Observation of singly charged barium ions in a buffer gas

Towards a functional barium-tagging system for use in
the Enriched Xenon Observatory

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Abstract

The observation of the rare neutrinoless-double-beta-decay will have major implications for both neutrino physics and particle physics as a whole. In particular, a positive observation will confirm the Majorana nature of the neutrino and demonstrate that lepton number is not a strictly conserved quantity. Furthermore, a measurement of the half-life of the decay will lend itself to a measurement of the absolute mass scale of the neutrino. In this work, the sensitivity to observing the neutrinoless-double-beta-decay of ^{136}Xe is analyzed for the EXO experiment, and the benefits of the inclusion of a barium-tagging system in a future tonne-scale detector are demonstrated. In addition, the sensitivity to single barium ion identification is analyzed for two different methods of observation: continuous excitation and intermodulation. The intermodulation technique was determined to be the more sensitive as single barium ions were observed with a relatively high signal-to-noise ratio of ~ 4 for a one second observation time. Single ions were not observed using the continuous excitation technique.

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Chapter 1

Introduction

Results from neutrino oscillation experiments showing that neutrinos are massive constitute the first experimental evidence inconsistent with the Standard Model of particle physics (SM). In order to guide and develop new theories that take massive neutrinos into account, the absolute mass scale and the answer to the question of whether or not the neutrino is its own antiparticle will need to be determined. Up to now it has been impossible to directly measure the mass scale of the neutrino, although, neutrino oscillation data [1] and cosmological considerations [2] suggest neutrino masses on the order of 0.01-0.1 eV. This small mass makes the whole task of measuring them a rather daunting, yet highly rewarding one as well. Indeed, one only needs to look at the multitude of neutrino mass experiments currently running, under construction, or being planned across the world to appreciate the importance of a measurement such as this.

Meeting the challenges of a neutrino mass measurement demands the introduction of entirely new experimental techniques in the fields of radiation detection, isotopic enrichment, and radioactive-background reduction, to name a few. A leader in the development of such novel techniques has been the Enriched Xenon Observatory (EXO) experiment – a xenon scintillation detector with the aim of measuring the rare neutrinoless-double-beta-decay ($0\nu\beta\beta$ -decay) of ^{136}Xe . If this reaction is observed, the half-life of the decay

will give a measurement of the neutrino mass scale, as well as imply that the neutrino is its own antiparticle. The best current limit on this decay puts the half-life $> 10^{25}$ years, implying that only a handful of $0\nu\beta\beta$ events are expected over the lifetime of the experiment. For this reason, the reduction of the natural radioactive backgrounds – all of which can obscure or fake a $0\nu\beta\beta$ event – is of paramount importance to the success of the experiment. The focus of this work was to aid in the development of a new background-reduction technique only available to a xenon detector – the tagging via laser induced fluorescence of the daughter barium ion produced in the $0\nu\beta\beta$ decay [3]. Specifically, a system to trap and detect single barium ions was set up and characterized with a focus on maximizing the sensitivity to single-ion observation.

The structure of the thesis is as follows. In Chapter 2 the theory and properties of the neutrino are presented. I start with an overview of the history of the neutrino beginning with its conception over 80 years ago, and leading up to the neutrino oscillation experiments of the 1990s and early 2000s. The theory of double-beta decay is then presented and the importance of a $0\nu\beta\beta$ -decay measurement is established.

In Chapter 3 the experiments searching for the $0\nu\beta\beta$ -decay are discussed. A large portion of my work in parallel with the barium-tagging work was to calculate the sensitivity and discovery potential for the EXO experiment. For this reason I go into some detail about how the sensitivity and discovery potential are calculated and which experimental factors influence them the most. Following this is a detailed introduction to the EXO-200 experiment which is the currently operating prototype detector for the EXO collaboration. After presenting the results of the sensitivity analysis for the EXO-200 detector I discuss a proposed tonne-scale upgrade to EXO-200 - the nEXO detector. I end the chapter by introducing the barium-tagging concept and demonstrating, through the discovery potential of nEXO, the benefits of including the barium-tagging technique in the future detector. This will solidify the motivation for the rest of the thesis.

In the next three chapters I outline the theory needed to understand the trapping and

identification of barium ions. In Chapter 4 the theory of charged particle confinement is presented, with a focus on what are called “linear Paul traps” - the type of trap used in this work. In Chapter 5 the theory of laser induced fluorescence is presented and the quantum dynamics of the barium ion are investigated. Finally, in Chapter 6 I give some of the practical considerations of detecting barium ions via laser induced fluorescence and introduce two broad methods of barium ion observation - continuous excitation and intermodulation.

In Chapter 7 the experimental setup used to detect the barium ions is introduced, and in Chapter 8 the measurements of barium ion fluorescence are presented. The conclusions are summarized in Chapter 9, and a brief review of the statistical techniques used is given in Appendix A.

Chapter 2

Neutrinos

2.1 History of neutrinos

2.1.1 Conception

The neutrino stands out among the subatomic particles as the most mysterious. It has been dogged by controversies from its conception nearly a hundred years ago continuing right up until the present time. The first controversy, the one that gave birth to the neutrino, was termed the “energy crisis” and stemmed from the observation that the energy spectrum of electrons emitted in beta decays of pure substances were continuous. At the time, only three subatomic particles were known to physicists - the electron, proton, and photon - and thus the continuous beta decay spectrum posed a serious problem for the seemingly sacred law of energy conservation. The argument was as follows: in beta decay an atomic nucleus X emits an electron e^- and subsequently is converted into the nucleus of a different element Y ,

$$X \rightarrow Y + e^- \tag{2.1.1}$$

In doing so it loses energy equal to the mass difference $\Delta M = M_X - M_Y$ where M_X and M_Y are the nuclear masses before and after the decay respectively. For energy to be conserved the electron must therefore be emitted with a kinetic energy $K_e = \Delta M - m_e$. Since ΔM is constant for a given initial nucleus then so too should be K_e (note that K_e is termed the “Q-value” of the decay).

Thus, when C. Ellis and W. Wooster published their results in 1927 [4] conclusively showing that the electron spectrum emitted from a beta decay was in fact continuous, it seemed that energy conservation did not apply here. Wolfgang Pauli, then a professor in theoretical physics at the Federal Institute of Technology, was driven to a rather less drastic conclusion and in 1930 predicted the existence of a new, neutral particle that also existed in the nucleus and which he termed a “neutron”. This new particle would also be emitted in β -decays and thus provide an explanation for the energy crisis.

At first Pauli’s idea was considered only as a remote possibility, even himself regarding it as a “desperate way out”. However, it was only a few years later that the neutrino was theoretically established with Enrico Fermi’s famous theory on beta decay [5]. In his classic paper, Fermi shows that beta decay is specifically a reaction that converts a neutron, n , into a proton, p , with the release of an electron and an (anti)neutrino,

$$n \rightarrow p^+ + e^- + \bar{\nu} \quad (2.1.2)$$

A consequence of Fermi’s theory is that the shape of the β -decay spectra - especially the high-energy tail near the Q-value¹) depends on the mass of the neutrino. This then provided a means to predict the neutrino’s mass, and indeed, Fermi used this to conclude that the neutrino had to have a zero or very small mass (as compared to the electron).

¹The Q-value is the difference in mass-energy between the initial particles and final particles in a nuclear decay - for the case of β -decay it represents the maximum kinetic energy attainable by the electron.

2.1.2 Discovery

Another implication of Fermi's theory, pointed out by the theorists Hans Bethe and Rudolf Peierls, was that the inverse reaction to β -decay should also occur. That is, an anti-neutrino, $\bar{\nu}$, is absorbed by a proton converting it into a neutron with the release of a positron, e^+ ,

$$\bar{\nu} + p^+ \rightarrow n + e^+ \quad (2.1.3)$$

They calculated that the cross section for this process should be on the order of 10^{-44} cm² [6], and, not surprisingly, concluded that it would be essentially impossible to ever detect a neutrino via inverse β -decay... what they did not foresee was just how abundant the neutrino was.

With the advent of the atomic bomb during World War II it became known to physicists that the nuclear fission of uranium was a source of a huge number of neutrinos. Fred Reines and Clyde Cowan, having worked on the atomic bomb project, knew of this abundant neutrino source and devised a plan to place a detector capable of observing the inverse β -decay reaction next to a nuclear reactor². A backbone to their idea lay in the unique signature that should accompany such a reaction - two 511 keV γ -rays followed by a neutron capture γ -ray an average of t_0 seconds later³. This would allow them to untangle their signal from the myriad of other background processes that would be occurring so close to a nuclear reactor.

The detector they used was a large tank of liquid-scintillator instrumented with photomultiplier tubes (PMTs). The scintillator was proton rich and was thus used as both source and detector. Cadmium chloride was added to increase the probability that the

²Originally they were going to set an atomic bomb off beside the detector but deemed that the reactor would be much simpler.

³The positron produced in inverse β -decay almost instantly annihilates with an electron producing two 511-keV γ -rays. The neutron, after bouncing around for some characteristic amount of time, t_0 , will be captured by a nucleus also emitting a high energy gamma ray.

neutron be captured within their detector⁴. The final apparatus weighed around 10 tonnes and was placed a mere 11 meters from the core of the Savannah River reactor in South Carolina. In 1956, after just over a year of running and 100 days of data collected, Reines and Cowan had definitively detected the first neutrinos [7] with a measured cross section very close to that calculated by Bethe and Peierls. Finally, 20 years after it was first postulated, the neutrino had been observed.

2.1.3 Properties

Over the next few decades major advancements in the area of neutrino physics were being made. A first hint at parity violation was discovered in 1956 by Tsung-Dao Lee and Chen-Ning Yang at the Berkeley National Laboratory (BNL) in the decays of neutral kaons [8]. This was later confirmed in 1957 by Chien-Siung Wu - having teamed up with Lee and Yang - in her famous experiment on the beta decay of a ^{60}Co nuclei [9]. Lee and Yang believed that the neutrino was the culprit behind their observed parity violation and published a paper later that year showing that parity violation naturally arose if the neutrino were massless [10]. Only one year later at BNL the neutrino was determined to be left-handed [11].

A few years later, again at BNL, the neutrino was determined to come in two flavours - one associated with muons and one associated with electrons [12]. Furthermore, the discovery of the tau particle [13] and the full development of the electro-weak theory provided strong evidence for another neutrino associated with the tau. The primary evidence for *three* neutrinos came in 1989 from precision measurements of the Z-boson lifetime⁵ at the Large Electron Positron collider (LEP). The combined results of the four LEP experiments (ALEPH, DELPHI, L3 and OPAL) puts the number of neutrinos at $N_\nu = 2.984 \pm 0.008$ [14]. The tau neutrino was finally observed in 2001 by the DONUT

⁴Cadmium has a relatively large neutron capture cross section.

⁵The lifetime of the Z-boson depends on the number of decay paths it has available to it and thus depends on the number of neutrinos that exist. The measured lifetime agreed perfectly with the three neutrino assumption.

experiment [15].

2.1.4 Dirac or Majorana

There is another question about the neutrino that scientists would like to know but have not answered yet - is the neutrino its own antiparticle? The question was raised late in the 1930's by a young Italian physicist, Ettore Majorana, in a paper he published [16] in competition for a university chair position. In his now famous paper, Majorana develops a theory akin to Dirac's where he represents neutral particles as their own antiparticles. These particles can still be left- or right-handed but do not represent distinct particles as in the case of electrons and positrons. Naturally, the neutrino is a good candidate for this type of particle. Early experiments to try and decide if nature prefers the neutrino to be Dirac or Majorana did not take into account parity violation and thus failed to say anything towards this problem. Still the question has not been answered, however, there is hope in one certain type of experiment aiming to observe a decay, that, paradoxically, does not produce any neutrinos.... but more on this later.

2.2 Neutrino oscillations

At this point in the history of the neutrino we know that the neutrino is very lightweight, it is left-handed, and it comes in three flavours: the electron neutrino (ν_e), the muon neutrino (ν_μ), and the tau neutrino (ν_τ). It also may or may not be of Majorana type. As of today we do know that at least two neutrinos must have some mass from observations of a phenomenon only available to a handful of particles - particle oscillation. The story of neutrino oscillations begins with another controversy of which the neutrino played an integral role - the solar neutrino problem.

2.2.1 Solar neutrino problem

In the 1930's Charles Critchfield and Hans Bethe developed a mechanism by which the sun generates its power. The so called proton-proton mechanism (pp-mechanism for short) converts hydrogen into helium, releasing energy in the process. The process can proceed along three main paths:

PP-I Chain

PP-II Chain

PP-III Chain

The fluxes of neutrinos produced in these reactions were calculated by John Bahcall [17, 18] who was prompted to do so by a request from an experimental physicist, Raymond Davis. Davis had been working on an experiment first proposed by Bruno Pontecorvo to detect anti-neutrinos from a nuclear reactor. The idea was based on the capture of a neutrino on a ${}^{37}\text{Cl}$ isotope thereby converting it to ${}^{37}\text{Ar}$,

By collecting and counting the number of ${}^{37}\text{Ar}$ isotopes the number of neutrino interactions could be deduced. For the above reaction to proceed, however, the neutrino must have an energy greater than ~ 1 MeV. The pp-neutrinos (Equation 2.2.1) have a maximum energy of about 0.3 MeV and will thus be useless in such an experiment. However, the neutrino produced in the boron decay (Equation 2.2.8) emits a relatively high-energy neutrino and is thus the reaction of choice.

In 1964, having now teamed up with Bahcall, Davis proposed a chlorine type experiment which would definitively detect neutrinos produced in the sun's interior. Housed at the Homestake Gold Mine in South Dakota, the detector contained 38000 liters of carbon-tetrachloride which would serve as the chlorine target. The first results were published in 1968 [19] and provided an upper limit on solar neutrinos about 2-3 times lower than what was predicted by Bahcall. It was not long after this that the detection of solar neutrinos, and therefore the existence of the pp-chain of reactions, was confirmed [20]. The number of neutrinos were still being produced in much lower numbers than what the solar model predicted and this discrepancy between theory and experiment became known as the “solar neutrino problem”.

The same year that Davis' first results were published, Pontecorvo proposed that the solar neutrino problem could be explained if neutrinos could oscillate between different flavours [21]. Particle oscillation had been predicted to exist in the neutral kaon system by Gell-Mann and Pais in 1955 [22] and was later confirmed in experiment by Leon Lederman in 1956 [23]. This led Pontecorvo to argue that, if neutrinos could oscillate, then a large fraction of the electron neutrinos coming from the sun could turn into a muon or tau neutrino before reaching Earth. As these flavours would not interact in Davis' detector this would explain the observed deficit.

The first strong evidence for neutrino oscillations came in 1998 from the Super Kamiokande collaboration who observed oscillations in neutrinos produced from cosmic ray interactions in the atmosphere [24]. In 2001, the Sudbury Neutrino Observatory (SNO) definitively measured oscillations in neutrinos coming from the sun [25] - finally laying to rest the solar neutrino problem⁶. Since then, a series of other oscillation experiments have confirmed the existence of neutrino oscillations in neutrinos produced in reactors, the atmosphere, and particle accelerators (KamLAND, LSND, OPERA, T2K...) [26, 27, 28, 29]. Perhaps a more important consequence of observing neutrino oscillations, as will soon be apparent, was that experimenters were finally able to say with confidence that (at least two) neutrinos did in fact have a non-zero mass.

2.2.2 Theory

For neutrino oscillations to occur in nature there must exist a difference between the neutrino's flavour eigenstates (ν_e , ν_μ , ν_τ) and its mass eigenstates (ν_1 , ν_2 , ν_3). The flavour eigenstates are the particles that actually interact with things and are what we observe in a detector. The mass eigenstates are those states that actually propagate through space and have a well defined mass. The flavour and mass eigenstates are not

⁶The SNO experiment was special in particular since it was not only able to measure electron neutrino flux, but also the entire neutrino flux (ν_e , ν_μ , and ν_τ). This allowed SNO to show that although the electron neutrino flux is lower than predicted by Bahcall's theory, the total neutrino flux is almost exactly equal to his prediction.

independent, however, as they are orthogonal superpositions of each other, related via,

$$\nu_\alpha = \sum U_{\alpha,i}^* \nu_i \quad (2.2.10)$$

where $U_{\alpha,i}$ is a matrix element of the Pontecorvo-Maki-Nakagawa-Sakata (PMNS) matrix [30]. This matrix is parameterized as,

$$U_{PMNS} = \begin{bmatrix} c_{12}c_{13}e^{\frac{i\phi_1}{2}} & c_{13}s_{12} & s_{13}e^{-i\delta} \\ -s_{12}c_{23} - c_{12}s_{23}s_{13}e^{i\delta} & (c_{12}c_{23} - s_{12}s_{23}s_{13}e^{i\delta})e^{\frac{i\phi_2}{2}} & s_{23}c_{13} \\ s_{12}s_{23} - c_{12}c_{23}s_{13}e^{i\delta} & -c_{12}s_{23} - s_{12}c_{23}s_{13}e^{i\delta} & c_{23}c_{13} \end{bmatrix} \quad (2.2.11)$$

where c_{ij} (s_{ij}) is taken to mean $\cos \theta_{ij}$ ($\sin \theta_{ij}$). The PMNS matrix is thus seen to depend on three mixing angles (θ_{12} , θ_{13} , θ_{23}) and one CP-violating phase, δ , if the neutrino is of Dirac type, and two additional CP-violating phases, ϕ_i , if the neutrino is of Majorana type.

The PMNS matrix determines the amount of mixing between the states, however, neutrino oscillation also depends on the squared mass-differences between the mass eigenstates⁷, Δm_{21}^2 and Δm_{32}^2 where,

$$\Delta m_{ij}^2 = \Delta m_i^2 - \Delta m_j^2 \quad (2.2.12)$$

To see how, consider the survival probability - the probability that a neutrino produced in some flavour α remains in that flavour after some time t . In the case of only two neutrinos the survival probability takes the simple form [31],

$$P_{\alpha\alpha} = 1 - \sin^2 \theta \sin^2 \left(1.27 \frac{L}{E} \Delta m^2 \right) \quad (2.2.13)$$

where L is the distance travelled by the neutrino in km, E is the neutrino's energy in GeV,

⁷There is a third mass difference Δm_{31}^2 but it is not independent since $\Delta m_{31}^2 = \Delta m_{32}^2 - \Delta m_{21}^2$.

Table 2.1: Experimental measurements of the neutrino mixing parameters. The values are taken from a global analysis of all neutrino oscillation experiments [1]. The parameters Δm_{31}^2 , θ_{23} , and θ_{13} depend on the choice of mass hierarchy (NH for normal, IH for inverted) assumed in the analysis and thus have two different values quoted.

Parameter		Measurement
$\Delta m_{21}^2 (\text{eV}^2)$		$7.54^{+0.26}_{-0.22} \times 10^{-5}$
$\Delta m_{31}^2 - \frac{1}{2}\Delta m_{21}^2 (\text{eV}^2)$	NH	$2.43 \pm 0.06 \times 10^{-3}$
$\Delta m_{31}^2 + \frac{1}{2}\Delta m_{21}^2 (\text{eV}^2)$	IH	$2.38 \pm 0.06 \times 10^{-3}$
$\sin^2 \theta_{12}$		0.308 ± 0.017
$\sin^2 \theta_{23}$	NH	$0.437^{+0.033}_{-0.023}$
	IH	$0.455^{+0.039}_{-0.031}$
$\sin^2 \theta_{13}$	NH	$0.0234^{+0.0020}_{-0.0019}$
	IH	$0.0240^{+0.0019}_{-0.0022}$

θ is the two-neutrino mixing angle, and Δm^2 is the squared mass difference between the two neutrino states in eV^2 . One thing that is immediately apparent is that if $\Delta m^2 = 0$ then oscillations do not occur. This result holds in the case of three neutrinos.

Unfortunately, neutrino oscillation experiments are not able to determine the absolute mass scale of the neutrino or the sign of Δm_{31}^2 . They can, however, put constraints on the allowed neutrino mass parameter-space using the best estimates for the neutrino mixing parameters (given in Table 2.1 for reference). In the next chapter these parameters are used in just this way (see Figure 3.2.1).

2.2.3 Mass hierarchy

The fact that Δm_{21}^2 is positive implies that the ν_2 mass eigenstate is heavier than the ν_1 mass eigenstate. Unfortunately, however, neutrino oscillation experiments are unable to determine the sign of the mass splitting Δm_{32}^2 and thus the mass hierarchy of the neutrino sector is not known. The knowledge that $m_{\nu_2} > m_{\nu_1}$ allows us to narrow the different orderings in mass to three different possibilities. The first, the normal hierarchy, has $m_{\nu_3} > m_{\nu_2} > m_{\nu_1}$ and requires that $\Delta m_{32}^2 > 0$. The second possibility, the inverted

Figure 2.2.1: Graphical depiction of the normal and inverted neutrino mass hierarchies. The shading of the bars represents the approximate flavour content of the neutrino mass states with the electron flavour as white, muon as gray, and tau as black.

hierarchy⁸, has $m_{\nu_2} > m_{\nu_1} > m_{\nu_3}$ and would require $\Delta m_{32}^2 < 0$. The last possibility, which is not really independent of the others, is the degenerate hierarchy which occurs if the absolute neutrino mass scale is on the order of 0.1 eV. In this situation the mass splittings are essentially negligible as compared to the masses. A graphical depiction of the normal and inverted hierarchies is shown in Figure 2.2.1.

2.2.4 Mass

The mass of the neutrino is much harder to measure than the properties already mentioned. This is primarily due to the fact that the neutrino interacts so weakly with regular matter but also because its mass must be very small. Original attempts at measuring the neutrino's mass relied on the same idea as Fermi had used to infer that the neutrino mass must be small - compare the spectra of electrons produced in β -decays and look for small deviations from theory in the region near the Q-value.

The main issue with this procedure is purely statistics - the probability for an electron to have an energy near the Q-value can be many orders of magnitude smaller than the probability at the peak of the distribution. Experimenters are thus forced to use highly radioactive β -decay sources, however, the choice of source also matters since the proportion of electrons in the endpoint region increases with decreasing Q-value of the

⁸It is called the inverted hierarchy due to the fact that ν_e , which is composed of almost no ν_3 , would not be the lightest eigenstate and so the hierarchy is inverted as compared to the charged leptons.

source. For this reason experimenters use tritium - a neutron rich form of hydrogen that β -decays with a half-life of 12.3 years and has an unusually low Q-value of 18.6 keV.

In 1972, an experiment of this kind was conducted by Karl-Erik Bergkvist at the University of Stockholm in which an upper limit was placed on the neutrino mass of 60 eV [32]. Later experiments would improve on this technique bringing the limit to less than 9.4 eV by the year 1990 [33]. The most sensitive β -ray spectrometers to date are the MAC-E (Magnetic Adiabatic Collimation combined with an Electrostatic filter) type with current limits on the neutrino mass of < 2.5 eV and < 2.3 eV produced by the Troitsk [34] and Mainz [35] neutrino mass experiments respectively. A planned experiment, KATRIN [39, 40] is a giant MAC-E type spectrometer that has a predicted sensitivity of 0.2 eV and plans to start collecting data sometime in the year 2016.

Investigating the endpoint of the β -decay spectrum is perhaps the simplest method of weighing the neutrino as it is direct and model independent. It is, however, not the only method. A limit of < 5.7 eV (95% CL) [41] has been placed by measuring the energy dependence in the arrival times of neutrinos produced in the supernova SN1987a. Cosmology can also say something about the neutrino mass - specifically the sum of the three neutrino masses - due to the effect neutrinos have on large scale structures in the universe. The best current limit from this method is $\sum m_\nu < 0.18$ eV (95% CL) [2]. The last method, and the most important for this work, depends on the measurement of the half-life of the $0\nu\beta\beta$ -decay of a candidate isotope. This is discussed in detail in the following section.

2.3 Double-beta decay

The single β -decay has already played a vital role in the history of the neutrino, among other things, leading to its discovery. For the future, physicists put their hopes in another, more exotic form of β -decay to finally put an end to some of the major questions

unanswered in the world of the neutrino - is the neutrino its own antiparticle, and what is the absolute mass scale of the neutrino?

2.3.1 Two-neutrino-double-beta decay

The two-neutrino-double-beta-decay ($2\nu\beta\beta$ -decay) is a standard model (SM) allowed second-order weak process in which two regular β -decays essentially occur simultaneously increasing the charge number of the decaying nucleus by two units, $M_A(Z, A) \rightarrow M_A(Z+2, A)$. The decay was first hypothesized in 1935 by Maria Goeppert-Mayer [36] and has since been confirmed to exist by experiment in a number of different isotopes.

In theory, $2\nu\beta\beta$ -decay will proceed in any nucleus in which two simultaneous β -decays are energetically allowed. However, due to the fact that single beta decays are much more likely to occur, it would not be feasible in an experiment to separate the genuine $2\nu\beta\beta$ -decays from two coincident normal β -decays occurring in the same region. Thus, to observe $2\nu\beta\beta$ -decays, one must choose a nucleus in which the β -decay is energetically forbidden and the $2\nu\beta\beta$ -decay is not.

In general, whether a nucleus undergoes a weak decay depends on the dependence of its mass, M_A , on its charge, Z . This dependence can be approximated by a parabolic function [37],

$$M_A(Z, A) = \text{const} + 2b_{\text{sym}} \frac{(A/2 - Z)^2}{A^2} + b_{\text{coul}} \frac{Z^2}{A^{1/3}} + Zm_e + \delta \quad (2.3.1)$$

where A is the mass number, Z is the charge number, $b_{\text{sym}} \sim 50$ MeV is a coefficient depending on symmetry considerations, $b_{\text{coul}} \sim 0.7$ MeV represents a coulombic energy term, and m_e is the electron mass. The value of δ depends on the relative number of neutrons to protons in the nucleus and can be roughly approximated by,

$$\delta = \begin{cases} \frac{+12}{A^{1/3}} & \text{for odd } Z, \text{ even } A \\ 0 & \text{for odd } A \\ \frac{-12}{A^{1/3}} & \text{for even } Z, \text{ even } A \end{cases} \quad (2.3.2)$$

As a result, nuclei with odd A will typically have only one stable isotope. This in turn implies that unstable nuclei will decay via β -decay if its charge is smaller than the stable nuclei, or via positron emission or electron capture if its charge is larger than the stable nuclei. For the case of even A , due to the pairing term δ , there will be two possibilities depending on whether Z is odd or even. This results in the typical case that there exist two even-even nuclei that are less massive than their nearest neighbour odd nuclei, and are thus stable against the first order weak decays ($\beta^\pm$ -decay, or electron capture). Since these isotopes do not generally have the same mass, the heavier can decay into the lighter via $2\nu\beta\beta$ -decay. Thus, experimental observation of $2\nu\beta\beta$ -decay is only available in even-even nuclei.

The half-life of the $2\nu\beta\beta$ -decay, $T_{1/2}^{2\nu}$, can be calculated theoretically via [37],

$$[T_{1/2}^{2\nu}]^{-1} = G^{2\nu} [M^{2\nu}]^2 \quad (2.3.3)$$

where $G^{2\nu}(E_0, Z)$ is the leptonic phase space factor, E_0 is the total energy available in the decay, and $M^{2\nu}$ is a nuclear matrix element (NME). The phase space factor, $G^{2\nu}$, can be calculated exactly for a given isotope (values listed in Table 2.2), however, the nuclear matrix element, $M^{2\nu}$, can only be approximated using theory and thus will contain some level of uncertainty in its evaluation.

2.3.2 Neutrinoless-double-beta decay

In 1939, Wendell Furry realized that $\beta\beta$ -decay may proceed without the release of two neutrinos provided that the neutrinos are Majorana particles [38]. The decay proceeds

via the so-called “Racah sequence” - a two step process where the neutrino emitted in the first step and absorbed in the second step is virtual,

$$n \rightarrow p + e^- + \nu' \quad (2.3.4)$$

$$\nu' + n \rightarrow p + e^- \quad (2.3.5)$$

As the first step emits an anti-neutrino and the second step⁹ is initiated with a neutrino, this process will only occur if the neutrino and anti-neutrino are in fact the same particle, i.e., are Majorana in nature. Thus, it is generally accepted that any positive observation of $0\nu\beta\beta$ -decay implies that the neutrino is a Majorana particle. It is also worth mentioning that an observation will also imply lepton non-conservation since there are two leptons (electrons) present in the final state and no leptons in the initial state.

As we know from parity violation, the condition that the neutrino is of Majorana type is not enough to ensure the reaction proceeds as stated above. This is due to the fact that the neutrino emitted in the first step is right-handed while a left-handed neutrino is necessary to induce the second step. If the neutrino were massless then there is nothing more that can be done, however, if the neutrino is massive, then the neutrino emitted in the first step will contain a small left-handed component. One important consequence of this is that the rate at which the decay proceeds will depend on the amplitude of the left-handed component, which in turn is proportional to the neutrino’s mass. It is in this way that the $0\nu\beta\beta$ -decay can be used as an experimental probe for the mass of the neutrino.

The $0\nu\beta\beta$ -decay half-life, $T_{1/2}^{0\nu}$, can be calculated via [37],

⁹This can be thought of as an induced β -decay and is the reaction used by Davis in his famous ^{37}Cl experiment.

$$[T_{1/2}^{0\nu}]^{-1} = G^{0\nu}(E_0, Z) [M^{0\nu}]^2 \frac{\langle m_{\beta\beta} \rangle^2}{m_e^2} \quad (2.3.6)$$

where the parameters are as defined above for the $2\nu\beta\beta$ half-life. The parameter $\langle m_{\beta\beta} \rangle$ is an effective Majorana-neutrino mass term that depends on the neutrino mixing parameters via,

$$\langle m_{\beta\beta} \rangle = \sum U_{e,i}^2 m_i \quad (2.3.7)$$

where m_i represents the mass of the i^{th} neutrino mass eigenstate. As for the $2\nu\beta\beta$ case, the phase-space factor, $G^{0\nu}$, can be calculated to whatever precision is required (values listed in Table 2.2). On the other hand, the nuclear matrix element, $M^{0\nu}$, will involve some level of uncertainty in its calculation.

2.3.3 Double-beta decay energy spectra

From the last section we can see that an observation of $0\nu\beta\beta$ -decay simultaneously implies that the neutrino is a Majorana particle, the neutrino is massive, and lepton number is not a strictly conserved quantity. In addition, if one is able to measure the decay half-life then the absolute neutrino mass can also be inferred. It should come as no surprise that the search for $0\nu\beta\beta$ -decay is a top priority in physics today.

Current limits place the $\beta\beta$ -decay half-life greater than 10^{22} to 10^{25} yrs, depending on the isotope in question, and thus any positive measurement constitutes an enormous experimental challenge. Natural radioactive isotopes such as ^{238}U and ^{232}Th can easily mask the wanted signal and thus attention to component purity and detector location are of primary importance. In addition, even if one were able to perfectly rid the detector of radioactive contamination, one would still have to contend with the much more likely $2\nu\beta\beta$ -decay which cannot be reduced. In order to get around this, one is forced to measure the energy spectrum of the two electrons emitted.

The $2\nu\beta\beta$ -decay will give a continuum spectrum, similar to a regular β -decay, since the emitted neutrinos can carry away any energy the electrons do not. The shape of the spectrum can be represented analytically if one uses the Primakoff-Rosen approximation [37],

$$\frac{dN_{2\nu}}{dK} \sim K(T_0 - K)^5 \left(1 + 2K + \frac{4K^2}{3} + \frac{K^3}{3} + \frac{K^4}{30} \right) \quad (2.3.8)$$

where K is the sum of the kinetic energies of the two electrons, and T_0 is the decay Q-value in units of the electron mass. For the $0\nu\beta\beta$ case the sum electron spectrum is simply a δ -function located at the Q-value (the summed kinetic energy of the electrons must equal the Q-value),

$$\frac{dN_{0\nu}}{dK} \sim \delta(K - T_0) \quad (2.3.9)$$

The two functions 2.3.8 and 2.3.9 are plotted in Figure 2.3.1. In reality, the detector will have finite resolution and the 0ν peak will not be as sharp. More realistic spectra are also shown in Figure 2.3.1 where the $0\nu\beta\beta$ peak has been smeared out by assuming various energy resolutions. From this plot it is clear that the energy resolution of the detector will have a large effect on the separation between the $0\nu\beta\beta$ - and $2\nu\beta\beta$ -decays.

A list of the known $0\nu\beta\beta$ -decay candidate isotopes are given in Table 2.2 along with their Q-values, natural abundances, and phase space factors for both the 0ν and 2ν decays. Table 2.3 lists calculated nuclear matrix elements for the $0\nu\beta\beta$ a selection of the isotopes in Table 2.2. The nuclear matrix elements in Table 2.3 are calculated using two different approximations: the microscopic interacting boson model (IBM-2) [42], and the quasi-random-phase-approximation (QRPA) [43].

Figure 2.3.1: Typical sum-electron energy spectra for $2\nu\beta\beta$ - and $0\nu\beta\beta$ -decays for a Q -value of 2458 keV assuming: a) perfect energy resolution, and b) 0.5 and 5.0% energy resolutions. The 2ν spectrum is calculated using the Primakoff-Rosen approximation (Equation 2.3.8). In all cases the 0ν spectra are normalized to 1/1000 of the 2ν . The independent parameter is the electron kinetic energy divided by the Q -value.

Table 2.2: $0\nu\beta\beta$ -decay candidate isotopes. The end-point energies and phase-space factors are taken from reference [44]. The natural abundances are taken from reference [45].

Isotope	Abundance (%)	Q -value (keV)	$G^{2\nu}$ (10^{-21}yr^{-1})	$G^{0\nu}$ (10^{-15}yr^{-1})
$^{48}\text{Ca} \rightarrow ^{48}\text{Ti}$	0.187	4272.26	40330	64.34
$^{76}\text{Ge} \rightarrow ^{76}\text{Se}$	7.83	2039.061	124.9	6.128
$^{82}\text{Se} \rightarrow ^{82}\text{Kr}$	8.73	2995.12	4139	26.35
$^{96}\text{Zr} \rightarrow ^{96}\text{Mo}$	2.80	3350.37	17676	53.37
$^{100}\text{Mo} \rightarrow ^{100}\text{Ru}$	9.67	3034.4	8579	41.28
$^{110}\text{Pd} \rightarrow ^{110}\text{Cd}$	11.72	2017.85	357.1	12.49
$^{116}\text{Cd} \rightarrow ^{116}\text{Sn}$	7.49	2813.5	7168	43.31
$^{124}\text{Sn} \rightarrow ^{124}\text{Te}$	5.79	2286.97	1434	23.44
$^{128}\text{Te} \rightarrow ^{128}\text{Xe}$	31.74	865.87	0.6971	1.524
$^{130}\text{Te} \rightarrow ^{130}\text{Xe}$	34.08	2526.97	3965	36.88
$^{136}\text{Xe} \rightarrow ^{136}\text{Ba}$	8.858	2457.83	3716	37.81
$^{148}\text{Nd} \rightarrow ^{148}\text{Sm}$	5.7	1928.75	842.3	26.19
$^{150}\text{Nd} \rightarrow ^{150}\text{Sm}$	5.6	3371.38	94470	163.5
$^{154}\text{Sm} \rightarrow ^{154}\text{Gd}$	22.75	1215.03	24.87	7.819
$^{160}\text{Gd} \rightarrow ^{160}\text{Dy}$	21.86	1729.69	502.6	24.79
$^{198}\text{Pt} \rightarrow ^{198}\text{Hg}$	7.163	1047.17	39.83	19.59
$^{232}\text{Th} \rightarrow ^{232}\text{U}$	100	842.15	29.33	36.12
$^{238}\text{U} \rightarrow ^{238}\text{Pu}$	99.2742	1144.98	37.78	87.16

Table 2.3: Nuclear matrix elements for the $2\nu\beta\beta$ - and $0\nu\beta\beta$ -decays. The nuclear matrix elements calculated using the IBM-2 method are from reference [42] and those calculated using the QRPA method are from reference [43].

Isotope	$M^{0\nu}$ (dimensionless)	
	IBM-2	QRPA
$^{48}\text{Ca} \rightarrow ^{48}\text{Ti}$	2.82	0.871
$^{76}\text{Ge} \rightarrow ^{76}\text{Se}$	7.54	8.31
$^{82}\text{Se} \rightarrow ^{82}\text{Kr}$	6.01	7.47
$^{96}\text{Zr} \rightarrow ^{96}\text{Mo}$	4.56	4.38
$^{100}\text{Mo} \rightarrow ^{100}\text{Ru}$	6.80	8.70
$^{110}\text{Pd} \rightarrow ^{110}\text{Cd}$	6.52	9.28
$^{116}\text{Cd} \rightarrow ^{116}\text{Sn}$	4.99	6.51
$^{124}\text{Sn} \rightarrow ^{124}\text{Te}$	5.14	4.12
$^{128}\text{Te} \rightarrow ^{128}\text{Xe}$	6.60	7.34
$^{130}\text{Te} \rightarrow ^{130}\text{Xe}$	5.96	6.26
$^{136}\text{Xe} \rightarrow ^{136}\text{Ba}$	4.91	3.51

Chapter 3

Experimental searches for $0\nu\beta\beta$

In the previous chapter we saw how the measurement of the rare $0\nu\beta\beta$ -decay could lead to the resolution of some major questions still open in the world of the neutrino. In this chapter the experiments that are working towards a $0\nu\beta\beta$ observation are discussed. All of these experiments will depend in some way or another on counting the number of occurrences of an event in a fixed region-of-interest (ROI). Specifically, for a $0\nu\beta\beta$ -decay search, we want to count the number of events for which we see two electrons with a summed energy in the region around the Q-value of the respective isotope. This basic type of experiment is often termed a “counting experiment”. An analysis of the ability of this type of experiment to observe a small signal on top of a large background will provide insight into the experimental aspects that are important for a $0\nu\beta\beta$ -decay search.

Thus, I begin the chapter by describing the sensitivity and discovery potential of a simple counting experiment. I then introduce some of the experimental techniques employed in a $0\nu\beta\beta$ -decay search by discussing a selection of the experiments that are currently operating. I then follow this by describing in detail the experiment for which this work is a part of - the EXO experiment. I begin by describing the various analysis and background reduction techniques that are employed in the currently operating EXO-200 detector, the prototype experiment for the EXO collaboration. The recently published

results and an analysis of the expected sensitivity for the detector are then presented. Following this is a discussion of the nEXO detector - a tonne scale upgrade to the EXO200 experiment. I focus on the pros and cons of increasing the mass of a liquid xenon time projection chamber (TPC) and describe a background reduction technique that is unique to ^{136}Xe experiments and is of major interest for this work: Ba-tagging. Finally, I end by presenting the results of the sensitivity and discovery potential analysis that was completed for the nEXO experiment. This will quantitatively show the benefit of having the Ba-tagging technique included in the future detector.

3.1 Counting experiments

In order to discuss the ability of a counting experiment to observe the $0\nu\beta\beta$ -decay we will need to introduce two concepts: the sensitivity and discovery potential of an experiment. These concepts are based on the principles of statistical hypothesis testing and confidence intervals and therefore I provide a general overview of this theory in Appendix A.

In words, the sensitivity of an experiment gives the most likely statistical limit that an experiment will put on an unobserved effect. In the case of a search for $0\nu\beta\beta$, it will help an experiment predict how much of the neutrino mass parameter-space they can hope to exclude (with a given confidence level). In section A.4.1 I give the mathematical definition of the “sensitivity” as the median upper-limit with a confidence level of 90%.

The discovery potential is a similar concept to the sensitivity but instead wants to quantify what kind of signal an experiment could hope to discover. For a $0\nu\beta\beta$ search this is essentially asking “How many counts in the ROI would we need to see in order to claim a discovery?”. The usual condition of a “5-sigma” effect for a discovery claim is used here. In section A.4.2 I give the mathematical definition of the discovery potential as the smallest number of signal events (the least-detectable-signal) needed to have a “P-value” of 3×10^{-7} (see Section A.1) in half of an ensemble of identical experiments. I

will illustrate the calculation of the sensitivity and discovery potential of a $0\nu\beta\beta$ -decay experiment using the example of the simple counting experiment.

3.1.1 Sensitivity

To this end we assume the experiment is looking for the $0\nu\beta\beta$ signal, s , by counting the number of occurrences of an event in a predefined region-of-interest (ROI). We will assume the case that there is some expected number of background counts b in our ROI such that the total number of events observed, N , will be given by the sum of b and the wanted signal s . The probability distribution function (pdf) of N is simply the Poisson distribution with a mean of $\mu = s + b$,

$$P(N | \mu) = \frac{\mu^N}{N!} e^{-\mu} \quad (3.1.1)$$

If we assume that the background is known exactly then we can use the above to write the pdf for the signal s (using a prime to indicate the measured signal),

$$P(s' | s + b) = \frac{(s + b)^{s'+b}}{(s' + b)!} e^{-(s+b)} \quad (3.1.2)$$

A natural estimator for the value of s is clearly the observed number of signal events $s' = N - b$.

The upper limit on s given an observation s'_{obs} is given by,

$$\beta = \sum_{s'=0}^{s'_{obs}} P(s' | s_{up} + b) \quad (3.1.3)$$

where s_{up} is the upper limit on the number of counts, and we have replaced the integral from Equation A.3.10 with a summation since the Poisson is a discrete pdf.

Unfortunately, this method does not always work, since, for large b and small s it can happen that a non-negative solution s_{up} does not exist. As this *is* the case for most $0\nu\beta\beta$ -

decay searches one should instead use a more robust method to determine s_{up} proposed by Feldman and Cousins [65]. This method uses an ordering principle based on ratios of likelihood functions and always gives physical upper limits.

Regardless of how the upper limit is calculated, if we assume that the pdf of s_{up} is given by $h(s_{up}|\mu)$, then the sensitivity is given by Equation A.4.1,

$$0.5 = \sum_{s_{up}=0}^{\tilde{s}_{up}} h(s_{up}|b) \quad (3.1.4)$$

If we use the Feldman-Cousins method to calculate the upper limit we find the distributions of $h(s_{up}|b)$ for various values of b as shown in Figure 3.1.1. The medians of these distributions are indicated on the plot and represent the sensitivities of the experiments. Taking the $b = 5$ case as an example, the sensitivity would be $\tilde{s}_{up} = 5.0$, that is, the experiment would be able to place a 90% upper limit at least as good as $s \leq 5$ half of the time.

The sensitivity is plotted as a function of b in Figure 3.1.2. The dotted curve represents the sensitivity as calculated using the Feldman-Cousins method, whereas the solid line represents a “Gaussian” approximation where the sensitivity is given by,

$$\tilde{s}_{up}^{gaus} = 1.64\sqrt{b} \quad (3.1.5)$$

This derives from the fact that the Poisson distribution approaches a Gaussian as b gets large.

The above approximation is not seen to be a great representation of the more robust method, however, it is useful as a metric to compare the sensitivities of different $0\nu\beta\beta$ -decay experiments. To see why, consider one of these experiments. The number of signal counts expected in a $0\nu\beta\beta$ -decay search is given by,

$$s = \epsilon \frac{\ln 2}{T_{1/2}^{0\nu}} \frac{\xi m N_A}{M} t \quad (3.1.6)$$

Figure 3.1.1: Distribution of the upper limits for a Poisson signal s and a background of $b = 5, 25$, and 50 . The Feldman-Cousins method was used to calculate the upper limits. The medians of the distributions are indicated by the vertical lines.

Figure 3.1.2: Dependence of $\tilde{s}_{up}$ on b calculated using the Feldman-Cousins method (dots) and the Gaussian approximation (solid line).

where ϵ is the signal detection efficiency, m is the $0\nu\beta\beta$ source mass, ξ is the mass enrichment fraction of source material in the $0\nu\beta\beta$ isotope, M is the $0\nu\beta\beta$ isotope molar mass, and N_A is Avogadro's number. The expected half-life sensitivity of such an experiment is calculated by replacing the signal counts s with the sensitivity $\widetilde{s}_{up}$,

$$\widetilde{T}_{1/2}^{0\nu} = \epsilon \frac{\ln 2}{\widetilde{s}_{up}} \frac{\xi m N_A}{M} t \quad (3.1.7)$$

This can be further converted into a mass sensitivity using Equation 2.3.6,

$$\langle \widetilde{m_{\beta\beta}} \rangle_{up} = \sqrt{\frac{\widetilde{s}_{up} m_e^2 M}{\ln(2) \epsilon \xi m N_A t G^{0\nu} |M^{0\nu}|^2}} \quad (3.1.8)$$

Using the Gaussian approximation for $\widetilde{s}_{up}$ gives the mass sensitivity explicitly as a function of b ,

$$\langle \widetilde{m_{\beta\beta}} \rangle_{up} = \sqrt{\frac{1.64\sqrt{b} m_e^2 M}{\ln(2) \epsilon \xi m N_A t G^{0\nu} |M^{0\nu}|^2}} \quad (3.1.9)$$

We can rewrite the above in a more suitable form by gathering all parameters not under an experimenter's control into an overall constant α ,

$$\alpha = \sqrt{\frac{1.64 m_e^2 M}{\ln(2) N_A G^{0\nu} |M^{0\nu}|^2}} \quad (3.1.10)$$

Equation 3.1.8 then becomes,

$$\langle \widetilde{m_{\beta\beta}} \rangle_{up} = \alpha \cdot \frac{b^{1/4}}{\sqrt{\epsilon t \xi m}} \quad (3.1.11)$$

In most (if not all) experiments the number of expected background events is proportional to the mass-exposure and the width of the energy ROI, ΔE ,

$$b = R \xi m t \Delta E \quad (3.1.12)$$

where R is a rate with units of background counts per isotope-source-mass per time per ROI unit. As all $0\nu\beta\beta$ -decay experiments have to measure the energy of the outgoing electrons a common standard ROI is given by the full-width-half-maximum (FWHM) of a standard Gaussian function with a variance defined by the energy resolution at the $0\nu\beta\beta$ -decay Q-value. Using this definition R has units of $(\text{mass}^{-1}\text{time}^{-1}\text{energy}^{-1})$ and is commonly called the background index. The mass sensitivity becomes,

$$\widetilde{\langle m_{\beta\beta} \rangle}_{up} = \alpha \cdot \left(\frac{R \Delta E_{FWHM}}{\epsilon^2 \xi m t} \right)^{1/4} \quad (3.1.13)$$

One important thing to take away from the above formula is that the sensitivity depends very weakly on the mass-exposure when the backgrounds are high. Thus, simply scaling an experiment's size or running for longer times will not provide much in the setting of better mass limits. It should be reiterated that Equation 3.1.13 is valid only in the high background limit and under the assumption that the background is proportional to the mass-exposure and energy ROI. Despite this, it is a useful tool for comparing the ability of various experimental sensitivities to the $0\nu\beta\beta$ -decay. A similar equation for the half-life sensitivity is given below,

$$\widetilde{T_{1/2}^{0\nu}} = \frac{\epsilon}{\alpha'^2} \sqrt{\frac{\xi m t}{R \Delta E_{FWHM}}} \quad (3.1.14)$$

where,

$$\alpha' = \sqrt{\frac{G^{0\nu} |M^{0\nu}|^2}{m_e^2}} \alpha = \sqrt{\frac{1.64 M}{\ln(2) N_A}} \quad (3.1.15)$$

Equation 3.1.14 is useful for comparing experimental sensitivities that involve the same isotope as the half-life is independent of theoretical uncertainties.

3.1.2 Discovery potential

To illustrate the calculation of the discovery potential, we again consider the experiment that is looking for the $0\nu\beta\beta$ signal, s , by counting the number of occurrences of an event in a predefined region-of-interest (ROI). For simplicity we will use the observed number of events N as our test statistic and assume the background b is known with certainty. The pdf of N is again the Poisson with mean $\mu = s + b$. We will define the null hypothesis H_0 as the hypothesis that the signal does not exist. The pdf of N under H_0 is then,

$$P(N | H_0) = \frac{b^N}{N!} e^{-b} \quad (3.1.16)$$

There will be an infinite number of alternative hypotheses H_i that are characterized by the infinite number of values that s could take on. The pdfs of N under these hypotheses are,

$$P(N | H_i) = \frac{\mu_i^N}{N!} e^{-\mu_i} \quad (3.1.17)$$

where $\mu_i = s_i + b$. The least detectable signal s_{lds} for a $5\text{-}\sigma$ discovery is given by Equations A.4.3 and A.4.4,

$$\sum_{N=N_{cut}}^{\infty} P(N | H_0 : s = 0) = 3 \times 10^{-7} \quad (3.1.18)$$

$$\sum_{N=0}^{N_{cut}} P(N | H_i : s = s_{lds}) = 0.5 \quad (3.1.19)$$

The procedure for determining s_{lds} is somewhat of an iterative one. We first solve Equation 3.1.18 to determine the value of N_{cut} and then try different values of s until we satisfy the above relations.

The discovery potential calculated this way is shown in Figure 3.1.3 as a function of the expected background b . The solid line in this plot again represents the Gaus-

Figure 3.1.3: The least-detectable-signal (discovery potential) of a simple counting experiment as a function of the expected background b .

sian approximation obtained by simply setting $s_{lds} = 5\sqrt{b}$. As shown, the Gaussian approximation again under-estimates the values obtained using the more robust method. However, one can use the Gaussian approximation to derive an approximation for the mass and half-life discovery potential for a $0\nu\beta\beta$ -decay search. We can immediately write the half-life and mass discovery potential by noticing that $s_{lds} = \frac{5}{1.64}\widetilde{s}_{up}$ such that we get,

$$\langle m_{\beta\beta} \rangle_{Disc} = \sqrt{\frac{5}{1.64}} \cdot \widetilde{\langle m_{\beta\beta} \rangle}_{up} \quad (3.1.20)$$

and,

$$(T_{1/2}^{0\nu})_{Disc} = \frac{1.64}{5} \widetilde{T_{1/2}^{0\nu}} \quad (3.1.21)$$

3.2 $0\nu\beta\beta$ -decay experiments

In the last section the sensitivity of a $0\nu\beta\beta$ -decay search was investigated from the standpoint of a simple counting experiment. In this section I will describe a selection of the actual experiments searching for the $0\nu\beta\beta$ -decay with a focus put on three broad categories: high resolution spectroscopy types, the large liquid-scintillator types, and those using time projection chambers (TPCs).

3.2.1 High resolution spectroscopy experiments

These experiments are characterized by their excellent energy resolution and use the ^{76}Ge isotope or ^{130}Te isotopes as $0\nu\beta\beta$ -decay candidates.

The experiments using ^{76}Ge employ high-purity germanium diodes enriched in the target isotope as both the detector and source. Ge diodes offer an excellent energy resolution and, since they are widely used in particle physics, techniques for their design are well established. The dominant experiment using this technique is the GERDA experiment [48].

GERDA (GERmanium Detector Array) is located at the underground Gran Sasso National Laboratory (LNGS) of INFN, Italy. The Ge diodes are created from crystals enriched to $\sim 86\%$ in the ^{76}Ge isotope with a total mass of ~ 20 kg. The first phase of the experiment ran from November 2011 to May 2013 and collected a total exposure of 21.6 kg·y. The diodes have a resolution of about 0.2% at the ^{76}Ge Q-value (2039 keV) and have a measured background index of $\sim 1\text{kg}^{-1}\text{y}^{-1}\text{keV}^{-1}$ (FWHM ROI) which allowed them to produce a limit on the $0\nu\beta\beta$ -decay half-life of $T_{1/2}^{0\nu} > 2.1 \times 10^{25}$ y (90% CL)[46]. The collaboration is currently commissioning the second phase of the experiment where they plan to improve their background index by an order of magnitude and double their source mass with the hopes of setting a limit of $\sim 1.5 \times 10^{26}$ y[47].

Experiments investigating the ^{130}Te isotope employ TeO_2 bolometers to measure the energy of the two outgoing electrons. Bolometers boast similar energy resolutions to Ge detectors and similar signal efficiencies. In addition, the natural abundance of the ^{130}Te isotope is quite high at $\sim 34\%$ and thus enrichment is not as important. The dominant experiment in this category is CUORE [49].

The first phase of CUORE (Cryogenic Underground Observatory for Rare Events) was a prototype experiment, CUORICINO [51], which employed a source mass of 41 kg of natural TeO_2 or ~ 12 kg of ^{130}Te . The experiment ran from 2003 until 2008 collecting a total exposure of ~ 20 kg·y allowing them to produce a limit of $T_{1/2}^{0\nu} > 2.8 \times 10^{24}$ y (90%

CL) on the ^{130}Te $0\nu\beta\beta$ -decay half-life. The next phase, CUORE0, was constructed mainly to test the design goals of CUORE and has been running since 2013. The first results were published this year [52] where they quote a background index of $0.06 \text{ kg}^{-1}\text{y}^{-1}\text{keV}^{-1}$ in a 5 keV FWHM energy ROI. With a total exposure of $9.8 \text{ kg}\cdot\text{y}$ they produced a limit on the ^{130}Te $0\nu\beta\beta$ -decay half-life of $T_{1/2}^{0\nu} > 2.9 \times 10^{24} \text{ y}$ (90% CL). In the final phase of the experiment, known simply as CUORE, the collaboration will increase their source mass to $\sim 200 \text{ kg}$ of ^{130}Te and hope to achieve a background rate of $\sim 0.01 \text{ kg}^{-1}\text{y}^{-1}\text{keV}^{-1}$ in the 5 keV FWHM window. The projected half-life sensitivity of the experiment is $\sim 9.5 \times 10^{25} \text{ y}$ after five years running[50].

3.2.2 Large liquid-scintillator experiments

The large liquid-scintillator experiments employ a large tank of liquid scintillator into which the candidate $0\nu\beta\beta$ -decay isotope is dissolved. The scintillator itself is surrounded by a large buffer volume providing excellent shielding against external backgrounds. The main advantage, in addition to the shielding, is the ability to dissolve a large mass of $0\nu\beta\beta$ -decay isotope into the scintillator. Unfortunately, they suffer from relatively poor energy resolution as compared to the other $0\nu\beta\beta$ -decay experiments. The dominant experiment in this category is KamLAND-Zen [53, 54].

The KamLAND-Zen experiment uses the $0\nu\beta\beta$ -decay isotope ^{136}Xe which is dissolved into the Kamioka Liquid Scintillator Antineutrino Detector (KamLAND). In their first phase of running [55] (October 2011 to June 2012) about 320 kg of 91% enriched Xe were dissolved in the detector. The measured energy resolution was $\sim 4\%$ at the ^{136}Xe Q-value (2458 keV) and the background index was determined to be $\sim 2 \times 10^{-4} \text{ kg}^{-1}\text{y}^{-1}\text{keV}^{-1}$. With a total exposure of $89.5 \text{ kg}\cdot\text{y}$, the experiment produced a half-life limit on the $0\nu\beta\beta$ -decay of ^{136}Xe of $T_{1/2}^{0\nu} > 1.9 \times 10^{25} \text{ y}$ at 90% CL. A second phase of the experiment reduced a problematic background in their detector by an order of magnitude and collected data between December 2013 and May 2014. A preliminary analysis combining the phase 1

and 2 data-sets produced a limit on the $0\nu\beta\beta$ -decay half-life of $T_{1/2}^{0\nu} > 2.6 \times 10^{25}$ y [56]. The collaboration plans to increase the mass of enriched Xe dissolved to ~ 1000 kg, and, with various other improvements, hopes to reach a half-life sensitivity on the order of 10^{27} y after five years of running [56].

3.2.3 Time projection chambers

The experiments in this category exclusively use the ^{136}Xe isotope. The xenon itself is a suitable detector material offering both charge and scintillation signals and thus Xe enriched in the ^{136}Xe isotope is used as both source and detector in these experiments. The main advantage over the other types of experiments is the ability to reject backgrounds using topological information, in addition to the energy measurement. There are two main experiments in this category: EXO [57] and NEXT [60, 61, 62].

The EXO (Enriched Xenon Observatory) collaboration is a xenon TPC searching for the $0\nu\beta\beta$ -decay of ^{136}Xe . A prototype experiment, EXO-200 [58], contains ~ 200 kg of liquefied-xenon enriched to 81% in the $0\nu\beta\beta$ -decay isotope, ^{136}Xe . Despite the high density of liquid xenon, the experiment has demonstrated an energy resolution of $\sim 1.5\%$ at the ^{136}Xe Q-value achieved by combining the scintillation and ionization signals measured. The most recent $0\nu\beta\beta$ -decay search [59] had a measured background index of $\sim 2 \times 10^{-3} \text{ kg}^{-1} \text{ y}^{-1} \text{ keV}^{-1}$ (FWHM ROI) and, with a total of $100 \text{ kg} \cdot \text{y}$ exposure, produced a limit on the $0\nu\beta\beta$ -decay half-life of $T_{1/2}^{0\nu} > 1.1 \times 10^{25}$ y. The EXO-200 detector, and a proposed tonne-scale experiment, nEXO, will be discussed in detail in the next section.

The NEXT (Neutrino Experiment with a Xenon TPC) collaboration is currently planning and designing NEXT-100, a 100 kg high-pressure gaseous Xe TPC enriched to 90% in the ^{136}Xe isotope. The experiment will take advantage of event topology in order to reject backgrounds, which, combined with a demonstrated energy resolution of $< 1\%$ [64] at the ^{136}Xe Q-value, gives an expected background index of $\sim 5 \times 10^{-4} \text{ kg}^{-1} \text{ y}^{-1} \text{ keV}^{-1}$ (FWHM ROI). The expected sensitivity is $\sim 1 \times 10^{26}$ y after 5 years of running [63]. The

Table 3.1: Experimental limits on the $0\nu\beta\beta$ -decay half-lives produced by the GERDA, CUORICINO, KamLAND-ZEN, and EXO-200 collaborations. The limits in all cases are at a 90% CL and were converted to a corresponding mass limit using the phase space factors, and nuclear matrix elements listed in Tables 2.2 and 2.3 respectively.

Experiment	Isotope	Exposure (kg · y)	$T_{1/2}^{0\nu}$ (y)	$\langle m_{\beta\beta} \rangle$, IBM-2 (meV)	$\langle m_{\beta\beta} \rangle$, QRPA (meV)
GERDA	^{76}Ge	22	$> 2.8 \times 10^{24}$	518	469
CUORICINO	^{130}Te	20	$> 2.9 \times 10^{24}$	262	249
KamLAND - ZEN	^{136}Xe	90	$> 2.6 \times 10^{25}$	105	147
EXO-200	^{136}Xe	100	$> 1.1 \times 10^{25}$	161	226

collaboration is currently building the NEW detector, a 10 kg prototype planned to start running this year. The ultimate goal of the experiment is to use the knowledge gained from NEXT-100 to build a tonne scale experiment.

Table 3.1 summarizes the different parameters and limits of the experiments above that have already made measurements. For the matrix elements shown, the current best limit is from the KamLAND-Zen experiment putting $\langle m_{\beta\beta} \rangle < 105 - 147$ meV, depending on the nuclear matrix element used. Combining the limits produced from $0\nu\beta\beta$ -decay and cosmology, as well as the best estimates of the neutrino mixing parameters (Table 2.1), we can plot the allowed region in $\langle m_{\beta\beta} \rangle, m_{\nu}^{\min}$ -space (where $m_{\nu}^{\min}$ is the mass of the lightest neutrino), as is shown in Figure 3.2.1. From this we can see that in order to completely probe the inverted hierarchy, the next generation of $0\nu\beta\beta$ -decay experiments will have to probe neutrino masses in the 10 meV range.

3.3 EXO-200

The EXO experiment is a multi-phase program devoted to observing the $0\nu\beta\beta$ -decay of ^{136}Xe . The first phase of the program, EXO-200, is a 200 kg liquid-xenon (LXe) TPC

Figure 3.2.1: Allowed regions of the neutrino mass parameter space. The y-axis is the effective neutrino mass, and the x-axis is the mass of the lightest neutrino. The solid lines correspond to the central values of the neutrino mixing parameters, and the dashed lines are for the $\pm 2\sigma$ interval on the mixing parameters.

that is currently operating in the Waste Isolation Pilot Plant (WIPP) in Carlsbad, NM.

The EXO-200 detector (Figures 3.3.1 and 3.3.2) is a relatively compact cylindrical TPC roughly 40 cm in diameter and 44 cm in length and is divided into two drift regions by a central cathode. The liquid xenon is enriched to 80.6% in the ^{136}Xe isotope and kept at a temperature and pressure of 167 K and 147 kPa, respectively. The entire chamber is surrounded by a double-walled cryostat containing the coolant HFE-7000 (HFE, hydro-fluoro-ethers) which provides temperature uniformity throughout. Lead shielding reduces external backgrounds and plastic scintillators are installed on four of the six sides of the detector to act as active muon vetoes. At any given time only 175 kg of the xenon is in liquid form with the rest circulated in gaseous form within the gas handling and purification systems.

Physics events occurring in the TPC produce both ionization and scintillation signals in the xenon. The ionization signal is collected on two anodes on either end of the detector. The anodes themselves are composed of two planes of crossed wires (U- and

Figure 3.3.1: Schematic of the EXO-200 LXe vessel. Adapted from [58].

Figure 3.3.2: Cartoon diagram of the EXO-200 detector setup. Adapted from [58].

Figure 3.3.3: Calibration source locations (S2, S5, S8, S11) for the EXO-200 detector. Adapted from [67]

V-wires). The first plane (V) is a shielding plane and is biased to produce full electron transparency. In addition, the V-wires also produce useful induction signals which are combined with the charge collected on the second plane (U). The wires are oriented at an angle of 60° to each other allowing X-Y reconstruction of the signal location.

The scintillation light ($\lambda = 178$ nm) produced is collected at two planes of large-area-avalanche-photo-diodes (LAAPDs) located behind the charge collection wires. Ultra-violet reflective teflon-PTFE tiles are installed on the walls of the TPC to increase light collection. The time difference between the prompt scintillation light and ionization signal gives the charge drift time and allows for the reconstruction of the events' Z-position in the detector.

The detector is calibrated frequently using various gamma-ray sources - ^{137}Cs , ^{60}Co , ^{228}Th , and ^{226}Ra . The sources are placed at specific locations (Figure 3.3.3) near the detector (within the HFE) using copper guide tubes.

The energy variable used (rotated energy, E) in the analysis is created as a linear combination of the ionization (E_I) and scintillation (E_S) signals,

Figure 3.3.4: Left: definition of the rotated energy. Right: comparison of resolution for the ionization, scintillation, and rotated energy channels.

$$E = E_S \sin \theta + E_I \cos \theta \quad (3.3.1)$$

vastly improving the energy resolution as compared to that for either individual channel, as shown in Figure 3.3.4. This is possible due to the fact that the ionization signal anti-correlates with the scintillation signal [66]. The ideal rotation angle, θ , is determined from the calibration data. Using this method an energy resolution of 1.53% at the Q-value was achieved in the most recent $0\nu\beta\beta$ -decay analysis [59].

The event reconstruction is composed of three steps: signal finding, parameter estimation, and clustering/event reconstruction. The first stage, signal finding, identifies potential physics events by matching predefined models to the raw waveforms produced. The second stage, parameter estimation, builds a χ^2 function from the waveforms identified in the first stage with the signal model and minimizes it to determine signal parameters such as amplitude, arrival time, and uncertainties. In the final stage, the energy and positions of the events are reconstructed. This is accomplished by first bundling signals together into charge clusters (CCs) based on their relative positions in space, and then associating different charge clusters to one event or another based on their relative positions in time. The final reconstructed physics events are classified as single-site (SS) if they contain only one associated CC, and as multi-site (MS) otherwise. The overall

signal reconstruction efficiency for $0\nu\beta\beta$ events was measured to be $84.6 \pm 8.6\%$ [59].

Radioactive contamination is mainly due to the daughter products of the ^{232}Th and ^{238}U decay chains. However, if not considered, cosmogenic activation of the detector components can also become substantial. From the ^{232}Th chain the main concern is the gamma produced in the decay of ^{208}Tl , which, at 2614 keV (100%), is relatively close to the ^{136}Xe Q-value. From ^{238}U comes the very problematic isotope ^{214}Bi , which has two β -decay paths with end-points of 2663 keV (1.7%) and 3272 keV (18.2%), and also a gamma at 2447.8 keV (1.5%). From cosmogenic activation of copper we get the ^{60}Co isotope which releases two prominent gammas at 1173.2 keV and 1332.5 keV (99.88%) in succession which causes a sum peak at 2505.7 keV.

In order to reduce the effects of the aforementioned backgrounds many steps and advanced techniques were used in the design, construction, and analysis stages. Firstly, the detector (especially components near the LXe) was constructed from components that were carefully screened for radiopurity and the entire detector's design was optimized to reduce the mass of passive components as much as possible. Any components that had the possibility of cosmogenic activation (specifically those made from copper) were shipped on land and machined in a dedicated underground shop, and all components were degreased and etched before being assembled.

Background rejection using event topology is realized in EXO in two ways. The first is based on the SS or MS classification given to an event during reconstruction, and the second uses a parameter called the standoff-distance¹, s_d . The majority of backgrounds in EXO-200 are gamma-rays originating from sources outside of the liquid xenon. This has two physical consequences: first, gamma-rays tend to Compton scatter and thus will more likely cause a MS event to occur in the detector, and second, there will be somewhat of an exponential dependence on the standoff distance due to the attenuation of gammas travelling through the xenon. Thus, the MS data-set should be primarily composed of

¹Standoff-distance is defined as the minimum separation between an event and any wall of the LXe vessel.

background gamma-events, while the SS data-set should contain more electron-like events (such as $0\nu\beta\beta$ -decay). In addition, gamma-like backgrounds will have an exponential-like standoff-distance distribution, while the signal events should be distributed uniformly throughout the detector.

In order to extract a final physics result, a multivariate log-likelihood fit is performed on the SS and MS data-sets simultaneously. The shapes of the energy and standoff-distance distributions for each background (and signal) and the expected fraction of SS events are estimated using a detailed simulation of the detector built using the GEANT4 physics simulation package.

In 2013 and 2014 EXO-200 published two significant results. The first result was a measurement of the $2\nu\beta\beta$ half-life of ^{136}Xe of $T_{1/2}^{2\nu} = 2.165 \pm 0.016$ (stat.) ± 0.059 (systematic) $\times 10^{21}\text{y}$ [67]. This is the most precise measurement of any $2\nu\beta\beta$ half-life to date. The best-fit to the measured energy and standoff distributions is shown in Figure 3.3.5.

The second publication updated a limit on the $0\nu\beta\beta$ -decay half-life of ^{136}Xe and was mentioned in the first chapter. Apart from various support work during the final analysis, I was tasked to calculate the sensitivity of the EXO-200 experiment based on the backgrounds expected and the running time of the experiment. The method used to do this is somewhat more complicated than the example of the simple counting experiment and will use the theory of parameter estimation using the maximum likelihood method. This theory is presented in section A.2 and a full example of the sensitivity and discovery potential calculation is given in section A.5. The only difference from the given example is that the background shapes are replaced by the simulated distributions, and the pdfs are functions of two variables: energy and standoff-distance.

The distribution of the calculated upper limits on the number $0\nu\beta\beta$ -decay events is shown in Figure 3.3.6. The median upper limit calculated from this distribution is $\widetilde{s}_{up} = 14.04$ counts. At this point we can use all of the equations derived in the last section for the half-life and mass sensitivities. The half-life sensitivity calculated in this way is

Figure 3.3.5: The EXO-200 energy distributions for the $2\nu\beta\beta$ -decay half-life measurement in [67]. a) The distribution of the SS-events. The standoff-distance distribution is shown in the inset in the top right. b) Distribution of MS-events. c) Normalized residuals defined as the difference between the best-fit model and the data divided by the error on the data.

Figure 3.3.6: The distribution of upper limits on the number of $0\nu\beta\beta$ -decay events for the EXO-200 detector. The line in the center of the shaded region indicates the median of the distribution, while the shaded region represents a 68% interval around the median. The vertical line on the right indicates the observed upper limit from the analysis given in [59].

1.9×10^{25} y which corresponds to a mass sensitivity of 123 meV (IBM-2) or 172 meV (QRPA).

We can compare this to the sensitivity expected using the approximations of $\tilde{s}_{up}$ derived in section 3.1.1. Using the background index, efficiency, and mass-exposure given in [59] we calculate a half-life sensitivity of $\tilde{T}_{1/2}^{0\nu} > 2.9 \times 10^{25}$ y using the Gaussian approximation, Equation 3.1.14. If we use the Feldman-Cousins method for $\tilde{s}_{up}$ we get $\tilde{T}_{1/2}^{0\nu} > 2.4 \times 10^{25}$ y. Both of these are larger than the sensitivity calculated using the more complex method but this is to be expected since the backgrounds were assumed to be known without error for these simple approximations. The method outlined in section A.5 is capable of including uncertainties in the fit and should thus give the more accurate prediction. Despite this, it is seen that the simple methods give a reasonable approximation to the real thing.

3.4 nEXO

The success of the EXO-200 experiment has led the collaboration to start developing the nEXO detector. Technically its own collaboration, the nEXO group is currently designing

a 5-tonne LXe detector which is being designed with the goal of completely ruling out the inverted hierarchy (or discovering a neutrino mass if it exists there) in 5-10 years running time. The design of the detector is run under the principle of “as close as possible to EXO-200” so many of the things discussed in the previous section apply here. However, in comparing a 5-tonne detector to a ~ 200 kg detector there are some major differences.

The number one benefit of increasing the detector size is clearly to increase the mass of the active isotope under investigation. However, as we saw in section 3.1.1, if the backgrounds are high then one only gains a small improvement in experimental sensitivity by increasing the source mass. Luckily, the increase in source mass has two other important benefits that will impact the background. Firstly, event containment improves which leads to a higher fraction of events (specifically the Compton scatters) being deposited entirely inside the detector and reconstructed properly. Secondly, the internal parts of the detector become less sensitive to external backgrounds due to the increased mass-attenuation of the γ -rays.

There are, of course, many problems created by increasing the size of the detector so drastically. The large mass implies that engineering will be critical to ensure the survival of the detector and everyone who works around it. Temperature stability over such a large volume also becomes a challenge. Finally, the susceptibility to radioactive contaminants during the construction phase also increases with the complexity and size, and, if not properly controlled, could ruin any benefit the increased mass would bring.

As the detector is built, other improvements uncovered by constant research and development will almost certainly improve the success of the nEXO experiment. One of these research paths, which is the secret weapon of a ^{136}Xe detector, has to do with the identification of the ^{136}Ba ion produced in the $\beta\beta$ -decay. As this was the main motivation for much of this work I will describe it in detail.

3.4.1 Barium-tagging

Barium-tagging [3] is a simple concept - identify the daughter ^{136}Ba produced in the $\beta\beta$ -decay of ^{136}Xe and use this to reject all non- $\beta\beta$ backgrounds. The Ba-tagging scheme is broadly composed of four main steps:

1. A ^{136}Xe atom decays inside the xenon vessel producing a $^{136}\text{Ba}^{++}$ ion.
2. The $^{136}\text{Ba}^{++}$ ion is extracted from the vessel into a low pressure region.
3. The $^{136}\text{Ba}^{++}$ ion is converted to its singly charged form, $^{136}\text{Ba}^+$.
4. The $^{136}\text{Ba}^+$ ion is identified via laser induced fluorescence.

In reality, the actual implementation is an extreme experimental challenge. The $^{136}\text{Ba}^{++}$ ion will be produced at some random location in the detector and drifted to an exit where it can be extracted. It then has to be propagated through a pressure differential spanning orders of magnitude into a collision cell where it is converted to the singly charged species. Following this, it must then be transferred to an ion trap where it can finally be identified via laser induced fluorescence. This all must be done relatively quickly, since, according to the measured half-life of the regular $2\nu\beta\beta$ -decay, roughly one decay should occur in a 5-tonne volume every 4.5 s. Finally, this all has to be done with essentially 100% efficiency.

This may sound impossible but the work is already under way with many of the steps outlined above already demonstrated. Namely, collaborators at Stanford University have already demonstrated trapping and identification of single ions [68, 69, 70, 71], and also have developed a so-called RF-funnel that can extract ions from a 10 bar region to a 10^{-6}mbar region [72]. Here at Carleton, the charge-conversion of $^{136}\text{Ba}^{++}$ to $^{136}\text{Ba}^+$ using tri-ethyl-amine (TEA) has been demonstrated [73], and a barium ion selection system has been constructed [74] for the use in a planned system to measure the absolute efficiency of charge conversion plus ion trapping and identification. My work focused

on developing the ion trapping and identification system here at Carleton and will be discussed at length in the following chapters.

The importance of including the Ba-tagging scheme in a future detector is best demonstrated through an analysis of the sensitivity and discovery potential with and without Ba-tagging present.

3.4.2 Sensitivity and discovery potential of nEXO

The calculations of the sensitivity and discovery potential for the nEXO experiment also follow the method presented in section A.5. Again, the only difference will be the backgrounds considered and the pdfs used to describe them. The results of the full analysis are presented here.

The first step to calculate the discovery potential or sensitivity is to determine a general concept of how the detector will be set up and then simulate all the backgrounds expected in the experiment. A schematic of the detector is shown in Figure 3.4.1. The ^{238}U and ^{232}Th natural decay chains were simulated in each considered component, and the cosmogenically activated ^{60}Co was simulated in each of the copper components. An additional contamination of ^{222}Rn was added to the HFE and the LXe. Inside the vessel, the problematic ^{214}Bi was added, and a neutrino-induced reaction that produces ^{137}Xe has also been considered. Of course each of the $\beta\beta$ -decays were also simulated. Combining all of this with the expected radioactive activities for the components provides the expected background contamination. The SS energy and standoff distributions for the simulated backgrounds are shown in Figure 3.4.2, and a plot of the expected background index given as a function of the internal mass of LXe is shown in Figure 3.4.3.

Once the expected background and component pdfs are known, then it is simply a matter of applying the method developed in Appendix A. The resulting least-detectable-signal and median 90% upper limits are shown in Figure 3.4.4 as a function of the live-time of the detector.

Figure 3.4.1: Illustration showing the main components simulated for the nEXO experiment. The dimensions are not to scale. The details of the LXe TPC included in the simulation has not been shown here. Note that HDPE stands for high-density polyethylene

Figure 3.4.2: The simulated SS energy and standoff distributions for the nEXO experiment.

Figure 3.4.3: The background index of nEXO as a function of the internal Xe mass considered for the calculation. The large increase in backgrounds as more mass is considered is mostly caused by the reduction in attenuation of external γ -rays as the standoff from the detector walls is decreased. This is the self-shielding effect.

Figure 3.4.4: The sensitivity (median 90% upper limit) and the discovery potential (minimum $0\nu\beta\beta$ -decay counts needed to make a discovery) as a function of the live-time for the nEXO detector.

Figure 3.4.5: The sensitivity (median 90% upper limit) and the discovery potential (minimum $0\nu\beta\beta$ -decay counts needed to make a discovery) as a function of the live-time for the nEXO detector when barium-tagging is included.

So far we have not considered the case that barium-tagging is implemented. If nEXO does implement barium-tagging successfully then, as far as the calculation is concerned, all of the non- $\beta\beta$ backgrounds can simply be neglected. Following the same procedure as used to determine the sensitivity and discovery without Ba-tagging we get the least-detectable-signal and median 90% upper limits as shown in Figure 3.4.5.

Combining the results from the cases with and without barium-tagging, we can plot the sensitivity and discovery potential for the half-life and neutrino mass as shown in Figure 3.4.6 and 3.4.7 for the two different matrix elements we have been considering. These plots clearly demonstrate the power of the Ba-tagging technique. As shown, without Ba-tagging, the detector will just be able to cover the inverted hierarchy, depending on the nuclear matrix element used, and will certainly not be able to discover an effect above this point. With Ba-tagging implemented, however, the detector will easily be sensitive to the entire inverted hierarchy and even begin to creep into the normal hierarchy. In addition, if the neutrino masses do follow the inverted hierarchy, then the detector with Ba-tagging will quite likely discover it. This provides the main motivation for this work.

Figure 3.4.6: Discovery potential and half-life sensitivity of the nEXO detector. It is assumed that the first five years of the experiment will be run without Ba-tagging included, and the next five years with Ba-tagging included. The horizontal bands indicate the regions where the neutrino masses will have the inverted hierarchy or normal hierarchy configuration. The dashed vertical line in the center separates the experiment into two phases: the first phase is without Ba-tagging, and the second phase includes Ba-tagging. The effective neutrino masses listed on the y -axis on the left used the nuclear matrix element from the IBM-2 calculation. The y -axis on the left has units of years.

Figure 3.4.7: Discovery potential and half-life sensitivity of the nEXO detector. It is assumed that the first five years of the experiment will be run without Ba-tagging included, and the next five years with Ba-tagging included. The horizontal bands indicate the regions where the neutrino masses will have the inverted hierarchy or normal hierarchy configuration. The dashed vertical line in the center separates the experiment into two phases: the first phase is without Ba-tagging, and the second phase includes Ba-tagging. The effective neutrino masses listed on the y -axis on the left used the nuclear matrix element from the QRPA calculation. The y -axis on the left has units of years.

Chapter 4

Charged particle confinement

The idea of confining charged particles using electromagnetic fields has been around since the middle of the 20th century. The first such device, the so called Penning-trap, uses hyperbolic electrodes to produce a static quadrupole electric field for confinement along the trap axis, and a strong static magnetic field about which the ions oscillate. This design is appealing due to the use of static fields only, however, it is not particularly suited to spectroscopic measurements due to energy splittings caused by the strong magnetic field.

The ion trap used in this work is a form of the radio-frequency (RF) Paul trap which does away with the magnetic field, instead confining particles using an oscillating electric field. The theory of Paul traps is essentially identical to any other quadrupole device (mass spectrometer, ion guide, etc.) and thus the theory is developed in the context of a generic linear quadrupole (LQ).

4.1 Adiabatic approximation

The motion of an ion inside an LQ can be solved analytically, however, it is more insightful to analyze the ion motion under the so-called adiabatic approximation that was developed by Kapitza, as described by Gerlich in [75]. The main assumption of the adiabatic approximation is that the electric field intensity varies only weakly over the oscillation of

the ion. Thus, we consider a particle of mass m and charge Q under the action of a one-dimensional, oscillating electric field $E(r, t)$. We assume E is composed of an oscillatory part, $E_0(r) \cos \Omega t$, and a static part, $E_s(r)$. The particle motion is therefore governed by,

$$m\ddot{r}(t) = Q [E_0(r) \cos \Omega t + E_s(r)] \quad (4.1.1)$$

It is easiest to begin by neglecting the static part of the field such that,

$$m\ddot{r}(t) = Q E_0(r) \cos \Omega t \quad (4.1.2)$$

Now, if we assume complete position independence for the field, i.e., it is homogeneous, we can immediately get the solution,

$$r(t) = -\frac{QE_0}{m\Omega^2} \cos \Omega t \quad (4.1.3)$$

which describes the simple harmonic motion of an ion with average position $r = 0$.

If we now consider the case that the field is weakly inhomogeneous, then we can guess that the particle motion will be composed of a rapidly oscillating part (micromotion) as given in Equation 4.1.3, and a slowly drifting part (macromotion). On this basis we assume a solution of the form,

$$r(t) = R_0(t) - \frac{q}{m\Omega^2} E_0(t) \cos \Omega t \quad (4.1.4)$$

where R_0 represents the slow, macromotion of the ion.

Expanding the field around R_0 and keeping only the first two terms gives,

$$E(r, t) = E_0(R_0) \cos \Omega t - \frac{Q}{m\Omega^2} \frac{dE_0}{dr} \cos^2 \Omega t \quad (4.1.5)$$

such that the force on the ion is,

$$m\ddot{r}(t) = m\ddot{R}_0 + QE_0(r) \cos \Omega t = Q \left[E_0(R_0) \cos \Omega t - \frac{Q}{m\Omega^2} E_0 \frac{dE_0}{dr} \cos^2 \Omega t \right] \quad (4.1.6)$$

Now, the field amplitude E_0 will have some time dependence due to its dependence on $r(t)$. If we assume this dependence is dominated by the macromotion R_0 then we can set $E(r) \approx E(R_0)$ and cancel the two $\cos \Omega t$ terms in Equation 4.1.6 to get,

$$m\ddot{R}_0 = -\frac{Q^2}{2m\Omega^2} \frac{d}{dr} (E_0 \cos \Omega t)^2 \quad (4.1.7)$$

Finally, taking the time average gives,

$$m\ddot{R}_0 = -\frac{Q^2}{4m\Omega^2} \frac{d}{dr} \langle E_0^2 \rangle \quad (4.1.8)$$

What this equation shows is that the effect of the field inhomogeneity is to produce a time-averaged force on the particle that is inversely proportional to the particle's mass, squared-frequency, and is independent of the sign of the particle's charge. In addition, the dependence on the gradient of E_0^2 shows that the particle is always pushed towards areas of weaker field intensity.

If we now account for the static field in Equation 4.1.1 and rewrite it as the negative gradient of a static potential $E_s = -\nabla \Phi_s$ then we simply need to modify Equation 4.1.8 as below,

$$m\ddot{R}_0 = -\nabla \left[\frac{Q^2 \langle E_0^2 \rangle}{4m\Omega^2} + Q\Phi_s \right] \quad (4.1.9)$$

What the above equation shows is that the motion of the ion can be described by an effective potential,

$$\Phi_{eff}(R_0) = \frac{Q \langle E_0^2(R_0) \rangle}{4m\Omega^2} + \Phi_s(R_0) \quad (4.1.10)$$

such that the force is given by,

$$m\ddot{R}_0 = -Q\nabla\Phi_{eff}(R_0) \quad (4.1.11)$$

The ion energy can be determined by integrating the above equation once,

$$T_0 = \frac{1}{2}m\dot{R}_0^2 + \frac{Q^2\langle E_0^2 \rangle}{4m\Omega^2} + Q\Phi_s \quad (4.1.12)$$

where T_0 is the total energy of the ion and is a constant of the motion. The first term in the above equation is easily recognized as the kinetic energy due to the macromotion of the ion. Furthermore, it is not hard to show that the second term in Equation 4.1.12 is just the time averaged kinetic energy of the micromotion, $\langle \frac{1}{2}m\dot{R}_1^2 \rangle$. Thus, Equation 4.1.12 shows that the ion's energy is shared between three degrees of freedom: the kinetic energy associated with the macromotion, $\frac{1}{2}m\dot{R}_0^2$, micromotion, $\langle \frac{1}{2}m\dot{R}_1^2 \rangle = \frac{Q^2\langle E_0^2 \rangle}{4m\Omega^2}$, and a potential energy due to the static field, $Q\Phi_s$. These results are easily generalized to three-dimensions.

4.2 The linear quadrupole

The linear quadrupole is composed of four parallel, cylindrical electrodes with an RF voltage $\pm \frac{1}{2}V \cos \Omega t$ applied such that adjacent (opposite) electrodes are completely out of phase (in phase) with each other (see Figure 4.2.1). This (approximately¹) produces an oscillating quadrupolar potential at the center of the trap[77],

$$\Phi_{RF} = \frac{1}{2}V \cos \Omega t \left(\frac{x^2 - y^2}{r_0^2} \right) \quad (4.2.1)$$

¹Ideally one would use hyperbolic electrodes to produce a purely quadrupole field. However, due to the complexities of machining them, hyperbolic electrodes are seldom used. Instead, cylindrical electrodes are used with dimensions such that the ratio of electrode radius to trap radius is in the range $r_e/r_0 = 1.12 - 1.3$ as this produces the best approximation to a purely quadrupole field near the center of the trap[76].

where r_0 is the radius of the trap.

The electric field is obtained by taking the gradient of Equation 4.2.1,

$$\vec{E}_{RF} = -\nabla\Phi_{RF} = -V \cos \Omega t \left(\frac{x\hat{\mathbf{x}} - y\hat{\mathbf{y}}}{r_0^2} \right) \quad (4.2.2)$$

such that,

$$\langle E_{RF}^2 \rangle = \left\langle \vec{E}_{RF} \cdot \vec{E}_{RF} \right\rangle = \frac{V^2}{2r_0^4} r^2 \quad (4.2.3)$$

Inserting the above into Equation 4.1.10 gives the effective potential,

$$\Phi_{eff} = \frac{QV^2}{4m\Omega^2 r_0^4} r^2 \quad (4.2.4)$$

We can thus see that the effective potential is just that of a two-dimensional harmonic oscillator with characteristic depth,

$$D_{RF} = \frac{Q^2 V^2}{4m\Omega^2 r_0^2} \quad (4.2.5)$$

and secular frequency,

$$\omega = \omega_x = \omega_y = \frac{QV}{\sqrt{2}m\Omega r_0^2} \quad (4.2.6)$$

The addition of a static voltage U to the electrodes breaks the x - y symmetry and modifies the effective potential,

$$\Phi_{eff} = \frac{1}{2r_0^2} \left[\left(\frac{QV^2}{2m\Omega^2 r_0^2} + U \right) x^2 + \left(\frac{QV^2}{2m\Omega^2 r_0^2} - U \right) y^2 \right] \quad (4.2.7)$$

The secular frequencies become,

$$\omega_{x,y} = \sqrt{\frac{Q^2 V^2}{2m^2 \Omega^2 r_0^4} \pm \frac{QU}{mr_0^2}} \quad (4.2.8)$$

Figure 4.2.1: Front end view of the linear quadrupole.

The maximum depth of the trap is reduced by $\frac{1}{2}Q|U|$ regardless of the sign of U ,

$$D_{RF} = \frac{Q^2 V^2}{4m\Omega^2 r_0^2} - \frac{1}{2}Q|U| \quad (4.2.9)$$

and particle confinement ceases when,

$$|U| \leq \frac{QV^2}{2m\Omega^2 r_0^2} \quad (4.2.10)$$

The adiabatic approximation is appealing due to its simplicity and because it offers many insights into the motion of ions in charged particle traps. It however relies on the assumption that the field varies only weakly over the particle oscillation, implying the condition that $\omega_{x,y} \ll \Omega$. The full solution to Equation 4.1.1 for the LQ can be obtained analytically and is discussed in the next section.

4.3 Mathieu equations

In this section the full solution to Equation 4.1.1 is discussed for an ion in a combined oscillating and static field,

$$E_{x,y} = \mp(U + V \cos \Omega t) \frac{x\hat{\mathbf{x}} - y\hat{\mathbf{y}}}{r_0^2} \quad (4.3.1)$$

The equations of motion are,

$$\begin{aligned} m\ddot{x}(t) &= -\frac{Q}{r_0^2} [U + V \cos \Omega t] x \\ m\ddot{y}(t) &= +\frac{Q}{r_0^2} [U + V \cos \Omega t] y \end{aligned} \quad (4.3.2)$$

If we define the variables,

$$\tau = \frac{1}{2}\Omega t \quad (4.3.3)$$

$$a_x = -a_y = \frac{4QU}{m\Omega^2 r_0^2} \quad (4.3.4)$$

$$q_x = -q_y = \frac{2QV}{m\Omega^2 r_0^2} \quad (4.3.5)$$

and insert these into Equation 4.3.2 we get,

$$\ddot{u}(\tau) + [a_u - 2q_u \cos 2\tau]u = 0, \quad u \in x, y \quad (4.3.6)$$

The above is the general form of the homogeneous Mathieu equation. The solutions that produce stable ion trajectories take the form of a Fourier series [77],

$$u(\tau) = A_u \sum_{n=-\infty}^{+\infty} C_{2n,u} \cos[(\beta_u + 2n)\tau] + B_u \sum_{n=-\infty}^{+\infty} C_{2n,u} \sin[(\beta_u + 2n)\tau] \quad (4.3.7)$$

where A_u and B_u are constants defined by the initial conditions $[u_0, \dot{u}_0]$, the $C_{2n,u}$ are coefficients that depend only on the parameters a_u and q_u , and β_u is an overall stability parameter also depending only on a_u and q_u . The coefficients can be determined by a recursion relation obtained by differentiating the above twice and inserting the result into Equation 4.3.6,

$$\frac{C_{2n,u}}{C_{2n,u\mp 2}} = -\frac{q_u}{(2n + \beta_u)^2 - a_u - \frac{q_u^2}{(2n \pm 2 + \beta_u)^2 - a_u - \dots}} \quad (4.3.8)$$

The value of β is determined by a continued fraction expression [77],

$$\begin{aligned} \beta_u^2 &= a_u + f_u(\beta_u) + f_u(-\beta_u) \\ f_u(\beta_u) &= \frac{q_u^2}{(2 + \beta_u)^2 - a_u - \frac{q_u^2}{(4 + \beta_u)^2 - a_u - \dots}} \end{aligned} \quad (4.3.9)$$

The strict criterion for stability is that β_u is a real, non-integer number. Since β_u only depends on a_u and q_u , this implies that stable trajectories can be defined by regions in a_u, q_u -space that are bounded by curves of integer β_u . Of particular interest is the region bounded by $0 < \beta_u < 1$ as this is exclusively used for particle confinement. This region is plotted in Figure 4.3.1 where the vertex along the x -axis indicates the maximum value of q that still produces a stable trajectory ($q_{max} = 0.908$).

Inspection of Equation 4.3.7 reveals that the characteristic oscillation frequencies of the ion are given by,

$$\omega_{n,u} = (\beta_u + 2n) \frac{\Omega}{2} \quad (4.3.10)$$

Figure 4.3.1: Region of stability for a LQ, defined by $0 < \beta_u < 0$, $u \in x, y$. The maximum value of q that still produces a stable trajectory occurs at $q = 0.908$.

The fundamental frequency of oscillation is given when $n = 0$ and represents the slow, macromotion of the ion. The $n = 1$ and higher order frequencies are responsible for the rapidly oscillating micromotion.

It is interesting to note that, in the case that a_u and q_u are small ($a_u, q_u \ll 1$), it becomes a good approximation to truncate the solution 4.3.7 after the first two terms. In this case the stability parameter can be written,

$$\beta_u \approx \sqrt{\frac{1}{2}q_u^2 + a_u} \quad (4.3.11)$$

With this definition the fundamental frequency becomes,

$$\omega_{0,u} = \frac{\Omega}{2} \sqrt{\frac{1}{2}q_u^2 + a_u} = \sqrt{\frac{Q^2 V^2}{2m^2 \Omega^2 r_0^4} \pm \frac{QU}{mr_0^2}} \quad (4.3.12)$$

which is exactly what was found using the adiabatic approximation.

Figure 4.4.1: Segmented linear Paul trap. The segments on either end are grounded, while the center segment has the confining potential U_L applied.

4.4 The linear Paul trap

The preceding analysis was given in the context of a generic LQ device. To operate the LQ as a Paul trap a method of longitudinal confinement must be added in order for the ions to be confined in all three dimensions. In this work this was accomplished by segmenting the parallel electrodes and applying a static longitudinal voltage U_L as in Figure 4.4.1.

Referring to this figure, we can see that the application of U_L sets up a potential minimum along the trap axis at the center of the second rod. This potential is approximately parabolic along the trap axis and is given by,

$$\Phi_L(r = 0, z) = U_L \frac{z^2}{z_0^2} \quad (4.4.1)$$

where z_0 is half the length of the trapping segment.

Unfortunately, away from the trap axis the potential does pick up a radial component due to the fact that the field lines originating from the inside faces of the electrodes cannot all converge at the center of the trap. To see how this happens we consider

Laplace's equation in the region of the trap center in cylindrical coordinates (r, ϕ, z) ,

$$\nabla^2 \Phi_L = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \Phi_L \right) + \frac{\partial^2}{\partial z^2} \Phi_L = 0 \quad (4.4.2)$$

where we have neglected the azimuthal term as Φ_L is independent of this coordinate. Inserting Equation 4.4.1 and integrating the above twice gives,

$$\Phi_L(r) = -\frac{1}{2} U_L \frac{r^2}{z_0^2} \quad (4.4.3)$$

The total radial depth of the potential well is thus composed of a component due to the oscillating field (Equation 4.2.9 with $U = 0$) and a component from the longitudinal confining potential,

$$\begin{aligned} D_r &= \frac{Q^2 V^2}{4m\Omega^2 r_0^2} - \frac{1}{2} Q U_L \frac{r_0^2}{z_0^2} \\ &= \frac{QV}{8} |q_u| - \frac{1}{2} Q U_L \frac{r_0^2}{z_0^2} \end{aligned} \quad (4.4.4)$$

The depth along the trap axis is simply,

$$D_L = Q U_L \quad (4.4.5)$$

Unfortunately, what these two equations imply is that the longitudinal confinement cannot be made as large as one wants due to the radial component of the field.

Note that we have neglected the static voltage U in the above derivation as it is almost always the case for a Paul trap that the static voltage U is set to zero. This configuration produces the largest combined acceptance region in both the x and y directions (see Figure 4.7.4)².

²It might be beneficial, however, to use a non-zero U as this will increase the acceptance in the x direction if $U > 0$ and in the y direction if $U < 0$.

4.5 The quadrupole mass filter (QMF)

Another common application of the LQ device is for the use as a quadrupole mass spectrometer/filter (QMF). As the stability parameter q is inversely proportional to the mass of the ion, with $U = 0$ the QMF acts as a high pass filter with a lower mass limit given by,

$$m_{low} = \frac{2QV}{q_{max}\Omega^2 r_0^2} \quad (4.5.1)$$

where $q_{max} = 0.908$ for $U = 0$. If, however, $U \neq 0$, the QMF becomes a band pass filter with a lower limit given by Equation 4.5.1 and an upper limit given by,

$$m_{up} = \frac{2QV}{q_{low}\Omega^2 r_0^2} \quad (4.5.2)$$

In this case q_{max} (q_{low}) are the maximum (minimum) values of q for the given a that still lie within the stability region (Figure 4.3.1).

The QMF can also be used to produce a high resolution mass spectrum by quickly scanning over the value of U . In this way the only ions emitted from the QMF will be those with “instantaneous” values of a and q that lie within the stability region. If one then measures the number of particles with a detector placed on the output side of the QMF and the timing of the scan and detector counts are appropriately calibrated then a mass spectrum can be created.

The above discussion must be taken with some caution, however, since it is assumed that the ion spends enough time within the trap such that it becomes unstable and falls on one of the QMF electrodes. This in turn depends on the length of the trap and the energy of the ions entering it. It is for this reason that the trap length and ion energy are the most crucial parameters defining the mass-resolution of the QMF.

4.6 Trapping in a buffer gas

So far we have only considered the case of an isolated ion. In reality we will need some way to cool the ions in order to trap them. There are a few different methods that may be used to do this inside a linear Paul trap, however, in this work a buffer gas is exclusively used and only this method will be discussed.

Due to its simplicity, buffer gas cooling was the earliest method used for cooling heavy ions. The method involves filling the trap with an inert gas at a pressure in the range of 10^{-6} - 10^{-3} mbar and relies on collisions between the ion and the buffer gas to achieve ion cooling. The efficiency of cooling depends on the average energy exchanged in each collision and thus on the relative mass of the buffer gas molecule and ion in question. The downside of this method is that individual collisions can cause a significant change in an ion's trajectory which can potentially knock it out of the trap. The degree to which this happens also depends on the relative masses of the ion and buffer gas.

In each collision there will be some degree of randomness introduced into the phase of the micromotion of the ion. As the micromotion must always be coherent with the driving frequency of the trap this randomness is subsequently transferred to the macromotion of the ion. The overall effect is that the ion draws energy from the RF field and is heated up (RF-heating). If, however, the average ion collision removes more energy from the ion than that provided from RF-heating an overall cooling effect will occur.

If we write the mass of the buffer gas as M_B and the mass of the ion as M_I then there exist three different regimes of buffer gas cooling,

1. Stable regime, $M_B \ll M_I$
2. Efficient cooling regime, $M_B \sim M_I$
3. Unstable regime, $M_B \gg M_I$

In the stable regime the buffer gas is much less massive than the ion and each collision

will only slightly shift the ion's trajectory. RF-heating is thus negligible and there is an overall cooling effect with a small probability for the ion to be lost.

In the second regime the ion and buffer gas masses are approximately equal and thus the greatest amount of energy is exchanged leading to rapid cooling. However, the number of collisions producing a large shift in the ion trajectory will be larger in this case leading to a higher probability for the ion to be ejected from the trap.

In the case that the buffer gas is heavier than the ion, each collision will introduce a high degree of randomness into the ion's micromotion, RF-heating will be substantial, and the ion will be quickly lost.

Whether or not one works in the first regime or near the second depends on the experimental conditions and goals. For example, if one needs long trapping times then it is clear that one should choose $M_B \ll M_I$. However, if the ions entering the trap are too energetic then the buffer gas may not be able to cool them and the ions may not get trapped at all. The main goal of this work was to demonstrate the trapping and identification of single barium ions and thus the only concern was with ion lifetime. For this reason helium was used exclusively as a buffer gas.

4.7 Acceptance and emittance of the linear quadrupole

For the Ba-tagging technique to succeed, the $^{136}\text{Ba}^{++}$ created in the $0\nu\beta\beta$ -decay must be extracted from the LXe chamber, converted into a singly charged species, trapped, and identified with as close to 100% efficiency as is possible. In order to maximize efficient transfer of the ion between the different stages involved in the Ba-tagging scheme, the acceptance of each component must be optimally matched to the emittance of the preceding stage. In this section the acceptance of a LQ is investigated.

The analysis given here follows that given in [78] and assumes the ideal case of an infinitely long ion trap without the presence of a buffer gas. To begin, we start with the

Mathieu equation (Equation 4.3.6), which is reproduced here for convenience (with an extra initial phase, τ_0 , included),

$$\ddot{u}(\tau) + [a_u - 2q_u \cos 2(\tau - \tau_0)]u = 0, \quad u \in x, y \quad (4.7.1)$$

We can represent the solution to the above as a series of step functions of width T , where, if we take T to be small enough, then we should approach the actual solution as given in Equation 4.3.7. Let us consider the first such interval of width T and let,

$$h^2 = a_u - 2q_u \cos 2(\tau - \tau_0) \quad (4.7.2)$$

where we assume that h^2 is constant over the interval.

Now, if $h^2 > 0$ then the Mathieu equation takes the form,

$$\ddot{u}(\tau) + h^2 u = 0$$

The solution to the above is well known. The position and velocity of the ion are given by,

$$u(\tau) = u_0 \cos h\tau + \frac{\dot{u}_0}{h} \sin h\tau \quad (4.7.3)$$

$$\dot{u}(\tau) = -u_0 \sin h\tau + \dot{u}_0 \cos h\tau \quad (4.7.4)$$

where u_0 and $\dot{u}_0$ are the initial position and velocity of the ion respectively. We can equivalently represent this solution as,

$$\begin{bmatrix} u \\ \dot{u} \end{bmatrix}_T = M_1 \begin{bmatrix} u_0 \\ \dot{u}_0 \end{bmatrix} \quad (4.7.5)$$

where,

$$M_1 = \begin{bmatrix} \cos h\tau & \frac{\sin h\tau}{h} \\ -h \sin h\tau & \cos h\tau \end{bmatrix} \quad (4.7.6)$$

After another interval of size T the solution takes the form,

$$\begin{aligned} \begin{bmatrix} u \\ \dot{u} \end{bmatrix}_{2T} &= M_2 \begin{bmatrix} u \\ \dot{u} \end{bmatrix}_T \\ &= M_2 \cdot M_1 \begin{bmatrix} u_0 \\ \dot{u}_0 \end{bmatrix} \end{aligned} \quad (4.7.7)$$

If we continue in this fashion we can determine the solution after n such intervals as,

$$\begin{bmatrix} u \\ \dot{u} \end{bmatrix}_{nT} = M_n \begin{bmatrix} u \\ \dot{u} \end{bmatrix}_{(n-1)T} \quad (4.7.8)$$

$$= M_n \cdot M_{n-1} \cdot M_{n-2} \cdot \dots \cdot M_2 \cdot M_1 \begin{bmatrix} u_0 \\ \dot{u}_0 \end{bmatrix} \quad (4.7.9)$$

If $h^2 < 0$ over the interval in question, then we simply need to replace the trigonometric functions with their hyperbolic forms (and remove the minus sign on the $-h \sin h\tau$ term as the derivative of $\cosh$ does not pick up a minus sign). The position and velocity after the first interval is,

$$\begin{bmatrix} u \\ \dot{u} \end{bmatrix}_T = M'_1 \begin{bmatrix} u_0 \\ \dot{u}_0 \end{bmatrix} \quad (4.7.10)$$

where,

$$M'_1 = \begin{bmatrix} \cosh h\tau & \frac{\sinh h\tau}{h} \\ h \sinh h\tau & \cosh h\tau \end{bmatrix} \quad (4.7.11)$$

After n intervals we have,

$$\begin{bmatrix} u \\ \dot{u} \end{bmatrix}_{nT} = M'_n \cdot M'_{n-1} \cdot M'_{n-2} \cdots M'_2 \cdot M'_1 \begin{bmatrix} u_0 \\ \dot{u}_0 \end{bmatrix} \quad (4.7.12)$$

By combining the M_n and M'_n matrices appropriately it is easy to determine the matrix for one complete field cycle, M_Ω . We can then proceed to determine the solution after any integer number N of field cycles,

$$\begin{bmatrix} u \\ \dot{u} \end{bmatrix}_N = M_\Omega^N \begin{bmatrix} u_0 \\ \dot{u}_0 \end{bmatrix} \quad (4.7.13)$$

This method is particularly easy to use since it only involves matrix multiplications and the interval width T can be made as small as one wants in order to get the required precision. More importantly for our purpose, the matrix elements M_Ω contain useful information about the acceptance of the ion trap.

If one uses M_Ω to calculate u and $\dot{u}$ for a given initial phase τ_0 the points will fall on an ellipse in $[u, \dot{u}]$ phase space given by [78],

$$\Gamma u^2 + 2A u \dot{u} + B \dot{u}^2 = \epsilon \quad (4.7.14)$$

with,

$$A = \sqrt{B\Gamma - 1} \quad (4.7.15)$$

The parameters Γ , A , and B are determined from the matrix elements of M_Ω . In fact, it can be shown that [78],

$$M_{\Omega} = \begin{bmatrix} \cos \pi\beta + A \sin \pi\beta & B \sin \pi\beta \\ -\Gamma \sin \pi\beta & \cos \pi\beta - A \sin \pi\beta \end{bmatrix} \quad (4.7.16)$$

where β is the usual Mathieu stability parameter. The parameter ϵ is called the acceptance and is equal to the area of the ellipse divided by π .

We can determine the major and minor axes by comparing Equation 4.7.14 to the standard equation of an ellipse centered at the origin with it's major axes rotated by an angle θ . Doing this we find that,

$$L_{maj} = \sqrt{\frac{\epsilon}{2}} \left(\sqrt{\frac{B+\Gamma}{2} + 1} + \sqrt{\frac{B+\Gamma}{2} - 1} \right) \quad (4.7.17)$$

$$L_{min} = \sqrt{\frac{\epsilon}{2}} \left(\sqrt{\frac{B+\Gamma}{2} + 1} - \sqrt{\frac{B+\Gamma}{2} - 1} \right) \quad (4.7.18)$$

where the angle θ is given by,

$$\tan 2\theta = -\frac{2A}{B-\Gamma} \quad (4.7.19)$$

For a given initial phase, the maximum displacement along a trajectory is given by [79],

$$u_{max} = \sqrt{\epsilon B_{max}} \quad (4.7.20)$$

where B_{max} is the maximum value of B regardless of the initial phase.

Physically, the ellipse represents all combinations of initial u and $\dot{u}$ that, for a given initial phase, will produce a stable trajectory. To make a connection to the physical ion trap we just need to set u_{max} (as given in Equation 4.7.20) equal to the radius of the trap, r_0 . The area of the ellipse thus scales as Ωr_0^2 and is given by³,

³The factor of Ω comes from the fact that we have been using the normalized velocity, $\dot{u} = \frac{du}{d\tau} = \frac{2}{\Omega} \frac{du}{dt}$.

$$Area = \pi\epsilon = \frac{r_0^2}{B_{max}} \quad (4.7.21)$$

Since the x and y directions are both scaled according to the above, the total acceptance area of the trap scales as $\Omega^2 r_0^4$.

Ellipses for three different initial phases are shown in Figure 4.7.1 for the operating parameters $a = 0$ ($U = 0$), and $q = 0.416$. The 100% transmission window, regardless of the initial phase, can be built by the intersection of all such ellipses. This is shown in Figure 4.7.2 for the operational parameters $a = 0$, and $q = 0.416$. In this case, since $a = 0$, the 100% transmission window is identical for both the x and y directions. If, however, the parameter $a \neq 0$, the acceptance window will be larger (smaller) in the x (y) coordinate for $a_x > 0$ ($a_y < 0$) and *vice versa* for $a_x < 0$ ($a_y > 0$). This is illustrated in Figure 4.7.3 where $a = 0.0832$ and $q = 0.416$. Setting $a \neq 0$ may be beneficial for an ion trap, if, for example, one is attempting to match the acceptance of the trap to a highly polarized beam of ions. However, the total combined area of the x and y windows does decrease as a increases, so caution should be used if considering this option (see Figure 4.7.4).

As is evident from Figure 4.7.1 the region in phase space that produces 100% transmission efficiency shrinks substantially if we consider a random initial phase for the ion. If the phase of the ion entering the trap was matched to the initial phase of the RF as the ion enters then it is not unreasonable to think that a large increase in transmission efficiency could be achieved. Unfortunately, there was not enough time to investigate this phenomenon further.

The preceding analysis has assumed the ideal case of an infinitely long ion trap without the presence of fringing fields (distortions around the ends of the trap). In general, for a finite sized trap, the acceptance will increase as the trap decreases in length. The reason for this, as mentioned in the last section, is that the ion does not spend enough time inside the trap to become unstable. If fringing fields are present then the acceptance of

Figure 4.7.1: Acceptance ellipses for initial phases of $\tau_0 = 0$ (solid line), $\tau_0 = \frac{\pi}{4}$ (small dashes), $\tau_0 = \frac{\pi}{2}$ (large dashes). The operational parameters are $a = 0$, and $q = 0.416$ in all cases.

Figure 4.7.2: Visualization of the 100% transmission window for the operational parameters $a = 0$, and $q = 0.416$. When $a = 0$ the window has the same area for both the x and y directions.

Figure 4.7.3: Comparison of 100% transmission windows for the x -coordinate (left) and the y -coordinate (right). The operational parameters are $a = 0.0832$ and $q = 0.416$ with $a_x > 0$.

Figure 4.7.4: The area of the 100% transmission windows as a function of the stability parameter a for the x and y coordinates (left) and the total combined area for both coordinates (right). It is assumed $a_x > 0$ and $q = 0.416$ in all cases.

Figure 4.8.1: Schematic of a sextupole (left) and octupole (right).

the trap can be increased or decreased depending on the length of the region in which the fringing fields are present. For a detailed analysis on the effect of fringing fields see [78].

Finally, if a buffer gas is used to cool the ions then the acceptance of the ion trap will be slightly larger, since, on average, the ion will decrease in speed as it travels through the ion trap allowing it to have a wider range of stable radial displacements.

4.8 Multipole ion guides

In a similar manner to the quadrupole trap, higher order multipole traps may be constructed by simply adding more rods to the trap structure. In general, the n^{th} order multipole trap will contain $2n$ parallel rods equally spaced around a cylinder of radius r_0 at angular positions $j\pi/n$, $j = 1, 2, 3 \dots$. The RF voltage is applied exactly as in the quadrupole case with adjacent electrodes being completely out of phase with one another. As an example Figure 4.8.1 shows configurations for the $n = 3$ (sextupole) and $n = 4$ (octupole) multipoles.

The theory of confinement for multipole traps is similar to that of quadrupoles and can also be described by an effective potential [77],

$$\Phi_{eff} = \frac{QV^2n^2}{4m\Omega^2r_0^2} \left(\frac{r}{r_0} \right)^{2n-2} \quad (4.8.1)$$

This potential is plotted for $n = 2, 3, 4, 10, 20$ in Figure 4.8.2. We can immediately see from this that as n gets large the radial intensity of the multipole field remains small for a larger range of distances from the trap axis. This has the consequence that the acceptance of the multipole increases with n . It is due to this increased acceptance that higher order multipoles are almost always used as either ion guides (for propagating ions between different system components) or as collision cells (used for breaking up large molecules, or to induce charge exchange reactions). A sextupole and octupole are being investigated at Carleton for use in the charge exchange reaction to convert $\text{Ba}^{++} \rightarrow \text{Ba}^+$.

Figure 4.8.2: Effective potentials for a quadrupole, sextupole, octupole, and a 20-pole as a function of the distance from the center of the trap.

Chapter 5

Theory of laser induced fluorescence

The next step in the Ba-tagging scheme is the identification of the $^{136}\text{Ba}^+$ ion. For this task the singly-charged barium ion is ideal as it has many properties that make it particularly suited for spectroscopic measurements. In fact, the Ba^+ ion was used in the first attempt ever to detect trapped ions via laser fluorescence [80] and was also the first atomic particle to ever be optically observed [81].

The electronic structure of Ba^+ is that of a xenon core with a lone 6s valence electron. It has two dipole allowed transitions between its lowest lying energy levels, $6^2\text{P}_{1/2} \rightarrow 6^2\text{S}_{1/2}$ (494 nm, blue) and $6^2\text{P}_{1/2} \rightarrow 5^2\text{D}_{3/2}$ (650 nm, red). Furthermore, the $\text{D} \rightarrow \text{S}$ transition is dipole forbidden but has an allowed quadrupole transition making the $5^2\text{D}_{3/2}$ state metastable with the extremely long lifetime of $34.5 \pm 3.5\text{s}$ [82].

In this chapter the theory of atom-photon interactions is discussed with the goal of deriving a means to calculate the fluorescence rate of the Ba^+ ion. In the first section the Ba-II system is introduced and the energy level structure is specified. This is followed by a description of atom-photon interactions with a focus on electric-dipole transitions. After this, the density matrix formalism which is used to analyze the dynamics is introduced, and, finally, I end by deriving the optical Bloch equations which can be integrated to determine the expected fluorescence rate.

Figure 5.1.1: Energy level scheme for the Ba-II system.

Table 5.1: Vacuum wavelength (λ) [83], branching ratio (BR), spontaneous emission rate (Γ), and reduced dipole matrix element ($\langle ||D|| \rangle$) for the Ba-II transitions [85]. The wavelengths were given in air in [83] and converted to vacuum wavelengths using refractive indices taken from [84].

Transition	$\lambda(\text{nm})$	BR (%)	Γ (MHz)	$\langle D \rangle$ (a.u.)
$6^2P_{1/2} \rightarrow 6^2S_{1/2}$	493.54544	72.93 ± 0.02	92.1 ± 0.9	3.305 ± 0.014
$6^2P_{1/2} \rightarrow 5^2D_{3/2}$	649.86934	27.07 ± 0.02	34.2 ± 0.3	3.042 ± 0.016

5.1 The Ba-II system

We define the Ba-II system to be the three lowest lying energy levels of the Ba^+ ion as shown in Figure 5.1.1. The important properties of the transitions are summarized in Table 5.1.

The system is made to fluoresce using a laser tuned to the $S \rightarrow P$ transition to create an excited P-state population which will spontaneously decay according to the branching ratios given in Table 5.1. Eventually all the ions would be pumped into the meta-stable D-state and fluorescence would cease. For this reason another laser tuned to the $D \rightarrow P$ transition is necessary to pump the ions back into the P-state such that fluorescence can resume. The Ba-II system is an example of what is called a three-level Λ -system.

5.2 The dipole interaction

The interaction of a charged particle with electromagnetic fields is treated in detail in many introductory quantum mechanics texts and thus I will only summarize the main aspects. What is presented here follows that given in [86].

We will assume for now only two states labeled by $|2\rangle$ and $|1\rangle$ with energies $E_2 = \hbar\omega_2$ and $E_1 = \hbar\omega_1$ respectively, where $E_2 > E_1$. In the presence of a monochromatic plane wave of frequency ω , and vector amplitude $\vec{A}_0$,

$$\vec{A} = 2\vec{A}_0 \cos\left(\frac{\omega}{c}\hat{n} \cdot \vec{r} - \omega t\right) \quad (5.2.1)$$

the Hamiltonian can be written as,

$$H = \frac{\vec{p}^2}{2m_e} - e\phi(\vec{r}) - \frac{e}{m_e c} \vec{A} \cdot \vec{p} \quad (5.2.2)$$

where the Coulomb gauge ($\vec{\nabla} \cdot \vec{A} = 0$) is assumed, the $|\vec{A}|^2$ term has been neglected, and $\phi(\vec{r})$ is the electric potential.

Expanding the cosine in the vector potential $\vec{A}$ gives,

$$\vec{A} = \vec{A}_0 [e^{i(\omega/c)\hat{n} \cdot \vec{r} - i\omega t} + e^{-i(\omega/c)\hat{n} \cdot \vec{r} + i\omega t}] \quad (5.2.3)$$

and inserting this into the Hamiltonian we get,

$$H = H_0 + H_+ + H_-$$

$$H_0 = \frac{\vec{p}^2}{2m_e} + e\phi(\vec{r})$$

$$H_{\pm} = -\frac{e}{m_e c} \vec{A}_0 \cdot \vec{p} e^{i(\omega/c)\hat{n} \cdot \vec{r} \mp i\omega t} \quad (5.2.4)$$

The radiation field couples the two states through the $H_{\pm}$ terms with H_+ corresponding to photon absorption and H_- photon emission. The transition rates are obtained using Fermi's golden rule,

$$\Gamma_{\pm} = \frac{2\pi}{\hbar^2} \frac{e^2}{m_e^2 c^2} |\langle 2 | \vec{A}_0 \cdot \vec{p} e^{\pm i(\omega/c)\hat{n} \cdot \vec{r}} | 1 \rangle|^2 \delta(\omega_{21} - \omega) \quad (5.2.5)$$

where $\delta(\omega_{21} - \omega)$ is an energy conserving δ -function and $\omega_{21} = \omega_2 - \omega_1$. In the case where spontaneous emission is present, the excited state will have a lifetime given by the inverse of the spontaneous decay rate, Γ . The δ -function is thus interpreted as the limiting case $\Gamma \rightarrow 0$,

$$\pi \delta(\omega_{21} - \omega) = \lim_{\Gamma \rightarrow 0} \frac{\Gamma/2}{(\omega_{21} - \omega)^2 + \Gamma^2/4} \quad (5.2.6)$$

In the electric dipole approximation only the first term of the expansion $e^{i(\omega/c)\hat{n} \cdot \vec{r}} = 1 + i\frac{\omega}{c}\hat{n} \cdot \vec{r} + \dots$ is kept such that the transition rate can be written,

$$\Gamma_{\pm} = \frac{2\pi}{\hbar^2} \frac{e^2}{m_e^2} |\langle 2 | \vec{A}_0 \cdot \vec{p} | 1 \rangle|^2 \delta(\omega_{21} - \omega) \quad (5.2.7)$$

Furthermore, due to the fact that,

$$\vec{E} = -\frac{1}{c} \frac{\partial \vec{A}}{\partial t} \quad (5.2.8)$$

the dipole approximation implies the intuitive result,

$$\frac{e}{m_e c^2} \vec{A} \cdot \vec{p} = e \vec{E} \cdot \vec{r} \quad (5.2.9)$$

where the above represents the energy of a dipole with moment $\vec{d} = e\vec{r}$ in an electric field $\vec{E}$.

The matrix element can now be written,

$$\Gamma_{\pm} = \frac{2\pi}{\hbar^2} |\vec{E} \cdot \langle 2|\vec{d}|1 \rangle|^2 \delta(\omega_{21} - \omega) \quad (5.2.10)$$

Assuming a spontaneous decay rate of Γ we get our final result for the transition rate,

$$\Gamma_{\pm} = \frac{|\vec{E} \cdot \langle 2|\vec{d}|1 \rangle|^2}{\hbar^2} \frac{\Gamma}{\delta^2 + \Gamma^2/4} \quad (5.2.11)$$

where $\delta = \omega_{12} - \omega$ is the detuning from resonance.

The above result displays some of the general aspects of the interaction of photons and atoms. First, we can see that the transition rate is proportional to the intensity of the incident laser and the rate is equal for both absorption and emission of photons, $\Gamma_+ = \Gamma_-$. In addition, since the dipole operator $\vec{d}$ is a spherical tensor of rank 1, the Wigner-Eckart theorem can be applied to immediately read off the usual dipole selection rules $\Delta l = l' - l = \pm 1$ and $\Delta m = m' - m = 0, \pm 1$. Finally, the linewidth of the transition will have a Lorentzian profile with full-width-half-maximum (FWHM) given by the spontaneous decay rate Γ .

5.3 Density matrix formalism

The density matrix formalism is a powerful tool to analyze the statistical dynamics of a quantum system and is specifically useful in the presence of spontaneous emission. In this section the properties and time evolution of the density matrix are investigated. The theory presented here follows that given in [86].

5.3.1 The density matrix

Consider an ensemble of particles. If all of the particles are in some definite state $|\alpha\rangle$ then the ensemble is said to be pure and we can represent the entire ensemble by the state $|\alpha\rangle$. The expectation value of some operator A for a pure ensemble is just the usual

quantum mechanical expression,

$$\begin{aligned}\langle A \rangle &= \langle \alpha | A | \alpha \rangle \\ &= \sum_j |\langle a_j | \alpha \rangle|^2 \langle a_j | A | a_j \rangle\end{aligned}\tag{5.3.1}$$

where $|a_j\rangle$ is an eigenstate of A . In a more general basis defined by the states $|b_k\rangle$ we can write the above as,

$$\begin{aligned}\langle A \rangle &= \sum_i \sum_j \langle \alpha | b_i \rangle \langle b_i | A | b_j \rangle \langle b_j | \alpha \rangle \\ &= \sum_i \sum_j (\langle b_j | \alpha \rangle \langle \alpha | b_i \rangle) \langle b_i | A | b_j \rangle\end{aligned}\tag{5.3.2}$$

For a system in which each particle is in some state $|\alpha_i\rangle$ the ensemble will be a random mixture of different states, and, not suprisingly, is called a mixed ensemble. We can no longer represent the ensemble by a single state and Equation 5.3.1 no longer applies. To represent the expectation value of A for a mixed ensemble we introduce the fractional populations w_i which represent the probability of a particle in the ensemble to be in the state $|\alpha_i\rangle$.

The expectation value A is then calculated as the ensemble average,

$$\begin{aligned}[A] &= \sum_i w_i \langle \alpha_i | A | \alpha_i \rangle \\ &= \sum_i \sum_j w_i |\langle a_j | \alpha_i \rangle|^2 \langle a_j | A | a_j \rangle\end{aligned}\tag{5.3.3}$$

where the square brackets around A are to remind us that this is an ensemble average.

In the more general basis we have,

$$[A] = \sum_j \sum_k \left(\sum_i w_i \langle b_k | \alpha_i \rangle \langle \alpha_i | b_j \rangle \right) \langle b_j | A | b_k \rangle \quad (5.3.4)$$

The term in brackets is independent of the specific operator A and thus only describes the properties of the ensemble. This leads us to define the Hermitian operator ρ , called the density operator,

$$\rho \equiv \sum_i w_i |\alpha_i\rangle \langle \alpha_i| \quad (5.3.5)$$

with corresponding density matrix elements,

$$\langle b_k | \rho | b_j \rangle = \sum_i w_i \langle b_k | \alpha_i \rangle \langle \alpha_i | b_j \rangle \quad (5.3.6)$$

The expectation value of A then becomes,

$$\begin{aligned} [A] &= \sum_j \sum_k \langle b_k | \rho | b_j \rangle \langle b_j | A | b_k \rangle \\ &= \text{Tr}(\rho A) \end{aligned} \quad (5.3.7)$$

Since the trace is independent of a change-of-basis, the above relation can be used with whatever basis is convenient.

The density matrix has the property that it is normalized,

$$\text{Tr}(\rho) = 1$$

and, if it represents a pure state,

$$\text{Tr}(\rho^2) = 1$$

5.3.2 Time evolution of the density operator

Consider a state $|\alpha(t)\rangle$ that evolves in time according to some unitary time evolution operator $\mathcal{U}$ such that,

$$|\alpha(t)\rangle = \mathcal{U}(t, t_0) |\alpha(t_0)\rangle \quad (5.3.8)$$

where the state at some initial time t_0 is represented by $|\alpha(t_0)\rangle$. Obviously we have,

$$\mathcal{U}(t_0, t_0) = 1 \quad (5.3.9)$$

The Schrödinger equation for the state $|\alpha(t)\rangle$ is,

$$i\hbar \frac{d}{dt} |\alpha(t)\rangle = \mathcal{H} |\alpha(t)\rangle \quad (5.3.10)$$

which we can rewrite using Equation 5.3.8 as,

$$i\hbar \left(\frac{\partial \mathcal{U}(t, t_0)}{\partial t} |\alpha(t_0)\rangle + \mathcal{U}(t, t_0) \frac{\partial |\alpha(t_0)\rangle}{\partial t} \right) = \mathcal{H} \mathcal{U}(t, t_0) |\alpha(t_0)\rangle \quad (5.3.11)$$

Now, the state $|\alpha(t_0)\rangle$ does not depend on time and thus the second term on the left-hand-side of Equation 5.3.11 is zero. We then have,

$$i\hbar \frac{\partial \mathcal{U}(t, t_0)}{\partial t} |\alpha(t_0)\rangle = \mathcal{H} \mathcal{U}(t, t_0) |\alpha(t_0)\rangle \quad (5.3.12)$$

or,

$$i\hbar \frac{\partial \mathcal{U}(t, t_0)}{\partial t} = \mathcal{H} \mathcal{U}(t, t_0) \quad (5.3.13)$$

which is just the Schrödinger equation for the operator $\mathcal{U}(t, t_0)$. If $\mathcal{H}$ does not depend explicitly on time then the solution to the above is,

$$\begin{aligned}
\mathcal{U}(t, t_0) &= \mathcal{U}(t_0, t_0) e^{-\frac{i}{\hbar} \mathcal{H}(t-t_0)} \\
&= e^{-\frac{i}{\hbar} \mathcal{H}(t-t_0)}
\end{aligned} \tag{5.3.14}$$

If we now consider some operator A that evolves with the state $|\alpha(t)\rangle$ according to Equation 5.3.8 then we can write the expectation value of A as,

$$\begin{aligned}
\langle A \rangle &= \langle \alpha(t) | A | \alpha(t) \rangle \\
&= \langle \alpha(t_0) | \mathcal{U}^\dagger(t, t_0) A \mathcal{U}(t, t_0) | \alpha(t_0) \rangle \\
&= \langle \alpha(t_0) | A_0 | \alpha(t_0) \rangle
\end{aligned}$$

where A_0 has been defined as,

$$A_0 = \mathcal{U}^\dagger(t, t_0) A \mathcal{U}(t, t_0) \tag{5.3.15}$$

Multiplying the above on the right by $\mathcal{U}^\dagger$ and on the left by $\mathcal{U}$ we get,

$$A = \mathcal{U}(t, t_0) A_0 \mathcal{U}^\dagger(t, t_0) \tag{5.3.16}$$

The time evolution of A is then obtained by differentiating the above. We get,

$$\frac{dA}{dt} = \frac{\partial \mathcal{U}}{\partial t} A_0 \mathcal{U}^\dagger + \mathcal{U} \frac{\partial A_0}{\partial t} \mathcal{U}^\dagger + \mathcal{U} A_0 \frac{\partial \mathcal{U}^\dagger}{\partial t} \tag{5.3.17}$$

Using Equation 5.3.13 and 5.3.16 we can rewrite the above as,

$$\begin{aligned}
\frac{dA}{dt} &= -\frac{i}{\hbar}\mathcal{H}A + \frac{\partial A}{\partial t} + \frac{i}{\hbar}A\mathcal{H} \\
&= -\frac{i}{\hbar}[\mathcal{H}, A] + \frac{\partial A}{\partial t}
\end{aligned} \tag{5.3.18}$$

If A is the density matrix ρ then the above equation is known as the Liouville equation,

$$\begin{aligned}
\frac{d\rho}{dt} &= -\frac{i}{\hbar}[\mathcal{H}, \rho] + \frac{\partial \rho}{\partial t} \\
&= -\frac{i}{\hbar}[\mathcal{H}, \rho] + \gamma\rho
\end{aligned} \tag{5.3.19}$$

where γ is commonly termed the relaxation matrix. In our case γ represents the spontaneous emission rates between the states $|n\rangle$ and is given by,

$$\gamma_{mn} = \frac{1}{2}(\Gamma_n + \Gamma_m) \tag{5.3.20}$$

such that,

$$(\gamma\rho)_{mn} = \gamma_{mn} \cdot \rho_{mn} \tag{5.3.21}$$

5.4 The optical Bloch equations

We now have all the tools needed to describe the full quantum dynamics of the Ba-II system. The theory presented here follows that given in [88].

To fix the notation used, I will represent the eigenstates of the Ba-II system as,

$$\begin{aligned}
|a\rangle &= |6^2P_{1/2}\rangle \\
|b\rangle &= |6^2S_{1/2}\rangle \\
|c\rangle &= |5^2D_{3/2}\rangle
\end{aligned} \tag{5.4.1}$$

Furthermore, let ω_B (ω_R) be the frequency of the laser corresponding to the $S \rightarrow P$ ($D \rightarrow P$) transition, and $\hbar\omega_j$, $j \in [a, b, c]$ is the energy of the j^{th} level. In the presence of the two laser fields the Hamiltonian of the system is given by,

$$H = H_0 + H_B + H_R \tag{5.4.2}$$

where the terms H_B and H_R represent the interaction of the ion with the laser fields, and H_0 is the Hamiltonian of the unperturbed system such that,

$$H_0 |j\rangle = \hbar\omega_j |j\rangle \tag{5.4.3}$$

In the dipole approximation we can represent H_B and H_R as,

$$H_j = -\vec{d} \cdot \vec{E}_j \cos \omega_j t, \quad j \in [B, R] \tag{5.4.4}$$

where $\vec{E}_B$ ($\vec{E}_R$) is the vector amplitude of the blue (red) laser, and $\vec{d}$ is the dipole operator for the transition between states $|m\rangle$ and $|n\rangle$,

$$\vec{d} = \vec{d}_{nm}(|n\rangle \langle m| + |m\rangle \langle n|) \tag{5.4.5}$$

where $\vec{d}_{nm}$ is the dipole matrix element. The term $|m\rangle \langle n|$ is just the raising operator which raises a state from $|n\rangle$ to $|m\rangle$. Likewise, $|n\rangle \langle m|$ lowers a state from $|m\rangle$ to $|n\rangle$. Rewriting H_B and H_R in terms of the raising and lowering operators and expanding the

cosines gives,

$$H_B = -\frac{1}{2} \langle b | \vec{d} | a \rangle \cdot \vec{E}_B \left[|a\rangle \langle b| (e^{i\omega_B t} + e^{-i\omega_B t}) + |b\rangle \langle a| (e^{i\omega_B t} + e^{-i\omega_B t}) \right] \quad (5.4.6)$$

$$H_R = -\frac{1}{2} \langle c | \vec{d} | a \rangle \cdot \vec{E}_R \left[|a\rangle \langle c| (e^{i\omega_R t} + e^{-i\omega_R t}) + |c\rangle \langle a| (e^{i\omega_R t} + e^{-i\omega_R t}) \right] \quad (5.4.7)$$

We interpret the terms $e^{\pm i\omega_j t}$ as corresponding to photon emission and absorption respectively.

To simplify Equation 5.4.6 and 5.4.7 we apply the rotating wave approximation (RWA) and ignore terms like $\langle a | |b\rangle e^{i\omega_B t}$ and $\langle b | |a\rangle e^{-i\omega_B t}$ which correspond to photon absorption with the state lowering from $|a\rangle$ to $|b\rangle$, and photon emission with the state raising from $|b\rangle$ to $|a\rangle$. Thus, under the RWA, we have,

$$H_B^{RWA} = \frac{1}{2} \hbar \Omega_B \left(|a\rangle \langle b| e^{i\omega_B t} + |b\rangle \langle a| e^{-i\omega_B t} \right) \quad (5.4.8)$$

$$H_R^{RWA} = \frac{1}{2} \hbar \Omega_R \left(|a\rangle \langle c| e^{i\omega_R t} + |c\rangle \langle a| e^{-i\omega_R t} \right) \quad (5.4.9)$$

where Ω_B and Ω_R are the Rabi frequencies,

$$\hbar \Omega_B = -\langle b | \vec{d} | a \rangle \cdot \vec{E}_B \quad (5.4.10)$$

$$\hbar \Omega_R = -\langle c | \vec{d} | a \rangle \cdot \vec{E}_R \quad (5.4.11)$$

The total Hamiltonian for the system under the RWA approximation is thus,

$$\begin{aligned}
H_{RWA} &= H_0 + H_B^{RWA} + H_R^{RWA} \\
&= \begin{bmatrix} \hbar\omega_a & \frac{1}{2}\hbar\Omega_B e^{-i\omega_B t} & \frac{1}{2}\hbar\Omega_R e^{-i\omega_R t} \\ \frac{1}{2}\hbar\Omega_B e^{i\omega_B t} & \hbar\omega_b & 0 \\ \frac{1}{2}\hbar\Omega_R e^{i\omega_R t} & 0 & \hbar\omega_c \end{bmatrix} \quad (5.4.12)
\end{aligned}$$

One of our assumptions in deriving the Liouville equation was that the Hamiltonian did not depend explicitly on time and thus the above form is not compatible. Fortunately, there exists a basis related by a unitary transformation in which H_{RWA} does not depend on time. It is easily checked that the transformation,

$$\mathcal{H}_{RWA} \rightarrow T \mathcal{H}_{RWA} T^\dagger - i\hbar T \frac{d}{dt} T^\dagger \quad (5.4.13)$$

with T given by,

$$T = \begin{bmatrix} 1 & 0 & 0 \\ 0 & e^{-i\omega_B t} & 0 \\ 0 & 0 & e^{-i\omega_R t} \end{bmatrix} \quad (5.4.14)$$

has this property. Applying the transformation and shifting the ground state energy we get,

$$\begin{aligned}
\tilde{\mathcal{H}}_{RWA} &= \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega_B & \frac{1}{2}\Omega_R \\ \frac{1}{2}\Omega_B & \omega_B - \omega_{ab} & 0 \\ \frac{1}{2}\Omega_R & 0 & \omega_R - \omega_{ac} \end{bmatrix} \quad (5.4.15)
\end{aligned}$$

$$= \frac{\hbar}{2} \begin{bmatrix} 0 & \Omega_B & \Omega_R \\ \Omega_B & 2\delta_B & 0 \\ \Omega_R & 0 & 2\delta_R \end{bmatrix} \quad (5.4.16)$$

where we have introduced the detunings,

$$\delta_B = \omega_B - \omega_{ab} \quad (5.4.17)$$

$$\delta_R = \omega_R - \omega_{ac} \quad (5.4.18)$$

The density matrix for our three-level system is given by,

$$\rho = \begin{bmatrix} \rho_{aa} & \rho_{ab} & \rho_{ac} \\ \rho_{ba} & \rho_{bb} & \rho_{bc} \\ \rho_{ca} & \rho_{cb} & \rho_{cc} \end{bmatrix} \quad (5.4.19)$$

The diagonal elements give the populations of the three states, while the off diagonal terms are called the coherences and contain information about the relative phases between the states.

Since we have transformed the Hamiltonian we must also transform the density matrix for things to be consistent. Doing this we get,

$$\begin{aligned} \tilde{\rho} &= T\rho T^\dagger \\ &= \begin{bmatrix} \rho_{aa} & \rho_{ab}e^{i\omega_B t} & \rho_{ac}e^{i\omega_R t} \\ \rho_{ba}e^{-i\omega_B t} & \rho_{bb} & \rho_{bc}e^{i(\omega_R - \omega_B)t} \\ \rho_{ca}e^{-i\omega_R t} & \rho_{cb}e^{-i(\omega_R - \omega_B)t} & \rho_{cc} \end{bmatrix} \\ &= \begin{bmatrix} \rho_{aa} & \tilde{\rho}_{ab} & \tilde{\rho}_{ac} \\ \tilde{\rho}_{ba} & \rho_{bb} & \tilde{\rho}_{bc} \\ \tilde{\rho}_{ca} & \tilde{\rho}_{cb} & \rho_{cc} \end{bmatrix} \end{aligned} \quad (5.4.20)$$

where we have introduced the “slow” variables,

$$\tilde{\rho}_{ab} = \rho_{ab} e^{i\omega_B t} \quad (5.4.21)$$

$$\tilde{\rho}_{ac} = \rho_{ac} e^{i\omega_R t} \quad (5.4.22)$$

$$\tilde{\rho}_{bc} = \rho_{bc} e^{i(\omega_R - \omega_B)t} \quad (5.4.23)$$

The relaxation matrix is determined by the spontaneous emission rates for the two transitions. If Γ_B is the rate of spontaneous emission for the $P \rightarrow S$ transition and Γ_R is that for the $P \rightarrow D$ transition, then, with $\Gamma = \Gamma_B + \Gamma_R$, the relaxation matrix is,

$$\gamma\rho = \begin{bmatrix} -\Gamma\rho_{aa} & -\frac{1}{2}\Gamma\tilde{\rho}_{ab} & -\frac{1}{2}\Gamma\tilde{\rho}_{ac} \\ -\frac{1}{2}\Gamma\tilde{\rho}_{ba} & \Gamma_{ab}\rho_{bb} & 0 \\ -\frac{1}{2}\Gamma\tilde{\rho}_{ca} & 0 & \Gamma_{ac}\rho_{cc} \end{bmatrix} \quad (5.4.24)$$

The Liouville equation then becomes,

$$\frac{d\tilde{\rho}}{dt} = -\frac{i}{\hbar}[\tilde{\mathcal{H}}, \tilde{\rho}] + \gamma\tilde{\rho} \quad (5.4.25)$$

The above gives us nine differential equations for the matrix elements of ρ , however, due to the normalization and Hermiticity conditions, only five of these are independent. These equations are known as the Optical Bloch Equations (OBEs). The equations for the diagonal elements are,

$$\frac{\partial\rho_{aa}}{\partial t} = \frac{i}{2} [\Omega_B(\tilde{\rho}_{ab} - \tilde{\rho}_{ba}) + \Omega_R(\tilde{\rho}_{ac} - \tilde{\rho}_{ca})] - \Gamma\rho_{aa} \quad (5.4.26)$$

$$\frac{\partial\rho_{bb}}{\partial t} = -\frac{i}{2}\Omega(\tilde{\rho}_{ab} - \tilde{\rho}_{ba}) + \Gamma_{ab}\rho_{aa} \quad (5.4.27)$$

$$\frac{\partial\rho_{cc}}{\partial t} = -\frac{i}{2}\Omega_R(\tilde{\rho}_{ac} - \tilde{\rho}_{ca}) + \Gamma_{ac}\rho_{cc} \quad (5.4.28)$$

and the off diagonal elements are,

$$\frac{\partial \tilde{\rho}_{ab}}{\partial t} = \frac{i}{2} [\Omega_B(\rho_{aa} - \rho_{bb}) - \Omega_R \tilde{\rho}_{cb} + 2\delta_B \tilde{\rho}_{ab}] - \frac{1}{2} \Gamma \tilde{\rho}_{ab} \quad (5.4.29)$$

$$\frac{\partial \tilde{\rho}_{ac}}{\partial t} = \frac{i}{2} [\Omega_R(\rho_{aa} - \rho_{cc}) - \Omega_B \tilde{\rho}_{bc} + 2\delta_R \tilde{\rho}_{ac}] - \frac{1}{2} \Gamma \tilde{\rho}_{ac} \quad (5.4.30)$$

$$\frac{\partial \tilde{\rho}_{bc}}{\partial t} = \frac{i}{2} [\Omega_R \tilde{\rho}_{ba} - \Omega_B \tilde{\rho}_{ac} + 2(\delta_R - \delta_B) \tilde{\rho}_{bc}] \quad (5.4.31)$$

The solution to the above equations will depend on the exact time structure of the lasers and in general will have to be integrated numerically to determine the populations as a function of time. In the case that the laser beams are continuously on (or at least the pulses are long compared to the lifetime of the P -state) then the equations can be solved in the steady-state by setting the time derivatives to zero. This has been done in [89] and the results are quoted here. The steady-state P-population is,

$$\rho_{aa} = \frac{4}{Z} (\delta_B - \delta_R)^2 \Omega_B^2 \Omega_R^2 \Gamma \quad (5.4.32)$$

where,

$$\begin{aligned} Z &= 8(\delta_B - \delta_R)^2 \Omega_B^2 \Omega_R^2 \Gamma + 4(\delta_B - \delta_R)^2 \Gamma^2 Y \\ &+ 16(\delta_B - \delta_R)^2 (\delta_B^2 \Omega_R^2 \Gamma_B + \delta_R^2 \Omega_B^2 \Gamma_R) \\ &- 8\delta_B (\delta_B - \delta_R) \Omega_R^4 \Gamma_B + 8\delta_R (\delta_B - \delta_R) \Omega_B^4 \Gamma_R \\ &+ (\Omega_B^2 + \Omega_R^2)^2 Y \end{aligned} \quad (5.4.33)$$

and,

$$Y = \Omega_B^2 \Gamma_R + \Omega_R^2 \Gamma_B \quad (5.4.34)$$

The functional form of ρ_{aa} is complicated, however, one thing that is immediately apparent is the presence of the $(\delta_B - \delta_R)$ term in the numerator of Equation 5.4.32. This indicates that the P-state population goes to zero when the detunings of the two lasers are equal. The phenomenon is called the two-photon resonance, and, physically, it represents a coherent transition between the S and D states that bypasses the P-state altogether.

The total fluorescence rate for the Ba-II system is finally obtained by multiplying the P-state population by the spontaneous decay rates. For the $P \rightarrow S$ transition the rate is,

$$\Gamma_B \rho_{aa} \tag{5.4.35}$$

and for the $P \rightarrow D$ transition,

$$\Gamma_R \rho_{aa} \tag{5.4.36}$$

Chapter 6

Practical considerations of laser induced fluorescence

In the last chapter we laid out how the fluorescence rates for the Ba-II transitions can be determined. Experimentally, what we actually measure is the total integrated number of photons over some period of time τ in a detector. If we let η represent the acceptance of the detector (which includes effects such as quantum efficiency, and solid-angle coverage) and the laser induced fluorescence (LIF) rate by Γ , then the total detected number of LIF photons is given by,

$$N_\gamma = \eta \int_0^\tau \Gamma dt \tag{6.0.1}$$

Although the decay rates are large for the Ba-II system, in practice η and ρ_{aa} can be prohibitively small. In addition, there will always be some non-zero rate of background counts Γ_{bkg} in our detector such that the total measured light, N_{tot} , will contain both our fluorescence signal N_γ , and a background signal N_{bkg} ,

$$N_{tot} = N_\gamma + N_{bkg} \quad (6.0.2)$$

$$= \eta \int_0^\tau (\Gamma + \Gamma_{bkg}) dt \quad (6.0.3)$$

The combination of these factors can make single-ion observation a difficult task. The purpose of this chapter is to investigate the experimental parameters that will effect the measured fluorescence signal. I divide the chapter into two broad sections motivated by whether or not the steady-state solutions to the OBEs can be used.

For what follows it will be useful to note that,

$$|\Omega_{B,R}| = \beta_{B,R} \frac{\sqrt{P_{B,R}}}{w_{B,R}} \quad (6.0.4)$$

$$\begin{aligned} \beta_B &\approx 184 \frac{\text{mm} \cdot \text{MHz}}{\sqrt{\text{mW}}} \\ \beta_R &\approx 169 \frac{\text{mm} \cdot \text{MHz}}{\sqrt{\text{mW}}} \end{aligned} \quad (6.0.5)$$

where $P_{B,R}$ and $w_{B,R}$ are the power and beam waist of either the blue or red laser respectively.

6.1 Continuous excitation

In the case that the excitation lasers are either continuously on (or pulsed such that the pulse widths are long compared to the P-state lifetime) then the population of the P-state can be described in the steady-state. Since, by definition, the steady state solution ρ_{aa} is independent of time, we can rewrite Equation 6.0.1 as,

$$N_\gamma = \eta \Gamma_B \rho_{aa} \tau \quad (6.1.1)$$

Thus, to maximize N_γ we simply need to maximize ρ_{aa} as given in Equation 5.4.32.

6.1.1 Natural linewidths

If we hold the detuning of the red laser fixed and scan the blue laser across the resonance frequency, lineshapes as in Figure 6.1.1 are produced. We can clearly see the dip in fluorescence at the two-photon resonance when the detunings are set equal. What is somewhat surprising from these plots is that the maximum ρ_{aa} occurs when both lasers are detuned from resonance.

The values of δ_B and δ_R that maximize ρ_{aa} for given Ω_B and Ω_R can in principle be solved analytically, however, this will involve finding the roots of a fourth degree polynomial and thus the functional form does not provide much insight. Instead I plot the value of δ_B as a function of the δ_R that maximizes ρ_{aa} as well as the maximum value of ρ_{aa} as a function of δ_R in Figure 6.1.2.

Although the functional forms are not simple, we can see that if the detunings are set in the region of the maximum, then there should not be very much dependence on their specific values. It also shows that the detunings should differ in sign to achieve the highest values for ρ_{aa} . These conclusions also hold for different values of the Rabi frequencies, Ω_B and Ω_R .

In Figure 6.1.3 the linewidths are shown for fixed $\delta_R = 0$ and $\Omega_R = 2\pi(40 \text{ MHz})$ and various values of Ω_B . For the first three plots ($\Omega_B = 50, 100, 200 \text{ MHz}$) we can see that the maximum P-state population increases with Ω_B . However, as Ω_B increases further, we see a substantial increase in the width of the distribution and eventually a decrease in the maximum P-state population. This effect is termed power broadening.

We can understand how the power broadening works if we consider the simpler case of

Figure 6.1.1: P-state populations as a function of the detuning of the blue laser for different fixed values of the red detuning. In all cases $\Omega_B = 2\pi(10 \text{ MHz})$ and $\Omega_R = 2\pi(40 \text{ MHz})$.

Figure 6.1.2: Maximum P-state population (solid line, left axis) and blue detuning at the maximum (dashed line, right axis) as a function of the detuning of the red laser. The vertical and horizontal lines indicate the maximum value of ρ_{aa} . In this case the maximum is $\rho_{aa} = 0.17$ and it occurs at $\delta_B = -78$ MHz and $\delta_R = 133$ MHz. The Rabi frequencies were set to $\Omega_B = 2\pi(10 \text{ MHz})$ and $\Omega_R = 2\pi(40 \text{ MHz})$.

a two-level system. The same formalism used in the treatment of the three-level system can be used here and it is not hard to find that the steady-state population for the excited state is given by,

$$\rho = \frac{\Omega^2}{2(\Gamma^2 + \Omega^2 + \delta^2)} \quad (6.1.2)$$

where Ω , Γ , and δ are the Rabi frequency, spontaneous emission rate, and detuning for this single transition. Defining the saturation parameter,

$$s_0 = \frac{\Omega^2}{\Gamma^2} \quad (6.1.3)$$

we can rewrite Equation 6.1.2 as,

$$\rho = \frac{s_0}{2 \left[1 + s_0 + \left(\frac{\delta}{\Gamma} \right)^2 \right]} \quad (6.1.4)$$

Figure 6.1.3: Power-broadened linewidths for different values of Ω_B . In all cases $\delta_R = 0$ and $\Omega_R = 2\pi(40 \text{ MHz})$.

This is just a Lorentzian function and has a FWHM given by,

$$\Gamma_{FWHM} = \Gamma\sqrt{1 + s_0} \quad (6.1.5)$$

For low laser intensities such that $s_0 \ll 1$ the excited state population increases linearly with s_0 and the width of the line is simply the spontaneous emission rate. As the laser intensity gets large such that $s_0 \gg 1$ the excited state population saturates and the width increases as the square-root of s_0 . Similar results can also be demonstrated in the case of a three-level system [90].

6.1.2 Doppler broadening

In practice one will not observe the linewidths discussed in the previous section as there will be other line-broadening mechanisms at work. Of specific interest to this work is the broadening caused by the presence of a buffer gas. There are two types of broadening that this can cause, collisional and Doppler. The former is not very important, since, as a general rule of thumb, the width is broadened by $\sim 1 \text{ MHz/mbar}$ [91] of gas pressure. In the range of pressures at which we are working ($10^{-4} - 10^{-3} \text{ mbar}$) the linewidths should be broadened by $\lesssim 1 \text{ kHz}$. This is much less than the natural linewidth and we will thus neglect any effects from collisional broadening.

Doppler (or thermal) broadening, on the other hand, can be substantial. This type of broadening is caused by the motion of the ions in the gas and is a type of *inhomogeneous* broadening since it depends on the frequency of the interacting laser. Motion of the ion along the laser direction will cause the frequency of the laser, observed in the ion's rest frame, to be shifted according to the Doppler effect. If we assume that the laser is aligned with the z-axis, then the frequency observed by the ion is,

$$\omega' = \omega \left[1 - \frac{\nu_z}{c} \right] \quad (6.1.6)$$

where ω is the laser frequency, ν_z is the z-velocity of the ion, and c is the speed-of-light. In order for the ion to absorb a photon, the observed frequency must equal the resonant frequency, ω_0 , of the transition in question. Thus, absorption only occurs when,

$$\omega_0 = \omega \left[1 - \frac{\nu_z}{c} \right] \quad (6.1.7)$$

From the perspective of an interaction taking place, we can equivalently look at this as though the ion is motionless but has a shifted resonant frequency given by,

$$\omega'_0 = \frac{\omega_0}{\left[1 - \frac{\nu_z}{c} \right]} \quad (6.1.8)$$

If we assume a Maxwell-Boltzmann velocity distribution then it is not hard to show that the Doppler broadened linewidth is given by,

$$\begin{aligned} g(\omega'_0 - \omega_0) &= \frac{1}{\omega_0} \sqrt{\frac{mc^2}{2\pi k_B T}} \exp \left[-\frac{mc^2}{2k_B T} \cdot \frac{(\omega'_0 - \omega_0)^2}{\omega_0^2} \right] \\ g(\delta) &= \sqrt{\frac{1}{2\pi\sigma^2}} \exp \left[-\frac{\delta^2}{2\sigma^2} \right] \end{aligned} \quad (6.1.9)$$

where k_B is Boltzmann's constant, m is the mass of the ion, T is the ion temperature, $\delta = \omega'_0 - \omega_0$ is the detuning from resonance, and σ is the width of the distribution given by,

$$\sigma = \frac{\omega_0}{c} \sqrt{\frac{k_B T}{m}} \quad (6.1.10)$$

The experimental linewidth for the Ba-II system in the steady state is then given by the convolution of Equation 6.1.9 with the steady-state solution of ρ_{aa} . As both the red and blue laser are needed to induce fluorescence the Doppler broadening of both lasers will have to be considered. Since,

$$\delta_B = \omega'_{ab} - \omega_{ab} = \omega_{ab} \left(\frac{c}{\nu_z} - 1 \right)^{-1} \quad (6.1.11)$$

$$\delta_R = \omega'_{ac} - \omega_{ac} = \omega_{ac} \left(\frac{c}{\nu_z} - 1 \right)^{-1} \quad (6.1.12)$$

we have that,

$$\delta_R = \frac{\omega_{ac}}{\omega_{ab}} \delta_B \quad (6.1.13)$$

Using the above relation we can write our final result as,

$$g_{Doppler}(\delta_B, \delta_R) = \int_{-\infty}^{\infty} \rho_{aa}(\delta_B - t_B, \delta_R - \frac{\omega_{ac}}{\omega_{ab}} t_B) g(t_B) dt_B \quad (6.1.14)$$

$$= \sqrt{\frac{1}{2\pi\sigma_B^2}} \int_{-\infty}^{\infty} \rho_{aa}(\delta_B - t_B, \delta_R - \frac{\omega_{ac}}{\omega_{ab}} t_B) \exp\left[-\frac{t_B^2}{2\sigma_B^2}\right] dt_B \quad (6.1.15)$$

The Doppler broadened linewidths for various detunings are shown in Figure 6.1.4 for values of $\Omega_B = 2\pi(10 \text{ MHz})$, $\Omega_R = 2\pi(40 \text{ MHz})$, and $T = 298 \text{ K}$. Comparing these to the linewidths in Figure 6.1.1 we can see that the Doppler broadening causes an increase in the widths of the lines, as well as a substantial decrease in the maximum P-state population.

To determine the optimal detunings in the presence of Doppler broadening we can follow the same method as done for the natural linewidths (Figure 6.1.2) and plot the maximum P-state population and value of δ_B at the maximum against the corresponding δ_R . This is done in Figure 6.1.5. We can see from this that the values of δ_B and δ_R at the maximum are shifted slightly from the non-broadened case and, as already mentioned, there is a significant decrease in the maximum P-state population. Despite this, we do observe the same general conclusions as were found for the previous case - if the detunings are set in the region of the maximum then there is a weak dependence on them, and, for the largest P-state populations, the detunings should be set opposite in sign.

As it is possible to cool the buffer gas it is possible to decrease the ion temperature to the point where Doppler broadening is negligible. Figure 6.1.6 shows the dependence of the linewidths on temperatures ranging from 1 - 298 K. What we can see is that at very low temperatures the linewidth does indeed approach the natural linewidth (compare to Figure 6.1.1). However, the peak of the distribution quickly decreases as the temperature increases until, at $T \sim 60 \text{ K}$, the Doppler width is roughly equal to the width of the P-state. As the temperature is increased further the maximum population continues to decrease, albeit at a slower rate, and the width of the distribution slowly increases.

Figure 6.1.4: Doppler broadened linewidths for $\Omega_B = 2\pi(10\text{ MHz})$, and $\Omega_R = 2\pi(40\text{ MHz})$. The temperature of the ion is 298 K.

Figure 6.1.5: Maximum P-state population (solid line, left axis) and blue detuning at the maximum (dashed line, right axis) as a function of the detuning of the red laser for the Doppler broadened lines. In this case the maximum is $\rho_{aa} = 0.051$ and it occurs at $\delta_B = -69$ MHz and $\delta_R = 178$ MHz. The Rabi frequencies were set to $\Omega_B = 2\pi(10$ MHz) and $\Omega_R = 2\pi(40$ MHz) and the temperature was 298 K.

What we can gather from this is that there is not much to be gained from decreasing the temperature of the buffer gas, since, even at the temperature of liquid nitrogen, the maximum P-state population is approaching that at room temperature.

6.1.3 Experimental considerations

The main experimental consideration is the scattered light from the excitation lasers, which, itself, can be divided into two different components: light scattered from components of the ion trap, and light that is scattered by the buffer gas. The latter is called Rayleigh scattering and is a well known effect that depends on the fourth power of the frequency of the scattered light. At the pressures used in this work the background due to Rayleigh scattering is negligible and it will thus be neglected.

Scattering from components of the ion trap depends on the particular aspects of the experiment. In our case, we are trying to launch two lasers into a linear Paul trap in

Figure 6.1.6: Doppler broadened linewidths as a function of temperature. In all cases $\Omega_B = 2\pi(10\text{ MHz})$ and $\Omega_R = 2\pi(40\text{ MHz})$.

such a way that the lasers are aligned with the trap axis. To simplify the analysis of this setup I will make the following assumptions:

1. The rate of scattered light increases linearly with the laser power.
2. The laser beam is circular with a Gaussian profile.
3. The scattered light is entirely due to the beam coming into contact with the electrodes of the linear Paul trap.
4. The laser is perfectly aligned with the trap axis.
5. Only blue light is observed - red is perfectly filtered away

The first is not really an assumption as the rate of scattering should be proportional to the power of the scattered laser. The second and third assumptions imply that no matter how small we make the beam, there will always be some component that comes into contact with the electrodes of the Paul trap and is thus scattered into our detector. The next assumption simplifies things greatly, since, in general, the laser can drift in both position (relative to trap center) and direction (relative to trap axis). Finally, the last assumption implies that we only need to consider scattering from the blue laser and is motivated by the fact that the blue fluorescence rate is roughly three times higher than the red fluorescence rate.

Mathematically these assumptions imply the following,

$$\Gamma_{bkg} = P_B \Theta \exp \left(-\sqrt{2 \ln 2} \frac{r_0^2}{w_B^2} \right) \quad (6.1.16)$$

where Γ_{bkg} is the background rate, P_B and w_B are respectively the power and beam waist of the blue laser, r_0 is the radius of the ion trap, and Θ is a proportionality constant depending on the specific geometry of the ion trap.

The dependence of Γ_{bkg} on the beam waist complicates things greatly as the Rabi frequency Ω_B truly depends on the energy density of the laser field and thus also on w_B ,

$$\Omega_B \propto E_B \propto \frac{\sqrt{P_B}}{w_B} \quad (6.1.17)$$

Furthermore, there will also be dependence on the beam waist due to the fact that the ions are not localized in space but will have some spatial distribution in the direction perpendicular to the laser beam. This implies that making the beam smaller in order to decrease Γ_{bkg} will eventually have a negative impact on the fluorescence rate due to a decreased proportion of time that the ion is interacting with the laser field. For these reasons I will make the final assumption that the laser beam waist is fixed and will use a value of $w_B = w_R = 1$ mm throughout the rest of the chapter (this is the beam diameter that was used in this work). The background rate is then given by,

$$\Gamma_{bkg} = P_B \frac{\theta}{E_\gamma} \quad (6.1.18)$$

where E_γ is the energy of the blue photons and θ is a dimensionless constant representing the fraction of the blue laser power that is scattered. In terms of Ω_B we have,

$$\Gamma_{bkg} = \frac{\theta}{E_\gamma \beta_B^2} \Omega_B^2 \quad (6.1.19)$$

where β_B is defined in Equation 6.0.5 and is written here assuming that the laser diameter is 1 mm.

To determine the effect this will have on the sensitivity of a single-ion observation we can plot the maximum theoretical ρ_{aa} in the presence of Doppler broadening as a function of Ω_B for different values of Ω_R in Figure 6.1.7. This plot shows that ρ_{aa} certainly does not increase with Ω_B^2 and more closely follows a square-root dependence. Since Γ_{bkg} does increase with Ω_B^2 we can expect that the single-ion sensitivity will depend strongly on the value of θ .

Defining the signal to noise ratio (SNR) as,

Table 6.1: Experimental parameters needed to achieve the maximum SNR using continuous excitation of the ion. In this table $d\lambda_B$ and $d\lambda_R$ are the equivalent wavelength detunings calculated from δ_B and δ_R respectively.

$\frac{\delta_B}{2\pi}$ (MHz)	$\frac{\delta_R}{2\pi}$ (MHz)	$d\lambda_B$ (fm)	$d\lambda_R$ (fm)	Ω_B (MHz)	Ω_R (MHz)	ρ_{aa}
∓ 5.3	± 6.4	± 4.3	∓ 8.9	70	50	0.047
∓ 7.2	± 11	± 5.8	∓ 16	94	100	0.081
∓ 10	± 16	± 8.3	∓ 22	107	200	0.14
∓ 13	± 28	± 11	∓ 39	113	400	0.19

$$\begin{aligned}
\text{SNR} &= \frac{N_{tot} - N_{bkg}}{\sqrt{N_{tot} + N_{bkg}}} \\
&\approx \frac{N_\gamma}{\sqrt{N_\gamma + 2N_{bkg}}}
\end{aligned} \tag{6.1.20}$$

and, using Equation 6.1.1, the SNR for a one second scan becomes,

$$\begin{aligned}
\text{SNR} &= \frac{\eta \Gamma_B \rho_{aa}}{\sqrt{\eta \Gamma_B \rho_{aa} + 2\eta \Gamma_{bkg}}} \\
&= \frac{\Gamma_B \rho_{aa} \sqrt{\eta}}{\sqrt{\Gamma_B \rho_{aa} + 2 \frac{\theta \Omega_B^2}{E_\gamma \beta_B^2}}}
\end{aligned} \tag{6.1.21}$$

In Figure 6.1.8 $\sqrt{\frac{1}{\eta}} \text{SNR}$ is plotted against Ω_B for various values of Ω_R and θ . As we can see, the SNR peaks in the range $\Omega_B \approx 50$ to 100 MHz, with the peak slowly drifting to higher values as Ω_R is increased. Caution should be taken when using these plots since this represents the best possible scenario with all experimental parameters perfectly tuned. Fluctuations in laser intensity, imperfect laser alignment, and electronic noise can drastically reduce the SNR and these curves should thus be used only as a guide.

To summarize this section, the values of δ_B , δ_R , Ω_B , and Ω_R needed to approximately achieve the maxima on the SNR plots in Figure 6.1.8 are given in Table 6.1.

Figure 6.1.7: Maximum P-state population in the presence of Doppler broadening as a function of Ω_B for various values of Ω_R . The optimum detunings were assumed for each point, and the temperature of the ^{138}Ba ion was set to 298 K.

Figure 6.1.8: Best case scenario for the SNR using continuous excitation of the ions for different values of the parameter θ and Ω_R .

6.1.4 Experimental techniques

To experimentally observe fluorescence using continuous excitation two different techniques were used: direct observation, and synchronous photon counting. I will briefly describe these below.

6.1.4.1 Direct observation

Direct observation is the simplest method and uses a CCD camera or some other imaging device to directly observe the fluorescing ions. Taking the example of a CCD, direct observation has the benefit of high quantum efficiency and excellent spatial resolution, both of which directly impact the expected SNR of a measurement. The quantum efficiency enters the SNR calculation via the detector efficiency η and thus there is a square-root dependence on this parameter. More importantly than this is the ability to reject backgrounds based on spatial information. The fluorescing ions will be localized at the center of the trapping region whereas the scattered background light should mostly be coming from the edges of the trap electrodes. By cutting out those regions of an image that contain a high density of scattered photons the background parameter θ can be reduced drastically.

The technique is not without its drawbacks, however. The time needed to expose an image and then read it out can be prohibitively long (on the order of seconds) and thus limits the total frame-rate that one can achieve. For the proposed nEXO experiment we calculated that we should expect about one barium ion produced every five seconds in the chamber and thus fast observation will be important. Only the highest-end CCD cameras have frame-rates that would be useful for a barium-tagging scheme.

In addition, cameras are usually bulky and will generally need collection optics in order to get close to the trapping region. The camera will need to be focused as well in order to make use of the spatial rejection. Background contributions from CCDs include dark counts from thermally excited electrons, and readout noise due to the electronic

circuits. For most modern CCDs, these are usually quite small. Finally, CCDs are susceptible to false positives due to interaction with cosmic rays and may need more than one exposure in order to identify a barium ion with high-confidence.

6.1.4.2 Synchronous photon counting

This technique is based on counting the fluorescence photons produced using a PMT or similar device. In this technique the red laser is modulated on-off at a relatively low rate, in the 100-10000 Hz range and thus allows us to use the steady-state solution to the OBEs. As fluorescence will only occur if both lasers are on, an on-off modulation in the fluorescence signal will be produced. The photons are counted as a function of time using a multi-channel analyzer (MCA) or similar device and the fluorescence signal is obtained by subtracting the counts in the off period from the counts in the on period.

The main benefit of this technique is that there is always a live measurement of the scattered background light from the unmodulated blue laser and thus, if we are in the saturation regime, the measurement will be robust against slow fluctuations or drifts in the laser intensities. Observations can also be made at a high rate limited only by the modulation frequency chosen. Finally, since PMTs can be made rather compact, collection optics may not be necessary as the detector can be placed as close as one can get to the trapping region (without being under vacuum).

6.2 Intermodulation

In the case that the steady-state solution for ρ_{aa} cannot be used, the OBEs must be integrated numerically. In this section I describe the time-dependent dynamics of the Ba-II system for the specific technique of *intermodulation*.

The intermodulation technique involves modulating the excitation lasers in such a way that both lasers are never on at the same time. If we let $\vec{E}'_B$ and $\vec{E}'_R$ be the chopped

blue and red laser fields, then we can mathematically represent this as,

$$\vec{E}'_B = \vec{E}_B \Pi(2\pi f_{mod}t) \quad (6.2.1)$$

$$\vec{E}'_R = \vec{E}_R \Pi(2\pi f_{mod}t + \pi) \quad (6.2.2)$$

where Π is the square-wave function, and f_{mod} is the modulation frequency.

To better understand the technique consider only one period $\tau_{mod} = 1/f_{mod}$ of the modulation. In the first half of the period the blue laser is on and the red is off. The ion transitions between the S and P states until it falls into the metastable D-state and is trapped.

In the second half of the modulation period the blue laser is off and the red laser is turned on. The ion transitions between the D and P states, until, inevitably, it falls back into the S state, emits a blue photon, and is again trapped. The power of the technique comes from the fact that any blue light observed in the second half of the modulation cycle must have come from the ion since the blue laser is not on and scattered light will not be present. Thus, in principle, this technique allows a background free measurement of the ion. An illustration of the technique is given in Figure 6.2.1.

The above description was somewhat of an ideal case since there could be some probability that the ion does not transition to the D-state when the blue laser is on, and also some probability that the ion will not transition back to the S state when the red laser is on. If we let p_D be the probability of finding the ion in the D-state after the first half of the period and p_S be the probability for the ion to be in the S-state after the second half, then the number of photons emitted per modulation period is $p_{mod} = p_D \cdot p_S$. We can write the total fluorescence rate as,

$$\begin{aligned}
 \Gamma_{mod} &= p_D p_S f_{mod} \\
 &= p_{mod} f_{mod}
 \end{aligned}
 \tag{6.2.3}$$

The probabilities p_D and p_S are related to the populations ρ_{cc} and ρ_{bb} via,

$$p_D = \rho_{cc}(t_0 + \frac{1}{2}\tau_{mod}) \tag{6.2.4}$$

$$p_S = \rho_{bb}(t_0 + \tau_{mod}) \tag{6.2.5}$$

where t_0 defines the start of the modulation cycle. The probabilities p_D and p_S will depend on the power of lasers as well as the length of time the lasers are on.

We can analyze how long we need to leave the lasers on as a function of the laser intensity to get a certain probability to be in either the D or S states. This is done in Figure 6.2.2 where the time needed to ensure 99% probability (τ_{99}) of occupation for the S or D states is plotted against the respective Rabi frequency. In these plots the y -axis has a logarithmic scale to make it clear that these are not simply exponential decays.

As shown, there is a point beyond which increasing the laser power does not decrease the time needed to populate the respective state. This is a similar effect to the saturation phenomenon encountered for the power-broadened linewidths in the steady-state case. For the D-state, this “saturation time” is $\tau_D^{sat} \approx 0.3 \mu s$ and for the S-state it is $\tau_S^{sat} \approx 0.1 \mu s$. Although these do not correspond to the spontaneous decay rates, it is interesting to note that $\tau_D^{sat}/\tau_S^{sat} \approx \Gamma_B/\Gamma_R$.

The sum of τ_D^{sat} and τ_S^{sat} sets a limit on how fast the modulation can be,

$$f_{mod}^{max} = \frac{1}{\tau_D^{sat} + \tau_S^{sat}} = 2.5 \text{ MHz} \tag{6.2.6}$$

Figure 6.2.1: Illustration of the intermodulation technique showing one full period of the modulation. In the first half, the blue laser is on and the probability to be in the D-state increases. In the second half, the red laser is on and the D-state quickly depopulates to the S-state. The oscillations at the start of each half-period are due to oscillations between the S and P states (first half) and D and P states (second half). The observation of the emitted photon occurs in the second half when the blue laser is off. For this particular case $f_{mod} = 1$ MHz, $\Omega_B = \Omega_R = 2\pi(100 \text{ MHz})$, and $\delta_B = \delta_R = 0$.

Figure 6.2.2: Time needed to reach a 99% probability of occupation of the D-state (left) and S-state (right) as a function of Ω_B and Ω_R respectively. The detunings are $\delta_B = \delta_R = 0$.

Figure 6.2.3: Illustration of the intermodulation technique with unequal duty-cycles for the two lasers. In the first $3/4$ of the period the blue laser is on and the probability to be in the D-state increases to 99% at $0.3 \mu\text{s}$. In the last $1/4$ of the pulse the red laser is on and the D-state quickly depopulates to the S-state which reaches 99% occupancy at the end of the period. For this particular case $f_{\text{mod}} = 2.5 \text{ MHz}$, $\Omega_B = \Omega_R = 2\pi(50 \text{ MHz})$, and $\delta_B = \delta_R = 0$. The total photon emission rate would be $\sim 2.45 \text{ MHz}$.

which can be achieved (in theory) by changing the duty cycle¹ of the modulating square-wave functions to $\frac{\tau_D^{\text{sat}}}{\tau_D^{\text{sat}} + \tau_S^{\text{sat}}}$ for the blue laser and $\frac{\tau_S^{\text{sat}}}{\tau_D^{\text{sat}} + \tau_S^{\text{sat}}}$ for the red laser. This situation is illustrated in Figure 6.2.3 for $\Omega_B = \Omega_R = 2\pi(50 \text{ MHz})$ where the duty cycle has been set to 75% for the blue laser and 25% for the red laser.

6.2.1 Linewidths

I have so far made no mention of the effect detuning the lasers has on the fluorescence rate using the intermodulation technique and have kept $\delta_B = \delta_R = 0$ for all of the previous examples². To investigate the dependence of the fluorescence rate on the detunings we will follow the same method as used for the continuous excitation case. However, the

¹The duty cycle of the square wave function is defined as the fraction of a full period for which the square wave equals unity.

²We are able to set the detunings equal since both lasers are never on simultaneously and thus the two-photon resonance effect does not occur.

linewidths produced in this way do not have the same interpretation as in the continuous excitation case and thus need some explanation.

First, we are not interested in the P-state population but instead the probability to get one photon per modulation period. This probability, p_{mod} , can be factored into the individual probabilities p_D , that depends only on the properties of the blue laser, and p_S , that depends only on the red laser. This implies that, for a given set of f_{mod} , Ω_B , Ω_R and corresponding duty cycles, p_{mod} can be completely determined at any δ_B and δ_R using the linewidths determined for p_D and p_S separately. These linewidths are shown in Figure 6.2.4 for $f_{mod} = 1$ MHz and various values of Ω_B and Ω_R . The duty cycles are both assumed to be 50%.

For low intensities the linewidths have a Lorentzian profile with a width equal to the total lifetime of the P-state. This is to be expected from the discussion of the dipole interaction given in the previous chapter. However, as Ω_B and Ω_R increase further, the length of time the lasers are on (0.5 μ s in this case) surpasses the saturation times for the D and S states and the probability saturates. As the intensity increases further, the probability saturates for a larger range of detunings and will continue to do so without limit. Fortunately, this method does not need any optimization of the detunings since the maximum probabilities always occur when the lasers are on resonance.

6.2.2 Doppler broadening

To analyze the effect of Doppler broadening we will again assume a Maxwell-Boltzmann velocity distribution, however, this time we will have to apply the broadening separately to each of the red and blue linewidths. Following the same procedure used in the last section we have,

$$g_D(\delta_B) = \sqrt{\frac{1}{2\pi\sigma_B^2}} \int_{-\infty}^{\infty} p_D(\delta_B - t_B) \exp\left(-\frac{t_B^2}{2\sigma_B^2}\right) dt_B \quad (6.2.7)$$

Figure 6.2.4: Linewidths using the intermodulation technique. The plots on the left are obtained by scanning the blue laser across the $S \rightarrow P$ transition, and the plots on the right by scanning the red laser across the $D \rightarrow P$ transition.

Figure 6.2.5: Doppler broadened linewidths for the intermodulation technique.

$$g_S(\delta_R) = \sqrt{\frac{1}{2\pi\sigma_R^2}} \int_{-\infty}^{\infty} p_S(\delta_R - t_R) \exp\left(-\frac{t_R^2}{2\sigma_R^2}\right) dt_R \quad (6.2.8)$$

For low intensities, the natural linewidths are just Lorentzians and thus the convolutions are described by what is called a Voigt function. For the higher intensities the Doppler broadened spectra are Voigt-like, however, the tops of the distributions are flattened due to the saturation effects described previously. Overall, the same conclusions can be drawn as for the continuous excitation case - Doppler broadening decreases the peak probabilities and increases the widths of the lines.

6.2.3 Experimental considerations

In principle, the scattered blue light should not be an issue since the blue laser is off during the measurement. In practice, this is not the case due to the method used for creating the laser “pulses”. In this work an acousto-optical modulator (AOM) is used to modulate the laser beams on and off and this has some important consequences for the single-ion sensitivity.

Firstly, the AOM has a “rise time” which describes the time needed to modulate the laser from the off state to the on state. This will have two effects. The first is a decrease in the integrated power seen by the ion for a fixed pulse width and thus a respective decrease in the probabilities p_D and p_S . In practice, AOM rise times are generally small enough that this will not be a problem. The second effect has to do with the background. A finite rise-time implies that there will be leakage of blue light into our “signal” region. For high modulation frequencies the leakage can be significant enough to decrease the SNR of the measurement below useful levels. In order to get around this, one can introduce a time separation between the end of the blue pulse and the start of the red pulse. Unfortunately, if one wants to keep the same probability of observing a photon per cycle then the modulation frequency, and thus fluorescence rate, will have to be decreased.

The second issue with AOMs is that, even in the off-state, there is always some light that leaks through and this will be proportional to the intensity of the lasers. The level of leakage is described by the extinction ratio (ϵ_{AOM}) of the AOM which is defined as the ratio of laser power throughput when the AOM is in the on-state to that in the off-state. Fortunately, the extinction ratios for most modern AOMs are quite high (> 50 dB) and, considering that this is applied to the scattered light, the effect can usually be neglected.

The last thing to consider for this technique has to do with the physical limits of the photon detector used. Since, in this method, we generally run the laser at high intensities to ensure population of the D-state, the amount of light falling on the detector during the blue pulse can be substantial. This can be a problem for photon detectors since they can

be damaged if the intensity of the light is too high (photomultiplier tubes, for example). In practice, this will limit the intensity of the blue laser pulse and it may be necessary to use lower modulation frequencies in order to ensure D-state population.

Chapter 7

Experimental systems

7.1 Linear Paul trap

The linear Paul trap used in this work was previously designed and built by collaborators at Stanford University. Unfortunately, the engineering drawings and design files for the trap were not available and the only information on the trap was contained in previous PhD theses of EXO collaborators [71, 68]. As a result many of the design characteristics of the trap had to be reverse engineered and this took up a large portion of the time available in this work. The description of the trap given here is a combination of what was measured and what has been described previously.

7.1.1 Geometry

A schematic of the ion trap is given in Figure 7.1.1. The ion trap has a total length of ~ 710 mm and is composed of sixteen segments. Each individual segment is composed of four parallel, stainless-steel (SS) electrodes of radius 6 mm centered around a circle of 2.66 mm. The first twelve and last two segments are for transport and cooling of the ions entering the trap and are each 49.5 mm in length. The 13th and 14th segments are the trapping segments and have a reduced length of 4.5 mm to provide better ion

Figure 7.1.1: Schematic showing the dimensions of the ion trap (not to scale).

localization.

The electrodes are positioned relative to each other on macor mounts using SS dowel pins to ensure precise placement. The electrodes are then fixed into place with vented, silver-plated SS screws¹ which also provide an electrical connection to the RF and DC potentials. Each macor mount supports two electrodes and is itself attached to a precision ground SS support structure. The relative positions of the mounts are also defined using dowel pins and fixed into position using silver plated screws.

Each support structure defines half of the ion trap with each half joined together using SS plates that fix the relative positions of the supports, again using SS dowel pins. Once assembled, the entire trap structure is a stand alone component that is inserted into an ultra-high-vacuum (UHV) chamber.

¹The screws are vented by drilling a hole through the middle to prevent virtual leaks when under vacuum. The screws are silver plated to prevent cold welding of the SS.

7.1.2 Electrical

From our discussion of the theory of linear quadrupoles we know that the stability parameters a_u and q_u must lie within the stability region as shown in Figure 4.3.1. For this work we did not apply a DC bias field so that $a_u = 0$. Thus, for stable confinement we need $|q_u| < 0.908$. Having fixed the trap radius at $r_0 = 2.66$ mm and since we are interested in trapping a single Ba ion (assuming $^{138}\text{Ba}^+$) the last two free parameters determining q_u are the frequency and amplitude of the RF driving field. We therefore have the condition that,

$$\frac{V}{f^2} < 180 \frac{\text{V}}{\text{MHz}^2} \quad (7.1.1)$$

where $2\pi f = \Omega$. For a typical value of $f = 1.4$ MHz we would need $V < 355$ V.

To achieve the required voltage and frequency a tuned RF amplifier was designed and built at Carleton to amplify a 1.2 - 1.5 MHz RF signal to voltages in the range 50 - 125 V. The RF signal was supplied by a digital signal generator². The schematic of the amplifier is included in Figure 7.1.2 for reference. The configuration most commonly used has $f = 1.45$ MHz and $V = 100$ V such that the stability parameter q and radial potential depth D_r (not considering the effect of DC) are calculated to be,

$$|q_u| = 0.26 \quad (7.1.2)$$

$$D_r = 3.25 \text{ eV} \quad (7.1.3)$$

The longitudinal confining DC potentials were supplied to the trap using sixteen analog outputs controlled via a computer user interface. The outputs are supplied by the driving electronics of an EXTREL MAX500HT Quadrupole Mass Filter that was used by my predecessor as a means to select singly and double charged barium ions. The

²Model 4005DDS by B&K Precision

Figure 7.1.2: Schematic of the tuned RF amplifier.

analog outputs are capable of producing DC potentials of ± 10 V.

The most common configuration used for the DC potentials on the trap electrodes is shown below (starting from the entrance),

$$U = \{0, -0.25, -0.50, \dots, -2.75, -6, -6, -2.75, 10\} \text{V} \quad (7.1.4)$$

where the trapping segments (segments 13 and 14) are at -6 V and the adjacent segments are at -2.75 V. The longitudinal potential depth is thus 3.25 eV. This modifies the radial depth according to Equation 4.4.4 to $D_r \approx 2.9$ eV.

The DC is mixed with the RF through a series of parallel RC circuits to isolate the RF from the DC drive and provide unattenuated transmission to the trap electrodes. The time constant of the circuit is ~ 10 ms ($R = 100$ k Ω , $C = 100$ nF). A schematic showing the electrical connections for two of sixteen identical circuits is given in Figure 7.1.3. The mixing circuits are housed in four separate boxes that are directly attached to vacuum feedthroughs on the ion trap chamber to minimize capacitive load on the system. The mixed DC and RF signals are delivered to the trap inside the vacuum chamber via Kapton-insulated wires. The total capacitance of the system is estimated to be ~ 250 pF.

Figure 7.1.3: RF and DC mixing circuit. Two of sixteen identical segments are shown.

7.2 Buffer gas and vacuum systems

Barium is itself a very reactive metal and readily reacts with oxygen and other electronegative impurities. Thus, in order to efficiently trap the barium ions, the entire vacuum system must be constructed out of UHV material with the ability to be baked-out to temperatures in excess of 100°C.

The vacuum chamber used to house the ion trap is itself a 30-litre volume, SS, custom built chamber by Trinos Vacuum. The chamber is fitted with five 8" conflat ports for easy experimenter access, and numerous 2.75" and 4.5" ports for laser access and electrical feedthroughs. The observation port is itself a 6" conflat located on the top of the chamber and is fitted with a re-entrant window that allows one to place the fluorescence detector a mere 24.5 mm from the center of the trap. The chamber is pumped to vacuum using an 400 L/s turbomolecular pump³ and is backed by an 10 L/s dry scroll pump⁴. A small diaphragm pump is added to pump down the gas line if one wanted to change the buffer gas.

³Edwards nEXT400D

⁴Edwards nXDS10i

The buffer gas is fed into the chamber using the system shown in Figure 7.2.1. The buffer gas is first fed through a passive getter⁵ that is capable of purifying He to < 100 ppt for the contaminants of concern (O_2 , H_2O). Pressure is monitored inside the chamber using two dual Pirani/cold cathode pressure gauges⁶. A flow-regulating valve⁷ regulates the pressure inside the chamber using the pressure signal on one of the gauges and is able to keep the pressure constant to within 0.5%. Unfortunately, the pressure gauges are a source of background light believed to be due to the hot filaments of the Pirani circuits. More importantly, the gauges are a source of barium ions, a point which will be discussed shortly. As a result, when searching for single-ions, the pressure inside the system is fixed using the manual control on the regulating valve and the gauges are turned off. The pressure set in this way is observed to be stable within 1% over measurement times of a few hours.

For initial pump downs after the chamber was exposed to the atmosphere the entire system is baked at $110^\circ C$ for at least two days to remove any water present. After the bake-out, the system is flushed a number of times with helium which is observed to increase the vacuum substantially. A residual-gas-analyzer⁸ (RGA) is used to monitor the contaminants inside the system to ensure they are at acceptable levels after every bake-out. An example RGA spectrum taken after a two-day bake-out is shown in Figure 7.2.2 where it is seen that the partial pressure of water is $\sim 10^{-9}$ mbar.

7.3 Ion loading

To load the trap with barium ions a small chunk of pure barium metal is placed inside the chamber near the entrance to the ion trap. A pulsed UV laser⁹ directed at the barium chunk ablates a cluster of ions free from the metal. Some of the liberated ions drift into

⁵SAES MicroTorr MC1-902f

⁶Pfeiffer Vacuum PKR261

⁷Pfeiffer Vacuum UDV146 with RVG 050 C Control Unit

⁸SRS RGA300

⁹Newport Explorer DPSS 349

Figure 7.2.1: Buffer gas supply system.

Figure 7.2.2: Example RGA spectrum showing the main contaminants present after a ~two day bake-out.

the entrance of the trap and subsequently fall down the potential gradient to the trap center. The maximum energy of each pulse is 120 μJ , however, the laser is usually run at the lowest energy possible while still observing ions. The number of ions produced in this manner is highly variable but always produces orders of magnitude more than what is wanted for observing single-ions. This can be useful when tuning different parameters (laser alignment, for example), however, it has the drawback that one has to wait for the trap to empty in order to get the required number for making measurements. Since maximizing ion life-time in the trap is one of the priorities, this can take a long time. Another method of ion loading (that was not intended for this purpose) uses the pressure gauge as an ionization source.

A major source of frustration during this work was due to a phenomenon which we termed “self-loading” of the ion trap. If the trap was left to its own means, without the UV laser on, barium ions (as observed from their fluorescence) slowly started filling the trap. For obvious reasons, this self-loading was a massive barrier standing in the way of single-ion observation. After a long, drawn out process¹⁰ it was finally discovered that the pressure gauges were the source of the self-loading.

Since the chunk of barium metal has a vapor pressure there will be free atoms of barium inside the vacuum chamber. These atoms are not a problem by themselves since atomic barium will not be trapped or interact with the fluorescence lasers. However, the pressure gauges work by ionizing gas molecules. In addition to the barium atoms being directly ionized by the gauges, any helium (the main constituent of the buffer gas) that was ionized would immediately ionize any barium that was around due to the large difference in their ionization potentials. The overall effect was the production of a large number of barium ions from which only a few had to make it into our trapping region to cause problems. The upside of this was that when the gauge was turned off the ions

¹⁰This really was a pain since the pressure gauges were one of the first things checked. However, due to a series of unfortunate circumstances we ruled out the pressure gauges as the source of self-loading. It was only by accident that the gauges were discovered to indeed be the culprits.

were no longer produced. As the number of ions produced in this manner was far less than that produced by the UV laser, the ion gauge turned out to be a very useful tool to load the trap. Consequently, the ion gauge became the primary means to load the trap and was in fact the loading method used when single ions were first observed.

7.4 Fluorescence collection

Fluorescence light from the trapped ions was detected using a CCD camera¹¹ or a PMT¹² depending on the specific method used. The CCD was exclusively used for the direct observation type of experiment, whereas the PMT was used for the synchronous-photon counting and the intermodulation techniques.

The CCD was a rectangular pixel array of dimensions of 3326×2504 pixels or 17.96×3.52 mm. The camera was fitted with a filter wheel, intended for use as astronomy filters, from which the blue filter was used to reject the scattered red light. An imaging lens¹³ with a focal length of 50mm was used to focus the fluorescent light onto the CCD. As the camera was quite bulky, a combination of lens tubes were also employed in order to get the lens close to the re-entrant window so that the fluorescence could be imaged. The camera was attached to a micrometer driven, three-axis mount in order to accurately position it over the center of the trap, and to aid in focusing. The specified quantum efficiency at 493 nm is ~ 0.49 and the overall collection efficiency of the CCD plus collection optics was estimated to be 0.5% if the fluorescence light was emitted as a point source. The main specifications of the CCD (including the estimated dark counts and read noise) are summarized in Table 7.1.

The PMT was a current-output type and contained a circular gallium-arsenide-phosphide (GaAsP) cathode with a diameter of 5 mm. The PMT module was small enough to place up against the re-entrant window, however, it was calculated that the collection

¹¹SBIG STF8300M

¹²Hamamatsu H7422-40P

¹³ASAHI Super Multi-Coated Pentax-M, 50mm, F1.4

Table 7.1: Specifications of the SBIG STF8300-M CCD camera and optics setup.

CCD	Kodak KAF-8300M	
Dimensions	3326×2504 px	17.96×13.52 mm
Quantum efficiency	~0.5 @ 493 nm	
Dark counts	0.02 e-/px/s @ -10 C	
Read noise	~9 e- rms	
Digitization rate	10 Mpx/s	
Quantum efficiency	~0.5 @ 493 nm	
Optics collection efficiency	~1 % (point source)	
Total collection efficiency	~ 0.5%	

Table 7.2: Specifications of the Hamamatsu H7422-40P PMT and optics setup.

Photocathode material	GaAsP
Photocathode dimensions	5 mm diam.
Quantum efficiency	~ 0.37 @ 493 nm
Dark counts	300 /s
Max. current-output	2 μ A
Optics collection efficiency	~2 % (point source)
Total collection efficiency	~0.7 %

efficiency could be increased somewhat if a collection lens was also used. The lens used was a 16.0mm effective-focal-length (EFL) condenser lens¹⁴ that was placed ~ 30 mm in front of the photocathode. A 24 nm narrow-band dielectric filter centered at 500 nm¹⁵ was placed in front of the lens to reject the scattered red light. The entire PMT module and optics could be attached to the same three-axis mount used for the camera and thus precise positioning of the PMT was available (this was not as necessary as for the camera since we did not need to form a focused image). The quantum efficiency of the PMT at 493 nm is ~ 0.37 , and the total light collection efficiency of the PMT plus optics was estimated to be $\sim 0.7\%$. The main specifications of the PMT are listed in Table 7.2.

To count the current pulses produced by the PMT an amplifier/discriminator¹⁶ able to count up to a rate up of ~ 100 MHz. The output TTL pulses were then counted by a

¹⁴Thorlabs ACL25416U-A

¹⁵It is advantageous to choose the center wavelength larger than wanted as the bandpass tends to smaller wavelengths as the angle-of-incidence is increased.

¹⁶F-100T Amplifier-Discriminator from Advanced Research Instruments Corporation

multi-channel-analyzer¹⁷ (MCA) that was connected to a PC. The MCA is able to count pulses up to a rate of 150 MHz with a minimum dwell-time of 100 ns.

7.5 Fluorescence lasers

The blue fluorescence laser used was really a state-of-the-art device and a pleasure to work with. A schematic of the laser setup is shown in Figure 7.5.1. As there is no known diode yet that produces light directly at 493 nm, the blue light is produced by frequency doubling an infra-red (IR) beam at 986.7 nm. The IR diode is a Toptica DL pro that produces 986.7 nm light at ~ 1 W. The beam is directed into a grating in the “Littrow” set-up such that the first-order diffracted beam is directed back into the laser diode in order to control the laser frequency. The stabilized beam is then directed through an optical isolator to prevent any other feedback into the diode and then into a tapered amplifier to increase the power of laser. The output beam from the amplifier will be highly elliptical and thus the beam is propagated through a series of optics to match the mode of the beam to the fundamental mode of the doubling cavity.

The doubling cavity itself is designed in a bow-tie-ring geometry and consists of two plane and two concave mirrors. The beam enters the cavity through a partially transmitting mirror M1 (see Figure 7.5.1) and produces a beam waist between mirrors M1 and M2. Mirror M2 is mounted on a piezo crystal which is driven by a regulator signal generated by a Pound-Drever Hall control unit. The second-harmonic-generation (SHG) crystal is placed at the beam waist between mirrors M3 and M4 and the beam is extracted through the partially transmitting mirror M4. A photo-diode located behind mirror M1 monitors the intra-cavity power. The output beam is finally passed through some shaping optics to produce a near diffraction-limited Gaussian beam with a power of ~ 300 mW. The frequency stability without external locking is already within ± 10 MHz.

The 649 nm light needed for the D $\rightarrow$ P transition is directly available from a diode

¹⁷ORTEC MCA-pci

and is produced using a Toptica DL pro. The design of the diode is identical to the DL pro used to generate the 986.7 nm light. The red beam, however, lacks shaping optics at the output and is slightly elliptical with the ratio of the major to minor diameters of about 1.5. The output power is ~ 400 mW.

In order to lock the beams to the Ba-II transition frequencies, the 986.7 and 649 nm beams are sampled and fed into a Fizeau interferometer¹⁸. A dedicated, frequency stabilized helium-neon (He-Ne) laser¹⁹ provides a constant frequency reference to the interferometer. The interferometer then provides an analog output signal which is fed back into the laser control electronics in order to lock the beam to the desired wavelength. Due to the tunability of the diodes, frequencies within ± 10 GHz of the center frequency for the blue laser (much less for the red laser) may be set directly from a computer, with the optional ability to set the regulating signal to a function of time (such as a sine, or square wave). The total frequency stability of the lasers locked to the interferometer has been demonstrated to be ± 1 MHz for at least three hours. A plot of the frequencies of both lasers while they are locked to the interferometer are shown in Figure 7.5.2. The two peaks near the middle of the plot are due to the door of the laboratory opening and closing, which, due to the slight pressure shift in the room, causes the refractive index and thus the laser frequencies to shift. Both lasers quickly relock, and display < 1 MHz stability again.

Before being launched into the ion trap, the red and blue laser beams are overlapped using a dichroic mirror. They are then directed at a system of mirrors which provide more-or-less independent control over the direction and position of the beam with respect to the trap axis. An overall schematic of the laser systems is shown in Figure 7.5.3.

As the diameters of the lasers are an important parameter for ion fluorescence the beams were characterized using a beam profiler²⁰. The spatial profile of both lasers

¹⁸High Finesse WS-U 10

¹⁹SIOS SL 03

²⁰Newport LBP-VIS

Figure 7.5.1: Schematic of the Toptica TA-SHG pro 493 nm laser. The mirrors of the resonant cavity are represented by M1 - M4.

Figure 7.5.2: Frequencies of the two lasers as a function of time while locked to the interferometer. The top represents the blue laser, and the bottom the red. The peaks near the middle of the plot represent a slight shift in the pressure due to the laboratory door opening and closing. The rellocking capability of the lasers is evident.

Figure 7.5.3: Schematic of the laser systems used. The blue and red lasers are sampled by the interferometer to monitor the laser frequencies. A dedicated frequency-stabilized He-Ne laser is used as a calibration reference for the interferometer. A computer interface allows one to set a desired frequency for the lasers. The two beams are overlapped using a dichroic mirror before being launched into the ion trap.

coming directly out of the diodes are shown in Figure 7.5.4. The blue laser is seen to have a nearly perfect Gaussian profile with a waist in the horizontal and vertical directions of 1.9 and 2.1 mm respectively. The red laser profile is not so nice due to the fact that the red laser does not have beam shaping optics at the output of the diode. To produce a nicer beam profile two cylindrical lenses (focal lengths of 50mm and 75mm), combined with an aperture, were used to produce the beam profile shown in Figure 7.5.5. As shown, the profile is much closer to that of an ideal Gaussian beam. The horizontal and vertical waists after shaping are 2.5 and 2.0 mm respectively.

7.6 Laser modulation

To observe ions using the synchronous photon counting and intermodulation techniques the amplitude of the lasers had to be modulated on and off. This was accomplished using acousto-optical modulators (AOMs). The principles of operation of the AOM are discussed briefly here.

Figure 7.5.4: Spatial profiles of the blue (left) and red (right) lasers. The blue laser has a nearly perfect Gaussian profile with a horizontal waist of 1.9 mm, and a vertical waist of 2.1 mm. The red laser lacks shaping optics at the output of the diode and has a much worse profile.

Figure 7.5.5: Spatial profile of the red laser after the shaping optics. The beam is much more Gaussian and has a horizontal and vertical waist of 2.5 and 2.0 mm respectively.

An acousto-optic interaction takes place whenever a laser beam and an acoustic wave are present in an optical medium. The acoustic wave generates a refractive index wave that essentially acts as a grating, diffracting the laser beam into several orders. If the AOM is properly designed, then most of the laser beam power will be diffracted into the first order beam. The AOM works as an amplitude modulator by diffracting the laser beam such that the first order diffracted beam is lined up with a slit or aperture. When the AOM is on (acoustic wave is present) the beam is diffracted and passes through the aperture. When it is off (acoustic wave not present), the beam is not diffracted and is blocked by the aperture (see Figure 7.6.1).

As previously mentioned, there is an intrinsic risetime necessary for the beam to go from the full off state to the full on state. This is due to the transit time of the acoustic wave across the diameter of the beam and results in a risetime that is dependent on the beam waist inside the optical medium. For the AOM used with the blue laser²¹ the risetime was quoted at 110 ns / mm beam diameter. The beam emitted from the diode has a diameter of roughly 2 mm and thus the risetime would be ~ 220 ns. For the photon counting technique, where the modulation frequencies are low, this risetime is sufficient. However, for the intermodulation technique the modulation frequencies are high and thus to decrease the risetime the beam was focused into the AOM using a 500 mm focal length lens. This produced a beam waist inside the AOM of roughly 0.15 mm and thus a risetime of ~ 20 ns.

For the red laser the AOM used²² had a risetime of 150 ns / mm beam diameter. A 200 mm focal length lens was used to focus the red beam into the AOM such that the beam waist was roughly 0.06 mm producing a risetime of ~ 10 ns.

One other effect to take into account for the AOMs is that they produce a shift in the frequency of the outgoing beam that is equal to the frequency of the acoustic wave. This is a Doppler frequency shift. For the blue AOM the shift is 85 MHz and for the red

²¹NEOS AO Modulator 35085-0.5

²²NEOS AO Modulator 23080-1

Figure 7.6.1: Schematic of how the AOM accomplishes laser beam modulation. At the top the AOM is off, the beam is not diffracted and the beam is blocked by the aperture. At the bottom, the AOM is on, the beam is diffracted, and the first order beam passes through the aperture. The first order beam is then sent to the ion trap to initiate fluorescence.

it is 80 MHz. These frequency shifts are taken into account when tuning the frequencies of the diode lasers.

Chapter 8

Experimental results

In this chapter I present the results and measurements obtained from the fluorescence of Ba^+ . In the first section I present an analysis of the background scattered light and use this to make an estimate of the SNR for single ion observation. I then present the results of the searches for single ions using the three different techniques attempted. I follow this by presenting some of the measured linewidths taken while both lasers are on (continuous excitation) and for the intermodulation case.

8.1 Background light

Keeping the scattered background light at a minimum was certainly the most challenging aspect of this work. Drifts in the beam alignment forced us to have to realign the lasers every day which made comparison and repetition of results taken at different times difficult. Nonetheless, it was important to try to quantify the rate of scattered light as this should predict the sensitivity to the single ion observation.

The counting rate of the scattered light is plotted in Figure 8.1.1. The left side of this figure shows the scattering rate as a function of time and displays the overall stability of the laser power. The scattering rate is quite steady in time with deviations within 1%. The right side of the plot shows the scattering rate as a function of the blue laser power.

Figure 8.1.1: Analysis of the scattered background light. The left side of the plot shows the stability in time of the scattered laser light for different powers of the blue laser. The right side of the plot shows the rate of scattered light detected as a function of the power of the blue laser. Note that the error bars on the points on the right plot are too small to be seen.

We can see from this plot that the rate of scattering increases relatively linearly with the blue power over most of the range of powers tested. There is somewhat of a deviation from this at the highest counting rates and this is believed to be due to the counting rate approaching the pulse-pair resolution of the amplifier/discriminator.

In order to use this information to predict the SNR (using Equation 6.1.21) for observing single ions the scattered light must be expressed as a fraction of the blue power. We can calculate this by fitting the curve in Figure 8.1.1 with a line and determining the slope. Due to the non-linear behaviour of the curve at higher laser powers, only the linear portion of the curve is fitted (we do not normally run at such high laser powers anyway). The fit is shown in Figure 8.1.2. The slope was determined to be $0.009 \text{ MHz} / \mu\text{W}$ which we can convert into a percentage using the energy of the blue photons. We find that the scattering rate is $3.6 \times 10^{-7}\%$ of the power of the blue laser. We note that this number will also include the detector acceptance since what is shown is the *measured* scattered rate. Thus, if we use the estimated 0.7% detector efficiency then we find a background parameter of roughly 5×10^{-5} . Looking at Figure 6.1.8 we can estimate the SNR for a

Figure 8.1.2: Fit to the rate of the background light. The slope determines the fraction of light that is scattered from the blue laser.

one second observation to be roughly 4 *if* everything is tuned perfectly.

8.2 Barium ion observation

8.2.1 Direct detection (CCD Imaging)

The first technique attempted to see single ions used the CCD camera. The CCD was used early on in this work as a trouble-shooting tool and, unfortunately, single-ions were never observed. This technique was successful at observing more than one barium ion and was in fact the first technique for which we observed anything.

Two images taken of the barium ions inside the trap are shown in Figure 8.2.1. In the picture on the left the potential minimum along the trap axis is set over the two trapping electrodes such that the ions should be localized in the center of these two electrodes. On the right side of the figure the potential minimum is set to be over only one of the trapping electrodes such that the ions should be localized at the center of only this electrode. Although no critical analysis was completed on this data it does serve nicely to illustrate that ions are indeed being trapped, and that we have control over

Figure 8.2.1: Trapped barium ions inside of the linear ion trap. For the picture on the left the potential minimum along the trap axis was set over the two trapping electrodes. In the right picture the potential minimum was only over one of the two trapping electrodes. The laser enters from the right. The scale of the image is indicated by the size of the trapping electrode and the gap between the two electrodes.

their position in the trap by setting the DC potentials suitably.

8.2.2 Synchronous photon counting

Unfortunately, this technique was also unsuccessful at observing single ions, although the observation of many ions was easily accomplished. To illustrate the technique, an example of one of the “scans” used to analyze the ion signal is shown in Figure 8.2.2. What is shown is the ion fluorescence rate over one full period of the red laser modulation cycle. The modulation frequency is set to 100 Hz, and the blue and red laser powers were set to 40 μ W, and 5 mW respectively. In the first 5 ms both the red and blue lasers are on, the ions are fluorescing, and the count rate is high (this is a lot of ions!). In the last 5 ms the red laser is turned off and the fluorescence ceases quickly. To determine the fluorescence rate the first half of the period is integrated to get the signal plus background

Figure 8.2.2: Example of a scan obtained using the synchronous photon counting technique. For the first 5 ms the red and blue lasers are both on and the ions are fluorescing. In the last 5 ms the red laser is turned off and the fluorescence quickly stops. The inset in the top right shows the first 0.2 ms of the scan where a peak is observed to occur. The modulation frequency was set to 100 Hz, and the blue and red laser powers were 40 μ W and 5 mW respectively.

counts, and the second half of the period is integrated to get the background counts. The signal is then the difference between these two numbers.

One interesting thing to note is the sharp peak at the very start of the scan which I have highlighted in the inset of the figure. This peak is present in every scan that is taken, regardless of the laser powers, modulation frequency, or any other experimental parameter. This is a real effect and is predicted to occur by the optical Bloch equations (Equations 5.4.26 to 5.4.31). A theoretical scan is shown in Figure 8.2.3 for the same modulation frequency and laser powers as used for the scan in Figure 8.2.2. The peak in the P-state population at the start of the scan is clearly visible, although somewhat higher than the peak in the experimental scan. The effect of Doppler broadening has not been considered for the theoretical scan, but it is clear from the experimental one that the peak is still present for this case.

One final measurement that is worth showing is the dependence of the fluorescence

Figure 8.2.3: Theoretical scan for the same experimental parameters as used in Figure 8.2.2 obtained by solving the optical Bloch equations. The peak at the start of the scan is clearly visible. Doppler broadening has not been considered for this scan.

light on the power of the blue laser. This dependence is shown in Figure 8.2.4. To obtain this plot the trap was filled with ions and scans like the one shown in Figure 8.2.2 were obtained for blue laser powers in the range 5 - 325 μW and a red laser power of 5 mW. The red laser wavelength was shifted by 80 MHz to avoid the two photon resonance. As we are very much in the region where the signal is greater than the background, the SNR will simply follow a square-root dependence on the laser power. If we compare this to the theoretical dependence on the laser power as shown in Figure 6.1.7 we can see that the shape matches well with what is expected.

8.2.3 Intermodulation

This was the only technique successful at observing single ions. For this reason I will go through the analysis of the data in some detail. To start, I will present the proof that single-ions were indeed observed and then show how this evidence was obtained. Note that, in all single ion searches, the He buffer gas pressure was set to 5×10^{-3} mbar.

Figure 8.2.4: Dependence of the Ba^+ fluorescence rate on the power of the blue laser. This scan was taken with the red laser set at 5 mW with a detuning of 80 MHz.

In order to know that single ions have been observed, the fluorescence rate is monitored as a function of time and a discrete step-like pattern should appear as single ions are ejected from the trap. These steps should also be approximately equal in size as each ion should have the same fluorescence rate. Thus, by analyzing the background rate before or during the measurement one can infer the time when the last ion is in the trap.

Figures 8.2.5 and 8.2.6 display the expected step-like behaviour perfectly. In both figures the fluorescence rate is plotted as a function of time on the left with each data point representing five seconds of integration time. In Figure 8.2.5 there are initially four ions inside the trap and one by one they are ejected. To determine the SNR for single ion observation the data points in the time series are histogrammed and fitted with a Gaussian function to determine the mean and width of each ion signal. The fluorescence rate is determined by the difference in means between the bottom two histograms which represent the single ion and background rates respectively. The single-ion fluorescence rate was found to be 60 ± 13 Hz which corresponds to a SNR of 4.6. The lifetime

Figure 8.2.5: Evidence for the observation of single ions in the linear ion trap. Initially there were four ions trapped until one by one they were ejected leaving the trap empty. The SNR ratio for single ion observation was found to be 4.6 for a five second observation time.

of the single ion in the trap for this data set was determined to be 550 seconds.

In Figure 8.2.6 only three ions were initially in the trap. In this case the single ion signal was determined to be 53 ± 6 Hz corresponding to a SNR of ~ 8.8 again for five seconds of integration time. The lifetime of this ion was found to be much less at 145 seconds. The observation of single ions was the main goal of this work and these two figures show that this goal was achieved.

The process for obtaining the above figures was as follows. First, the trap was loaded with ions by turning the Pirani/cold cathode pressure gauge on. The fluorescence signal was then monitored while the lasers were carefully aligned to produce the highest signal rate and lowest background rate. The pressure gauge was then turned off and the fluorescence rate was monitored until the discrete steps were observed. For both of the above plots the modulation frequency was set at 0.2 MHz, and the laser powers were 1 mW and 5 mW for the blue and red lasers respectively. The duty cycle of the blue laser pulse was set to 56% or a pulse width of $2.8 \mu\text{s}$. I will discuss the choice of the duty cycle for the red laser below. The pressure was relatively high at 5×10^{-3} mbar He.

Figure 8.2.6: Evidence for the observation of single ions in the linear ion trap. Initially there were three ions trapped until one by one they were ejected leaving the trap empty. The SNR ratio for single ion observation was found to be 8.8 for a five second integration time.

Since the intermodulation technique does not simply output a fluorescence rate some analysis was necessary to extract a rate from the raw data. A scan obtained from the multi-channel analyzer (MCA) is shown in Figure 8.2.7 with *no* fluorescence present. In the first $2.8 \mu\text{s}$ the blue laser is on and the scattered photon rate is high. Once the blue laser is turned off there is approximately one bin (100 ns) where the risetime of the AOM has an effect and the laser power ramps down. The cause of the higher background counts in the subsequent bins is most likely due to noise in the PMT from the high count rate experienced in the first part of the scan. The last 6 bins of the scan are relatively background free and this is where we want to start the red laser pulse.

The same scan as shown in Figure 8.2.7 is plotted in Figure 8.2.8 this time with ions in the trap. The red pulse was set to start at the beginning of the 46th bin (4th from last) and the fluorescence signal is clearly visible above the background. The fluorescence signal is then calculated by integrating these 4 bins.

Figure 8.2.7: Example of an intermodulation scan showing the blue laser pulse (first 2.8 μs) and the level of the background where the red pulse, and thus fluorescence signal, should start (last four bins).

Figure 8.2.8: Example of an intermodulation scan showing the fluorescence signal which occurs in the last four bins when the red laser is turned on.

8.3 Linewidths

8.3.1 Continuous excitation

As single ions were never observed using a technique where both lasers were on, single-ion linewidths were not obtained using this method. However, linewidths were obtained by filling the trap with many ions and scanning the laser frequencies across the resonance frequency of the transition. The blue laser frequency was easily tuned using software provided with the interferometer. One simply had to input the desired frequency and the laser would then lock there, provided that the frequency was in the free tuning range of the laser. For the blue laser this tuning range was roughly ± 1 GHz which more than covered the frequencies needed to generate a linewidth. The free tuning range of the red laser was significantly less than the blue, so linewidths are only presented for the case that the blue laser is scanned over the transition frequencies.

A linewidth obtained by scanning the blue laser frequency is shown in Figure 8.3.1. To obtain this linewidth the blue and red laser powers were set to 40 μW and 500 μW respectively, and the red laser had a detuning of roughly 80 MHz (below the resonance). We can compare this to a theoretical linewidth using the beam diameters of the lasers presented in the previous chapter to calculate the expected Rabi frequencies. We get $\Omega_B \approx 18 \text{ MHz}$ and $\Omega_R \approx 53 \text{ MHz}$. Using a temperature of 300 K we get the linewidth shown in Figure 8.3.2 (plotted with the experimental linewidth). The theoretical linewidth matches perfectly the experimental linewidth for blue detunings $\delta_B > 0$ MHz, however, for negative detunings the shapes differ significantly.

The most likely reason for this is that the beam diameters or laser powers are not well characterized at the trapping region (we can only measure beam properties outside of the vacuum chamber). In addition, since we are using a source of natural barium and this linewidth was taken with many ions in the trap, we can expect that there will be more than just the ^{138}Ba isotope present. This is somewhat problematic since there is a shift in

Figure 8.3.1: Experimental linewidth obtained by scanning the blue laser frequency across the resonant frequency of the $P \rightarrow S$ transition. The blue and red laser powers were set to $40 \mu\text{W}$ and $500 \mu\text{W}$ respectively, and the detuning of the red laser was set to 80 MHz (below resonance).

the resonant frequencies for the two transitions for different isotopes, termed an isotope shift. The odd isotopes, ^{135}Ba and ^{137}Ba , are both stable with natural abundances of 6.592% and 11.232% [45] respectively. However, due to the hyperfine interaction, the odd isotopes have large enough isotope shifts that we should not see their effect on the linewidths. The ^{136}Ba isotope on the other hand has an isotope shift well within the range of the linewidths shown - $\nu_{138} - \nu_{136} = 179.4 \text{ MHz}$ for the $P \rightarrow S$ transition [92] and -68.0 MHz for the $P \rightarrow D$ transition [93]. This isotope also has a natural abundance of 7.854% [45] making it likely that it was present when this linewidth was taken. This would have the effect of increasing the right side of the linewidth which is possibly why the shape agrees so well with theory (as in, it is a coincidence).

Despite this, the qualitative features expected of the experimental linewidth are present. Namely, the dip in fluorescence corresponding to the two-photon-resonance is clearly visible and it does not dip down to zero fluorescence due to the effect of Doppler broadening. In addition, the overall shape does appear to have a Voigt-like profile.

Figure 8.3.2: Comparison of the theoretical (dashed line) and experimental linewidth obtained by scanning the blue laser. The Rabi frequencies for the theoretical linewidth were set at $\Omega_B = 18$ MHz and $\Omega_R = 53$ MHz respectively, and the detuning of the blue laser was set to -80 MHz.

8.3.2 Intermodulation

Since single ions were observed using this method it was possible to measure the linewidth of a single ion. The procedure for measuring the linewidth was difficult since ions do not have an infinite lifetime in the trap and one needs roughly five to ten minutes to set up and take the linewidth. Nearly every attempt to obtain a linewidth failed due to the ion being ejected before the laser frequency scan had finished. To lessen this constraint the trap was filled with many ions and linewidths were repeatedly measured until it was observed that all of the ions had left the trap. Due to time constraints only a couple of these scans were obtained successfully. The best scan obtained using this method is shown in Figure 8.3.3. This linewidth was taken with the blue and red laser powers set to 1 mW and 5 mW respectively. A modulation frequency of 0.2 MHz was used and the timing of the blue and red pulses was set the same as was used to observe the single ion steps (section 8.2.3).

As was done in the previous section, we can calculate the expected linewidth given

Figure 8.3.3: Single ion linewidth obtained using the intermodulation technique. The blue and red laser powers were set to 1 mW and 5 mW respectively.

the experimental parameters used. In this case we have $\Omega_B = 92 \text{ MHz}$ ¹ and again using a temperature of 300 K we get the linewidth shown in Figure 8.3.4. For the single ion the theoretical curve matches the experimental data much better which leads us to think that at least part of the discrepancy for the continuous-excitation linewidth is likely due to contamination by ^{136}Ba . Unfortunately, the statistics here are low, and whether or not this would look as good as it does now with more statistics is debatable. It is comforting, however, that the theoretical curve does approximately predict what is observed in experiment.

¹The red is not important since it just changes the normalization, which, due to the fact that we fit the normalization, does not matter (see section 6.2.1).

Figure 8.3.4: Comparison of the theoretical linewidth with the experimental linewidth obtained by scanning the blue laser. The Rabi frequency of the blue laser was $\Omega_B = 92$ MHz.

Chapter 9

Summary and conclusions

In this work we have investigated the importance of an observation of the $0\nu\beta\beta$ -decay and showed how this can lead to the development of physical models beyond the standard model and a better understanding of the nature of the neutrino. The ability of experiments searching for the $0\nu\beta\beta$ -decay were investigated using the concepts of sensitivity and discovery potential. In particular, we found that experiments will have to employ advanced background reduction techniques in order to have a chance of observing the $0\nu\beta\beta$ -decay, since, simply increasing source mass and exposure times will not provide much gain in sensitivity if the search is limited by background contamination. One such technique, barium-tagging, is a particularly powerful technique that has the potential of reducing background contamination to the point where the entire inverted hierarchy and a portion of the normal hierarchy of neutrino masses may be excluded with high confidence. Furthermore, if a neutrino mass does exist in the region of the inverted hierarchy, then an experiment employing the barium-tagging technique will likely discover it. This, in turn, provides the main motivation for the experimental portion of this work, which was to set up and demonstrate a working system for the trapping and identification of single barium ions in a linear Paul trap - a necessary step in a fully functional Ba-tagging system.

Throughout the body of the thesis the theory of ion trapping and laser induced fluorescence was presented with a particular focus on how to maximize the efficiency and sensitivity to observing single ions. In particular, two general methods for detecting the fluorescing ions were investigated: continuous excitation and intermodulation. For the continuous excitation methods the experimental parameters of primary concern to a high signal-to-noise ratio observation of single ions were discussed in detail. It was shown that a careful tuning of the fluorescence laser amplitudes and frequencies can provide an extremely high signal-to-noise ratio (SNR) even in the presence of a buffer gas. To achieve this observation in practice, the amount of background light must be kept at a minimum and the efficiency of observing the fluorescent photons maximized. For the intermodulation technique, the timing and widths of the blue and red laser pulses are of critical importance due to the practical aspects of how the laser pulses are created. In principle, the SNR for this method should be very high due to the measurement being relatively background free.

At the end of this report I present the analysis and results of the search for single-ions using the experimental system described in Chapter 7. An analysis of the background light contaminating the single-ion observation is given in order to estimate what the ion sensitivity should be. A signal-to-noise ratio of 4 was predicted for a one second observation, provided that all experimental parameters were perfectly tuned. The observation of barium ions using direct detection (CCD imaging) was presented and control over the ions in the ion trap was demonstrated. Barium ions were also observed using the continuous excitation technique of synchronous photon counting, and the measured dependence of the fluorescence rate on the blue laser power was comparable to what was expected from theory. Unfortunately, single barium ions were never observed using either of the continuous excitation techniques. The reasons for this are not entirely known, however, it is expected that the efficiency of photon detection is most likely much lower than expected. The intermodulation technique *was* successful at observing single ions and the

analysis of the single ion signal yielded a maximum signal-to-noise ratio of 8.8 in five seconds of observation time, which, roughly corresponds to a one second signal-to-noise ratio of 4. This demonstrated that the intermodulation technique is the more sensitive technique for observing single-ions.

I end the thesis by presenting measurements of the linewidths of the barium ion using the continuous excitation and intermodulation techniques. The linewidth obtained for the continuous excitation method was taken with many ions in the trap and was thus susceptible to contamination by other naturally occurring isotopes of barium. Specifically, by comparing the linewidth obtained with that expected from theory, it was concluded that the ^{136}Ba isotope was most likely present with the more abundant ^{138}Ba isotope. The linewidth presented for the intermodulation technique was taken with a single ion in the trap and was much better described by what was theoretically expected.

In conclusion, the intermodulation technique was found to be much more sensitive to single ion observation than the continuous excitation techniques. The relatively background free measurement of the fluorescence photons provides a high SNR, and, combined with the ability for fast detection, makes it a suitable technique to be used in a future tonne scale ^{136}Xe detector looking to employ the barium-tagging technique.

Appendix A

Statistics

The statistical theory of hypothesis testing, limit-setting, and parameter estimation is presented here for reference following the treatment given in [95].

A.1 Hypothesis testing

The main goal of a statistical hypothesis test is to make some kind of statement on how well the observed data agree with prior expectations. Traditionally the hypothesis in question is called the null hypothesis and is represented by H_0 . It is often the case that we wish to compare the validity of H_0 to some alternative hypothesis, H_1 . In order to see how to do this let us consider an experiment that has made a series of n measurements of some variable that we will represent as $\vec{x} = \{x_1, x_2, \dots, x_n\}$. Normally, a test statistic $t(\vec{x})$ is constructed from the data in order to reduce the dimensionality of the analysis without losing discrimination power. The test statistic may be a simple function of the data, a complicated function, or may just be the data itself. Whatever the test statistic may be, the important thing is that the two hypotheses H_0 and H_1 will specify (we will assume) unique probability distributions for the test statistic, $f(t | H_0)$ and $f(t | H_1)$ respectively. By comparing the likelihood of getting the observed test statistic under the assumption of H_0 or H_1 one can then make a statement as to the probability that the

data came from H_0 or H_1 .

It is best to understand this using an example. Suppose the pdfs for t under the assumptions of H_0 and H_1 are as shown in Figure A.1.1. To determine if the observed value of the test statistic t_{obs} is more compatible with H_0 or H_1 one defines an acceptance region for t defined by a boundary t_{cut} such that if $t_{obs} < t_{cut}$ we accept H_0 , and if $t_{obs} > t_{cut}$ we accept H_1 . There will of course be some probability α to reject H_0 even if it is true (called an error of the first kind), and, referring to Figure A.1.1, we can see that α is given by,

$$\alpha = \int_{t_{cut}}^{\infty} f(t | H_0) dt \quad (\text{A.1.1})$$

Likewise, the probability that H_0 is accepted if H_1 is actually true (error of the second kind) is given by,

$$\beta = \int_{-\infty}^{t_{cut}} f(t | H_1) dt \quad (\text{A.1.2})$$

The probability α is also called the significance of the test. The power of the test is defined as $1 - \beta$ and represents the ability of an experiment to discriminate against the alternative hypothesis H_1 .

The hypothesis test illustrated above is a useful tool especially when an experiment is trying to decide if an event belongs to one class or another, or if a particle is an electron or a muon for example. A closely related concept to the significance of a hypothesis test is the so called P-value which is defined by,

$$P_{val} = \int_{t_{obs}}^{\infty} f(t | H_0) dt \quad (\text{A.1.3})$$

In contrast to α , which is fixed before the experiment and represents the significance of a *test*, the P-value represents the significance of the experimental *result* and is thus a random variable. In fact, we could equivalently express the decision to accept or reject

Figure A.1.1: Illustration of hypothesis testing. The pdf on the left is under the assumption of H_0 and the pdf on the right is under H_1 . If the observed value of the test statistic is less than t_{cut} then H_0 is accepted, and rejected otherwise.

H_0 based on the P-value, since, $P_{val} \leq \alpha$ implies $t_{obs} \geq t_{cut}$ and *vice versa*. In a literal sense, the P-value expresses the probability of obtaining the observed result or something more extreme if the null hypothesis were actually true.

We will use the P-value to define statistically what we mean by a discovery. Using the now common standard of a “five-sigma significance” our condition for discovery is,

$$P_{val} \leq \frac{1}{\sqrt{2\pi}} \int_5^\infty \exp\left(\frac{-x^2}{2}\right) dx \approx 3 \times 10^{-7}$$

which simply states that we require the P-value to pass a significance test with α equal to the integral of the tail above 5σ for a standard Gaussian function.

A.2 Parameter estimation

Consider an experiment that measures a random variable x with some pdf $f(x)$. We will assume that we do not know the exact form of the pdf but that we do know that it can

be parameterized as $f(x|\vec{\theta})$ according to some unknown set of parameters $\vec{\theta} = \{\theta_i\}$. The goal of parameter estimation, then, is to determine the “best” estimates for the value of $\vec{\theta}$ based on the observed data. A common technique of parameter estimation is the method of maximum likelihood, and, as this is the method used by EXO, it’s the only one I will discuss.

To illustrate the maximum likelihood method we again consider an experiment measuring x and assume that the experiment has finished and obtained n independent measurements $\vec{x} = \{x_1, x_2, \dots, x_n\}$. Since the probability of observing any one x_i is given by $f(x_i|\vec{\theta})$ we can write the total probability of observing all the x_n as,

$$f(x_1|\vec{\theta}) \cdot f(x_2|\vec{\theta}) \cdots f(x_n|\vec{\theta}) = \prod_{i=1}^n f(x_i|\vec{\theta}) \quad (\text{A.2.1})$$

As we do not know the value of $\vec{\theta}$ we cannot calculate the exact likelihood of obtaining the observed data. However, we can expect that if we choose a value of $\vec{\theta}$ close to the true value then the probability of obtaining the data, as calculated by the above, should be relatively high. This motivates us to define the likelihood-function,

$$L(\vec{\theta}) = \prod_{i=1}^n f(x_i|\vec{\theta}) \quad (\text{A.2.2})$$

The maximum likelihood method is then simply defined by maximizing the above function and taking the resulting value of $\vec{\theta}$ as the estimators to the true values. If we represent the estimators using a $\vec{\theta}'$ then, mathematically, we have,

$$\left. \frac{\partial L(\theta_i)}{\partial \theta_i} \right|_{\theta_i=\theta'_i} = 0 \quad (\text{A.2.3})$$

Instead of maximizing the likelihood function, it is more common to minimize the negative-log-likelihood function defined by,

$$NLL(\vec{\theta}) = -\log L(\vec{\theta}) = -\sum_{i=1}^n \log [f(x_i | \vec{\theta})] \quad (\text{A.2.4})$$

Since the logarithm is a monotonically increasing function, minimizing the above will give the exact same estimators as maximizing $L(\theta)$. The process of determining the parameters by minimizing a likelihood function is often termed as a “fit to the data” and the parameters $\vec{\theta}$ are the “fit results”.

A.2.1 Extended likelihood function

It is often the case that the pdf $f(x | \vec{\theta})$ is a simple sum of m known, component pdfs $f_i(x)$, with unknown relative fractions θ_i ,

$$f(x | \vec{\theta}) = \sum_{i=1}^m \theta_i f_i(x) \quad (\text{A.2.5})$$

Since the total pdf must still be normalized we clearly have the condition that,

$$\sum_i^m \theta_i = 1 \quad (\text{A.2.6})$$

.

In this situation it is useful to treat the number of observations n as a Poisson random variable with mean value ν . The total likelihood function should then be multiplied by the Poisson probability for getting n observations,

$$L(\vec{\theta}) = \frac{\nu^n}{n!} e^{-\nu} \prod_{i=1}^n f(x_i | \vec{\theta}) \quad (\text{A.2.7})$$

which is called the *extended* likelihood function. Consequently, the extended negative-log-likelihood function¹ is,

¹We have neglected the $\log n!$ term as the estimators $\vec{\theta}$ only depend on the derivatives of NLL and thus constant terms will have no effect.

$$NLL(\vec{\theta}) = -n \log \nu + \nu - \sum_{i=1}^n \log [f(x_i | \vec{\theta})] \quad (\text{A.2.8})$$

$$= \nu - \sum_{i=1}^n \log \left[\sum_{j=1}^m \nu \theta_j f(x_i) \right] \quad (\text{A.2.9})$$

Due to the fact that the θ_i sum to unity, we can define μ_j as the expected number of events of type j where,

$$\mu_j = \nu \theta_j \quad (\text{A.2.10})$$

and,

$$\sum_{j=1}^m \mu_j = \nu \quad (\text{A.2.11})$$

Using these *yield parameters* μ_j we can redefine the likelihood function as,

$$NLL(\vec{\mu}) = \sum_{j=1}^m \mu_j - \sum_{i=1}^n \log \left[\sum_{j=1}^m \mu_j f(x_i) \right] \quad (\text{A.2.12})$$

A.2.2 Binned likelihood function

If the number of observations made in experiment is large then the minimization of the likelihood function can take a long time. A common technique to reduce the complexity of the calculation is to bin the data first and then build a likelihood function from the histogram created. In this case we simply have to modify the likelihood function given above as follows,

$$NLL(\vec{\mu}) = \sum_{j=1}^m \mu_j - \sum_{i=1}^N n_i \log \left[\sum_{j=1}^m \mu_{i,j} \right] \quad (\text{A.2.13})$$

where N is the number of bins and n_i is the observed number of events in bin i . The $\mu_{i,j}$ represent the expected number of events in the i^{th} bin and are given by,

$$\mu_{i,j} = \mu_j \int_{\text{bin } i} f_j(x | \theta) dx \quad (\text{A.2.14})$$

A.2.3 Error propagation in a likelihood estimation

It is commonly the case that the yield parameters μ_j can themselves be expressed as a function of some fixed quantity η_j and an unknown parameter ν_j ,

$$\mu_j = \epsilon_j \eta_j \quad (\text{A.2.15})$$

such that the likelihood function can be written as,

$$NLL(\vec{\nu}) = \sum_{j=1}^m \nu_j \eta_j - \sum_{i=1}^n \log \left[\sum_{j=1}^m \nu_j \eta_j f(x_i) \right] \quad (\text{A.2.16})$$

If the values of the η_j are known exactly then minimizing the above will not change the final results for the μ_j . Now let us assume that the η_j are known to have some uncertainty in their values defined by σ_{η_j} . We can include this uncertainty in our likelihood function by allowing the values of the η_j to float in the fit and including a constraint term that reflects the level of uncertainty on the η_j . We will assume that the parameters η_j are not correlated and are Gaussian distributed with a mean of $\bar{\eta}_j$ and a standard deviation given by σ_{η_j} . In this case the constraint term will be given by the Gaussian pdf,

$$C_j(\eta_j) = \frac{1}{\sqrt{2\pi\sigma_j^2}} \exp \left[-\frac{(\eta_j - \bar{\eta}_j)^2}{2\sigma_j^2} \right] \quad (\text{A.2.17})$$

We then need to simply multiply our likelihood function (equation A.2.7) to include this uncertainty in the fit. Doing this, and taking the negative-logarithm gives,

$$NLL(\vec{\nu}, \vec{\eta}) = \sum_{j=1}^m \nu_j \eta_j - \sum_{i=1}^n \log \left[\sum_{j=1}^m \nu_j \eta_j f(x_i) \right] + \sum_{j=1}^m \frac{(\eta_j - \bar{\eta}_j)^2}{2\sigma_{\eta_j}^2} \quad (\text{A.2.18})$$

where we again have neglected constant terms that will not affect the final results for the parameters.

A.3 Confidence intervals

Consider an experiment that measures some random variable x in order to determine a parameter θ . Once the experiment is over and the estimator θ' is determined, the next step is to report the result to the community. A common way of doing this is to report a confidence interval (CI) for θ that reflects the statistical variance in the measurement. Let us assume that the estimator obtained in the experiment is θ'_{obs} . Since the actual value of θ'_{obs} depends on a random sample of data, it itself will be a random variable characterized by some pdf $g(\theta'|\theta)$ that depends on the true value of the parameter θ . We will again assume that we do not know the exact form of $g(\theta'|\theta)$ but that this is only due to our not knowing the true value of θ .

We can define two numbers u_α and u_β such that the probabilities to observe $\theta' \geq u_\alpha$ or $\theta' \leq u_\beta$ are given by α and β respectively. Since the pdf of θ' is given by $g(\theta'|\theta)$ we can write the probabilities α and β as,

$$\alpha = P(\theta' \geq u_\alpha) = \int_{u_\alpha}^{\infty} g(\theta'|\theta) d\theta' \quad (\text{A.3.1})$$

$$\beta = P(\theta' \leq u_\beta) = \int_{-\infty}^{u_\beta} g(\theta'|\theta) d\theta' \quad (\text{A.3.2})$$

We assume that the probabilities α and β are fixed beforehand such that the specific values of u_α and u_β are determined by the true value θ , that is, $u_\alpha = u_\alpha(\theta)$ and $u_\beta = u_\beta(\theta)$.

If we further assume that $u_\alpha(\theta)$ and $u_\beta(\theta)$ are monotonically increasing functions of θ then we can define their inverse functions by,

$$a(\theta) = u_\alpha^{-1}(\theta) \quad (\text{A.3.3})$$

$$b(\theta) = u_\beta^{-1}(\theta) \quad (\text{A.3.4})$$

If we apply the inverse functions to the inequalities $\theta' \geq u_\alpha$ or $\theta' \leq u_\beta$ we get,

$$a(\theta') \geq \theta \quad (\text{A.3.5})$$

$$b(\theta') \leq \theta \quad (\text{A.3.6})$$

which allows us to rewrite equations A.3.1 and A.3.2 as,

$$\alpha = P(a(\theta') \geq \theta)$$

$$\beta = P(b(\theta') \leq \theta)$$

Combining these together gives us the probability of finding $b(\theta') \leq \theta \leq a(\theta')$,

$$P(b(\theta') \leq \theta \leq a(\theta')) = 1 - \alpha - \beta \quad (\text{A.3.7})$$

If $a(\theta')$ and $b(\theta')$ are evaluated at the observed value of θ'_{obs} , then the interval $[a(\theta'_{obs}), b(\theta'_{obs})]$ is called a confidence interval for θ at a confidence level (CL) of $1 - \alpha - \beta$.

In order to calculate the values of $a(\theta')$ and $b(\theta')$ we note that $a(\theta')$ represents the hypothetical value of θ such that a fraction α of repeated experiments would produce a value of θ' larger than the one observed. Likewise, $b(\theta')$ represents the value of θ such

that a fraction β of repeated experiments would produce a value of θ' lower than the one observed. Writing this out mathematically gives us,

$$\alpha = P(\theta' \geq \theta'_{obs} | a) = \int_{\theta'_{obs}}^{\infty} g(\theta' | a) d\theta' \quad (\text{A.3.8})$$

$$\beta = P(\theta' \leq \theta'_{obs} | b) = \int_{-\infty}^{\theta'_{obs}} g(\theta' | b) d\theta' \quad (\text{A.3.9})$$

which is the standard way of determining the limits a and b . We note that, from the above definition, there will be some ambiguity in the CI since there are any number of ways to choose α and β to give the same CL. One therefore has to define some condition on the α and β to fix the CI. A common choice is the central CI which takes $\alpha = \beta$.

A common scenario in particle physics occurs when one is searching for a new effect that will be characterized by $\theta \neq 0$ if the effect exists and $\theta = 0$ if it does not. If the experiment is run and θ' is found to be significantly non-zero (as decided by a hypothesis test) then a confidence interval for the parameter can be reported and the effect discovered. More times than not, however, the experiment is run and the parameter θ' is consistent with zero. In these cases it does not make sense to report a confidence interval and instead the experiment will report an upper limit on the parameter. In order to this we simply set $\alpha = 0$ in our definition of the confidence interval such that the upper limit is given by,

$$\beta = \int_{-\infty}^{\theta'_{obs}} g(\theta' | \theta_{up}) d\theta' \quad (\text{A.3.10})$$

where I have written $b = \theta_{up}$ to remind ourselves that this is a limit and not a confidence interval. The CL of the limit is simply,

$$\text{CL} = 1 - \beta \quad (\text{A.3.11})$$

A.4 Sensitivity and discovery potential

So far we have only been considering examples where an experiment has been run and a result on the parameter θ is obtained, be it a limit or a confidence interval. The sensitivity and discovery potential are a means to characterize the ability to see a new effect for a planned experiment. In this section I give the formal definitions for the sensitivity and discovery potential of a planned experiment.

A.4.1 Sensitivity

The sensitivity of an experiment wants to answer the question “What is the most likely limit that will be placed on a new effect assuming that the effect does not exist?”. In order to answer this question consider the upper limit θ_{up} derived in the last section. Since the upper limit depends on the observed value of the parameter θ'_{obs} , the upper limit will itself be a random variable defined by some pdf $h(\theta_{up} | \theta)$ that depends on the true value of the parameter θ . The formal definition of the sensitivity is the median upper limit at a confidence level of 90%, assuming the parameter $\theta = 0$. Thus, the sensitivity $\tilde{\theta}_{up}$ is found by solving,

$$\begin{aligned} 0.5 &= \int_{\tilde{\theta}_{up}}^{\infty} h(\theta_{up} | \theta = 0, \beta = 0.1) d\theta_{up} \\ &= \int_0^{\tilde{\theta}_{up}} h(\theta_{up} | \theta = 0, \beta = 0.1) d\theta_{up} \end{aligned} \quad (\text{A.4.1})$$

From the above definition we can see the true meaning of the sensitivity. If an experiment has a sensitivity of $\tilde{\theta}_{up}$ then that experiment can expect to put a limit on the parameter θ at least as good as $\tilde{\theta}_{up}$ in half of an ensemble of identically run experiments.

A.4.2 Discovery potential

The discovery potential of an experiment wants to answer the question “What kind of signal would we need in order to make a discovery at the ‘5-sigma’ level?”. Where the sensitivity closely relates to the concept of limit setting, the discovery potential relates to the concept of hypothesis testing. Thus, consider an experiment that is trying to discover a new effect that will be characterized by the parameter θ being non-zero. We will call the null hypothesis H_0 the hypothesis that the effect does not exist. We will assume that the pdf for our test statistic t has the pdf $f(t|\theta)$ where the form is known for a given θ . We define the null hypothesis as the hypothesis that no new signal exists, such that the pdf of t under H_0 is $f(t|H_0 : \theta = 0)$. From our definition of a discovery in the first section we have the condition,

$$P_{val} = \int_{t_{obs}}^{\infty} f(t|H_0 : \theta = 0)dt \leq 3 \times 10^{-7} \quad (\text{A.4.2})$$

There are, in general, an infinite number of alternative hypotheses H_i , where each H_i is characterized by a specific choice of $\theta \neq 0$. We formally define the discovery potential by the smallest value of θ that would satisfy the discovery condition given above in half of an ensemble of identically run experiments. Thus, if we represent this *least-detectable-signal* as θ_{lds} then we can determine its value by solving,

$$\int_{t_{cut}}^{\infty} f(t|H_0 : \theta = 0)dt = 3 \times 10^{-7} \quad (\text{A.4.3})$$

$$\int_{-\infty}^{t_{cut}} g(t|H_i : \theta = \theta_{lds})dt = 0.5 \quad (\text{A.4.4})$$

These will be the definitions used when determining the discovery potential for the EXO experiment.

A.5 Example calculation of sensitivity and discovery potential for the EXO experiment

I will now calculate the sensitivity and discovery potential for a more complex situation that mirrors the method used for EXO. For simplicity I will only consider one background component corresponding to the $2\nu\beta\beta$ -decay and I will assume that the shape of the pdf is given by the Primakoff-Rosen approximation (equation 2.3.8). However, due to the low probability to get any events near the Q-value, I will set the Q-value for the $2\nu\beta\beta$ pdfs somewhat higher than the actual Q-value for the decay of ^{136}Xe to artificially increase the background in this region. For the $0\nu\beta\beta$ decay I will assume a Gaussian pdf with an energy resolution of 1.5% at the Q-value. The total energy spectrum is given by the sum of the two component pdfs,

$$f(x) = \sum_{i=1}^2 \theta_i \cdot f_i(x) \quad (\text{A.5.1})$$

where f_1 (f_2) and θ_1 (θ_2) is the pdf and relative fraction for the $0\nu\beta\beta$ ($2\nu\beta\beta$) decay respectively, and the random variable x is the outgoing energy of the two electrons emitted.

We will further assume that the data $\vec{x}$ can be subdivided into two different categories based on some other information from the detector, and that we know to within 10% the relative fractions of events that should occur in each of the two categories, for each of the two components considered. If we represent these fractions as η_i then we have the two pdfs,

$$f_a(x) = \sum_{i=1}^2 \eta_i \cdot \theta_i \cdot f_{a,i}(x) \quad (\text{A.5.2})$$

$$f_b(x) = \sum_{i=1}^2 (1 - \eta_i) \cdot \theta_i \cdot f_{b,i}(x) \quad (\text{A.5.3})$$

Table A.1: Assumptions used for the EXO discovery potential and sensitivity example calculation.

$0\nu\beta\beta$	
Category a , pdf	Gaussian pdf centered at 2457.83 keV, with 1.5% resolution
Category b , pdf	Gaussian pdf centered at 2457.83 keV, with 2.0% resolution
η	0.80 ± 0.08
$2\nu\beta\beta$	
Category a , pdf	Primakoff-Rosen with Q-value = 2650 keV
Category b , pdf	Primakoff-Rosen with Q-value = 2750 keV
η	0.90 ± 0.08
Rate	1.4×10^6 decays/yr/tonne

Figure A.5.1: The shape of the $2\nu\beta\beta$ pdfs (left) and $0\nu\beta\beta$ pdfs (right) used in the calculation.

where the subscript a represents the first category and the subscript b represents the second category. This is similar to categorizing the EXO backgrounds as being single-site or multi-site. We will also assume that the pdfs of each of the two components are slightly different between the two categories, $f_{a,i} \neq f_{b,i}$. Finally, to make things specific, I will assume that $\eta_1 = 0.8 \pm 0.08$ and $\eta_2 = 0.9 \pm 0.09$, and, to stay true to EXO, I will consider the case of a 1-tonne detector filled with ^{136}Xe such that the rate of the background $2\nu\beta\beta$ -decays is known from the half-life of the decay (which I will assume is known with certainty). The summary of the assumptions made are given in Table A.1 and the shapes of the pdfs are shown in Figure A.5.1 for each of the two categories.

We can build the extended, binned likelihood functions for our total pdfs given in equations A.5.2 and A.5.3 as was done in section A.2. We get,

$$NLL(\vec{\nu}, \vec{\eta}) = NLL_a(\vec{\nu}, \vec{\eta}) + NLL_b(\vec{\nu}, \vec{\eta}) + \sum_{j=1}^2 \frac{(\eta_j - \bar{\eta}_j)^2}{2\sigma_{\eta_j}^2} \quad (\text{A.5.4})$$

$$NLL_a(\vec{\nu}, \vec{\eta}) = \sum_{j=1}^2 \nu_j \eta_j - \sum_{i=1}^N n_i \log \left[\sum_{j=1}^2 \nu_{i,j} \eta_j \right] \quad (\text{A.5.5})$$

$$NLL_b(\vec{\nu}, \vec{\eta}) = \sum_{j=1}^2 \nu_j (1 - \eta_j) - \sum_{i=1}^N n_i \log \left[\sum_{j=1}^2 \nu_{i,j} (1 - \eta_j) \right] \quad (\text{A.5.6})$$

where the parameters ν_j represent the expected number of events for each of the two components, and the n_i are the number of events observed in the i^{th} bin. The parameter of interest for a $0\nu\beta\beta$ -decay search is clearly $\nu_1 = \nu_{0\nu\beta\beta}$.

At this point I will discuss the sensitivity and discovery potential calculations separately.

A.5.1 Sensitivity

As we do not currently have a dataset we proceed using an ensemble method whereby we generate a “fake” dataset based on the expected number of background events. If we then fit this dataset by minimizing the likelihood functions just defined, we can determine an upper limit for the parameter of interest ν_1 . Note also, as described in [94], the proper way to treat uncertainties on parameters in *pseudo-experiments* of this kind is to draw a new mean value for the uncertain parameter (in this case the η_j) with each new fake dataset and use this value for $\bar{\eta}_j$ in the constraint term (equation A.5.4).

By repeating this procedure many times and histogramming each resulting upper limit we can determine the pdf of $\nu_{1,up}$. The median of this distribution will correspond to the sensitivity of the experiment. We will give the following example assuming the

Figure A.5.2: Example of one of the fits to a fake dataset generated to determine the distribution of upper limits. The $0\nu\beta\beta$ -decay pdf has been normalized to the upper limit found for this pseudo-experiment ($s_{up} = 64.7$).

detector has run for five years.

An example of a fit to one of the fake datasets is shown in Figure A.5.2 where the $0\nu\beta\beta$ pdf has been normalized to the resulting 90% upper limit on ν_1 which was found, in this case, to be $\nu_{1,up} = 64.7$. The resulting distribution of upper limits is shown in Figure A.5.3 where the median is observed to be $\tilde{s}_{up} = 63.6$.

A.5.2 Discovery potential

To calculate the discovery potential we need to first define the test statistic we will use. There are many different test statistics possible, however, the test statistic that gives the maximum power for a given significance level is the likelihood ratio[95],

$$t'(x) = \frac{L(\theta|H_0)}{L(\theta|H_1)} \quad (\text{A.5.7})$$

Here, $L(\theta|H_0)$ is the likelihood function under the assumption that the signal does not exist, and $L(\theta|H_1)$ is the likelihood function under the assumption that it does. We will actually use the negative-log-likelihood ratio which is simply the negative logarithm

Figure A.5.3: Distribution of the 90% CL upper limits for the sample calculation of the EXO sensitivity. The median of the distribution represents the sensitivity and is indicated by the dashed line.

of the above. In our case, the likelihood function under H_0 will just be equation A.5.4 with the signal parameter ν_1 fixed to zero. The likelihood function under H_1 is just equation A.5.4. Thus the test statistic we will use is,

$$t(x) = NLL(\vec{\nu}, \vec{\eta}) - NLL(\vec{\nu}, \vec{\eta} | \nu_1 = 0) \quad (\text{A.5.8})$$

We calculate the discovery potential using a similar ensemble method as used for the sensitivity. However, this time, we want the distribution of $t(x)$ instead of the upper limits. Furthermore, we will also need to simulate experiments where we actually generate a signal in order to determine the pdf of $t(x)$ under various alternative hypotheses. Finally, since we are interested in the location of the test statistic such that the “P-value” is less than $\sim 3 \times 10^{-7}$ we will need to simulate a lot more datasets than in the case for sensitivity. A good rule of thumb is to generate at least ten-times more statistics than is needed to achieve the given probability. For discovery, this means we have to generate and fit $\sim 30 \times 10^6$ datasets. The amount of calculation time needed to do this

cannot be underestimated. When I calculated the discovery potential for the planned nEXO experiment it took roughly a week running on 50 computer cores to generate the discovery potential for one assumed background model. For this reason I will illustrate the calculation of the discovery potential using the slightly less demanding constraint of a 4-sigma effect. This means we need to modify the right-hand-side of equation to be $\sim 3 \times 10^{-5}$.

The steps taken to calculate the discovery potential are follows:

1. Determine the pdf of $t(x)$ under the null hypothesis. To do this a fake dataset is generated assuming no signal is present. The dataset is then fit with the likelihood function (equation A.5.4) with the signal parameter fixed to zero to determine $NLL(\vec{\nu}, \vec{\eta} | \nu_1 = 0)$. The same dataset is then fit with the same likelihood function, however, this time with the signal parameter free in the fit. The value of $t(x)$ is then calculated and histogrammed. This process is repeated many times to build the distribution of $t(x)$.
2. Once the pdf of $t(x)$ is known, we have to determine the value t_{cut} such that equation A.4.3 is satisfied (with the corresponding 4-sigma probability in this case).
3. Step 1 is then repeated, this time with a signal component ν_1 in the generated datasets, in order to determine the distributions of $t(x)$ under each alternative hypothesis tested (different values of s).
4. Step 3 is repeated until we find an alternative hypothesis that satisfies equation A.4.4. The corresponding signal for this hypothesis is the least detectable signal and gives the discovery potential of the experiment.

The result of the first two steps is shown in Figure A.5.4 where the cut value was determined to be $t_{cut} = 3.42628 \times 10^6$. The pdfs of $t(x)$ under the assumption of signals ranging from $\nu_1 = 10 \rightarrow 1000$ were then determined and the probability $\beta = \int_{-\infty}^{t_{cut}} g(t | H_i : \theta =$

Figure A.5.4: The distribution of the NLL ratios assuming that no signal exists. The value of the test statistic that satisfies the condition given in equation A.4.3 is indicated by the vertical dashed line. Roughly 3.5 million datasets were generated to produce the above distribution.

$\theta_{lds})dt$ was calculated. This probability is plotted against the corresponding signal in Figure A.5.5. The least-detectable-signal that satisfied equation A.4.4 was determined to be $\nu_{lds} = 1415$ counts. This is not a small number of counts by the standards of $0\nu\beta\beta$ -decay searches but it illustrates the method nicely.

The actual calculation for the EXO experiment is not much different than what was presented here. The main differences lie in the number of backgrounds considered as well as the inclusion of more parameters in the fit to reflect higher levels of uncertainty. The results of these calculations are presented in chapter 3.

Figure A.5.5: The probability $\beta = \int_{-\infty}^{t_{cut}} g(t | H_i : \theta = \theta_{lds}) dt$ as a function of the signal counts assumed. The point along the curve that gives the probability equal to 0.5 is the least-detectable-signal.

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