>
<td>225</td>
<td>-4.2</td>
</tr>
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</table>
Chapter 4: NMR Probes

4.1 Overview

Here we describe the novel nuclear magnetic resonance probes developed for our experiment. The probes measure the precessional frequency of protons in our magnetic field, allowing us to characterize the strength of the magnetic field at a given location. The probes are unique because unlike conventional NMR probes, which use tuned resonant circuits, these probes behave like transmission lines. As such they are able to operate over an extremely large bandwidth, and thus magnetic field range, without any tuning intervention by the user. Here we describe the important aspects of probe design, construction, and operation. The material presented here was originally published in [48].

4.2 Abstract

We have developed an easy to construct, non-resonant wideband NMR probe. The probe is of the saddle coil geometry and is designed such that the coil itself forms a transmission line. The probe thus requires no tuning or matching elements. We use the probe with a spectrometer whose duplexer circuitry employs a simple RF switch instead of the more common $\lambda/4$ lines, so the entire probe and spectrometer perform in an essentially frequency-independent manner. Despite being designed with electro- and magnetostatic formulas, the probe performs well at frequencies up to 150 MHz and beyond. We expect that with additional design effort, the probe could be modified for
use at significantly higher frequencies. Because our construction method relies on commercial circuit fabrication techniques, identical probes can be easily and accurately produced.

4.3 Introduction

Most nuclear magnetic resonance measurements use resonant tuned probes for generation and detection of radiofrequency (RF) magnetic fields. However, a variety of applications require probes capable of wideband operation. Here we present the design for such a probe, measure its performance, and describe our experiences of constructing probes of this type.

The design of our probes has been driven by a specific experiment underway in our group, with the goal to study electroweak interactions inside molecules. This experiment requires the ability to shim and monitor a magnetic field in the range 0.1-0.6 T, to a homogeneity of 0.1 ppm, with rapid adjustments to large changes in the field strength. In order to do so, we will use an array of thirty-two spatially distributed NMR probes to provide a precise real-time map of the magnetic field. Because we will not have physical access to the probes in order to tune them as we change the field, we need a probe which is non-resonant and automatically impedance matched for a broad range of frequencies. Although we are using the probe for this one particular application (as a precision NMR teslameter), the basic design appears to lend itself to other potential applications. We discuss several possibilities near the end of this article.
4.4 Discussion

4.4.1 Principles

Our probe is a saddle coil designed such that the coil itself constitutes a transmission line. A saddle coil is similar to a Helmholtz coil, but with the conductors constrained to lie on the surface of a cylinder [49]. Unlike conventionally tuned probes, which rely on matching networks to impedance match the probe to the spectrometer, a properly terminated transmission line probe presents a constant impedance to the spectrometer, independent of RF frequency. Furthermore, as it contains no resonant circuit, a transmission line probe is inherently equally sensitive to all NMR signals within its bandwidth, and requires no tuning to move between frequencies of interest. These two characteristics make transmission line probes fundamentally different from other types of NMR probes which employ transmission lines. In solid state NMR for example, there are many designs for multiply tuned cross-polarization magic angle spinning probes which use transmission lines as tuning and matching elements [50]. In our design, however, the probe itself is a transmission line: no tuning or matching is required. Note that this is also distinct from the situation in which a transmission line with a carefully chosen electrical length is attached to a probe in order to move the tuning and matching elements farther from the coil.

The basics of transmission lines can be found in standard RF engineering texts, e.g. Pozar [51]. A key feature to recall in the context of this work is that the characteristic impedance of a transmission line is determined purely by its geometry and by the
materials involved in its construction. When terminated with a load which matches its characteristic impedance, no reflections will appear on the line. In a loss-free transmission line with uniformly distributed inductance and capacitance, the impedance is independent of frequency and is given by:

\[ Z_0 = \sqrt{\frac{L/l}{C/l}} \]  

(4.1)

where \( L/l \) is the inductance per unit length and \( C/l \) is the capacitance per unit length. The basic premise behind a transmission line NMR probe then is to match the distributed inductance, which appears due to the probe coils, with an appropriate distributed capacitance, in order to give the desired characteristic impedance. The line can then be terminated with a resistor of the same value. The desired characteristic impedance is determined by the spectrometer, and in most cases is 50 \( \Omega \).

4.4.2 Previous Work

The use of transmission line-like probes in NMR was originally discussed by Lowe, Engelsberg, and Whitson [52,53] in the mid 70’s, then recently revisited by Kubo and Ichikawa [54]. A significant difference between these authors’ work and our own is that their probes are modeled as delay lines consisting of a finite number of cascaded LC circuit sections. This is natural for their designs, because their probes are constructed with a small number of discrete capacitors and wire inductors arranged in segments, forming a classic delay line. These probes are thus described by a characteristic impedance

\[ Z = Z_0 \sqrt{1 - \frac{\omega^2}{\omega_c^2}} \]  

(4.2)
where $\omega_c$ is a cutoff frequency above which no signal will propagate. The cutoff frequency is determined by $\omega_c = \sqrt{1/L_u C_u}$, where $L_u$ and $C_u$ are the inductance and capacitance per circuit section.

In a transmission line, the inductance and capacitance are distributed continuously along the length of the line rather than appearing in discrete cascaded sections. There are then essentially an infinite number of these sections, each of infinitesimal length, with the important consequence that there is (in principle) no cutoff frequency. Since our probes have a continuously distributed capacitance and inductance (with no discrete elements used in the construction), they are best treated as transmission lines rather than delay lines. It is interesting to note that Kubo and Ichikawa [54] found that after including the mutual inductances between adjacent circuit segments, the impedance of discrete delay line probes could also be well described by the transmission line formula.

As mentioned by Kubo and Ichikawa [54], very few transmission line probes have been put into practical use, most likely due to the difficulty encountered until now in their construction. We have only found two examples of transmission line coils: that of Stokes [55], who used a solenoidal coil to measure the T1 of $^{19}$F in difluorotetrachloroethane over a range of 18-80 MHz, and a low field ESR gaussmeter developed by Gebhardt and Dormann [56]. Both were solenoidal in design and required somewhat involved construction and calibration techniques. Our design constitutes a straightforward method of constructing transmission line probes. An important benefit of our method is that we can produce large quantities of practically identical probes with ease. Furthermore, our
design is the first which uses a saddle coil, thus making it particularly appropriate for superconducting solenoidal magnets.

4.5 Design

4.5.1 Method

The basic idea behind our probe is to make a saddle coil pattern out of traces on a flexible circuit board. We model the probe inductance, which is dominated by the saddle coil loops, as being uniformly distributed along the length of the coil trace. Then, for a given coil size and number of loops, we control the capacitance per unit length by adjusting the trace width. The construction method is similar to that of planar surface microcoils [57], but because our circuit board is flexible, we can wrap it around a sample tube to create a saddle coil.

Calculating the characteristic impedance of our transmission line probe consists of two parts, the capacitance and the inductance. Because the size of our coils is small compared to the wavelength of interest, we calculate the electrical characteristics of our probe for DC fields. To design probes for use at higher frequencies, of course, these assumptions will break down and more sophisticated modeling will need to be employed. We chose trace widths large compared to the thickness of the flex substrate, so that the capacitance could be modeled as that of a simple parallel plate capacitor, with

$$C = \frac{\varepsilon A}{d}$$  \hspace{1cm} (4.3)
Here $A$ is the surface area of the entire probe trace, $d$ is the thickness of the dielectric, and $\varepsilon_r$ is its effective dielectric constant. The effective dielectric constant [58] combines the effects of the dielectrics above and below the transmission line. It is dependent on the trace width and thickness. A typical ratio of the effective dielectric to the material dielectric for our various probe geometries is 0.88 to 0.89. To calculate the inductance of our coils, we initially made a simple estimate using the Biot-Savart law. We then used the results of Mohan et al. [59] for determining the inductance of surface-patterned spiral inductors. For various probe geometries the Mohan result was greater than the Biot-Savart approximation by around 18-20%.

To verify our calculations, we directly measure the total capacitance and inductance of our probes. To measure $C$, we remove the terminating resistor from the probe (so that the probe looks like an open circuit at DC) and read off the capacitance at the lowest frequency available (1 MHz) from a network analyzer. Using this method, our capacitance predictions match our measurements within 10-20% (see Table 4-1). To measure the inductance, we replace the resistor with a short circuit. These measurements, also made at 1 MHz, match the predicted values within 20-40% (see Table 4-1).

### Table 4-1: Electrical and mechanical parameters for three probe designs.
Experimental values were measured at 1 MHz, predicted values are at DC. The diameter refers to the total probe diameter, or the distance between the centers of the coils when the probe is wrapped around a sample tube.

<table>
<thead>
<tr>
<th></th>
<th>Diam.</th>
<th># loops</th>
<th>$C_{\text{meas}}$</th>
<th>$L_{\text{meas}}$</th>
<th>$\frac{C_{\text{meas}}}{C_{\text{pred}}}$</th>
<th>$\frac{L_{\text{meas}}}{L_{\text{pred}}}$</th>
<th>$Z_0 = \frac{L_{\text{meas}}}{\sqrt{C_{\text{meas}}}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Probe #2</td>
<td>0.5&quot;</td>
<td>3</td>
<td>375pF</td>
<td>769nH</td>
<td>0.86</td>
<td>0.76</td>
<td>45.3</td>
</tr>
<tr>
<td>Probe #3</td>
<td>0.5&quot;</td>
<td>2</td>
<td>138pF</td>
<td>343nH</td>
<td>1.02</td>
<td>0.95</td>
<td>49.8</td>
</tr>
<tr>
<td>Probe #4</td>
<td>15mm</td>
<td>2</td>
<td>69pF</td>
<td>230nH</td>
<td>1.10</td>
<td>1.43</td>
<td>57.7</td>
</tr>
</tbody>
</table>

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4.5.2 Potential Pitfalls

We have considered two possible types of deviations from ideal behavior in our probes: impedance mismatches and non-uniform distribution of inductance and capacitance. Impedance mismatches can occur at the "launch lines" which connect the coaxial connector to the coil, as well as at the interface between the coils and the stripline which connects one coil to the other. Reflections at the coax to coil connection reduce the total power delivered to the coil, as well as signal received from it. Reflections at the coil to stripline interface can unbalance the currents in the two sides of the saddle coil, and hence introduce inhomogeneity in the RF field produced and detected by the probe [54]. Furthermore, reflections can also cause the current amplitude to vary along the length of the probe, which can introduce additional spatial variation to the field.

The second form of deviation from the ideal that our probes may experience is the non-uniform distribution of capacitance and inductance. In this case the theoretically infinite segmentation of the transmission line becomes effectively finite, and our probe will behave in a manner similar to the delay line probe discussed above, albeit with a potentially high number of circuit sections. This has two effects. The first is the appearance of a cutoff frequency, as described in Eq. (4.2). The second is the appearance of a decay frequency, above which a signal will still propagate but will not effectively couple to the spins. This is due to phase delays between adjacent circuit segments [54]. The magnetic fields produced by the phase-delayed currents can interfere destructively, thus reducing the total RF field. The decay frequency is probe volume dependent; for cylindrical coils it scales as one over the radius cubed [54]. Since our probes have
relatively large radii, if their inductance and capacitance are not uniformly distributed it is possible that this could be a limiting factor for higher frequencies.

4.6 Construction

The flex material we have used is the Pyralux LF brand by DuPont, which consists of 1 oz. copper (thickness = 34.2 μm) deposited on both sides of a Kapton polyimide dielectric substrate. For this copper thickness the dielectric is available in thicknesses ranging from 76 to 127 μm. The thinner varieties are susceptible to permanent damage by creasing (similar to tin foil), and care must be taken when handling them. A dielectric coverlay can be deposited over the final design in order to lend mechanical rigidity and electrical insulation.

We have constructed three different types of probe. The first (probe #1) was machined on a CNC mill, making use of a vacuum chuck to restrain the flexible circuit material during cuts. This probe has two loops, and is rolled to form a saddle coil one inch in diameter. It is fabricated on Pyralux LF 9151R substrate (dielectric thickness of 0.007”, \( \varepsilon = 3.7 \)). Difficulties in machining make this an unreliable method for producing multiple identical probes. However, this prototype probe showed excellent impedance characteristics and gave us confidence in our basic modeling.

A more convenient and reliable way of producing these probes is to have them fabricated by commercial flex circuit manufacturing houses. We have used two different manufacturers to produce three different types of probe. The first (probe #2) is a one
inch diameter, three loop probe on Pyralux LF 7010R, with trace widths of 0.042". It has a 0.002" thick dielectric (Kapton + adhesive) with a dielectric constant \( \varepsilon = 3.7 \). The second (probe #3) is a one inch diameter two loop probe on a non-specified 1oz. copper laminate, with trace widths of 0.074" and a dielectric with a thickness of 0.007" and \( \varepsilon = 3.5 \). Our most recent probe design (probe #4) is a 15 mm diameter two loop probe printed on Pyralux LF 9151R, with trace widths of 0.062" and a dielectric thickness of 0.007", \( \varepsilon = 3.7 \). The coverlay for probes 2-4 is LF 1510, with thickness 0.0015".

Connections to and from the probe are made via a U.FL standard miniature (0.81 mm diam.) coaxial cable produced by Hirose. The transmission line is terminated on the flex circuit, with a 1/4 Watt 50 ohm non-magnetic chip resistor from SRT Technologies (model CHR-1206). The straight lines of conductor between the coaxial launch and the coils are standard 50Ω microstrip transmission lines. A photograph and mechanical drawing of probe #4 can be found in Figure 4-1 and Figure 4-2. Once manufactured, the probe is secured via plastic cable ties around a tube appropriate for the sample of choice. We later discovered that the cable ties were a significant source of field perturbation, and should be replaced with a more appropriate fastener.

Because of the limited thicknesses of dielectric available, the capacitance per unit length of the microstrip can be designed only over a fairly restricted range. This limitation can cause difficulties when attempting to specify the value of \( C/l \) needed to match the large \( L/l \) of the coil loops as necessary to make a \( Z_0 = 50 \Omega \) line. We found that trying to meet this capacitance requirement effectively limited us to no more than three loops.
Figure 4-1: Photograph of probe #4. The U.FL RF connector is located outside the picture, at the end of the microstrip "launch line" extending down past the bottom of the picture.

4.7 Results

A natural measure for an initial evaluation of the electrical properties of our probes is the reflection coefficient $\Gamma$, defined as the ratio of the reflected voltage to the incident voltage:

$$\Gamma = \frac{V_{ref}}{V_{inc}}$$  \hspace{1cm} (4.4)

This quantity is easily measured with modern network analyzers and is simple to interpret. In particular, this coefficient determines the fraction of power delivered to the load:

$$P = P_0 (1 - |\Gamma|^2)$$  \hspace{1cm} (4.5)
Figure 4-2: Mechanical drawing of front side of probe #4. The circuit is wrapped around a sample tube to form a volume coil that is 15 mm in diameter and 15 mm long. Trace widths are 0.062". A ground conductor is patterned on the back underneath the traces. Connections between the front and back are made via 0.007" diameter circular plated through holes. The T shaped lines are where the terminating resistor sits. The coaxial connector is on a tail several inches away, outside the drawing. The long “tail” was incorporated into the design when we found that the U.FL connectors contain a nickel flash of sufficient volume to affect our NMR measurements when located too near the probe.

Here $P_0$ is the incident power and $P$ is the power delivered to the load. In our situation the load is either the probe or the receiver amplifier, depending on whether the spectrometer is transmitting or receiving.

The transmitted power as a function of frequency for probe #4 can be seen in Figure 4-3. The measurements were taken with a Hewlett-Packard 8712C Network Analyzer. As seen in Figure 4-3, the probe is extremely well matched to a 50 Ω load over the frequency range of 5 to 26 MHz (0.1 to 0.6 T) required for our experiment; essentially all of the
Figure 4-3: Measurement of the unreflected fraction of power, \((1-|\Gamma|^2)\), vs. frequency, for probe #4. \(\Gamma \equiv \frac{V_{\text{reflected}}}{V_{\text{incident}}}\).

applied power is transmitted to the load. At higher frequencies, however, the probe begins to reflect larger fractions of the power. At 150 MHz nearly 90% of the applied power is transmitted, but at 300 MHz almost half of it is reflected.

Having a large transmission coefficient does not ensure that power is delivered in the form of an RF magnetic field, however. Field strength can be affected either by the decay frequency effects discussed above, or by current being shunted through the capacitance of the probe instead of flowing through the coils as desired. Hence, a more
stringent test of our probes requires measuring the RF field produced by the probe as a function of both frequency and position. To do this, we made a small pickup coil of one ~1.2 mm radius loop terminated with a 50 Ω resistor. In separate measurements, we verified that this pickup coil has a useful bandwidth of DC-300 MHz. To measure the RF field at various positions in the probe, we mounted the pickup coil on a translation stage near the center of a probe. The probe was driven by a function generator, and the voltage induced in the pickup coil was amplified and monitored with an oscilloscope. The RF field from the probe was investigated at frequencies up to 300 MHz using this setup. At low frequencies, the behavior of the probes agreed well with our expectations based on modeling with DC currents. At higher frequencies, substantial changes appeared both in the spatial distribution of the RF field, and in its magnitude. Figure 4-4 and Figure 4-5 show this behavior.

Figure 4-4 shows the measured RF field strength, as a function of position along the axis which connects the saddle coil centers, for various frequencies. At low frequencies, we expect a field minimum halfway between the coils, as observed. As the frequency is increased, the field minimum shifts away from the center, and towards the second coil in the probe (the coil furthest from the launch connector, and closest to the terminating resistor). At 300MHz, the field minimum shifted more than a millimeter from the center. Similar data showed that, in contrast, the RF field distribution along the central axis of the saddle coil cylinder (perpendicular to the line between coil centers) was relatively insensitive to the frequency. This indicates that the reason for the change in transverse field distribution is simply that the field produced by the first coil is stronger than that of
Figure 4-4: RF field strength versus position for probe #4 along the axis connecting the centers of the coils. Measured results are shown at several different frequencies, as well as the calculated field profile at DC. The zero of the x-axis is defined as the fitted position of the field minimum at low frequency. To highlight the changes in the spatial distribution of the field, results from all frequencies are normalized to the same value at x=0. A shift in the position of the field minimum, as well as an increase in the spatial curvature of the field, is evident at high frequencies.

the second. We suspect that this may be due to reflections at the transitions from the coils themselves to the thin microstrip trace connecting the coils.

Results for the absolute RF field strength as a function of frequency appear in Figure 4-5. To account for the varying spatial distribution of the field, this measurement was performed while tracking the field minimum along the axis connecting the coils. The measured values take into account the measured amplifier gain vs. frequency, and include
Figure 4-5: Measured and predicted RF field strength vs. frequency for probe #4, at an input power of +16 dBm (40 mW). As discussed in the text, the field strength for each frequency is measured at the position corresponding to the local minimum near the center of the saddle coils. The absolute accuracy of these measurements is at the 10-20% level. The predicted field strength is based on magnetostatic calculations, with corrections for the measured voltage reflection from the probe.

corrections for the measured pickup coil behavior as a function of frequency. We compare the measured values with predictions based on the calculated low-frequency behavior. The predictions include corrections for the measured reflection coefficient of the probe and the measured transmission properties of the U.FL cable. The measured and predicted values agree to within 25% for all frequencies up to 150MHz, but begin to diverge substantially at higher frequencies. Considering that the probe was designed using electro- and magnetostatic formulae, the frequency range of good performance is
Figure 4-6: Gradient echo image of the RF field produced by probe #4 at 170 MHz. The coils are near the upper left and lower right corners. This is a nutation angle map made from a 3.0 mm axial slice taken near the longitudinal center of the probe. TR = 3 s; TE = 3 ms; 2 Averages; Spectral Width = 50 kHz; FOV = 1.6 x 1.6 cm, 50 x 50 pixels.

surprisingly broad. Several possible reasons for deviation from the predicted behavior were mentioned earlier; however, determination of the source of the degradation in performance at high frequencies will require further investigation.

In order to further characterize the RF field homogeneity of our probe, we used the 4 T magnet at the Yale Magnetic Resonance Research Center to make a gradient echo image of probe #4's RF field at 170 MHz. As seen in Figure 4-6, the probe field is unbalanced, with greater sensitivity near one coil and less near the other. The overall behavior is consistent with our pickup-coil measurements. Both techniques indicate that at frequencies above about 100 MHz, the expected region of RF field homogeneity near the center of the saddle coils shifts to one side and becomes narrower. We note in passing
that using the probe with a commercial Bruker console required no extra consideration beyond a cable adaptor.

We have tested the NMR performance of our probes in an Oxford 85/310HR horizontal superconducting magnet run at a proton frequency near 22 MHz. The field was shimmed to approximately 0.1 ppm inside a 5 cm long, 15 mm diameter cylinder along the bore. The spectrometer is home built and will be described elsewhere. The important characteristic in terms of its RF performance is that we have replaced the traditional \( \lambda/4 \) line and crossed-diode based protection circuit with a simple electromechanical reed relay (Figure 4-7). At the center of the magnet and using a long, cylindrical water sample doped with 68.5 mM CuSO\(_4\), we can measure the proton frequency with a typical standard deviation in the center frequency of around 0.4 Hz for data from a single free induction decay. Under these conditions the linewidths for probe #4 are around 12.5 Hz, while the linewidths for probe #3 are around 25 Hz. After switching to pure water we see linewidths of around 6 Hz for probe #4. We believe the residual line widths are a result of the inhomogeneous B0 field; this is consistent with the fact that the measured linewidth for probe #4 is smaller than that of probe #3, due to the smaller volume sampled by probe #4. These widths were measured with the CuSO\(_4\) sample occupying the entire central volume of the probes. Later a reduced volume sample holder was introduced, effectively limiting the sample volume to a long cylinder of 5 mm diameter. Under this condition probe #4’s linewidth dropped to 8 Hz with CuSO\(_4\) and 3 Hz with pure water.
Figure 4-7: Broadband duplexer. Relay is Coto 2341. The rated switching time is 0.5 ms and the carry current rating is 1.5 A. The frequency range of this relay is unspecified, but it performs well below 22 MHz. Relays with similar specifications, specifically designed to act as 50 Ω transmission lines for signals with RF frequencies up to the GHz range, are available if needed.

In order to verify the broadband performance of our probe, we obtained spectra from \(^1\text{H}\) (in \(\text{H}_2\text{O}\) and in \(\text{H}_2\text{PO}_4\)), \(^{19}\text{F}\) (in perfluorotributylamine [Fluorinert, FC-43]) and \(^{31}\text{P}\) (in \(\text{H}_2\text{PO}_4\)). Examples can be seen in Figure 4-8. Switching between the various nuclei required changing only the samples and the applied RF frequency.

4.8 Possible Improvements

We anticipate that this probe might be useful in a wider range of circumstances than in our experiment, so here we discuss some possible improvements in the design. Clearly, operation up to higher frequencies would be desirable for many applications. A first step in this direction would be simply to make the probes smaller. Making the characteristic
length of the probe smaller may reduce the effects of reflections. Evidence for this can be seen in Figure 4-9: The useful frequency range (defined loosely as frequencies below the first peak in the voltage reflection coefficient) for the 15 mm diameter probe #4 is significantly broader than that of the 25.4 mm diameter probe #3. Furthermore, if the capacitance and inductance are unevenly distributed along the probe, thus causing it to behave more like a delay line rather than a transmission line, then one would expect the phase delays between sections to decrease thus raising the decay frequency. Finally, additional improvements in coil design may be possible by more accurately predicting the probe’s behavior at higher frequencies (e.g. by using commercial RF electromagnetic simulation packages).
Figure 4-8: Sequentially obtained spectra from various nuclei. The (a) $^1$H and (c) $^{31}$P spectra come from $\text{H}_3\text{PO}_4$ (85% by mass); the (b) $^{19}$F spectrum comes from perfluorotributylamine (Fluorinert, FC-43). Samples were held in a 17x120 mm standard 15mL Falcon plastic tube. Both the $^1$H and $^{31}$P spectra are the average of 100 FID's, each of 150,000 samples with a 2s delay between each FID acquisition. The $^{19}$F spectrum is the average of 100 FID's, each of 45,000 samples, with a 2s delay between each FID acquisition. In all cases the sampling rate was 1MHz. The only adjustment made to the spectrometer between acquiring these signals was a change in the drive frequency and the tipping pulse time. The tipping pulse was 80μs for $^1$H and $^{19}$F, 200μs for $^{31}$P.
Figure 4-9: Voltage reflection coefficient vs. frequency for probes #3 and #4. The smaller size of probe #4 pushes the peak in reflected power to a higher frequency.

4.9 Potential Applications

The utility of a transmission line probe goes well beyond that of our particular application (a high-precision, broadband NMR teslameter for a physics experiment). Other potential uses include:

- The ability to simultaneously acquire the spectra of several nuclei without the need for multiply tuned probes.
- Use in solid-state NMR, where the spectral frequency ranges are often too large for conventional tuned probes [54].
- NMR of nuclei with large quadrupolar couplings, where frequency sweeps are typically employed [54].
- Study of samples whose magnetic properties are dependent on the external field strength, since with a non-resonant probe one can change the strength of the static field continuously [54].
- Use in cryogenic studies to avoid tuning difficulties due to resonant circuit components at different temperatures [60].
- Use in short time scale experiments, since non-resonant probes should have better transient recovery times [52].
- Reduction in Johnson noise by locating the terminating resistor in a low temperature bath [54].
- Use in nuclear quadrupole resonance detection schemes, e.g. for explosives and narcotics, where the signals intrinsically cover a broad range of frequencies [61] and fast transient recovery time can increase the signal to noise for fast decaying-signals [51, 61].

4.10 Conclusion

We have designed and constructed an easy to manufacture, tuning free, broadband NMR probe. The probe coil itself constitutes a transmission line, which ensures a good impedance match to a standard spectrometer across over 200 MHz of bandwidth. Construction of our probe is easily performed by printing the coil design onto a flexible circuit board, and design of new probe geometries should be straightforward.
Chapter 5: Fluorescence Detection

5.1 Introduction and Overview

We have recently made a substantial improvement to the signal sizes in the anapole experiment by implementing a new, more efficient detection scheme based on two-photon excitation. This chapter reviews the previous detection method and then describes the methods and results obtained with the new technique.

Recall that we are interested in two hyperfine-rotational states in our molecules, $|a\rangle$ and $|b\rangle$. Level $|a\rangle$ is initially depopulated in Chamber 2, then the levels are brought to near degeneracy as they pass through the magnetic field. An electric field forces the molecules to change state, shifting some of the population from $|b\rangle$ back to $|a\rangle$. In addition to this Stark effect, weak-interaction processes also contribute to the population transfer. Depending on the sign of the electric field, the weak-interaction mixing either adds to the Stark mixing or subtracts from it, thus the term Stark interference. The difference between the population transferred with a positive electric field versus that transferred with a negative electric field is proportional to the strength of the weak interaction. Measuring this population transfer is thus critical to the success of our experiment.

Our original strategy for detecting the final population in the $|b\rangle$ state was to drive a single photon transition from $|b\rangle$ to a higher state, then to look for fluorescence at the
same wavelength as the higher state decayed back to the ground state (Figure 5-1). In principle this is fine, but exciting and detecting at the same wavelength leads to problems with scattered laser light contributing to the background. Some of our efforts to control this background scatter are described in 5.3.2. Further disadvantages to this strategy are that the near infrared wavelengths we use to excite the molecules are in ranges where photomultiplier tubes (PMT's) have very low quantum efficiencies and small active areas. Compounding the difficulty is the magnetic field, which causes a dramatically lower gain for the type of PMT required by the single photon scheme. In order to mitigate these effects we found it necessary to place the PMT at the end of a flexible liquid lightguide that is long enough to move it out of the magnetic field. The signal loss in this configuration is substantial.

A better detection strategy employs a two photon excitation scheme. Under such a scheme two lasers of different frequencies couple initial and final states in the molecule via an intermediate state. When the final state decays, the resulting fluorescence is at a different frequency than either of the excitation lasers, thus allowing for essentially background-free detection. The cost is the complication of an additional laser, as well as loss due to the finite lifetime of the intermediate state. This loss can occur when molecules which have been excited to the intermediate state decay before they can be subsequently excited to the final state.
$A^2\Pi_{\frac{1}{2}}, J_P = \left(\frac{3}{2}\right)^+$

$X^2\Sigma_{\frac{1}{2}}, N_P = 0^+, J_P = \left(\frac{1}{2}\right)^+$

Figure 5-1: Single photon excitation scheme. The molecules are driven from the ground state $X$ to excited state $A$. They then decay, with the resulting fluorescence detected at 860 nm. The $A$ state can decay to many hyperfine-rotational sublevels of the ground state, thus in practice once the upper state has decayed we see no more photons from the molecule.

In addition to the dramatic reduction in background, a two photon scheme benefits from the fact that in our molecules the emitted fluorescence is in the blue, where significantly more efficient PMTs are available. Also, here we have found PMTs that are much less susceptible to magnetic field effects, thus allowing us to forgo the long liquid lightguide in favor of a short quartz rod. Furthermore, elimination of background concerns allows us to increase geometric collection efficiency. This increase in the collection efficiency due to a better geometry, PMT, and lightguide, as well as the reduction in background, are the primary advantages of the two photon scheme.
One important aspect of the two photon excitation method is the need to minimize losses due to the finite lifetime of the intermediate state. There is a control protocol known as STImulated Raman Adiabatic Passage (STIRAP) [62,63], which employs a counterintuitive temporal ordering of the two excitation lasers, that has been demonstrated to produce complete population transfer between the initial and final states with virtually no loss from the intermediate state. However, because of the sensitivity of STIRAP to external parameters, as well as further complications in applying it to magnetic sublevels whose degeneracy has been lifted by Zeeman shifts [64] (as is our case), we designed our detection scheme with the goal of providing for STIRAP where possible but keeping the main focus on increasing as much as possible our signal-to-noise in Chamber 3. To that end we pursued the program described here.

The organization of this chapter is as follows: First we give an overview of the states of our molecule. Then we describe the single-photon detection scheme, followed by the two-photon scheme, and finally our attempts to optimize the signal in the anapole beam.

5.2 Molecular States

Here we describe the molecular states of interest in barium monofluoride (BaF), the first molecule of our experimental series. We are interested in parity violation which depends on nuclear spin, which in this case refers to the spin of the significantly heavier Ba nucleus. In particular, for parity violation measurements we plan to use $^{137}$Ba$^{19}$F, where $I_{Ba} = 3/2$ and $I_F = 1/2$. However, for beam diagnostics and proof of principle experiments with the two photon detection system, we use the seven times more
abundant $^{138}$Ba$^{19}$F. Unfortunately $^{138}$Ba has no nuclear spin, so it cannot be used for NSD-PNC experiments. To be clear, the majority of discussions in this chapter will be concerned with $^{138}$Ba$^{19}$F.

Barium fluoride is a diatomic free radical. We can consider its electronic structure to consist of a valence electron shared unequally by two nuclei surrounding a closed inner core of electrons. We effectively treat the molecule as having a single electron. The states of BaF can be represented by the following glyph:

$$Y^{2\Sigma+1}A_{\Omega}$$

Here $Y$ is a naming label for the electronic state, $\Sigma$ is the projection of the electronic spin $S$ onto the internuclear axis, $\Lambda$ is the projection of the electronic orbital angular momentum $L$ onto the internuclear axis, and $\Omega$ is the projection of the total angular momentum onto the internuclear axis. In general $J = N + L + S$, where $N$ refers to the rotational angular momentum of the molecule about the internuclear axis. Much like the familiar $s, p, d$ values of $L$ for atoms, in molecules the values of $\Lambda$ take on the labels $\Sigma, \Pi, \Delta$ etc. The vibrational level of the molecule is labeled with an additional quantum number $\nu$, although in all cases of current interest $\nu = 0$ so we ignore it. When nuclear spin is included, $F = J + I$ is the total angular momentum of the molecule, with projection $\Omega$, on the internuclear axis. If there are two nuclei present, $F = F_1 + I_2$, where $F_1 = J + I_1$.

The appropriate couplings of angular momenta in molecules depend on the particular state, and are specified by the Hund's cases [65]. Relevant to the problem at hand are
Hund's cases (a) and (b). In Hund's case (a), the spin-orbit coupling is much larger than the rotational energy, and good quantum numbers are \( J, \Lambda, S, \Sigma, \) and \( \Omega = \Lambda + \Sigma. \) In Hund's case (b), the spin-orbit coupling is much smaller than the rotational energy. Good quantum numbers for Hund's case (b) are \( J, \Lambda, S, \) and \( N. \)

The parity of a Hund's case (b) state is given by \( P = (-1)^N. \) The parity of a Hund's case (a) state cannot be written as a comparably simple formula. Rather, the parity eigenstates are superpositions of states with \( \pm \Omega. \)

The low lying electronic states of BaF are shown in Figure 5-2. There are three

![Diagram of electronic states](image)

**Figure 5-2:** Electronic states of BaF. The two photon scheme will drive transitions between both the X and A state and the A and D state. Fluorescence is emitted by the decay of the D state near 413nm.
electronic states of interest to us: X, A, and D. The X and the A states are used for the one-photon scheme, and the D state is added in the two-photon scheme. For the parity violation experiment our two levels \( |a\rangle \) and \( |b\rangle \) are different \( m_r \) sublevels of the \( N^p = 0^+ \) and \( N^p = 1^- \) rotational levels of the X electronic ground state. The otherwise degenerate \( m_r \) sublevels are split in energy by the magnetic field, and we are free to choose pairs of sublevels which cross at different field strengths. A plot of the \( N=0,1 \) levels as a function of magnetic field are seen in Figure 5-3.

![137 BaF rotation/hfs Zeeman shifts (l=3/2)](image)

**Figure 5-3:** \(^{137}\text{BaF}\) ground state level crossings. The plot shows hyperfine/rotational energy levels as a function of magnetic field. \( F_1 = I_{Ba} + J \). In this case \( L = 0 \), so \( J = S \). The notation is \( F = F_1 + I_F \). Here the effects of the fluorine hyperfine interaction, which is at the 30 MHz level, are not visible.
5.3 Single-Photon Excitation

5.3.1 Overview

In the original single photon excitation scheme, we drive transitions between the

\( X^2 \Sigma_{1/2}, \ N^p = 0^+, \ J^p = \left(\frac{1}{2}\right)^+ \) state and the \( A^2 \Pi_{1/2}, \ J^p = \left(\frac{3}{2}\right)^- \) state. This transition we will henceforth call simply \( X \rightarrow A \). Its wavelength is around 860 nm. We then detect fluorescence from the decay of the A state. A diagram can be found in Figure 5-1.

In order to know where to tune the laser when driving transitions such as this, as well as to understand any spectra we observe, we must determine the energy splittings of these states. The rotational Hamiltonian, when neglecting nuclear spin, can include terms contributions from the term (electronic) energy, rotational energy, spin-orbit interaction, spin-rotation interaction, and lambda doubling. The Hamiltonian for a \( ^2 \Pi \) state such as the A state is given by [66]:

\[
H_A = T + BJ(J + 1) + AT^l(L) \cdot T^l(S) + (-1)^{J-1/2} \frac{1}{2} [p + 2q] \left[ J + \frac{1}{2} \right].
\]  (5.1)

Here \( T \) is the electronic energy, \( B \) is the rotational constant, \( A \) is the spin-orbit coupling constant, and \( p \) and \( q \) are lambda doubling parameters. The A state is a Hund’s case (a) state, so its parity is not described by a simple formula. As described in [66], the spin-orbit coupling, \( H = AT^l(L) \cdot T^l(S) \), yields an on-diagonal energy term of \( E = A \Lambda \Sigma \). We are considering the \( \Pi_{1/2} \) substate where \( \Omega = 1/2 \). Since \( \Omega = \Lambda + \Sigma \), and \( \Lambda = 1 \) this
means \( \Sigma = -1/2 \). Thus the spin-orbit shift to the A state is \(-1/2 \, A\). There is no spin-
rotation contribution to the A state.

The relevant constants were determined to high precision in [72]. Using these constants (given in Table 5-2), as well as the Hamiltonian and constants for the X state (described in 5.4.1), we calculate the transition frequency to be 11631.374 \( \text{cm}^{-1} \). We observe the transition at 11631.28 \( \text{cm}^{-1} \), as determined by a standard laser wavelength meter. Since the accuracy of the wavemeter used in the measurement is quoted as 0.1 \( \text{cm}^{-1} \), the agreement is reasonable.

The apparatus used in the single-photon excitation scheme is very similar to that used in the two-photon scheme, which will be described in 5.4.2. The important differences, as mentioned in the introduction, are the photomultiplier tube and lightguide used, as well as the fluorescence collection mirrors. A combination of the PMT’s small area, its small quantum efficiency at 860 nm, the lightguide’s insertion loss, and the mirrors’ reflection efficiency limited our total collection efficiency dramatically. D. Rahmlow estimates the collection efficiency using the single photon apparatus to be \(-0.0002\).

### 5.3.2 Limitations

An important limitation on the signal-to-noise of the single-photon scheme is background produced by laser scatter. Because we are exciting and detecting at the same wavelength, photons from our detection laser are indistinguishable from photons from our molecules. Considerable time and effort therefore were devoted to reducing the background rates in Chamber 3, where the signals are both smallest and most critical. In addition to typical
considerations such as installing Brewster-angle windows on the optical access paths, a series of light baffles made of electro-sharpened tungsten washers was installed. To produce these washers, tungsten sheets were cut into discs, which were then electro-sharpened in a 2 M solution of KOH. All washers had an outer diameter of $\frac{1}{2}$" and inner diameters of 5 mm, 6 mm, or 7 mm. To perform the electro-sharpening an electrode was placed through the center of the hole in the washer, and a current of 2 A was maintained between this central electrode and a second electrode placed in the KOH bath. A holding structure shown in Figure 5-4 maintained the position of the center electrode in the washer as well as the washer in the bath. During the sharpening time the solution was constantly agitated by a Troemner Model 500 stirring device, with the stir power set to 2.5. The 2 A current was measured when the stirrer was on. A total of eight production quality washers, four with an inner diameter of 5 mm and four with an inner diameter of 6 mm, were made in this fashion. Sharpening time was 3 minutes for the 5 mm washers and 2.5 minutes for the 6 mm washers. After sharpening, a special blackening paint, AZTech MLS-85-SP-C, was applied via pneumatic sprayer. The model of sprayer, Preval, was found to be critical to achieving an even flat coat; other types of sprayers gave unacceptable paint finishes. The finished washers were mounted inside Thorlabs $\frac{1}{2}$" lens tubes, and affixed to a translation stage mounted on the inside of Chamber 3. By carefully adjusting this translation stage, and thus the alignment of the washers relative to the incoming laser beam, the laser scatter measured in the absence of a molecular beam was minimized. The resulting background rate of 5-10 kHz was a considerable improvement over the initial rate of 470 kHz.
5.3.3 Results

Using the single-photon excitation scheme, typical signal rates in Chamber 3 for $^{138}\text{Ba}^{19}\text{F}$ (with the beam running at a 10 Hz repetition rate) are 2 counts / pulse. Using this scheme our group has been able to perform spectroscopy of $^{138}\text{Ba}^{19}\text{F}$. Additional results achieved with the single-photon excitation scheme will be described in detail in the forthcoming dissertation by D. Rahmlow.

Figure 5-4: Holding apparatus for tungsten washers during electro-sharpening. The edge of the tungsten washer is held between the two steel washers on the rod on the left, while the rod on the right passes through the center of the tungsten washer.
5.4 Two-Photon Excitation

5.4.1 Overview

In the two photon excitation scheme we excite the $X \rightarrow A$ transition at 860 nm and the $A \rightarrow D$ transition at 797 nm. We detect fluorescence from the D state at 413 nm. A diagram of the process can be seen in Figure 5-5. In the D state we have used two final sublevels, either the $D^2 \Sigma_{1/2}$, $N^p = 2^+$, $J^p = \left( \frac{3}{2} \right)^+$ or

the $D^2 \Sigma_{1/2}$, $N^p = 2^+$, $J^p = \left( \frac{5}{2} \right)^*$ states. Both of these transitions have wavelengths around 797 nm.

The effective Hamiltonian for a Hund’s case (b) state such as the D state is given by:

$$H_D = T + BN(N + 1) + AT^l{(L \cdot T^l(S))} + \gamma N \cdot S.$$  \hspace{1cm} (5.2)

Here $T$ is the electronic energy, $B$ is the rotational constant, $A$ is the spin-orbit coupling constant, and $\gamma$ is the spin-rotation constant. Both D states as well as the X state are Hund’s case (b) states, so their parity is described by $P = (-1)^N$. Constants are determined in [72] and are tabulated in Table 5-2. Similarly to the A state, for the D states the spin-orbit coupling yields an on-diagonal energy term of $E = A \Lambda \Sigma$. However here $\Lambda = 0$, so there is no spin-orbit contribution. The spin-rotation term $H = \gamma N \cdot S$ yields an on-diagonal energy term [66] of $E = \frac{\gamma}{2} \left[ J(J + 1) - N(N + 1) - S(S + 1) \right]$. The
Figure 5-5: Two photon excitation scheme. Molecules are driven from the ground state $X$ to an intermediate state $A$, then to the final state $D$. Fluorescence is then detected at 413 nm. Due to the number of sublevels available for the upper state to decay to, in practice we do not expect to see multiple photons from the same molecule.

The $A \rightarrow D$ ($J = 3/2$) transition is calculated to be at 124546.588 cm$^{-1}$, and the $A \rightarrow D$ ($J = 5/2$) transition at 12546.606 cm$^{-1}$. These have been measured (as detailed in 5.5.3) to be at 12546.66 cm$^{-1}$ and 12546.68 cm$^{-1}$. As in the single photon case, this is within the 0.1 cm$^{-1}$ accuracy of our wavemeter.
5.4.2 Apparatus

The excitation and detection apparatus consists of three subsystems. One is an optical board where the 797 nm light originates. We refer to this as the Beam Production Board. It is physically located on the main optical table. A second subsystem is an optical board where the 797 nm and 860 nm beams are overlapped then delivered to the molecules. We refer to this as the Beam Combination Board. The combination board is attached to the side of Chamber 3, and is connected to the production board via a single mode polarization-maintaining fiber optic cable. Inside Chamber 3 there is a set of specially designed optical mirrors. These mirrors, in combination with a quartz lightguide and photomultiplier tube, will be collectively referred to as the Fluorescence Detection.

Figure 5-6: Overview of the two-photon detection scheme apparatus as installed in the anapole beam.
subsystem. An overview diagram of the entire apparatus as used in the anapole beam experiment can be found in Figure 5-6. A photograph of Chamber 3 featuring the collection mirrors can be found in Figure 5-7.

5.4.2.1 Beam Production Board

A schematic of the 797 nm beam production board can be found in Figure 5-8. The 797 nm beam originates from a 160 mW Toptica DL100L diode laser. An anamorphic prism pair reshapes the initially elliptical beam to have a more circular intensity profile. An OFR I0-λ series optical isolator protects the diode from back-reflections. The beam then passes through a series of half-wave plate (HWP) and polarizing beam splitter cube (PBS) combinations. The half-wave plate rotates the polarization angle of the incoming
linearly polarized beam. The PBS allows the horizontally polarized portion of the beam to pass through it, while it reflects at 90 degrees the vertically polarized portion. Thus when used in series the HWP-PBS combination acts like a beam splitter where the user can select the amount of power which exits in each direction. There are three such combinations on the production board. The first in the series sends light to a wavemeter, the second to Chamber 3, and the third is available for future use. Light which passes through all three HWP-PBS combinations is aligned into a Fabry-Perot (FP) cavity for locking. The locking mechanism is a software servo loop written in Labview by D. Rahmlow. It uses the 860 nm laser (which is itself locked in a different setup) as a reference, and attempts to maintain a constant separation between the 797 nm and 860 nm laser frequencies via a feedback loop. The 860 nm reference beam is overlapped with the 797 nm beam prior to the FP cavity via a HWP-PBS combination.

An $f = 250$ mm plano-convex singlet lens is used to focus the 860 nm beam through the FP. A PBS splits the beams on the far end of the cavity, and the transmitted power of each is read via a PDA100A Si amplified photodiode from Thorlabs. Bandpass filters are present in the 860 nm beam path to better isolate its signal from that of the 797 nm light. Standard BNC cables connect the photodiodes to the computer. The variable gain transimpedance amplifier present in each photodiode is set to a gain of 50 dB.

The light which branches off to Chamber 3 typically carries as much power as is practical. Because of this it has two additional isolators to reduce potential optical feedback into the laser diode. A half wave plate is in place between the two isolators in order to maximize the total power transmitted. We note in passing that significantly
Figure 5-8: Production board schematic. 1-anamorphic prism pair. 2-isolator. 3-HWP/PBS. 4-HWP. 5-PBS. 6-f=25 cm lens. 7-Fabry-Perot. 8-bandpass filter.

greater transmission efficiency was realized after replacing the original multi-order @ 780 nm HWP with a zero order @ 808 nm plate.

When the 797 nm laser is run at a current of 234 mA, a total of 100 mW exits the laser head. The maximum rated current for the 797 laser is 262 mA. After the anamorphic prism pair and first isolator, a total of 83 mW is available for all optical paths to share. Losses due to the isolators in the Chamber 3 branch result in a total power of 47 mW available to be fiber-coupled to the experiment. The three isolators in the beam path have insertion losses of 17%, 23% and 20% respectively. These isolators are OFR Body Type II isolators, which are designed for a fixed wavelength. Their polarizers are fixed in
place and cannot be rotated. The first isolator is for 850 nm, and the second and third are for 780 nm. Thus in the future if more power is desired, replacing the isolators with models optimized for operation at 797 nm may help.

Connections between the production board and the combination board, as well as the wavemeter, are made via Thorlabs FS-LS-4616 Single Mode Polarization Maintaining fiber. The free space to fiber couplers are Thorlabs/OFR PAF-X-11-PC-B fiber port collimators. We have regularly achieved total fiber coupling efficiencies of 65-67% using this setup. The output polarization of the laser beams is determined by the orientation of the collimators. When the output collimators are oriented to maximize transmission through the PBS, 90% of the coupled laser power is available with the horizontal polarization required by the experiment.

We note that when the setup was moved to the Anapole Beam laboratory (after original setup and use in two other laboratory rooms), the coupling efficiencies through the single mode fibers were found to be substantially reduced: typical efficiency here was only 50% rather than ~66% as previously found. It is possible that this is due to higher vibrations in the Anapole lab, in part potentially because of the location of laboratory electronics on the main experimental optical table. Attempts to improve the vibration isolation of the beam production board through standard techniques (e.g. mounting the board on rubber stopper feet and on vibration insulating foam) did not yield an improvement however.
5.4.2.2 Beam Combination Board

A schematic of the beam combination board appears in Figure 5-9. A photograph appears in Figure 5-10. The combination board is mounted on three translation stages: one controls the height, a second controls the position along the molecular beam, and a third controls the distance from the center of Chamber 3.

Optical inputs to the board are via Thorlabs F810FC-780 fiber output collimation lenses mounted in Thorlabs K6X six-axis kinematic optic mounts. These lenses produce beams with slightly elliptical outputs. The 797 nm beam has a vertical beam waist of 2.9 mm and a horizontal beam waist of 3.6 mm. For the 860 nm beam these waists are 3.4 mm.

![Combination board schematic diagram](image)

Figure 5-9: Combination board schematic diagram.
and 3.3 mm. Here the waist refers to the $1/e^2$ radius in intensity as measured by a CCD camera. The 797 nm beam output collimation is sufficiently good that a maximum variation in beam waist of 18% is observed over 2.5 meters in beam propagation distance from the collimating lens. The collimation of the 860 nm beam output has not been measured. It is important to note that the fiber output collimators are specified for a fixed wavelength of 780 nm. This fixed wavelength design [67] results in a wavelength dependent degree of collimation. The next closest wavelength available however was 635 nm.

After entering the board the lasers pass through PBS cubes, which ensure that the beams contain only horizontally polarized light. This is necessary to address the proper magnetic sublevels of the states of interest. The beams are then focused with $f = 400$ mm cylindrical lenses. The purpose of the cylindrical focusing is to allow the beams to stay relatively large in the vertical dimension while narrowing their profile and increasing their intensity in the horizontal dimension. In this way we can intercept as much of the molecular beam as possible while still maintaining the tight beam focus required for STIRAP. After passing through the cylindrical lenses, the beams are overlapped via a dichroic mirror. The measured insertion loss of the dichroic in the 860 nm beam is $\sim20\%$, while for the 797 nm beam it is very little. After the dichroic mirror, the beams propagate through Chamber 3. In situ determination of the laser beams’ sizes and overlap, at the position of the molecular beam, is made by picking off a small portion of both beams with a Thorlabs BSP10-B1 beam sampler along the way. Images of the sampled beam are taken with a Mightex 1.3 megapixel 8-bit monochrome CMOS
windowless camera (Model MCE-B013-UW). A sample image can be found in Figure 5-12.

Because of diffraction effects and imperfect collimation, each beam’s minimum waist is not located exactly one focal length from the cylindrical lens. In order to produce similar spot sizes at the same distance from the dichroic mirror, we found that the 797 nm beam’s cylindrical lens should be placed 3 cm farther from the dichroic mirror than the 860 nm beam’s lens.
Since the two-photon excitation depends critically on the overlap between the two excitation lasers, great care was taken to ensure that the 797 nm and 860 nm beams were parallel. To verify this, intensity profiles were taken with a CCD camera at different points along both laser beams. This can be seen in Figure 5-12. These data were then fit to Gaussian profiles, and beam waists as well as peak separations were extracted. The results, shown in Table 5-1, demonstrate that the beams are substantially parallel. This can be seen by noting that the separation between the 797 nm beam peak and the 860 nm beam peak varies by less than 4.7 μm, or 0.8%, over a 2 cm range. As is also seen in Table 5-1, the waist of each beam is sensitive to the position along the beam path. Because we would like both lasers’ foci to be at the intersection with the molecular beam, this has important implications for both the positioning of each laser as well as the
position of the beam combination board. When making the measurements described above, we were careful to match the optical path length from the dichroic mirror to the camera as closely as possible to the optical path length from the mirror to the center of the chamber. However, the uncertainty in these distance measurements is likely at least a few millimeters. The impact of this uncertainty is mitigated somewhat however by the fact that the combination board is on a translation stage with a range of 254 mm. The fluorescence signal can be optimized by tuning the board position. Note however that this requires scanning the relative beam position (described below) at each point in the translation stage scan.

A second important experimental requirement is to be able to precisely control the relative position of the two beams. By mounting the dichroic mirror on a standard Thorlabs translation stage with a Mituyo KK-350 precision micrometer, we are able to control the beam separation to 0.001 mm. A plot of the relative separation of the two laser beams as a function of the micrometer reading can be found in Figure 5-13.

**Table 5-1**: Beam waists for the 797 nm beam and 860 nm beam at three positions along the laser beam path. The 0 mm measurement is taken at the same distance from the dichroic mirror as the center of the chamber. The -10 mm point is farther from the mirror than the center of the chamber, while the +10 mm point is closer. At each position the separation between the peaks of both lasers is also recorded.

<table>
<thead>
<tr>
<th>Position along laser:</th>
<th>-10 mm</th>
<th>0 mm</th>
<th>+10 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>797 nm beam waist ((\mu m))</td>
<td>44</td>
<td>58</td>
<td>157</td>
</tr>
<tr>
<td>860 nm beam waist ((\mu m))</td>
<td>47</td>
<td>66</td>
<td>117</td>
</tr>
<tr>
<td>separation between peak centers ((\mu m))</td>
<td>588</td>
<td>583</td>
<td>586</td>
</tr>
</tbody>
</table>
Figure 5-12: Beam profiles taken with a CCD camera at three positions along the optical path. The 0 mm (red) trace is taken at the same distance from the dichroic mirror as the center of the chamber. The -10 mm trace is farther from the mirror than the center of the chamber, while the +10 mm trace is closer. Each trace shows both the 797 nm beam (left peak) and the 860 nm beam (right peak). The beams are deliberately offset for clarity. The z-axis is along the horizontal dimension perpendicular to the laser optical paths. As tabulated in Table 5-1, over a 2 cm range the separation between the peaks varies by less than 4.7 μm, or 0.8%, indicating that the beams are substantially parallel. Beam waists can be found in Table 5-1.
Figure 5-13: Separation between the centers of the 860 nm and 797 nm laser beams, as a function of dichroic mirror position. As the micrometer reading increases, the 797 moves further upstream. By changing the relative position of the beams we can optimize conditions for STIRAP. The beam centers are overlapped at 1.111 mm.

5.4.2.3 Fluorescence Detection

The primary components of the fluorescence detection system are a set of optical mirrors, a 1” OD quartz lightguide, and a Hamamatsu R7600U-200SELECT photomultiplier tube. Photographs of the mirrors in Chamber 3 as well as the PMT in its holding apparatus are in Figure 5-7 and Figure 5-14.

The mirror set was specially designed by Matthew Lawlor as part of an undergraduate research project with our group. The lower half of the set is an ellipsoidal mirror, while
the upper half is a spherical mirror. The operational principle is that the majority of light emitted within the mirror set will be focused at the upper focus of the ellipse. A diagram can be found in Figure 5-15. Lawlor calculated optimal design parameters through numerical simulation. The mirrors were manufactured by the Gibbs Machine Shop and diamond turned by NU-TEK Precision Optical Corp. The surface coating was a “semi-enhanced” aluminum, with an expected reflectivity of 92% in the 370-440 nm range. Optimum collection efficiency theoretically occurs when the light pipe is positioned at the upper focus of the ellipsoidal mirror, which is located 0.6” below the top surface of the mirror set.

The photomultiplier tube has a quantum efficiency at 413 nm of around 37%. In order to help reduce any background due to the excitation lasers, a Newport FSR-BG40 color glass filter is located immediately prior to the PMT. Its transmission at 413 nm is around 85-87%, but at 797 nm and 860 nm it is near zero. Signals from the PMT are amplified by a Stanford Research Systems SR445A 350 MHz preamplifier. The first two channels of this amplifier are chained together to give a voltage gain of ~25. The total DC offset was carefully zeroed. The PMT is operated in a photon counting mode, and the threshold was chosen as follows: first, the number of counts as a function of threshold under various lighting conditions is recorded. Then, the difference between the number of counts at a given threshold is subtracted from the number of counts at a threshold which is one step higher. For example, the number of counts at a threshold value of -92 mV is subtracted from the number of counts at -90 mV. We loosely term this the “derivative” of the PMT counts. A plot of this derivative as a function of threshold voltage can be
found in Figure 5-16. Each point in this plot the extra number of counts you will see at a
given threshold relative to the threshold immediately below it. By comparing this change
under different lighting scenarios, a threshold can be chosen which minimizes
unnecessary background but still remains sensitive to signals of interest. Using this
technique a threshold of -0.034 mV was chosen. This is a somewhat conservative value
however, and the potential for performance increase should be investigated.

Figure 5-14: Photograph of PMT and lightguide. The lightguide passes through two
custom vacuum fittings in order to keep it from sliding when the chamber is at
$10^{-7}$ torr. A copper tube is placed around the lightguide to help reduce scatter from
the room lights.
Figure 5-15: Collection mirror diagram. The top mirror (red) is a spherical surface, while the bottom mirror (blue) is shaped like an ellipse. The center of the spherical mirror (yellow sun) coincides with the lower focus of the elliptical mirror (also yellow sun). The upper focus of the elliptical mirror coincides with the entrance to the light pipe (red sun). When molecules emit light near the focus of the ellipsoidal mirror (yellow sun), rays that hit the ellipse go to the other focus of the ellipse, while rays that hit the sphere go back through the center, then hit the ellipse, and are subsequently reflected to the other focus of the ellipse. We estimate the geometric collection efficiency of this mirror set to be 80-85%.
Figure 5-16: “Derivative” of PMT counts as a function of threshold. See the text for a description of the derivative. Each point is essentially the extra number of counts you will see at a given threshold relative to the threshold immediately below it. The PMT is being operated in a photon counting mode, thus the threshold is the voltage level above which a signal is registered as a count. The black trace is with minimal room lighting, the red trace is with no room lighting. The tall peak on the right is electronic noise.

5.5 Proof of Principle

5.5.1 Introduction

A key element to successful state detection using a two-photon resonance is knowledge of both of the transition frequencies required. We knew through prior experiments using
the single photon scheme where to tune the 860 nm laser. However, the $A \rightarrow D$
transition frequency had to be explicitly verified. Because the number of molecules in
Chamber 3 is small, measuring the transition frequency in situ was anticipated to be
difficult. Furthermore, installing the two-photon detection apparatus involved removing
the single photon detection apparatus. Thus in order to maximize chances of success in
the anapole beam, we first performed a separate proof of principle experiment using a
buffer gas source.

An independent experimental team in the DeMille group is developing a source of cold
polar molecules based on a buffer gas cooling technique [68]. A drawing of the buffer
gas chamber can be found in Figure 5-17, and a schematic diagram of the chamber as
used with our two-photon detection apparatus can be found in Figure 5-18. Briefly, a
BaF$_2$ pressed target is vaporized via laser ablation inside a cell of helium gas maintained
at 4 K. BaF produced in the ablation thermalizes with the helium, then exits the cell
through a small hole, producing a beam of BaF outside the cell. The beam typically
contains $10^{11}$ molecules per shot, with a translational temperature of $\sim 4$ K.
Characteristics of the beam are usually measured via absorption techniques.
Figure 5-17: Buffer gas source drawing. A layer of coconut charcoal, not shown here, maintains the vacuum inside the heat shield.

5.5.2 Buffer Gas Setup

Both the beam combination board and beam production board were installed in the Buffer Gas Lab to perform this experiment. The beam production board was located on their optical table, and the combination board was mounted near the side of the buffer gas chamber such that the two laser beams intersected the molecular beam approximately 3 mm from the cell aperture. Absorption measurements both inside and outside the cell allowed us to ensure that the 860 nm laser was on resonance with the molecules.
Figure 5-18: Top view schematic diagram of buffer gas source. Molecules are formed via laser ablation of a BaF$_2$ target. They thermalize with helium gas which is maintained at 4K (green), then exit the cell to the right through a small hole. There they interact with our 797 nm and 860 nm lasers. Fluorescence emitted at 413 nm, is detected by a photomultiplier tube (not shown), which is mounted underneath the antechamber (light blue).

5.5.3 Buffer Gas Results

Results of scanning the 797 nm laser frequency are shown in Figure 5-19. The two peaks are associated with the $J = 3/2$ and $J = 5/2$ levels of the D state. Their calculated splitting is 0.018 cm$^{-1}$, as described in 5.4.1. The separation of their measured central
peak values, as seen in Figure 5-19 is 0.02 cm\(^{-1}\). The predicted location of the \(J = 5/2\)
peak was 12546.606 cm\(^{-1}\) while the measured location was 12546.68 cm\(^{-1}\); the difference
of 0.074 cm\(^{-1}\) (2.2 GHz) is within the wavemeter accuracy of 0.1 cm\(^{-1}\). This experiment
successfully verified the 797 nm peak location and gave an overall validation of the
experimental technique of two-photon detection.

![Graph](image)

**Figure 5-19: Fluorescence vs. 797 nm laser frequency detuning, using the buffer gas cell
apparatus.** The two peaks correspond to the \(J=3/2\) and \(J=5/2\) levels of the D state. The
measured peak separation is 559 MHz; the calculated separation is 534 MHz. The peak
widths are 73 and 69 MHz. This is likely due to a combination of power broadening and
Doppler broadening.
5.6 Optimization

One of the potential areas for improvement of the two-photon excitation scheme is to attempt to minimize loss due to decay of the A state. Loss can occur when molecules which have been excited from X to A decay before they can be subsequently excited from A to D. One strategy for minimizing this loss is to employ a control protocol known as STImulated Raman Adiabatic Passage, or STIRAP. An excellent review of STIRAP can be found in [63]. STIRAP is based on the premise of transferring the molecular population from the initial state (in our case X) to the final state (D) without ever populating the potentially lossy intermediate state (A).

To completely transfer the population of the X state to the D state via the STIRAP method, and thus not lose any signal due to the decay of the A state, we must insure that at a minimum the following experimental conditions are met:

1) The lasers must be on resonance with a two-photon transition between X and D.

2) The laser phase must not change appreciably during the interaction time:
\[ 1/T < \Delta \nu, \]  
where \( T \) is the interaction time and \( \Delta \nu \) is the laser linewidth.

3) The power must be sufficiently high. The requirement can be expressed in the form: \( N_R = \Omega_R \cdot T > 10 \), where \( N_R \) is the number of Rabi cycles and \( \Omega_R \) is the Rabi frequency. The Rabi frequency for a single-photon transition is given
by $\Omega_r = \frac{dE}{2\pi\hbar}$, where $d$ is the dipole matrix element of the transition and the electric field $E$ is related to the laser intensity via $I = \frac{1}{2} c \varepsilon_0 E^2$. The factor of $2\pi$ in the denominator expresses it in Hz. Because we have two beams, the criterion becomes their combined Rabi cycle number [63]:

$$N_{R2\gamma} = \sqrt{(N_{797})^2 + (N_{860})^2} > 10.$$

4) Counterintuitive beam ordering: The 797 nm ($A \rightarrow D$) laser must begin to interact with the molecules before the 860 nm ($X \rightarrow A$) laser becomes appreciable. More precisely, the interaction with the 797 nm beam must turn on before the interaction with the 860 nm beam, then remain on during part of the time the interaction with the 860 nm beam is on, then turn off before the interaction with the 860 nm beam turns off. This is “counterintuitive” because the 797 nm beam is tuned to resonance between two states which initially have no population. The naïve expectation would have been that one must first move the molecules from the initial state to the intermediate state, then from the intermediate state to the final state. However, greater population transfer is reported to occur when the intermediate-to-final state laser precedes the initial-to-intermediate state laser. As is reviewed in [63], reported in [69], as well as seen in many of the studies listed in [62], a broad plateau of high transfer efficiency can be found when the intermediate-to-final state laser precedes the initial-to-intermediate state laser by a distance of around twice the beam diameter.
5.6.1 Linewidth

The linewidths of the 797 nm and 860 nm lasers were measured to be 940 kHz and 160 kHz respectively. This measurement was performed via the delayed self-heterodyne technique [70] using a device developed by C. Bruzewicz and S. Falke for the DeMille group. The basic idea is to send laser light modulated by an acousto-optic modulator (AOM) down two optical paths, one of which is five kilometers longer than the other. After following the separate paths the light is recombined and the output collected by a photodiode. The spectrum of the photodiode signal is related to the autocorrelation of the laser lineshape [71]. If the spectrum is fit to a Lorentzian, the laser linewidth is the FWHM divided by 2. This is derived rigorously in [71], but can be basically understood by realizing that the autocorrelation of a Lorentzian is a Lorentzian with twice the width. Results for both lasers can be found in Figure 5-20 and Figure 5-21. The 797 nm beam has a linewidth of 670 kHz, while the 860 nm beam has a linewidth of 120 kHz. D. Rahmlow has measured the velocity of molecules in our beam to be around 650 m/s. As shown in 5.4.2.2, our lasers are adjusted to have waists of 58 microns and 66 microns for the 797 nm and 860 nm beams respectively. Thus for our calculated molecular fly through times of $T(797) = 0.18 \mu s$ and $T(860) = 0.20 \mu s$, the constraints in (2) would require laser linewidths of 5.6 MHz and 5.1 MHz, a condition which is easily met.
Figure 5-20: Linewidth of the 860 nm laser. The FWHM when fit to a Lorentzian is 0.240 MHz, yielding a laser linewidth of 120 kHz.

Figure 5-21: Linewidth of the 797 nm laser. The FWHM when fit to a Lorentzian is 1.33 MHz, yielding a laser linewidth of 670 kHz.
5.6.2 Rabi Frequency

Complete population transfer cannot occur unless there is sufficient laser intensity. The intensity is conveniently parameterized in terms of the Rabi frequency \( \Omega_R = \frac{dE}{2\pi\hbar} \) described above. The transitions of interest in barium fluoride have dipole matrix elements with magnitude \( d \sim 1 \text{ eA}_0 \). The beam waist for the 797 nm beam, at the position where the molecular and laser beams intersect, is 58 \( \mu \text{m} \) in the horizontal direction along the beam, and 2.026 mm in the vertical direction. For the 860 nm beam, these numbers are 64 \( \mu \text{m} \) and 2.065 mm respectively. Using these waists, and nominal powers of 18 mW for the 797 nm beam and 6 mW for the 860 nm beam, our lasers have intensities of 98 kW/m\(^2\) for the 797 nm beam and 29 kW/m\(^2\) for the 860 nm beam. The Rabi frequencies for the two steps of the transition are thus expected to be roughly \( \Omega_{797} \sim 130 \) MHz for the 797 nm beam and \( \Omega_{860} \sim 70 \) MHz for the 860 nm beam. When multiplied by the interaction times found above, these give products of \( N_{797} = \Omega_{797} \cdot T_{797} \approx 23 \) and \( N_{860} = \Omega_{860} \cdot T_{860} \approx 14 \). Their combined Rabi cycle number is

\[
N_{R2y} = \sqrt{(N_{797})^2 + (N_{860})^2} \sim 27,
\]

thus meeting requirement (3) from above.

5.6.3 Counterintuitive Beam Order

We allow for the counterintuitive beam ordering by making the position of the 797 laser adjustable relative to that of the 860. The mechanism for this is described in 5.4.2.2. The
calibration of the relative positioning of the beams is described in 5.4.2.2, and the results of the experiment in 5.7.

5.7 Anapole Beam Experiment

5.7.1 Overview

Here we present the results of our optimization efforts using the actual beam from the anapole experiment as a molecule source. The following experiments were performed in the context of maximizing the signal in Chamber 3, with the additional goal of investigating the potential for STIRAP. Two distinct experiments were performed; the first was an attempt to ascertain whether or not we were saturating the transitions in the two-photon scheme, the second was an exploration of how our signal magnitude depended on the relative beam position. A third experiment was performed with the 860 nm laser far off resonance.

All of the following experiments depend on measurements of the fluorescence signal in Chamber 3. In order to ensure that variations in signal size due to laser intensity were not a contributing background, we measured the power of both lasers as a function of time. The results can be found in Figure 5-22. The standard deviation of both beams over four minutes is 0.03 mW.
Figure 5-22: Laser power vs. time as measured after output collimators on combination board. The power was intentionally turned down so as to be in the range of the power meter’s DC output. The standard deviation of both beams is 0.03 mW.

5.7.2 Saturation Experiment

By measuring the Chamber 3 fluorescence signal as a function of laser power, we can learn whether or not we are saturating our two-photon transition. If we are not saturating the transition it is likely that we can increase our signal by increasing our laser power. To investigate this we performed a series of measurements of the Chamber 3 fluorescence at a variety of powers for each of the two laser beams. The power of a given laser was controlled via the introduction of an absorptive neutral density filter into the beam line.
For the 797 nm beam the filter was positioned immediately following the PBS cube on
the combination board, while for the 860 nm beam the filter was positioned immediately
prior to entering the fiber. These locations were chosen for experimental convenience.

At each power we scanned the 797 nm laser frequency over a range of 120 MHz in 2
MHz steps. We simultaneously recorded the fluorescence signal in Chambers 2 and 3.
In order to control for variations in the ablation yield, we then normalized the signal
observed in Chamber 3 to that of Chamber 2. By fitting this ratio to a Gaussian
distribution and extracting the amplitude, we were able to characterize the Chamber 3
fluorescence signal size as a function of laser power.

The results for dependence of the signal size on 797 nm laser power can be seen in Figure
5-23, and a similar plot for the 860 nm laser power can be found in Figure 5-24. These
measurements give a strong indication that we are saturating the transitions.
Figure 5-23: Normalized Chamber 3 fluorescence signal vs. 797 nm laser beam power.

Figure 5-24: Normalized Chamber 3 fluorescence signal vs. 860 nm laser beam power.
5.7.3 Relative Position Experiment

One of the potential signatures of population transfer via STImulated Raman Adiabatic Passage (STIRAP) is an asymmetry in the signal size as a function of relative position between the laser beams. Specifically, it has been shown in many cases that an increase in total population transfer can be achieved when the molecules interact first with the laser driving the second step of the transition (in our case the 797 nm beam, exciting the $A \rightarrow D$ transition), and then with laser driving the first step (in our case the 860 nm beam, exciting the $X \rightarrow A$ transition). From the molecules' point of view, the 797 nm laser should turn on first, then the 860 nm laser should turn on, making both beams present. The 797 nm laser should then turn off, followed shortly by the 860 nm laser turning off. To accomplish this we spatially separate the laser beams, so that as the molecules propagate along the molecular beam they first encounter the 797 nm laser, then later the 860 nm laser. As described in 5.4.2.2, the overlap between the lasers can be varied by translating the dichroic mirror on the beam combination board.

We looked for evidence of STIRAP with our detection scheme by tuning the relative position of the beams to a given value, then scanning the 797 nm laser frequency. As described in [63], a plateau of high transfer efficiency should be found in the region near a relative separation of $2w_0$, where $w_0$ is the beam waist at the interaction point with the molecules. The waists of our lasers are 58 $\mu$m for the 797 nm beam and 64 $\mu$m for the 860 nm beam. Thus we expect maximum transfer efficiency near a beam separation around 120 $\mu$m. To look for this, we scanned the relative position over a range of $\pm 200$ $\mu$m in steps of 25 $\mu$m. At each step we swept the 797 nm laser frequency 130
MHz in 2 MHz steps across the peak. A typical scan can of the 797 nm laser frequency at a given relative laser beam separation can be seen in Figure 5-25. As in the saturation study described above (5.7.2), we controlled for variation in molecule production by normalizing the signal observed in Chamber 3 to that of Chamber 2. By fitting this ratio to a Gaussian distribution and extracting the amplitude, we were able to characterize the fraction of detected molecules as a function of beam separation. Figure 5-26 shows a plot of the normalized Chamber 3 signal as a function of relative beam position. In this plot we do not see evidence of STIRAP. The region of maximal transfer efficiency is centered near zero beam separation, and appears symmetric independent of laser ordering. This indicates that the factor of 2 to 4 enhancement in signal we would expect to see if the entire X state population were being transferred to the D state remains to be gained. One caveat to interpreting this plot is that scans similar to Figure 5-25, but at differing beam separation, sometimes show multiple peak structure. The origin of these peaks remains to be determined. If they are, for example, differing magnetic sublevels of the D state, we should be fitting the region of a given peak rather than fitting the entire curve to a single Gaussian. This same feature was observed in the data recorded for the saturation study. In the saturation study however the evidence was strong enough to indicate saturation that the importance of any multipeak structure was obviated.
Figure 5-25: Scan of 797 nm laser frequency. This scan was taken when the 797 nm beam preceded the 860 nm beam by 0.1 mm. The signal is normalized to the fluorescence in Chamber 2. A neutral density filter of ND = 0.5 was placed in the path of the 860 nm laser going to Chamber 2 in order to mitigate any reduction in Chamber 3 signal due to optical pumping.

Figure 5-26: Chamber 3 fluorescence as a function of the spatial separation between the 797 nm and 860 nm laser beams. The data give no indication of STIRAP.
5.7.4 Current Status

The implementation of the two-photon excitation has resulted in a significant improvement over the single photon scheme. Under the single photon scheme, typical signals in Chamber 3 were 1 to 3 counts per pulse depending on the degree of effort put into optimization. Under the two-photon scheme, typical non-optimized signals in Chamber 3 are 30 to 45 counts per pulse.

Comparing our expected gain in detection efficiency to our realized gain is not exact due to the uncertainties in characterizing the coupling efficiencies in the single photon scheme. This is particularly true for the geometric and light guide factors. Estimated expected improvements lie between factors of 15 and 800.

The factor of ~20 improvement we have achieved makes an important difference in terms of experimental convenience: the signals are now large enough that they can be monitored and optimized by eye in real time with minimal need for averaging. The truly critical difference however is in the implications this performance gain has for the parity violation measurement: now, for the first time, we have enough signal to try to make the anapole measurement in $^{137}$BaF.

In the future we expect to be able to improve this result even further. Once we find the right conditions for STIRAP, our signal should increase by a factor of 2 to 4. Also, optimizing parameters such as the beam combination board position or the light guide height could result in another factor of 2 to 3. In the long term, once the buffer gas beam
source technology matures enough to be installed in the anapole beam, the initial molecular beam flux could increase by two orders of magnitude.

5.7.5 Molecular Constant Tables

Table 5-2: Molecular constants in cm⁻¹ for $^{138}$Ba$^{19}$F taken from the effective constants table in [72]. v=0 in all cases.

<table>
<thead>
<tr>
<th></th>
<th>$X^2\Sigma_{v/2}$</th>
<th>$A^2\Pi_{v/2}$</th>
<th>$D^2\Sigma_{v/2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>T</td>
<td>0</td>
<td>11946.403 (1)</td>
<td>24176.608 (1)</td>
</tr>
<tr>
<td>B</td>
<td>0.2159509 (22)</td>
<td>0.2117889 (27)</td>
<td>0.2274137 (22)</td>
</tr>
<tr>
<td>A</td>
<td>x</td>
<td>632.161 (1)</td>
<td>x</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>2.7246×10⁻³ (61)</td>
<td>x</td>
<td>7.130×10⁻³ (25)</td>
</tr>
<tr>
<td>p</td>
<td>x</td>
<td>-0.257310 (17)</td>
<td>x</td>
</tr>
<tr>
<td>q</td>
<td>x</td>
<td>-0.0840×10⁻³ (29)</td>
<td>x</td>
</tr>
</tbody>
</table>
Chapter 6: Conclusion

Our study of nuclear spin-dependent parity nonconservation using Stark interference in diatomic molecules is an emerging method full of promise. What began in 2002 with literally an empty room is now an experiment poised to make a major impact on our understanding of nuclear weak forces and neutral weak couplings. The work described here has been an important stepping stone towards our long term goals of an “anapole factory” and the ensuing extraction of fundamental Standard Model parameters.

Our magnetic field can now be automatically shimmed to a uniformity better than 0.4 ppm, and with minor manual intervention this can be reduced to 0.03 ppm. Even the automatically shimmed field is now good enough to make a parity violation measurement.

Work on the magnetic field has produced new technology in the form of novel nuclear magnetic resonance probes. The potential utility of these probes, which are currently under review by the United States Patent and Trademark Office, extends well beyond our current physics experiment. We sincerely hope they may prove useful in other applications.

With the increase in detection efficiency due to the two-photon excitation scheme, we now for the first time have large enough signals to make a parity violation measurement.
In short, we have made strong progress towards our ultimate experimental goals. Naturally, there are several areas we can identify for further improvement. A clear understanding of why the maximum uniformity of magnetic field readings over the array does not correspond to the maximum uniformity of readings on axis will no doubt go far towards improving the shimming results. Furthermore, understanding why the two-photon detection efficiency is not as high as anticipated should help increase our signal sizes in Chamber 3.

While there is clearly much work left to be done to measure and understand nuclear spin dependent parity violation, the work described herein has been an important contribution to our experimental program.
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