EXPERIMENTAL DETERMINATION OF THE NUCLEAR MAGNETIC OCTUPOLE MOMENT OF $^{137}$Ba

NICHOLAS CHARLES LEWTY
MPhys (Hons) Physics with instrumentation, University of Leeds
M.Sc. Physics, University of Leeds

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Declaration

I hereby declare that the thesis is my original work and it has been written by me in its entirety. I have duly acknowledged all the sources of information which have been used in the thesis.

The thesis has also not been submitted for any degree in any university previously.

NICHOLAS C. LEWTY
January 29, 2014
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Undertaking a PhD project is a once in a lifetime expedition, and with all great expeditions one cannot reach the finishing line without a great support team. I would first like to offer my deepest gratitude to my doctoral adviser, Professor Murray Barrett without him, this PhD project would not exist. He is a perfect example of what is required to complete a PhD, setting an extremely high standard not only for himself, but for the rest of his group. He is also an ultimate source of technical knowledge for solving problems that one regularly encounters while undertaking a PhD. The knowledge I have gained from him will be instrumental in all of my future plans.

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This thesis describes the measurement of the nuclear magnetic octupole moment of $^{137}\text{Ba}$. The measurement utilizes radio frequency (rf) spectroscopy and optical shelving to chart the hyperfine structure of meta-stable 5D states in $^{137}\text{Ba}$ and provided the most precise value of the nuclear magnetic octupole moment in any atom to date.

The measurements are performed on a singly charged barium ion trapped within a linear Paul trap. The idea of the experiment is to measure the hyperfine intervals to a high precision. This then allows the extraction of the hyperfine constants, one of which, $(C)$, can be related to the nuclear magnetic octupole moment. Hence, the observation of the hyperfine $(C)$ constant constitutes observation of the octupole moment.

The first experiment involved measuring the hyperfine intervals of 5D$_{3/2}$ manifold to an accuracy of a few Hertz. The second experiment measured the hyperfine intervals of the 5D$_{5/2}$ manifold to almost the same accuracy as the 5D$_{3/2}$ manifold. The second experiment suffers from two additional error sources compared with the 5D$_{3/2}$ manifold measurement. The first error source is caused by the rf used to drive the trap causing an ac-Zeeman shift to the hyperfine intervals. The second comes from the hyperfine interaction mixing the two 5D manifolds together causing a magnetic field dependent perturbation. The method used for fitting the hyperfine structure in the 5D$_{5/2}$ manifold allows for the extraction of the Landé g-factor for that manifold. We obtained a value of $g_J$ that is an order of magnitude improvement over the next best measurement at the time.

These two hyperfine interval measurements give us three ways to obtain the octupole moment, providing a consistency test between the measurements. We found that all three values for the octupole moment agreed within their errors. All of this work culminated in establishing a nuclear magnetic octupole moment value of

$$\Omega(^{137}\text{Ba}^+) = 0.05061(56) \ (\mu_N \times b).$$
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The main results of this thesis have been reported in the following articles


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List of Acronyms

ac  alternating current

AOM  Acousto-optical modulator

AR  anti-reflective

ASE  Amplified Stimulated Emission

BBO  Beta Barium Borate

CCD  Charge Coupled Device

CPM  Critical Phase Matching

CQT  Center for Quantum Technologie

dc  direct current

ECDL  External Cavity Diode Laser

EOM  Electro-optical modulator

GPS  Global Positioning System

IR  Infra-Red

ITO  Indium Tin Oxide

LBO  Lithium Triborate

LD  Lamb-Dicke

NCPM  Non-critical Phase Matching
NIR  Near Infra-red
NSM  Nuclear Shell Model
PNC  Parity non-conservation
PBS  Polarizing Beam Splitter
PDH  Pound-Drever-Hall
PPKTP  Periodically Poled Potassium Titanyl Phosphate
PZT  Lead Zirconium Titanate crystal
QIP  Quantum Information Processing
rf  radio frequency
RMS  Root Mean Squared
RWA  Rotating Wave Approximation
SHG  Second Harmonic Generation
SPCM  Single Photon Counting Module
TEC  Thermo-electric Cooler
UV  ultra-violet
Chapter 1

Introduction

It is widely regarded that atomic physics was born when Ernest Rutherford attempted to test J.J. Thompson’s plum pudding model of the atom. Rutherford’s experiment discovered the atomic nucleus, which proved Thompson’s model was incorrect. Interplay like this is at the heart of scientific research. Theories are constructed that explain and predict observable processes, which in turn are tested in experiments. These experiments validate the theories, but sometimes are at odds with them and yield genuine new discoveries. From an experimental point of view, the more control there is over a physical system, the better it is suited to test theories and look for effects beyond current understanding.

In recent decades remarkable experimental control has been achieved over trapped ions and their coherent interactions with lasers. The high degree of control has made trapped ions an ideal system to probe atomic structure [1, 2]. They have also been used to perform basic quantum physics experiments [3, 4], which previously had only been envisioned as thought experiments. The high accuracy with which trapped ions can be manipulated is also the reason why they have become one of the best test beds for Quantum Information Processing (QIP) [5, 6, 7] and why they currently provide one of the best time standards [8, 9]. Accuracy in control and measurement has reached levels where it has become feasible to use the interaction of trapped ions with lasers to look for signatures beyond the Standard Model of particle physics [10, 11].

Given the large difference in energy between the nuclear interactions and the electromagnetic interactions, it is perhaps surprising that the tools of atomic physics would be able to probe the nuclear structure of atoms. Nevertheless, early optical spectra
measurements in 1884 by A.A. Michelson in thallium [12] revealed an unknown line splitting that could not be explained by physics at that time. In 1924 Wolfgang Pauli proposed the existence of a small nuclear magnetic moment. The nucleus can then be treated as a massive point charge that possesses intrinsic spin angular momentum $I$. This intrinsic nuclear spin generates an electro-magnetic field, which couples to the orbiting electrons and leads to a measurable energy level splitting, solving the mystery of the unknown spectral line splitting in thallium. This effect is known as the hyperfine interaction.

(a) (b) (c) (d)

Figure 1.1: Schematic of nuclear moments. (a) is the charge of the nucleus which the zeroth order moment, also known as the electric monopole. (b) is the magnetic dipole which is the first order moment. (c) is the electric quadrapole which is the second order moment. (d) is the magnetic octupole which is the third order moment.

This simple description for the hyperfine interaction is incomplete because the nucleus has internal structure. The internal structure means the electric and magnetic fields generated by the nucleus can not be described fully by a Coulomb potential and magnetic dipole field. Instead, the structure allows higher order electric and magnetic multipole moments of the fields to exist. The nuclear structure can be expanded in terms of these higher order moments via the multipole expansion [13]. These multipole moments couple to the orbiting electrons and produce higher order corrections to the energies of the hyperfine sub levels, resulting in a deviation from the magnetic dipole interaction. The first four nuclear moments are shown schematically in Fig. 1.1. The next nuclear moment after the magnetic dipole is the electric quadrapole and its existence was confirmed by measurement in indium in 1937 [14]. After the electric quadrapole comes the magnetic octupole and this was observed in iodine in 1954 [15]. Since then, the nuclear magnetic octupole moment has been observed in a host of other atoms, for example in Gd[16], Eu[17], Cs[18], Rb[19] and Yb[20]. These higher order
nuclear moments were observed through measurements of the hyperfine structure. This is linked to the nuclear moments through the hyperfine structure constants, where the nuclear magnetic dipole moment is related to the hyperfine (A) constant, the electric quadrupole to the hyperfine (B) constant and the magnetic octupole to the hyperfine (C) constant [21]. Hence, it is possible to obtain information about the nuclear structure through measurements of the hyperfine structure [22].

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Schematic showing fine structure and hyperfine structure splittings of the 5D manifolds of $^{137}$Ba$^+$.}
\end{figure}

In order to measure the nuclear multipole moments the hyperfine splittings must be measured to a very high precision as the nuclear moments after the magnetic dipole only lead to a small perturbation. The size of the perturbation gets smaller the higher the order the nuclear moment. Beyond observing the nuclear octupole moment, singly ionized barium is a good candidate for observing possible physics beyond the standard model, because it is an alkali-like atom. Being alkali-like means singly ionized barium only has one valance electron, which means its electronic structure is simple and well understood. This makes it simpler to work with and allows for high accuracy atomic structure calculation to be performed [23]. Barium has a large nucleus $Z = 56$ meaning that it can take advantage of the heavy atom scaling $Z^3$ [24] in the application to a Parity non-conservation (PNC) measurement [25] at low energy. Barium is also an excellent candidate for observing possible fluctuation in the fine structure constant $\alpha$ [26], due to its large fine structure splitting depicted in Fig. 1.2. Since all these measurements rely on precision measurements of the level structure of Barium, observing the octupole moment will equip the laboratory with the technologies required for performing precision atomic structure measurements.
Furthermore, measurement of the nuclear magnetic octupole moment may help to spur the development of a new model to describe the nucleus, which would replace the Nuclear Shell Model (NSM). The NSM is analogous to the atomic shell model and was developed in 1949 following independent work by several physicists [27, 28]. However, comparison between the nuclear octupole moment for $^{133}$Cs [18] yielded a factor of 40 discrepancy between the model and the measured value, suggesting it is only relevant for estimating the magnetic dipole moment. A high accuracy measurement of nuclear octupole moment in $^{137}$Ba$^+$ would act as a rigorous test for any new theories developed.

The measurement of the magnetic octupole moment in a trapped Ba$^+$ has been proposed in [29]. To observe the nuclear octupole moment the hyperfine structure of the low lying metastable 5D states shown in Fig. 1.2, we would need to measure the levels down to an accuracy better than 10 Hz. Although the hyperfine intervals can be measured in a variety of ways, the simplest and most accurate one is to use rf spin resonance techniques [30]. Measurement of the hyperfine structure from one manifold would lead to a value for the octupole moment, but this value would include a theoretical correction factor. A correction factor free measurement of the octupole moment can be achieved by measuring the hyperfine structure of both 5D manifolds. In this research project, we aim to determine the hyperfine structure constants of both 5D manifolds to an accuracy of below a few Hz by measuring the hyperfine intervals, with a combination of high precision rf spectroscopy and shelving techniques [31, 32] on singly trapped $^{137}$Ba$^+$. The measurement of the hyperfine intervals from both manifolds yields three different values for the octupole moment, one from each manifold and a combination of the two. These three different values can be used as a self consistency check. Furthermore, comparing measured hyperfine structure constants obtained from hyperfine intervals with calculated values allows one to experimentally assess the accuracy of the structure calculations [33]. These calculations play a crucial role in the interpretation of experiments looking for physics beyond the standard model.

1.1 Thesis outline

The nuclear magnetic octupole moment is measured in two different experiments, one measuring the hyperfine intervals of the $5D_{3/2}$ manifold and the other measuring the intervals of the $5D_{5/2}$ manifold. These two measurements rely on the same procedures,
which are described in general terms preceding the measurement chapters. The two hyperfine interval measurements are split into two separate chapters to highlight the different problems faced with each measurement. The organization of this thesis follows the structure: theory, experimental apparatus, experimental methods, measurements, results and conclusion.

In Chapter 2, we start with the theoretical discussion of the hyperfine interaction. This discussion develops further to include the effect of the hyperfine mixing between the two 5D manifolds, which are separated by the fine structure splitting. From here we move onto the theoretical considerations that go into performing coherent transitions. The hyperfine interval measurement relies heavily on the ability to perform coherent state transitions and rf spectroscopy between the hyperfine states.

In Chapter 3, we cover the equipment required to perform the measurement of the hyperfine intervals. We start with the vacuum system, ion trap and oven, which are used for creating and confining the ion. We next discuss the lasers systems that are used to manipulate the ions. To access all the required wavelengths we use diode lasers and frequency doubled diode lasers. For maintaining the laser at a fixed frequency, we utilize the stability of optical reference cavities. In this chapter we also cover the method we use to photo-ionize barium. Next we describe the rf source used for driving the hyperfine transitions. Fluorescence detection is used to check whether an rf transition occurred and we conclude this chapter by describing the imaging system used to collect the fluorescence.

Chapter 4 covers the experimental procedure used to conduct the hyperfine interval measurements. We start with optical pumping which is used to prepare the ion in a well defined state. We then move onto the more advanced technique of two color Raman transitions. Next we discuss the procedure of measuring the hyperfine energy splitting using rf transitions. The chapter progresses to state detection, which is necessary to detect whether a hyperfine transition took place.

The measurements performed in this thesis are covered in Chapter 5 and Chapter 6. Chapter 5 concentrates on the hyperfine interval measurements performed in the 5D_{3/2} manifold. It also covers the majority of the errors sources encountered in the octupole measurement. Chapter 6 covers the the hyperfine interval measurements performed in the 5D_{5/2} manifold. In this chapter the influence of the alternating current (ac) Zeeman shift due to the trap is explained in detail. We also give an account of how
the hyperfine mixing between the two 5D manifolds affect the measurement. Finally, we conclude this chapter by presenting the measured hyperfine structure constants.

The thesis is summarized in Chapter 7, where we present and compare the three values we obtained for the the nuclear magnetic octupole moment. We also discuss how the value of the octupole moment compares with theory. We then discuss future research directions we intend to investigate with the setup developed in this research project.
Chapter 2

Theory

In this chapter we cover the important theoretical concepts that underpin the physics of the experiment. We begin with a discussion of the hyperfine structure in Section 2.1. Although we are interested in the hyperfine intervals at zero magnetic field, measurements are performed at a finite field. This further changes the level structure which is covered in Section 2.2. Finally we discuss coherent interactions with electro-magnetic fields in Section 2.3.

2.1 Hyperfine interaction

The coarse atomic energy structure arises from interactions of the electrons with the spherically symmetric Coulomb potential, due to the electric monopole of the nucleus. The exact level structure however, depends on the electrons’ interaction with the higher order moments of the nucleus, which gives rise to the hyperfine structure. Thus, measuring the hyperfine intervals very accurately can yield information about the structure of the nucleus. The first three moments beyond the electric monopole are the magnetic dipole, electric quadrapole and magnetic octupole moments respectively. In general, a nucleus of spin $I$ posses $2I$ of these moments. The hyperfine interaction then can be viewed as the perturbation to energy of the electrons moving in the field of the monopole, due to these higher order moments.

Observing higher order nuclear moments is very challenging because the higher the moment, the smaller the associated shift to the energy levels. In addition, the $2^k$-pole moment only contributes in first order to hyperfine levels with electronic angular
moment $J \geq \frac{k}{2}$ [34]. Thus in order to observe the nuclear octupole moment, $k = 3$, we need to work with an atom of $I \geq \frac{3}{2}$ and in a manifold with $J \geq \frac{3}{2}$. Furthermore, measuring the hyperfine intervals to the level necessary to detect the effect of the octupole requires long integration times. All these requirements are satisfied by the two low lying 5D manifolds in $^{137}\text{Ba}^+$, whose structure is shown in Fig. 2.1.

Figure 2.1: Hyperfine states of 5D manifolds showing hyperfine intervals as $\delta W_k$, where $k = 0, 1, 2$ for the 5D $\frac{3}{2}$ manifold and $k = 1, 2, 3$ for 5D $\frac{5}{2}$ manifold. The 5D $\frac{3}{2}$ manifold contains a total of 16 magnetic sub-states and the 5D $\frac{5}{2}$ manifold a total of 24.

2.1.1 Hyperfine structure

We follow the treatment presented in [35] to describe the hyperfine interaction. The hyperfine interaction can be expressed as a sum over scalar products of spherical tensors

$$H_{\text{HFI}} = \sum_k T^n_k \cdot T^e_k,$$

(2.1)

where $T^n_k$ and $T^e_k$ are spherical tensors of rank $k$ acting in the nuclear and electronic spaces respectively and are defined in [35]. As $H_{\text{HFI}}$ is a scalar operator in the combined space, it is convenient to work in the conventional basis formed by coupling nuclear, $|I m_I\rangle$, and electronic, $|J m_J\rangle$, states

$$|\gamma IJ F m_F\rangle = \sum_{m_I, m_J} C^{F m_F}_{I m_I, J m_J} |I m_I\rangle |J m_J\rangle,$$

(2.2)

where $C^{F m_F}_{I m_I, J m_J}$ are Clebsch-Gordon coefficients and $\gamma$ encapsulates the remaining electronic quantum numbers. In order to calculate the necessary dipole elements, we can apply the Wigner-Eckert on the nuclear and electronic spaces, which gives

$$\langle \gamma' I' J' F' m'_F |H_{\text{HFI}}| \gamma IJ F m_F\rangle = \delta_{F' F} \delta_{m'_F, m_F} (-1)^{I' + J' + F} \times \sum_k \left\{ I \atop J' \atop I \atop k \right\} \langle I || T^e_k || I \rangle \langle I || T^n_k || I \rangle \langle \gamma' J' || T^e_k || \gamma J \rangle,$$

(2.3)
where \{\} is the Wigner 6J symbol. Here we follow the normalization convention for the reduced matrix elements \( \langle I || T_k^m || I \rangle \) and \( \langle \gamma' J' || T_k^m || \gamma J \rangle \) such that the Wigner-Eckert theorem takes the form

\[
\langle \gamma' J' m' || T_{k,q} || \gamma J \rangle = (-1)^{2k} \sqrt{2j+1} \langle \gamma' J' || T_k || \gamma J \rangle \langle j' m' || j m ; k q \rangle
\] (2.4)

where \( T_q \) is the \( q \) component of the spherical tensor of rank \( k \), \( T^{(k)} \), \( j, j' \) are angular momenta with \( m \) and \( m' \) their respective projections along the quantization axis. Note that from this convention it follows that

\[
\langle \gamma' J' || T_k || \gamma J \rangle = (-1)^{(j-j')} \langle \gamma J || T_k || \gamma' J' \rangle^* \] (2.5)

which leads to the change in sign of the off diagonal elements in Table 2.1 and its conjugate\(^1\).

In order to calculate the energy corrections to the state described by quantum numbers \( \gamma \) and \( J \) we treat \( H_{\text{HF}} \) as a small perturbation. First order corrections are then given by

\[
W_F^{(1)} = \langle \gamma J F m_F || H_{\text{HF}} || \gamma J F m_F \rangle = (-1)^{I+J+F} \sum_k \{ \begin{array}{ccc} I & J & F \\ J & I & k \end{array} \} \langle I || T_k^m || I \rangle \langle \gamma J || T_k^m || \gamma J \rangle.
\] (2.6)

The first order correction corresponds to the conventional expansion in terms of the hyperfine constants, which in turn are related to the nuclear moments. Focusing on the first three terms of the sum in Eq. (2.6) we can define the hyperfine constants (A), (B), (C) and relate them to the respective nuclear moments. We get

\[
A = \langle T_1^m || I (T_1^e) J \rangle = \frac{\mu}{I J} \langle T_1^e || \rangle J,
\] (2.7)

\[
B = 4 \langle T_2^m || I (T_2^e) J \rangle = 2Q \langle T_2^e || \rangle J
\] (2.8)

and

\[
C = \langle T_3^m || I (T_3^e) J \rangle = -\Omega \langle T_3^e || \rangle J,
\] (2.9)

\(^1\) Using this normalization procedure leads to Eq. (2.3) being in agreement with Eq. (3) from [35]. However, in earlier work by the same authors there is a sign difference in Eq. (3) of [34] which disagrees by a factor of \((-1)^{(j-j')}\) with Eq. (2.3).
where $\mu$ represents the magnetic dipole, $Q$ the electric quadrapole and $\Omega$ the magnetic octupole moment. $\langle T^n_k \rangle_l$ is the expectation value of the zero-component operator of spherical tensors $T_k$ in the “stretched state”. This is related to the reduced matrix element through the expression
\[
\langle T_k \rangle_l = \left( \begin{pmatrix} I & k & I \\ -I & 0 & I \end{pmatrix} \right) (I || T^n_k || I),
\]
where $(\cdot)$ is the Wigner 3J symbol. From atomic structure calculations [33, 36], the expectation value of the spherical tensors in the electronic space are computed and are given in Table 2.2.

To the accuracy to which we perform our measurements it is necessary to consider second order corrections to the energy of the states described by $\gamma$ and $J$. In general, these second order contributions include coupling between states of different $J$ and are given by
\[
W^{(2)}_F = \sum_{\gamma', J'} \frac{\langle \gamma' J' F' m_F' | H_{HFI} | \gamma J F m_F \rangle \langle \gamma J F m_F | H_{HFI} | \gamma' J' F' m_F' \rangle}{E_{\gamma J} - E_{\gamma' J'}},
\]
where $(\cdot)$ is the Wigner 3J symbol. From atomic structure calculations [33, 36], the expectation value of the spherical tensors in the electronic space are computed and are given in Table 2.2.

To evaluate these expressions we use $\langle D_{5/2}^0 || T^0_1 || D_{5/2}^0 \rangle$ and $\langle D_{5/2}^0 || T^0_2 || D_{5/2}^0 \rangle$ as given in Table 2.1 and use the values of the dipole moment [37]
\[
\mu = 0.937365(20) \mu_N
\]
and the quadrupole moment [38]
\[
Q = 0.235(3) \text{ b},
\]
where $\mu_N$ is the Bohr magneton and b is the barn unit of area. The coefficients in front of $\eta$ and $\zeta$ in Eqs. (2.18) to (2.23) can be found from the Wigner 6J symbols
\[
\frac{\cal c}_\eta^{(W_F)} = \left\{ \begin{pmatrix} 3/2 & 5/2 & F \\ 3/2 & 3/2 & 1 \end{pmatrix} \right\}^2,
\]
\[
\frac{\cal c}_\zeta^{(W_F)} = \left\{ \begin{pmatrix} 3/2 & 5/2 & F \\ 3/2 & 3/2 & 1 \end{pmatrix} \right\} \left\{ \begin{pmatrix} 3/2 & 5/2 & F \\ 3/2 & 3/2 & 2 \end{pmatrix} \right\}.
\]
Table 2.1: Off diagonal elements of electronic spherical tensors of rank k (k>0) $T_k^e$ [40].

<table>
<thead>
<tr>
<th>Elements</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle D_0^{5/2}</td>
<td></td>
<td>T_1^e</td>
</tr>
<tr>
<td>$\langle D_0^{5/2}</td>
<td></td>
<td>T_2^e</td>
</tr>
<tr>
<td>$\langle D_0^{5/2}</td>
<td></td>
<td>T_3^e</td>
</tr>
</tbody>
</table>

By evaluating higher order corrections beyond $\eta$ and $\zeta$ one can show that these are much smaller and negligible for the scope of this work [39].

Combining the first and second order we get energy corrections $W_F = W_F^{(1)} + W_F^{(2)}$, and we can use Eqs. (2.6) and (2.11) to express the hyperfine energy intervals $\delta W_F = W_F - W_{F+1}$, as follows:

$$\delta W_0^{(3/2)} = -A + B - 56C - \frac{1}{100} \eta + \frac{\zeta}{100} \sqrt{\frac{7}{3}},$$

$$\delta W_1^{(3/2)} = -2A + B + 28C - \frac{1}{75} \eta,$$

$$\delta W_2^{(3/2)} = -3A - B - 8C - \frac{1}{300} \eta - \frac{\zeta}{20} \sqrt{\frac{3}{7}},$$

$$\delta W_1^{(5/2)} = -2A + \frac{4}{5} B - \frac{96}{5} C - \frac{1}{75} \eta,$$

$$\delta W_2^{(5/2)} = -3A + \frac{9}{20} B + \frac{81}{5} C - \frac{1}{300} \eta - \frac{1}{20} \sqrt{\frac{3}{7}} \zeta,$$

$$\delta W_3^{(5/2)} = -4A - \frac{4}{5} B - \frac{32}{5} C + \frac{2}{75} \eta + \frac{2}{25} \sqrt{\frac{3}{7}} \zeta.$$ (2.23)

Note that these relations give the hyperfine (A), (B) and (C) in terms of the measurable hyperfine intervals, $\delta W_k^{(J)}$. For example the hyperfine constant (C) for the 5D3/2 and 5D5/2 manifolds are given by

$$C(5D_{3/2}) = -\frac{1}{80} \delta W_0^{(3/2)} + \frac{1}{100} \delta W_1^{(3/2)} - \frac{1}{400} \delta W_2^{(3/2)} - \frac{1}{2000\sqrt{21}} \zeta.$$ (2.24)

and

$$C(5D_{5/2}) = \frac{1}{40} \delta W_1^{(5/2)} + \frac{1}{35} \delta W_2^{(5/2)} - \frac{1}{112} \delta W_3^{(5/2)} + \frac{1}{200\sqrt{21}} \zeta,$$ (2.25)

respectively. From Eq. (2.9) these constants are related to the nuclear magnetic octupole moment via the matrix elements $\langle T_3^e \rangle_J$ given in Table 2.2. Thus,

$$C(5D_{3/2}) = 0.584(6) \left( \frac{\Omega}{\mu_N \times b} \right) \text{kHz}.$$ (2.26)
Table 2.2: Diagonal elements of electronic spherical tensors of rank $k$ ($k > 0$) $T_k^e$.  

<table>
<thead>
<tr>
<th>Elements</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle T_1^e \rangle_{3/2}$</td>
<td>454.5(15)</td>
<td>MHz/$\mu_N$</td>
</tr>
<tr>
<td>$\langle T_1^e \rangle_{5/2}$</td>
<td>−46.8(20)</td>
<td>MHz/$\mu_N$</td>
</tr>
<tr>
<td>$\langle T_2^e \rangle_{3/2}$</td>
<td>94.9(13)</td>
<td>MHz/b</td>
</tr>
<tr>
<td>$\langle T_2^e \rangle_{5/2}$</td>
<td>126.9(18)</td>
<td>MHz/b</td>
</tr>
<tr>
<td>$\langle T_3^e \rangle_{3/2}$</td>
<td>−584(6)</td>
<td>Hz/((\mu_N \times b))</td>
</tr>
<tr>
<td>$\langle T_3^e \rangle_{5/2}$</td>
<td>250(10)</td>
<td>Hz/((\mu_N \times b))</td>
</tr>
</tbody>
</table>

*From a private communication with B.K. Sahoo.

and

$$C(5D_{5/2}) = -0.25(1) \left( \frac{\Omega}{\mu_N \times b} \right) \text{kHz}. \quad (2.27)$$

These expressions provide an independent determination of the octupole moment and a consistency check in the associated atomic structure calculations. Combining the measurements of the hyperfine ($C$) constants from the $5D_{3/2}$ and $5D_{5/2}$ manifolds, the correction factor, $\zeta$, can be eliminated resulting in the equations

$$C(5D_{3/2}) + \frac{1}{10} C(5D_{5/2}) = -\frac{1}{80} \delta W^{(3/2)}_0 + \frac{1}{100} \delta W^{(3/2)}_1 - \frac{1}{400} \delta W^{(3/2)}_2$$

$$- \frac{1}{400} \delta W^{(5/2)}_1 + \frac{1}{350} \delta W^{(5/2)}_2 - \frac{1}{1120} \delta W^{(5/2)}_3 \quad (2.28)$$

and

$$C(5D_{3/2}) + \frac{1}{10} C(5D_{5/2}) = 0.559(6) \left( \frac{\Omega}{\mu_N \times b} \right) \text{kHz}, \quad (2.29)$$

which provides a value for the octupole moment independent of second order corrections. However, we note Eq. (2.29) is still limited by the accuracy of atomic structure calculations.

### 2.2 Effect of external magnetic fields on the hyperfine structure

To first order, the effect of an external magnetic field can be viewed as a linear Zeeman shift, but due to the accuracy with which we measure the hyperfine intervals, we must
go to a more accurate description that includes second order Zeeman shifts. In addition, the picture that the magnetic field causes a small perturbation to the hyperfine levels only holds in the weak field limit, where the Zeeman energy is much smaller than the hyperfine splitting. In the 5D_3/2 manifold, we work in the weak field limit so the effect of the magnetic field on the hyperfine structure can be modeled using first and second order perturbation theory. This is discussed in Section 2.2.1. For the 5D_5/2 manifold, however, the close proximity of the hyperfine states does not permit a simple perturbation type analysis and a full theory of the hyperfine plus Zeeman interaction must be used. This is described in Section 2.2.2.

2.2.1 Effect of magnetic field on 5D_3/2 manifold

In the weak field limit the first order Zeeman shift to the hyperfine levels in an atom with nuclear spin caused by an external magnetic field is given by [41]

$$\Delta E_z = g_F \mu_B m_F B. \quad (2.30)$$

The Landé $g_F$ factor is given by

$$g_F = g_J \frac{F(F + 1) + J(J + 1) - I(I + 1)}{2F(F + 1)} + g_I \frac{F(F + 1) - J(J + 1) + I(I + 1)}{2F(F + 1)}, \quad (2.31)$$

where $g_J$ and $g_I$ are the Landé $g$-factors for the total electronic angular momentum and the nuclear spin angular momentum respectively. For the special case of the 5D_3/2 state, $I = J$ and Eq. (2.31) reduces to

$$g_F = \frac{g_J + g_I}{2} \quad (2.32)$$

which is independent of $F$. We measure the hyperfine intervals between the $m_{F'} = m_F = 0$ states. To first order, $m_F = 0$ states have no field dependence. In this case, the Zeeman shift from second order perturbation theory is given by

$$\Delta E_{zF}^{(2)} = \sum_{F'} \frac{|\langle (IJ)Fm_F | g_J \mu_B J_z B | (IJ)F'm_{F'} \rangle|^2}{E_F - E_{F'}} \quad (2.33)$$

which has a quadratic form. The resulting second order Zeeman shift coefficients for the 5D_3/2 states are given in Table 2.3.
Figure 2.2: Magnetic field dependence of the $m_F$ states in the 5D$_{5/2}$ manifold for the $F = 3$ and $F = 4$ hyperfine levels. The states in the legend represent the hyperfine states in the weak field limit.
Table 2.3: Second order Zeeman shift coefficients for 5D$_{3/2}$ manifold

<table>
<thead>
<tr>
<th>Interval</th>
<th>Second order Zeeman shift</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\delta W_0$</td>
<td>$-18.58$ kHz/G²</td>
<td></td>
</tr>
<tr>
<td>$\delta W_1$</td>
<td>$5.719$ kHz/G²</td>
<td></td>
</tr>
<tr>
<td>$\delta W_2$</td>
<td>$1.155$ kHz/G²</td>
<td></td>
</tr>
</tbody>
</table>

2.2.2 Effect of magnetic field on 5D$_{5/2}$ manifold

In the 5D$_{5/2}$ manifold the Zeeman interaction can no longer be viewed as a weak perturbation because hyperfine intervals are smaller than in the 5D$_{3/2}$. The $F = 3$ and $F = 4$ states are only separated by 490 kHz [12] and the hyperfine states become highly mixed even for small fields. The $F = 1$ and $F = 2$ states have a larger splitting, but nonetheless their energies significantly deviate from those given by a simple perturbation treatment. Therefore, the Zeeman and hyperfine interaction must be treated on an equal basis. To find energies we diagonalize the full Hamiltonian $H_{\text{HFI}} + H_z$, which is described in further detail in appendix A.

From this Hamiltonian, the magnetic field dependence of the $m_F$ states of the $F = 3$ and $F = 4$ states is mapped out and is shown in Fig. 2.2. Note that, even for small magnetic fields, there is a significant amount of mixing between the $F = 3$ and $F = 4$ states. As a consequence, the shifts of the states which in the zero field limit would correspond to $|F'' = 3, m_{F''} = 0\rangle$ and $|F'' = 4, m_{F''} = 0\rangle$, but at intermediate field are mixed with other Zeeman states, are large and therefore the accuracy in determining the hyperfine intervals is greatly limited by magnetic field fluctuations. Instead it is much more favorable to use states $|\pm\rangle$, as proposed in [29], which in the zero field limit would correspond to $|F'' = 3, m_{F''} = -1\rangle$ and $|F'' = 4, m_{F''} = 1\rangle$.

In the magnetic field range of $0.4 - 2$ G, where we perform our measurement, these states are weakly dependent on the magnetic field as shown in Fig. 2.2. Over the same magnetic field range, $|1\rangle \equiv |F'' = 1, m_{F''} = 0\rangle$, and $|2\rangle \equiv |F'' = 2, m_{F''} = 0\rangle$ are also only weakly dependent on the magnetic field. Therefore measurements are carried out on the transitions $|2\rangle \leftrightarrow |1\rangle$ and $|2\rangle \leftrightarrow |\pm\rangle$. The magnetic field dependence of these transitions are shown in Fig. 2.3 (a), (b) and (c). The magnetic field dependence of the transition between the $|2\rangle \leftrightarrow |F'' = 3, m_{F''} = 0\rangle$ state is shown in Fig. 2.3 (d) to demonstrate its large magnetic field dependence (do note the scale of the axis is in
kHz) and its deviation from a simple quadratic structure represented by the dashed line. It is also worth noting that the $|2\rangle \leftrightarrow |-\rangle$ transition contains two turning points in its dependence on the magnetic field and the $|2\rangle \leftrightarrow |+\rangle$ transition contains one. The $|\rangle$ state containing two turning points is a consequence of mixing with the $F = 1$ and $F = 2$ levels. If mixing with these states is neglected as done in [29], then the $|-\rangle$ state only contains one turning point.

Figure 2.3: Plots of the hyperfine intervals of 5D$_{5/2}$ manifold as a function of magnetic field for (a) $|2\rangle \leftrightarrow |1\rangle$, (b) $|2\rangle \leftrightarrow |-\rangle$ and (c) $|2\rangle \leftrightarrow |+\rangle$. (d) $|2\rangle \leftrightarrow |F'' = 3, m_{F'} = 0\rangle$. The dashed line represents the magnetic field dependence expected from second order perturbation theory.
2.3 Coherent photon-ion interactions

Coherent state manipulation is a general necessity in the majority of atomic physics experiments such as, QIP [43, 44, 45], atomic clocks [46, 47], ground state cooling [48, 49] and precision spectroscopy measurements [30, 40, 50, 51]. In this research project we perform coherent rf transitions between hyperfine states and coherent Raman transitions between the 6S_{1/2} manifold and the 5D manifolds using optical transitions.

In this section we give a brief description of the theory describing these processes. A more detailed description of this interaction can be found in most atomic physics textbooks [52, 53]. The discussion presented here has been split into four parts. In Section 2.3.1, we cover the theory describing the interaction between a two-level atom and monochromatic electro-magnetic radiation. In Section 2.3.2 we apply the two-level atom model to rf hyperfine transitions. In Section 2.3.3, we show that Raman transitions involving three levels can be simplified to a two-level model by adiabatic elimination of the excited state. In Section 2.3.4, we discuss decoherence mechanisms that affect the efficiency of coherent state manipulation.

2.3.1 Two-level atom

In this section we give a brief summary of the theory describing a two-level atom, highlighting the important results. More detailed treatments are given in [41, 53, 54]. Throughout this summary we will ignore spontaneous emission from the excited state.

A two-level atom consists of a ground state, \( |g\rangle \), and an excited state, \( |e\rangle \), separated by an energy splitting of \( \hbar \omega_0 \). These two levels can be coupled together by the application of an electro-magnetic field oscillating at a frequency \( \omega_l \), where the atom-field detuning is defined as \( \delta = \omega_l - \omega_0 \). For the resonant case, \( \delta = 0 \), the state of atom will oscillate between the \( |g\rangle \) state and the \( |e\rangle \) state at a frequency equal to the Rabi rate \( \Omega \), characterizing the strength of the interaction. These Rabi oscillations correspond to the periodic stimulated absorption and stimulated emission of radiation.

In the case of non-zero detuning, \( \delta \), and in the Rotating Wave Approximation (RWA) [54] the probability of being in the \( |g\rangle \) or \( |e\rangle \) state for an atom initially in \( |g\rangle \) state at
\[ |e\rangle \quad \delta \quad |g\rangle \]

\[ \Omega, \omega_l \quad \omega_0 \]

Figure 2.4: The two-level atom. The energy levels \( |e\rangle \) and \( |g\rangle \) are separated by a transition frequency of \( \omega_0 \). The atom is driven by a monochromatic plane wave of frequency \( \omega_l \). The atom-light interaction is governed by two key parameters, the detuning \( \delta = \omega_l - \omega_0 \) and Rabi rate \( \Omega \), which describes the strength of the coupling.

\[ t = 0 \] is given by

\[ |c_g(t)|^2 = 1 - \frac{\Omega^2}{\Omega^2 + \delta^2} \sin^2 \left( \frac{t}{2} \sqrt{\frac{\Omega^2}{\delta^2}} \right), \tag{2.34} \]

\[ |c_e(t)|^2 = \frac{\Omega^2}{\Omega^2 + \delta^2} \sin^2 \left( \frac{t}{2} \sqrt{\frac{\Omega^2}{\delta^2}} \right), \tag{2.35} \]

respectively, where \( t \) is the interaction time. On resonance, a pulse of duration \( T = \pi / \Omega \) is known as a \( \pi \) pulse, which will cause the two-level atom to undergo a state transfer from \( |g\rangle \) to \( |e\rangle \).

For the off-resonant case \( \delta \gg \Omega \), the probability of the atom undergoing a state transfer approaches zero. However the atom-field interaction causes a shift to the energy levels. In a two-level atom the corresponding shift to \( |g\rangle \) is

\[ \Delta E_g = \frac{\hbar \Omega^2}{4\delta}, \tag{2.36} \]

which is known as an ac-Stark shift. The ac-Stark shift is one of the most significant error sources encountered in this work.

### 2.3.2 Rf transitions

Rf transitions between hyperfine states can be modeled as a two-level atom interacting with an electro-magnetic field, which is described in Section 2.3.1. This simplification
to a two-level system is valid as long as the detuning of the rf frequency to transitions to other states is much larger than the Rabi rate $\Omega$, coupling the states of interest. Furthermore the lifetime of the hyperfine state in the 5D manifolds is very long so spontaneous emission can be neglected.

Transitions between hyperfine state $|F_m \rangle$ and $|F_m' \rangle$ are coupled by a magnetic dipole transition and the strength of this coupling is given by the Rabi rate \[ \Omega = \frac{g_J \mu_B B_0}{\hbar} |\langle (IJ)F'm_F'|J_q|(IJ)Fm_F \rangle|, \] (2.37)
where $B_0$ is the amplitude of the magnetic field, $|\langle (IJ)F'm_F'|J_q|(IJ)Fm_F \rangle|$ is the dipole matrix element between the two states, and $J_q$ is the operator coupling the states, where $q$ takes the values $q = (+, -, z)$. We define this operator in appendix D and also provide the relevant matrix elements.

In addition, each state has an ac-Zeeman shift due to off-resonant coupling to other Zeeman states in the 5D manifolds. The ac-Zeeman shift is found by summation over all possible state couplings \[ \Delta E_{(F,m_F)} = (g_J \mu_B B_0)^2 \sum_{F',m_F',q} \frac{|\langle (IJ)F'm_F'|J_q|(IJ)Fm_F \rangle|^2}{E_{(F,m_F)} - E_{(F',m_F')} - \hbar \omega_l}, \] (2.38)
where the matrix elements $\langle (IJ)F'm_F'|J_q|(IJ)Fm_F \rangle$ are given in appendix D. This ac-Zeeman shift will be a source of systematic errors in the measurement of the hyperfine intervals. In the description of a two-level atom the RWA is used, which neglects counter rotating terms. Inclusion of these terms tends to shift energy levels apart by \[ \Delta E_{BS}^{g,e} = \pm \frac{\hbar \Omega^2}{4(\omega_0 + \omega_l)}, \] (2.39)
where the minus sign goes with the $|g\rangle$ state and the plus sign with the $|e\rangle$ state. This is known as the Bloch-Siegert shift and is often important for rf transitions where $\Omega \sim \omega_l$ is possible [55]. For example in $^{137}$Ba$^+$ the smallest hyperfine interval frequency measured is $\delta W_2^{(5/2)} \approx 60$ MHz and for Rabi rates $\Omega$ approaching 20 kHz this effect can become a significant shift.

2.3.3 Two photon Raman transitions
A Raman transition is used to coherently transition population from one long lived state $|g_1\rangle$ to a second long lived state $|g_2\rangle$ via a virtual state. The virtual state is
Figure 2.5: Λ-type Raman transition between two ground states. In the case of $^{137}\text{Ba}^+$ this is between the $6S_{1/2}$ and either the $5D_{3/2}$ or $5D_{5/2}$ states. The process takes place through a virtual state that is detuned from the excited by a detuning $\Delta$.

Consider a three level system consisting of two ground states, $|g_1\rangle$, $|g_2\rangle$, and an excited state $|e\rangle$. A laser beam can be used to couple states $|g_1\rangle$ and $|e\rangle$ together, with a Rabi rate defined as $\Omega_1$. A second laser beam can be used to couple states $|g_2\rangle$ and $|e\rangle$ together, with a Rabi rate defined as $\Omega_2$. The detuning from the excited state can be defined as $\Delta = \omega_1 - \omega_{g_1,e}$ and the detuning of the two lasers from Raman resonance as $\delta = \omega_1 - \omega_2 - \omega_0$. In the regime where $\Delta \gg (\Omega_1, \Omega_2, \Gamma_e, \delta)$, the probability of the ion being in the excited state will be very small. Therefore, the excited state can be adiabatically eliminated [56, 57] and the three level system can be simplified to an effective two-level system, where the Rabi rate between the ground states is described by the effective Rabi rate, alternatively known as the Raman Rabi rate,

$$\Omega_R = \frac{\Omega_1\Omega_2}{2\Delta}. \tag{2.40}$$
Here the individual Rabi rates $\Omega_1$ and $\Omega_2$ can be expressed as

$$\Omega = \frac{2d_{e,g}}{\hbar w_0} \sqrt{\frac{P}{\pi \epsilon_0}}, \quad (2.41)$$

where $w_0$ is the waist of the beam, $P$ is the power of the beam and $d_{e,g}$ is the dipole matrix element between the states.

Due to off-resonant coupling to the excited state, $|e\rangle$, Stark shifts must be accounted for in ground states, $|g_1\rangle$, and $|g_2\rangle$, by

$$\Delta E_{gi} = \hbar \frac{\Omega_i^2}{4\Delta}, \quad (2.42)$$

where $i = 1, 2$. These Stark shifts lead to the detuning $\delta$ being defined as the effective detuning

$$\delta_e = \delta + \left( \frac{\Omega_1^2 - \Omega_2^2}{4\Delta} \right), \quad (2.43)$$

which is the sum of the original detuning plus the differential Stark shift. The probability of the ion being in $|g_1\rangle$ or in $|g_2\rangle$ state is given by the two-level results in Eqs. (2.34) and (2.35), where $|g_1\rangle = |g\rangle$ and $|g_2\rangle = |e\rangle$ in these equations. Furthermore the Rabi rate $\Omega$ and detuning $\delta$ in these equations must be replaced by the Raman Rabi rate $\Omega_R$ and the effective detuning $\delta_e$ respectively.

### 2.3.4 Decoherence mechanisms

In our treatment of coherent rf and Raman transitions we have neglected all decoherence terms. The presence of spontaneous emission from the exited state, ion motion, electromagnetic radiation phase jitter and magnetic field noise all contribute to reducing the efficiency of the coherent transitions.

For rf transitions, spontaneous emission can be neglected because the 5D manifolds are metastable. For the Raman transitions, spontaneous emission can not be fundamentally eliminated because even though we modeled the system as a two-level atom, there will still always be a small amount of population in the excited state. This results in decoherence through decay out of the excited state via the emission of a photon. The mean number of photons $N$ scattered from the excited is given as [41]

$$N = Rt = \frac{\pi R}{\Omega_R}, \quad (2.44)$$
where \( t \) is the interaction time equivalent to a π pulse and \( R \) the scattering rate defined as

\[
R = \rho_e \Gamma, \tag{2.45}
\]

where \( \Gamma \) is the natural decay rate of the \(|e\rangle\) state and \( \rho_e \) is probability of being in the excited state, which in turn is given by [41]

\[
\rho_e \approx \frac{\Omega_1^2}{4\Delta^2} + \frac{\Omega_2^2}{4\Delta^2}. \tag{2.46}
\]

Eq. (2.46) is minimized if the individual Rabi rates are equal, \( \Omega_1 \approx \Omega_2 \), leading to

\[
N \approx \frac{\pi \Gamma}{2\Delta}, \tag{2.47}
\]

which is independent of the Raman Rabi rate. In our experiment \( N < 0.001 \) so the effect of spontaneous emission can be neglected.

Magnetic field noise causes fluctuations in the atom-field detuning via the Zeeman interaction. When \( \Omega \) is much larger than the fluctuations, \( \Omega \gg \delta\nu \), then the fluctuations will not have much effect on the transfer efficiency. For the rf transitions this will be the limiting factor on how slowly we can drive the transitions and hence, how accurately the hyperfine intervals can be measured. For Raman transitions magnetic field noise will have a negligible effect on the efficiency of the transitions because \( \Omega_R \gg \delta\nu \).

This leaves ion motion and phase jitter to consider. More generally, the interaction of a two-level atom with a laser field is described by

\[
\Omega = |\Omega| e^{i(\vec{k} \cdot \vec{x} - \omega_l t + \phi(t))}, \tag{2.48}
\]

where \(|\Omega|\) is the previously determined Rabi rate, \( \vec{k} \cdot \vec{x} \) an additional term due to coupling between the field and the ion’s motion and a phase term \( \phi(t) \) associated with the phase fluctuations of the field. In the specific case of the rf transitions, phase noise is not an issue when using modern oscillators. In the case of the Raman transitions, the phase term is determined by the relative phase between the two lasers. For Raman transitions between hyperfine states of the same manifold the two driving beams originate from a single laser, so the two arms are phase coherent. For transitions between the \( 6S_{1/2} \) and \( 5D \) levels, this is not the case and phase jitter arises from inherent incoherence between the phases of the two lasers involved. A detailed description of this effect is given in [57]. The relative phase noise between the two Raman beam arms can be reduced by locking both lasers to the same optical cavity, as discussed in Section 3.5.4.
Finally we consider ion motion. Here we only give a brief summary of the effects, a more detailed treatment can be found in [58]. If we consider only the vibrational coupling along the $x$-direction, the term $\vec{k} \cdot \vec{x}$ can be expressed in terms of the ladder operators, $\hat{a}$ and $\hat{a}^\dagger$, for the corresponding quantized states

$$\vec{k} \cdot \vec{x} = \eta (\hat{a} - \hat{a}^\dagger),$$

(2.49)

where $\eta$ is the Lamb-Dicke (LD) parameter defined as

$$\eta = k_x \sqrt{\frac{\hbar}{2m\omega_{\text{trap}}}}.$$  

(2.50)

For rf transitions the motional coupling term $\vec{k} \cdot \vec{x}$ is negligible because the amplitude of the ion motion is much smaller than the wavelength of the driving field. For Raman transitions $\vec{k} \rightarrow \Delta \vec{k}$ and the motional coupling is only negligible if $\Delta k_x$ is small compared to the ion’s motional amplitude. For the case of Raman transitions between hyperfine states of the same manifold, $\vec{k}_1 \approx \vec{k}_2$, and choosing a co-propagating laser beam alignment the LD parameter can be made small. In this regime Raman transitions are insensitive to ion motion. Raman transitions in this thesis are used for shelving to the 5D states, which means that $\vec{k}_1 \neq \vec{k}_2$. Therefore, independent of beam alignment the transitions will always be sensitive to motion.
Chapter 3

Equipment

This chapter covers the equipment required for performing a wide range of experiments on singly ionized barium. Our experiments are performed on a single barium ion, which is trapped and isolated from the environment. We manipulate the state of the ion by addressing it with coherent light fields generated from a variety of lasers. The state of the ion is determined by imaging its fluorescence after a controlled interaction.

In Section 3.1 we describe the vacuum system used for isolating the ion from the environment. In Section 3.2 the ion trap is described and the oven used to create a beam of atomic barium, which is ionized inside the trap. In Section 3.3 we detail the diode lasers used to manipulate the ion and how they are built. In Section 3.4 we describe how optical frequency doubling is used to obtain wavelengths that are currently unavailable with standard diode lasers. In Section 3.5 we describe how the different lasers are stabilized to reference cavities. The method we use to photo-ionize barium is discussed in Section 3.6. The rf antenna used to drive the hyperfine transition used for measuring the hyperfine intervals is discussed in Section 3.7. In the final Section 3.8 the imaging system is explained.

3.1 Vacuum system

Ultra high vacuum is necessary in atomic physics experiments to isolate the ion from the environment. Isolation from the environment limits collisions with background gas particles, which, through chemical reactions, are the main cause of ion loss. The vacuum chamber is a custom-designed octagonal piece manufactured by Kimble Physics
shown in Fig. 3.1. The custom design allows for a more compact setup compared to using a standard sized octagon, providing a shorter distance between ion and imaging lens. The chamber is connected to a 20 liter per second ion pump and a titanium sublimation pump. After baking the chamber out for three days at 150°C, firing the titanium sublimation pump and turning on the ion pump, a pressure of $10^{-11}$ Torr is readily achievable. At this pressure ion lifetimes of many hours or even days are possible, which is more than sufficient for the experiments performed in this thesis.

![Figure 3.1: Schematic of our octagonal vacuum chamber with laser beams and ion trap shown.](image)

### 3.2 Ion trap

There are three pieces of equipment required for trapping an ion. An ion trap, an atomic source and an ionization source. The typical procedure for loading an ion is to lock the ionization lasers, heat up the oven, ionize the barium from the atomic beam in the center of the trap, cool the trapped ions with a laser and detect ion fluorescence on a camera. For our experiments, the ionization is done by photo-ionization and is
discussed separately in Section 3.6. The ion trap and oven (atomic beam source) are discussed below.

3.2.1 Linear Paul trap

![Figure 3.2: Test linear Paul trap. This ion trap was initially used to first trap ions and benchmark our equipment.](image)

![Figure 3.3: Experimental linear Paul trap with cavity. This ion trap is used for all the experiments performed in this research.](image)

An ion trap is a device that is used to localize a charged particle. The trap works on the principle of using electric potentials to confine the particle via Coulomb’s law. According to Earnshaws theorem [59], electrostatic potentials alone can not be used to achieve three dimensional trapping of a charged particle. This problem was overcome by Wolfgang Paul in the early 1950’s [60, 61], where he used an additional oscillating electric field to provide complete confinement. There are numerous different ways in which to implement an ion trap [62, 63, 64, 65, 66, 67] but we have chosen to use a linear Paul trap, shown in Figs. 3.2 and 3.3, for its simple construction. The theory of how an ion trap operates is documented in [58]. A schematic drawing of all of the electrodes required to operate a linear Paul trap is shown in Fig. 3.4.

In brief, an rf potential is applied to two diagonally opposed electrodes to provide confinement in two dimensions. A pair of endcaps are held at a fixed direct current (dc) voltage to provide confinement in the third dimension. A second set of diagonally opposed electrodes known as bias electrodes are held at a dc potential to provide splitting of the radial trapping frequency modes.

Over the course of this research two linear Paul traps have been built. The first linear Paul trap was used to initially trap an ion and benchmark our equipment and will be referred to as the “test trap”. The second trap was used for all the experiments undertaken in this research and will be referred to as the “experiment trap”. They are in essence of the same design. The experiment trap is approximately a factor of two
**Figure 3.4:** Schematic of a linear Paul trap showing electrical connections. An rf potential is applied to two diagonally opposed electrodes to provide confinement in two dimensions. The other set of diagonally opposed electrodes are held at a dc potential to provide splitting of the radial trapping frequency modes. A pair of endcaps are held at a fixed dc voltage to provide confinement in the axial direction.

smaller than the test trap, which allows for the implementation of an optical cavity around the ion trap. The details of the traps are tabulated in Table 3.1 and pictures of both the traps are shown in Figs. 3.2 and 3.3.

**Table 3.1:** Ion trap parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Experiment trap</th>
<th>Test trap</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>rf drive frequency</td>
<td>$2\pi \times 5.3$</td>
<td>$2\pi \times 3.4$</td>
<td>MHz</td>
</tr>
<tr>
<td>rf amplitude (peak)</td>
<td>125</td>
<td>500</td>
<td>V</td>
</tr>
<tr>
<td>End cap voltage</td>
<td>30</td>
<td>5</td>
<td>V</td>
</tr>
<tr>
<td>End cap separation</td>
<td>2.4</td>
<td>5</td>
<td>mm</td>
</tr>
<tr>
<td>Rod separation</td>
<td>2</td>
<td>3.6</td>
<td>mm</td>
</tr>
<tr>
<td>Rod diameter</td>
<td>0.45</td>
<td>1.2</td>
<td>mm</td>
</tr>
</tbody>
</table>

The experiment trap consists of four stainless steel rods of diameter 0.45 mm whose centers are arranged on the vertices of a 2 mm square, giving a minimum ion electrode distance of 1.4 mm. The trap has an optical cavity aligned perpendicular to the axial direction of this trap. The optical cavity was the focus of my colleague’s research which can be found in [58, 68, 69]. The optical cavity is not required for any part of this research and is lowered so that its optical axis is not aligned with the ion. Hence, the optical cavity has no effect on the experiments in this thesis. A $2\pi \times 5.3$ MHz rf potential with an amplitude of 125 V is applied via a step-up transformer (with
a step up factor of 25) to two diagonally opposing electrodes. The rf source used
to drive the trap comes from a frequency synthesizer that passes through a variable
attenuator for convenient amplitude control and is then amplified up to 2 W. A dc
voltage of around 0.1 V is applied to the bias electrodes, which ensures a splitting of the
transverse trapping frequencies and rotates the principle axes of the trap with respect
to the propagation direction of the cooling lasers. Axial confinement is provided by
two rod endcap electrodes separated by 2.4 mm and held at 33 V. Further adjustment
is provided by two aluminum shields (see Fig. 3.3) that are used to shield the ion
from the cavity mirrors. They can be set independently, at a fixed potential in the
range of -10 V to 10 V. The ability to independently set the all the dc confinement
potentials allows us to compensate for the effects of stray static electric fields. The
ability to compensate the effect of stray electric fields allows us to minimize micromotion
[70] down to the 10 nm level [68]. Using this configuration, the measured trapping
frequencies are \((\omega_x, \omega_y, \omega_z)/2\pi \approx (1.1, 1.0, 0.4)\) MHz.

3.2.2 Barium oven

A resistively heated oven provides a source of neutral barium which then is ionized
inside the trap. Due to the compact nature of the chamber and the ion trap, the oven
design has two main constraints. One it must be small in size to fit within the chamber
and two it must use relatively low power to heat it up, so the radiation it gives off does
not heat up sensitive parts of the ion trap. The design of the oven depends on how
hot it has to get. From experimental work performed by another ion trapping group
working with barium [71] we know an oven temperature of around 300 °C is required
for a suitable atomic beam flux for ionization. Increasing the oven temperature much
above this level could result in excessive barium coating of the ion trap, which could
lead to shorts.

The oven is made from a stainless steel tube with a 2.5 mm inner diameter and a
25 μm wall thickness. The tube is cut to a 10 mm length and has one end folded up
and spot welded shut. Small chunks of Barium are placed in the tube under an inert
environment to reduce oxidation. The tube end is then folded up and spot welded on
the other end. Two 25 μm diameter tantalum wires are spot welded to the ends of the
tube as shown in Fig. 3.5. These wires heat up to glow red hot indicating they are around
700 °C in temperature when 3 A is passed through them. The resistive heating of the
wires and stainless steel tube act as a heat source to heat up the pieces of barium to the required temperature. A small hole that is placed in the middle of the tube directs the atomic barium beam towards the trap.

3.3 Lasers

A coherent light field is necessary for addressing the various transitions in barium which are shown in Fig. 3.6. These coherent light fields originate from lasers. In this thesis we only work with diode lasers [72] as these are cheaper and more simpler to operate than other types of lasers. The only major issue with diode lasers is that they do not cover the full frequency spectrum, but through the application of frequency doubling

**Figure 3.5:** Foil parcel oven with pin hole in center and 0.25 mm diameter tantalum wires spot welded to the ends to provide heat and electrical contact.

**Figure 3.6:** Energy level diagram for Ba$^+$, showing all the transition that are addressed in this work and the laser wavelengths required to address them.
all the required wavelengths can be obtained. At the start of this research diodes with wavelengths between 410 nm to 645 nm were not available, hence the requirement for Second Harmonic Generation (SHG) to produce light at 455 nm, 493 nm and 615 nm, where as the range 645 nm to 2 µm is accessible with commercial diodes. The required lasers, their wavelengths and the source they are derived from is given in Table 3.2. The lasers discussed in this section have been grouped in terms of the technology used to implement them. We also dedicate an entire separate section to the equipment used to implement SHG, which is discussed in Section 3.4. The laser discussion is split up into four smaller sections. We start with a brief overview of the uses of each laser and the technology used to construct them in Section 3.3.1. In Section 3.3.2 we discuss lasers originating from laser diodes. Section 3.3.3 covers a laser housing design that allows the laser diode to be cooled to well below 0°C, which is useful for temperature tuning the gain profile of the laser diode. In Section 3.3.4 we describe a laser that originates from a gain chip [73].

Table 3.2: Laser wavelengths required for working with $^{137}$Ba$^+$. All wavelength are measured using the lab’s wavemeter, which has been calibrated to an atomic transition in Rb.

<table>
<thead>
<tr>
<th>Role</th>
<th>Transition addressed</th>
<th>Wavelength vacuum ( nm)</th>
<th>Laser type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ionization</td>
<td>$6s^2S_0 \leftrightarrow 6s6p^3P_1$</td>
<td>791.351</td>
<td>Diode</td>
</tr>
<tr>
<td>Ionization</td>
<td>$6s6p^3P_1 \leftrightarrow 6p^2^3P_1$</td>
<td>450.719</td>
<td>Diode</td>
</tr>
<tr>
<td>Doppler cooling</td>
<td>$6S_{1/2} \leftrightarrow 6P_{1/2}$</td>
<td>493.545</td>
<td>SHG diode</td>
</tr>
<tr>
<td>Repumping</td>
<td>$5D_{3/2} \leftrightarrow 6P_{1/2}$</td>
<td>649.869</td>
<td>Diode</td>
</tr>
<tr>
<td>Repumping</td>
<td>$5D_{5/2} \leftrightarrow 6P_{3/2}$</td>
<td>614.341</td>
<td>SHG gain chip</td>
</tr>
<tr>
<td>Shelving</td>
<td>$6S_{1/2} \leftrightarrow 6P_{3/2}$</td>
<td>455.403</td>
<td>SHG diode</td>
</tr>
</tbody>
</table>

3.3.1 Overview of laser system

Operating a barium ion trap experiment requires a lot of lasers, in this section we give a brief overview of all the lasers used throughout the experiments in this thesis. As barium has two low lying metastable 5D manifolds the majority of the lasers in this thesis operate in pairs to address certain states in the ion and as such will be discussed in pairs. The first pair of lasers are used to address the 493 nm and 650 nm transitions
which are concerned with Doppler cooling and fluorescing the ion. The 493 nm radiation is derived from a commercial frequency doubled External Cavity Diode Laser (ECDL) and 650 nm from a home built ECDL, which is a standardized design used throughout our lab and is described in Section 3.3.2. These two lasers are covered in further detail in [58]. The next pair of lasers again have wavelengths at 493 nm and 650 nm but are used for Raman transitions to the 5D_{3/2} state. These lasers can not originate from the same sources as the Doppler cooling lasers due to the large detuning at which the Raman lasers have to operate. The 493 nm Raman beam comes from a second commercial frequency doubled ECDL and the 650 nm Raman beam from an injection locked high power 650 nm laser, which is of the design discussed in Section 3.3.3. The 650 nm high power laser diode, is injection locked with a narrow linewidth cavity stabilized laser described in [58]. These two lasers systems are all the laser used for addressing the 6S_{1/2}, 6P_{1/2} and 5D_{3/2} states.

For coherent transitions to the 5D_{5/2} state we use a pair of Raman lasers at 455 nm and 615 nm. The ion is returned from the 5D_{5/2} state back into the Doppler cooling cycle by using a repump laser at 615 nm. The 455 nm light originates from a 911 nm ECDL, where the laser design is given in Section 3.3.2. The 911 nm laser undergoes frequency doubling to 455 nm via intra-cavity doubling which is discussed in Section 3.4.1. The 614 nm Raman and repump lasers are derived from a 1230 nm gain chip covered in Section 3.3.4, which is then frequency doubled by a waveguide doubling crystal detailed in Section 3.4.2. We have a further two ECDLs lasers used at 791 nm and 450 nm for the ionization of barium, these also come under the same description of ECDLs given in Section 3.3.2.

3.3.2 Diode laser design

Our standard home build ECDL was designed by Kyle J. Arnold and is covered in further detail in his thesis [74]. The design is a modification of the T Hansch design [75]. The main modification is that the diffraction grating is mounted on a Lead Zirconium Titanate crystal (PZT) that is glued to a two axis mirror mount instead of being mounted on a flexure mount. This has the advantage of being easier to tune and the freedom to use a larger range of grating angles than just 45° ± 5° of the flexure mount. This is useful because of the limited range of diffraction grating line spacings which are commercially available. Our standard diode laser design is shown in Fig. 3.7.
Figure 3.7: Standard ECDL housing showing temperature stabilized laser diode housing, grating mounted on a mirror mount for easy tunability and output mirror for laser to exit housing through Brewster angled window. The laser housing can be hermetically sealed.

The housing is machined from a single piece of aluminum and black anodized. The laser diode sits inside a collimation tube supplied by Thorlabs. The collimation tube is placed in an aluminum housing that sits on top of a Thermo-electric Cooler (TEC) to stabilize its temperature. The laser diode housing is fixed to the main laser housing using plastic screws to limit the flow of heat in or out of the laser diode housing. The mirror mount holding the diffraction grating and PZT is glued in position after rough alignment. The zeroth order light that comes off the diffraction grating is directed out of the housing using a mirror that is just glued in position. Finally the laser beam leaves the housing through a Brewster angle window. The window means once the lid is in place the housing is completely sealed, as all joints on the housing use rubber seals. The laser diodes are driven and locked using home built electronics designed by Professor M.D. Barrett.

3.3.3 High power 650 nm laser

To achieve efficient Raman transitions between the $6S_{1/2}$ and the $5D_{3/2}$ state the off-resonant excitation must be low. For low off-resonant excitation the detuning of the lasers with respect to the $6P_{1/2}$ state must be large. According to Eq. (2.40) increasing the detuning will decrease the Raman rate. To keep the Raman rate high the intensity of the 493/650 nm beams must be high. All the commercially available higher power
650 nm laser diodes have a room temperature wavelength of around 659 nm. This is a problem as the resonance wavelength of barium is around 650 nm. To get the laser diode to this wavelength it must be cooled down to move the center wavelength of the gain medium to 650 nm. Moving the gain material 9 nm requires a substantial amount of cooling, which creates problems with ice/condensation build up and having to remove large amounts of heat from the laser diode housing.

![Cut through drawing of high power 650 nm laser housing showing the two stage TEC cooling and heat sink used to remove heat from housing. The heat sink can be replaced with a water cooled heat sink for more efficient heat removal. The housing is made of perspex to limit heat transfer from environment and to lower thermal contact between heat sink and housing.](image)

**Figure 3.8:** Cut through drawing of high power 650 nm laser housing showing the two stage TEC cooling and heat sink used to remove heat from housing. The heat sink can be replaced with a water cooled heat sink for more efficient heat removal. The housing is made of perspex to limit heat transfer from environment and to lower thermal contact between heat sink and housing.

We use a Perspex based design shown in Fig. 3.8 to limit the heat conduction paths to the laser diode housing. A two stage TEC system is connected to a water cooled block to bring the laser diode housing down to a temperature of −35 °C. The upper TEC is used for temperature stabilizing the laser diode housing. Two further TECs are run at a fixed current to remove as much heat as possible from a base plate, which the upper TEC is mounted on. The heat produced by the two lower TECs is removed by water cooling as it is the most efficient way to remove the large amount of heat generated. The perspex housing can either be maintained under vacuum or in a dry

\(^{1}\text{HL6545MG}\)
atmosphere of argon. Either of which ensures that ice does not build up around the laser diode.

### 3.3.4 1228 nm gain chip

A gain chip is used to produce 1228 nm light that can be frequency doubled to produce 614 nm for the Raman and repump laser systems. A gain chip differs from a laser diode as its front and/or back facet is anti-reflective (AR) coated. This means it does not have a lasing cavity and is therefore just a laser gain medium. If current is passed through a gain chip it will not lase, but instead produce incoherent broad band light know as Amplified Stimulated Emission (ASE). To make a gain chip lase, a cavity must be placed around it. In our case, the gain chip has an AR-coated front facet and a 95% reflective coating on the back facet. A diffraction grating is placed in front and forms a low finesse cavity with the rear facet of the gain chip. This low finesse cavity makes the gain chip lase.

In our experiment we use a gain chip\(^1\) with a gain profile that is centered at 1240 nm and is over 100 nm wide. It is attached to a mount that is temperature stabilized via a TEC. Due to the way it is mounted on its heat sink it is difficult to align an aspherical lens to collimate its output. For alignment the asphere is mounted in a square lens mount, which is the mounted to a 5-axis translation stage. The translation stage allows precise positioning of the lens to collimate the output ASE. Collimating the ASE does not ensure collimation of the lasing mode, it is just done for initial alignment to help with diffraction grating placement. Next a 1200 lines/mm holographic diffraction grating is glued to a PZT, which is attached to a two axis mirror mount. The grating is then aligned so that the first order is retro-reflected back into the gain chip. This causes the gain chip to lase on a certain mode, which changes the spatial output mode of the light and the collimation lens must then be re-aligned. The grating is adjusted until the gain chip lases at the required operating wavelength. Then the lens is re-adjusted to ensure the gain chip produces its peak power and that the spatial mode out of the gain chip has a profile as Gaussian as possible to help with fibre coupling. Finally the lens mount is glued into place using Norland 63 ultra-violet (UV) curing epoxy.

\(^{1}\)SAF1145H manufactured by Covega
3.4 Second Harmonic Generation

In this section we describe the experimental aspect of SHG, for a theoretical discussion of SHG see appendix B. Three laser systems rely on SHG to obtain the target wavelength, 455 nm, 493 nm and 615 nm. The 455 nm and 615 nm systems are home built, whereas the 493 nm is a commercial system. We employ two different methods of frequency doubling, the first uses intra-cavity doubling to produce the 455 nm light and the second uses a newer technique of single pass waveguide doubling to produce the 615 nm. The intra-cavity doubling is discussed in Section 3.4.1 and the single pass waveguide doubling is described in Section 3.4.2.

3.4.1 Intra-cavity doubling of 910 nm to 455 nm

From Eq. (B.7) it can be seen that very high laser power is required to get high conversion efficiency in single-pass configurations. An enhancement cavity is required for optimal frequency conversion of a low power (< 500 mW) laser beam. For intra-cavity doubling we must make three consideration to maximize the conversion efficiency: nonlinear crystal type, crystal dimensions and cavity loss matching.

The first decision to be made is what is the best crystal to use, as this decision dictates the rest of the doubling cavity requirements. The best crystal to use would be one that provides a large single pass conversion factor. The single pass conversion factor depends on many parameters that are discussed in appendix B.2. There are many different crystals that can be used to double 911 nm to 455 nm, the most popular of these are: Beta Barium Borate (BBO), Lithium Triborate (LBO), KNbO$_3$ and Periodically Poled Potassium Titanyl Phosphate (PPKTP). These crystals have different phase matching parameters: BBO and LBO use Critical Phase Matching (CPM), whereas KNbO$_3$ and PPKTP use Non-critical Phase Matching (NCPM). It is advantageous to use a NCPM crystal as there is no walk off factor, which greatly enhances single pass doubling efficiency. It was not possible to get an NCPM crystal as KNbO$_3$ is very difficult to obtain and PPKTP is expensive. So instead we opted to use BBO as it is readily available and cheap.

The dimensions of the crystal affects the overall doubling efficiency, so requires careful consideration. The most important dimension to consider is the length of the crystal, as in the ideal case the conversion efficiency scales with the interaction length.
As BBO is not a perfect crystal, it has some absorption losses. Therefore there is an optimum crystal length for input power. The crystal length determines the optimum waist inside the crystal. The optimal crystal length for BBO can be found from Eq. (B.7). When we assume these typical parameters $\xi = 1.39$, $\alpha_1 = 0.3\% \text{/ cm}$, $\alpha_2 = 4\% \text{/ cm}$, $P_{\text{in}} = 150 \text{ mW}$, $\eta_{\text{coup}} = 80\%$, $v = 0.75\%$ and $t = 1\%$, as shown in Fig. 3.9 a crystal length of $L = 10 \text{ mm}$ is optimal. For this crystal length, the optimum waist defined in Eq. (B.9) is determined to be $w_0 = 25 \mu\text{m}$.

**Figure 3.9:** Plot showing how length of BBO crystal affects second harmonic power for certain initial parameters.

**Figure 3.10:** SHG conversion efficiency for varying input power. The different plots show how different input couplers affect SHG conversion efficiency.

The final criteria to determine is the optimum reflectivity of the input coupler of the bow-tie doubling cavity, which is dependent on the total losses $\mathcal{L}$ in the doubling cavity. It is hard to accurately estimate the losses in a cavity before it is built, so some trial and error is required. The input coupler choice can be narrowed down by knowing the maximum input power $P_{\text{in}}$ and the crystals single pass conversion factor. The input power has to be high when using BBO as its single pass conversion factor is low due to its large walk off angle. This also means that the fundamental beam depletion is not really a problem for the regime we are working in. A plot of input couplers versus conversion efficiency is shown in Fig. 3.10. For the regime we are working in we would need an input power above 500 mW before an input coupler with a lower than 99% reflectivity is required. If the absorption and scattering losses in the cavity were larger
than 1% then this would become a 98% input coupler. It is found from trial and error that an input coupler with a transmission of \( t = 1\% \) is the best choice for our setup.

The final part of setting up a doubling cavity is to choose the arm lengths of the doubling cavity, the folding angle and the radius of curvature of the focusing mirror. The design aspect is discussed in appendix B.3. As BBO has a large walk off angle the output mode of 455 nm beam is astigmatic and highly elliptical, even though the fundamental mode is round at the center of the crystal. The poor spatial mode is corrected with the use of cylindrical lenses on the output. The doubling cavity is locked using the Pound-Drever-Hall (PDH) technique detailed in Section 3.5.4. When the doubling cavity was initially built it provided 15 mW of 455 nm, but over time this has decayed down to a maximum of 6 mW. The most likely cause for this is water absorption by BBO as it is highly hydrophobic. Remedies to this would be to place the doubling cavity in a hermetically sealed housing, heat the crystal to slow moisture absorption or replace the crystal with a different type. After the end of the work described in this thesis we replaced the BBO crystal with a PPKTP crystal, which yields 60 mW of output power.

### 3.4.2 Single pass waveguide doubling of 1228 nm to 614 nm

Single pass waveguide doubling offers advantages over intra-cavity doubling because it does not use a cavity. The lack of a cavity means no locking electronics are required to stabilize the length of the cavity. This also means single pass waveguide doubling has the advantage of being always on. Disadvantages to waveguide doubling are that it suffers from lower conversion with respect to intra-cavity doubling and the output mode is far from a perfect Gaussian mode. A single pass waveguide doubling crystal is used for the generation of the 614 nm laser light, both Raman and repump. The theory of single pass waveguide doubling is covered in appendix B.4.

Our setup for the 614 nm repump light is similar to the setup detailed in [76]. In brief, the 1228 nm beam is focused down using an aspherical lens with a 3 mm focal length. The waveguide doubling crystal is mounted on a 6 axis fibre launch stage to ensure good alignment between the input beam and the waveguide. About 15% of the 1228 nm light is coupled into the guide, which is due to mode mismatch between the guide and laser. The poor mode matching is the limiting factor in the overall conversion efficiency, especially because the output power scales as the input power...
squared. From the 1228 nm laser power of 30 mW the setup only provides 100 µW of light for the experiment. We did not optimize the mode matching further as this 100 µW is much more power than is required for repumping the state.

For the 614 nm Raman light a fibre pigtailed waveguide doubling crystal is used. The fibre pigtail does not offer any improvements to conversion efficiency as mode matching between the fibre and the guide is still poor. It does however, offer a nice Gaussian output mode that is easier to work with. From the 1228 nm laser power of 45 mW the setup provides 300 µW of power, about 50 µW of power is sent to the reference cavity, which is used to lock the fundamental laser. The rest of the 614 nm light passes through an Acousto-optical modulator (AOM) and is fibre coupled to provide 150 µW of light to the experiment chamber.

3.5 Reference cavities

In order to address and manipulate the ion with laser beams, one faces two challenges. The first challenge is the laser frequency has to be centered at the frequency of the atomic transition and the second challenge is the laser has to be narrow enough to address the transition. For the Doppler cooling beams the maximum drift range of the laser must be on the order of the atomic transition linewidths and the laser linewidth must be smaller than the atomic linewidths. Typical atomic transition linewidths in Ba⁺ are less than 20 MHz wide, so making a laser that satisfies the above criteria is relatively simple to achieve. For a Raman transition the same criteria applies except the drift of the two lasers should be less than 10% of the Raman rate \( \Omega_R \) and the relative linewidths of the two lasers should be less than 1% of the Raman rate \( \Omega_R \). Fulfilling these criteria for a pair of Raman beams that are generated from the same laser is relatively simple, but for Raman beams from two separate laser this is technically challenging. The difficulty comes from the fact that two independent lasers will be inherently phase incoherent and will drift at different rates.

To satisfy the above requirements one needs a narrow stable frequency reference with which to lock the lasers, of which there are a few different options available. One could use the atomic transition itself [77], but this would require building a second ion trap just for locking the lasers. Another option is to use a hollow cathode lamp, which has previously been used as a reference source for a 650 nm laser [78], but due to how
the device operates the reference features are over 1 GHz wide. Atomic vapor cells with similar atomic transitions to Ba$^+$ have been used as locking references before for example, molecular tellurium has lines near 493 nm [79] and a molecular iodine has lines near 650 nm [80]. Molecular references suffer from having broad transitions so locking the laser tightly to these features is not possible. An alternative method is to use optical cavities, which allow for narrow reference features via the adjustment of the length of the cavity and the quality of the mirror coatings. Optical cavities have the advantage of broadband mirror coatings, which allows for all the lasers used in this research to be locked to the same cavities. A disadvantage of optical cavities is the length of the cavity drifts around with changes in the ambient temperature and a laser locked to the cavity will track this drift. The cavity drift is a problem for Raman beams that originate from different lasers because the frequency drift of each beam will be different due to the large difference in their wavelengths. Put a different way, one beam will be drifting with respect to the other at a rate proportional to the ratio between their wavelengths. Therefore any change in the cavity length will mean the Raman resonance condition will not be satisfied. A further disadvantage of using a cavity is that they have discrete locking features which are separated by the free spectral range defined as

$$\text{FSR} = \frac{c}{2l}$$

where $c$ is the speed of light and $l$ is the length of the cavity. This issue can be resolved by using an offset locking technique that uses a broadband Electro-optical modulator (EOM), which spans at least one free spectral range.

The most pressing issue with stabilizing the length of the cavity is keeping the length of the cavity spacer fixed. To do this we employ a three pronged approach of using a low temperature coefficient cavity spacer material, active temperature stabilization techniques and provide thermal isolation from the lab environment. In this section we discuss two different reference cavity designs. In Section 3.5.1 we cover a cavity housing design that uses expanding foam to provide thermal isolation. In Section 3.5.2 we discuss a cavity housing design that uses multiple heat shields to provide thermal isolation. Both the cavity housing designs share similar spacer material, both use active temperature stabilization and both are kept under high vacuum. The high vacuum provides two functions. One it ensures that air currents can not conduct heat from/to the cavity spacer and two it stops the optical path length fluctuating due to changes
in air pressure. We also made improvements to the heat shield cavity design, which is covered in Section 3.5.2.1. The two designs and the improved second design are compared in Section 3.5.3. In Section 3.5.4 we explain the scheme we used to lock the lasers to the reference cavities.

### 3.5.1 Foam insulated reference cavity

![Diagram of foam insulated cavity](image)

**Figure 3.11:** Schematic of foam insulated cavity showing Zerodur cavity space rubber mounted to a 2.75” tee-piece. The tee-piece is temperature controlled and mounted in a Perspex box using Teflon mounts. The box is filled with expanding foam to limit the effect of environmental temperature fluctuations.

The foam insulated reference cavity design take advantage of the low thermal conductivity of expanding foam to limit the effects ambient temperature changes have on the cavity spacer. The design of the foam insulated reference cavity is shown in Fig. 3.11. Two ATFilms cavity mirrors with a broadband coating from 650 nm to 1100 nm, with a radius of curvature of 10 cm and a wavelength dependent finesse of around 2000 are fixed to a low drift Zerodur space [81]. The mirrors are held in place with a tiny amount of Norland 81 UV curing epoxy. The Zerodur spacer is approxi-
mately 10 cm long and is of grade type 0, which means it has a temperature coefficient of $0.0 \pm 0.02 \times 10^{-6} \text{K}^{-1}$. The cavity is placed inside a 2.75 inch tee-piece depicted in Fig. 3.12 and the cavity is held in place with three small pieces of Viton rubber to ensure minimal thermal conduction to the vacuum chamber. Optical access to the vacuum chamber is provided by quartz windows. The chamber is pumped down to $1 \times 10^{-7} \text{Torr}$ and baked out at $110 \degree \text{C}$ for two days. A copper pinch off tube is used to seal the vacuum chamber.

![Figure 3.12: Image showing vacuum tee-piece with heater tapes and thermistor mounted in perspex box via Teflon mounts, before insulating foam is added.](image)

![Figure 3.13: Linewidth of ATFilms mirrors at 780 nm measured by adding sidebands to laser to be used as a frequency marker in order to calibrate time data saved by the scope. The sidebands are generated at 52 MHz. The cavity mirrors reflectivity lead to a linewidth of 1.4 MHz at 780 nm.](image)

The vacuum chamber has two Omega heater tapes stuck to the side of it to provide a heat source for temperature stabilization. Three thermistors are fixed using Torr Seal to the vacuum housing for redundancy purposes. The vacuum chamber is placed in two Teflon mounts and placed inside a clear Perspex box. The box is filled with expanding foam, which is impeded from covering the windows due to the Teflon spacers. The box is sealed and stabilized two degrees above ambient temperature to a precision of 10 mK. The overall drift of the cavity can either be monitored with respect to
a laser referenced to an atomic transition in $^{87}$Rb or by a laser referenced to Center for Quantum Technologies (CQTs) shared optical frequency comb [82]. An image of the cavity under construction and the linewidth of the cavity at 780 nm are shown in Fig. 3.13. The linewidth of the cavity is measured by adding sidebands to the laser which act as frequency markers in order to calibrate the time data saved by the scope. Drift data was taken at various times throughout a month a typical dataset over the period of two days is shown in Fig. 3.14. The average drift rate for this cavity is found to be 25 Hz/sec. This cavity is used as a reference for the 493 nm and 650 nm Raman lasers.

![Graph showing drift of reference cavity](image)

*Figure 3.14:* Plot showing drift of reference cavity one. The drift rate extracted from this plot represents a drift rate of 11 Hz/sec. The spread between points is due to the underlying frequency stability of the probing laser. An average over several datasets like the one shown in this figure yields an average drift rate of 25 Hz/sec.

### 3.5.2 Heat shield reference cavity

The heat shield reference cavity uses a number of radiation shields to limit the effects ambient temperature changes have on the cavity spacer. A schematic diagram of the design of the heat shield reference cavity is shown in Fig. 3.15. The construction of the heat shield reference cavity is similar in the initial phase to the foam insulated reference cavity. The main difference being it uses two cylindrical heat shields to limit thermalization due to the effect of blackbody radiation as discussed in appendix C. The same mirrors with a radius of curvature of 20 cm are used and fixed with Torr Seal to a 5 cm long cavity spacer of type 1 grade Zerodur. Type 1 Zerodur has a temperature
Figure 3.15: Schematic of heat shield insulated cavity showing Zerodur cavity spacer mounted on a pedestal using three pieces of rubber. The cavity spacer is then placed inside an aluminium cylindrical inner heat shield, which contains a hole in both ends for optical access. This heat shield is then placed inside a second temperature stabilized outer heat shield being separated by six small pieces of rubber. The outer heat shield is mounted again with 6 small pieces of rubber inside a custom 6" nipple. This nipple is evacuated meaning thermalization is dominated by radiation. The heat shields ensure low thermal conduction with the environment.
coefficient of $0.0 \pm 0.05 \times 10^{-6} \text{K}^{-1}$. The Zerodur has two slots cut into it around the middle of the cylinder this allows two aluminum semi-circle profiles to locate within the slots cut in the Zerodur. The aluminum mounts sit on three pieces of Viton rubber that sits on an aluminum pedestal shown in Fig. 3.16. This pedestal is mounted inside an inner aluminum heat shield. The inner heat shield sits on three pieces of Viton rubber inside an outer heat shield. The outer heat shield is temperature stabilized to $27^\circ \text{C}$ in the same way as the foam insulated cavity discussed in Section 3.5.1.

The outer heat shield has two Indium Tin Oxide (ITO) coated widows glued with Torr Seal to its top and bottom caps shown in Fig. 3.19. These windows allow visible and Near Infra-red (NIR) radiation to pass through with minimal attenuation, but they are highly reflective to far-IR radiation, which is where the bulk of the blackbody radiation spectrum at room temperature lies. The ITO coating has the effect of reducing the emissivity of the window from glasses value of 0.9 to a value similar to the surrounding aluminum, which is typically around 0.1. The window is in contact with aluminum shield, therefore shield and ITO window are the same temperature. The whole setup...
is placed inside a 6.0 inch vacuum nipple that has two 2.75 inch nipples welded to it. The 2.75 inch nipples act as feedthroughs for the wires used in the active temperature stabilization system and another for the copper pinch off tube. Optical axis is provided by two quartz 2.75 inch view ports, which are locate in the top and bottom ports. The cavity drift can then be monitored by using the same method used to measure the drift of the foam insulated cavity. The residual drift of the cavity is given in Fig. 3.18. The average drift rate for this cavity is found to be 25 Hz/sec. The linewidth measurement of the cavity at 911 nm is shown in Fig. 3.17. The drift rate of the this cavity is higher than what we expected and this could be due to the Torr seal used to fix the mirrors changing length due to temperature fluctuations. This cavity is used as a reference for the 455 nm and 615 nm Raman lasers.

![Graph](image)

**Figure 3.18:** Drift of reference cavity two measured over two different time periods. (a) compares the passive drift of the cavity against the drift when the outer heat shield has been temperature stabilized. (b) shows the drift of the cavity a month after the temperature stabilization has been engaged showing the drift over three days. This plot shows the cavity drifts less than 1 MHz in a three day time period.

### 3.5.2.1 Improvements to heat shield reference cavity

A third cavity of the same design as the heat shield reference cavity has been built to see if the average drift of the heat shield cavity can be reduced further. This cavity employs Layertec mirrors with a coating spanning 1450 nm to 1800 nm and a radius of curvature of 100 cm for future use with a 1762 nm laser, which is needed to address the $6S_1/2$ to $5D_5/2$ quadrupole transition. The mirrors are fixed with the same UV
Figure 3.19: Image showing outer heat-shield for reference cavity. The outer shield has had ITO coated window glued over the openings in the end caps. The ITO window reflects far-IR radiation responsible for the majority thermal heat, but it allows visible and near-IR to pass with minimal attenuation.

Figure 3.20: Linewidth of Layertec mirrors at 1762 nm measured using sideband method specified in Section 3.5.1. Sidebands not shown in plot to allow more detail of peak fit to be seen. The cavity mirrors have a linewidth of 550 kHz at 1762 nm.

curing epoxy, the amount used is much less than the Torr Seal, so any change in its length should have a negligible effect on the total cavity length. A small five liter per second ion pump is used to ensure the chamber pressure does not gradually increase over time, which would change the index of refraction inside the vacuum chamber. Type 0 Zerodur is used for the cavity spacer, which offers an improvement in length stability over type 1. As the ITO coated windows were difficult to acquire they were not used in the construction of this cavity. The lack of ITO coated windows allows us to make a comparison with cavity number two, to access their effect. The cavity drift is monitored with a 1560 nm laser, which is referenced to the frequency comb. The linewidth of the cavity at 1762 nm is shown in Fig. 3.20. The residual cavity drift can be seen in Fig. 3.21. The average drift rate for this cavity is 10 Hz/sec.
Figure 3.21: Plot showing drift of reference cavity three in the best case scenario. This drift measurement represents the best drift rate obtained from monitoring the cavity over a period of a month. The drift rate extracted from this data represents a drift rate of less than 1 Hz/sec.

Table 3.3: Comparison of drift rates between cavities designs. Cavity number one corresponds to the foam insulated, cavity number two corresponds the heat shield insulated with ITO windows, cavity number three corresponds the heat shield insulated with an ion pump and without ITO windows.

<table>
<thead>
<tr>
<th>Cavity number</th>
<th>Zerodur grade</th>
<th>Average drift rate</th>
<th>Max drift rate</th>
<th>Drift range</th>
</tr>
</thead>
<tbody>
<tr>
<td>One</td>
<td>0</td>
<td>25 Hz/sec</td>
<td>50 Hz/sec</td>
<td>± 2 MHz</td>
</tr>
<tr>
<td>Two</td>
<td>1</td>
<td>20 Hz/sec</td>
<td>50 Hz/sec</td>
<td>± 2 MHz</td>
</tr>
<tr>
<td>Three</td>
<td>0</td>
<td>5 Hz/sec</td>
<td>20 Hz/sec</td>
<td>± 1 MHz</td>
</tr>
</tbody>
</table>

3.5.3 Comparison of Cavities

Drift data has been collected over the period of at least two weeks to ascertain the average drift rates between the different cavity designs. The drift data is only compared once the cavities have been temperature stabilized for over one week to ensure they have reached equilibrium. The drift rates for all the cavity designs tested are shown in Table 3.3. A conclusion of the effect the ITO windows have on the overall drift rate could not be made, due to the fact cavity 3 had the addition of an ion pump and a better grade of Zerodur, whereas cavity 2 did not. We can only assume the effect is negligible because cavity 3 has a lower drift rate compared with cavity 2. The current drift rates of the cavities are acceptable for performing the experiments covered in this
thesis because over the time period of one typical measurement, 15 minutes, the cavity will drift less than 15 kHz, which is within our acceptable limits.

The drift issues of the cavities can be improved further by implementing a servo method that monitors the drift of the cavities and provides a correction to the frequency of the laser beam of interest. The monitoring can be done by implementing a master laser setup, which is tightly locked < 10 kHz to an atomic reference or referenced to the frequency comb. The master laser can be used to track the drift of a cavity through adjusting the modulation frequency of a broadband EOM. This frequency can then be adjusted for the wavelength difference between the master laser and the target laser by a frequency multiplication board, which is then feed to an AOM acting on the target laser to compensate for the drift of the cavity. Improvements to the passive stability would also be helped by gold coating the outside surface of the heat shields, as gold is good at reflecting far-IR radiation and providing a second housing around the reference cavities, which is also actively temperature stabilized.

3.5.4 Laser locking scheme

Our lasers are locked to our reference cavities using the PDH technique [83]. This technique combined with a fast feedback loop allows our lasers to be stabilized to below 10 kHz to an optical cavity with a linewidth of around 1 MHz. The way we implement the lock for the 455 nm and 615 nm lasers is shown schematically in Fig. 3.22. This locking scheme is used for all of the lasers in our lab. The key component in the locking scheme is the EOspace\(^1\) EOM, which allows us to arbitrarily lock our lasers to any frequency within the laser diodes tuning range, as the bandwidth of the EOspace is larger than the free spectral range of our cavities.

In detail, the 911 nm laser passes through a Polarizing Beam Splitter (PBS), where the majority of the power is split off for doubling to 455 nm. The doubled light passes through an AOM to provide optical switching of the beam and is fibre coupled to the experiment chamber. The rest of the 911 nm light is sent through an EOspace EOM to modulate the laser beam. It is then sent through a second EOM at \(\approx 20\) MHz to provide modulation for the PDH lock-in detection (not shown in Fig. 3.22). The light passes through a second PBS, a dichroic mirror used for combining in a master laser beam (optional), which is used for tracking the drift of the cavity and a \(\lambda/4\) waveplate.

\(^1\)http://www.eospace.com/ EOspace part number: PM-3K5-10-PFA-PFA-850-UL
Figure 3.22: Schematic of Raman laser lock setup for 455/614 nm Raman laser system. The broadband cavity mirror coatings allow for the 910 nm and 614 nm laser to be locked to the same cavity, which is important for Raman transition as it reduces the two beams relative linewidths. The locking scheme is almost standard for all the ion trap lasers. The setup allows for an optional reference beam to be coupled into the cavity to probe the drift rate. The key component in the setup is the EOspace, which allows a sideband of the laser light to be adjusted from 100 MHz to 10 GHz.
Table 3.4: Stable barium isotopic abundances [84], masses, nuclear spin and isotope shift [85]. The stable isotopes of $^{130}\text{Ba}$ and $^{132}\text{Ba}$ are not included in this table as their abundance is negligible ($<0.1\%$).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mass (amu)</th>
<th>Abundance</th>
<th>Nuclear spin</th>
<th>Isotope shift relative to $^{138}\text{Ba}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{134}\text{Ba}$</td>
<td>133.904486</td>
<td>2.42 %</td>
<td>0</td>
<td>122 MHz</td>
</tr>
<tr>
<td>$^{135}\text{Ba}$</td>
<td>134.905665</td>
<td>6.59 %</td>
<td>3/2</td>
<td>220 MHz</td>
</tr>
<tr>
<td>$^{136}\text{Ba}$</td>
<td>135.904553</td>
<td>7.85 %</td>
<td>0</td>
<td>109 MHz</td>
</tr>
<tr>
<td>$^{137}\text{Ba}$</td>
<td>136.905812</td>
<td>11.23 %</td>
<td>3/2</td>
<td>183 MHz</td>
</tr>
<tr>
<td>$^{138}\text{Ba}$</td>
<td>137.905232</td>
<td>71.70 %</td>
<td>0</td>
<td>–</td>
</tr>
</tbody>
</table>

After that the light is sent to the reference cavity and the non resonant light is reflected back through the $\lambda/4$ waveplate, which has effectively rotated the polarization by 90 degrees compared to the input beam. The reflected light passes through the PBS and on to a fast photo-diode, which can detect the modulation in the laser beam. The 614 nm laser follows the same setup except it has already been doubled so does not require the SHG stage and does not require an EOspace as only one laser needs to be scanned to satisfy the Raman resonance condition defined in Eq. (2.40). We do not use the 455 nm or the 1230 nm for locking due to the optical bandwidth of the cavity mirrors.

### 3.6 Photo-ionization

We use photo-ionization to ionize barium because it is isotope selective, which is important as barium has seven stable isotopes. The abundances of these isotopes are given in Table 3.4. Furthermore photo-ionization does not cause significant charging of the ion trap compared to ionization via electron bombardment. We implement a three-photon ionization technique, where the first transition provides isotope selectivity. In Section 3.6.1 we discuss the atomic levels involved in the photo-ionization scheme and the ionization laser. In Section 3.6.2 we discuss the design of the barium vapor cell, which is used as a reference for stabilizing the lasers. In Section 3.6.3 we compare our method to photo-ionization methods of other groups.
Figure 3.23: Energy level diagram for $^{137}$Ba showing the three transition used to ionize barium and their respective wavelengths.
3.6.1 Barium spectroscopy and ionization lasers

Our photo-ionization procedure involves three steps and is partly based on [86, 87]. The first step is to excite the atom from $6s^2^1S_0$ ground state to the $6s6p^3P_1$ excited state with a narrow linewidth laser at 791 nm. Since this transition is spin forbidden its linewidth is only approximately 50 kHz [88]. This is much smaller than the isotope shifts tabulated in Table 3.4 and thus this transition provides isotope selectivity. The second step is to excite the atom from the $6s6p^3P_1$ state to the $6p^2^3P_1$ state via a second laser at 450 nm. From here the electron is stripped from the atom with a large electric field provided by a high power 650 nm laser. The relevant level scheme is shown in Fig. 3.23.

![Neutral barium spectrum from 791 nm transition. The frequency axis is mapped to the profile according the relative isotope shift between $^{134}$Ba and $^{136}$Ba [85].](image)

For loading the isotope of interest, namely $^{137}$Ba, we rely on the isotope selectivity of the 791 nm transition. A typical saturated absorption spectrum of this transition is
shown in Fig. 3.24, where the peaks due to the different isotopes are clearly resolved. By locking the 791 nm laser to the corresponding feature one can ensure that it has the correct frequency. However, the isotope selectivity when ionizing in the experiment chamber could be affected by the following factors: power broadening of the transition and Doppler shifts in the transition frequency. Power broadening can easily be avoided as long as the laser intensity is approximately equal to the saturation intensity of the transition. Since Doppler shifts are caused by the relative angle between the ionization lasers and the atomic beam used for the loading the ion trap, it can be eliminated by orientating the atomic beam to be perpendicular with respect to ionization lasers. The residual Doppler broadening due to the divergence of the atomic beam persists in any case. This can potentially broaden the width of the loading spectra to 100 MHz (FWHM) as reported in [86]. In our case the atomic beam in the experiment trap produced by the oven is orientated perpendicular to the laser beam and the atomic beam is collimated by the use of a small opening before arriving at the trap, limiting the divergence angle, which helps reduce broadening effects.

In order to make sure that the second step in the ionization happens at a high enough rate we lock the 450 nm laser to the transition of interest. Upon locking the 791 nm laser, sweeping the frequency of the 450 nm laser over the transition resonances gives a Doppler-free absorption profile as shown in Fig. 3.25. The profile is Doppler-free because the two-photon transition happens very fast compared to changes of the atom velocity. Only velocity classes that have been excited in the first step can be excited in the second step. Further details of this can be found in [89]. In the work presented here we use this profile to lock the 450 nm laser.

For the third step we use a high powered 650 nm laser beam to ionizes the ion from the $6p^2 3P_1$ state. Initially we relied on the 450 nm laser to perform this step, which meant that we needed much higher powers than the saturation intensity. The frequency of this laser is sufficiently close to the $6S_{1/2}$ to $6P_{3/2}$ transition in Ba+, so that off-resonant excitation of the ion due to 450 nm beam would causes it to decay to the metastable dark state $5D_{5/2}$. In order to avoid this we lower the power in the 450 nm beam and use the high power 650 nm Raman beam to perform the final step in the ionization process. An alternative remedy is to combine 615 nm repump light with the ionization beams to de-populate the metastable dark state.
Overall we get an ionization rate of approximately 0.2 ions/sec. This can easily be increased by increasing the oven temperature slightly, which results in multiple ions being loaded per second.

![Graph showing Doppler-free absorption profile of Ba for the transition $6s6p^3P_1 \rightarrow 6p^2^3P_1$. The frequency axis is mapped to the profile according to the relative isotope shift at state $6s6p^3P_1$ between $^{138}$Ba and $^{136}$Ba.]

**Figure 3.25:** The Doppler-free absorption profile of Ba for the transition $6s6p^3P_1 \rightarrow 6p^2^3P_1$. The frequency axis is mapped to the profile according to the relative isotope shift at state $6s6p^3P_1$ between $^{138}$Ba and $^{136}$Ba.

### 3.6.2 Reference cell

To implement the ionization scheme the 791 nm and 450 nm lasers must be on resonance with the relevant transitions in the isotope of interest. This is achieved by locking the lasers to the atomic transitions in a barium vapor reference cell. A schematic of the setup is shown in Fig. 3.26.

To achieve a clear saturated absorption profile the optical density of barium must be high enough that a significant proportion of the 791 nm beam is absorbed. The optical
Figure 3.26: The barium spectroscopy setup utilizes a heat pipe oven design to obtain the required vapor temperature, which gives the necessary optical depth. The cell is over 0.5 m long to ensure the windows remain at room temperature and also to maximize the interaction length. The cell utilizes a buffer gases of argon to ensure barium vapor does not condense on the windows, which would lead to attenuation of the probe beams. The 791 nm is in a saturation absorption configuration, this enables the individual isotopes of barium to be resolved. The 455 nm beam is combined with the 791 nm via a dichroic mirror.

The density is proportional to the vapor pressure of barium, which in the temperature range of 700-1200 K is given by

$$\log P(\text{Pa}) = 9.733 - \frac{9304}{T},$$

where $P$ is the pressure in pascals and $T$ is the temperature in Kelvin. At room temperature the vapor pressure of barium is very low and is not sufficient to provide a saturated absorption profile. Hence, the reference cell has to be heated up 500°C to increase the vapor pressure to the required optical depth.

The reference cell design utilizes a heat pipe oven design, which is described in [91]. It consists of a stainless steel tube which contains a small piece of solid barium. Both ends of the tube are sealed with 2.75 inch diameter quartz windows. After a piece of solid barium is put in place, the cell is evacuated down to $10^{-6}$ Torr and then argon is back filled to a pressure of $10^{-2}$ Torr. The argon acts as a buffer gas to contain the barium vapor and prevent it from condensing on the windows. We operate the reference cell below the melting point of barium and at a vapor pressure lower than the argon pressure, which results in barium migrating and condensing to cooler parts of the tube, requiring the barium to be refilled in the cell after about six months of continuous use.
To obtain the saturated absorption spectra for transition $^1S_0 \rightarrow ^3P_1$, a 791 nm laser beam with approximately 12 mW power and 10 mm diameter is sent into the cell with vertical polarization to saturate the atomic transition. After passing through the cell, the beam is retro-reflected to probe the saturation profiles. A quarter waveplate and optical attenuator are placed in the reflection path to rotate the polarization and reduce the reflected beam power by $\sim 20$ times. Upon completing a round trip, the beam is transmitted through the PBS and picked up by a photodiode, as shown in Fig. 3.26. Sweeping the laser frequency over the atomic resonances results in the spectrum shown in Fig. 3.24. The 450 nm laser is combined with the 791 nm via a dichroic mirror. It then passes through the cell once before it separated with a second dichroic mirror and detected with a photo-diode. Locking the 791 nm and sweeping the 450 nm laser results in the spectrum shown in Fig. 3.25.

3.6.3 Comparison to other ionization methods

It is interesting to compare our method to photo-ionization schemes implemented by different groups such as [86, 87, 92]. We achieve isotope selectivity through use of a 791 nm transition, whereas the author in [92] reports single step isotope selective ionization via a two-photon transition using a single 413 nm laser. Using only one laser greatly simplifies this step, however 413 nm sources are not readily available and are expensive. Our scheme in turn relies on readily available cheaper diodes.

Our ionization scheme uses laser light in well defined spatial modes which has advantages over methods using incoherent light sources such as the LED’s as used in [87]. Blue light impinging on metal surfaces can release free-electrons due the photo-electric effect. These electrons lead to charge build up which dissipates throughout the experiment. This results in time-varying stray fields which limits the effectiveness of micromotion compensation. This is particularly problematic in barium experiments where barium deposition on the electrodes lowers the work function to such a degree that wavelengths below 500 nm is sufficient to induce the photo-electric effect. Indeed, we have observed charge build up if the 493 nm cooling light impinges on the trap electrodes.

The method presented here only relies on the use of commercially available laser diodes, which are much cheaper and simpler to use compared to the nitrogen gas laser used in [86]. The nitrogen laser provides light at 317 nm, which is difficult to work with.
as it can not be fibre coupled and is absorbed heavily by standard boro-silicate glass, of which our viewports are made from. Additionally, the 450 nm laser used in our scheme could be replaced by a 457 nm laser, which excites the atom from the 6s6p3P1 level to the 6p23P1 level. A laser tuned to resonance with this transition, is approx 2 THz detuned from the 6S1/2 ↔ 6P3/2 transition of Ba+ and can therefore potentially be used as Raman laser for the 6S1/2 to 6P3/2 transitions.

3.7 Antenna design

To drive the hyperfine transitions an rf source is required. As the ion trap was not initially setup with this experiment in mind, the rf source had to be provided from outside the chamber. Creating an antenna for the rf source is not too difficult, but coupling enough power into the chamber is more challenging. The problem is that the frequency range of interest is in the hundred’s of MHz range and therefore the wavelength is on the order of a meter. Since the largest windows have a diameter of 100 mm the chamber effectively acts like a Faraday cage, attenuating any wavelength that is larger than the window diameter. The antenna used to drive the hyperfine transitions is a λ/4 whip antenna [93] design which is relatively simple and easy to make. The first antenna was made from a 15 cm long M6 threaded rod mounted onto a Teflon stand. A λ/4 wave whip antenna is a resonant design when placed on an infinite ground plane such as a laser table. The antenna is impedance matched by the use of a simple tank circuit. The resonant design of the antenna meant we would need a different antenna for each wavelength. We found from experimental evidence that by applying up to a 1 W in power to the antenna meant that even in the worst case scenario we could drive all rf transitions in the 5D3/2 manifold at ≈ 1 kHz Rabi rate. This meant we only required one antenna for the hyperfine splittings measurement of the 5D3/2 manifold. For the 5D5/2 manifold the 15 cm antenna was not able to drive the transitions at a reasonable rate so a 70 cm λ/4 whip antenna was made instead. The antenna’s are driven from a computer controller signal generator that has been synchronized to a Global Positioning System (GPS) stabilized Rb based atomic clock1. The rf signal passes through an rf switch that is connected to the software running the experiment.

1Precision test systems limited model GPS10RBN
3.8 Imaging system

Figure 3.27: A schematic diagram of the imaging system. The imaging system consist of a multi-element array imaging lens, which can resolve single ions spaced by $\approx 2 \, \mu m$. An iris is used to spacial filter out unwanted background light. The iris is imaged onto either a CCD camera or SPCM using a pair of acromats. An interference filter at 493 nm is used to further remove any background light that can lead to false photon counts. A flipper mirror to direct light to either the SPCM or the CCD camera. The inset image shows single $^{137}$Ba$^+$ ion trapped in the experiment ion trap.

The imaging system is used for collecting and counting the ion’s fluorescence. A schematic representation of the imaging system is given in Fig. 3.27. In brief, the ion florescence is collected through a 43 mm diameter multi-element custom designed lens (diffraction limited for 493 nm)$^1$. The lens is designed to compensate for refraction caused by a 2.75 inch vacuum view port. The lens has a focal length of 105 mm. The collected light is focused down using a 300 mm acromat and reflected off a 99.9%

$^1$http://www.photongear.com/
reflective mirror through a 75 μm pin hole to spatially filter out unwanted background light. The light then passes through a narrow band interference filter\(^1\) at 493 nm. The pin hole is imaged with 100 mm and 150 mm acromats in back to back configuration, which focuses the fluorescence onto a Charge Coupled Device (CCD) or Single Photon Counting Module (SPCM) depending on the position of a flipper mirror. The SPCM has a dark count rate of less than 150 photons per second and a dead time of 30 ns. Overall the imaging system collects 0.25(5)±% of the florescence into the SPCM. This value includes the quantum efficiency of the SPCM at 493 nm and all other losses. The SPCM has a quantum efficiency of 45% at 493 nm and 72% at 650 nm.

\(^{\text{1}}\)www.semrock.com
Chapter 4

Experimental Methods

In this chapter we explain the different processes that are used to measure the hyperfine transition frequency. The measurement of a hyperfine interval follows this procedure, cool the ion, prepare the ion in a known starting state in the 5D manifold of interest, perform an rf transition between two hyperfine states in this manifold, shelve the ion to the detection state and finally perform detection. To perform the hyperfine interval measurement five experimental techniques are utilized, Doppler cooling, optical pumping, Raman transitions, rf transitions and fluorescence detection, as illustrated in Fig. 4.1. Further information about ion loading and cooling procedures can be found [58].

In Section 4.1 we describe how we implement Doppler cooling. In Section 4.2 we cover optical pumping, which is used to place the ion in an initial state. In Section 4.3 we describe the two-color Raman transition that are used to shelve the ion into the 5D manifolds. To measure the energy splitting of the hyperfine intervals an rf spectroscopy procedure is utilized which is described in Section 4.4. Section 4.5 details the fluorescence detection that is used to test whether a hyperfine transition took place or not. These five sections cover the individual steps required to perform the octupole measurement.

4.1 Doppler cooling

The experiment relies on Raman transitions, in order to perform them efficiently, the ion must be Doppler cooled to a low vibrational state [94]. Doppler cooling is achieved
Figure 4.1: Hyperfine interval measurement steps for 5D$_{3/2}$ manifold. (a) the ion is cooled to the Doppler limit. (b) the ion is optically pumped into the \( |F = 2, m_F = 2 \rangle \) of the 6S$_{1/2}$ manifold. (c) the ion is shelved to the \( |F'' = F, m_{F''} = 0 \rangle \) by a two color Raman transition. (d) an rf pulse is applied to drive the rf transition. (e) the ion is mapped to the 5D$_{5/2}$ state for detection by another Raman transition and an optical pumping step at 455 nm. (f) fluorescence detection is performed on the ion.
by driving the $6S_{1/2}$ to $6P_{1/2}$ transitions at 493 nm and repumping on the $5D_{3/2}$ to $6P_{1/2}$ transitions at 650 nm. The 493 nm laser is passed through two EOMs in order to generate sidebands to address all the hyperfine states of $6S_{1/2}$ and $6P_{1/2}$ manifolds. Similarly, to address all the hyperfine states of the $5D_{3/2}$ manifold, the repump laser at 650 nm is split into four beams, frequency shifted by AOMs, and then recombined into a single fibre. All laser fields are linearly polarized perpendicular to a magnetic field of greater than 0.3 G. This configuration avoids unwanted dark states in both the cooling and detection cycles. Additionally, the field ensures a well defined quantization axis for optical pumping and state preparation.

4.2 Optical pumping

Optical pumping is an essential tool in atomic physics experiments [44], as it provides a method for placing an atomic system into a well defined state. Optical pumping is the process where an atom repeatedly scatters resonant light until it decays via spontaneous emission into a so-called dark state. This dark state is decoupled from the resonant light and the atom no longer scatters. Optical pumping relies on finding an atomic level structure that has a strong scattering transition and a long lived state that the excited state can decay into, which is decoupled from the resonant light used for scattering. One such structure is a three level system with a metastable ground state which is far detuned from the scattering transitions resonant frequency. Another utilizes polarization modes of light and a well defined quantization axis to decouple a magnetic sub level from the scattering transitions. We exploit both of these optical pumping schemes for measuring the octupole moment.

To initialize the ion in a well defined state we pump it into the $|F = 2, m_F = 2\rangle$ state of the $6S_{1/2}$ manifold. After performing Doppler cooling the ion is in an undefined state in $6S_{1/2}$ manifold and is pumped into the initial state by applying resonant light at 493 nm and 650 nm with a specific polarization, as illustrated in Fig. 4.2. The 493 nm is modulated by an EOM to address both the hyperfine state in the $6S_{1/2}$ manifold, which makes sure no unwanted population ends up in the $F = 1$ state. The 650 nm beam repumps any population that has fallen into the $5D_{3/2}$ state. If $\sigma^+$ polarized light is used to drive the $6S_{1/2}$ to $6P_{1/2}$ the ion is pumped into the $|F = 2, m_F = 2\rangle$ state by the process of absorption and spontaneous emission. The ion will remain in
Figure 4.2: Schematic of optical pumping used to prepare the ion in the \( |F = 2, m_F = 2\rangle \) state of the \( 6S_{1/2} \) manifold. A 493 nm beam with \( \sigma^+ \) polarization is used optically pump the ion into the \( |F = 2, m_F = 2\rangle \) state. The 493 nm laser has frequency sidebands added to ensure all hyperfine levels are addressed. A laser at 650 nm is used to repump any population that has fallen into the \( 5D_{3/2} \) manifold.

Once the ion is in \( |F = 2, m_F = 2\rangle \), the \( |F = 2, m_F = 2\rangle \) once it has decayed there because the \( \sigma^+ \) polarized light decouples it from the excited state. The efficiency of this process is limited by the quality of the polarization and we readily achieve a \( |F = 2, m_F = 2\rangle \) state population of greater than 99%.

Along with state initialization, optical pumping can be used for shelving the ion into the dark state for use in the detection process. Starting from the \( |F = 2, m_F = 2\rangle \) state optical pumping is performed by applying a 10 \( \mu s \) resonant pulse of 455 nm laser light to excite the ion up to the \( 6P_{3/2} \) state. From there it can fall back into the scattering cycle by decaying back to the \( 6S_{1/2} \) state or it leaves the scattering cycle and falls into either \( 5D \) state. In our detection scheme the \( 5D_{3/2} \) state acts as the dark state because it is decoupled from the detection cycle. The efficiency of this scheme is limited by the 13% probability the ion has of decaying into the \( 5D_{3/2} \) state. In theory any population that decays into the \( 5D_{3/2} \) state can be placed back in the pumping cycle through the application of a laser at 587 nm. In our detection scheme this is not possible as \( 5D_{3/2} \) state acts as a bright state and repumping at 587 nm would disturb this state. The scheme is shown schematically in Fig. 4.3. More details on the fluorescence detection schemes are given in Section 4.5.1.
Figure 4.3: Schematic of optical pumping to 5D$_{5/2}$ state used to transfer the ion from the 6S$_{1/2}$ to the 5D$_{5/2}$ manifold. This is done by the application of a resonant beam at 455 nm. The ion has a 13\% chance to fall into the 5D$_{3/2}$ manifold which would result in an incorrect state discrimination event in our detection scheme.

4.3 Two-color Raman transitions

For the hyperfine splittings measurements the ion needs to be transferred to a specific \( F \) and \( m_F \) level of the 5D manifold of interest from the \( |F = 2, m_F = 2\rangle \) state in the 6S$_{1/2}$ manifold. We achieve this through a two-color Raman transition. Typically this would be done by direct shelving through a quadrapole transition [95, 96]. Driving a quadrapole transition is technically challenging as it puts stringent requirements on the laser linewidth and typically involves slow transition rates. The slower transition rates mean noise sources such as magnetic field fluctuations become a serious concern. A two-color Raman transition can overcome the limitations of direct driving as the Raman transition acts through two dipole transitions. This enables faster transition rates and reduces the individual laser linewidth requirements.

The Raman transition process works by applying a \( \pi \) pulse (explained in Section 2.3.2) of the two Raman beams. The two-color radiation transfers the ion from the initial state to the final state with near unit probability. To ensure this process takes place with optimal efficiency the Raman resonance condition must be satisfied. In order to find the Raman resonance the frequency of one the laser beams is varied in frequency steps at which the transition is repeated enough times to find the probability that the ion has been transferred to the final state. Mapping out the full frequency dependence
results in a sinc function spectrum (see Eq. (2.35)) like the one shown Fig. 4.4. The center frequency of the sinc function corresponds to the resonance frequency.

The two Raman transitions to the $5D_{3/2}$ and $5D_{5/2}$ states have slight differences and will be discussed in Section 4.3.1 and in Section 4.3.2 respectively.

![Figure 4.4: Plot showing Raman scan between $|F = 2, m_F = 2\rangle$ of the $6S_{1/2}$ to $|F'' = 2, m_{F''} = 0\rangle$ of the $5D_{5/2}$ level.](image)

Fundamentally the efficiency of a Raman transition is limited by the process of spontaneous emission. However, additional imperfection due to effects from magnetic field fluctuations, imbalanced ac-Stark shifts, phase jitter of the driving lasers and finite ion temperature reduce the efficiency below this fundamental limit. The first three effects can be viewed as causing changes to the transition frequency or changes to the laser field, whereas the finite ion temperature can viewed as a deviation from the dipole approximation [41].

Phase jitter causes the two Raman lasers to dephase on time scales longer than their relative linewidths. On these long time scales the interaction between the ion and the lasers is no longer coherent and the transition efficiency drops. Magnetic field fluctuations on the other hand cause the levels to shift, which in turn changes the Raman resonance condition. Finally unbalanced Stark shifts cause a relative shift of the energy levels if the beam intensities are imbalanced or the driving pulse is not a perfect rectangle.

The finite ion temperature manifests itself in the transition efficiency through the fact that the LD parameter defined in Eq. (2.50) can not be reduced to zero. In our
specific case this can be explained by the fact we are performing Raman transitions between states that are separated by optical frequencies. The LD parameter compares the transition wavelength being addressed to the ground state wavefunction spread of the ion, which is not negligible in this case. For Raman transitions between hyperfine states on the other hand, the LD parameter is negligible because the wavelength scale for microwave transitions is much larger than the wavefunction of the ion.

These individual error sources are discussed separately in more detail for state transfers to the 5D_{3/2} and 5D_{5/2}, as they have different effects in the two situations, which are given in Sections 4.3.1 and 4.3.2 respectively.

4.3.1 Transitions to the D_{3/2} manifold

The 493/650 nm Raman transition takes the ion from the \(|F = 2, m_F = 2\rangle\) state of the 6S_{1/2} manifold to the \(|F'' = k, m_{F''} = 0\rangle\) state of the 5D_{3/2} manifold, where \(k = 0, 1, 2\). The shelving scheme is shown schematically in Fig. 4.5 (a). We only shelve to the \(|F'' = k, m_{F''} = 0\rangle\) of the 5D_{3/2} manifold because from these states all the other states involved in measuring the hyperfine intervals can be reached via an rf transition, as explained in more detail in Section 5.1. Shelving to the 5D_{3/2} manifold is performed by two co-propagation laser beams at 493 nm and 650 nm. This is done to minimize the LD parameter. The availability of chamber viewports limits the polarization that can be set for the beams. The Raman beams are aligned perpendicular with respect to the magnetic field which results in perpendicular polarization. Ideally, the polarization should be \(\sigma^-\) for the 493 nm and \(\sigma^+\) for the 650 nm beam to address the states of interest because this ensures all the optical power is in the correct polarization mode. Perpendicular polarization has the effect of putting both beams in a superposition of \(\sigma^-\) and \(\sigma^+\), effectively reducing the power by 1/2 for the polarization of interest.

The intensity of the 493 nm and 650 nm beams are set to be equal and with the available power we achieve a Raman rate of \(\Omega_R = 1\) MHz for a Raman detuning of \(\Delta = 500\) GHz. The two beams pass through separate AOMs for switching to achieve precise pulse lengths. The AOMs have been synchronized to ensure perfect pulse overlap between the two beams. The limiting factor when using AOMs for switching is that they have a finite rise time that can be up to 50 ns in length. For pulses faster than 1 \(\mu s\) this becomes a problem as the pulse shape is more trapezoid than square, which ends up effecting the transfer efficiency. At these short pulse times this is the limiting factor
where as for longer pulse times the magnetic field fluctuations and laser phase jitter limit the transfer efficiency.

\[ \Delta \approx 500 \text{ GHz} \]

\[ \Delta \approx 20 \text{ GHz} \]

**Figure 4.5:** Two-color Raman shelving process. (a) Raman shelving process to 5D_{3/2} manifold. (b) Raman shelving process to 5D_{5/2} manifold.

Using the parameters described above we achieve a Raman transition efficiency of 95\%. To try to understand the mechanism that causes the efficiency to be lower than unity we performed a Rabi oscillations experiment to measure the decoherence time for the Raman transition. The measurement is shown in Fig. 4.6, which gives a dephasing time constant of 4.4 \( \mu s \). Going through the prominent error source mentioned in Section 4.3 the dephasing due to the phase jitter of the lasers can be neglected as the lasers relative linewidth is less than 1 kHz, which is much smaller than the Raman rate. Shelving problems from magnetic field noise can also be ignored as this only becomes a problem when the Raman rate is on the order of 1 kHz. This leaves dephasing due to the finite ion temperature and imbalanced ac-Stark shifts to be investigated. The axial trapping frequency of 400 kHz gives a LD parameter of \( \eta_z = 0.021 \) and the Doppler cooling limit gives a \( \langle n_z \rangle \approx 25 \) [68], which leads to a dephasing time constant of \( \tau \approx 10 \mu s \). If the ion has a temperature twice the Doppler limit then this would explain the dephasing time. The shelving efficiency can be improved by increasing the trapping frequencies to minimize the LD parameter or by performing resolved sideband cooling [97] to minimize the ion temperature. In addition, Stark shifts could be better balanced by increasing the intensities of the 650 nm beam relative to the 493 nm beam,
to compensate for the relative transition strengths. The shelving efficiency being less than unity does not affect the accuracy with which the hyperfine intervals can be measured. It only affects how much data needs to be collected to achieve the desired accuracy. By re-scaling our data for better shelving efficiency, we estimate that we would only need half as many frequency steps to achieve the same accuracy in the fit if we had unity shelving.

Figure 4.6: Plot showing Rabi flopping between \(|F = 2, m_F = 2\rangle\) of the 6S\(_{1/2}\) to \(|F'' = 0, m_{F''} = 0\rangle\) of the 5D\(_{5/2}\) level. The plot shows dephasing with a time constant of 4.4 \(\mu\)s.

### 4.3.2 Transitions to the D\(_{5/2}\) manifold

The 455/614 nm Raman transition takes the ion from the \(|F = 2, m_F = 2\rangle\) state of the 6S\(_{1/2}\) manifold to the \(|F'' = 2, m_{F''} = 0\rangle\) state of the 5D\(_{5/2}\) manifold. The Raman transition process to the 5D\(_{5/2}\) manifold is shown in Fig. 4.5 (b). We are only interested in shelving to the \(|F'' = 2, m_{F''} = 0\rangle\) of the 5D\(_{5/2}\) manifold, because all other states involved in the hyperfine interval measurements can be reached from this state as explained in Section 6.1. The process of Raman shelving is the same method as described in Section 4.3.1, the only difference being the beams are parallel to the magnetic field so the polarization is set to \(\sigma^-\) for the 455 nm beam and \(\sigma^+\) for the 614 nm beam. The beam intensities have been set to give a Raman rate of 1 MHz at a detuning of 20 GHz. The detuning is small compared with the detuning used for the 493/650 nm Raman transition because there is only 150 \(\mu\)W of power available at 614 nm. The smaller detuning is necessary to be able to achieve a Raman rate of
1 MHz, but is still large enough that off-resonant scattering out of the 5D\textsubscript{5/2} manifold caused by the Raman beams is not an issue. Using these parameters we can achieve a Raman transfer efficiency of greater than 92%.

![Graph](image_url)

**Figure 4.7:** Plot showing Rabi flopping between $|F = 2, m_F = 2\rangle$ of the 6S\textsubscript{1/2} to $|F'' = 2, m_{F''} = 0\rangle$ of the 5D\textsubscript{5/2} level. The plot shows dephasing with a time constant of 6 $\mu$s.

Again the reason for the lower than unity Raman transition efficiency is investigated using the Rabi oscillations method described in Section 4.3.1. The dephasing of the transition can be seen in Fig. 4.7, where the transition decays with a time constant $\tau = 6$ $\mu$s. The sources of dephasing described in Section 4.3.1 and also applicable to this Raman transition. The LD parameter for this Raman transition is $\eta_z = 0.024$, which is almost the same as for the Raman transition to the 5D\textsubscript{3/2} manifold. This means the dephasing time constant due to ion motion will be about the same, partially explaining the measured decay time. The imbalanced Stark shift could be a bigger issue here due to the lack of power at 614 nm, which means the intensity of this beam is much lower compared to the 455 nm beam. One way to increase the intensity at 615 nm is to increase the power for this beam. Increasing the intensity by reducing the waist size of the 614 nm beam with the respect to 455 nm beam is not possible as the beams emanate from the same fibre, therefore they pass through the same lens, which defines the ratio of their waists. Another method is to decrease the power of the 455 nm and reduce the detuning to maintain the same Raman rate, but the transition would then be more susceptible to spontaneous emission. Overall shelving via this method is limited to 92%, which is a slight improvement over the method using optical pumping.
which achieved 87% efficiency.

### 4.4 Rf hyperfine transitions

We use rf transitions for measuring the frequency separation of the hyperfine intervals in the 5D$_{3/2}$ and 5D$_{5/2}$ manifolds. Rf transitions are typically used for inducing hyperfine transitions in atomic systems [98] and for precision spectroscopy of the hyperfine structure of atoms [99]. In $^{138}$Ba$^+$ for example they have been used to measure the Zeeman splitting of the 6S$_{1/2}$ and 5D$_{3/2}$ states in [30] and for precise measurements of light shifts in [50]. Alternatively we could measure the hyperfine intervals relative to the 6S$_{1/2}$ state via optical transitions either directly through a quadrupole transition or through a Raman transition like the one described in [32]. However, the use of rf transitions is preferred because they are easier to implement and imparts less of an ac-Stark shift to the states of interest.

A simple picture of the rf transition is that of an electro-magnetic field interacting with a two-level atom as described in Section 2.3.2. In order to induce a transition between hyperfine levels a $\pi$ pulse of the rf field is applied to the ion. To check whether a transition is successful, state selective fluorescence detection described in Section 4.5 can be used for example. By varying the frequency of the rf field, the full frequency spectrum can be mapped out resulting in the characteristic sinc function. From this the center can be obtained via a fit to an accuracy of approximately 100 times smaller than the width of the feature, which is given by the Rabi rate.

In the ideal situation the lifetime of the 5D states would be the limiting factor in how slowly the rf transition can be driven and thus of the accuracy of the hyperfine transition frequency. In practice we are limited by noise sources. The most significant noise source is magnetic field fluctuations. To get an idea of the level of dephasing caused by magnetic field fluctuations we perform Rabi oscillations between the $|F'' = 0, m_{F''} = 0\rangle$ and $|F'' = 1, m_{F''} = 1\rangle$ states in the 5D$_{3/2}$ manifold, which is shown in Fig. 4.8 (a). The decoherence time for this transition is $\tau = 2.5$ ms, which relates to a magnetic field jitter of less than 1 mG, which is consistent with the specified current noise of the power supply used for driving the magnetic field coils. This decoherence value can be compared to an rf transition between the $|F'' = 2, m_{F''} = 0\rangle$ and the $|-\rangle$ in the 5D$_{5/2}$ manifold shown in Fig. 4.8 (b), which for a magnetic field of $B = 1.684$ G is almost
Figure 4.8: Rf Rabi flopping between hyperfine states. (a) Rabi flopping on rf transition, between $|F'' = 0, m_{F''} = 0\rangle$ to $|F'' = 1, m_{F''} = 1\rangle$ states of the 5D$_{3/2}$ manifold. Magnetic field fluctuations cause state dephasing on a time constant of $\tau = 2.5$ ms. (b) Rabi flopping on rf transition, between $|F'' = 2, m_{F''} = 0\rangle$ to $|+\rangle$ states of the 5D$_{5/2}$ manifold. As this transition is magnetic field independent its decoherence time constant is $\tau = 250$ ms.

field insensitive. This transition has a decoherence time of $\tau = 250$ ms suggesting that the principal decoherence source is magnetic field jitter. So in practice the achievable Rabi rate and thus measurement accuracy is limited by magnetic field noise. We also considered the effect of line noise but deemed triggering the experiment on it would lead to biasing of the measurement, due to the different time scales at which the magnetic field splitting and hyperfine interval splittings were made.

4.5 Detection

For the hyperfine intervals measurement we want to detect whether a hyperfine transition took place. To do this we map either the initial or the final hyperfine state onto a dark state. After this is done we apply the fluorescence detection lasers and collect the fluorescence of the ion. The amount of fluorescence collected tells us if the ion is in the bright state or the dark state. Bright states are coupled to a highly fluorescing transition on which the ion can repeatedly scatter. Dark states are off-resonant with or decoupled from the excitation lasers, and the fluorescence is suppressed. More generally, the dark state may be coherently transferred to a far off-resonant state to enhance its immunity to excitation [31, 100].
Figure 4.9: Schematic showing the detection scheme that is used in this work. The scheme relies on being able to map an initial and final hyperfine states onto bright and dark states. This utilizes the larger fine structure splitting between the 5D manifold, as off-resonant scatter from the 5D_{3/2} manifold induced by the 650 nm beam is negligible. In this scheme the 5D_{3/2} manifold is the bright state and the 5D_{5/2} manifold is the dark state.

In the hyperfine intervals measurement, fluorescence detection is applied by using the 493/650 nm lasers to fluoresce the ion. We use the 5D_{5/2} state as the dark state and the 5D_{3/2} state as the bright state, which is illustrated in Fig. 4.9. The 5D_{5/2} state provides a robust dark state, which can not be depumped by the fluorescence lasers because the 650 nm beam is 27 THz detuned from the 6P_{3/2} ↔ 5D_{5/2} transition. For the case of hyperfine interval measurements in the 5D_{3/2} manifold, the initial state of the hyperfine transition is mapped to the dark state and the final state to the bright state. This situation is reversed for hyperfine interval measurements in the 5D_{5/2} manifold, where the initial state is bright and the final state is dark. The detection scheme used for the hyperfine intervals measurement in the 5D_{3/2} manifold is discussed in Section 4.5.1 and the 5D_{5/2} manifold in Section 4.5.2.

4.5.1 Detection method used for hyperfine interval measurement in 5D_{3/2} manifold

To detect whether a hyperfine transition has taken place in the 5D_{3/2} manifold we implement the following scheme. After the rf transition step has been performed, the ion undergoes a Raman transition, as described in Section 4.3.1, which takes the ion
from the \(|F'' = k, m_{F''} = 0\) of the 5D\(_{3/2}\) manifold to the \(|F = 2, m_F = 2\) state of the 6S\(_{1/2}\) manifold. From here the ion is optically pumped using a 455 nm laser into the dark 5D\(_{3/2}\) manifold. These two steps map the initial state of the hyperfine transition to the dark state. Next the fluorescence lasers are applied and the photons scattered by the ion are collected. After the fluorescence lasers have been applied, this state can be emptied by the application of the 614 nm repump laser to return the ion back to the ground state. This scheme is far from ideal as it is limited by the branching ratio of the 5D states from the 6P\(_{3/2}\) state, which limits the detection efficiency to 87%. In theory improvements to the 99% level could be achieved by replacing the 455 nm optical pumping laser with a Raman transition to the 5D\(_{5/2}\) state (see Fig. 4.10).

4.5.2 Detection method used for hyperfine interval measurement in 5D\(_{5/2}\) manifold

![Histograms showing occurrence for number of photons counted in a detection period of 800 \(\mu\)s for bright and dark state. (a) is the detection histogram produced when using the optical pumping method to place the ion in the dark state. (b) is the detection histogram produced when using a Raman transition to place the ion in the dark state. Each histogram consists of 10,000 detection experiments. The leakage of the dark state into the bright state is due to inefficiencies in the shelving process.

In the 5D\(_{5/2}\) manifold the mapping procedure is slightly different. After the \(\text{rf}\) pulse has been applied a second Raman transition takes the ion back from the \(|F'' = 2, m_{F''} = 0\) to the 6S\(_{1/2}\) manifold. Then fluorescence detection is performed and if the \(\text{rf}\) transition was successful the ion remains in the 5D\(_{5/2}\) manifold which is the dark
state otherwise it remains in the bright state. Next the 614 nm repump laser is applied to return the ion back into the cooling cycle. The detection efficiency of this scheme is limited by the efficiency with which the mapping step can be performed. Currently we can only perform Raman transitions to the 5D$_{5/2}$ state with a 92% efficiency due to technical limitations. The bright and dark state photon histograms using this scheme are shown in Fig. 4.10. These histograms are produced by counting the number of photons collected in a 800 µs window by our imaging system, which is described in Section 3.8. The process is repeated 10,000 times to build up a typical reference histogram. The vertical line in Fig. 4.10 represents the threshold number of counts for a detection period of 800 µs. The detection time of 800 µs was chosen to be fast but to provide a good enough contrast to perform the measurement. Overall the detection efficiency changes only how much data we need to achieve a certain accuracy. Any counts to the right of the line are considered a bright state detection event. The penetration of the dark state into the bright state is caused by the imperfect shelving to the 5D$_{5/2}$ state.
Chapter 5

Hyperfine Interval Measurement of D$_{3/2}$ Manifold

This chapter covers the hyperfine interval measurements of the 5D$_{3/2}$ manifold. The experiment performed in this chapter relies on the experimental techniques described in Chapter 4. In this chapter we first describe the procedure used to measure the hyperfine intervals. We then discuss the error sources that affect the experiment. The results obtained are then presented in Section 5.3 and finally the experiment is summarized in Section 5.4. The results in this chapter were reported in [51].

5.1 Method for measuring hyperfine intervals of 5D$_{3/2}$ manifold

The procedure to measure the hyperfine intervals is similar to that proposed in [30, 31] and the relevant level structure is given in Fig. 5.1. The ion is first loaded into the experimental ion trap (Section 3.2.1), Doppler cooled (Section 4.1) and optically pumped (Section 4.2) into the state $|F = 2, m_F = 2\rangle$ of the 6S$_{1/2}$ level. To measure the hyperfine interval, $\delta W_k$ defined in Eqs. (2.18) to (2.20), the ion is shelved to the state $|F'' = k, m_{F''} = 0\rangle$, where $k = 0, 1, 2$, of the 5D$_{3/2}$ manifold, using a two photon Raman transition (Section 4.3.1). An rf antenna (Section 3.7) is turned on to drive $F'' \leftrightarrow F'' + 1$ transitions.

To determine if the hyperfine transition occurred, we use a second Raman pulse to transfer the state $|F'' = k, m_{F''} = 0\rangle$ back to the $|F = 2, m_F = 2\rangle$ ground state.
Figure 5.1: Relevant levels of $^{137}\text{Ba}^+$ for the rf spectroscopy: The ion is prepared in the $|F = 2, m_F = 2\rangle$ state of the $6S_{1/2}$ level from where it is shelved to $|F'' = k, m_{F''} = 0\rangle$ of the $5D_{3/2}$ level with a pair of Raman beams which are red-detuned by $\Delta \approx 2\pi \times 500$ GHz from the $6P_{1/2}$ level. The rf transition is detected by shelving to the $5D_{5/2}$ level (see text).

and then optically pump on the $6S_{1/2}$ to $6P_{3/2}$ transition at 455 nm. If the hyperfine transition occurred, the ion will remain in the $|F'' = k + 1, m_{F''} = 0\rangle$ state of the $5D_{3/2}$ level, otherwise it will be shelved to the $5D_{5/2}$ level. Subsequent driving of the $6S_{1/2}$ to $6P_{1/2}$ and $5D_{3/2}$ to $6P_{1/2}$ transitions using the Doppler cooling beams provides a fluorescence measurement for the probability of driving the hyperfine transition: the ion being bright if the hyperfine transition took place and dark otherwise. The detection efficiency of this scheme is limited to 87% by the optical pumping step because of the branching ratio between the $6P_{3/2}$ and $5D_{3/2}$ which results in unwanted population of the $5D_{3/2}$ level. This does not impact on the accuracy at which we can measure the hyperfine transition probability, but only on the amount of averaging needed to achieve a particular level of accuracy.

We infer the zero field splittings, $\delta W_k$ from measurements at finite magnetic field. To a good approximation the magnetic field gives rise to a second order Zeeman shift of $m_{F''} = 0$ levels, which is quadratic in the magnetic field. We determine the exact magnetic field by measuring transitions between $|F'' = k + 1, m_{F''} = 0\rangle$ and $|F'' = k + 1, m_{F''} = \pm 1\rangle$. Half of the difference frequency between the $\Delta m_{F''} = 1$ and $\Delta m_{F''} = -1$ transitions is given by the linear shift $\mu_B g_F B/\hbar$ with any quadratic shifts canceled. By measuring the $\Delta m_{F''} = 0$ transition over a range of magnetic fields, each calibrated by
measurements of the $\Delta m_{F''} = \pm 1$ transitions, we can map out the full field dependence of the $\Delta m_{F''} = 0$ transition and interpolate to the desired zero field result. This also yields the second order Zeeman shift coefficient for the $\Delta m_{F''} = 0$ transitions providing a useful consistency check within our measurements.

**Figure 5.2:** Plot showing rf resonance scan of $|F'' = 0, m_{F''} = 0\rangle$ to $|F'' = 1, m_{F''} = 0\rangle$ of 5D$_{3/2}$ manifold at a magnetic field of $B = 1$ G

An example of a hyperfine splitting measurement for a fixed value of the magnetic field is shown in Fig. 5.2 where the transition probability for $|F'' = 0, m_{F''} = 0\rangle$ to $|F'' = 1, m_{F''} = 0\rangle$ is plotted as a function of the external rf frequency. The rf drive power is adjusted to give an on resonant $\pi$-pulse time of approximately 10 ms resulting in a resonance width (full width half maximum) of approximately 50 Hz. The resonance is scanned in 2 Hz frequency steps, and the probability of undergoing the hyperfine transition is determined by the average over 200 measurements. The data is fitted [101] via a $\chi^2$ minimization to the Rabi flopping function detailed in Eq. (2.35) with additional offset and amplitude parameters to account for imperfect shelving. From the fits we also extract the 68% confidence limits on the fit parameters giving a determination of the center transition frequency with an accuracy of about 1 Hz. At this same fixed magnetic field, a similar measurement is also performed for the $\Delta m_{F''} = \pm 1$ transitions. For this case we use a shorter $\pi$-pulse time of 0.5 ms and a larger step of 20 Hz for the resonances scans as these transitions are more sensitive to the magnetic field fluctuations and therefore more prone to decoherence effects. This gives a measurement of the resonant frequency for $\Delta m_{F''} = \pm 1$ transitions with an
accuracy of about 20 Hz which corresponds to a field accuracy of $\approx 50 \mu G$.

Measurements for the first hyperfine interval, $\delta W_0$, are shown in Fig. 5.3 (a). For each hyperfine interval we have taken two sets of data on separate days. For each set, we measured the transition frequency at 10 values of the field: five with the field above zero and five below. The inset in Fig. 5.3 (a) highlights two points taken at a similar magnetic field setting on separate days. Confidence limits from the fits to the resonance scans determine the vertical errors, which are smaller than the thickness of the lines shown in the inset. We fit the data for each hyperfine interval to a quadratic form, $\alpha_k B^2 - \delta W_k$, using a $\chi^2$ minimization. Since the second order Zeeman shift coefficients, $\alpha_k$, are determined by the three values of $\delta W_k$, we fit all three quadratic forms simultaneously. However, we may still determine a $\chi^2$ statistic for each data set. Since the $\alpha_k$ are only weakly dependent on the $\delta W_k$, the minimization procedure is equivalent to three independent single parameter fits with the $\alpha_k$ fixed to values consistent with the fitted values of $\delta W_k$.

In Table 5.2 we give the $\delta W_k$ along with the reduced $\chi^2$ for each fit. Errors reported here are again the 68% confidence limits extracted from the fits. For the field calibration and calculation of the $\alpha_k$ we have used values of $g_J$ and $g_I$ reported in [102] and [103] respectively. It is worth noting that the quadratic form used for the fitting is only an approximation. As the field strength increases, higher order terms become important, which can shift the zero field value extracted from a quadratic fit. The smaller the hyperfine splitting, the larger the effect. It is for this reason we have restricted our field values to $\sim 1$ G. Even within this restricted field range higher order terms could still have a small effect which is discussed in further detail in Section 5.2.4. The $\chi^2$ contributions of each data point are shown in Fig. 5.3. From these $\chi^2$ contribution plots it can be seen that not one data point contributes more than six to the $\chi^2$. This can be interpreted as every point being within three confidence intervals of the fitted curve.

5.2 Error sources

In this section we discuss possible error sources that can perturb the hyperfine intervals measurements. In brief, the largest contribution to the errors comes from the slow magnetic field drift between the calibration measurements of the magnetic field and
Figure 5.3: Plots of measured hyperfine intervals of 5D\textsubscript{3/2} manifold as a function of magnetic field for (a) $|F'' = 0, m_{F''} = 0\rangle \leftrightarrow |F'' = 1, m_{F''} = 0\rangle$ (b) $|F'' = 1, m_{F''} = 0\rangle \leftrightarrow |F'' = 2, m_{F''} = 0\rangle$ and (c) $|F'' = 2, m_{F''} = 0\rangle \leftrightarrow |F'' = 3, m_{F''} = 0\rangle$. The crosses represent the measured data and the solid black lines are fits to the data (See text for further details). To the right of each splitting plot is the contribution each point makes to the $\chi^2$. For each interval (×) and (+) markers represent data sets taken on different days, while datasets for different intervals were also taken on different days.
measurements of the hyperfine intervals. This leads to an error in the exact magnetic field at which the state is measured. The magnetic field drift is accounted for by the use of a horizontal error bar placed on the measurement points and the method to obtain a value for it is explained in Section 5.2.1. Additional systematic errors come from shifts due to coupling with stray ac and dc electro-magnetic fields. Shifts caused by electric fields are discussed in Section 5.2.2 and ac magnetic fields in Section 5.2.3. Finally, the effect of higher order terms unaccounted for in the fitting model is discussed in Section 5.2.4.

5.2.1 Magnetic field noise

The magnetic field noise can originate from two mechanisms. The first mechanism is noise from changes to the externally applied magnetic field, which is discussed in Section 5.2.1.1. The second is movement of the ion in a magnetic field gradient, which is described in Section 5.2.1.2.

5.2.1.1 Magnetic field drift

![Magnetic field drift measured over six hours. The magnetic field drift was measured on between the $|F'' = 2, m_{F''} = 0\rangle$ to the $|F'' = 3, m_{F''} = 1\rangle$ states.](image)

The time it takes to measure a single splitting is five minutes and at each magnetic field point we perform three separate splitting measurements. It is therefore necessary to consider the effects of magnetic field drifts which give rise to additional errors in the field calibration. We have monitored the variation of the magnetic field over the course
of a day by measuring the first order Zeeman shift of the $\Delta m_{F''} = +1$ transition. The magnetic field drift from these measurements is shown in Fig. 5.4. From this data we were able to extract a Root Mean Squared (RMS) magnetic field drift of 300 µG per five minutes.

For the analysis of the measurements presented in this chapter we are interested in what error this drift causes in the $B$-field we assign to a particular measurement of the $\Delta m_{F''} = 0$ transition. Each data point in Fig. 5.3 is derived from three separate splitting measurements. First we measure the $\Delta m_{F''} = +1$ and the $\Delta m_{F''} = -1$ transitions to determine the magnetic field. Then we measure the $\Delta m_{F''} = 0$ transition. The drift of the magnetic field during the time of these measurements will cause both an error in the estimation of the magnetic field and an error because the last measurement is at a later time than the magnetic field measurement. Depending on whether the drifts are uncorrelated or correlated we have to treat these errors as independent or dependent respectively. We will analyze both cases and show that they in fact lead to the same overall error.

First we look at the independent case. The magnetic field $B_+$ at the time of the $\Delta m_{F''} = +1$ measurement can be considered to be error free

$$B_+ = B_0.$$  \hspace{1cm} (5.1)

If $\sigma$ is the one standard deviation drift for the time it takes to make one measurement then the field at the time of $\Delta m_{F''} = -1$ transition becomes

$$B_- = B_0 \pm \sigma$$  \hspace{1cm} (5.2)

and adding the independent errors in quadrature the field at the time of the measurement of the $\Delta m_{F''} = 0$ transition is

$$B_a = B_0 \pm \sqrt{2}\sigma.$$  \hspace{1cm} (5.3)

The measured magnetic field, $B_M$ is obtained by taking half the difference of the first two splitting measurements. Since we considered the first magnetic field to be error free we get

$$B_M = B_0 \pm \frac{1}{2}\sigma.$$  \hspace{1cm} (5.4)
Adding this error in the estimation of $B_0$ to the error in the field $B_a$ we arrive at the total error in the magnetic field which is

$$\Delta B_T = \pm \left(\sqrt{\frac{1}{4} + 2}\right) \sigma = \pm \frac{3}{2} \sigma,$$  \hfill (5.5)

Next we look at the case where the errors are treated as being dependent. $B_+$ and $B_-$ are still given by Eqs. (5.1) and (5.2) respectively and the error in the measured magnetic field $B_M$ is given by Eq. (5.4). Since dependent errors add $B_a$ becomes

$$B_a = B_0 \pm 2\sigma. \hfill (5.6)$$

When evaluating the combined error in the final estimate $B_T$ the direction of the drifts becomes important. Since the drift direction is assumed to be the same for all three measurements the error from the magnetic field estimation compensates the error in $B_a$. We get

$$\Delta B_T = \pm \left(2 - \frac{1}{2}\right) \sigma = \pm \frac{3}{2} \sigma,$$  \hfill (5.7)

which is the same as for the independent case.

Taking $\sigma$ from the data presented above, we get an error of $450 \mu G$, which dominates the $50 \mu G$ error extracted from the fit of $\Delta m_{F'=1}$ transitions. The slow drift does not have any effect in the time scale of the rf $\pi$-pulse as slow drift happens at a much longer time scale compared to the rf $\pi$-pulse used in performing the interval measurement. An independent method of measuring the magnetic field fluctuations such as using a hall probe [104] was not used because the device could not be placed at the same position as the ion and as such would be an unreliable measurement.

### 5.2.1.2 Magnetic field gradient

Along with measuring the magnetic field drift we also explored the magnetic field gradient within the trap. We could only successfully measure this in the ion trap’s axial direction as the confinement in this axis is weaker than in the radial axes, meaning the ion can be more easily displaced in this axis by the application of a dc potential. An experiment was undertaken to move the ion in the axial direction and monitor any change in the frequency of the $\Delta m_{F'=1}$ transition. The ion’s position can be accurately measured using the imaging system’s camera. The position of the ion in
pixels on the camera is accurately known through centroid fitting of the image. The relative pixel position of the ion can be related to the relative position of the ion through a calibration factor. The magnetic field in the axial direction as a function of position from the trap center is shown in Fig. 5.5. From this data we estimate a field gradient of 2.5 mG/µm, which seems significantly large given our coil arrangement. The reason for it being so large remains unclear and we note that it is a factor of six larger than the gradient measured in a similar linear Paul trap [105].

Having such a large magnetic field gradient in the trap is a source for concern as a small displacement of the ion could lead to a large change in the magnetic field. As we have already measured the magnetic field drift in Section 5.2.1.1, we know that it is not changing by more than 450 µG throughout one measurement, which puts a bound of 200 nm on how far the ion can move in the axial direction of the trap during the same time frame.

5.2.2 Stark shifts

The effect of off-resonant stray ac and dc electric fields can lead to ac and dc-Stark shifts respectively, through coupling to other manifolds via electric dipole transitions. Leakage light from the lasers used in the experiment can cause ac-Stark shifts and the magnitude of this shift is discussed in Section 5.2.2.1. The electric field used for trapping the ion also causes an ac-Stark shift. If the ion is displaced from the trap’s
rf null the ion will experience an oscillating electric field from the trap. In the best case, the displacement will be the ion’s secular motion taking it in and out of the rf null. In the worst case, an uncompensated dc field forces the ion away from the trap’s rf null. Since the frequency of oscillation of the electric field is much smaller than the optical transition it couples to, we can treat the effect as a dc-Stark shift due to the time averaged electric field, which is discussed in Section 5.2.2.2.

5.2.2.1 Stark shift caused by lasers

Stray laser light entering the chamber can be a major source of systematic errors as a tiny amount of near resonant light leads to a significant shift in the hyperfine energy levels. We use AOMs for switching the beams on and off. These devices have a finite attenuation factor that has been measured to be greater than 45 dB. Even at this level, enough leakage light can enter the chamber to be an issue for the level of accuracy we are aiming to achieve. The only light shifts we are concerned with are the shifts that affect the 5D_{3/2} manifold. This means the only lasers that will be problematic are the 650 nm lasers, as these will be sufficiently close in detuning to cause a significant light shift. For the 650 nm shelving beam, it was found that 1 µW of leakage light at a detuning of 500 GHz was enough to shift the resonances by ~ 60 Hz, consistent with ac-Stark shift calculations. The estimate given in Table 5.1 is based on this measurement and the addition of a second AOM lowering the leakage power by at least a further 45 dB.

For the repumping beams an undetectable amount of leakage light could be a problem as the beams are at a small detuning. To ensure the effect of leakage light from the 650 nm repump is minimized two AOMs are used to provide double isolation. When both AOMs are off the amount of light at the chamber is undetectable with an optical power meter that is sensitive to 10 nW and also can not be seen by the human eye\(^1\). We can further bound the Stark shift from the knowledge of the 650 nm beam parameters and the attenuation factor of two AOMs. From Eqs. (2.41) and (2.42), taking the beam parameters of \(w_0 = 30 \mu \text{m}, \) \(P = 0.1 \text{pW} \) of estimated power through two AOMs and a detuning \(\Delta = 10 \text{ MHz} \) gives a Stark shift of 0.03 Hz. We conservatively increase this error to 0.5 Hz to cover any errors in the parameters. Note the Clebsch Gordon

\(^{1}\)The human eye can easily detect 1 pW of 650 nm light scattering off a white card in a darkened room.
coefficients have not been factored into the calculation of the Rabi rate. The effect of these will only reduce the actual light shift.

5.2.2.2 Stark shift caused by trap

The systematic effect of micromotion causes a second order Stark shift caused by a time averaged electric field. This frequency shift to the hyperfine level of interest due to the electric field from the trap is given by [70]

\[ \Delta E_s \approx \sigma_s \langle E_i^2 \rangle, \]

(5.8)

where \( \sigma_s \) is the dc-polarizability for the hyperfine level of interest and \( \langle E_i^2 \rangle \) is the time averaged square of the electric field at the ion’s position. The Stark shift constant can be found from perturbation theory detailed in [106], where in this case the Stark shift is primarily due to the dipole coupling between the 5D and 6P states. The electric field the ion experiences is given by

\[ \langle E_i^2 \rangle \approx \frac{m \omega_{rf}^2 k_b T_i a_i^2 + 2q_i^2}{2a_i + q_i^2} + 8 \left( \frac{q_i E_{dc} \cdot \hat{u}_i}{2a_i + q_i^2} \right)^2 \]

(5.9)

where \( \omega_{rf} \) is the trap drive frequency, \( m \) is the mass of the ion, \( T \) is the temperature of the ion, \( k_b \) is Boltzmann’s constant and \( E_{dc} \) is the stray electric field displacing the ion from the \( rf \) null. Definitions for the trap parameters \( a_i \) and \( q_i \) can be found in [70]. Here the first term is due to the unavoidable micromotion which scales with the ion temperature and the second term is due to excess micromotion. The first term is negligible if the ion is cooled to the Doppler limit. We next describe the method used to ensure excess micromotion is kept to a minimum and then we estimate the systematic error due to uncompensated excess micromotion.

To ensure excess micromotion is kept to a minimum we perform the micromotion compensation procedure illustrated in Fig. 5.6 and described by these steps. First the drive \( rf \) potential is set to its maximum value; the ion’s relative position at the upper \( rf \) value is found by using a centeroid fitting to the image of the ion on the CCD camera; the \( rf \) is then lowered to a value that approximately halves the radial trapping frequencies. If the ion moves position on the camera then excess micromotion is present. We compensate it by adjusting the dc bias potentials of the trap to return the ion to its initial position when the \( rf \) potential is set to its maximum value. This process is
**Figure 5.6:** Micromotion compensation procedure. (a) Ion in initial position when rf drive potential set max value. (b) rf potential lowered, ion displaced due to stray electric field. (c) Trap compensation potentials adjusted to return ion to initial position. (d) rf potential set to max value to see if ion moves.

repeated until no movement of the ion is seen when changing the rf drive potential from high to low. To compensate the micromotion along the trap axis that is perpendicular to the imaging systems focal plane, which we shall call the y-axis, the same procedure as described above is used, but the ion’s fluorescence is used as the feedback signal. The fluorescence feedback signal works on the principal that less photons are scattered if the ion is displaced from the center of the waist of the Doppler cooling beams. This method only works if the Doppler cooling beams position has been adjusted to give maximum fluorescence when the rf drive potential has been set to its maximum value. From the 493 nm beam waist and the imaging systems sensitivity to a change in the scattering rate, we get a displacement sensitivity of 1 µm for this axis.

For a particular trapping frequency $\omega_i$, the ion is displaced in that direction by a uncompensated dc electric field by the amount

$$u_i = \frac{eE_{dc} \cdot \hat{u}_i}{m \omega_i^2}.$$  

(5.10)

From the procedure outlined above we know that the ion does not move by more than a certain amount when going from the high to the low trapping potential. This in turn allows us to bound the size of the stray field. The largest bound for the difference in displacement is in the y-axis where we are only sensitive to 1 µm. This corresponds to a stray field of $E_{dc} \approx 15 \text{ V/m}$ and an ion displacement of 300 nm at the
high trapping potential. Using Eqs. (5.8) and (5.9) we calculate a Stark shift of $\sim 5$ mHz. Additionally, we adjusted the dc compensation potentials to induce micromotion and saw no measurable shift in the resonance. Therefore we can safely rule out dc-Stark shifts due to micromotion.

5.2.3 ac-Zeeman shifts

Transitions between Zeeman sublevels of either the same or different hyperfine states can be induced via magnetic dipole transitions, so off-resonant oscillating magnetic fields can cause ac-Zeeman shifts. There are three sources of oscillating magnetic fields in our experiment that need to be considered. The first comes from the rf-field used to drive the hyperfine transitions. The second comes from ion motion in the magnetic field gradient measured in Section 5.2.1.2. The third source comes from rf currents in the trap electrodes that generate an oscillating magnetic field.

The rf field used to measure the hyperfine intervals causes shifts through the other magnetic field components coupling to different states. The important couplings to consider are between hyperfine levels, where $\omega_{rf} \approx \delta W_k$. Coupling between Zeeman states of the same hyperfine level can be neglected because the detuning is large, $\omega_{rf} \gg \Delta_z$, where $\Delta_z$ is the Zeeman splitting.

For ion motion in a magnetic field gradient the shift can be viewed as an effective oscillating magnetic field for both the ion’s micromotion and its secular motion. The trap drive frequency is $\omega_{rf\, \text{trap}} = 5.3$ MHz and we operate at trapping frequencies $\omega_i < 1.2$ MHz, therefore the effect of coupling between hyperfine levels is negligible because $(\omega_{rf\, \text{trap}}, \omega_i) \ll \delta W_k$. The effect of coupling between Zeeman states of the same hyperfine level will be significant because $(\omega_{rf\, \text{trap}}, \omega_i) \sim \Delta_z$.

The field due to currents in the trap electrodes also oscillates at the trap drive frequency, so it couples predominantly to Zeeman states of the same hyperfine level. However since the amplitude of the B-field is much larger than for the one due to the ion motion we also have to consider the effect of coupling between different hyperfine levels. We only treat this shift in Section 6.2.1.1. At the time of the 5D$_{3/2}$ measurement we were not aware of this mechanism as its effect is too small to be observed in the 5D$_{3/2}$ manifold. In the 5D$_{5/2}$ manifold however, this effect will become significant and after we have analyzed it for the 5D$_{5/2}$ manifold we will show it is smaller than the majority of errors for the 5D$_{3/2}$ manifold.
5.2.3.1 Off resonant rf coupling due to antenna

\[
\begin{array}{cccccc}
\hat{m}_F'' & -2 & -1 & 0 & 1 & 2 \\
F'' & = & 2 & \approx & 2\delta W_0 & \\
\delta W_0 & & & & \\
F'' & = & 1 & \approx & 2\delta W_0 & \\
F'' & = & 0 & & & \\
\end{array}
\]

Figure 5.7: Schematic showing off-resonant coupling due to the rf driving field coupling to other levels. The levels that can be coupled together have been numbered so that they can be referred to in the text.

The rf field used to drive the hyperfine state transition can also couple to other levels causing the measured levels to be ac-Zeeman shifted. The levels involved are shown in Fig. 5.7. The following simple geometrical argument can be used to prove that shifts caused by off-resonant rf coupling will not be significant when measuring between the \(m_{F''} = 0\) states. The rf driving field used to measure the hyperfine interval of interest is represented by (1) in Fig. 5.7, this can not ac-Zeeman shift the splitting as it is on resonance. The next two transitions (2) and (3) connect the \(m_{F''} = \pm 1\) states, which are detuned from the rf field by the Zeeman energy. The detuning for the (2) and (3) transitions is equal and opposite for both states leading to a net shift of zero assuming equal powers for \(\sigma^+\) and \(\sigma^-\) polarization components. The next three transitions (4), (5) and (6), can cause a net shift to the upper level of the hyperfine interval of interest but as the hyperfine intervals follow almost an integer interval spacing the smallest detuning will be \(\approx \delta W_0\). The Rabi rate when driving the \(\Delta m_{F''} = 0\) transitions is \(\Omega \approx 2\pi \times 100\) Hz, which is much smaller than \(\delta W_k\) and leads to a negligible shift.

We now analyze the shift for the field calibration measurements. The shift for the \(\Delta m_{F''} = \pm 1\) will be different to the \(\Delta m_{F''} = 0\) because the Rabi rate for these
transitions is $\Omega = 2\pi \times 1$ kHz, which is significantly larger than for the $\Delta m_{F''} = 0$ transition. Also the coupling configurations for the field calibration measurements leads to a net shift. The effect of the off resonant rf coupling can be quantified from standard ac-Zeeman shift calculations, which are detailed in appendix D. In the worst case scenario this can lead to a a shift of $\approx 1$ Hz, which in turn corresponds to a -2 $\mu$G offset in the magnetic field calibration measurement. Even though this shift is much smaller than the error in the magnetic field it has the same effect on every point leading to a systematic under estimation in the magnetic field. The offset can alter the quadratic form in Fig. 5.3 (a) causing a shift in the zero-field point. This effect is largest for the $F'' = 0 \leftrightarrow F'' = 1$ transition, as it has the greatest curvature of all the splittings and gives a total shift of $\lesssim 0.1$ Hz.

5.2.3.2 ac-Zeeman shift due to ion motion in a magnetic field gradient

Ion motion can cause an ac-Zeeman shift because the ion oscillates in the magnetic field gradient measured in Section 5.2.1.2. Ion motion has two components, micromotion and secular motion and both can be viewed as external rf fields being applied at the rf trap drive frequency and the secular trap frequencies respectively. As all these frequencies are much smaller than the separation between the hyperfine levels and the amplitude is small, only coupling between the $m_{F''}$ states of the same $F''$ level need be considered as shown in Fig. 5.8. The shift to the $m_{F''} = 0$ can be neglected due to symmetry of $m_{F''}$ state coupling. This means only the $m_{F''} = \pm 1$ levels will be shifted. Therefore only the field calibration measurements will be effected. In this section we first calculate the ac-Zeeman shift due to micromotion and then move onto the secular motion.

For ion motion at the micromotion frequency the amplitude of the field can be calculated from the micromotion amplitude [70]

$$\Delta x_{i}^{\text{micro}} = \frac{1}{2} u_{0i} q_{i},$$  \hspace{1cm} (5.11)

where $u_{0i}$ is the ions displacement from the rf null. Substituting the ion’s displacement found in Section 5.2.2.2 into Eq. (5.11) gives a amplitude of approximately 35 nm, which corresponds to a magnetic field amplitude of $B_{rf} \approx 100 \mu$G from motion in the magnetic field gradient given in Section 5.2.1.2. This magnetic field oscillates at the trap drive frequency of $\omega_{tr} = 2\pi \times 5.3$ MHz and using standard ac-Zeeman shift calculations detailed in appendix D, one obtains a shifts of less than 2 mHz over the whole range.
of magnetic field points explored in the measurement. This in turn corresponds to a magnetic field calibration error of less than 10 nG, which has a negligible effect on the splitting values.

For motion at the secular frequencies, the amplitude of the field is given by the ion’s wavefunction spread defined as

\[ \sigma_i = \sqrt{\frac{k_B T}{m \omega_i^2}}, \]

(5.12)

where \( \omega_i^2 \) is the trapping frequency for the axis of interest, \( k_B \) is Boltzmann’s constant, \( m \) is the mass of the ion and \( T \) is the temperature of the ion. The ion’s temperature can be found from the Doppler cooling limit \[107\] expressed as

\[ T_D = \frac{\hbar \Gamma}{2k_B}, \]

(5.13)

where \( \Gamma \) is linewidth of the Doppler cooling transition. The axial direction will have the largest spread due to the fact the trapping frequency is the lowest for this axis. Using the Doppler cooling limit as the ion’s temperature, an axial trapping frequency of \( \omega_z = 2\pi \times 380 \) kHz and Eq. (5.12), we find a magnetic field amplitude of \( B_{rf} = 125 \) µG.

Figure 5.8: Schematic showing off-resonant rf relevant coupling between Zeeman states. The relevant coupling for the \( F'' = 3 \) level is not shown in the diagram as it is exactly the same as the \( F'' = 2 \).
The effect of the secular motion is larger than the micromotion because the detuning is only on the order of 10 kHz. The size of the shift will also be highly dependent on the magnetic field set point, as the detuning is dependent on the Zeeman splitting. Using standard ac-Zeeman shift calculations detailed in appendix D and the values defined above, one obtains a shifts of less than 0.5 Hz. However, we note that larger shifts occur when the Zeeman splitting and $\omega_z$ are resonant. This occurs at 0.65 G for the above parameters. In this case, we expect a maximum change to the $B$-field of $\pm1 \mu$G at magnetic filed values of 0.6 G and 0.7 G respectively. These resonances were observed in preliminary measurements, through effects to the rf hyperfine transitions. This effect in preliminary measurements would go away after a few minutes due to magnetic field drift. With knowledge of this problem, we specifically set the trappings frequencies to avoid resonance points. Full modeling of this shift leads to a shift of less than a 0.01 Hz in the $\delta W_0$ splitting.

Similar calculations have been performed for the radial trap frequencies. Because they are $\approx 1 \text{ MHz}$ the minimum detuning of the effective magnetic field from the Zeeman transitions is on the order of 500 kHz, so the overall shift is negligible.

5.2.4 Higher order terms of $\delta W_0$

The zero-field splittings for the $5D_{3/2}$ manifold are well described by treating the shift due to the magnetic field as a small perturbation to the energy of the $F$ states. In this weak field limit where the Zeeman energy is much smaller than the hyperfine intervals energy, the zero-field splitting for a particular hyperfine interval is independent of the exact value of other intervals and so each can be fitted to a quadratic independently. However it is still worth considering the effect of higher order terms neglected in this limit. To get an idea of the size this effect has on the zero field splitting values the experimental data is fitted to the full hyperfine plus Zeeman Hamiltonian. Over the range of field values we have used, we estimate that this effect could be as large as $\sim 0.5 \text{ Hz}$ for $\delta W_0$. For field values in the range $1 \sim 2 \text{ G}$ this systematic error would be comparable to the uncertainty in our current measurement.
Table 5.1: Estimates of the systematic errors.

<table>
<thead>
<tr>
<th>Source</th>
<th>Error estimation</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>ac-Zeeman shifts</td>
<td>~ 0.1</td>
<td>Hz</td>
</tr>
<tr>
<td>Off-resonant rf coupling</td>
<td>~ 0.1</td>
<td>Hz</td>
</tr>
<tr>
<td>Secular motion</td>
<td>≲ 0.01</td>
<td>Hz</td>
</tr>
<tr>
<td>Stark shifts</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Micromotion</td>
<td>~ 5</td>
<td>mHz</td>
</tr>
<tr>
<td>Stray Raman light</td>
<td>~ 60</td>
<td>mHz</td>
</tr>
<tr>
<td>Stray repump light</td>
<td>≲ 0.5</td>
<td>Hz</td>
</tr>
<tr>
<td>Higher order terms (δW₀)</td>
<td>~ 0.5</td>
<td>Hz</td>
</tr>
</tbody>
</table>

Table 5.2: Measured hyperfine intervals, δWₖ, for the 5D₃/2 manifold of ¹³⁷Ba⁺.

<table>
<thead>
<tr>
<th>Transition</th>
<th>−δW (Hz)</th>
<th>Reduced χ²</th>
</tr>
</thead>
<tbody>
<tr>
<td>F″=0 → F″=1</td>
<td>145 193 549.3 (2.8)</td>
<td>1.01</td>
</tr>
<tr>
<td>F″=1 → F″=2</td>
<td>334 921 347.13 (89)</td>
<td>1.60</td>
</tr>
<tr>
<td>F″=2 → F″=3</td>
<td>613 730 628.08 (22)</td>
<td>0.47</td>
</tr>
</tbody>
</table>

*Intervals affect by shift discovered while thesis under examination see appendix E for more details.

5.3 Results

The hyperfine coupling constants inferred from the measured values of δWₖ are given in Table 5.3. The systematic errors in Table 5.1 are listed as the largest for any of the hyperfine intervals. A separate analysis for each individual interval yields a systematic error of 0.5 Hz for each of them, which we have added to the errors given in Table 5.2. The correction terms were calculated from the expressions given in Eqs. (2.12) and (2.13), using the matrix elements ⟨5D₃/2||T₁||5D₅/2⟩ and ⟨5D₃/2||T₂||5D₅/2⟩ given in Table 2.1. In our paper [51] we used incorrect matrix elements in the calculations of the error terms, due to an error in the code used in the calculations of the off diagonal elements, for which we apologize. The magnetic moment 0.937365(20)µN and quadrupole moments 0.246(1)b were taken from [37] and [36] respectively.

The final values obtained for hyperfine constants (A) and (B) are within three
Table 5.3: 5D$_{3/2}$ hyperfine coupling constants.

<table>
<thead>
<tr>
<th></th>
<th>A (Hz)</th>
<th>B (Hz)</th>
<th>C (Hz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uncorr.</td>
<td>189730524.90(32)</td>
<td>44538793.7(10)</td>
<td>32.465(44)</td>
</tr>
<tr>
<td>$\eta$ corr.</td>
<td>805(16)</td>
<td>$-1610(32)$</td>
<td></td>
</tr>
<tr>
<td>$\zeta$ corr.</td>
<td>164.2(42)</td>
<td>411(10)</td>
<td>$-2.933(75)$</td>
</tr>
<tr>
<td>Corr.</td>
<td>189731494(17)</td>
<td>44537594(34)</td>
<td>29.533(86)</td>
</tr>
</tbody>
</table>

standard deviations of the results reported in [108] with a 30-fold reduction in the uncertainty. From the hyperfine (C) constant given in Table 5.3 the nuclear octupole moment can be determined, which is given in Eq. (5.14).

5.4 Summary

In conclusion we have performed high precision measurements of the hyperfine structure in $^{137}$Ba$^+$. Our measurements have greatly reduced the uncertainty of the currently available hyperfine structure constants and have provided an estimate of the nuclear octupole moment, $\Omega$, accurate to 1%. Approximately 10% of the estimated value for $\Omega$ is given by the correction factor, $\zeta$. In Section 7.1 we compare the octupole moments from hyperfine interval measurements in this and the 5D$_{5/2}$ manifold.

$$\Omega^{^{137}\text{Ba}^+ D_{3/2}} = 0.05057(54) \ (\mu_N \times b),$$  \ (5.14)
Chapter 6

Hyperfine Interval Measurement of D_{5/2} Manifold

This chapter covers the hyperfine interval measurements of the 5D_{5/2} manifold. Along with the measurement in Chapter 5, it completes the nuclear octupole moment measurement of $^{137}$Ba$. The experiment performed in this chapter relies on the experimental techniques described in Chapter 4. In this chapter we first describe the procedure we use to measure the hyperfine intervals. Then we cover the extra sources of errors that are specific to the hyperfine interval measurements in the 5D_{5/2} manifold. Then we present the results, in Section 6.3. The octupole moment obtained from measurements in this manifold and a summary of the experiment is given in Section 6.5. The work in this chapter was reported in [40].

6.1 Method for measuring hyperfine intervals of 5D_{5/2} manifold

The method used for measuring the hyperfine intervals in the 5D_{5/2} manifold is similar to one used in the hyperfine interval measurement of the 5D_{3/2} manifold which is covered in Section 5.1. Briefly, the ion is first Doppler cooled (Section 4.1) and optically pumped (Section 4.2) to the $|F = 2, m_F = 2\rangle$ ground state. The ion is then shelved to the $|F'' = 2, m_{F''} = 0\rangle$ state in the 5D_{5/2} manifold with $\approx 92\%$ efficiency using a two color Raman transition at 455 nm and 615 nm (Section 4.3.2). An rf antenna (Section 3.7) is used to drive the hyperfine transition of interest. To determine, whether
Figure 6.1: Schematic showing hyperfine transition measured by rf spectroscopy in the 5D₅/₂ manifold.

The hyperfine transition occurred we use a second Raman pulse to transfer the ion from the \(|F'' = 2, m_{F''} = 0\rangle\) state back to the \(|F = 2, m_F = 2\rangle\) ground state. The ion fluorescence from the Doppler cooling lasers is then counted using a single photon counting module. Thus, the fluorescence counts provide a probability measurement of the rf transition taking place: the ion being dark if the rf transition took place and bright otherwise. This process is repeated 200 times to obtain enough statistical data for one frequency point of an rf resonance scan and is repeated for different rf driving frequencies.

The rf resonance scans are taken by stepping the rf signal generator in 5 Hz steps. The rf signal power is adjusted until the \(\pi\)-pulse time for the resonant transition is approximately 5 ms. The scans are fitted via a \(\chi^2\) minimization [101] to the usual Rabi flopping function Eq. (2.35) with additional offset and amplitude parameters to account for imperfect shelving. An example of a fitted scan is shown in Fig. 6.2. Resonance scans are repeated over a range of magnetic fields to map out the field dependence of the measured hyperfine intervals. The magnetic field range covered varies depending on the hyperfine interval being addressed. The transition \(|F'' = 1, m_{F''} = 0\rangle\) to \(|F'' = 2, m_{F''} = 0\rangle\) has a quadratic shape so it is measured for both positive and negative magnetic fields in the range of -1 to 1 G. For the \(|F'' = 2, m_{F''} = 0\rangle\) to \(|-\rangle\) transition the magnetic field range covers 0.5 to 2 G to include both magnetic field independent points, which are defined in Section 2.2.2. For the \(|F'' = 2, m_{F''} = 0\rangle\) to \(|+\rangle\) transition the magnetic field range covers 0.35 to 1 G to include the magnetic field independent point. The magnetic field for each rf resonance scan is measured in the 5D₃/₂ manifold using the \(|F'' = 2, m_{F''} = 0\rangle \leftrightarrow |F'' = 3, m_{F''} = \pm 1\rangle\) transitions as described in Section 5.1.
Figure 6.2: Plot of rf resonance scan of $|F'' = 2, m_{F''} = 0\rangle$ to $| - \rangle$ transition taken at a magnetic field of 1.684 G.

The full magnetic field dependence of the three hyperfine intervals and their contributions to the reduced $\chi^2$ is plotted in Fig. 6.3. For each hyperfine interval the rf transition frequency is measured at ten magnetic field values. The magnetic field points have roughly equal spacing with a range of values depending on which hyperfine interval has been measured. Two data sets for each hyperfine interval were taken on separate days to minimize the potential for systematic errors due to the time at which the experiment was performed. The insets in the plots of Fig. 6.3 are used to highlight the size of the error bars. The vertical error bars represents 68% confidence intervals from the resonance scan fits. The horizontal error bars include the magnetic field drift plus the 68% confidence intervals from the magnetic field measurement scans.

6.2 Error sources

Error sources encountered in the hyperfine interval measurement of 5D$_{3/2}$ manifold described in Section 5.2, do not have a large impact here as they have an effect of less than 0.5 Hz, which is smaller than the statistical error on the hyperfine intervals. There is an additional error in this measurement compared to the 5D$_{3/2}$ measurement due to the rf drive causing an ac-Zeeman shift between the $F'' = 3$ to $F'' = 4$ levels, which is described in Section 6.2.1. The magnetic field drift in this experiment could be different compared to the experiment performed in the 5D$_{3/2}$ manifold, it is therefore re-measured as described in Section 6.2.2.
Figure 6.3: Plots of measured hyperfine intervals of 5D_{5/2} manifold as a function of magnetic field for (a) $\delta W_1 = |2 \leftrightarrow 1|$, (b) $\delta W'_2 = |2 \leftrightarrow -|$ and (c) $\delta W'_3 = |2 \leftrightarrow +|$. The crosses represent the measured data and the solid black lines are fits to the data (See text for further details). To the right of each splitting plot is the contribution each point makes to the $\chi^2$. For each interval ($\times$) and ($+$) markers represent data sets taken on different days, while datasets for different intervals were also taken on different days.
6.2.1 ac-Zeeman shift due to the ion trap

The rf field driving the trap electrodes induces currents that generate an rf magnetic field in the trap. The rf drive frequency is sufficiently close to transition frequencies between the $m_{F''}$ states of the $F'' = 3$ and $F'' = 4$ levels that a significant ac-Zeeman shift results, an effect which has previously been observed with trapped ions in [50]. In the ideal geometry the null in the electric field would occur at the center of the trap and magnetic fields from the induced currents would cancel. However, fabrication imperfections or design asymmetries result in a non-zero magnetic field at the ion’s position. To estimate the size of this field, we assume the rf electrodes carry equal currents. This gives rise to a zero magnetic field at the midpoint between the two electrodes. For a small displacement, $\delta$, from the midpoint position between the two rf electrodes, the magnitude of the $B$-field is then given by

$$B = \frac{8\mu_0 I}{2\pi a^2} \delta = \frac{8\mu_0 V_0 \Omega C}{2\pi a^2} \delta$$  \hspace{1cm} (6.1)$$

where $a$ is the separation of the electrodes, $V_0$ and $\Omega$ are the amplitude and frequency of the rf trapping potential, respectively, and $C$ is the electrode capacitance. In our system we have $a = 1.77$ mm, $V_0 = 120$ V, and $\Omega = 2\pi \times 10.6$ MHz. For our trap dimensions, the electrode capacitance is estimated to be $C \approx 10$ pF and, using $\delta = 25\mu$m as a typical dimensional tolerance, we estimate a $B$-field amplitude of approximately 10 mG.

For a given polarization and magnitude of the rf magnetic field, it is straightforward to calculate the ac-Zeeman shifts of each level as a function of the applied static magnetic field. Details of this calculation are given in appendix D. To a good approximation this can be done by neglecting mixing with $F'' = 1$ and $F'' = 2$ levels as done in [29]. For the estimated rf field amplitude of 10 mG, the resulting ac-Zeeman shift can be on the order of 10 Hz and thus must be accounted for.

In order to precisely measure the ac-Zeeman shift at a particular static field it is necessary to measure the hyperfine transitions for a range of rf drive strengths and extrapolate the results to $V_0 = 0$. However, this is only practical at the field independent points where magnetic field drifts do not shift the resonance frequency. Measurements for the field independent point in the $|2\rangle \leftrightarrow |{-}\rangle$ transition at 1.68 G are illustrated in Fig. 6.4. Due to the fact that the ac-Zeeman shift scales with the square of the $B$-field and thus with the square of the drive voltage $V_0$, a quadratic fit centered at an rf drive
voltage $V_0 = 0$ is used. Similar measurements at the other field independent points were inconclusive as the transition frequency did not significantly shift over the range of possible rf drive voltages.

![Figure 6.4: ac-Zeeman shift measured against applied trap potential at an rf drive frequency of 10.6 MHz and a magnetic field of $B = 1.685$ G](image)

Since the ac-Zeeman shifts can only be reliably measured at one magnetic field independent point, we scale the calculated values to coincide with the measured value. In principle this requires an estimation of the polarization components of the rf magnetic field. Although the components cannot be readily determined, according to our calculations the ac-Zeeman shifts due to a $\pi$ polarized field are about two orders of magnitude smaller than for the $\sigma^\pm$ components. For the expected field amplitude of 10 mG, shifts due to the $\pi$ component are less than 0.2 Hz and thus we can safely neglect them. Furthermore, a significant difference in the amplitude of the $\sigma^+$ and $\sigma^-$ components can only be present when there is a phase shift in the currents and thus voltages between the rf rods. This phase shift would result in micromotion that cannot be compensated. However, from the micromotion compensation level achieved in this trap [68], we can safely neglect any phase shift and assume equal contributions from the $\sigma^+$ and $\sigma^-$ components. We can thus scale the calculated ac-Zeeman shifts to the value obtained for the field independent point in the $|2\rangle \leftrightarrow |\rangle$ transition at 1.68 G assuming equal $\sigma^+$ and $\sigma^-$ components. We note that the estimated rf $B$-field amplitude based on this approach is 10.8(6) mG, which is consistent with the crude estimate above. We also note that the inferred ac-Zeeman shifts for the other field independent points are
approximately 2 Hz or less, which is on the order of the error of the rf resonance scan and are thus consistent with the inconclusive measurements at these points. From the ac-Zeeman shift calculations and the inferred rf B-field amplitude, the data points are corrected to remove the ac-Zeeman shift.

Note that one can also calculate at which magnetic field the different polarization components of the rf drive field become resonant with the ac-Zeeman shifted transitions. Initial experiments were performed at a driving frequency of 5.3 MHz where the lowest resonance in magnetic field is at approximately 1.5 G. Around this value of the magnetic field the ac-Zeeman shifts become huge. In order to avoid this problem, the final experiment were performed at a driving frequency of 10.6 MHz. This shifts the lowest resonance to approximately 3.1 G, which is far outside the range of magnetic field used in the experiment to determine the hyperfine intervals.

6.2.1.1 a.c. Zeeman shift in 5D\(^{3/2}\) manifold

The ac-Zeeman shift will also have a similar effect in the 5D\(^{3/2}\) manifold in which we measure the magnetic field. However since the splitting between the \(F\) levels is much larger than the splitting between the \(F = 3\) and \(F = 4\) levels in the 5D\(^{5/2}\) manifold, only coupling between the \(m_F\) states of the same \(F\) number is significant. Using the methods described above we get a shift of approximately 0.05 Hz, which translates into a magnetic field error of \(\approx -0.1 \, \mu\text{G}\). This shift has been added to the experimental data to see how it changes the fitted values. This has a negligible effect on the splittings.

Note that we only became aware of this effect after we had completed the hyperfine intervals measurement of the 5D\(^{3/2}\) manifold. However, we can still estimate the effect of the ac-Zeeman shift for our previous measurement, the corresponding shift to \(\delta W_0^{(3/2)}\) interval is 0.4 Hz and for the other intervals is less than 0.04 Hz. This shift is comparable to errors given in Table 5.1.

6.2.2 Magnetic field drift

The slow magnetic field drift effect on the measurement of the hyperfine intervals for the 5D\(^{5/2}\) manifold is the same effect as it was for the 5D\(^{3/2}\) measurements. For this reason we follow the same method that is used to obtain the magnetic field drift in 5D\(^{3/2}\) measurement, given in Section 5.2.1, for this measurement. The magnetic field drift is measured over a period of two hours to generate sufficient statistical data on the
RMS of this slow drift. The drift data taken is shown in Fig. 6.5. A typical experiment mapping out the field dependence of the hyperfine splittings would take about two hours. We determine the magnetic field to have 220 µG RMS drift over a 10 minute interval. This is a factor of four larger than the error in the fit of the magnetic field resonance scan. This is also a factor of two improvement over the slow magnetic field drift in the 5D\(_{3/2}\) measurement, which can be explained by the fact that the power supply used to drive the magnetic field coils has been changed to a model with better current stability\(^1\).

### 6.3 Results

Using a \(\chi^2\) minimization, we perform a global fit of all the measured hyperfine intervals and magnetic fields to the appropriate eigenvalues of the Zeeman Hamiltonian given in Eq. (A.1). The model has a total of 5 parameters: the three zero field hyperfine splittings, \(\delta W_F\), the Landé g-factor, \(g_J\), and a mixing coefficient, \(\beta_3\), that takes into account the effect of hyperfine mixing between the \(F = 3\) levels of the 5D\(_{5/2}\) and 5D\(_{3/2}\) manifolds. The full account of the effect of mixing is discussed in appendix A.

To a good approximation mixing only affects the energies of the \(|\pm\rangle\) states and can be incorporated into the single factor \(\beta_3\). Due to the sensitivity of the fit to \(g_J\), we

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\(^1\)Hameg HMP4030.
Table 6.1: Measured hyperfine intervals, $\delta W_k$, for the 5D$_{5/2}$ manifold of $^{137}$Ba$^+$. \\

<table>
<thead>
<tr>
<th>Transition</th>
<th>$\delta W$ (Hz)</th>
<th>Stat err.</th>
<th>Syst err.</th>
</tr>
</thead>
<tbody>
<tr>
<td>F=1 $\rightarrow$ F=2</td>
<td>71675902.4</td>
<td>$\pm 4.6$</td>
<td>$\pm 0.9$</td>
</tr>
<tr>
<td>F=2 $\rightarrow$ F=3</td>
<td>62872301.0</td>
<td>$\pm 1.4$</td>
<td>$\mp 2.2$</td>
</tr>
<tr>
<td>F=3 $\rightarrow$ F=4</td>
<td>503510.5</td>
<td>$\pm 2.6$</td>
<td>$\pm 3.2$</td>
</tr>
</tbody>
</table>

included it as a fitting parameter. For the mixing coefficient, we note that since $\beta_3$ depends on exactly the same matrix elements as the correction factors $\eta$ and $\zeta$, the coefficient $\beta_3$ is not independent of $\eta$ and $\zeta$. Moreover, as a function of the static B-field, both the $\text{ac}$-Zeeman shift and the hyperfine mixing give approximately linear shifts in the energies of the $|\pm\rangle$ states. Thus an error in one can be compensated to a degree by a change in the other. For these reasons we leave $\beta_3$ fixed to the theoretical value of $\beta_3 = 1.698(17) \times 10^{-5}$.

Fitting the experimental data gives a reduced $\chi^2 = 1.10$ and the resulting zero field values are given in Table 6.1. The individual contribution to the $\chi^2$ is given in Fig. 6.3, where only one point out of the entire data set is more than three confidence intervals out. The statistical errors given are the 68% confidence intervals extracted from the fits using standard statistical methods. The systematic error accounts for the uncertainty in the theoretical value for the $\beta_3$ parameter and the uncertainty in the measured $\text{ac}$-Zeeman shift. The derived systematic errors in the zero field splittings and $g_J$ correspond to the largest change in the results of the fit when changing the input parameters by their estimated one standard deviation error. For $g_J$ we obtain a value of

$$g_J = 1.20057(5)_{\text{Stat}}(2)_{\text{Syst}},$$

which is within 3$\sigma$ of the value reported in [109]. It also sits within 3$\sigma$ of a correction to their old value [110] but is one order of magnitude more accurate. We note that, if $\beta_3$ is included as a fitting parameter, the fitted value is 13% smaller than the theoretical one and the estimated zero field splittings do not change by more than twice the total error (statistical+systematic) with the most significant change occurring for $\delta W_3$.

The hyperfine coupling constants are determined from the obtained hyperfine intervals and are presented in Table 6.2. The statistical and systematic errors on the hyperfine intervals are added to give the total error on the hyperfine constants. Each
Table 6.2: 5D<sub>5/2</sub> hyperfine coupling constants.

<table>
<thead>
<tr>
<th></th>
<th>A (Hz)</th>
<th>B (Hz)</th>
<th>C (Hz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uncorr.</td>
<td>-12029724.1(9)</td>
<td>59519566.2(43)</td>
<td>-41.73(18)</td>
</tr>
<tr>
<td>η corr.</td>
<td>537(11)</td>
<td>5367(110)</td>
<td>-</td>
</tr>
<tr>
<td>ζ corr.</td>
<td>-46.9(12)</td>
<td>587(15)</td>
<td>29.33(75)</td>
</tr>
<tr>
<td>Corr.</td>
<td>-12029234(11)</td>
<td>59525520(110)</td>
<td>-12.41(77)</td>
</tr>
</tbody>
</table>

constant falls within one standard deviation of previous work [42] but all are three orders of magnitude more accurate. The octupole moment can be derived from the hyperfine C constant and is given in Eq. (6.7).

6.4 Experimental evidence for mixing between the 5D manifolds

The mixing between the 5D manifolds for the \( m_F = 0 \) states is accounted for by the use of a correction parameter \( ζ \) defined in Eq. (2.13). As we measure \(| + \rangle \) and \(| - \rangle \) states that contain mixtures of \( m_F = ±1 \) the mixing manifest itself in a different way. It now comes in as a first order perturbation to the Zeeman interaction and must be taken into account. Its effect is defined as a correction factor \( β_F \) given in Eq. (A.8) to the levels of interest. A full description of this perturbation is given in appendix A. Here we give further evidence of this mixing.

Consider the level structure of \(^{137}\text{Ba}^+ \) shown in Fig. 6.6, when driving the 493 nm and 650 nm transitions the ion only rarely decays into the 5D<sub>5/2</sub> state because this path is dipole forbidden, \( ΔJ = 2 \). The only remaining decay paths are an electric octupole transition<sup>1</sup>, collisions with background gas particles and/or through excitation to the 6P<sub>3/2</sub> level by stray light. If there is mixing between the 5D<sub>3/2</sub> and 5D<sub>5/2</sub> states then this decay should be enhanced because of the small component of the 5D<sub>3/2</sub> state. By comparing the decay rate to the one in \(^{138}\text{Ba}^+ \), where \( I = 0 \), it is possible to get an estimate of the degree of mixing.

<sup>1</sup>An experiment has been proposed to measure the branching ratio between 6P<sub>1/2</sub> and 5D<sub>5/2</sub> states in \(^{138}\text{Ba}^+ \) [111].
Figure 6.6: Schematic of the experiment to find the decay rate through forbidden transition at 686 nm. The idea of the experiment is to fluoresce on the 493 nm and 650 nm transitions and recorded the fluorescence data. For $^{138}\text{Ba}^+$ there is a very small probability of decaying to the 5D$_{5/2}$ manifold through an electric octupole transition, collisions with background gas particles and/or through excitation to the 6P$_{3/2}$ level by stray light. For $^{137}\text{Ba}^+$ this decay process can be enhanced through hyperfine mixing between the two 5D manifolds.

An experiment is performed to measure the decay rate from the 6P$_{1/2}$ level to the 5D$_{5/2}$ level. The level structure and decay channels involved in this experiment are shown in Fig. 6.6. The experiment is performed by monitoring the fluorescence of the ion on the transition between 6S$_{1/2}$ and 6P$_{1/2}$, while repumping on the transition between the 5D$_{3/2}$ and 6P$_{1/2}$. If the ion went dark for a period of 2-120 seconds, it is assumed to have fallen in to the 5D$_{5/2}$ state. This assumption could be tested as long as the ion fell back into the fluorescence cycle via the quadrupole transition between the 5D$_{5/2}$ and 6S$_{1/2}$ states. The lifetime of the low lying 5D$_{5/2}$ is approximately 30 seconds, therefore the ion should appear bright before three minutes passes. If the ion did not come bright after three minutes, it had either left the trap or the fluorescence laser had become unlocked. The fluorescence of the ion is recorded every second over the period of a week to build up enough statistical data. Fluorescence data collected over the period of a day is shown in Fig. 6.7.

We perform the same experiment on both $^{137}\text{Ba}^+$ and $^{138}\text{Ba}^+$ for comparison. As $^{138}\text{Ba}^+$ has no nuclear spin, it has no hyperfine structure and consequently there is no hyperfine interaction mixing between the 5D$_{3/2}$ and 5D$_{5/2}$ states. Therefore, the rate of decay to the 5D$_{5/2}$ state should be significantly smaller for $^{138}\text{Ba}^+$ compared to $^{137}\text{Ba}^+$. Moreover, if the number of dark events was the same for $^{137}\text{Ba}^+$ as it is
Then we could assume the mixing between the $5D_{3/2}$ and $5D_{5/2}$ states in $^{137}\text{Ba}^+$ is negligible or the other decay paths dominate.

Figure 6.7: Plot of $^{137}\text{Ba}^+$ fluorescence counts over a period of 22 hours. The red line shows the raw fluorescence data, where the drops indicate the ion most likely falling into the $5D_{5/2}$ state. The dashed black line represents the discriminated data, where we believe the ion decayed into the $5D_{5/2}$ state. This discriminated data is used to construct the histograms shown in Fig. 6.8. After the 22nd hour the fluorescence data drops to the background level as the ion has been lost from the trap.

The time period between the two dark events is extracted from the fluorescence data. This time period is the decay rate of the ion from the $6P_{1/2}$ level to the $5D_{5/2}$. The time the ion remained dark is also extracted and used to estimate the lifetime of the $5D_{5/2}$, which is compared to the published lifetime of 31.5(5) seconds [1]. This comparison acts as a self consistency check to make sure there is no repumping between the $5D_{5/2}$ and $6P_{3/2}$ levels. The results of this experiment are presented in Eqs. (6.3) to (6.6).

The measured values for the time constants between dark events are

$$\tau(^{137}\text{Ba}^+) = 35(3) \text{ minutes},$$  \hspace{1cm} (6.3)

$$\tau(^{138}\text{Ba}^+) = 120(12) \text{ minutes}.$$ \hspace{1cm} (6.4)

It can be seen that the $\tau(^{137}\text{Ba}^+)$ decay rate is almost four times faster than the $\tau(^{138}\text{Ba}^+)$ decay rate. The difference between the two isotopes adds further weight to
Figure 6.8: Plots of histograms of forbidden transition decay rates and lifetime of 5D_{5/2} manifold. (a) is a histogram of time interval between dark state events for $^{137}$Ba$^+$. (b) is a histogram of the amount of time the ion remained in the dark state for $^{137}$Ba$^+$. (c) is a histogram of time interval between dark state events for $^{138}$Ba$^+$. (d) is a histogram of the amount of time the ion remained in the dark for $^{138}$Ba$^+$. The dashed lines in the plots represents fit of data to an exponential function to extract relevant time constants.
hyperfine interaction causing mixing between the 5D states. The lifetime of the 5D_{5/2} state agrees for both $^{137}$Ba$^+$ and $^{138}$Ba$^+$, we obtain these lifetimes

$$\tau^{(137)5D_{5/2}} = 26(3) \text{ seconds}, \quad (6.5)$$

$$\tau^{(138)5D_{5/2}} = 25(2) \text{ seconds}, \quad (6.6)$$

showing that the dark events are indeed most probably due to the decay to the 5D_{5/2} state. The shorter state lifetimes compared with the published values [1] can be attributed to the fact this it is notoriously difficult to measure metastable state lifetimes due to effects from collisions with background gas particles [112]. From the bright state fluorescence counts the occupation percentage of the 6P_{1/2} state can be calculated. For this experiment the photon count rate for $^{137}$Ba$^+$ is 12(1) counts/ms. Using this photon count rate and the collection efficiency of the imaging system $\eta = 0.25(5)$ % this translates to a 6P_{1/2} state occupation of 5(1) %. From the Einstein A coefficient for the 6P_{1/2} to 5D_{3/2} transition, the 6P_{1/2} state occupation percentage and the fitted $\beta$ factor, the decay time constant from 6P_{1/2} to 5D_{5/2} is 30(6) minutes. The calculated value is within one standard deviation of our measured value. This add further weight to the validity of the mixing between the two 5D manifolds.

### 6.5 Summary

In summary, we have performed simultaneous high precision measurements of the hyperfine splittings of the 5D_{5/2} manifolds of $^{137}$Ba$^+$, that has provided a value for the octupole moment

$$\Omega \left( ^{137}\text{Ba}^+_{D_{5/2}} \right) = 0.0496(37) \left( \mu_N \times \text{b} \right), \quad (6.7)$$

that is accurate to the 10% level. The accuracy of the octupole moment is limited by the accuracy of atomic structure calculations. We have also provided an improved value of $g_J$ for the 5D_{5/2} manifold, which has a 10 fold improvement in accuracy over previous efforts [110]. Our measurements have sufficient precision that hyperfine mixing between the two fine structure levels must be taken into account. In Section 7.1 we compare the octupole moments from hyperfine interval measurements in this and the 5D_{3/2} manifold.
Chapter 7

Conclusion and Future Work

In the closing chapter, we summarize our finding throughout the course of this research in Section 7.1. We also discuss the direction in which the research can be extended in Section 7.2.

7.1 Conclusion

In summary, we have performed simultaneous high precision measurements of the hyperfine splittings of the $5D_{3/2}$ and $5D_{5/2}$ manifolds of $^{137}\text{Ba}^+$, which provide an independent measurement of the nuclear octupole moment and a self consistency check of the associated structure calculation. The aim of the research project was to observe the nuclear octupole moment in $^{137}\text{Ba}^+$, which can be used as the first step towards a PNC measurement. Our research project obtained three different values for the octupole moment, which can be obtained from the hyperfine (C) constants given in Eqs. (2.26), (2.28) and (6.7) and the Eqs. (2.26), (2.29) and (6.7), the results are

\[
\Omega\left(^{137}\text{Ba}_{D_{3/2}}^+\right) = 0.05057(54) \, (\mu_N \times b), \quad (7.1)
\]

\[
\Omega\left(^{137}\text{Ba}_{D_{5/2}}^+\right) = 0.0496(37) \, (\mu_N \times b), \quad (7.2)
\]

\[
\Omega\left(^{137}\text{Ba}^+\right) = 0.05061(56) \, (\mu_N \times b), \quad (7.3)
\]

which are all within one standard deviation of each other. The hyperfine (C) constants have been measured with an experimental accuracy to allow for nearly another order of
magnitude improvement in the accuracy of the nuclear octupole moment. The limiting factor in the accuracy of the octupole moment is the accuracy with which the diagonal matrix elements can be calculated. This measurement constitutes the most accurate value for the nuclear octupole moment in any atom to date.

In our first measurement we measured the hyperfine intervals of the $5D_{3/2}$ manifold to the Hertz level. These measurements have greatly reduced the uncertainty of the currently available hyperfine structure constants and have provided an estimate of the nuclear octupole moment, $\Omega$, accurate to 1%. Approximately 10% of the estimated value for $\Omega$ is given by the theoretical correction factor, $\zeta$. In our second measurement we measured the hyperfine intervals of the $5D_{5/2}$ manifold to an accuracy better than 5 Hz. The hyperfine interval measurements in the $5D_{5/2}$ manifold also spawned values for other important constants like the Lande $g_J$ factor for the $5D_{5/2}$ manifold. This provided an improved value of $g_J$ for the $5D_{5/2}$ manifold, which has a 10 fold improvement in accuracy over [110]. It should be noted that a newer measurement of $g_J$ for this manifold by the authors of [110] reported in [109] has improved $g_J$ value by a factor of five over our value. Our measurements have sufficient precision that the hyperfine mixing between the two fine structure levels must be taken into account. Although we are only sensitive to mixing of the $F = 3$ levels, we note that measurements of the splittings in the $5D_{3/2}$ manifold for transitions between $m_F = \pm 1$ states would also be dependent on the mixing of the $F = 1$ and 2 levels. Thus, in principle, our measurements could be improved such that all three mixing coefficients, $\beta_k$, become measurable quantities. This would provide a direct measurement of the reduced matrix elements $\langle D_{3/2}||T_k||D_{5/2}\rangle$.

In conclusion we have measured the nuclear magnetic octupole moment for $^{137}\text{Ba}^+$ down to the 1% accuracy level, that is limited by the accuracy with which the reduced matrix elements $\langle D_J||T_3||D_J \rangle$ can be calculated.

### 7.2 Future work

After performing the most accurate nuclear magnetic octupole measurement to date, it seems fitting that we should continue our research in the same direction. The preferred next step would be to perform the same measurement but using a different isotope of barium. The next logical isotope to choose would be $^{135}\text{Ba}^+$, as this is the next most
abundant isotope after $^{137}\text{Ba}^+$ to have a nuclear spin $I = 3/2$. A measurement on $^{135}\text{Ba}^+$ would have the effect of further improving theoretical models as direct comparisons between isotope could be performed. The technologies developed for the octupole measurement could allow us to perform experiments aiming to discover new physics beyond the standard model. One such experiment is a low energy PNC measurement. There has for a longtime been a proposal to perform a PNC measurement [25] in barium, which is explained in further detail in Section 7.2.1. Other than probing exotic nuclear physics in barium, optical frequency metrology is possible, which requires the detection and shelving technologies we developed for the octupole measurement. Barium has two optical quadrupole transitions that would make good clock transitions for an atomic clock, see Section 7.2.2 for further details. Another direction our group has already moved onto is to ion trapping lutetium, further details of the aims of the project and current status are given in Section 7.2.3.

7.2.1 Barium PNC measurement

The PNC measurement in a Barium ion has been the focus of the Washington group headed by Professor E. N. Forston for over two decades now. The idea was put forward in 1993 [25] and to this day the group has been working to make their measurement apparatus sensitive enough to perform this experiment. In this section we just briefly summarize their idea. All credit for this work must go to the Washington group. For a much more detailed discussion see [111, 113, 114].

Parity violation was experimentally demonstrated in 1957 [115] through the beta decay of $^{60}\text{Co}$. In itself PNC is strange as it implies the universe is some how left-handed. However, the phenomenon can be used as a tool for accepting or rejecting extensions to the standard model. Putting a limit on PNCs effect will allow certain extension to the standard model, like certain forms of super symmetry [116] to be ruled out. Similar to other atomic physics experiments like the measurement of the electron’s electric dipole moment [117].

A low-energy table top atomic physics experiment has the advantage of a much better signal to noise ratio than a high energy proton-proton scattering PNC measurement [118] and the experimental setup is typically less complex. Low-energy Stark interference measurements have already been performed in atomic cesium [119] which

\[\text{http://www.phys.washington.edu/~fortson/}\]
has put a bound on PNC. Performing a PNC measurement with a barium ion has
the advantage of improved systematic errors over the cesium experiment. The cesium
experiment was performed with an atomic beam, which has a relatively complicated
thermal distributions of atoms, whereas a trapped ion is ideally just a single isolated
particle at rest in free space.

\[ E_2 \Omega'' \]
\[ E_1^{\text{PNC}} \Omega' \]
\[ \delta \omega_{\text{PNC}} \sim 1 \text{ Hz} \]
\[ 2051 \text{ nm} \]
\[ 6S_{1/2} \]
\[ 5D_{3/2} \]

**Figure 7.1:** PNC measurement scheme. A coupling \( \Omega'' \) of \( 6S_{1/2} \) to \( 5D_{3/2} \) via the electric
quadrupole (E2) interaction can interfere with another coupling \( \Omega' \) meant to drive the
small electric dipole (E1\text{PNC}) interaction allowed by parity violation. This interference can
change the Zeeman shift in the \( 6S_{1/2} \), depending on the phase of the (E2) field.

The reason barium in particular is a good candidate for a low energy PNC mea-
surement is that the atomic number of barium is high and the atomic structure can
be calculated precisely. In general, matrix elements that enable such parity violating
transitions scale quickly with increasing atomic number Z by the so-called \( Z^3 \) scaling
law \[24\]. A barium ion PNC experiment would measure a light shift due to parity
violation that is induced when a particular combination of optical fields are applied.
This light shift splits the ground state \( 6S_{1/2} \) Zeeman sublevels and the energy shift, or
equivalently the ground state precession rate it implies, see Fig. 7.1 for further details.
The calculated size of a PNC violation signal in \( ^{138}\text{Ba}^+ \) would be on the order of \( \sim 1 \text{ Hz} \) \[113\]. We have already demonstrated with our measurements of the hyperfine
splitting that we can achieve this kind of accuracy. We have also shown that we have
good control over our systematic uncertainties, so we will have a good understanding
of how these would affect the PNC measurement. For performing the experiment we
would require a laser at 2051 nm to provide a light shift between the \( 6S_{1/2} \) state and
5D$_{3/2}$ state. It is possible to purchase a fibre laser at 2051 nm or we could build our own 2051 nm laser with the introduction of commercially available laser diodes \(^1\).

### 7.2.2 Barium as an atomic clock

Atomic clocks are the foundation of modern navigation devices. Without their precise timing, navigational positioning down to an accuracy of 10 m would not be possible. The precise timing in the satellites of the GPS is provided by two rubidium and two cesium atomic clocks with a clock stability of at least 10$^{-13}$. The clock quality factor $Q$ is given as

$$Q = \frac{\nu}{\Delta \nu}$$

and $\nu$ is the fundamental frequency of the oscillator. The clock quality factor is an indicator of how good the clock will perform, but it is not the only factor. Improvements can also be made to the signal to noise ratio, minimizing the time required to detect a 180° phase shift. The best clocks in the world are optical \(^2\) because the fundamental frequency is in the THz range, which gives high quality factors and seconds needed to detect a 180° phase shift. Currently the second most accurate atomic clock is based on an optical transition in an aluminum ion \(^3\) and has a stability of 8.6$\times$10$^{-18}$.

Barium would make a good candidate for an optical atomic clock as it has two quadrupole transition, 6S$_{1/2}$ to 5D$_{3/2}$ or 5D$_{5/2}$ states. The lifetime of the 5D states is on the order of 10 seconds, which means the transition linewidths $\Delta \nu$ are very narrow. As both transition are in the optical regime the quality factor for the clock would be $Q \approx 1 \times 10^{-16}$. Ionized barium has been considered previously as a candidate for an atomic clock where an extremely detailed account is given in \(^4\). The limiting factor in the barium clock’s absolute stability is the black body radiation shift, which at 25 °C would limit the its accuracy to $\approx \pm 4 \times 10^{-16}$ \(^5\). This shift can be reduced by placing the trap in a cryostat that could maintain a temperature of 77 K, equivalent to an uncertainty level on the 10$^{-18}$ level, which is comparable to today’s most accurate clock. Going cooler still would further improve the black body radiation uncertainty, but at the 10$^{-18}$ level other factors become a problem like the first and second order Doppler shifts. Due to the black body radiation barium is not the best choice for an atomic clock.

\(^1\)http://sarnoff.org/downloads/products/lasers/SAR-2050-DFB.pdf

\(^2\)http://sarnoff.org/downloads/products/lasers/SAR-2050-DFB.pdf

\(^3\)http://sarnoff.org/downloads/products/lasers/SAR-2050-DFB.pdf


atomic clock but it does offer the ability to produce the first in house built atomic clock for Singapore that could easily be synced to the CQT frequency comb.

### 7.2.3 Lutetium as an ion

![Energy Level Diagram](image)

**Figure 7.2:** Lu⁺ energy level diagram, showing relevant transitions. The cooling transition would be performed between the $^3P_0$ to $^3D_1$ states. The linewidth of this transition is $\Gamma = 2\pi \times 2.6$ MHz [122].

Recently we have begun actively trapping and cooling lutetium with the aims of exploring it as a potential atomic clock candidate. Lutetium makes a good candidate for an atomic clock as it has a number of highly forbidden transitions that have lifetimes measured in years. These ultra narrow transitions have potential for being used as a clock transition such as the one in Aluminum ion clock [121]. Singly ionized lutetium has two electrons in its valance shell meaning it has an atomic structure that is similar to neutral barium, which is shown in Fig. 7.2. The two valance electrons makes
lutetium a suitable candidate for implementing an atomic clock that has a much lower susceptibility to blackbody radiation [123].

Further to being an atomic clock candidate, lutetium isotopes also have the property of having large nuclear spins, with \(^{175}\text{Lu}\) and \(^{176}\text{Lu}\) having nuclear spins of \(I = 7/2\) and \(I = 7\) respectively [124]. This opens up the possibility to observing nuclear moments beyond the octupole as \(I > 2\) for both isotopes. Nuclear moments above the octupole have not been observed previously and observation would lead to a better understanding of the hyperfine structure of atoms and the structure of the nucleus. A total angular moment of \(J = 3\) means the 6\(^{th}\) order nuclear moment is observable. A measurement of this nuclear moment is possible by using rf transitions to measure the hyperfine intervals of the \(^3\text{D}_3\) state in either \(^{175}\text{Lu}\) or \(^{176}\text{Lu}\). Single ionized lutetium makes a nuclear moment measurement above the octupole relatively simple because the hyperfine splittings in the D state is on the order of GHz. This means that the issues due to hyperfine intervals being on the order of the Zeeman interaction are negligible and hyperfine states will not be mixed to an significant degree. Therefore the measurement should be more straight forward than measuring the hyperfine splitting of the \(^5\text{D}_{5/2}\) manifold in \(^{137}\text{Ba}^+\).

Our current progress with working with this ion is that we have it trapped and have fluoresced on the 646 nm cycling transition. The ion is trapped by first trapping barium, which is used as a sympathetic cooler to trapped lutetium. We confirmed we have trapped lutetium by measuring the axial trapping frequencies with just barium trapped or barium plus one dark ion. As trapping frequencies are mass dependent, there is a difference between trapping barium and trapping barium plus a dark ion. This difference in the frequencies can be used to calculate the mass of the dark ion confirming we have trapped lutetium to an accuracy of a couple of atomic mass units.
Appendix A

Hyperfine interaction model of 5D_{5/2} manifold

In this section we discuss the model used for fitting the data. This model incorporates the influence of hyperfine mixing between the 5D_{3/2} and 5D_{5/2} manifolds. In Ba\(^+\) the fine structure splitting of the 5D level is very large (24.0 THz) and thus the influence of any mixing between the 5D_{3/2} and 5D_{5/2} manifolds can be determined by perturbation theory.

Since the Zeeman interaction only mixes states with the same \(m_F\), we can restrict ourselves to a particular \(m_F\). Neglecting any mixing with the 5D_{3/2} levels, the Hamiltonian in the presence of a magnetic field for the \(m_F = 0, \pm 1\) levels of 5D_{5/2} is given by

\[
H_0 = \begin{pmatrix}
E_1 & 0 & 0 & 0 \\
0 & E_2 & 0 & 0 \\
0 & 0 & E_3 & 0 \\
0 & 0 & 0 & E_4
\end{pmatrix} + U_{m_F}^\dagger \begin{pmatrix}
-\frac{3}{2} & 0 & 0 & 0 \\
0 & -\frac{1}{2} & 0 & 0 \\
0 & 0 & \frac{1}{2} & 0 \\
0 & 0 & 0 & \frac{3}{2}
\end{pmatrix} U_{m_F} (g_J - g_I) \mu_B B, \quad (A.1)
\]

where \(U_{m_F}\) is the unitary transformation between the \(F\) and \(IJ\) bases for the \(m_F\) level of interest. We note that we have omitted a term \(g_I m_F \mu_B B\) which, being proportional to the identity matrix, simply adds to the energy eigenvalues and does not impact on any of the following discussion.

The Hamiltonian given in Eq. (A.1) neglects mixing of the 5D_{3/2} and 5D_{5/2} levels.
Strictly speaking, the hyperfine eigenstates are given by

\[ |D_{5/2}, F\rangle = \alpha_F |D^0_{5/2}, F\rangle + \beta_F |D^0_{3/2}, F\rangle, \quad (A.2) \]
\[ |D_{3/2}, F\rangle = \alpha_F |D^0_{3/2}, F\rangle - \beta_F |D^0_{5/2}, F\rangle, \quad (A.3) \]

where the coefficients \( \alpha_F, \beta_F \) are independent of \( m_F \) and satisfy \( \alpha_F^2 + \beta_F^2 = 1 \). The full Zeeman Hamiltonian, \( H_z = \mu_B (g_s S_z + g_L L_z + g_I I_z) B/h \), then has the form

\[
\begin{pmatrix}
H_a & H_{ab} \\
H_{ab}^\dagger & H_b
\end{pmatrix},
\]

where matrix elements of \( H_a, H_b \), and \( H_{ab} \) are given by

\[
\langle D_{5/2}, F | H_a | D_{5/2}, F' \rangle = \alpha_F \alpha_F' \langle D^0_{5/2}, F | H_z | D^0_{5/2}, F' \rangle
+ \alpha_F \beta_F' \langle D^0_{5/2}, F | H_z | D^0_{3/2}, F' \rangle
+ \beta_F \alpha_F' \langle D^0_{3/2}, F | H_z | D^0_{5/2}, F' \rangle
+ \beta_F \beta_F' \langle D^0_{3/2}, F | H_z | D^0_{5/2}, F' \rangle,
\]

\( (A.5) \)

\[
\langle D_{5/2}, F | H_{ab} | D_{3/2}, F' \rangle = \alpha_F \alpha_F' \langle D^0_{5/2}, F | H_z | D^0_{3/2}, F' \rangle
- \alpha_F \beta_F' \langle D^0_{5/2}, F | H_z | D^0_{5/2}, F' \rangle
+ \beta_F \alpha_F' \langle D^0_{3/2}, F | H_z | D^0_{3/2}, F' \rangle
- \beta_F \beta_F' \langle D^0_{3/2}, F | H_z | D^0_{5/2}, F' \rangle,
\]

\( (A.6) \)

\[
\langle D_{3/2}, F | H_b | D_{3/2}, F' \rangle = \alpha_F \alpha_F' \langle D^0_{3/2}, F | H_z | D^0_{3/2}, F' \rangle
- \alpha_F \beta_F' \langle D^0_{3/2}, F | H_z | D^0_{5/2}, F' \rangle
- \beta_F \alpha_F' \langle D^0_{5/2}, F | H_z | D^0_{3/2}, F' \rangle
+ \beta_F \beta_F' \langle D^0_{5/2}, F | H_z | D^0_{5/2}, F' \rangle,
\]

\( (A.7) \)

The parameters \( \beta_F \) can be determined from atomic structure calculations and to first
order in the hyperfine interaction we have \[35\]

\[
\beta_F = \frac{\langle D_{3/2}^0, F | H_{\text{HFI}} | D_{5/2}^0, F \rangle}{E_{D_{5/2}^0} - E_{D_{3/2}^0}} = \frac{(-1)^{F+1}}{E_{D_{5/2}^0} - E_{D_{3/2}^0}}
\times \sum_k \left\{ \begin{array}{c} 3/2 \\ 3/2 \end{array} \right\} F_k \langle D_{3/2}^0 || T_k^0 || D_{5/2}^0 \rangle \langle I || T_k^0 || I \rangle.
\] (A.8)

Accuracy of a few percent can be obtained by including only the \( k = 1 \) and \( 2 \) terms in the summation. The reduced matrix elements \( \langle I || T_k^0 || I \rangle \) can be determined from the nuclear multipole moments and, from \[35\], they are \( 2 \sqrt{\frac{5}{3}} \mu \) and \( \sqrt{\frac{5}{3}} Q \) for \( k = 1 \) and \( 2 \) respectively where \( \mu \) and \( Q \) are given in Eqs. (2.14) and (2.15), respectively. Using matrix elements \( \langle D_{3/2}^0 || T_1^0 || D_{5/2}^0 \rangle \) and \( \langle D_{3/2}^0 || T_2^0 || D_{5/2}^0 \rangle \) given in Table 2.1 we obtain \( \beta_1 = 0.915 \times 10^{-5}, \beta_2 = 1.478 \times 10^{-5}, \) and \( \beta_3 = 1.698 \times 10^{-5}. \)

The terms in \( H_a \) and \( H_b \) proportional to \( \alpha_F \alpha'_F \approx 1 \) are simply the elements of the Zeeman Hamiltonian neglecting any mixing. All the other terms can be treated as a perturbation. Elements of \( H_{ab} \) only influence the energy levels at second order giving shifts \( \sim (\mu_B B)^2/E_{FS} \) where \( E_{FS} \) is the fine structure splitting. For the \( B \) fields considered in this work this amounts to shifts \( \lesssim 0.25 \text{ Hz} \) and thus we can neglect \( H_{ab} \) altogether. Moreover, the terms proportional to \( \beta_F^2 \) will contribute at most by \( \sim \beta_F^2 \mu_B B \) which amounts to level shifts of only a few mHz. Thus, concerning the measurements in the \( 5D_{5/2} \) manifold, we need only to consider the terms in \( H_a \) that are proportional to \( \beta_F \) as a perturbation to the zero order Hamiltonian given in Eq. (A.1).

The matrix elements \( \langle D_{5/2}^0, F | H_z | D_{3/2}^0, F' \rangle \) are all proportional to \( (g_s - g_L) \mu_B B \) and the proportionality constants are given in Tables A.1 and A.2.

**Table A.1**: \( \langle D_{5/2}^0, F | H_z | D_{3/2}^0, F' \rangle \) elements scaled by \( (g_s - g_L) \mu_B B \) for \( m_F = 0. \)

<table>
<thead>
<tr>
<th>( F )</th>
<th>( F' )</th>
<th>3</th>
<th>2</th>
<th>1</th>
<th>0</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>1/5√5</td>
<td>0</td>
<td>−1/5√5</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1/5√3</td>
<td>0</td>
<td>−2/5√3</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>−2/5√3</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>−2/5√3</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>
Table A.2: \( \langle D_{5/2}^0, F | H_Z | D_{3/2}^0, F' \rangle \) elements scaled by \((g_s - g_L)\mu_B B\) for \( m_F = \pm 1 \).

<table>
<thead>
<tr>
<th>( F )</th>
<th>( F' )</th>
<th>3</th>
<th>2</th>
<th>1</th>
<th>0</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>( \frac{3}{10} \sqrt{\frac{3}{5}} )</td>
<td>( \mp \frac{3}{10} \sqrt{\frac{5}{7}} )</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>( \frac{2}{5} \sqrt{\frac{2}{105}} )</td>
<td>( \mp \frac{1}{10} \sqrt{\frac{7}{3}} )</td>
<td>( -\frac{3}{10} \sqrt{\frac{7}{5}} )</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>( \pm \frac{1}{5\sqrt{5}} )</td>
<td>( -\frac{8}{5\sqrt{15}} )</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>( -\sqrt{\frac{3}{14}} )</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>

For the \( m_F = 0 \) case, the perturbation has the form

\[
\begin{pmatrix}
0 & \frac{1}{5\sqrt{5}}\beta_2 - \frac{1}{5} \sqrt{\frac{21}{5}}\beta_1 & 0 & 0 \\
\frac{1}{5\sqrt{5}}\beta_2 - \frac{1}{5} \sqrt{\frac{21}{5}}\beta_1 & 0 & -2 \sqrt{\frac{6}{5}}\beta_2 + \frac{1}{5} \sqrt{\frac{35}{3}}\beta_3 & 0 \\
0 & -2 \sqrt{\frac{6}{5}}\beta_2 + \frac{1}{5} \sqrt{\frac{35}{3}}\beta_3 & 0 & -2 \sqrt{\frac{35}{3}}\beta_3 \\
0 & 0 & -2 \sqrt{\frac{35}{3}}\beta_3 & 0
\end{pmatrix} \quad (A.9)
\]

Due to the fact that only \( F = 3 \) and \( F = 4 \) levels are mixed significantly by the Zeeman interaction, the unitary transformation that diagonalizes Eq. (A.1) has the approximate form

\[
\begin{pmatrix}
\mathbb{I} & 0 \\
0 & R
\end{pmatrix} \quad (A.10)
\]

where \( R \) is a rotation matrix that depends on the strength of the magnetic field. Consequently, in the basis of states that diagonalizes Eq. (A.1), there are no significant diagonal elements of the perturbation associated with the states \(|1, 0\rangle \) and \(|2, 0\rangle \). Calculations confirm shifts of \(< 1 \text{ Hz} \) for magnetic fields of \(< 2 \text{ G} \). Thus, for the energies of these two states we can neglect mixing with the 5D_{3/2} manifold altogether. We note that the lack of diagonal elements in the perturbation for the \( m_F = 0 \) case is a consequence of the magnetic dipole selection rule that \( m_F' = 0 \leftrightarrow m_F = 0 \) is forbidden when \( \Delta F = 0 \). Consequently, our previous measurements for the 5D_{3/2} level were not affected by hyperfine mixing of the fine structure levels.
For the $m_F = \pm 1$ case, the perturbation has the form

$$
\begin{pmatrix}
\pm \frac{3}{5}\beta_1 & -\frac{3}{10}\sqrt{\frac{7}{5}}\beta_1 + \frac{1}{10}\sqrt{\frac{3}{5}}\beta_2 & 0 & 0 \\
-\frac{3}{10}\sqrt{\frac{7}{5}}\beta_1 + \frac{1}{10}\sqrt{\frac{3}{5}}\beta_2 & \mp \frac{1}{5}\sqrt{\frac{5}{3}}\beta_2 & -\frac{8}{5}\frac{1}{\sqrt{15}}\beta_2 + \frac{2}{5}\sqrt{\frac{2}{105}}\beta_3 & 0 \\
0 & -\frac{8}{5}\frac{1}{\sqrt{15}}\beta_2 + \frac{2}{5}\sqrt{\frac{2}{105}}\beta_3 & \mp \frac{2}{5}\frac{1}{\sqrt{14}}\beta_3 & -\sqrt{\frac{3}{10}}\beta_3 \\
0 & 0 & -\sqrt{\frac{3}{10}}\beta_3 & 0 \\
\end{pmatrix}
$$

(A.11)

The states of interest here are those associated with the $F = 3$ and $F = 4$ levels. From the approximate form of the unitary transformation that diagonalizes Eq. (A.1), these levels are only influenced by the terms proportional to $\beta_3$, and we note that these elements provide level shifts on the order of $\beta_3\mu_B B/h \sim 2\pi \times 25\text{ Hz}$. For the purposes of modeling the energy levels of interest we therefore use Eq. (A.1) for the $m_F = 0$ states measured, while for the $F = 3$ and $F = 4$ levels we include the perturbation

$$
\begin{pmatrix}
0 & 0 & 0 & 0 \\
0 & 0 & \frac{2}{5}\sqrt{\frac{2}{105}} & 0 \\
0 & \frac{2}{5}\sqrt{\frac{2}{105}} & \mp \frac{2}{5}\frac{1}{\sqrt{14}} & -\sqrt{\frac{3}{10}} \\
0 & 0 & -\sqrt{\frac{3}{10}} & 0 \\
\end{pmatrix}
\beta_3(g_s - g_L)\mu_B B,
$$

(A.12)

in Eq. (A.1).

Note that while writing this thesis it was realized that the mixing could have a significant shift on the $m_F = \pm 1$ states in the $5D_{3/2}$ manifold. To estimate the size of this shift a similar analysis can be carried out. The $m_F = \pm 1$ states in the $5D_{3/2}$ manifold where used to determine the magnetic fields and hence any mixing will result in an error in the magnetic field measurement. The induced shift in the magnetic field is field dependent and worst for the $F = 1$ states where it is $\approx 15\mu\text{G}$ at 1 G. The resulting shift in the intervals of the $5D_{3/2}$ hyperfine intervals measurement is largest for the $\delta W^{(3/2)}_0$, where it is $\approx 0.3\text{ Hz}$. In the intervals of the $5D_{5/2}$ manifold the largest shift is in the $\delta W^{(5/2)}_1$, where it is $\approx 0.5\text{ Hz}$. In both cases the errors are well within in the stated errors.
Appendix B

Nonlinear Optics

The field of nonlinear optics is vast and includes a large number of effects but of these effects only one is of interest in our research. The effect of Second harmonic generation (SHG) or frequency doubling is of great use to us as it allows to access wavelength of laser light not available as a diode laser. In this section we will cover the theoretical details of of SHG. This section will concentrate on phase matching and conversion efficiency in the nonlinear medium. The basics principle of SHG is to take two pump photons at frequency $\omega$ and through their interaction with a nonlinear medium are converted into a single photon with a frequency $2\omega$, which is covered in greater detail in [125].

B.1 Phase matching

For SHG to take place it must obey conservation of energy

$$\omega_3 = \omega_1 + \omega_2,$$

(B.1)

where $\omega_1 \equiv \omega_2$ for SHG and $2\omega_1 \equiv \omega_3$. Hence the wave-vectors within the nonlinear medium must also obey this relationship

$$\Delta k = \vec{k}_1 + \vec{k}_2 - \vec{k}_3.$$

(B.2)

Due to dispersion in optical mediums the refractive index of the medium must be considered, and so the wave-vector are given by

$$\vec{k}_n = \frac{2\pi n_n}{\lambda_n},$$

(B.3)
where \( n = 1, 2, 3 \). If all the light propagates in the same direction then only the magnitude of the wave-vector needs to be considered \(|\vec{k}_n| = k_n\). The aim of SHG is to convert as much of the fundamental light to the second harmonic as possible. This is achieved by ensuring the phase velocities of the fundamental and doubled light remain the same inside the crystal. The effect of phase mismatch on conversion efficiency can be expressed by a sinc function [126]

\[
\eta_{\text{shg}} \propto \frac{\sin^2 \left( \frac{\Delta k l}{2} \right)}{\left( \Delta k l / 2 \right)^2}.
\]  

(B.4)

Phase matching \( \Delta k = 0 \) between the fundamental and the harmonic can be achieved using birifringent crystals. The ordinary (o) and the extraordinary (e) polarization modes can be exploited by angle, temperature and polarization tuning so that the condition is satisfied

\[
n(\omega) + n(\omega) = 2n(2\omega).
\]

(B.5)

There are two types of phase matching methods that can be exploited to satisfy Eq. (B.5). For type I (ooe), the fundamental waves have the same polarization while the second harmonic has a polarization perpendicular to the incident waves. This has the advantage that a single fundamental polarization can be used. For type II (oeo), the two fundamental waves have different polarization and it requires the splitting of the fundamental beam. This generally yields a significantly smaller conversion efficiency and is used when the birefringence is relatively strong. As the index of refraction varies substantially with the angle of incidence the easiest way to satisfy Eq. (B.5) is by angle tuning, which is known as CPM. The phase matching angle can be found from this relation [126]

\[
\frac{1}{n^2(\theta, \omega)} = \frac{\sin^2 \theta}{n_o(\omega)} + \frac{\cos^2 \theta}{n_e(\omega)}.
\]

(B.6)

where \( \theta \) is the phase matching angle. The phase matching angle is normally described as the cut of the nonlinear crystal and is the angle between the optical propagation axis and the principal axis of the crystal.

The second method is to use the effect of temperature to tune the refractive indices \( n_o \) and \( n_e \), with respect to each other. This only works for crystals that have a refractive index that varies greatly with temperature. This method is known as NCPM and has the advantage of being able to set the optical axis to be perpendicular with respect to the crystal axis, which means the fundamental beam will not suffer from spatial
walk off. Spatial walk off occurs only for a beam with extraordinary polarization, propagating at some angle $\theta$ against the optical axes, so that the refractive index $n_e$ and the phase velocity become dependent on that angle. Walk off can be a big problem for CPM because it causes the conversion efficiency to be greatly reduced.

**B.2 SHG conversion efficiency**

For Gaussian beams the conversion efficiency of a nonlinear crystal can be described by Boyd-Kliemann theory [127]. The second harmonic power in single pass can be described as

$$P_{2\omega} = \left( \frac{16\pi^2 d_{\text{eff}}^2}{\lambda^2 \epsilon_0 n^2 \epsilon_0} \right) \times L e^{-\left(\alpha_1 + \alpha_2/2\right)L} \times h(\sigma, \beta, \xi) \times P_{\omega}^2, \quad (B.7)$$

where $L$ is the length of the crystal, $n$ is the refractive index of the crystal at phase matching, $\lambda$ is the fundamental wavelength, $\epsilon_0$ is the vacuum permittivity, $c_0$ is the speed of light in vacuum, $d_{\text{eff}}$ is effective nonlinear coefficient, $P_{\omega}$ is the power of the fundamental beam, $\alpha_1, \alpha_2$ are absorptive losses at the fundamental and second harmonic frequency respectively. The function $h(\sigma, \beta, \xi)$ is the Boyd-Kliemann harmonic factor, which must be optimized for various parameters.

The harmonic factor parameters are given as $\sigma$ the phase mismatch factor

$$\sigma = \frac{1}{2} b \Delta k, \quad (B.8)$$

where $b$

$$b = \frac{2\pi w_0^2}{\lambda} \quad (B.9)$$

is the confocal parameter and $w_0$ is the waist inside the crystal. The intensity profile at the beam waist is defined as

$$I(r) = \frac{P_{\omega}}{\pi w_0^2/2} \ e^{-2\frac{r^2}{w_0^2}}. \quad (B.10)$$

The parameter $\beta$ is the walk-off factor

$$\beta = \frac{D}{2} \sqrt{\frac{2\pi n L}{\lambda}}, \quad (B.11)$$
where \( \rho \) is the walk off angle inside the nonlinear crystal. The final parameter is \( \xi \), which is the focusing parameter

\[
\xi = \frac{L}{b} \quad \text{(B.12)}
\]

The focusing parameter fixes the optimum waist for chosen crystal length. The harmonic function is given by

\[
h(\sigma, \beta, \xi) = \frac{1}{4\xi} \int_{-\xi}^{\xi} \int_{-\xi}^{\xi} e^{i\sigma(\tau-\tau')} e^{-\beta^{2}(\tau-\tau')^{2}/\xi} \frac{d\tau d\tau'}{(1+i\tau)(1-i\tau')}, \quad \text{(B.13)}
\]

which can be calculated numerical for certain SHG cases. The function is always calculated with the phase mismatch factor maximized \( h_m(\beta, \xi) = \max[(\sigma, \beta, \xi)]_{\sigma} \).

To improve the calculation speed of Eq. (B.13) the authors of [128] found an approximate analytic solution to the harmonic factor for maximized phase mismatch. The approximate analytic solution is given by

\[
h_m(\beta, \xi) = \frac{h_{mm}(\beta)\gamma(\beta)\xi}{|\xi - \xi_m(\beta)|n(\beta) + \gamma(\beta)\xi}, \quad \text{(B.14)}
\]

where \( \xi_m(\beta) \) is

\[
\xi_m(\beta) = \frac{2.84 + 1.39\beta^2}{1 + 0.1\beta + \beta^2}, \quad \text{(B.15)}
\]

\( h_{mm}(\beta) \) is

\[
h_{mm}(\beta) = \frac{1.068}{1 - \sqrt{0.7\beta + 1.62}\beta}, \quad \text{(B.16)}
\]

\( n(\beta) \) is

\[
n(\beta) = \frac{1.91 + 1.83\beta}{1 + \beta}, \quad \text{(B.17)}
\]

and \( \gamma(\beta) \) is

\[
\gamma(\beta) = \frac{|\xi_m(0)|n(0)}{h_{mm}(0)} e^{-\beta} + 13 \left(1 - e^{-\beta/3}\right). \quad \text{(B.18)}
\]

Using Eq. (B.14) which is less than 5% different from Eq. (B.13) a plot of the harmonic factor against the focusing parameter for different values of walk off is given in Fig. B.1. From this figure it can be seen that the harmonic function has a maximum value of 1.068 for \( \beta = 0 \) and \( \xi = 2.84 \), but for \( \beta > 1 \) the harmonic function is maximized at \( \xi = 1.39 \).
Figure B.1: Harmonic factor plotted against focusing parameter for different values of walk off factor.
B.3 Cavity enhanced SHG

Unless the fundamental source for SHG is a pulse or very high power CW laser then the conversion from fundamental to second harmonic will be very poor in single pass configuration. To achieve high conversion efficiency starting from medium to low power CW (≈50 mW to 1 W) laser a cavity is normally used to enhance the intensity in the crystal, although breakthrough waveguide technology allows reasonable conversion in single pass now, see Section 3.4.2. Up to 85% conversion has been achieved [129, 130] using cavity enhancement in a laser with less than 1 W output. The improvement in conversion efficiency comes from the cavity enhancing the fundamental power, which is given by

\[ A = \frac{t}{(1 - \sqrt{1 - t\mathcal{L})}}^2, \]  

(B.19)

where \( t \) is the transmission of the output coupler and \( \mathcal{L} \) is the losses within the cavity. The power circulating in the cavity is then just

\[ P_{\text{cav}} = A\eta_{\text{koup}}P_{\text{in}}, \]  

(B.20)

where \( \eta_{\text{koup}} \) is the coupling efficiency of the incident light onto the cavity. The second harmonic power scales as the input power squared. A small enhancement due to a doubling cavity will lead to a large increase in the second harmonic power. The losses \( \mathcal{L} \) are defined as

\[ \mathcal{L} = (1 - \alpha_1 L)(1 - \eta_{\text{shg}} P_{\text{cav}})(1 - v), \]  

(B.21)

where \( v \) is other cavity losses due to crystal interface and losses off mirrors.

![Figure B.2: Optimum output transmission for doubling cavity losses](image-url)
The output coupler’s transmission $t$ should match the losses in the cavity to ensure optimum conversion, which is known as impedance matching. As the conversion becomes larger the losses in the cavity become greater, this is because the enhancement of the cavity depends on how much fundamental light is converted to second harmonic light which also depends on the enhancement of the cavity. This circular problem can be solved numerically using a minimization function to find the optimum output coupler for expected conversion efficiency. There is also a simpler relation that relates cavity losses to optimal input coupler, which is given by \[ r_{\text{opt}} = 1 - (\mathcal{L} + \sqrt{\eta_{\text{shg}} P_{\text{in}}}) \], (B.22)
which is plotted in Fig. B.2.

Figure B.3: Schematic showing bow-tie doubling cavity

A doubling cavity can be of many designs, but the most popular is the bow-tie design shown in Fig. B.3, as this is a running wave design so second harmonic light is only emitted in one direction. The nonlinear crystal’s length fixes the optimum waist for the optimum focusing parameter. The waist inside the nonlinear crystal determines the lengths, folding angle and radius of curvature for the doubling cavity. A full complement of design equations are given in \[132\].

B.4 Waveguide enhanced SHG

A waveguide improves doubling conversion efficiency by confining the light field tightly within the guide. The waveguide confinement means the waist of the fundamental beam
Figure B.4: Schematic of waveguide doubling crystal. The end view shows the waveguides in the top surface of the crystal. The waveguides have approximate dimensions of $3 \times 3 \, \mu m$ and there are roughly 30 in the crystal. The top view shows the polling of the crystal to achieve quasi-phase matching.

can remain small less than 10 \( \mu m \) throughout the length of the crystal. The single pass waveguide doubling crystal is manufactured by AdvR$^1$ from PPKTP. A schematic of the crystal design is given in Fig. B.4. The crystal is periodically polled to achieve quasi-phase matching [133] at 1228 nm. Phase matching is achieved by temperature tuning the crystal so that the polling period is matched to the wavelength of the fundamental light. The crystal is temperature stabilized by resistively heating the mount, which the crystal sits on. The crystal has waveguides etched into the top surface via ion exchange [134]. The waveguides have an average mode field diameter of $3 \, \mu m$. The waveguides act as light guide to keep the intensity of the 1228 nm high throughout the interaction length of the crystal, which is 20 mm long.

$^1$www.advr-inc.com
Appendix C

Thermalization of Blackbody Radiation

In this appendix we briefly describe the considerations that went into designing the heat shield reference cavity detailed in Section 3.5.2. The focus of the design is to minimize heat conductivity to the Zerodur cavity spacer, in order to minimize temperature changes which would alter the cavity length and hence its resonance frequency. We implement three measures to achieve this goal. The first is to limit heat conduction by minimizing contact of the cavity spacer to the environment. To minimize thermal contact to the vacuum nipple small pieces of rubber are used to mount the Zerodur spacer. The rubber provides two benefits, first is it has a low thermal conductivity and second it helps to damp out mechanical vibrations. The second measure is to remove the air from the vacuum nipple as this eliminates heat transfer via convection. The third is to minimize heat transfer via black-body radiation, which is the focus of this section.

Heat transfer via radiation is minimized by the use of heat shields. A good heat shield will be highly reflective to far-Infra-Red (IR) radiation, where the peak of the black-body radiation spectrum is at room temperature. Metals for example are excellent at reflecting far-IR, but can suffer from oxidization, which can turn them into good emitters/absorbers of far-IR. For the heat shields we chose aluminum as it is easy to machine and only develops a thin oxide layer, which does not overly effect its reflective properties. A better choice would have been gold coated copper as gold does not tarnish and has the highest reflectivity to far-IR radiation of all the metals, but its
cost made it prohibitive. Further reductions in heat transfer can be gained by adjusting
the geometry of the heat shield design. In order to justify our design choice we briefly
model the heat transfer in our cavity design, which is shown again in Fig. C.1. A more
detailed discussion of heat transfer can be found in [135].

\begin{align*}
\tau &= R_H C_H, \\
R_H &= \text{thermal resistance between the bodies and } C_H = \text{heat capacitance of one body. The heat capacitance can be represented as } \\
C_H &= C_m M,
\end{align*}

where $C_m$ is the material specific heat capacity and $M$ is the mass of the body. The thermal resistance can be modeled via an electrical analogue, where all thermal resistances are added up appropriately. The thermal resistances of the cavity system can be split into two different types. The first comes from contact resistance through the rubber mounts. The second is due to black-body radiation which includes the effects
due to the emissivity factors and the geometry factors of the surfaces involved [135]. Thermal contact resistance is defined as

$$R_c = \frac{kx}{A},$$

where \(x\) is the length of the material, \(A\) is the contact surface area between the bodies and \(k\) is the thermal resistivity of the material. The thermal resistance due to the emissivity factor is defined as

$$R_\varepsilon = h_r \sum_{i,j} \frac{1 - \varepsilon_i}{\varepsilon_i A_i} + \frac{1}{A_i F_{i \rightarrow j}},$$

where \(A_i\) is the emitting surface area, \(\varepsilon_i\) is the emissivity factor of the surface, \(h_r\) is the radiation heat transfer coefficient and \(F_{i \rightarrow j}\) is the shape factor for the emitting surface onto a receiving surface, which is a measure of how much one surface sees of the other surface. In the sum \(i\) and \(j\) run over all possible configurations. Here \(h_r\) is given as

$$h_r = \sigma T_m^3,$$

where \(\sigma\) is Stefan-Blotzmanns constant and \(T_m\) is the average temperature of the emitting surface.

Figure C.2: Schematic of concentric cylinders for calculating shape factor.

For our cavity design there are only two relevant shape factors that need to be considered. The first is the view factors between two concentric cylinders as illustrated.
in Fig. C.2 and defined as

\[ F_{1-2} = 1, \]  \hspace{1cm} (C.6)
\[ F_{2-1} = \frac{r_1}{r_2}. \]  \hspace{1cm} (C.7)

The second is the view factor between two stacked discs as illustrated in Fig. C.3 and defined as

\[ F_{1-2} = \frac{1}{2} \left( X - \sqrt{X^2 - \left( \frac{R_2}{R_1} \right)^2} \right), \]  \hspace{1cm} (C.8)

where

\[ R_1 = \frac{r_1}{h}, \]  \hspace{1cm} (C.9)
\[ R_2 = \frac{r_2}{h}, \]  \hspace{1cm} (C.10)
\[ X = 1 + \left( 1 + \frac{R_2^2}{R_1^2} \right). \]  \hspace{1cm} (C.11)

Figure C.3: Schematic of stacked discs for calculating shape factor.

Our cavity design can be view as a set of stacked cans, where the first can is the Zerodur cavity spacer, as illustrated in Fig. C.1. Using the heat shields dimensions and properties the thermalization time constant can be calculated between each shield. The time constants between each heat shield can be calculated from the parameters given in Table C.1. This gives time constants of 13.5 hours for heat transfer between the cavity spacer to inner heat shield; 11.3 hours between inner and outer shields; 4.2 hours between outer shield and vacuum nipple. These agree roughly with the total measured time constant of 36 hours for the whole system, where the discrepancy is most likely due variation in emissivity factors for the material of choice.
Table C.1: List of cavity parameters, where $L$ is component length, $R_1$ is inner radius and $R_2$ is outer radius. The emissivity factors given are taken from [135].

<table>
<thead>
<tr>
<th>Component</th>
<th>Material</th>
<th>Emissivity</th>
<th>$L$ (mm)</th>
<th>$R_1$ (mm)</th>
<th>$R_2$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cavity spacer</td>
<td>Zerodur</td>
<td>0.9</td>
<td>50</td>
<td>-</td>
<td>15</td>
</tr>
<tr>
<td>Inner shield</td>
<td>Aluminum</td>
<td>0.1</td>
<td>200</td>
<td>25</td>
<td>30</td>
</tr>
<tr>
<td>Outer shield</td>
<td>Aluminum</td>
<td>0.1</td>
<td>260</td>
<td>40</td>
<td>45</td>
</tr>
<tr>
<td>Vacuum nipple</td>
<td>Stainless steel</td>
<td>0.4</td>
<td>270</td>
<td>50</td>
<td>53</td>
</tr>
</tbody>
</table>
Appendix D

Calculation of ac-Zeeman shifts from various sources

In this section we discuss the ac-Zeeman shift calculations for both manifolds caused by various rf magnetic fields. These include ac-Zeeman shifts caused by off-resonant rf coupling from the driving antenna, ion motion in a magnetic field gradient and the rf magnetic field caused by trap electrodes. The calculations for all these shifts are similar. The only differences are the hyperfine states involved, the amplitude of the applied magnetic field and the frequency of the rf field. The interaction of an atom with total electronic spin \( J \) and an applied rf B-field \( B_{\text{rf}} \) is described by the interaction Hamiltonian [106]

\[
H_{\text{INT}} = \frac{g_J \mu_B B_{\text{rf}} \cdot \vec{J}}{\hbar}.
\]  

(D.1)

For the treatment presented here we neglect the similar coupling to the nuclear spin \( I \) because \( g_I \ll g_J \). In particular we are interested in Zeeman shifts to different \( |F_m\rangle \) states, which can be related back to \( |I_m\rangle \) states via

\[
|F_m\rangle = \sum_{m,J} C_{Fm,Fm,I}^{Fm,I} |I_m\rangle |J_m\rangle,
\]  

(D.2)

where \( C_{Fm,Fm,I}^{Fm,I} \) are Clebsch-Gordon coefficients. The ac-Zeeman effect can be calculated by following the treatment given in [106] (page 467.), which leads to a shift

\[
\Delta E_a = -\frac{\hbar \Omega^2}{4} \sum_k |\langle k | \hat{J}_q | a \rangle|^2 \left[ \frac{1}{\omega_{ak} - \omega_{\text{rf}}} + \frac{1}{\omega_{ak} + \omega_{\text{rf}}} \right],
\]

(D.3)
where \( \Omega_0 = \frac{\mu_B B_{rf} J_B}{\hbar} \) is the Rabi rate relating back to the two-level picture (see Eq. (2.37)) \( B_{rf} \) is the amplitude of magnetic field, \( \omega_{ak} \) is the frequency difference between the levels of interest and \( \omega_{rf} \) is the frequency of the magnetic field. Note that the second term in the square brackets is the counter-rotating term. Here \( |a\rangle \) and \( |k\rangle \) represent the states \( |Fm_F\rangle \) and \( |F'm_{F'}\rangle \) respectively, which are operated on by \( \hat{J}_q \), where \( q = (-, +, z) \).

The operation of \( \hat{J}_+ \) on the \( |j, m\rangle \) gives
\[
\hat{J}_+ |j, m\rangle = \sqrt{j(j+1) - m(m+1)} |j, m+1\rangle,
\]
(D.4)

\( \hat{J}_- \) is given as the transpose of \( \hat{J}_+ \) and \( \hat{J}_z \) is given as
\[
\hat{J}_z = \frac{1}{2} (\hat{J}_+ \hat{J}_- - \hat{J}_- \hat{J}_+),
\]
(D.5)

In appendix D.1 we calculate the \textit{ac}-Zeeman shift on the magnetic field calibration measurements due to off-resonant rf coupling from the antenna and rf fields from the trap in the 5D\(_{3/2}\) manifold. We also give the relevant matrix elements involved in the calculations. In appendix D.2 we explain how the \textit{ac}-Zeeman shift calculation was performed in the 5D\(_{5/2}\) manifold and give the relevant matrix elements involved in the calculations. For all these calculations we assume equal powers of all field components.

### D.1 \textit{ac}-Zeeman shift calculations and relevant matrix elements for 5D\(_{3/2}\) manifold

![Figure D.1: Schematic showing off-resonant coupling configuration which will cause a shift to the \( |F'' = 0, m_{F''} = 0\rangle \) causing the magnetic field calibration value to be incorrectly measured.](image)

First we look at the off-resonant \textit{rf} coupling from the antenna used to drive the hyperfine transitions. When performing the \( \Delta m_F = +1 \) transition measurement the \( |F'' = 0, m_{F''} = 0\rangle \) state is shifted by off-resonant coupling to the \( |F'' = 1, m_{F''} = 0\rangle \).
state and $|F'' = 1, m_{F''} = -1\rangle$ state, as illustrated in Fig. D.1. The net shift can be written as

$$\Delta E = \frac{\hbar \Omega_z^2}{8\Delta_x} + \frac{\hbar \Omega_{-z}^2}{4\Delta_x} = \frac{5}{2} \left( \frac{\hbar \Omega_z^2}{8\Delta_x} \right) + \frac{5}{4} \left( \frac{\hbar \Omega_{-z}^2}{4\Delta_x} \right).$$  \hspace{1cm} \text{(D.6)}$$

Here $\Omega_z$ is the Rabi rate for the $\Delta m_{F''} = 0$ transition, $\Omega_{-z}$ is the Rabi rate for the $\Delta m_{F''} = -1$ transition and $\Delta_x$ is the Zeeman splitting. The coefficients relating $\Omega_z$ and $\Omega_{-z}$ come from the relevant matrix elements listed in Fig. D.2. Note that we have neglected the counter-rotating terms because $\Omega_0 \ll (\omega_{ak} + \omega_{rf})$. The value for $\Omega_0$ can be related to the Rabi rate $\Omega_+$ for the $\Delta m_{F''} = 1$ transition that is driven by

$$\Omega_+ = \frac{5}{2} \Omega_0.$$

Finally we get a shift

$$\Delta E = \frac{\hbar \Omega_z^2}{4\Delta_x}.$$ \hspace{1cm} \text{(D.8)}$$

To evaluate this expression for the magnetic field calibration measurements we use $\Omega_+ \approx 1$ kHz and $\Delta_x$ according to the magnetic field.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure_d2.png}
\caption{Matrix elements squared $|(F'm_{F'}|\hat{J}_q|Fm_F)|^2$ between different $F$ and $m_F$ states for 5D\textsubscript{3/2} manifold.}
\end{figure}

For an rf magnetic field generated by the trap electrodes the shift induced on the $|F'' = 1, m_{F''} = -1\rangle$ in the limit that $\omega_{rf} > \Delta_x$, can be written as

$$\Delta E = \frac{\hbar \Omega_0^2}{2} \frac{1}{4} \left[ \frac{1}{\omega_{rf} - \Delta_x} - \frac{1}{\Delta_x + \omega_{rf}} \right].$$  \hspace{1cm} \text{(D.9)}$$
Figure D.3: Matrix elements squared $|(F'M_F|\hat{J}_z|Fm_F)|^2$ between different $m_F$ states in the same $F$ level for 5D$_{3/2}$ manifold.

The additional factor of $\frac{1}{2}$ at the front comes from the matrix elements squared, which can be found in Fig. D.3 and here we need to take the counter-rotating terms into account because $\Omega_0 \ll (\Delta_{\chi} + \omega_{rf})$. To evaluate the shifts we use $B_{rf} \approx 3.5 \mu\text{G}$ and $\omega_{rf} \approx 2\pi \times 5.3 \text{ MHz}$. The remaining ac-Zeeman shifts can be found by performing similar calculations between the relevant states.

D.2 ac-Zeeman shift calculations and relevant matrix elements for 5D$_{5/2}$ manifold

The same calculation described in appendix D.1 can be performed in the 5D$_{5/2}$ manifold for the $F''=3$ and $F''=4$ levels. In the 5D$_{5/2}$ manifold, due to the mixing, the states are defined by the approximate form

$$|+\rangle \approx \sin \theta_+ |F''=3, m_{F''}=+1\rangle + \cos \theta_+ |F''=4, m_{F''}=+1\rangle,$$

$$|-\rangle \approx \cos \theta_- |F''=3, m_{F''}=-1\rangle - \sin \theta_- |F''=4, m_{F''}=-1\rangle$$

(D.10) (D.11)
Figure D.4: Matrix elements squared $|\langle F'm_{F'} | \hat{J}_q | Fm_F \rangle |^2$ between different $F$ and $m_F$ states for 5D$_{5/2}$ manifold. Here the vertical arrows represent $|\langle F'm_{F'} | \hat{J}_z | Fm_F \rangle |^2$ matrix elements and diagonal arrows represent $|\langle F'm_{F'} | \hat{J}_- | Fm_F \rangle |^2$ matrix elements.

Figure D.5: Matrix elements squared between different $m_F$ states in the same $F$ level for 5D$_{5/2}$ manifold. Here the transitions represent $|\langle F'm_{F'} | \hat{J}_\pm | Fm_F \rangle |^2$. 
Table D.1: ac-Zeeman shift in 5D$_{5/2}$ manifold due to rf trapping currents.

<table>
<thead>
<tr>
<th>Magnetic field point (G)</th>
<th>Shift (Hz)</th>
<th>Magnetic field point (G)</th>
<th>Shift (Hz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.54</td>
<td>-1.68</td>
<td>0.40</td>
<td>0.79</td>
</tr>
<tr>
<td>0.70</td>
<td>-2.12</td>
<td>0.46</td>
<td>0.91</td>
</tr>
<tr>
<td>0.87</td>
<td>-2.59</td>
<td>0.51</td>
<td>1.04</td>
</tr>
<tr>
<td>1.03</td>
<td>-3.10</td>
<td>0.57</td>
<td>1.18</td>
</tr>
<tr>
<td>1.20</td>
<td>-3.65</td>
<td>0.62</td>
<td>1.31</td>
</tr>
<tr>
<td>1.36</td>
<td>-4.27</td>
<td>0.68</td>
<td>1.45</td>
</tr>
<tr>
<td>1.52</td>
<td>-4.98</td>
<td>0.73</td>
<td>1.60</td>
</tr>
<tr>
<td>1.68</td>
<td>-5.80</td>
<td>0.79</td>
<td>1.75</td>
</tr>
<tr>
<td>1.85</td>
<td>-6.78</td>
<td>0.84</td>
<td>1.90</td>
</tr>
<tr>
<td>2.01</td>
<td>-7.99</td>
<td>0.90</td>
<td>2.05</td>
</tr>
</tbody>
</table>

for positive magnetic fields, which neglects mixing with the $F'' = 1$, and $F'' = 2$ states. For the ac-Zeeman shift calculations the approximate form is a good enough representation for our measurement precision. The mixing angles $\theta_\pm$ are functions of the applied magnetic field with $\theta_\pm \in (\frac{\pi}{7}, \frac{\pi}{4})$ over the magnetic field range of 0.4 – 2 G explored in this work. In order to evaluate the shifts at a particular $B$-field to the energy of the corresponding mixed state we use the matrix elements listed in Figs. D.4 and D.5. The resulting shifts in the error are given in Table D.1.
Appendix E

Quadrupole shift

In this section, we describe an extra shift to the hyperfine intervals that came to our attention while the thesis was under examination. The shift is caused by static electric field gradients coupling to the quadrupole transition between the $6S_{1/2}$ manifold and the two $5D$ manifolds. The static gradients are due primary to the potential applied to the endcaps and as such can be related to trap frequency in the axial direction.

To describe the effect of the quadrupole shift we follow the treatment given in [136].

The Hamiltonian, $H_Q$, describes the interaction of external electric-field gradients with the atomic quadrupole moment. In the principal-axis frame it has the form

$$H_Q = -2A \Theta_0^{(2)'} + \sqrt{\frac{2}{3}} \epsilon A \left( \Theta_2^{(2)'} + \Theta_2^{(2)''} \right). \quad \text{(E.1)}$$

In the regime where the energy shifts due to $H_Q$ are small relative to the Zeeman shifts, which is normally the case, $H_Q$ can be treated as a perturbation. The matrix elements of the operators $\Theta_q^{(2)'}$ in the principal axis frame are given as

$$\langle \gamma J F m_F | \Theta_q^{(2)'} | \gamma J F m_F \rangle = \frac{(-1)^{F - m_F - q}}{\sqrt{(2F + 3)(2F + 2)(2F + 1)2F(2F - 1)}} f(A, \epsilon, \alpha, \beta) (\gamma J F m_F | \Theta | \gamma J F m_F),$$

where $D_{0-q}^{(2)s}(\omega)$ is defined in [136]. The energy shift to the $|F, m_F\rangle$ state is given by

$$\Delta E = \langle \gamma J F m_F | H_Q | \gamma J F m_F \rangle = \frac{-2(3m_F^2 - F(F + 1))}{[(2F + 3)(2F + 2)(2F + 1)2F(2F - 1)]^{1/2}} f(A, \epsilon, \alpha, \beta) (\gamma J F m_F | \Theta | \gamma J F m_F),$$

\footnote{Note that the author of [111] provides a similar treatment but in his thesis Eq. (7.41) differs by a factor of minus two compared with Eq. (44) of [136].}
where
\[ f(A, \epsilon, \alpha, \beta) = A((3 \cos \beta^2 - 1) - \epsilon \sin \beta^2(\cos \alpha^2 - \sin \alpha^2)), \]  
(E.4)

where \( \alpha \) and \( \beta \) are defined in [136]. For our trap geometry Eq. (E.4) reduces to \( \frac{1}{2} A \). In the center of the trap in the principal-axis frame the ion sees a potential [136]
\[ \Phi(x, y, z) = A[(x^2 + y^2 - 2z^2) + \epsilon(x^2 - y^2)], \]  
(E.5)

where the \( A \) parameter is given in terms of the axial trapping frequency as
\[ A = \frac{\omega_z^2 m}{4e}. \]  
(E.6)

Finally the matrix element \( (\gamma J F m_F || \Theta^{(2)} || \gamma J F m_F) \) can be expressed as
\[ (\gamma J F m_F || \Theta^{(2)} || \gamma J F m_F) = (-1)^{I+J+F}(2F+1) \left( \begin{array}{cc} J & 2 \\ F & I \end{array} \right) \left( \begin{array}{cc} J & 2 \\ -J & 0 \end{array} \right)^{-1} \Theta(\gamma, J), \]  
(E.7)

where \( \Theta(\gamma, J) \) is quadrupole moment of an atomic level \( |\gamma, J\rangle \). Here we use the quadrupole moments \( \Theta^{(2)}(5D_{3/2}) = 2.297ea_0^2 \) and \( \Theta^{(2)}(5D_{5/2}) = 3.397ea_0^2 \), which can be found in [137]. For the \( 5D_{3/2} \) and \( 5D_{5/2} \) measurements the trapping frequencies were \( \omega_z = 2\pi \times 403 \pm 1 \) and \( \omega_z = 2\pi \times 384 \pm 1 \) respectively, which leads to the shifts listed in Table E.1 and Table E.2.

<table>
<thead>
<tr>
<th>Table E.1: Quadrupole shifts for 5D_{3/2} states</th>
</tr>
</thead>
<tbody>
<tr>
<td>( F, m_F )</td>
</tr>
<tr>
<td>----------------</td>
</tr>
<tr>
<td>0, 0</td>
</tr>
<tr>
<td>1, 0</td>
</tr>
<tr>
<td>2, 0</td>
</tr>
<tr>
<td>3, 0</td>
</tr>
</tbody>
</table>

E.1 Quadrupole shift in 5D_{5/2} manifold

For the \( 5D_{5/2} \) manifold the calculation of the quadrupole shift is complicated by the fact that we measure the hyperfine intervals using the mixed states \(|+\rangle \) and \(|-\rangle \) defined in Eqs. (D.10) and (D.11) respectively. In order to calculate the perturbation to the
Table E.2: Quadrupole shifts for 5D\(3/2\) states

<table>
<thead>
<tr>
<th>(F, m_F)</th>
<th>Shift (Hz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,0</td>
<td>1.33</td>
</tr>
<tr>
<td>2,0</td>
<td>0.68</td>
</tr>
<tr>
<td>3,1</td>
<td>0.78</td>
</tr>
<tr>
<td>3,-1</td>
<td>0.78</td>
</tr>
<tr>
<td>4,1</td>
<td>1.4</td>
</tr>
<tr>
<td>4,-1</td>
<td>1.4</td>
</tr>
</tbody>
</table>

energy of these states due to static electric field gradients we need the off-diagonal elements

\[
\langle \gamma J F' m_F | H_Q | \gamma J F m_F \rangle = (-1)^{F'-m_F} \left( \begin{array}{cc} F & 2 \\ -m_F & m_F \end{array} \right) f(A, \eta, \alpha, \beta)(\gamma J F' m_F || \Theta^{(2)} || \gamma J F m_F),
\]

where

\[
(\gamma J F' m_F || \Theta^{(2)} || \gamma J F m_F) = (-1)^{I'+J'+F} \sqrt{(2F'+1)(2F+1)} \left( \begin{array}{cc} J & 2 \\ F & I \end{array} \right) \left( \begin{array}{cc} J & 2 \\ -J & 0 \end{array} \right)^{-1} \Theta(\gamma, J).
\]

For each magnetic point the intervals were measured at we can find the mixing angle and therefore the shift to the energies of the \(|+\rangle\) and \(|-\rangle\) states. Then using the approximate form, Eqs. (D.10) and (D.11), we can put the shift into the fit of the intervals and find the corresponding shift to the intervals. In the magnetic field range the intervals were measured, the \(|+\rangle\) state is shifted by 0.67 Hz to 0.48 Hz with increasing magnetic field and the \(|-\rangle\) state by 0.37 Hz to 0.47 Hz.

E.2 Conclusion

The quadrupole shift in the 5D\(3/2\) manifold has significant effect on the \(\delta W_1\) and \(\delta W_2\) hyperfine interval values, as the shift is larger than the total error for these intervals. For the hyperfine constants and the octupole moment for 5D\(3/2\) manifold the quadrupole shift has negligible effect due to the accuracy being limited by uncertainties in the
nuclear structure calculations. In the 5D_{5/2} manifold the shift is insignificant as the shift to each interval is \( \approx 0.5 \) Hz, which is much smaller than either the statistical or systematic uncertainties on the hyperfine intervals.
References


[73] [http://www.innolume.com/products/Gain-chips.htm](http://www.innolume.com/products/Gain-chips.htm).


