

Towards the world's first nuclear clock

by

Aaron C. Hellinger

B.S., Northeastern State University, 2013

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AN ABSTRACT OF A DISSERTATION

submitted in partial fulfillment of the  
requirements for the degree

DOCTOR OF PHILOSOPHY

Department of Mechanical and Nuclear Engineering  
Carl R. Ice College of Engineering

KANSAS STATE UNIVERSITY  
Manhattan, Kansas

2022

# Abstract

Since the concept of time was invented, scientists have striven to measure it as accurately as possible. Time-keeping devices have evolved throughout the years from ancient clepsydrae water clocks to the most sophisticated atomic clocks. It has now been theorized that a nuclear clock based on an isomeric transition of  $^{229}\text{Th}$  will become the new time standard with a precision error that decreases from  $10^{-15}$  to  $10^{-20}$  without any improvement upon the accuracy of the measurement of a second [1]. Before a prototype nuclear clock can be created, an exact energy measurement of the lowest known isomeric transition in  $^{229}\text{Th}$ , approximately 8 eV, must be performed. Current laser technology has become advanced enough that the idea of creating a laser with a wavelength near the transition energy, approximately 150 nm, is no longer an unrealistic idea.

This thesis covers the efforts taken to construct an apparatus that collects  $^{229}\text{Th}^{3+}$  ions. A custom-made  $^{229}\text{Th}$  beam line has been designed and constructed that gathers decays from  $^{233}\text{U}$  and initially filters out all negatively charged ions and electrons inside a gas stopping cell that is backfilled with ultra high purity helium gas that is used to control charge exchange as well as cool the ions as they progress forward. These ions are focused into a beam using a 2D radiofrequency (RF) funnel and gathered by an RF quadrupole (RFQ). The newly gathered beam is guided into a mass separation quadrupole that filters out all undesired ions. The new  $^{229}\text{Th}^{3+}$  ion beam is then guided into a linear Paul ion trap for laser interrogation.

While this thesis work does not complete the goal of trapping approximately 1,000,000  $^{229}\text{Th}^{3+}$  ions (explained in Chapter 6), a fraction of the goal has been achieved. As of the time this thesis was written, the optimized ion trap has been able to contain 374,000  $^{229}\text{Th}^{3+}$  ions, which is a factor of 200 times greater than the next largest linear Paul ion trap used for the detection of  $^{229m}\text{Th}$ . Many improvements to the original apparatus have been performed and additional improvements are in the design process.



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Approved by:

Co-Major Professor  
William L. Dunn

Approved by:

Co-Major Professor  
Walter J. McNeil

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# Abstract

Since the concept of time was invented, scientists have striven to measure it as accurately as possible. Time-keeping devices have evolved throughout the years from ancient clepsydrae water clocks to the most sophisticated atomic clocks. It has now been theorized that a nuclear clock based on an isomeric transition of  $^{229}\text{Th}$  will become the new time standard with a precision error that decreases from  $10^{-15}$  to  $10^{-20}$  without any improvement upon the accuracy of the measurement of a second [1]. Before a prototype nuclear clock can be created, an exact energy measurement of the lowest known isomeric transition in  $^{229}\text{Th}$ , approximately 8 eV, must be performed. Current laser technology has become advanced enough that the idea of creating a laser with a wavelength near the transition energy, approximately 150 nm, is no longer an unrealistic idea.

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# Acknowledgments

I would like to thank my advisors and mentors from Kansas State, Dr. Bill Dunn and Dr. Walter McNeil. I cannot express enough gratification that I have for the knowledge and professionalism that you two have passed to me. I will always appreciate the effort that you have taken to mentor me into the scientist I am today. Without either of you, there's no telling where I would be today.

I am very thankful to have been funded by the Consortium for Nonproliferation Enabling Capabilities (CNEC) during my time as a graduate student. Without CNEC, I would not have learned to present research in front of a large crowd nor would I have had the opportunity to intern/work at a national laboratory as a graduate student which would end up shaping my career path.

I owe Dr. Jason Harke (formerly Burke) a great deal of thanks for taking me in as a graduate student at Lawrence Livermore National Lab (LLNL) and teaching me what it is like to work in a national lab environment. My time as an intern at LLNL made me want to become a staff researcher at a national lab. Then I was offered a PhD project to work on which ended up doubling the skills that I had learned while at Kansas State.

Finally, I would like to thank Alexa Stewart. Without her, I do not think I would have finished writing my dissertation. All the long nights of writing while you study for your architectural exams finally paid off!

# Dedication

I dedicate this work to my parents, Cyrena Hellinger and William Varney. Without the endless love from my mother and my father's constant motivational talks, I would have never made it to graduate school to begin with. Thank you so much for always being there for me and motivating me to further myself.

# Chapter 1

## Introduction

According to the Merriam-Webster dictionary [23], time can be defined as "the measured or measurable period during which an action, process, or condition exists or continues." There are many applications for precise timekeeping such as, but not limited to, GPS, telecommunications, and high-speed internet. Increased knowledge of atomic structure and developing laser technology has made it possible to record time with an error of 1 second per 140,000,000 years. Laser technology has only recently advanced enough to allow the direct manipulation of a nucleus for the first time ever. Using non-standard models during the search for dark matter, it has been predicted that fundamental constants such as electron-to-proton mass ratio and the fine structure constant are changing with time [1]. It is now vital to create a clock that will be in the running to redefine the second with a precision of up to  $10^5$  greater than the current standard atomic clocks. This will allow further testing of these fundamental constants.

### 1.1 History of Timekeeping

Timekeeping is a practice that can be traced back to at least 5,000 years ago. In approximately 2000 BC, the Sumerians invented the sexagesimal system for time measurement which is still being used today. Ancient Egyptians divided the day into two 12-hour halves while using large obelisks to track the movement of the sun. Ancient Mesoamericans were the first to invent a

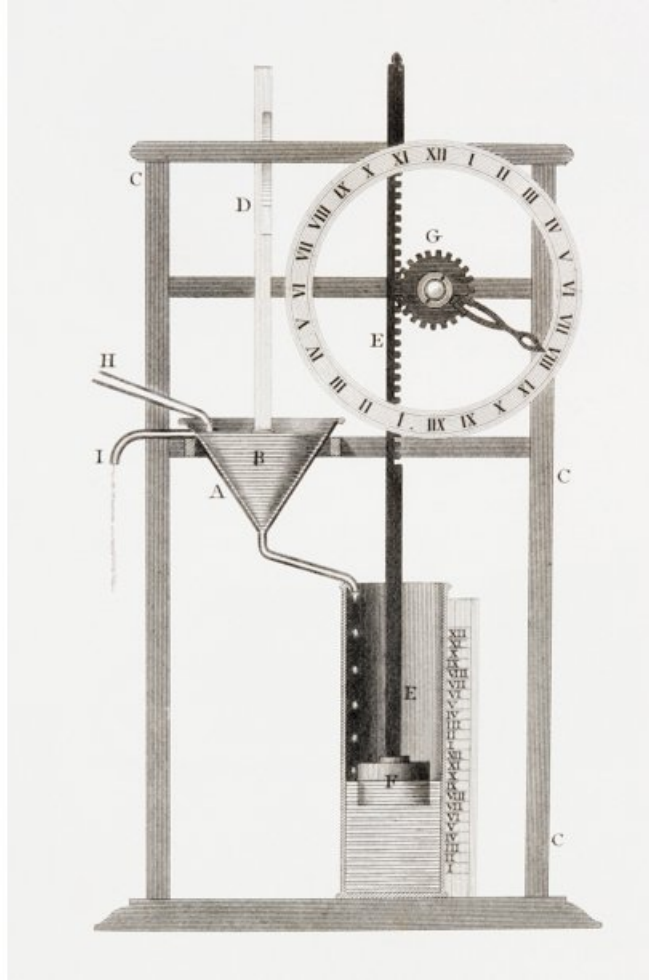


Figure 1.1: Schematic of a clepsydrae. Water drips from the funnel (B) into a chamber (E) where (F) is a very buoyant object that lifts as the water rises to move the gears that changes the clock [2].

calendar that produced a 360-day year. Although not well recorded, Prehistoric European stone circles (such as Stonehenge) are thought to be associated with timekeeping, as well. The most common ancient clock was the clepsydra, or water clock. A schematic of a clepsydrae can be seen in Figure 1.1. These clocks were found in ancient Greece, Rome, Persia, and China. There are two types of water clocks: outflow and inflow. The outflow water clock measures time by draining a container full of water at a constant rate. Markings inside the container designate how much time has passed. The inflow water clock works in the opposite way. A container is filled with water at a constant rate while markings inside the container designate how much time has passed.



Figure 1.2: An ancient sundial on the castle walls of Saint Michael's Mount in Cornwall, England [3].

Since ancient times, there have been different timekeeping devices invented that were not intended to measure time with high precision. These devices include: candle clocks, incense clocks, sundials, hourglasses, and astronomical clocks. Figure 1.2 shows a photograph of an ancient sundial that is located at Saint Michael's Mount in Cornwall, England. It was not until the 1580s, when Galileo first introduced the concept of a pendulum to regulate a clock, that precise timekeeping became compelling to scientists. The first pendulum clock was invented in 1656 by Christiaan Huygens that had an error of less than one minute per day [24]. A picture of a pendulum clock can be seen in Figure 1.3. This clock's error was eventually refined to less than 10 seconds per day. At that time, it was known that temperature played a drastic role in the error. In 1721, George Graham improved the accuracy to 1 second per day by compensating for changes in pendulum length due to temperature variations. The first accurate, non-pendulum based clock was a marine chronometer invented by John Harrison. It was accurate up to one-fifth of a second per day and was used to determine longitude within one-half of a degree after his voyage to the West Indies. This invention opened the way to pocket watches and wristwatches. Other advance-



ments to pendulum technology eventually led to an accuracy of one-hundredth of a second in a day.

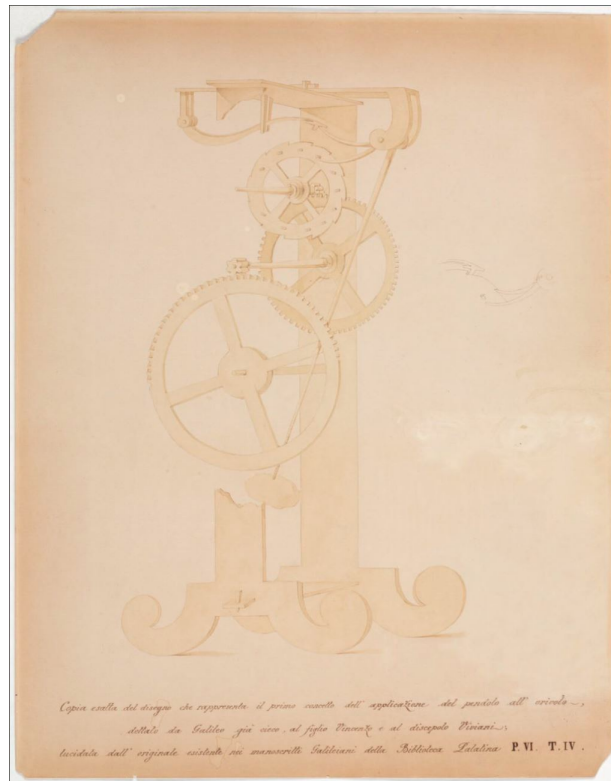


Figure 1.3: An ancient pendulum clock [4].

Pendulum-based clocks became obsolete after the invention of quartz crystal oscillators in 1927. Quartz clocks work by taking advantage of the piezoelectric properties of quartz. When a charge is placed on a quartz crystal, the crystal will bend and vibrate at a specific frequency. However, this frequency is size and temperature dependent. The age of the crystal also produces frequency shifts. Therefore, no two quartz clocks are exactly the same. Precise crystal growth has allowed most modern quartz clocks to vibrate at a frequency of 32,768 Hz, which gives an accuracy of about 15 seconds in 30 days. Quartz clocks were the standard for timekeeping up until the ultra-precise atomic clock was invented.

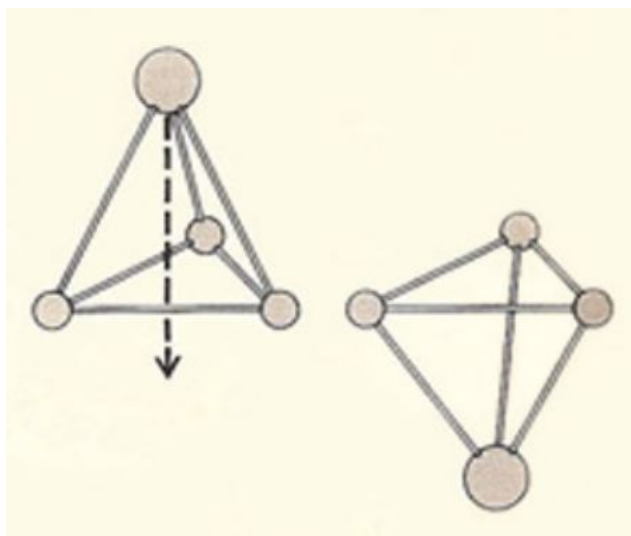


Figure 1.4: 3D structure of an ammonia molecule. The larger circle is the nitrogen atom while the smaller three circles are hydrogen atoms. Note the nitrogen displacement through the base of the pyramidal structure. This is how an ammonia molecule oscillates [5].

### 1.1.1 The First Atomic Clock

Lord Kelvin was the first person to predict that atoms could be used to create a new time and frequency standard [7]. In order to realize this prediction, a large frame of research in microwave spectroscopy of atoms and molecules was needed, as well as reliable electronics for microwave measurements. By the late 1940s, the vision of an atomic frequency had become achievable. In 1948, Harold Lyons began work on a frequency standard at the National Bureau of Standards (NBS) that used an ammonia absorption line at 23,870.1 MHz. The development was finalized and made public in January of 1949. Lyons used the following to describe the atomic clock: "The 'pendulums' which regulate them are the vibrating parts of atoms or molecules. So steady are these oscillations that atomic clocks keep better time than the spinning earth itself" [25]. The ammonia molecule exists in a pyramidal structure with the three hydrogen atoms in the shape of a triangular base with the nitrogen atom at the apex. When enough energy excites the ammonia, the nitrogen is moved through the base of the pyramidal structure, and forms another apex at the opposite side, as shown in Figure 1.4. It then proceeds to oscillate between these two positions at a frequency of 23,870.1 MHz.

The ammonia clock works as follows: a quartz crystal's frequency is fine-tuned to match the

resonant frequency of the ammonia molecule with the help of an oscillator circuit. These oscillations are then converted into radio waves by a small antenna and are fed into a long chamber that contains ammonia gas. If the radio waves are oscillating at the same frequency as an ammonia molecule, then most of the radio energy will be absorbed by that molecule and recorded. However, if the radio waves are not oscillating on the resonant frequency of an ammonia molecule, then the radio waves will pass through the chamber without being absorbed and enter a feedback circuit that adjusts the frequency of the oscillator circuit. This type of clock has two major flaws. Because the gas is freely moving, molecules will collide with other molecules as well as the walls of the chamber. This creates a broadening of the resonant frequency of the ammonia. The Doppler effect is also observed. When the ammonia molecules are traveling away from the antenna that radiates the radio waves, the radio waves see the frequency as slightly lower than it really is. The opposite is also true. When the ammonia is traveling towards the antenna, the frequency is higher. Both the collisions and Doppler effect limit the possible precision of this design. After realizing the flaws of the design, the error per second of the first version of the atomic clock decreased to  $1.00 \times 10^{-8}$ . This equates to an error of approximately 1 second in every 1,157 days (3.17 years).

### 1.1.2 Current Time Standard

There were three major design improvements that were made from the first prototype. The first two were discussed in the previous section: eliminating collisions and reducing the Doppler effect. Because resonant frequencies follow a Gaussian distribution, another improvement that was made was to find an atom/molecule that had a much narrower resonant frequency. All of these improvements were accomplished by Louis Essen in 1955 at the National Physical Laboratory (NPL) in the United Kingdom. This design utilized a continuous, uni-directional flow of atoms to eliminate collisional effects and reduce Doppler effects. A new isotope was also introduced,  $^{133}\text{Cs}$ , which has a narrower resonant frequency than ammonia.  $^{133}\text{Cs}$  has a frequency of 9.192 GHz, in comparison to ammonia's 23,870.1 MHz (23.870 GHz). The mechanism that "oscillates" is different than that of an ammonia molecule. In  $^{133}\text{Cs}$ , the outer electron shell is populated by

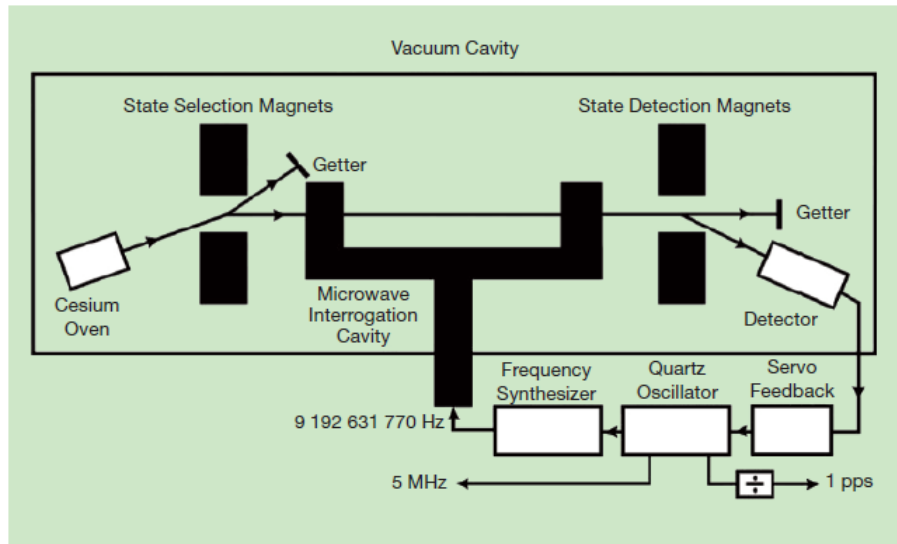


Figure 1.5: Schematic of the first atomic clocks [6].

only a single electron. When cesium atoms enter an electromagnetic field with a frequency of 9.192 GHz, the outer electron absorbs the energy from the electric field and flips orientation.

The process that the first cesium atomic clock used is as follows: the cesium was heated to a gaseous state inside an electric furnace. The atoms were then discharged and focused into a beam (which eliminates collisional and Doppler effects). The beam was directed into a magnetic field which selects only certain ions to pass. Next, the beam entered an electromagnetic field whose frequency is 9.192 GHz. Finally, it passed by a second magnet which deflected the beam into a receiver. If the frequency of the electromagnetic field was different than the  $^{133}\text{Cs}$  resonant frequency, then the ions will collide with the magnet and the current at the receiver was minimal. The receiver acts as a frequency check, and a circuit was added that will adjust the electric field until the current at the receiver was at a maximum. A visualization of the atomic clock process can be seen in Figure 1.5. This NPL design was accurate to one second in one billion seconds, or an error of one second in every 31 years, which was the current record at that time. A photograph of the first cesium atomic clock that was used as a time standard, NBS-1, can be seen in Figure 1.6.

Before 1967, the definition of a second had an astronomical basis, or, 1/86,400 of the time that it takes the Earth to rotate once on its axis. There are many errors to take into account, from the wobble of Earth's rotation to the slowing of Earth's rotation due to tidal forces from the moon.

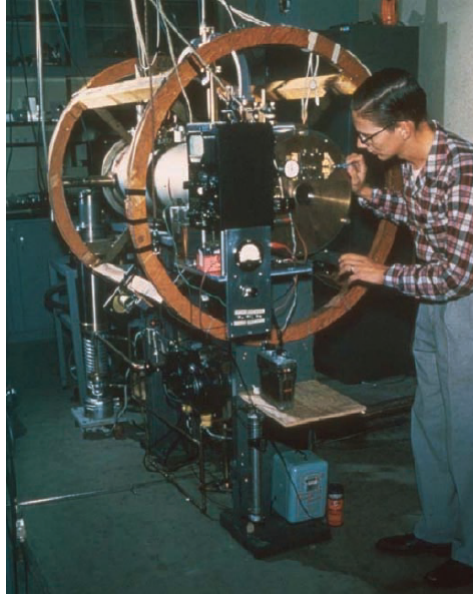


Figure 1.6: NBS-1, the first atomic clock that was used as a time standard in the United States [7].

Therefore, in 1967, the 13th General Conference on Weights and Measures redefined the second as "9,192,631,770 periods of the radiation of the ground state hyperfine transition in cesium" [6]. By that time, the standard cesium clocks had an error of 1 in 50 billion, or an error of 1 second in every 1,585 years. In 1997, an amendment was added to the definition due to calculations made by Wayne Itano in the early 1980s. It was shown that the resonant frequency for  $^{133}\text{Cs}$  can be manipulated by blackbody radiation. This is an important note and is a large reason for the topic of this thesis, which will be discussed later. Thus, a "perfect" realization of the SI second would require no present magnetic/electric disturbances while the cesium atoms are at rest.

### 1.1.3 Improvements to the Cesium Standard

Seven different cesium beam devices served as U.S. national primary frequency standards at NBS between the years 1959-1998, which later became the National Institute of Standards and Technology (NIST) in 1988. Each device consists of lasers that directed the thermal cesium beam through microwave cavities. These devices and their accuracies will be briefly described. NBS-1 was created in 1948 and was the first cesium clock ever created. Improvements were made on the original design until 1959, which the error was reported as 1 second every 3,170 years. NBS-1

standard was replaced by NBS-2 on January 1, 1960. The length of the microwave cavity was increased by a factor of three, which reduced the error in the resonant frequency of  $^{133}\text{Cs}$ . This resulted in a time error of 1 second every 3,963 years. During this time, work on NBS-3 had already begun, increasing the length of the microwave cavity in an attempt to increase the number of excited cesium atoms, therefore, increasing the precision of the device. In 1969, NBS-3 had an error of 1 second every 63,419 years. NBS-4 was created as a joint project between NBS and Hewlett-Packard and not intended to become a frequency standard because its cavity was much shorter than the other NBS beam lines. However, it served in conjunction with NBS-5 and NBS-6 as a comparison. NBS-4 had a reported error of 1 second every 105,699 years. NBS-5 and NBS-6 are the same beam line but with extensive modifications. NBS-5 became the frequency standard in 1973 with a microwave cavity about 8 cm longer than NBS-3 and about 6 meters long in total. The reported error for NBS-5 is 1 second in every 158,549 years. Modifications to NBS-5 included reducing the velocity of the beam further, reducing low-velocity collisional scattering further, and filtering of high velocity atoms. These modifications allowed NBS-6, which became the standard in 1975, to achieve an error of 1 second every 396,372 years. NIST-7 (created after NBS changed to NIST), was fundamentally different than the NBS clocks. New, narrow-linewidth lasers were available for use which replaced the magnets and detection system. This was beneficial for the following: while using magnets worked well, separating out the undesired energy states, this left only a limited number of ions to pass through the beam. Lasers, however, can optically excite as many atoms as possible, more quickly, which generates a much stronger signal and increases statistics. In 1993, NIST-7 was completed. The length of the cavity region was shortened, as well as the entire apparatus, yet the accuracy was nearly 20 times higher than NBS-6. NIST-7 was accurate to 1 second in 6,341,958 years.

After NIST-7, a new type of design, the cesium fountain, became the frequency standard. In the 1950s, Jerrold Zacharias introduced a concept of building a vertical cesium beam line that allowed gravity to slow the atoms. Using this design, the atoms would pass through the microwave cavity twice, once while traveling upwards, letting gravity slow the atoms down and eventually reverse their path, and then pass through the microwave cavity again while traveling downwards. This would increase the exposure of the cesium atoms to the microwave cavity by a full second,

which narrows the error on the resonant frequency. Unfortunately, his initial design never saw a signal. It was predicted that this was due to a collision problem between the cesium atoms. The slow atoms after the gravitational fall were being scattered out of the beam by the fast ions travelling upwards. It wasn't until laser cooling was introduced that the cesium fountain became viable. The NIST-F1 cesium fountain was operational in 1998.

A schematic of the cesium fountain can be viewed in Figure 1.8 and works as follows: a sample of cesium ions are laser cooled at the intersection of six laser beams. The cloud of cesium ions is then accelerated upward by detuning the cooling lasers so that the ions travel with the laser. The cloud eventually enters the microwave cavity for the first time and excites a majority of the cesium atoms. The cesium atoms then exit the cavity, allow gravitational forces to reverse the trajectories, and re-enter the cavity around one second later which excites another large number of atoms. After exiting the cavity, the newly excited atoms are detected using a probe laser tuned to the 9.192 GHz transition. The increased interaction time between the microwave cavity and cesium beam allowed the NIST-F1 cesium fountain to reach an accuracy of approximately  $4 \times 10^{-16}$ , or an error of 1 second every 79,274,480 years. The limiting factors of NIST-F1 were narrowed down to the interactions between the thermal radiation emitted by the walls of the vacuum chamber and cesium atoms as well as a density shift. The density shift is caused by cesium collisions during the initial launch of the beam. NIST-F2 was proposed in order to eradicate these problems. NIST-F2 was not fully operational until April 2014, 16 years after NIST-F1. The key difference between NIST-F1 and NIST-F2 is that NIST-F2 operates in cryogenic temperatures which eliminates the thermal radiation problem of NIST-F1. This change improved the error of the cesium fountain to 1 second in every 198 million years. NIST-F2 remains the frequency standard to this day. A table showcasing how the cesium atomic clocks improved over time can be seen in Table 1.1. The cesium atomic clocks constructed by NBS/NIST can be viewed in Figure 1.7. Other atomic clocks, such as the mercury-ion clock and maser clocks, exist but are not described in this thesis.

Table 1.1: NIST Cesium Standards and Associated Accuracies [6]

| Cesium Frequency Standard | Years as Standard | Resonant Frequency Width | Length of Microwave Cavity | Best Published Accuracy    | Seconds Lost per Year |
|---------------------------|-------------------|--------------------------|----------------------------|----------------------------|-----------------------|
| NBS-1                     | 1959-1960         | 300 Hz                   | 55 cm                      | $10^{-11}$                 | $3.5 \times 10^{-4}$  |
| NBS-2                     | 1960-1963         | 110 Hz                   | 164 cm                     | $8 \times 10^{-12}$        | $2.5 \times 10^{-4}$  |
| NBS-3                     | 1963-1970         | 48 Hz                    | 366 cm                     | $5 \times 10^{-13}$        | $1.6 \times 10^{-5}$  |
| NBS-4                     | NA*               | 130 Hz                   | 52.4 cm                    | $3 \times 10^{-13}$        | $9.5 \times 10^{-6}$  |
| NBS-5                     | 1972-1974         | 45 Hz                    | 374 cm                     | $2 \times 10^{-13}$        | $6.3 \times 10^{-6}$  |
| NBS-6                     | 1975-1993         | 26 Hz                    | 374 cm                     | $8 \times 10^{-14}$        | $2.5 \times 10^{-6}$  |
| NIST-7                    | 1993-1998         | 62 Hz                    | 155 cm                     | $5 \times 10^{-15}$        | $1.6 \times 10^{-7}$  |
| NIST-F1                   | 1998-present      | 1 Hz                     | 2.18 cm[26]                | $3.1 \times 10^{-16}$ [26] | $9.8 \times 10^{-9}$  |
| NIST-F2                   | 2014-present      | < 1 Hz                   | Not Reported               | $1.6 \times 10^{-16}$ [27] | $5.4 \times 10^{-9}$  |

\*NBS-4 never served as a time-standard.



## 1.2 Potential Applications for $^{229m}\text{Th}$

Using  $^{229m}\text{Th}$  as a new frequency standard has a number of applications. A few of those applications will be described in this section. The most important application is designing a nuclear clock that is predicted to have an increased accuracy of  $10^5$  over even the most accurate atomic clocks. Because some fundamental constants' consistency have undergone scrutiny over the last few decades, having a nuclear clock with greater accuracy, and precision, would mean the fundamental constants can undergo further study. Being able to manipulate an atom on the nuclear level also has other advantages such as a possible superior qubit used in quantum computing that can be created using direct excitation of a nucleus.

### 1.2.1 Nuclear Clock

The concept of a nuclear clock based on  $^{229m}\text{Th}$  was first proposed by Peik and Tamm in 2003 [28]. This thorium excited state has been predicted to have a lifetime of 2-4 hours, the linewidth of the nuclear transition should be on the order of 10  $\mu\text{Hz}$ , and its frequency is independent to external perturbations which is a large focus of Peik and Tamm. This, along with its extremely low nuclear state (5-9 eV), make it a prime candidate for a nuclear clock. As with cesium fountains, broadening and shifts of the resonant frequency can be reduced by cooling and trapping the ions. The exact energy of the isomer still eludes scientists. Once this energy is sufficiently evaluated, a nuclear clock can be created, which will work similarly to the cesium fountain standards: certain ions will be selected from a beam, cooled, directly excited by lasers, and then photons from the de-excitation will be detected. The frequency of the laser will be changed until the maximum number of photons from de-excitation are detected.

### 1.2.2 Temporal Variations of Fundamental Constants

Some scientists believe that some fundamental constants aren't so "constant". One of the first examples dates back to 1982 when Bekenstein questioned the constancy of the fine structure constant,  $\alpha$  [29]. It was shown that the fractional rate of change  $\dot{\alpha}/\alpha$  in a cosmological setting can

be compatible with astronomical constraints, therefore, are too insensitive to rule out variability. Bekenstein concluded that neither current astronomical constraints nor the state-of-the-art clocks at that time were sensitive enough to rule out variability. [30] and [31] provide further evidence for time fluctuations in the fine structure constant. Prestage's [32] work on atomic clocks while looking for these variations is a large influence on the topic of this thesis. Peik [33] also offers knowledge based on atomic clocks and using them to search for these temporal variations.

### 1.2.3 Other Applications

Another potential application of manipulating a nucleus at will would be the invention of a superior qubit for quantum computing. The term "qubit" stands for "quantum bit". Qubits have an advantage over regular bits in a computer because of the following: in computing, bits can only take the value of 0 or 1 to store information while qubits can take the value of 0 or 1 as well as a superposition of both. This means that a qubit can be described as a linear combination of 0 and 1, i.e.,

$$|\psi\rangle = \alpha|0\rangle + \beta|1\rangle \quad (1.1)$$

where  $\alpha$  and  $\beta$  are probabilities that can be described as complex numbers. Because the absolute squares of  $\alpha$  and  $\beta$  are probabilities, it follows that the probabilities must be constrained by the equation,

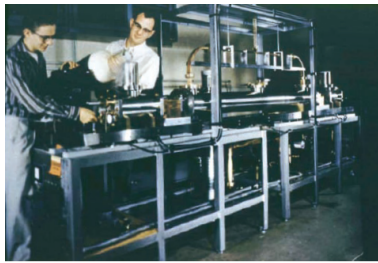
$$|\alpha|^2 + |\beta|^2 = 1. \quad (1.2)$$

Qubits also display quantum entanglement, which allows a transfer of information instantaneously. For example, consider you have two qubits that are miles apart. If one qubit is measured to have a state of  $|0\rangle$ , then the second qubit, if entangled, will display the exact same state.

### 1.2.4 Thesis Goals

The overall goal for this thesis is to create a custom linear  $^{229}\text{Th}^{3+}$  ion guide that mass separates and a custom linear Paul ion trap that traps at least  $10^6$  ions for laser interrogation (direct laser excitation). Initially, the desire is to detect  $^{229m}\text{Th}$  without the use of lasers. After correcting

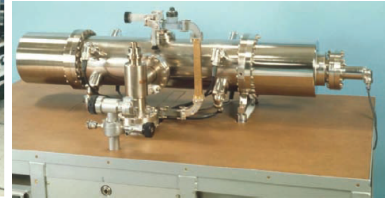
for the 5% solid angle, 10% detection efficiency of UV photons, and 2% branching ratio of  $^{233}\text{U}$  decaying to  $^{229m}\text{Th}$ , the MCP response would be approximately 100 counts per second. The MCP background count rate is approximately 10 counts per second. This would make the detection of the metastable state easily distinguishable from the noise from the MCP.



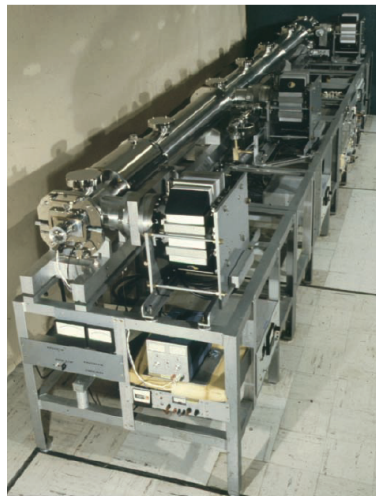
(a) NBS-2



(b) NBS-3



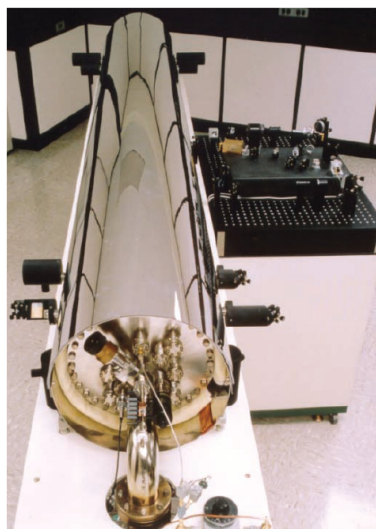
(c) NBS-4



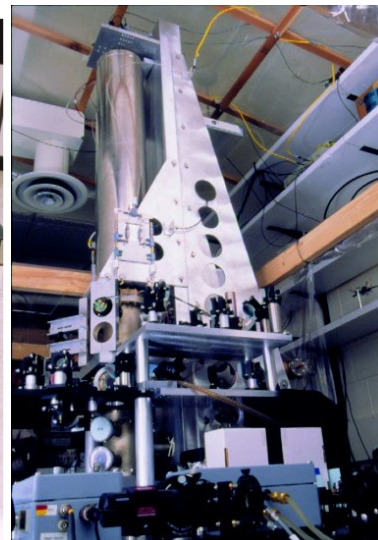
(d) NBS-5



(e) NBS-6



(f) NIST-7



(g) NIST-F1

Figure 1.7: The different cesium clocks at NIST (formerly NBS) [7].

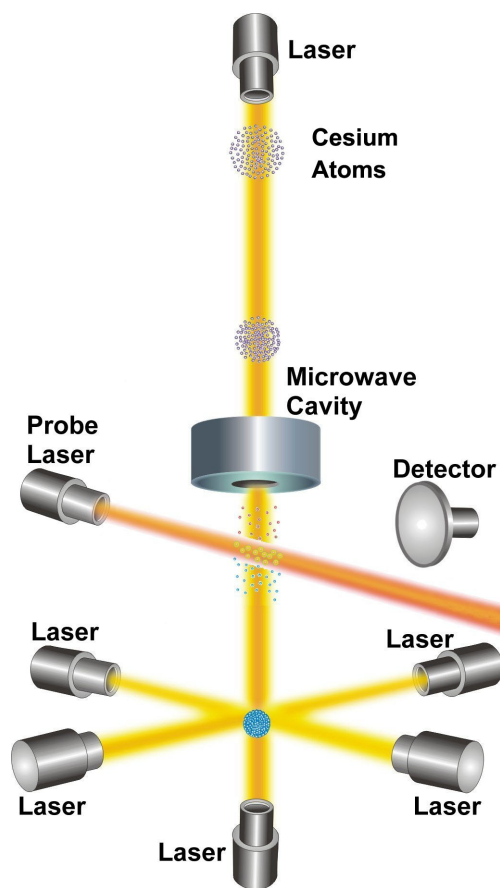


Figure 1.8: Schematic of how the NIST-F1/F2 cesium fountains works [6].

# Chapter 2

## History of $^{229m}\text{Th}$

In this chapter, previous theoretical and experimental investigations on  $^{229}\text{Th}$  will be described. First, how the lowest energy state of  $^{229}\text{Th}$  was discovered will be considered. Early progress on accuracy measurements and further proof of existence will follow. Then the first highly accepted value of the transition will be discussed along with recent progress. Other notable experiments assisting to the discovery of the physics behind  $^{229m}\text{Th}$  will be explained at the end of this chapter.

### 2.1 The Discovery and First Measurement of $^{229m}\text{Th}$

The existence of an indistinct, low-energy nuclear state in  $^{229}\text{Th}$  was first proposed by Kroger and Reich in 1976 [8]. During this study, they performed precise spectroscopy of the low-energy  $\gamma$ -rays of  $^{229}\text{Th}$  from the  $\alpha$  decay of  $^{233}\text{U}$ . A group of low-energy nuclear levels had already been identified previously, although their spin and parity were unknown. The group of four energy levels that needed analyzing were 29.16, 71.82, 125.45, and 195.76 keV. Initially, it was thought that these levels existed on the ground-state rotational band of  $K^\pi = 5/2^+$ . The conjectured  $K^\pi = 5/2^+$  was later rejected then proposed and accepted to be  $K^\pi = 3/2^+$ . The low-energy level scheme as depicted in [8] is shown in Figure 2.1. Kroger and Reich were then able to show that the 29.16 keV energy state had a spin of  $5/2^+$ , but were unable to find the missing band head. It was concluded, with indirect evidence, that this metastable state had a spin of  $3/2^+$  and was less

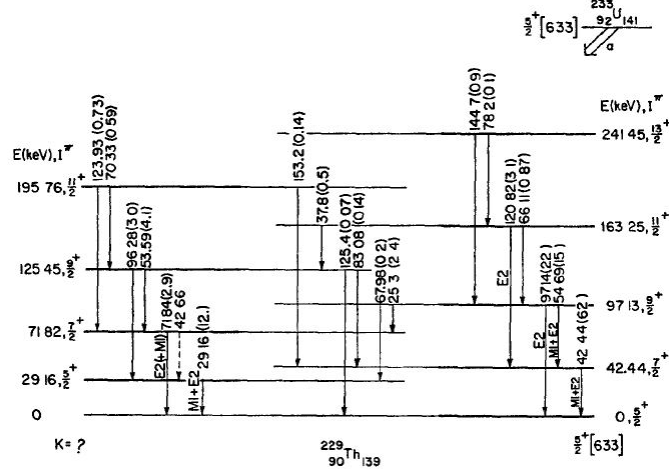


Figure 2.1: Low-level scheme of  $^{229}\text{Th}$  taken from [8] that shows the two lowest-lying positive-parity bands with their associated  $\gamma$ -ray transitions. The left side shows an unknown K-value which has been accepted as being  $\frac{3}{2}^+$  [631].

than 100 eV above the ground state. This energy state became what we know today as the lowest known nuclear state,  $^{229m}\text{Th}$ .

## 2.2 Further Evidence of Existence

In 1990, Reich and Helmer [9] presented a re-determination of their previous attempts to measure this energy state using their old data from 1976. The  $3/2^+$  band head energy was determined indirectly by using energy states that are well-known. The following three energy combinations were used to determine the band head energy:

$$\Delta_1 = E(97) - E(71) - E(25),$$

$$\Delta_2 = E(97) - E(67) - E(29),$$

$$\begin{aligned} \Delta_3 &= [E(148) - E(118)] - [E(146) - E(117)] \\ &= [E(148) - E(146)] - [E(118) - E(117)]. \end{aligned}$$

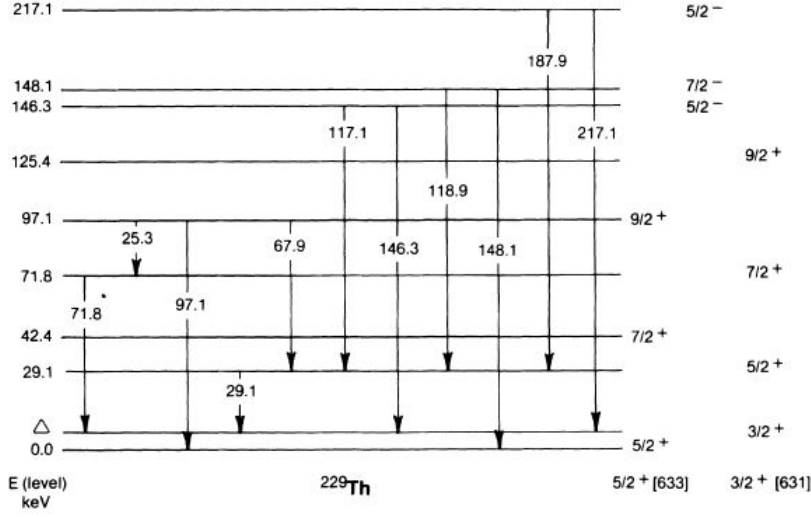


Figure 2.2: Updated version of Figure 2.1. Shown is the  $\frac{3}{2}^{+}$  band head as well as the  $\frac{5}{2}^{+}$  band head with their associated transitions used in determining the energy separation that was used to indirectly determine the transition energy of  $^{229m}\text{Th}$  [9].

The following equation is used to check the quality of the measurement:

$$0 = E(217) - E(187) - E(29).$$

Reich and Helmer found the three  $\Delta$  equations and check equation to be:

$$\Delta_1 = -0.003 \pm 0.005 \text{ keV},$$

$$\Delta_2 = -0.001 \pm 0.006 \text{ keV},$$

$$\Delta_3 = 0.002 \pm 0.008 \text{ keV},$$

$$- 0.002 \pm 0.003 \text{ keV}.$$

The first estimate of the 3/2+ band head was then shown to be

$$\Delta = -0.001 \pm 0.004 \text{ keV}.$$

After Reich and Helmer [11] indirectly estimated the energy of the band head, Burke et al. [10] provided additional support to the existence of the excited state using the  $^{230}\text{Th}(\text{d},\text{t})^{229}\text{Th}$



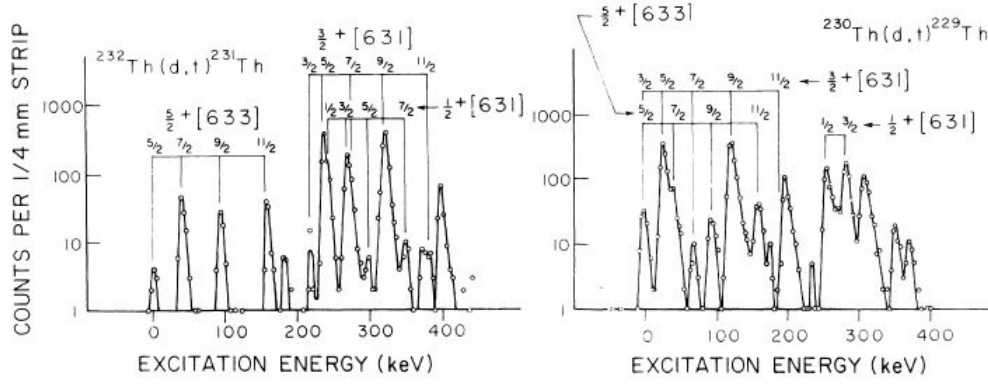


Figure 2.3: Triton spectra for the  $^{230}\text{Th}(d,t)^{229}\text{Th}$  (right) and  $^{232}\text{Th}(d,t)^{231}\text{Th}$  (left) reactions at an angle of  $\theta = 60^\circ$ . Rotational bands are grouped together with their corresponding Nilsson numbers. Taken from [10].

reaction. This experiment measured triton spectra at 17 different angles between  $5^\circ$  and  $80^\circ$  using a 17 MeV deuteron beam at the McMaster University Tandem Accelerator. Figure 2.3 shows the low energy triton spectra for an angle of  $\theta = 60^\circ$  for two different reactions,  $^{230}\text{Th}(d,t)^{229}\text{Th}$  and  $^{232}\text{Th}(d,t)^{231}\text{Th}$ . The energy resolution of the detector used was reported to be between 5.5-6.0 keV FWHM for the  $^{231}\text{Th}$  spectra and 7 keV FWHM for the  $^{229}\text{Th}$  spectra. The higher resolution was believed to be due to the greater target thickness (approximately 30 mg/cm<sup>2</sup> for the  $^{232}\text{Th}$  target and 40 mg/cm<sup>2</sup> for the  $^{230}\text{Th}$  target). The peaks in the  $^{229}\text{Th}$  plot agree within  $\pm 1$  keV with those of previously known levels up to several hundred keV. This provides assurance that the peaks are correctly associated with known energy levels.

There is a possibility that the proposed  $3/2^+$  level was misinterpreted. Kroger and Reich [8] considered that the levels directly above the lowest transition could be members of a  $5/2$  or different  $3/2$  band. The possibility of a  $3/2$  band starting at 29 keV is very unlikely due to the fact that the 125 keV level would have  $I^\pi=7/2^+$ . The peak for this level was very large, however, there is very little  $g_{7/2}$  strength expected. The results presented in [10] rule out all the possibilities that these levels might be a  $K^\pi=5/2^+$  band. Kroger and Reich in [8] also argue against a different  $3/2^+$  band.

The most important evidence that had been discovered is that the distributions of the (d,t) reaction strengths are virtually identical between  $^{229}\text{Th}$  and  $^{232}\text{Th}$ . Thus, the results in [10] give

| Detector | Material | Shape   | Volume<br>(cm <sup>3</sup> ) | Diam.<br>(mm) | Depth<br>(mm) | FWHM (eV) at |         |
|----------|----------|---------|------------------------------|---------------|---------------|--------------|---------|
|          |          |         |                              |               |               | 29 keV       | 122 keV |
| OR-12    | Ge       | coaxial | 114                          |               |               |              | 950     |
| PG-21    | Ge       | coaxial | 63                           |               |               |              | 900     |
| PG-16    | Ge       | planar  |                              | 16            | 10            | 390          |         |
| OR-16    | Ge       | planar  |                              | 10            | 7             | 270          |         |
| Si       | Si(Li)   | planar  |                              | 6             | 4             | 345          |         |

Figure 2.4: Detector specifications used in Helmer and Reich’s second experiment for determining the energy of  $^{229m}\text{Th}$  [11].

strong independent support for the assigned band structures and indirect evidence of a very low-energy nuclear transition in  $^{229}\text{Th}$ . However, this experiment was unable to provide a precise energy determination.

## 2.3 Improving Precision of Band Head Determination

The first experiment dedicated to determining the energy of  $^{229m}\text{Th}$  came from Helmer and Reich in 1994 [11]. The difference between their first experiment and this one is that they ”...considered more  $\gamma$  rays in  $^{229}\text{Th}$ , used more well-measured energy calibration and reference lines, used more detectors, used detectors with better low-energy resolution, more closely matched the counting rates in the  $\gamma$ -ray peaks whose relative energy is measured, and specifically considered certain systematic errors”. From this, they determined the energy level to be  $3.5 \pm 1.0$  eV. The following paragraph will briefly describe their experiment and how it differed from the previous experiment.

Their previous experiment had utilized only one Ge detector. The new experiment used 5 detectors in total, two high-purity coaxial Ge detectors, two planar Ge detectors, and a planar Si(Li) detector. The specifications of each detector are offered in Figure 2.4. Many calibration sources were used which include  $^{172}\text{Hf}$ ,  $^{172}\text{Lu}$ ,  $^{161}\text{Tb}$ ,  $^{241}\text{Am}$ ,  $^{182}\text{Ta}$ ,  $^{169}\text{Yb}$ ,  $^{152}\text{Eu}$ ,  $^{210}\text{Pb}$ , and  $^{170}\text{Tm}$ . Some gamma rays from  $^{241}\text{Am}$ ,  $^{172}\text{Hf}$ , and  $^{172}\text{Lu}$  were also measured during this experiment. Activity of sources used were chosen by attempting to match the number of events for  $\gamma$  rays of similar energies in order to reduce systematic errors due to peak shifts. Each source was created by depositing a solution containing the radionuclide onto a backing of polyimide film and then covering it after drying. The gamma rays considered for the energy determination are shown in

the following equations, similar to [8]:

$$\begin{aligned}\Delta_1 &= E(97.1) - E(71.8) - E(25.3), \\ \Delta_2 &= E(97.1) - E(67.9) - E(29.1), \\ \Delta_3 &= [E(148.1) - E(118.9)] - [E(146.3) - E(117.1)] \\ &= [E(148.1) - E(76.4)] - [E(146.3) - E(74.6)].\end{aligned}$$

which also include the two cascade crossover combinations:

$$\begin{aligned}217.1 &= 187.9 + 29.1, \\ 164.5 &= 135.3 + 29.1.\end{aligned}$$

There were multiple measurements made for each energy difference, for a total of 111 unique measurements. Finally, the peak fitting was done on an interactive system. For even greater detail of the experiment, including uncertainty analysis, measurement and analysis methods, and full range of results please see [11].

The first widely accepted value for this energy state comes in 2007 by Beck et al. [12]. The x-ray spectrometer (XRS) at the National Aeronautics and Space Administration (NASA) was used for this experiment. The XRS consists of 36 microcalorimeters, but only 28 HgTe pixels and 4 Bi pixels were active. Together, they have an energy resolution that is able to resolve the two specific doublets that the Ge detectors from previous experiments failed to resolve. These doublets are how the energy of  $^{229m}\text{Th}$  will be indirectly determined. The energy can be determined using the relationship

$$29.39 + 42.43 \text{ keV} = 42.63 + 29.18 + E(^{229m}\text{Th}) = 71.8 \text{ keV} \quad (2.1)$$

or

$$E(^{229m}\text{Th}) = \Delta E_{29} - \Delta E_{42} \quad (2.2)$$

where  $\Delta E_{29} = 29.39 - 29.18$  and  $\Delta E_{42} = 42.63 - 42.43$  keV and are the specific doublets that were previously unable to be resolved. The XRS has an approximate energy resolution of 6 eV

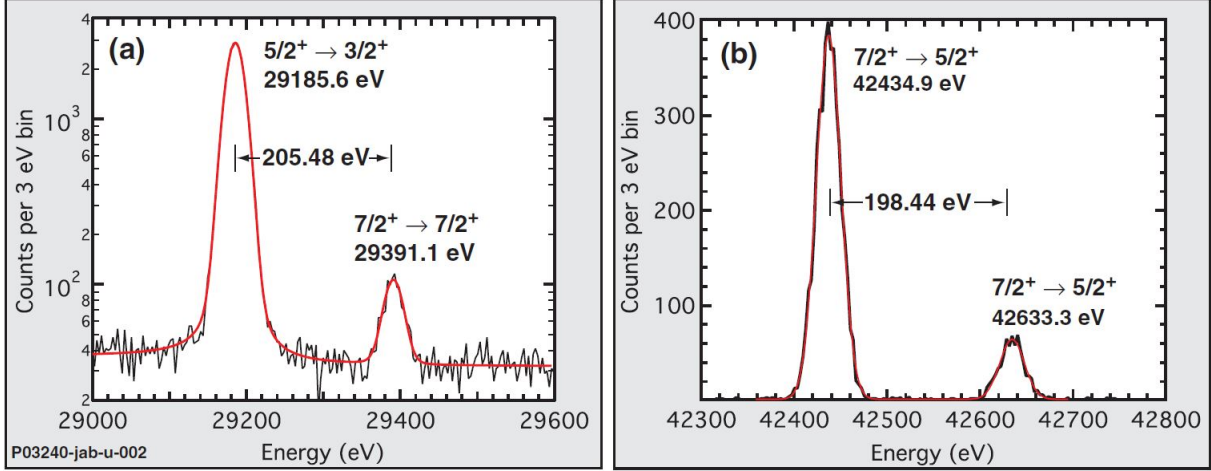


Figure 2.5: Data using the XRS showing the 29 keV doublet and 42 keV doublet. The 29 keV doublet consists of data from the sum of 11 data sets and 25 pixels while data from the 42 keV doublet is from the sum of 11 data sets using only pixel 0. The red line represents least-square fitting [12].

when cooled. When measuring higher energy photons (0.1 - 10 keV), the operating temperature was raised to 90 mK which changed the resolution to approximately 26 eV. The experiment counted  $^{229}\text{Th}$  photons after the alpha decay of  $^{233}\text{U}$ , which was placed inside the detector array for maximum solid angle. Figure 2.5 shows the results of the XRS experiment. From this data, it was determined that  $\Delta E_{29} - \Delta E_{42} = 7.0 \pm 0.5$  eV. However, a small correction is needed to account for the interband branching of the 29.19 keV state to the ground state. It was determined that the energy of  $^{229m}\text{Th}$  was  $7.6 \pm 0.5$  eV.

## 2.4 Recent Accepted Values

Wense et. al. in 2016 reported the first direct detection of the  $^{229}\text{Th}$  transition [34]. Taking advantage of the 6.31 eV ionization potential of  $^{229}\text{Th}$ , 1000  $^{229}\text{Th}^{2+}$  and  $^{229}\text{Th}^{3+}$  charge states per second were loaded onto a multi-channel plate (MCP) detector (model BOS-75-FO Beam Imaging Solutions) using a quadrupole ion guide and quadrupole mass filter. These charge states are created using an ultra-pure helium buffer gas that cools the ions and gives electrons to the  $^{229}\text{Th}$  atoms. Once the ions strike the MCP grid, they will neutralize. Because internal conversion is forbidden in higher charge states (see 3.2.2), once the ions are neutralized, internal conversion

will finally occur. With a 2% decay branch of  $^{233}\text{U}$  resulting in the  $^{229\text{m}}\text{Th}$  isomer [35], along with an approximate MCP efficiency of 1.5% for low energy electrons, only approximately 0.3 counts per second were hypothesized. Considerable effort went into proving that the signal was  $^{229\text{m}}\text{Th}$  and not an atomic signal or a different ion completely. Please refer to [34] for a description of these efforts. It was deduced that  $^{229\text{m}}\text{Th}$  must be constrained between 6.3 eV and 18.3 eV, which are the ionization potentials for the first charge state and third charge state, respectively.

Seiferle et al. in 2019 [36] claimed to have the first ever direct measurement of  $^{229\text{m}}\text{Th}$ . This value,  $8.28 \pm 0.17$  eV was found using the internal conversion decay of  $^{229\text{m}}\text{Th}$ . During internal conversion, the nuclear energy is transferred to the electron shell and then an electron is emitted. This electron's kinetic energy can then be measured. The experimental setup utilized the 2% decay branch in the  $\alpha$ -decay of  $^{233}\text{U}$  to bunch  $^{229\text{m}}\text{Th}$  isomers. Bunches contained approximately 400  $^{229}\text{Th}^{3+}$  ions.  $^{229}\text{Th}^{3+}$  are used due to internal conversion being impossible at the 3+ charge (see 3.2.2). The bunched  $^{229}\text{Th}^{3+}$  ions are guided toward two layers of graphene which neutralize the ions as they pass through. As soon as the charged thorium atoms neutralize, the internal conversion channel opens up, and the excited thorium atom decays within microseconds. The electrons emitted during internal conversion are then measured using a magnetic bottle-type retarding field electron spectrometer. An electrode is placed immediately after the graphene in order to attract stray electrons created during the interaction process between the charged thorium and the graphene. Description of statistical analysis can be found in [36].

In May 2020, Sikorsky et al. [37] published a magnetic micro-calorimeter experiment that determined the  $^{229\text{m}}\text{Th}$  isomer energy to be  $8.10 \pm 0.17$  eV. A 74 MBq (2,000  $\mu\text{Ci}$ )  $^{233}\text{U}$  aqueous solution was used that contained 2mm shields that stopped transmission of  $\alpha$  and  $\beta$  particles but allowed transmission of  $\gamma$  rays. The magnetic micro-calorimeter works by absorbing a  $\gamma$  ray using a gold absorber. The heat is then transferred and measured by a  $\text{Ag:Er}^{3+}$  paramagnetic sensor. The sensor has a constant current circulating within the superconducting coil that polarizes the magnetic moment of the sensor. A change of flux is driven by the temperature-induced change of magnetization which is read as a voltage drop. This experiment is similar to Beck's micro-calorimeter experiment [12] as Sikorsky measures the energy of  $^{229\text{m}}\text{Th}$  indirectly by directly measuring other known nuclear states and using the same process to deduce the energy as

equations 2.1 and 2.2. However, Sikorsky's setup uses an improved detection system over Beck's. This measurement also compliments Seiferle and Wense's  $8.28 \pm 0.17$  eV measurement [36] but does not improve on it.

## 2.5 Other Notable Experiments

Notable experiments that do not have to do with energy measurements will be described in this section. First, the half-life and lifetime measurements for  $^{229m}\text{Th}$  will be explained. Afterwards, a few structural studies that have been performed will be described. A list of other  $^{229}\text{Th}$  experiments will be given at the end of the chapter.

### 2.5.1 Half-life/Lifetime Measurements

Instead of energy evaluations, some research has gone into evaluating half-life and lifetime measurements of  $^{229m}\text{Th}$  [13; 38], which has proven to be as difficult to evaluate as the exact transition energy. Seiferle in 2017 [13] determined a rough estimate of the lifetime of various charge states of  $^{229m}\text{Th}$ . Measurements were taken with  $^{229}\text{Th}^{1+}$ ,  $^{229}\text{Th}^{2+}$ , and  $^{229}\text{Th}^{3+}$ . In this study, the various charged particles were led into the grid of an MCP which neutralized the charged particles. This lets IC become the dominating decay channel. The 2% branching ratio for the isomer was also utilized in this study. The experimental setup involves a  $^{233}\text{U}$  radioactive source that decays into a UHP helium-filled gas-stopper cell that is directed by an RF funnel into an RF quadrupole. This RF quadrupole uses a negative DC gradient to guide the ions. However, the last electrode in the guide is independently biased in order to create a potential well inside the RF quadrupole. Ions are loaded into this potential well for 80 ms and a 0.1  $\mu\text{s}$  switch opens the well and releases the bunched ions into a mass separating quadrupole which filters out all daughter products and undesired charge states of  $^{229}\text{Th}$  with 70% efficiency. The desired  $^{229}\text{Th}$  charge states are then guided into an MCP. Figure 2.6 shows the signal using  $^{229}\text{Th}^{2+}$ . Note that the exponential decays of each kinetic energy remain the same. This signifies that IC is occurring. Figure 2.7 shows the decay characteristics for 2+ and 3+  $^{229m}\text{Th}$ . The fit line is used to determine the half-life of

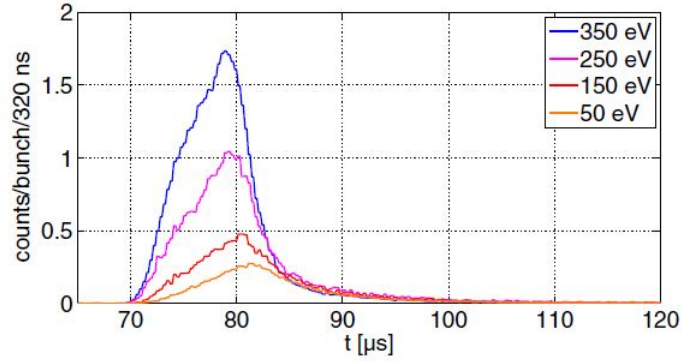


Figure 2.6: MCP signal of different kinetic energies of  $^{229}\text{Th}^{2+}$  and  $^{229}\text{Th}^{3+}$ . The exponential decay of each kinetic energy remains the same while the signal amplitude during impact is proportional to the velocity of the ion [13].

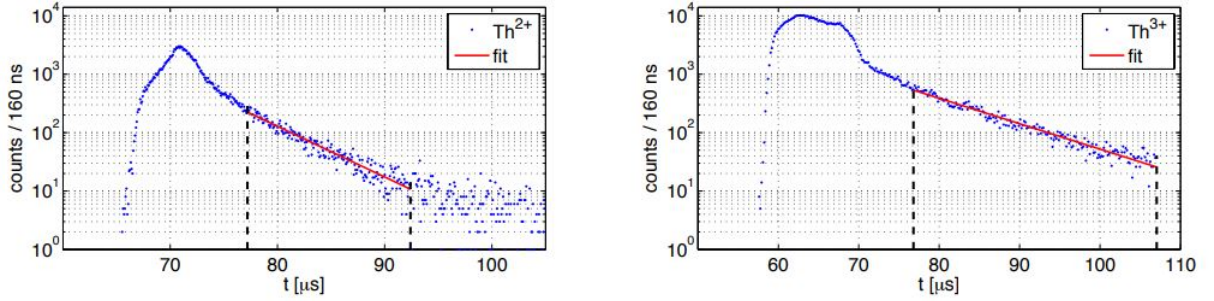


Figure 2.7: Plots of the time-dependent decay characteristics for  $^{229}\text{Th}^{2+}$  (Left) and  $^{229}\text{Th}^{3+}$  (Right). Isomeric half-life is indicated by the best fit line (Red) after charge recombination on the MCP surface [13].

the isomer. Unfortunately, no signal was obtained using  $^{229}\text{Th}^{1+}$ . This is most likely due to IC occurring before the ion strikes the MCP.

Tkalya in 2015 [38] used previous experimental data as well as a reduced probability matrix in order to estimate the half-life of the isomer and compare the results with [12; 39–41]. For a more detailed description of this process, please see [38]. The half-life of the isomer is given in terms of time and energy as

$$0.31 \times 10^6 \text{ s} \frac{\text{eV}^3}{\omega^3} \leq T_{1/2} \leq 2.1 \times 10^6 \text{ s} \frac{\text{eV}^3}{\omega^3},$$

where  $\omega = 7.8 \text{ eV}$  (the accepted mean energy value of  $^{229\text{m}}\text{Th}$ ).



## 2.5.2 Structural Studies

Thielking et al. [14] performed laser spectroscopic characterization of the hyperfine structure of  $^{229}\text{Th}$  in 2018. This setup consists of two separate experiments. One of the experiments, located at Physikalisch-Technische Bundesanstalt (PTB) in Brunswick, Germany, loaded an ion trap with  $^{229}\text{Th}$  and  $^{232}\text{Th}$ , both in the  $1+$  charge state. Because the source used was a  $\text{Th}(\text{NO}_3)_4$  solution that was dried on a tungsten target, this experiment will not contain the natural 2% branching ratio of  $^{229\text{m}}\text{Th}$ . Using laser ablation, the thorium ions were removed. Once the ions were trapped, three lasers were used to create  $^{229}\text{Th}^{2+}$ , while the  $^{232}\text{Th}$  remained untouched. One laser was used to excite the ground state to the  $24874\text{ cm}^{-1}$  state, while the next two lasers excited the ground state to the  $63258\text{ cm}^{-1}$  state. This continuous resonant pumping produces a cloud of approximate 1,000  $^{229}\text{Th}^{2+}$  ions. The next setup, located at Ludwig Maximilian University (LMU) in Munich, German, uses  $\alpha$  decay from  $^{233}\text{U}$  and then creates a beam which consists of all daughter products from the decay. The 2% decay branch will enable us to observe a  $^{229\text{m}}\text{Th}$  signal from this setup. All these particles, including the daughter particles, are trapped. The daughter products are claimed to not disturb the spectroscopic measurements. In both traps, argon (PTB) and helium (LMU) buffer gases are used to cool the ions to room temperature. The spectroscopic portion of the experiment is the same in both locations and utilizes three external lasers. See Figure 2.8 for a visual for the following explanation. A 459.1 nm laser is used to monitor the amount of  $^{229}\text{Th}^{2+}$  in the trap at a specific time which will be used to normalize the results. The other two lasers are directly used for spectroscopy: the first stage, 484.3 nm, is used to excite the ground state to the  $20711\text{ cm}^{-1}$  state while the 1164.3 nm laser, second stage, is used to pump the  $20711\text{ cm}^{-1}$  state to the  $29300\text{ cm}^{-1}$  state. Due to the spin states of the nuclear ground state and the nuclear isomer state, nine and eight hyperfine levels, respectively, are expected to be seen. For a detailed description of hyperfine levels, please see 3.2.3. To spectroscopically view the hyperfine levels, detuning of the lasers is necessary. Starting with the 484 nm laser, it is varied in 35 steps by  $\pm 120\text{ Mhz}$ . During each of these steps, the 1164 nm laser is scanned over the  $20711\text{ cm}^{-1} \rightarrow 29300\text{ cm}^{-1}$  transitions. Figure 2.9 shows the results of a second-stage frequency sweep during a stationary first-stage laser. Also shown is the 484.3 nm detuning to verify both  $^{232}\text{Th}^{2+}$  and



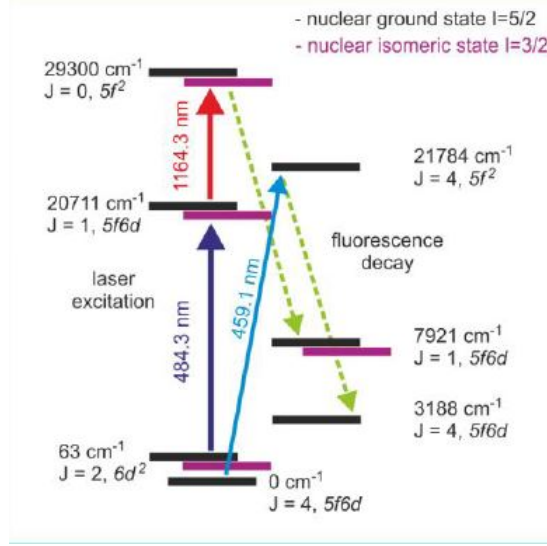


Figure 2.8: Atomic level scheme of  $^{229}\text{Th}^{2+}$ . The transitions and electronic configurations listed are ones used by [14] to determine the hyperfine structure. Energy of the states are listed in  $\text{cm}^{-1}$  (to convert reciprocal length to eV, use conversion  $(1 \text{ cm}^{-1})hc = 1.24 \times 10^{-4} \text{ eV}$ ). Solid arrows indicate direct laser excitation while the dashed arrows indicate decay channels.

$^{229}\text{Th}^{2+}$  contents inside the PTB ion trap. In the end, only seven out of eight resonances were observed in  $^{229m}\text{Th}^{2+}$ . The eighth resonance was calculated to have a signal-to-noise ratio that was unobservable. Figure 2.10 shows the hyperfine levels for the ground state, as well as the seven hyperfine levels that were observed for the isomeric state. From the bottom plot of the figure, there is a clear distinction between the PTB experiment where no isomer was trapped and the LMU experiment where 2% of the decays were in the desired isomeric state.

Robert Müller, also in 2018, performed a study on the hyperfine structure of both  $^{229}\text{Th}^{2+}$  and  $^{229m}\text{Th}^{2+}$  [42]. These data were taken during the same experiment as [14]. Precise measurements of the hyperfine coefficients using  $^{229}\text{Th}^{3+}$  were measured in 2012 [43]. Theoretical calculations, based on perturbation theory, of these coefficients were presented in [44], including the magnetic dipole moment  $\mu_{gr} = 0.36\mu_N$  and electric quadrupole moment  $Q_{gr} = 3.11 \text{ eb}$ . Müller used these moments to obtain hyperfine constants for the ground state as well as the first few excited states of  $^{229}\text{Th}^{3+}$  using the multi-configurational Dirac-Fock (MCDF) approach. The MCDF approach is explained in [45]. The  $^{229}\text{Th}^{3+}$  data, although not plotted, was said to offer great agreement to the theoretical data and provided a benchmark to the MCDF approach. Figure 2.8 shows the detailed level scheme for the low energy  $^{229}\text{Th}^{2+}$  charge state. Figure 2.11 plots the experimental

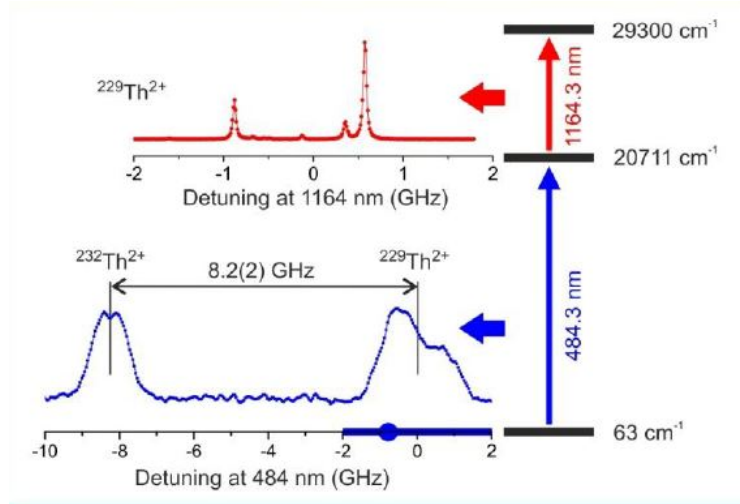


Figure 2.9: Laser detuning results for [14]. The bottom plot (Blue) shows the isotopic shifts of 8.2 GHz between  $^{232}\text{Th}^{2+}$  and  $^{229}\text{Th}^{2+}$ , which was obtained at PTB. The upper plot (Red) was obtained by fixing the first laser at -800 MHz and scanning the second laser between  $20711\text{ cm}^{-1}$  and  $29300\text{ cm}^{-1}$  (logarithmic scale).  $1\text{ cm}^{-1}$  is equivalent to  $10^7\text{ nm}$ .

data, theoretical data, and the MCDF approach. Note the agreement between the theoretical and MCDF approach. Only a few  $^{229}\text{Th}^{2+}$  states between  $0\text{ cm}^{-1}$  and  $29300\text{ cm}^{-1}$  were used while 45 states exist. The precision of this method would increase if all levels were used.

### 2.5.3 Other Studies

Many other studies have also been performed on  $^{229}\text{Th}$  that will not be explained in greater detail. Safronova et al. [46] performed experimental measurements of the hyperfine structure as well as the isotopic shift between  $^{229}\text{Th}^{2+}$  and  $^{232}\text{Th}^{2+}$ . Tkalya in 2019 [47] considered the decay of  $^{229m}\text{Th}$  by the atomic Rydberg states. Tkalya in 2020 also theoretically studied inelastic scattering of low energy electrons for excitation to create  $^{229m}\text{Th}$  [48] and also studied the half-life of the internal conversion channels of  $\text{Th}^-$  [49]. Other studies that may interest the reader include [50–57].

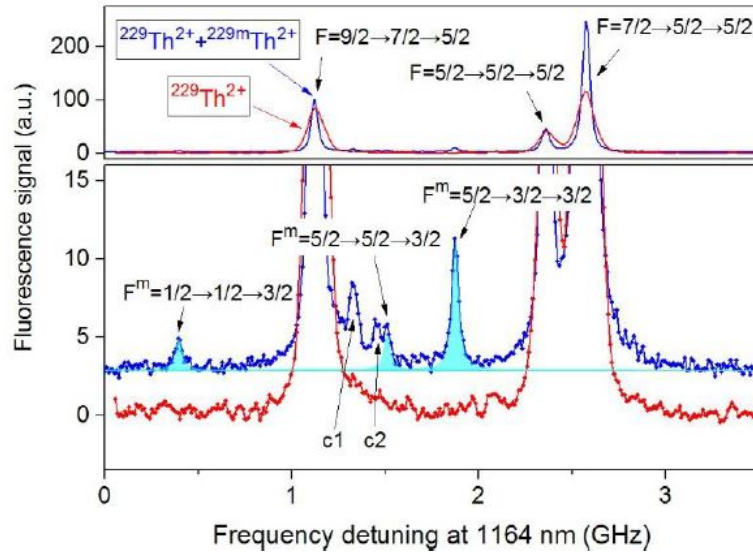


Figure 2.10: Two-stage laser excitation results with the first stage set to -800 MHz detuning with respect to the original hyperfine level. Red and blue colors indicate PTB and LMU traps, respectively. The bottom plot shows an enlarged view so the seven hyperfine levels can be seen. The total momenta  $F$  and  $F^m$  for the ground and isomeric states are shown, with the isomeric peaks highlighted in cyan. Note that the PTB experiment does not contain the isomeric state while the LMU experiment does.

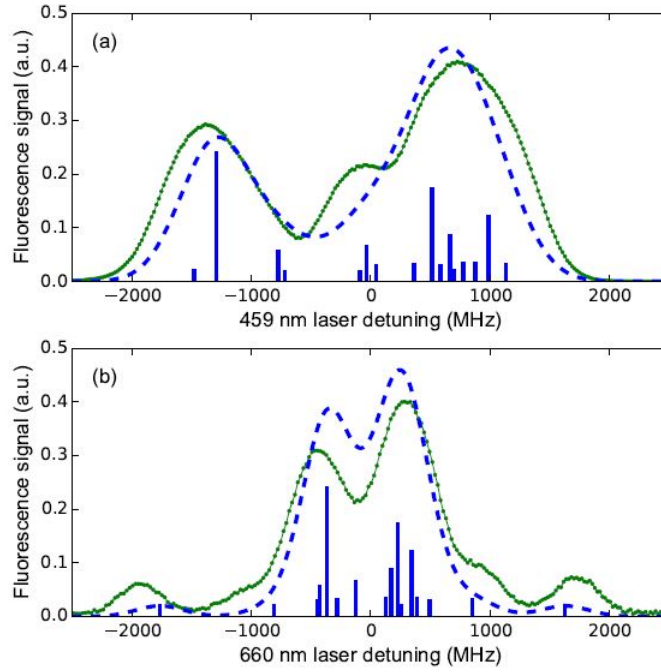


Figure 2.11:  $^{229}\text{Th}^{2+}$  hyperfine spectra. Referring to Figure 2.8, (a) is the transition from ground state  $0\text{ cm}^{-1}$  to the  $21784\text{ cm}^{-1}$  state that decays to the  $3188\text{ cm}^{-1}$  state, (b) is the transition from ground state  $0\text{ cm}^{-1}$  to  $15148\text{ cm}^{-1}$  state. Experimental data is represented by the blue bar, green connected dots are the MCDF approach, while the theoretical spectrum is represented by the blue dashed line.

# Chapter 3

## Theoretical Background

The fundamental theory behind how  $^{229m}\text{Th}$  was discovered and will be utilized in this experiment will be described in this chapter. The first section will describe the nuclear shell models and Nilsson models that led to the discovery of  $^{229m}\text{Th}$ . The second section will cover fundamental gamma decay and internal conversion as well as how it pertains to  $^{229m}\text{Th}$ . In the third section, radio frequency, quadrupoles, and mass separating quadrupoles will be covered in detail. The final section will cover atomic laser excitation by deriving the optical Bloch equations, using the equations to estimate the amount of ions excited inside a linear Paul ion trap, followed by an estimation of the half-life of  $^{229m}\text{Th}$ .

### 3.1 Nuclear Shell Model

First proposed in 1932 and developed in 1949, the nuclear shell model is a model of the nucleus of an atom that uses the Pauli Exclusion principle to describe the nucleus in terms of energy levels. Unfortunately, this model does not allow precise predictions of nuclear transitions in the eV range, only estimations that are then compared to experimental data. This section will include descriptions of the nuclear single particle shell model, Nilsson model, and rotational bands and it how it pertains to  $^{229m}\text{Th}$  and will closely follow [58].

### 3.1.1 Nuclear Single Particle Shell Model

The nuclear single particle shell model is the simplest of the single particle models. It is based on observation that the nuclear mass formula, which describes the nuclear masses quite well, fails for certain numbers of neutrons and protons. These numbers have been given the name "magic numbers", which correspond to the number of neutrons  $N = 20, 28, 50, 82, 126$  and proton number  $Z = 20, 28, 50, 82$ . This is due to a shell structure inside the nucleus, which is also observed in electrons (Coulomb force forcing electrons to occupy orbitals). At first, scientists believed that a single nucleon experienced the force of each individual nucleon. However, each nucleon is traveling rather quickly inside the nucleus. It has now been theorized that each nucleon is experiencing the average force of all nucleons combined.

In order to model the shell structure, one major assumption must be made. It is expected that in nuclei, the potential is more attractive in the center where the density is the highest while the attractiveness decreases as the density decreases toward the edge of the nucleus. Harmonic potential is then used to model this potential,

$$-\frac{\hbar^2}{2m}\Delta\psi(\vec{r}) + \frac{1}{2}mw^2r^2\psi(\vec{r}) = E\psi(\vec{r}) \quad (3.1)$$

where  $\hbar$  is the reduced Planck's constant,  $m$  is the mass of the nucleon,  $w$  is frequency of the nucleon,  $r$  is the distance from the center of the nucleus, and  $E$  is the energy of the nucleon. Realizing that  $r^2 = x^2 + y^2 + z^2$ , the Hamiltonian is actually a sum of an x, y, and z harmonic oscillator, and the eigenvalues are the sum of those oscillators,

$$E_{n_x n_y n_z} = (n_x + n_y + n_z + \frac{3}{2})\hbar w. \quad (3.2)$$

Equation 3.2 is disadvantageous due to the ignoring of rotational invariance on the potential. To fix this, the Schrödinger equation can be separated into radial coordinates as

$$R_{nl}(r)Y_{LM}(\Theta, \phi) \quad (3.3)$$

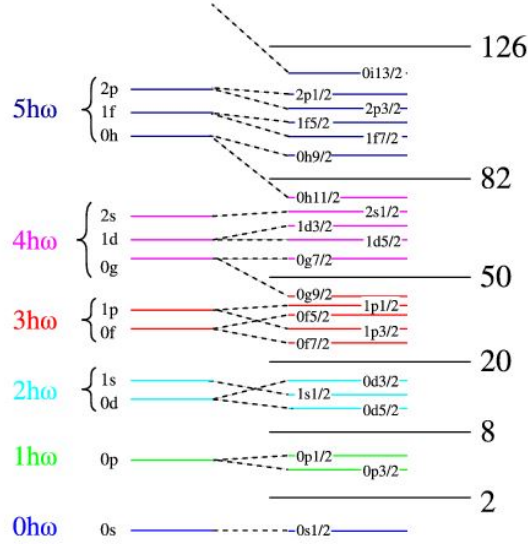


Figure 3.1: Visual representation of the shell model in nuclei [15]. Listed on the left side is the number harmonic oscillator quanta in each set of shells.

with  $Y$  being the spherical harmonics, we find

$$-\frac{\hbar^2}{2m} \frac{1}{r^2} \frac{\delta}{\delta r} \left( r^2 \frac{\delta}{\delta r} R(r) \right) + \frac{\hbar^2 L(L+1)}{r^2} R(r) + \frac{1}{2} m \omega^2 r^2 R(r) = E_{nL} R(r). \quad (3.4)$$

In this case, it can be shown that

$$E_{nL} = (2n + L + 3/2) \hbar \omega \quad (3.5)$$

where  $L$  is the orbital angular momentum of the state that Equation 3.2 neglected. Substituting  $s, p, d, f, g, h, i, j, \dots$  for  $L = 0, 1, 2, \dots$ , gives us the schematic of the shell structure shown in Figure 3.1.

### 3.1.2 Nilsson States

For many nuclei, an ellipsoidal shape is considered to be more stable than a spherical shape, especially with nuclei that do not have closed shells such as  $^{229}\text{Th}$ . Therefore, a deformed single-particle Hamiltonian was adopted to generate single-particle basis states in the following form, following [59]:

$$h(\mathbf{r}) = h_0 + h_\delta + a \mathbf{l} \cdot \mathbf{s} + b \mathbf{l}^2. \quad (3.6)$$

Here,  $h_0$  covers the spherical geometry and can be shown as an isotropic three-dimensional harmonic oscillator given as,

$$h_0 = -\frac{\hbar^2}{2\mu} \nabla^2 + \frac{1}{2} \mu \omega_0^2 r^2. \quad (3.7)$$

and  $h_\gamma$  describes the deformation as a quadrupole field as

$$h_\delta = -\frac{1}{3} \delta_{osc} \mu \omega_0^2 r^2 \sqrt{\frac{16\pi}{5}} Y_{20}(\theta, \phi), \quad (3.8)$$

where  $\mathbf{r}$  is the coordinate of the nucleon,  $\mu$  is its reduced mass,  $\omega_0$  is the frequency of the harmonic oscillator,  $Y_{20}(\theta, \phi)$  are spherical harmonics,  $a \mathbf{l} \cdot \mathbf{s}$  is the spin-orbit term and is required to account for nuclei with the magic number of protons and neutrons,  $b \mathbf{l}^2$  is used to give the proper ordering of single-particle states in the spherical limit, and  $\delta_{osc}$  is the deformation parameter and will be described later. The eigenvectors produced by Equation 3.6 are referred to as Nilsson states or Nilsson orbitals. For large deformations, the terms  $\mathbf{l} \cdot \mathbf{s}$  and  $\mathbf{l}^2$  can be ignored which allows the transformation into cylindrical coordinates which can now be written as

$$h = -\frac{\hbar^2}{2\mu} \nabla^2 + \frac{1}{2} \mu (\omega_3^2 x_3^2 + \omega_\perp^2 (x_1^2 + x_2^2)), \quad (3.9)$$

where  $\omega_3$  is the oscillator frequency along the symmetric axis and  $\omega_\perp$  is the oscillator frequency perpendicular to the symmetric axis. Both are given by

$$\omega_3 = \omega_0 \sqrt{1 - \frac{4}{3} \delta_{osc}} = \bar{\omega} (1 - \frac{2}{3} \delta_{osc}) \quad (3.10)$$

and

$$\omega_\perp = \omega_0 \sqrt{1 + \frac{2}{3} \delta_{osc}} = \bar{\omega} (1 + \frac{1}{3} \delta_{osc}), \quad (3.11)$$



and the deformation parameter in terms of frequencies can be written as

$$\delta_{osc} = \frac{\omega_{\perp} - \omega_3}{\bar{\omega}}, \quad (3.12)$$

where the average frequency  $\bar{\omega}$  is given by

$$\bar{\omega} = \frac{1}{3}(\omega_1 + \omega_2 + \omega_3) = \frac{1}{3}(2\omega_{\perp} + \omega_3) \approx \omega_0. \quad (3.13)$$

For large deformation, the single-particle energy can be expressed in terms of the oscillator quantum  $n_i$  along the three axes, assuming axial symmetry,

$$E_{Nn_3\Omega} = (n_3 + \frac{1}{2})\hbar\omega_3 + (n_2 + \frac{1}{2})\hbar\omega_2 + (n_1 + \frac{1}{2})\hbar\omega_1 = (n_3 + \frac{1}{2})\hbar\omega_3 + (n_{\perp} + 1)\hbar\omega_{\perp}, \quad (3.14)$$

where  $n_{\perp} = n_1 + n_2$  is the number of quanta perpendicular to the symmetry axis. Due to the total number of quanta being fixed, this leads to

$$N = n_3 + n_2 + n_1 = n_3 + n_{\perp}. \quad (3.15)$$

When the magic number correction is included, a  $1 \cdot s$ , degeneracy among states with different projections of angular momentum no longer occurs. Therefore, the orbital angular momentum, also, is a good quantum number. This leads to Nilsson states being described as a function of  $[Nn_3\lambda\Omega]$  which leads to

$$\Omega^{\pi}[Nn_3\lambda], \quad (3.16)$$

where  $\pi = (-1)^N$  is the parity of the state.

The deformation parameter  $\delta_{ocs}$  can also be defined in terms of the length of the ellipsoid structure  $b$  and the length orthogonal to the symmetry axis  $a$ ,

$$\delta_{osc} = \frac{2(b - a)}{b + a}. \quad (3.17)$$

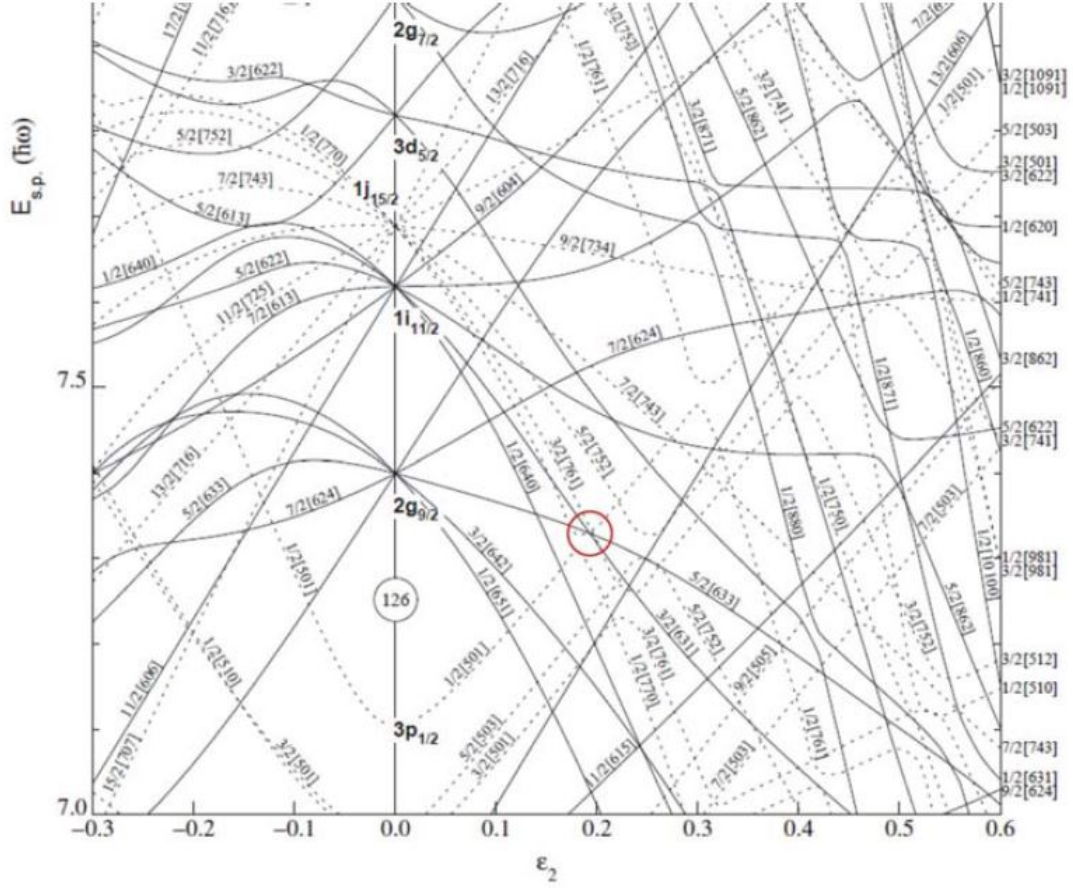


Figure 3.2: Nilsson diagram for neutron single-particle states. Single-particle energy is shown as a function of the  $\epsilon_2$ , the deformation parameter. Circled is the intersection of the  $5/2^+[633]$  ground state and the  $3/2^+[631]$  isomeric state of  $^{229}\text{Th}$  [16].

Instead of  $\delta_{osc}$ , the deformation parameter is often defined as  $\epsilon_2$ , which relates to  $\delta_{osc}$  as

$$\epsilon_2 = \delta_{osc} + \frac{1}{6}\delta_{osc}^2 + \frac{5}{18}\delta_{osc}^3 + \dots \quad (3.18)$$

This deformation parameter was estimated to be  $\epsilon_2 \approx 0.19$  [60] by using the electric quadrupole moment. Figure 3.2 displays a Nilsson diagram for different states in terms of deformation parameter  $\epsilon_2$ . The ground state and isomeric state of  $^{229}\text{Th}$  is circled in red.

### 3.1.3 Rotational Bands

A rotational band is simply a group of states that all have different total angular momenta  $J$  but share the same intrinsic state. The angular momentum  $\mathbf{J}$  of a rotating object is directly proportional to its angular velocity  $\omega$ ,

$$\mathbf{J} = I\omega, \quad (3.19)$$

where  $I$  is the moment of inertia. The rotational energy  $E_J$  can be written in terms of the proportionality of the square of the angular frequency which is also proportional to the square of angular momentum,

$$E_J = \frac{1}{2}I\omega^2 = \frac{1}{2I}J^2. \quad (3.20)$$

The rotational Hamiltonian can then be written as

$$H = \sum_{i=1}^3 \frac{\hbar^2}{2I_i} J_i^2, \quad (3.21)$$

where  $I_i$  is the moment of inertia along the  $i$ -th axis. For an axially symmetric object where 3 is the axis of symmetry, the moment of inertia along an intrinsic set of coordinate axes 1, 2, and 3 has the property  $I_1 = I_2 \equiv I$ . If  $I_3 = I$  then the shape would be spherical, which does not allow rotational degrees of freedom due to the inability to distinguish between wave functions before and after rotations. The Hamiltonian may now be written as

$$H = \frac{\hbar^2}{2I}(\mathbf{J}^2 - \mathbf{J}_3^2) + \frac{\hbar^2}{2I_3}\mathbf{J}_3^2. \quad (3.22)$$

In order to specify a rotational body's orientation in space, classically, three Euler angles  $(\alpha, \beta, \gamma)$  are required. For quantum mechanics, the three Euler angles must be described in quantum numbers in order to describe the rotational state. This can be done using the total angular momentum  $J$ , the projection of  $J$  along the quantization axis  $M$ , and the constant of motion  $K$ . Because  $J$ ,  $M$ , and  $K$  are functions of the Euler angles, they must be described in terms of a rotational matrix

$D_{MK}^J(\alpha, \beta, \gamma)$ . This function may be defined by the relation

$$Y_{JK}(\theta', \phi') = \sum_M D_{MK}^J(\alpha, \beta, \gamma) Y_{JM}(\theta, \phi), \quad (3.23)$$

where  $Y_{JK}(\theta', \phi')$  are spherical harmonics of order  $J$  in a coordinate system rotated by the Euler angles. To define parity, a linear combination of rotational matrices with both positive and negative  $K$ -values is required. The rotational wave function becomes

$$|JMK\rangle_{rot} = \sqrt{\frac{2K+1}{16\pi^2(2+\delta_{K0})}} \{D_{MK}^J(\alpha, \beta, \gamma) \pm (-1)^{J+K} D_{M-K}^J(\alpha, \beta, \gamma)\}. \quad (3.24)$$

Because the parity of a rotational state is given by Equation 3.24, there are clear limits to what  $J$ -values can be,

$$J = \begin{cases} 0, 2, 4, \dots & \text{for } K^\pi = 0^+ \\ 1, 3, 5, \dots & \text{for } K^\pi = 0^- \end{cases}$$

The energy of the state represented by the Hamiltonian in Equation 3.22 is given by

$$E_J = \frac{\hbar^2}{2I} J(J+1) + E_K, \quad (3.25)$$

where  $E_K$  represents the contributions from the intrinsic part of the wave function.

Each energy level of the Nilsson model described in Chapter 3.1.2 has a rotational band. Each rotational band has a ground state that is called the band head. In Kroger and Reich [8], direct evidence of the band head was the missing piece of the puzzle that led to the research of  $^{229m}\text{Th}$  today.

## 3.2 Gamma Decay and Internal Conversion

Most nuclear reactions leave the nucleus in an excited state. This excited nucleus usually decays by emission of one or more gamma rays until it reaches the ground state. However, for  $^{229m}\text{Th}$ , there are two decay channels that can be observed. These two channels are associated with

gamma decay of the approximately 8 eV photon (approximately 160 nm) and the internal conversion channel which transfers the energy of the nucleus to an electron which is consequently ejected. Unfortunately, the direct detection of the gamma decay channel has eluded scientists. The internal conversion electron, however, has successfully been measured which gives an indirect measurement of the excited state. The following sections will discuss the theory behind both decay channels and will closely follow [58].

### 3.2.1 Gamma Decay

In gamma decay, a nucleus goes from an excited state to a state of lower energy. The energy difference between these two states is then released as a gamma ray. These gamma rays typically have energies within the range of 0.1 to 10 MeV, with a few exceptions (one of which includes  $^{229m}\text{Th}$ ). These energies correspond to wavelengths between  $10^4$  and 100 fm. Wavelengths of this length are much shorter than visible light, which is  $10^6$  times longer than most gamma rays. Following [58], let's consider the decay of a nucleus of mass  $M$  at rest, from an initial excited state  $E_i$  to a final state  $E_f$ . To conserve linear momentum, the final nucleus will not be at rest but must have a recoil momentum  $p_R$  and corresponding recoil kinetic energy  $T_R$ , which is assumed to be nonrelativistic ( $T_R = p_R^2/2M$ ). Conservation of total energy and momentum give

$$\begin{aligned} E_i &= E_f + E_\gamma + T_R \\ 0 &= p_R + p_\gamma. \end{aligned} \tag{3.26}$$

It follows that  $p_R = p_\gamma$ ; the nucleus recoils with a momentum equal and opposite to that of the gamma ray. Defining  $\Delta E = E_i - E_f$  and using the relativistic relationship  $E_\gamma = cp_\gamma$ ,

$$\Delta E = E_\gamma + \frac{E_\gamma^2}{2Mc^2}, \tag{3.27}$$

which has the solution

$$E_\gamma = Mc^2 \left[ -1 \pm \left( 1 + 2 \frac{\Delta E}{Mc^2} \right)^{1/2} \right]. \tag{3.28}$$

Assuming the energy differences  $\Delta E$  are on the order of MeV while the rest energies are of order  $A \times 10^3$  MeV, where  $A$  is the mass number, it can be shown that  $\Delta E \ll Mc^2$ . Therefore, we only keep the first three terms in the expansion of the square root,

$$E_\gamma \approx \Delta E - \frac{\Delta E^2}{2Mc^2}, \quad (3.29)$$

which equals Equation 3.27 with the approximation  $\Delta E \approx E_\gamma$ . The comparison shows that the actual gamma-ray energy is somewhat diminished from the available energy  $\Delta E$ . However, the recoil energy for the nucleus when emitting low-energy gamma rays is negligible. This is beneficial when studying  $^{229m}\text{Th}$  as the measured gamma-ray energy should very closely represent the total energy of the metastable state.

### 3.2.2 Internal Conversion

Internal conversion (IC) is a process that competes with gamma-ray emission. During the IC process, the nuclear excitation energy after decay is transferred to an electron instead of emitting a gamma ray. The excited electron is then ejected from the atomic shell. IC can be visualized in the following way: when a nucleus de-excites either by a change in the rotational motion as a whole or as a nucleon jumping from one orbit to another, there is a sudden disturbance in the electromagnetic field. The innermost electrons, such as those in the K- and L- orbits, spend a large amount of time in the vicinity of the nucleus. It is possible that this disturbance will transfer its energy to the electron in the vicinity of the nucleus and eject it. Unlike beta decay, the electron emitted during IC is monoenergetic. The energy of this electron  $E_e$  is equivalent to the nuclear transition energy  $E_\gamma$  minus the binding energy of the electron  $E_b$ ,  $E_e = E_\gamma - E_b$ . This is important because this shows that IC can only occur when  $E_\gamma > E_b$ . Using one of the recent accepted values for  $^{229m}\text{Th}$  of 8.28 eV, the binding energy of the electron must not exceed 8.28 eV for IC to occur. For singly charged thorium,  $^{229}\text{Th}^{1+}$ ,  $E_b = 11.9$  eV, while for neutral  $^{229}\text{Th}$ ,  $E_b = 6.3$  eV. Because the binding energy increases for multi-charged isotopes, the IC process is energetically forbidden for all positive charges of  $^{229}\text{Th}$  but is expected to be the major decay

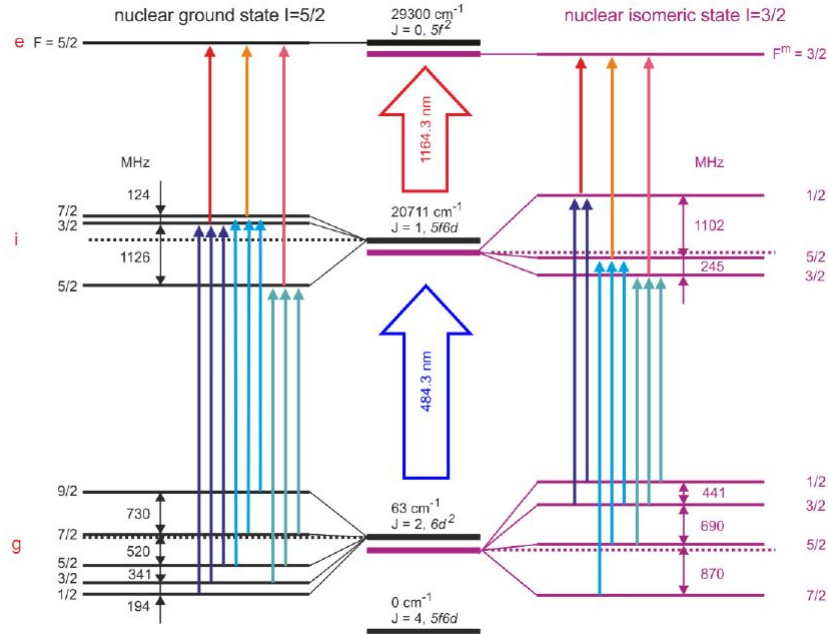


Figure 3.3: Detailed level scheme outlining the hyperfine levels of ground state and isomeric state of  $^{229}\text{Th}$ . These hyperfine levels are indicated by their total angular momentum  $F$  and  $F^m$  [14].

channel for neutral  $^{229}\text{Th}$ .

### 3.2.3 Hyperfine Structures

Hyperfine structure refers to the splitting of an atomic energy state when either an electric or magnetic field is applied. These effects are detrimental to the accuracy of a future nuclear clock. Hyperfine splitting arises from time-independent perturbation theory, which will not be covered in this thesis. Explicit numerical explanation of general hyperfine splitting is easiest when using a hydrogen atom, as it only contains one proton and one electron. For a detailed explanation of perturbation theory and hyperfine splitting of hydrogen, [61] is highly suggested. However, when looking at a complex ion such as  $^{229}\text{Th}^{3+}$ , the process of determining the energy splitting requires experimental approximations. Also, because most important quantities such as energy, magnetic dipole moment, and electric-quadrupole moment are not known to high precision, experimental approximations of the hyperfine splitting will continue to have a large error.

Hyperfine splitting occurs because of two different effects, the Stark effect and the Zeeman effect. The Stark effect, which has both linear and quadratic aspects, occurs when an outside elec-

| Type of shift         | Shift ( $\times 10^{-20}$ ) | Uncertainty ( $\times 10^{-20}$ ) |
|-----------------------|-----------------------------|-----------------------------------|
| Excess micromotion    | 10                          | 10                                |
| Gravitational         | 0                           | 10                                |
| Cooling laser Stark   | 0                           | 5                                 |
| Electric quadrupole   | 3                           | 3                                 |
| Secular motion        | 5                           | 1                                 |
| Linear Doppler        | 0                           | 1                                 |
| Linear Zeeman         | 0                           | 1                                 |
| Background collisions | 0                           | 1                                 |
| Blackbody radiation   | 0.013                       | 0.013                             |
| Clock laser Stark     | 0                           | $\ll 0.01$                        |
| Trapping field Stark  | 0                           | $\ll 0.01$                        |
| Quadratic Zeeman      | 0                           | 0                                 |
| Total                 | 18                          | 15                                |

Figure 3.4: List of expected systemic shifts with their uncertainties of a  $^{229m}\text{Th}^{3+}$  nuclear clock [17].

tric field is introduced to an excited atom. The quadratic Stark effect arises from non-degenerate time-independent perturbation theory, where the spin of the electron is ignored, whereas the linear Stark effect arises from degenerate time-independent perturbation theory, where the spin of the electron is taken into account. The Zeeman effect occurs when a magnetic field encounters the atomic shell. There are three types of Zeeman effects: normal, anomalous, and diamagnetic. The normal Zeeman effect is caused by an interaction with an orbital magnetic moment. Anomalous Zeeman effect is caused by the interaction with combined orbital and intrinsic magnetic moments. The diamagnetic Zeeman effect is caused by the interaction with a field-induced magnetic moment. Figure 3.4 shows a list of all the expected shifts due to hyperfine splitting in  $^{229m}\text{Th}$ , as well as other systemic shifts [17].

### 3.3 Radio Frequency and Quadrupoles

This section will be dedicated to describing the mechanisms of how the  $^{229}\text{Th}$  beam was focused and mass separated before ion trapping could occur which is a vital step in isolating the specific charge state needed to deny internal conversion.



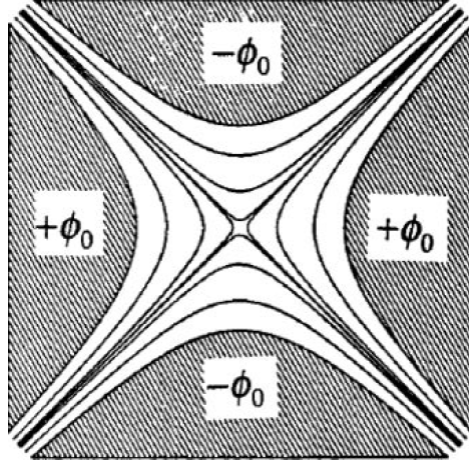


Figure 3.5: Hyperbolic quadrupoles showing equipotential lines [18].

### 3.3.1 Radio Frequency and Quadrupoles

Radio frequency (RF) is simply the oscillation rate of a voltage or a current, or the oscillation rate of a magnetic, electric, or electromagnetic field in the frequency range of 20 kHz to 300 GHz. RF has many different applications that cover many different fields of study from communications, medicine, energy, measurements, and mechanics.

A quadrupole is a system of electric charges with four parallel poles. When an oscillating RF current is added to a quadrupole with the two outputs phase-shifted  $180^\circ$ , charged particles passing through experience a focusing effect, see Figure 3.6. The electric potential of the linear quadrupole,  $\phi(x, y)$ , is given by

$$\phi(x, y) = \frac{x^2 - y^2}{r_0^2} \phi_0, \quad (3.30)$$

where  $x$  and  $y$  are Cartesian coordinates. This potential can be visualized in Figure 3.5. The parameter  $r_0$ , called the "field radius", is the distance from the center to the electrode. If balanced potentials, that is  $|\phi_0| = |-\phi_0|$ , are applied then it can be shown that the potential in the center of the quadrupole is zero. Because hyperbolic electrodes are expensive and difficult to precisely machine, round rods are often used as an approximate result. The optimum approximate result can be estimated by discussing the ratio  $r/r_0$ , where  $r$  is the radius of the round rod. This ratio has been discussed by many scientists with many different "optimum" values. The general consensus

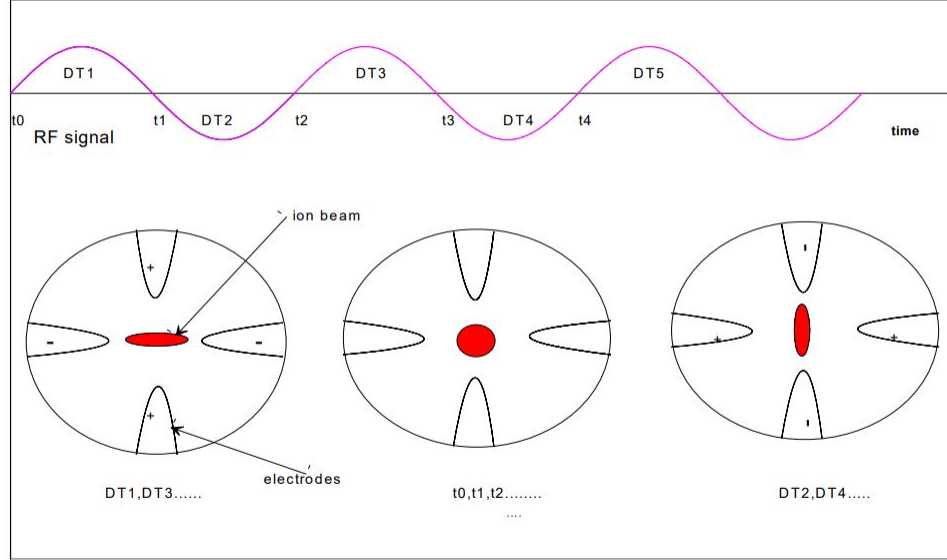


Figure 3.6: Visualization of the oscillation of RF in a quadrupole that leads to focusing of a charged particle beam [19]. Ions remain at constant velocity inside a RF quadrupole unless a voltage gradient is applied.

of the optimum ratio, however, is  $1.128 < r/r_0 < 1.148$ , with Douglass expressing that 1.128 produces the ideal electric field [18].

### 3.3.2 Mass Separation Quadrupole

The linear quadrupole developed for mass spectrometry didn't happen until 1958 [62]. Mass selection occurs when different DC voltages are applied to the diagonally adjacent rods. The potential applied to the quadrupole rods has both DC potential and time-varying parts, which is given by [18]

$$\phi_0(t) = U - V_{RF} \cos \Omega t, \quad (3.31)$$

where  $U$  is the DC voltage applied pole to ground,  $V_{RF}$  is a zero to peak alternating voltage applied pole to ground,  $\Omega$  is the operating frequency (which should be in the RF range), and  $t$  is time. The quadrupole potential is

$$\Phi(x, y, t) = \frac{x^2 - y^2}{r_0^2} (U - V_{RF} \cos \Omega t). \quad (3.32)$$

Ion motion in a vacuum is determined by Newton's second law of motion

$$\mathbf{F} = m \frac{d\mathbf{v}}{dt} \quad (3.33)$$

where  $\mathbf{F}$  is the force on an ion,  $m$  is the mass of the ion. and  $\mathbf{v}$  is the velocity of the ion. The force on a positive ion is

$$\mathbf{F} = -ze\nabla\Phi(x, y, t), \quad (3.34)$$

where  $z$  is the charge of the ion and  $e$  is the elementary charge constant. The electric field in the  $x$ -direction is

$$E_x = -\frac{\delta\Phi(x, y, t)}{\delta x} = -\frac{2x}{r_0^2}(U - V_{RF}\cos\Omega t) \quad (3.35)$$

and in the  $y$ -direction is

$$E_y = -\frac{\delta\Phi(x, y, t)}{\delta y} = -\frac{2y}{r_0^2}(U - V_{RF}\cos\Omega t). \quad (3.36)$$

The  $x$ - and  $y$ -direction electric fields are considered independent of each other. Next, ion motion and stability in the  $x$ - and  $y$ -direction will be discussed. The equation of motions for the  $x$ -direction can be written as

$$m \frac{d^2x}{dt^2} = -\frac{2zeU}{r_0^2}(U - V_{RF}\cos\Omega t). \quad (3.37)$$

Now the following variables will be introduced:

$$\xi = \frac{\Omega t}{2}, \quad a_x = \frac{8zeU}{mr_0^2\Omega^2}, \quad q_x = \frac{4zeV_{RF}}{mr_0^2\Omega^2}. \quad (3.38)$$

Equation 3.37 becomes

$$\frac{d^2x}{d\xi^2} + (a_x - 2q_x\cos 2\xi)x = 0. \quad (3.39)$$

Equation 3.39 is considered the Mathieu equation. Ion motion consists of oscillations with com-

binations of many different frequencies. The angular frequencies of ion oscillation are given by

$$\omega_n = (2n + \beta) \frac{\Omega}{2}, \quad (3.40)$$

where  $n = 0, \pm 1, \pm 2, \dots$ , and  $\beta$  is a function of the  $a_x$  and  $q_x$  parameters. From these equations, stability diagrams were created. Solutions to the Mathieu equation are considered either "stable" or "unstable". A solution is "stable" if the amplitude of the oscillation remains finite as  $t \rightarrow \infty$ . Conversely, a solution is considered "unstable" if the amplitude increases exponentially with time, which would cause the ion to either strike the rods or escape in between in a quadrupole. Figure 3.8 shows combinations of  $a_x$  and  $q_x$  that are stable.

Similarly, the equation of ion motion in the  $y$ -direction is

$$m \frac{d^2 y}{dt^2} = + \frac{2zeU}{r_0^2} (U - V_{RF} \cos \Omega t). \quad (3.41)$$

The difference between Equation 3.37 and Equation 3.41 is the sign difference on the right side of the equation. Using another change of variables, Equation 3.41 can also be written as the Mathieu Equation,

$$\xi = \frac{\Omega t}{2}, \quad a_y = - \frac{8zeU}{mr_0^2 \Omega^2}, \quad q_y = - \frac{4zeV_{RF}}{mr_0^2 \Omega^2}. \quad (3.42)$$

Substituting these variables back into Equation 3.41 produces the Mathieu equation in the  $y$ -direction,

$$\frac{d^2 y}{d\xi^2} + (a_y - 2q_y \cos 2\xi)y = 0. \quad (3.43)$$

Because Equation 3.39 and Equation 3.43 are similar, it is trivial to show that both  $x$ -direction and  $y$ -direction motion will be stable if  $a_x = a_y$  and  $q_x = q_y$ . Regions in Figure 3.8 where both the  $x$ - and  $y$ -motions are stable are called "stability regions". There are eight different stability regions shown. For the scope of this thesis, however, only the first stability region will be covered.

Figure 3.7 shows the first region of the stability diagram. The peak of the first stability region lies at  $a = 0.23699$  and  $q = 0.70600$  while the boundaries are at coordinates (0,0) and (0.908,

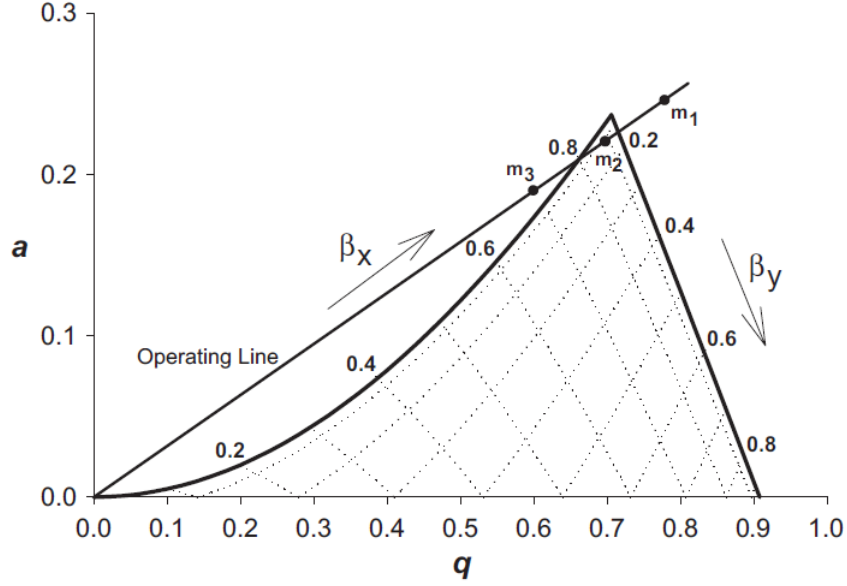


Figure 3.7: The first stability region with a mass separation operating line [18]. The operating line has equation  $a = \frac{2U}{V_{RF}}q$

0) with the axes being  $(q, a)$ . The operating line,  $\beta_x$ , has a slope of  $\frac{2U}{V_{RF}}$ . This means that for a given DC voltage and RF voltage, all masses of ions will fall on this line. It is optimal to have the mass and charge of the chosen ion to exist near the peak of the first stability region for the best resolution. Looking at Figure 3.7,  $m_2$  is located near the peak while  $m_1$  and  $m_3$  are being filtered out. Here,  $m_3$  is a heavier ion having a smaller  $a$  and  $q$  value while  $m_1$  is a lighter ion having a larger  $a$  and  $q$  value. If a detector was placed at the end of the mass separation quadrupole, you would detect only  $m_2$  ions. If, for instance, you want to detect  $m_3$  instead,  $U$  and  $V_{RF}$  would need to be adjusted accordingly. One could also change the resolution of the mass scan by changing the slope of the  $\beta_x$  line. Decreasing the slope of the line would allow a variety of ions to pass through the mass selector, therefore, contaminating the beam.

If ions do not spend a sufficient amount of time in the mass separation region, they may be transmitted even though they are outside of the stability region. Therefore it is necessary to make sure the ions are traveling at a velocity that allows them to complete a full oscillation while traveling inside the mass selector, either by slowing them down or making the quadrupole longer. The mass separation quadrupole used in this thesis does not have a voltage gradient applied, therefore ions will not be accelerated forward. However, if ions are slowed enough,

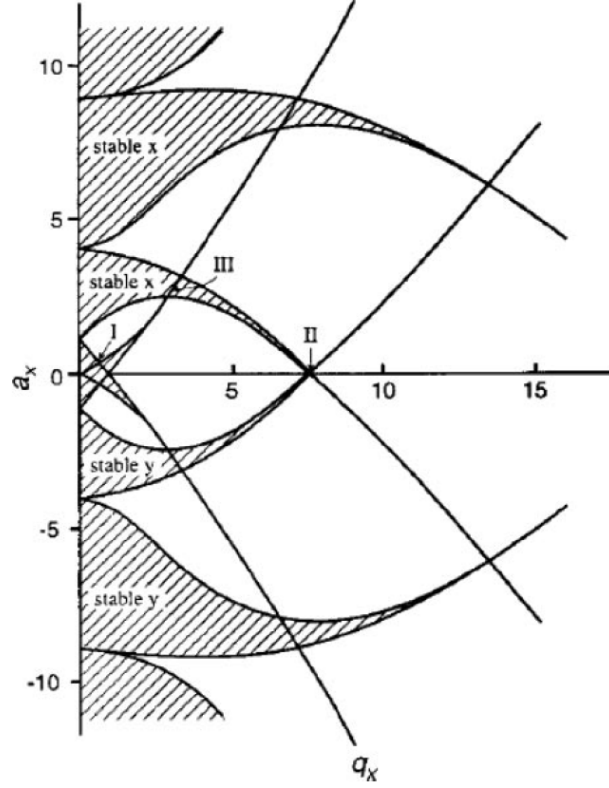


Figure 3.8: Regions of stability in the  $x$ - and  $y$ -directions in a linear quadrupole [18].

consecutive ions will interact by Coulomb repulsion and slowly force the ions forward out of the mass separation quadrupole.

### 3.4 Laser Excitation

Laser excitation is a necessary step in both determining the contents and the quantity of the contents, inside a linear Paul ion trap as well as searching for the isomeric state of  $^{229}\text{Th}$ . The optical Bloch equations will be derived and evaluated for an arbitrary amount of  $^{229}\text{Th}^{3+}$  ions inside an ion trap. Because little is known about laser/nucleus interactions, and because little knowledge of  $^{229m}\text{Th}$  exists in literature, no theory exists to predict the amount of isomers created while driving the  $^{229}\text{Th}$  nucleus with a laser. Therefore, a half-life estimation is made assuming an approximate isomer energy of 8 eV.

### 3.4.1 Optical Bloch Equations

The optical Bloch equations, sometimes called the Maxwell-Bloch equations, describe the coupling of light in a two-state quantum system. The corresponding description of the optical Bloch equations will closely follow [63]. The theory starts with the time-dependent Schrödinger equation,

$$\hat{H}\Psi(\mathbf{r}, t) = i\hbar \frac{\delta\Psi(\mathbf{r}, t)}{\delta t}, \quad (3.44)$$

where  $\hat{H}$  is the time-dependent quantum mechanical Hamiltonian,  $\Psi(\mathbf{r}, t)$  is the time-dependent wave function,  $i$  labels the different solutions,  $\hbar$  is the reduced Planck's constant, and  $\mathbf{r}$  symbolizes the coordinates of all the particles that makes up the atom. Solutions of the time-independent wave function take the form

$$\Psi_i(\mathbf{r}, t) = \psi_i(\mathbf{r}) \exp \frac{-iE_i t}{\hbar}. \quad (3.45)$$

Substitution of Equation 3.45 into Equation 3.44 gives the energy eigenvalue equation,

$$\hat{H}_A \psi_i(\mathbf{r}) = E_i \psi_i(\mathbf{r}). \quad (3.46)$$

The atomic states described by Equations 3.45 and 3.46 are known as "stationary states". A stationary state has the property that the average value is independent of time unless explicitly stated.

Only two specific eigenstates are of concern when calculating the Einstein coefficient  $B$ . These are labeled  $i = 1$  and  $2$  and are given by,

$$\begin{aligned} \Psi_1(\mathbf{r}, t) &= \psi_1(\mathbf{r}) \exp \frac{-iE_1 t}{\hbar} \\ \Psi_2(\mathbf{r}, t) &= \psi_2(\mathbf{r}) \exp \frac{-iE_2 t}{\hbar} \end{aligned} \quad (3.47)$$

Now consider the atom is introduced to an incident light beam which has electric and magnetic fields that vary with time. This interaction causes additional electromagnetic energy to be introduced to the system, which can be represented by an additional Hamiltonian  $\hat{H}_I$ . The total

Hamiltonian can then be expressed as

$$\hat{H} = \hat{H}_A + \hat{H}_I. \quad (3.48)$$

The system no longer has stationary states as the new total Hamiltonian is now dependent explicitly on time due to the addition of the incident light beam.

If the frequency of the incident light  $\omega$  is close to the resonant frequency of the state  $\omega_0$ , the wave function becomes a linear superposition

$$\Psi(\mathbf{r}, t) = C_1(t)\Psi_1(\mathbf{r}, t) + C_2(t)\Psi_2(\mathbf{r}, t). \quad (3.49)$$

The combinations of states vary with time and are limitless. However, if the state  $\Psi(\mathbf{r}, t)$  is normalized then

$$\int dV |\Psi(\mathbf{r}, t)|^2 = |C_1(t)|^2 + |C_2(t)|^2 = 1, \quad (3.50)$$

where the functions from Equation 3.47 are used and are normalized and orthogonal. Substitution of Equation 3.49 into Equation 3.44 and some simplification gives the equation for coefficients

$$\hat{H}(C_1\Psi_1 + C_2\Psi_2) = i\hbar \left( \Psi_1 \frac{dC_1}{dt} + \Psi_2 \frac{dC_2}{dt} \right). \quad (3.51)$$

Multiplying the left side of the previous equation with the complex conjugate  $\Psi_1^*$  and integrating over all space while substituting 3.47 and  $\hbar\omega_0 = E_2 - E_1$  will yield

$$\langle 1|\hat{H}_I|1\rangle C_1 + \exp(-i\omega_0 t) \langle 1|\hat{H}_I|2\rangle C_2 = i\hbar \frac{dC_1}{dt}. \quad (3.52)$$

For those not familiar with Dirac notation,

$$\langle 1|\hat{H}_I|1\rangle = \int dV \psi_1^* \hat{H}_I \psi_1. \quad (3.53)$$



Similarly, taking the complex conjugate  $\psi_2^*$  and multiplying the left side of Equation 3.51,

$$\langle 2|\hat{H}_I|2\rangle C_2 + \exp(i\omega_0 t)\langle 2|\hat{H}_I|1\rangle C_1 = i\hbar \frac{dC_2}{dt}. \quad (3.54)$$

Now, consider an atom interacting with a travelling electromagnetic wave. Looking at the electric and magnetic fields,

$$\mathbf{E} = \mathbf{E}_0 \cos(kz - \omega t) \quad (3.55)$$

$$\mathbf{B} = \mathbf{B}_0 \cos(kz - \omega t).$$

The Hamiltonian that describes the time-dependent interactions can be written as

$$\hat{H}_I = \hat{H}_{ED} = e\mathbf{D} \cdot \mathbf{E}_0 \cos(\omega t), \quad (3.56)$$

where  $e\mathbf{D}$  is the total electric dipole, and

$$\mathbf{D} = \sum_{\alpha=1}^Z \mathbf{r}_\alpha. \quad (3.57)$$

$\hat{H}_I$  is real and has odd parity, meaning, its sign changes during inversion of the nucleus, when  $\mathbf{r}_\alpha$  becomes negative. The diagonal matrix elements must nullify,

$$\langle 1|\hat{H}_{ED}|1\rangle = \langle 2|\hat{H}_{ED}|2\rangle = 0. \quad (3.58)$$

Because the states have opposite parity, the matrix elements not on the diagonal are related by,

$$\langle 2|\hat{H}_{ED}|1\rangle = \langle 1|\hat{H}_{ED}|2\rangle. \quad (3.59)$$

If  $\mathbf{E}_0$  is pointed in the  $x$ -direction, each matrix element becomes,

$$\langle 1|\hat{H}_{ED}|2\rangle = eE_0 X_{12} \cos(\omega t), \quad (3.60)$$

and

$$X_{12} = \int dV \psi_1^* X \psi_2, \quad (3.61)$$

where  $X$  is the  $x$ -component of the atomic dipole  $\mathbf{D}$ . Next, we define a variable  $\nu$  that has units of frequency and relate it as

$$\hbar\nu = eE_0 X_{12}. \quad (3.62)$$

Taking the results of the previous paragraph and substituting them into Equations 3.53 and 3.54 gives,

$$\nu \cos(\omega t) \exp(-i\omega_0 t) C_2 = i \frac{dC_1}{dt} \quad (3.63)$$

and

$$\nu \cos(\omega t) \exp(-i\omega_0 t) C_1 = i \frac{dC_2}{dt}, \quad (3.64)$$

where  $\omega_0$  is the frequency of the incident light. The incident oscillatory electromagnetic wave onto a two-level atom can be described exactly by equations 3.63, 3.64, and 3.49. A more general solution can now be obtained which calculates the optical excitation of an atom. Starting with the atomic density matrix  $\rho_{ij}$ ,

$$\begin{aligned} \rho_{11} &= |C_1|^2 = \frac{N_1}{N} \\ \rho_{22} &= |C_2|^2 = \frac{N_2}{N} \\ \rho_{12} &= C_1 C_2^* \\ \rho_{21} &= C_2 C_1^* = \rho_{12}^*. \end{aligned} \quad (3.65)$$

Equations of motion for the density matrix follow this form;

$$\frac{d\rho_{ij}}{dt} = C_i \frac{dC_j^*}{dt} + \frac{dC_i}{dt} C_j^*. \quad (3.66)$$

Substituting equations 3.63 and 3.64 into equation 3.66,

$$\frac{d\rho_{22}}{dt} = -\frac{d\rho_{11}}{dt} = -i\nu \cos(\omega t) \{ \exp(i\omega_0 t) \rho_{12} - \exp(-i\omega_0 t) \rho_{21} \} \quad (3.67)$$

$$\frac{d\rho_{12}}{dt} = \frac{d\rho_{21}^*}{dt} = i\nu \cos(\omega t) \exp(-i\omega_0 t) (\rho_{11} - \rho_{22}), \quad (3.68)$$

which are exact solutions for the density matrix. Applying the rotating-wave approximation allows the removal of the cosine oscillating term. The optical Bloch equations are then obtained,

$$\frac{d\rho_{22}}{dt} = -\frac{d\rho_{11}}{dt} = -\frac{1}{2}i\nu \left\{ \exp[i(\omega_0 - \omega)t] \rho_{12} - \exp[-i(\omega_0 - \omega)t] \rho_{21} \right\} \quad (3.69)$$

and

$$\frac{d\rho_{12}}{dt} = \frac{d\rho_{21}^*}{dt} = \frac{1}{2}i\nu \exp\{-i(\omega_0 - \omega)t\} (\rho_{11} - \rho_{22}), \quad (3.70)$$

where  $\omega_0 - \omega$  is the oscillation frequency of the incident light.

After deriving the optical Bloch equations, it is now necessary to derive the rate of atomic excitation so that the equation can be used to estimate the amount of charged  $^{229}\text{Th}$ , during lasing, at any given moment inside a linear Paul ion trap. The first step is to redefine terms;

$$\begin{aligned} \tilde{\rho}_{12} &= \exp\{i(\omega_0 - \omega)t\} \rho_{12} \\ \tilde{\rho}_{21} &= \exp\{-i(\omega_0 - \omega)t\} \rho_{21}. \end{aligned} \quad (3.71)$$

The optical Bloch equations then become

$$\frac{d\tilde{\rho}_{22}}{dt} = -\frac{d\tilde{\rho}_{11}}{dt} = -\frac{1}{2}i\nu (\tilde{\rho}_{12} - \tilde{\rho}_{21}) \quad (3.72)$$

and

$$\frac{d\tilde{\rho}_{12}}{dt} = \frac{d\tilde{\rho}_{21}^*}{dt} = \frac{1}{2}i\nu (\tilde{\rho}_{11} - \tilde{\rho}_{22}) + i(\omega_0 - \omega) \tilde{\rho}_{12}. \quad (3.73)$$

Here,  $\tilde{\rho}_{ij} = \rho_{ij}$ , where the tilde is used for variable consistency. Solutions of the above optical Bloch adaptation take the form of

$$\tilde{\rho}_{ij}(t) = \tilde{\rho}_{ij}(0) \exp(\lambda t). \quad (3.74)$$

The four exact solutions that result from the general solution will be written in matrix form as

$$\begin{bmatrix} -\lambda & 0 & \frac{1}{2}i\nu & -\frac{1}{2}i\nu \\ 0 & -\lambda & -\frac{1}{2}i\nu & \frac{1}{2}i\nu \\ \frac{1}{2}i\nu & -\frac{1}{2}i\nu & i(\omega_0 - \omega) - \lambda & 0 \\ -\frac{1}{2}i\nu & \frac{1}{2}i\nu & 0 & -i(\omega_0 - \omega) - \lambda \end{bmatrix} \begin{bmatrix} \tilde{\rho}_{11}(0) \\ \tilde{\rho}_{22}(0) \\ \tilde{\rho}_{12}(0) \\ \tilde{\rho}_{21}(0) \end{bmatrix} = 0. \quad (3.75)$$

The solutions for  $\lambda$  are obtained from the equation

$$\lambda^2 \{ \lambda^2 + (\omega_0 - \omega)^2 + \nu^2 \} = 0, \quad (3.76)$$

with roots

$$\begin{aligned} \lambda_1 &= 0 \\ \lambda_2 &= i\Omega \\ \lambda_3 &= -i\Omega, \end{aligned} \quad (3.77)$$

where

$$\Omega = \{ (\omega_0 - \omega)^2 + \nu^2 \}^{1/2}. \quad (3.78)$$

The density matrix therefore has solutions

$$\tilde{\rho}_{ij}(t) = \tilde{\rho}_{ij}^{(1)} + \tilde{\rho}_{ij}^{(2)} \exp(i\Omega t) + \tilde{\rho}_{ij}^{(3)} \exp(-i\Omega t). \quad (3.79)$$

The optical Bloch equation solutions are quite complicated when using arbitrary initial conditions. Because of this, special assumptions are used. Firstly, it is assumed that the bunch of atoms are initially all in the ground state, i.e.  $\tilde{\rho}_{22}(0) = \tilde{\rho}_{12}(0) = 0$ . The incident beam of light is considered to be one frequency, no detuning. The term "Rabi oscillations" is now introduced to describe the symmetrical oscillations, with no deviations, between the ground state and the excited state. Equations 3.72 and 3.73 then reduce to

$$\tilde{\rho}_{22}(t) = \sin^2\left(\frac{1}{2}\nu t\right). \quad (3.80)$$

The oscillation frequency  $\nu$ , introduced earlier, is called the "Rabi frequency" and is found as

$$\nu = \frac{eE_0 X_{12}}{\hbar} = \frac{eE_0 \mathbf{e} \cdot \mathbf{D}_{12}}{\hbar}. \quad (3.81)$$

The next step in the process is to add spontaneous emission to the optical Bloch equations, which will be the final step as we will ignore collisional effects. A spontaneous emission variable,  $\gamma_{sp}$ , is added to the optical Bloch equations,

$$\frac{d\tilde{\rho}_{22}}{dt} = -\frac{d\tilde{\rho}_{11}}{dt} = -\frac{1}{2}i\nu(\tilde{\rho}_{12} - \tilde{\rho}_{21}) - 2\gamma_{sp}\tilde{\rho}_{22} \quad (3.82)$$

and

$$\frac{d\tilde{\rho}_{12}}{dt} = \frac{d\tilde{\rho}_{21}^*}{dt} = \frac{1}{2}i\nu(\tilde{\rho}_{11} - \tilde{\rho}_{22}) + i(\omega_0 - \omega - \gamma_{sp})\tilde{\rho}_{12}. \quad (3.83)$$

This produces a steady-state-like equation that can be used to calculate the amount of excited atoms during a continuous incident beam without power or collisional broadening,

$$\tilde{\rho}_{22}(\infty) = \frac{\nu^2/4}{(\omega_0 - \omega)^2 + \gamma_{sp}^2 + (\nu^2/2)}. \quad (3.84)$$

Adding a power broadening term, i.e. adding a FWHM to the linewidth of the excited state, is fairly straightforward. The roots of the 4x4 determinant matrix shown in Equation 3.75 are now

$$\begin{aligned} \lambda_1 &= 0 \\ \lambda_2 &= -\gamma_{sp} \\ \lambda_3 &= -\frac{3}{2}\gamma_{sp} + i\lambda \\ \lambda_4 &= -\frac{3}{2}\gamma_{sp} - i\lambda, \end{aligned} \quad (3.85)$$

where

$$\lambda = (\nu^2 - \frac{1}{3}\gamma_{sp}^2)^{1/2}. \quad (3.86)$$

The excited-state population with a power broadening term now becomes

$$\tilde{\rho}_{22} = \frac{\nu^2/2}{2\gamma_{2p}^2 + \nu^2} = \left\{ 1 - \left[ \cos(\lambda t) + \frac{3\gamma_{sp}}{2\lambda} \sin(\lambda t) \right] \exp\left(-\frac{3\gamma_{sp}t}{2}\right) \right\}. \quad (3.87)$$

Next, collision broadening will be added, which takes into account the broadening of the excited state due to collisions with the buffer gas. The probability  $p(\tau)d\tau$  that an atom does not collide with a gas molecule during flight in the time  $\tau$  to  $\tau + d\tau$  is given by

$$p(\tau)d\tau = \frac{1}{\tau_0} \exp\left(-\frac{\tau}{\tau_0}\right)d\tau, \quad (3.88)$$

where  $\tau_0$  is the mean period of flight without collisions and is given by

$$\frac{1}{\tau_0} = \frac{4d^2N}{V} \left( \frac{\pi k_B T}{M} \right)^{1/2}, \quad (3.89)$$

where  $d$  is the distance between collisions and  $M$  is the atomic mass. If only elastic collisions are considered, then the number of excited states is taken as

$$\tilde{\rho}_{22}(\infty) = \frac{(\gamma/4\gamma_{sp})\nu^2}{(\omega_0 - \omega)^2 + \gamma^2 + (\gamma/2\gamma_{sp})\nu^2}. \quad (3.90)$$

The final step is to merge the decay rate equations with the optical Bloch equations.

$$\tilde{\rho}_{22}(t) = \frac{1}{4}\nu^2 \left\{ \frac{\gamma/\gamma_{sp}}{(\omega_0 - \omega)^2 + \gamma^2} + \frac{\left[ (2\gamma_{sp} - \gamma)/\gamma_{sp} \right] \exp(-2\gamma_{sp}t)}{(\omega_0 - \omega)^2 + (2\gamma_{sp} - \gamma)^2} - 2 \exp(-\gamma t) \right. \\ \left. \frac{\left[ (\omega_0 - \omega)^2 + \gamma(2\gamma_{sp} - \gamma) \right] \cos[(\omega_0 - \omega)t] + 2(\omega_0 - \omega)(\gamma - \gamma_{sp}) \sin[(\omega_0 - \omega)t]}{\left[ (\omega_0 - \omega)^2 + \gamma^2 \right] \left[ (\omega_0 - \omega)^2 + (2\gamma_{sp} - \gamma)^2 \right]} \right\}. \quad (3.91)$$

Equation 3.91 represents the number of excited atoms at a given time while incorporating both power broadening and collision broadening. This equation can be simplified by assuming large

collisional broadening. In this case,  $\gamma \gg \gamma_{sp}$ , which reduces Equation 3.91 to

$$\tilde{\rho}_{22}(t) = \frac{\nu^2 \gamma / 4 \gamma_{sp}}{(\omega_0 - \omega)^2 + \gamma^2} \{1 - \exp(-2\gamma_{sp} t)\}. \quad (3.92)$$

The rate equation associated with equation 3.92 is

$$\frac{d\tilde{\rho}_{22}(t)}{dt} = R - 2\gamma_{sp}\tilde{\rho}_{22}(t), \quad (3.93)$$

where

$$R = \frac{\frac{1}{2}\nu^2\gamma}{\left[(\omega_0 - \omega)^2 + \gamma^2\right]}. \quad (3.94)$$

### 3.4.2 <sup>229</sup>Th Atomic Excitation

Although the goal of this thesis is to construct an ion trap to trap <sup>229</sup>Th<sup>3+</sup> for the purposes of nuclear laser excitation, learning the realm of atomic excitation is a necessary step which allows the confirmation of the quantity of <sup>229</sup>Th<sup>3+</sup> ions inside the linear Paul trap. A two-step excitation will be performed on <sup>229</sup>Th<sup>3+</sup> shown by Figure 3.9. First, a 690 nm laser will be used to drive the 5F<sub>5/2</sub> ground state to the 6D<sub>5/2</sub> excited state. From there, a 1:8 branching ratio will occur to the 5F<sub>5/2</sub> ground state and the 5F<sub>7/2</sub> state, respectively [20]. Therefore, a 984 nm laser is needed to drive the 5F<sub>7/2</sub> state back to the 6D<sub>5/2</sub> state. This allows ions inside the ion trap to be driven at the 6D<sub>5/2</sub> state where the photons emitted during transition changes will be detected and processed to estimate the amount of ions inside the trap.

In the previous section, the number of excited atoms and the rate of excitation was derived using the optical Bloch equations. The next step is to evaluate the number of excitations inside a linear Paul ion trap. Because the previous section was purely an intensive derivation, a table of variables is provided. Table 3.1 lists the important variables that will be used in this section in order to evaluate the number excitations. Starting with equation 3.92, if it is assumed that as soon as the 690 nm state decays it gets reexcited immediately, then the Rabi frequency  $\nu$  can be evaluated using the inverse of the lifetime of the 690 nm atomic state, which is  $\tau = 0.676 \mu s$  [20]. This leads to a Rabi frequency of  $1.48 \times 10^6$  Hz, which is consequently equal to the

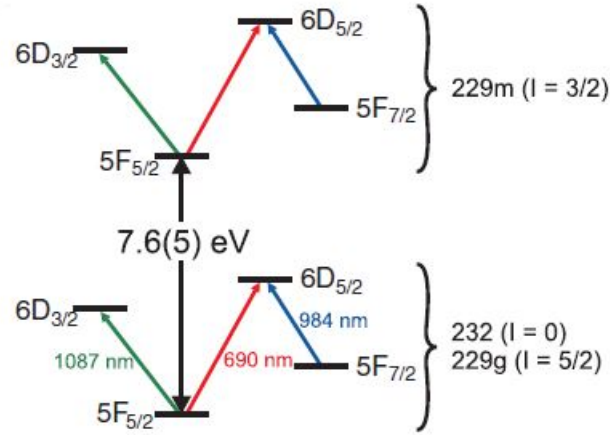


Figure 3.9: The atomic transitions of  $^{229}\text{Th}^{3+}$  that will be used to confirm the contents of the linear Paul ion trap by driving the  $6D_{5/2}$  state [20].

spontaneous decay coefficient  $\gamma_{sp}$ . However, the total decay coefficient  $\gamma$  is found by  $\gamma = \gamma_{sp} + \gamma_{coll}$ , where  $\gamma_{coll}$  is the collision coefficient. The collision coefficient can be evaluated by  $1/\tau_0$ , where  $\tau_0$  is the time between helium gas collisions for a single thorium atom. Using the Stopping and Range of Particles in Matter (SRIM) particle transport code, the range of  $^{229}\text{Th}^{3+}$  in helium gas at different pressures was obtained. Using this information in the SIMION particle transport program, the microscopic cross section for a collision between thorium and helium was estimated to be approximately  $3 \times 10^{-18} \text{ m}^2$ . Assuming an ideal gas at 3 mTorr, the density of helium is  $6.57 \times 10^{-4} \text{ g/m}^3$ . The energy of a  $^{229}\text{Th}$  atom entering the linear Paul trap is approximately 20 eV which corresponds to a velocity of approximately 130 m/s. Using this information, the time between gas collisions can be approximated as  $\tau_0 = 25 \text{ } \mu\text{s}$ , which gives  $\gamma_{coll} = 38,575 \text{ Hz}$ . Because  $\gamma$  is not much larger than  $\gamma_{sp}$ , then we must look at equation 3.91. Equation 3.91 becomes trivial to solve at saturation, or when  $t \rightarrow \infty$ . Also, assume that the fluctuation of the laser is negligible. The exponential terms disappear and equation 3.91 reduces to

$$\tilde{\rho}_{22}(t) = \frac{\nu^2}{4\gamma\gamma_{sp}}. \quad (3.95)$$

Substituting the appropriate quantities, the linear Paul ion trap should contain approximately 24.4% excited atoms at the saturation point. If there are 100,000 ions inside this ion trap, approx-



imately 24,400 will be excited. Because the Rabi frequency  $\nu$  is  $1.48 \times 10^6$  Hz, the number of photons released during saturation should be approximately  $3.61 \times 10^{10}$  photons/s.

| Optical Bloch Equations Variables |                                                |
|-----------------------------------|------------------------------------------------|
| Variable                          | Description                                    |
| $E_0$                             | Incident electric field strength               |
| $\omega$                          | Frequency of incident light                    |
| $\omega_0$                        | Frequency of desired atomic state              |
| $\nu$                             | Rabi frequency                                 |
| $\gamma$                          | Total decay coefficient                        |
| $\gamma_{sp}$                     | Spontaneous decay coefficient                  |
| $\gamma_{coll}$                   | Collision coefficient                          |
| $R$                               | Excitation Rate                                |
| $X_{12}$                          | $x$ -component of the transition dipole moment |
| $D_{12}$                          | Transition dipole moment                       |

Table 3.1: List of variables used in the optical Bloch equations and their descriptions.

### 3.4.3 $^{229m}\text{Th}$ Estimations

Because little is known about laser/nucleus interactions, minimal theoretical calculations exist. There has been effort to extend the optical Bloch equations into the nuclear realm, however that will not be a focus of this thesis as much is unknown about this type of interaction. Instead, attention will be brought to an estimation made by observing a transition between the same Nilsson states as the ground and isomeric state of  $^{229}\text{Th}$ .  $^{233}\text{U}$  possesses these same Nilsson states,  $3/2^+[631]$  and  $5/2^+[633]$ , and can be used to estimate the half-life of  $^{229m}\text{Th}$ , assuming that the internal conversion channel is closed. The  $\gamma$  transition from the  $3/2^+$  ground state to the  $5/2^+$  excited state is a magnetic M1 transition where the transition strength  $B(M1)$  has been experimentally found to be  $0.0060 \mu_N^2$  [11], where  $\mu_N$  is the Bohr magneton. This allows the estimation of the  $^{229m}\text{Th}$  half-life using the same Nilsson transition from  $^{233}\text{U}$ . The energies between the two Nilsson states can be scaled by  $E_\gamma^3$ . The  $^{233}\text{U}$  transition is well known with a half-life of 0.12 ns and an energy of 312 keV. If the energy of the thorium transition is assumed to be 8.1 eV, the half-life of the transition should be approximately 5 hours. There have been studies that have experimentally determined the half-life of the isomeric state of neutral  $^{229m}\text{Th}$ , which decays by

internal conversion [34]. However, the successful trapping and detection of  $^{229m}\text{Th}^{3+}$ , which cannot decay by IC, has eluded scientists.

Using the most recent accepted value of  $8.28 \pm 0.17$  eV for  $^{229m}\text{Th}$ , a corresponding wavelength of the transition of 146.73 nm to 152.80 nm is calculated. A typical broad linewidth laser in the 690 nm wavelength range will have a linewidth of 0.5 nm. If it is assumed that the UV laser will have this linewidth, it can be used to narrow the uncertainty further for the energy determination of the transition to an estimated  $\pm 0.03$  eV. A narrow linewidth laser can then be used to further reduce the uncertainty and hone in on the actual energy of the transition. Narrow linewidth lasers can have a linewidth of 0.0001 nm or smaller, which would take an unrealistic amount of time to scan the 146.73 nm to 152.80 nm region for the energy of the transition without a further decrease in uncertainty. A great amount of work has gone into reducing the uncertainty of the energy of this transition. However, more work needs to be done before a prototype nuclear clock can be constructed.

# Chapter 4

## Experimental Setup

The different studies in this thesis were designed to aid with the construction and understanding of a linear Paul ion trap for the direct detection of  $^{229m}\text{Th}$ . The majority of the equipment used was constructed and assembled on-site at Lawrence Livermore National Laboratory (LLNL). The studies described in this chapter include testing the following: newly constructed equipment and assemblies, modification of an RF oscillation circuit in order to stabilize the frequency, creation and testing of a gas stopping cell which utilizes a two-dimensional RF carpet as the beam initializer, designing one of the world's largest linear Paul ion traps, and a laser optics setup for atomic excitation. In addition, other studies will be described throughout this chapter.

To begin, this chapter will give an overall view of the apparatus constructed for this thesis. This will enable the reader to have a general understanding of the process of the creation and trapping of  $^{229}\text{Th}^{3+}$  ions. Simulations that were written using SIMION to model the electric fields and the trajectory of charged particles inside those electric fields inside the apparatus will follow. The construction of each individual piece of equipment will then be described. Once the reader has a grasp of the physical aspects of the apparatus, the physics behind the individual pieces will be explained. This includes: the initial formation of a positively charged ion beam, how radiofrequency quadrupoles and mass separation quadrupoles work, the experimentation behind the orifice plate, the physics behind the ion trapping system, and individual studies to ensure each section is working as expected. The laser optics system for atomic excitation of  $^{229}\text{Th}^{3+}$  will

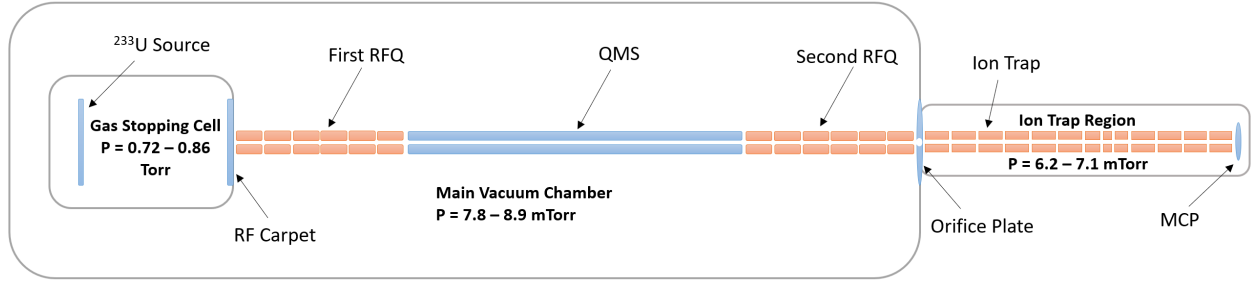


Figure 4.1: A conceptual two-dimensional view of the apparatus used in the work for developing a custom-made beam line and custom-made linear Paul ion trap for trapping and laser interrogation of  $^{229}\text{Th}^{3+}$ . Outlined are the three different vacuum chambers (gas stopping cell, main vacuum chamber, and the ion trapping region) with their respective operating ultrahigh purity helium gas pressures. Also shown are the individual sections of the apparatus which will be described in detail in the text.

follow ending with the electronics schematic used for both the microchannel plate detector (ion detection) and the photomultiplier tubes (photon detection).

## 4.1 Overview of the Apparatus

A two-dimensional conceptual view of the apparatus used for this thesis is shown in Figure 4.1. The first section incorporates a gas stopping cell filled with ultrahigh purity helium gas with an electroplated  $10\ \mu\text{Ci}$   $^{233}\text{U}$  source at the front. This source will  $\alpha$ -decay emitting a  $^{229}\text{Th}$  ion of various charge states with an energy of approximately 84 keV and an  $\alpha$  particle with energy of 4.8 MeV. The source plate that the  $^{233}\text{U}$  source is electroplated on is biased to +400 V which attracts all negatively-charged particles and applies an accelerating force to all positively-charged particles that inhibits them from stopping inside the helium-filled gas stopping cell. A series of 20 aluminum rings follow which are biased to continue accelerating all positively charged ions forward. A two-dimensional funnel system, called the RF carpet, sits at the end of the gas stopping cell and is used to initialize the beam. Initially, the beam consists of all positively charged particles that make it to the end of the gas stopping cell. The gas stopping cell is a separate vacuum chamber that sits inside the main vacuum chamber which will be discussed in Section 4.3.2. A gas jet created by a pressure differential between the gas stopping cell and the main vacuum chamber forcefully ejects the ions and helium gas at an energy of 570 - 580 eV. A positive bias

voltage on the extraction hole inhibits the positively-charged particles from colliding with the RF carpet during the ejection process. Following the gas stopping cell is an RF quadrupole (RFQ) ion guide system which is used to gather all particles after ejection from the gas stopping cell and guide them into the linear Paul ion trap. In the middle of the RFQ ion guide system lies a mass separation quadrupole (QMS) which is used to eject all charged particles out of the beam except  $^{229}\text{Th}^{3+}$ . At the end of the ion guide system lies the orifice plate (OP) which separates the ion trapping chamber that contains the continually-loading linear Paul ion trap and the microchannel plate detector (MCP). Because the ion trap is continually loading, the "half-life" of the ion trap becomes an important quantity and will be defined here. The half-life of the ion trap is simply the amount of time it takes for half of the trapped ions to escape the potential well created for ion trapping. Because there are residual gases inside the ion trapping region that will interact with  $^{229}\text{Th}^{3+}$  such as  $\text{N}_2$  and  $\text{O}_2$ , charge exchange can occur between the thorium ions and the residual gases which results in thorium stealing electrons. This causes the contents of the ion trap to become contaminated and eventually neutralize which will cause ejection from the ion trap. The longer the half-life of the ion trap, the greater the possible number of ions trapped before an equilibrium is attained. Each section of the apparatus is described in greater detail in Section 4.3.

## 4.2 Simulations

SIMION is a software package that allows users to model electric fields and trajectories of charged particles in those fields. Due to the way SIMION simulates adjustable voltages, quadrupole electrodes were modeled in diagonal pairs in order to successfully and easily model RF that is  $180^\circ$  out of phase which creates containment of the beam. Each electrode pair is modeled with its own DC voltage bias as well as applied peak-to-peak RF voltage. The overall quadrupole field which is created corresponds to Equation 3.32.

The experimental setup was modeled using SIMION starting from outside of the gas stopping cell. A geometry file was first created before adding electric fields, RF, gas collisions, and particles. The resulting geometry created by the geometry file is shown in Figure 4.2. The geometry file consists of both RFQs, QMS, orifice plate, the ion trap and the MCP. The multiple scripts that



Figure 4.2: The SIMION geometry used to model the trajectory of the  $^{229}\text{Th}^{3+}$  ions. Starting from the right, shown is the first radiofrequency quadrupole (RFQ), mass separation quadrupole (QMS), second RFQ, orifice place, ion trap, and the microchannel plate (MCP) detector.

input the DC bias, RF voltages and frequencies, and gas collisions were written in Lua. If no gas pressure is modeled, the simulation will run as if it were in vacuum, and trapping would be impossible. Modeling a gas accurately in SIMION requires approximating the collisional cross section between the gas particle (helium) and the ion chosen ( $^{229}\text{Th}^{3+}$ ). The SRIM (stopping and range of ions in matter) software was used to calculate the projected stopping range in a multitude of different helium gas pressures. By modeling the projected stopping range of  $^{229}\text{Th}^{3+}$  in the different pressures of helium that correspond to the pressures applied to the experimental apparatus, the three different cross sections needed were obtained using this process: first, SRIM was used to calculate the projected stopping range of  $^{229}\text{Th}^{3+}$  in different helium gas pressures corresponding to the different sections of the apparatus. These projected stopping ranges were noted. An example output can be viewed in Figure 4.3 for a main vacuum chamber helium pressure corresponding to 8 mTorr. One of SIMION's example programs simulates the stopping range of a particle in a gas if the cross section is known. Therefore, because the cross section of  $^{229}\text{Th}$  and helium atoms is not well known experimentally, the cross section was changed in SIMION until the projected stopping range of  $^{229}\text{Th}^{3+}$  in helium gas matched that of SRIM. This

cross section was then applied to the SIMION simulations created for this thesis. After the modeling of the proper DC voltage gradients with applied RF voltages and the helium gas, the ions were introduced. The introduction of ions requires many parameters, including the energies of the individual ions (can be randomized between two parameters), the angle of emission (can be randomized between two parameters), the mass and charge state, the number of ions, whether the ions are released as a beam or one at a time, and the coulomb forces associated if a beam is chosen. The energies of ions used were approximated by investigating the stopping power of the orifice plate, which is covered in Section 4.5.2. Using this setup, the efficiency of each section of the experiment could be simulated. The efficiency of the mass separation quadrupole was also inspected by changing the mass and charge of the ions to arbitrary values in an attempt to view the ejection/transmission process. In order to approximate the ion trapping efficiency, many different parameters had to be examined. This includes examining different values of the following: RF voltages, RF frequencies, initial ion velocities, beam intensities, potential well depths, and shapes of the potential well. Figure 4.4 shows the  $^{229}\text{Th}^{3+}$  ion traces for a simulation of 10  $^{229}\text{Th}^{3+}$  ions grouped together as a beam. As the ions slow, the beam becomes more confined due to less energy being being lost during helium interactions. This becomes apparent from Figure 4.4a. During the transition phase through the orifice plate which separates the main vacuum chamber from the ion trapping region, the beam diameter expands slightly because of the absence of RF in that region. When the beam enters the ion trap, the 10  $^{229}\text{Th}^{3+}$  ions are successfully trapped in the center which can be viewed in Figure 4.4c.



| Ion Energy | dE/dx Elec. | dE/dx Nuclear | Projected Range | Longitudinal Straggling | Lateral Straggling |
|------------|-------------|---------------|-----------------|-------------------------|--------------------|
| 3.99999 eV | 3.144E-10   | 1.411E-09     | 536.86 mm       | 47.26 mm                | 32.57 mm           |
| 4.49999 eV | 3.334E-10   | 1.533E-09     | 564.22 mm       | 50.02 mm                | 34.50 mm           |
| 4.99999 eV | 3.515E-10   | 1.650E-09     | 589.62 mm       | 52.58 mm                | 36.31 mm           |
| 5.49999 eV | 3.686E-10   | 1.765E-09     | 613.38 mm       | 54.97 mm                | 38.01 mm           |
| 5.99999 eV | 3.850E-10   | 1.875E-09     | 635.74 mm       | 57.22 mm                | 39.62 mm           |
| 6.49999 eV | 4.008E-10   | 1.983E-09     | 656.89 mm       | 59.35 mm                | 41.15 mm           |
| 6.99999 eV | 4.159E-10   | 2.088E-09     | 676.98 mm       | 61.38 mm                | 42.61 mm           |
| 7.99999 eV | 4.446E-10   | 2.291E-09     | 714.45 mm       | 65.27 mm                | 45.35 mm           |
| 8.99999 eV | 4.716E-10   | 2.485E-09     | 748.94 mm       | 68.84 mm                | 47.88 mm           |
| 9.99999 eV | 4.971E-10   | 2.672E-09     | 780.98 mm       | 72.15 mm                | 50.24 mm           |
| 10.9999 eV | 5.213E-10   | 2.853E-09     | 810.96 mm       | 75.25 mm                | 52.45 mm           |
| 11.9999 eV | 5.445E-10   | 3.028E-09     | 839.21 mm       | 78.16 mm                | 54.54 mm           |
| 12.9999 eV | 5.667E-10   | 3.199E-09     | 865.94 mm       | 80.92 mm                | 56.53 mm           |
| 13.9999 eV | 5.881E-10   | 3.364E-09     | 891.36 mm       | 83.55 mm                | 58.41 mm           |
| 14.9999 eV | 6.088E-10   | 3.526E-09     | 915.61 mm       | 86.05 mm                | 60.22 mm           |
| 15.9999 eV | 6.287E-10   | 3.684E-09     | 938.83 mm       | 88.45 mm                | 61.95 mm           |
| 16.9999 eV | 6.481E-10   | 3.839E-09     | 961.12 mm       | 90.76 mm                | 63.61 mm           |
| 17.9999 eV | 6.669E-10   | 3.990E-09     | 982.57 mm       | 92.98 mm                | 65.21 mm           |
| 19.9999 eV | 7.030E-10   | 4.284E-09     | 1.02 m          | 97.27 mm                | 68.24 mm           |
| 22.4999 eV | 7.456E-10   | 4.636E-09     | 1.07 m          | 102.29 mm               | 71.77 mm           |
| 24.9999 eV | 7.859E-10   | 4.975E-09     | 1.11 m          | 106.96 mm               | 75.06 mm           |
| 27.4999 eV | 8.243E-10   | 5.301E-09     | 1.16 m          | 111.32 mm               | 78.13 mm           |
| 29.9999 eV | 8.610E-10   | 5.617E-09     | 1.19 m          | 115.43 mm               | 81.03 mm           |
| 32.4999 eV | 8.961E-10   | 5.923E-09     | 1.23 m          | 119.32 mm               | 83.78 mm           |
| 34.9999 eV | 9.299E-10   | 6.220E-09     | 1.27 m          | 123.03 mm               | 86.40 mm           |
| 37.4999 eV | 9.626E-10   | 6.510E-09     | 1.30 m          | 126.56 mm               | 88.89 mm           |
| 39.9999 eV | 9.941E-10   | 6.792E-09     | 1.33 m          | 129.95 mm               | 91.28 mm           |
| 44.9999 eV | 1.054E-09   | 7.336E-09     | 1.39 m          | 136.47 mm               | 95.78 mm           |
| 49.9999 eV | 1.111E-09   | 7.858E-09     | 1.45 m          | 142.52 mm               | 99.97 mm           |
| 54.9999 eV | 1.166E-09   | 8.360E-09     | 1.50 m          | 148.19 mm               | 103.89 mm          |
| 59.9999 eV | 1.218E-09   | 8.844E-09     | 1.55 m          | 153.54 mm               | 107.58 mm          |
| 64.9999 eV | 1.267E-09   | 9.313E-09     | 1.60 m          | 158.60 mm               | 111.08 mm          |
| 69.9999 eV | 1.315E-09   | 9.767E-09     | 1.64 m          | 163.42 mm               | 114.41 mm          |
| 79.9999 eV | 1.406E-09   | 1.064E-08     | 1.73 m          | 172.64 mm               | 120.63 mm          |
| 89.9999 eV | 1.491E-09   | 1.147E-08     | 1.81 m          | 181.12 mm               | 126.37 mm          |
| 99.9999 eV | 1.572E-09   | 1.226E-08     | 1.88 m          | 189.01 mm               | 131.70 mm          |
| 110 eV     | 1.649E-09   | 1.302E-08     | 1.95 m          | 196.40 mm               | 136.70 mm          |
| 120 eV     | 1.722E-09   | 1.375E-08     | 2.01 m          | 203.37 mm               | 141.41 mm          |
| 130 eV     | 1.792E-09   | 1.445E-08     | 2.08 m          | 209.99 mm               | 145.88 mm          |
| 139.999 eV | 1.860E-09   | 1.513E-08     | 2.14 m          | 216.28 mm               | 150.13 mm          |
| 149.999 eV | 1.925E-09   | 1.580E-08     | 2.19 m          | 222.31 mm               | 154.18 mm          |
| 159.999 eV | 1.988E-09   | 1.644E-08     | 2.25 m          | 228.08 mm               | 158.07 mm          |
| 169.999 eV | 2.049E-09   | 1.706E-08     | 2.30 m          | 233.63 mm               | 161.81 mm          |
| 179.999 eV | 2.109E-09   | 1.767E-08     | 2.35 m          | 238.99 mm               | 165.41 mm          |
| 199.999 eV | 2.223E-09   | 1.885E-08     | 2.44 m          | 249.34 mm               | 172.23 mm          |
| 224.999 eV | 2.358E-09   | 2.025E-08     | 2.56 m          | 261.48 mm               | 180.18 mm          |
| 249.999 eV | 2.485E-09   | 2.158E-08     | 2.66 m          | 272.78 mm               | 187.59 mm          |
| 274.999 eV | 2.607E-09   | 2.286E-08     | 2.76 m          | 283.38 mm               | 194.55 mm          |
| 299.999 eV | 2.723E-09   | 2.408E-08     | 2.85 m          | 293.39 mm               | 201.12 mm          |
| 324.999 eV | 2.834E-09   | 2.525E-08     | 2.94 m          | 302.89 mm               | 207.35 mm          |
| 349.999 eV | 2.941E-09   | 2.638E-08     | 3.03 m          | 311.94 mm               | 213.30 mm          |
| 374.999 eV | 3.044E-09   | 2.748E-08     | 3.11 m          | 320.60 mm               | 218.98 mm          |
| 399.999 eV | 3.144E-09   | 2.854E-08     | 3.19 m          | 328.91 mm               | 224.44 mm          |
| 449.999 eV | 3.334E-09   | 3.057E-08     | 3.34 m          | 344.92 mm               | 234.75 mm          |
| 499.999 eV | 3.515E-09   | 3.250E-08     | 3.48 m          | 359.83 mm               | 244.39 mm          |
| 549.999 eV | 3.686E-09   | 3.433E-08     | 3.61 m          | 373.83 mm               | 253.46 mm          |
| 599.999 eV | 3.850E-09   | 3.608E-08     | 3.74 m          | 387.05 mm               | 262.05 mm          |

Figure 4.3: SRIM output using incident  $^{229}\text{Th}$  particles in a helium gas pressure of 8 mTorr which corresponds to the pressure inside the main vacuum chamber.

The results given by SIMION were used as a guideline for the construction of the beam line and the ion trap and will be discussed in the appropriate subsections in Section 4.5.



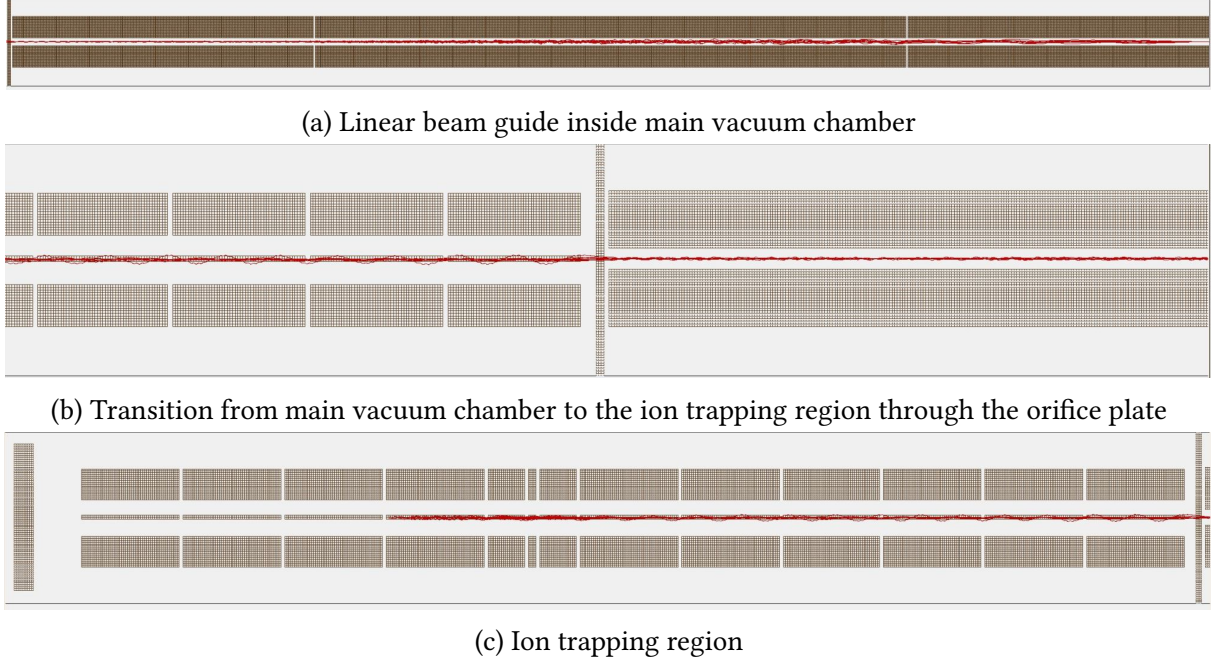


Figure 4.4: Particle traces of  $10^{229}\text{Th}^{3+}$  ions in a SIMION simulation split into (a) the beam path which contains (from right to left) the first RFQ, mass separation quadrupole, and the second RFQ, (b) the transition phase through the orifice plate (middle plate) which separates the main vacuum chamber (right side) and the ion trapping region (left side), and (c) the ion trapping region during trapping operation using the vane ion trap showing the orifice plate (right) and the MCP used for detection (left).

## 4.3 Construction

### 4.3.1 Radiofrequency Oscillators

Before describing the construction of each section of the experimental apparatus, the creation of the RF utilized by each section will be described. The RF oscillators drive the RF in the RF carpet, both RF quadrupoles (RFQ), mass separation quadrupole (QMS), and the ion trap. The RF oscillators used in this experiment were manually constructed due to the cost of commercially available oscillators existing upwards of \$20,000. By manually constructing the oscillators, the total cost was approximately \$300 per oscillator. This circuit design is highly based on [21], which utilizes two different circuits. However, the automatic feedback adjustment was removed in order to drive the RF at a set amplitude. An LC oscillating circuit, also called the tank circuit, generates the high voltage RF. A negative feedback automatic gain circuit (AGC) controls the maximum

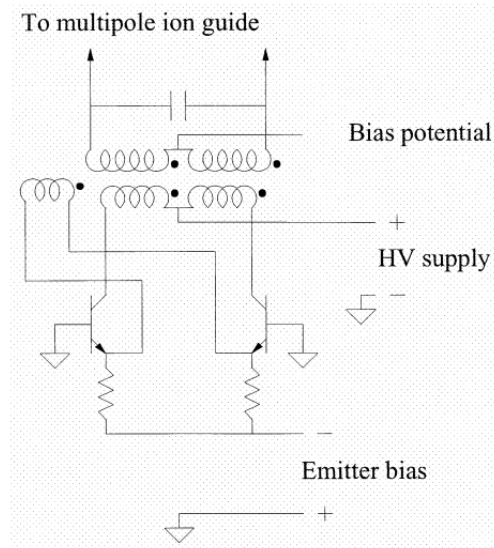


Figure 4.5: A simplified version of the oscillator circuit showing the transistor oscillator circuit (bottom) coupled to the LC tank circuit (top) separated by the coupling of the coils shown [21].

amplitude of the RF, which is where the feedback adjusting occurred. A simplified schematic of the oscillator is shown in Figure 4.5.

The ability to achieve oscillation starts with a small feedback coil and a pair of  $200\ \Omega$  series resistors that are connected between the transistor emitters. This converts the feedback voltage into a feedback current. The amplitude of the RF signal is set by the DC value of the emitter current which is set by the voltage applied across the emitter resistors. This feedback signal is  $180^\circ$  out of phase with the oscillator so that amplitude of oscillation increases until the emitter current is reduced to zero.

The main coil design sets the resonant frequency of the RF oscillator, which can later be changed by adding capacitance in parallel to the output of the RF signal. The main coil consists of a primary coil and a secondary coil wound around a 5.08 cm diameter PVC pipe so the coil keeps its shape. A feedback coil is wound on top of the primary coil. The tank circuit coil (secondary coil) is wound using 18-gauge copper magnet wire while the transistor coil (primary coil) and feedback coil are wound using 16-gauge copper magnet wire. It is important to achieve a high coupling between the primary and secondary coil or else the tank circuit transistors will attempt to oscillate at a frequency different than the frequency set by the inductance and capacitance of the tank circuit. The number of turns of the secondary coil determines the inductance of the

system. A small layer of epoxy was added to the wires to hold them in place while a 0.1 mm thick sheet of Mylar was wrapped around to insulate the coils. One extra loop exists simply as an extra feedback to the AGC circuit, which is wound over the center of the other coils. Figure 4.8 shows the main oscillator coil during construction. Each oscillator that was constructed has a slightly different resonant frequency due to the uncertainty of winding the coils by hand.

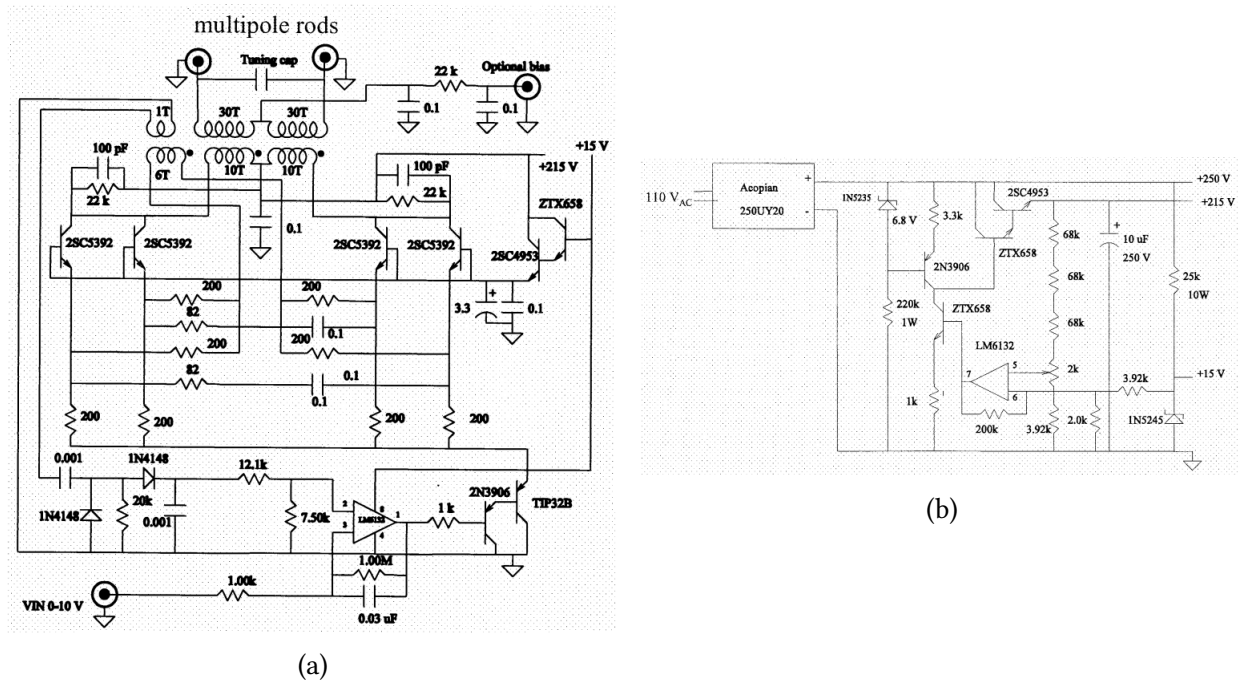


Figure 4.6: The schematics of the (a) oscillator and AGC circuit and (b) power supply circuit starting from the Acopian power supply. Resistance values are in ohms and capacitance values are in microfarads unless otherwise marked [21].

A considerable amount of heat is generated in this design due to the full power requirements of 40 W at 500 kHz and 30 W at 1 Mhz. Four parallel transistors are used to dissipate the majority of the heat. Metal heat sinks are attached to each transistor in the oscillator with a cooling fan blowing on each of them. This allows continuous operation without overheating the transistors. The required voltage per transistor is 215 V, 200 V between the collectors and bases and 15 V to power the op-amp.

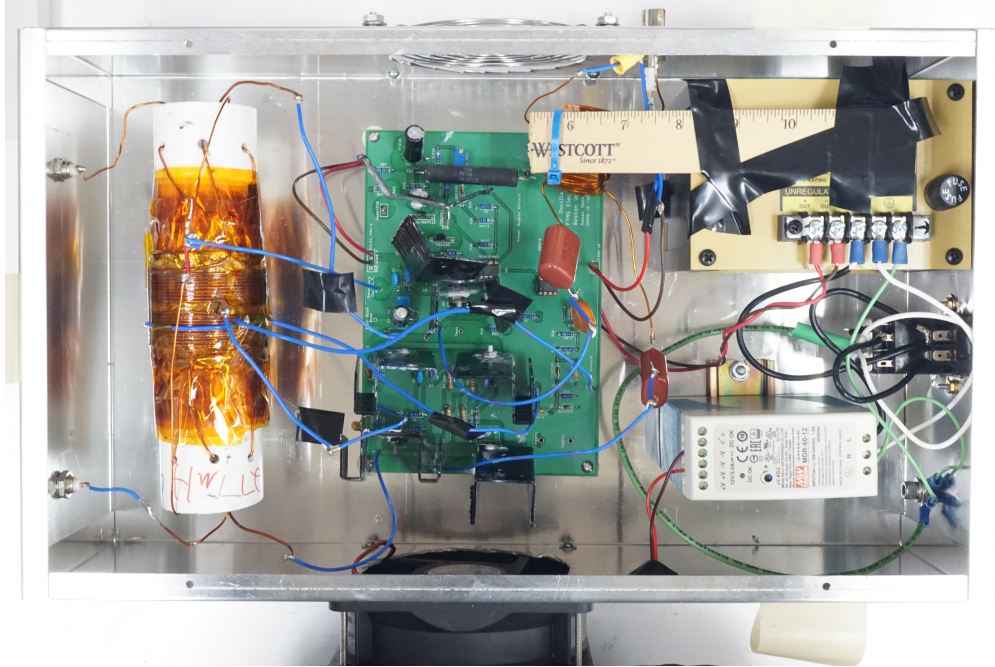


Figure 4.7: The inside of a finished RF oscillator showing both coils, circuit board, Acopian power supply, and cooling system. The ruler shown serves as a mount for the small coil to inhibit shorting.

Figure 4.7 shows the inside of a finished RF oscillator. The resonant frequencies for the RF carpet, first RFQ, QMS, second RFQ, and ion trap are 1.021 MHz, 729 kHz, 559 kHz, 516 kHz, and 942 kHz, respectively. The peak-to-peak voltage of each segment of the beam line also varies. For the RF carpet and both RFQs, the value of the RF voltage is set to the optimum value that day which is dependent on room temperature. For the RF carpet, the value varies between 100 V and 120 V, the first RFQ varies between 300 and 400 V, and the second RFQ varies between 120 and 180 V. The QMS has a set value of 230 V based on Section 3.3.2. The RF on the ion trap is set based on the maximum number of ions trapped inside, which is approximately 370 V. This value is dependent on room temperature. The RF voltages for each segment are constantly monitored using PR-55 BK Precision voltage probes and read through an oscilloscope. Due to the components of the RF oscillators heating up during use, a warm-up time is necessary of approximately 2 hours. This ensures that the components reach an equilibrium temperature and constant shifts in RF voltage will not occur.





Figure 4.8: A photograph of the main coil of the RF oscillator during winding. The secondary coil has been wound underneath and the primary coil is shown after epoxy has been applied. The next step is a layer of Mylar to keep the coils from shorting together. Following Mylar, the final step is the winding of the feedback coil

### 4.3.2 Main Vacuum Chamber

The main vacuum chamber shown in Figure 4.9 houses the gas stopping cell, both RFQs, the QMS, and the orifice plate which separates the main chamber from the ion trapping region. The outside dimensions of the main chamber are 73-inch length  $\times$  20-inch width  $\times$  11.5-inch height with 0.75-inch thick walls. The vacuum chamber has four turbomolecular pumps attached. The first turbomolecular pump located at the front of the main vacuum chamber is an Agilent-Varian TV 551 (551 L/s pump rate). Two Pfeiffer-Balzer TPH/TPU 330 (330 L/s pump rate) are attached directly on both sides of the chamber. A Pfeiffer TSU 261 (210 L/s pump rate) small turbomolecular pump is also attached to one side. Each turbomolecular pump has an Edwards 10/10i scroll pump attached to it as the final stage of pumping. There is also an Edwards 10/10i scroll pump attached directly to the vacuum chamber which is required to rough out (initialize the vacuum in an ultra-high vacuum system) the chamber before initial operation of the turbomolecular pumps. Between the main vacuum chamber and each turbomolecular pump is a gate valve that can be open or

closed at will to manipulate the pressure inside the main vacuum chamber. Multiple BNC and SHV feedthroughs are located throughout the chamber in order to apply DC potentials and RF voltages where necessary. Also attached to the main chamber is a residual gas analyzer (RGA). This is used to check the partial pressures of various gases inside and can also be used for leak testing. Vacuum lines are attached from the chamber to the gas stopping cell and the ion trapping region to assist in pumping to high vacuum when the experiment is inactive.

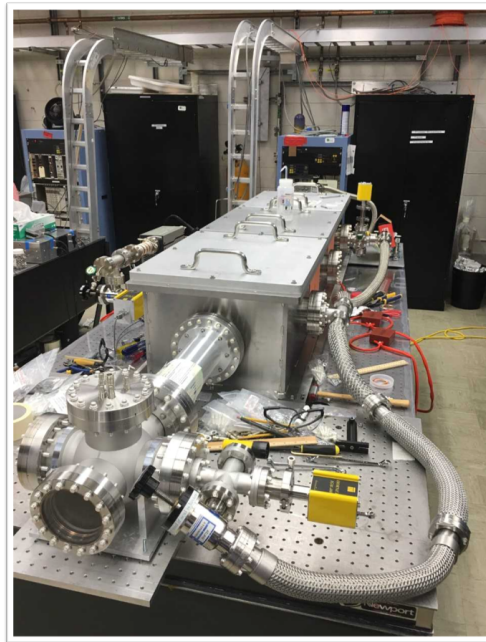


Figure 4.9: Picture of the main chamber with the ion trap section added. Note the placement of the vacuum lines that attach the main chamber to both the gas stopping cell and the ion trapping region.

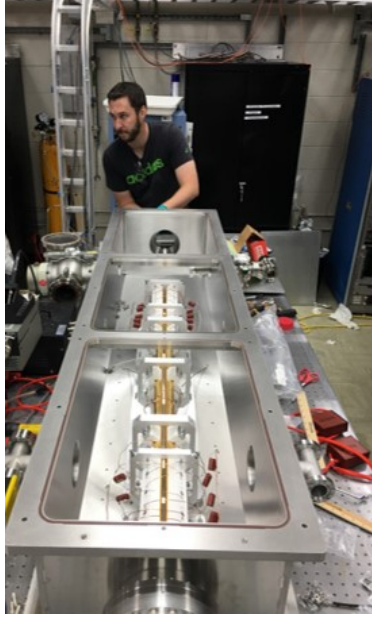


Figure 4.10: The inside of the main vacuum chamber which shows the RFQs and QMS. Pictured is the author, Aaron Hellinger.

### 4.3.3 Gas Stopping Cell

The gas stopping cell is the location at which the  $^{229}\text{Th}^{3+}$  beam begins formation. The gas stopping cell, which is a separate vacuum chamber from the main vacuum chamber, houses an electroplated  $10\ \mu\text{Ci}\ ^{233}\text{U}$  source which is screwed into an aluminum plate. Following the source plate are 20 aluminum rings on 1 cm thickness spaced 1 cm apart. At the end of the aluminum rings lies a two-dimensional RF funnel called the RF carpet. The gas stopping cell vacuum chamber is a cylindrical chamber that includes two 10-inch outer-diameter flanges on each end. The front flange has an inner diameter of 6 inches which is sealed by the RF carpet while the back of the gas stopping cell is sealed by a cap flange. The sides of the gas stopping cell include a BNC feedthrough that allows biasing of the source plate and subsequent aluminum rings and a helium injection site that administers helium into the gas stopping cell. During inactivity, the gas injection site also doubles as a way to pump high vacuum on the gas stopping cell. The cylindrical cell sits on top of a stand that raises the gas stopping cell's center to approximately 15 cm.

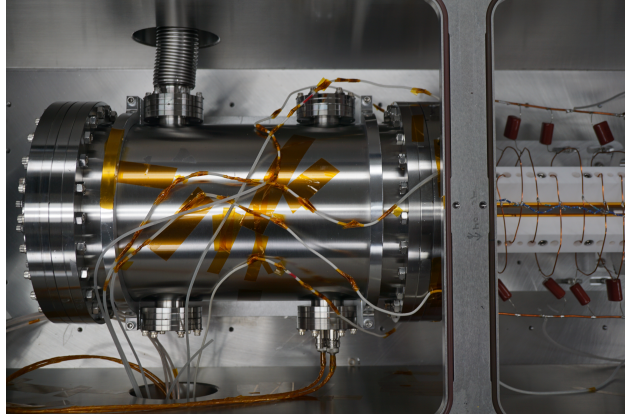


Figure 4.11: The gas stopping cell chamber inserted into the main vacuum chamber. To the right of the gas stopping cell is the first RFQ that collects the ions exiting out of the gas jet created by the extraction hole in the RF carpet. Also shown is the helium injection line (top) and the BNC feedthroughs (bottom).

The aluminum source plate and 20 subsequent aluminum rings are spaced 1 cm apart separated by ceramic spacers to prevent shorting. The ring electrodes are connected by  $1\text{ M}\Omega$  resistors in order to apply a voltage gradient to the aluminum rings. Directly after the final aluminum ring is the RF carpet which gathers all positively charged particles and focuses them into a 1 mm extraction hole. The design of the RF carpet is based on [64]. The RF carpet is placed flush between the last aluminum ring and the 10-inch flange to ensure proper sealing which creates a vacuum differential between the gas stopping cell and the main vacuum chamber. The RF carpet consists of gold-plated aluminum rings of two different sizes, thirteen 2.5 mm thick rings of decreasing diameters followed by seventy-four 0.25 mm thick rings of decreasing diameters. The RF carpet utilizes RF as well as a DC potential gradient in order to gently guide the ions toward the 1 mm extraction nozzle. The DC gradient section of the circuit consists of thirteen  $10\text{ M}\Omega$  resistors which connect the outer, larger electrodes and seventy-four  $2\text{ M}\Omega$  resistors which connect the smaller, inner electrodes. Capacitors are needed to block the DC current from passing into the RF current. The capacitance chosen is  $10\text{ }\mu\text{F}$  per ring. The extraction nozzle is an individually biased solid circle electrode with a small 1 mm drilled hole in which RF is not applied.



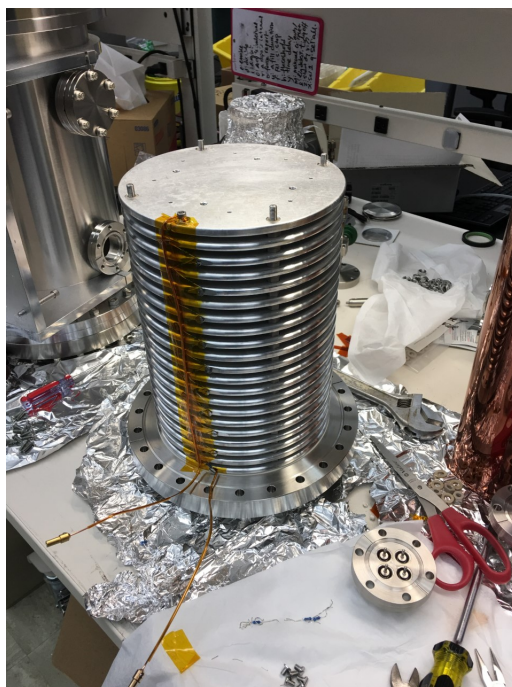


Figure 4.12: The inside of the gas stopping cell showing the 20 aluminum rings connected with  $1\text{ M}\Omega$  resistors, wrapped in Mylar, to create a voltage gradient to accelerate positively charged ions after decay from  $^{233}\text{U}$ .



Figure 4.13: Inside the rings of the gas stopping cell showing the RF carpet that is used to initially concentrate all positively-charged ions from the decay of  $^{233}\text{U}$  into a beam.

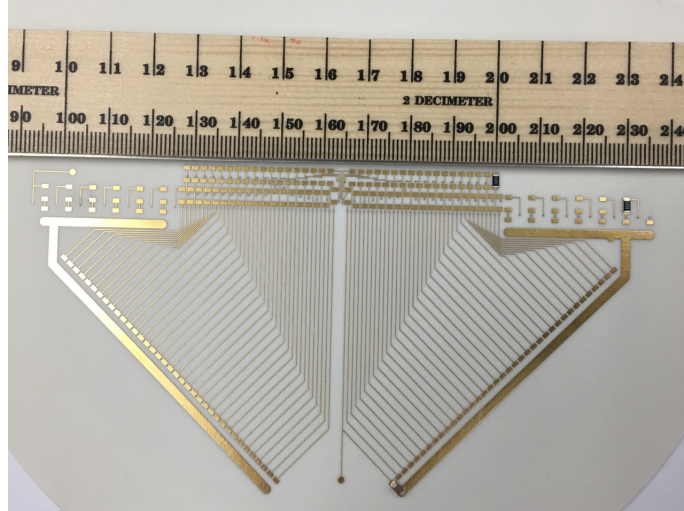


Figure 4.14: The electronic circuit of the RF carpet with a ruler for scale. The resistors used to create the DC voltage gradient are placed at the top, horizontal section. The capacitors used to separate the DC current from the RF current are placed on the diagonal sections.

#### 4.3.4 The Beam Line

The beam line consists of the first RFQ, QMS, the second RFQ, and then the orifice plate. Each RFQ pole consists of a 1-ft long, 2.29-cm diameter gold-coated aluminum rod that is separated into 6 segments spaced 1 mm apart. Each section is connected by a resistor to create a resistor network that applies a voltage gradient for acceleration/deceleration. The value of the resistance between each section of an RFQ is  $760\text{ k}\Omega$ . A  $5\text{ M}\Omega$  potentiometer is attached to the resistance network on the outside of the main vacuum chamber to fine-tune the voltage gradient of the RFQs. In order to apply RF voltage to the RFQs, copper wires are attached. Since the diagonal rods must be  $180^\circ$  out of phase (see Section 3.3), the copper wires are connected to different pairs of subsequent diagonal electrodes using capacitors that separate the RF current from the DC current. Figure 4.15 shows the wiring of the second RFQ, which is identical to the first RFQ.

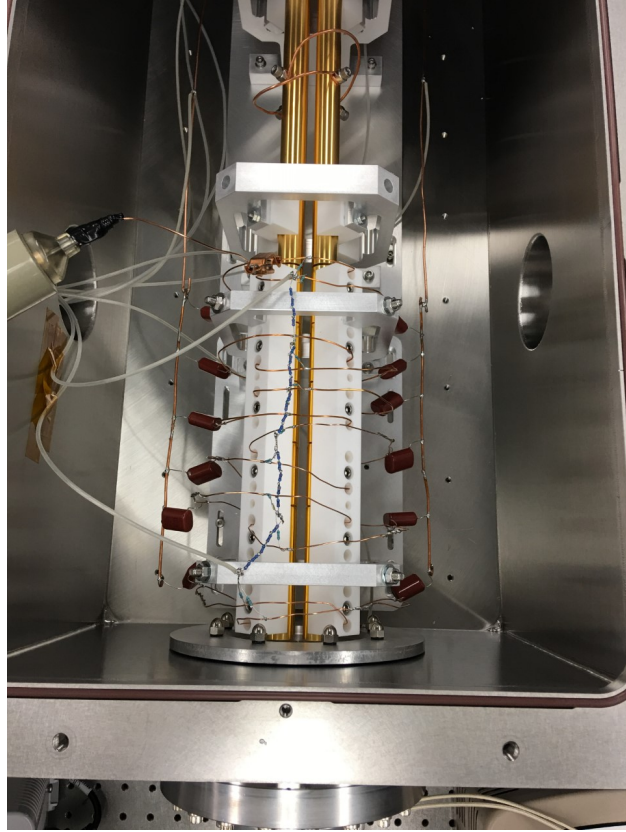


Figure 4.15: Picture of the second RFQ showing the soldered wiring inside the main vacuum chamber. Above the RFQ is the QMS. Below the RFQ is the orifice plate which separates the vacuum from the main vacuum chamber and the ion trapping region.

The QMS consists of four 2-foot non-segmented gold-plated aluminum rods. The QMS is 2 feet long to ensure that the ions spend ample time inside to increase the efficiency of mass selection. Each pair of diagonal rods are floated to a specific voltage difference that corresponds to  $U = 20 \text{ V}$  (see Section 3.3.2 for QMS theory). Since the QMS is created with solid rods, no voltage gradient can be applied. However, RF voltages are still needed to contain the ions inside the quadrupole. Only two capacitors are needed to filter the DC current from the RF since the rods are not segmented. From Section 3.3, the experimentally-determined optimum  $r/r_0$  value for quadrupoles is between 1.128 and 1.148. For the quadrupoles that have been created for this thesis, the radius  $r$  for each rod is 11.48 mm and the distance from the center to an electrode  $r_0$  is 8.3 mm. This sets the  $r/r_0$  value to be 1.38, which is greater than the experimentally determined optimum values from [18]. This error is due to a manufacturing error of the polyetheretherketone (PEEK) material that holds the RFQ rods.

The orifice plate is an aluminum disk with a hole in the center that was initially used to create a pressure gradient between the main vacuum chamber and the ion trapping region. Multiple plates were created with a variety of hole sizes. This was done to control the pressure gradient between the two regions. The orifice plate is biased to a voltage that allows the continuation of the acceleration of the thorium beam, usually 1 volt less than the last electrode on the second RFQ. The bias on the orifice plate can also be increased so that it stops the thorium beam. This can be used to measure the energy of the incoming ions.

#### **4.3.5 Linear Paul Ion Trap**

The two linear Paul ion trap designs were chosen for their large viewing solid angles as well as increased trapping capacity in comparison to other traps such as the ring electrode trap, planar trap, and even smaller linear Paul ion traps. These ion traps work by creating a potential well that, with the help of helium gas, oscillates ions in the ion trap until they lose their linear momentum and are trapped at the bottom of a potential well. During the course of this thesis, two linear Paul ion trap designs were used. The construction of both of these designs will be explained here. The goal of each design is to create an ion trap that maximizes the capacity of the ion trap without sacrificing solid angle for laser interrogation purposes. A typical linear Paul ion trap is constructed to trap few ions, in a line, in an attempt to view with a CCD camera as well as to laser interrogate solo ions for spectroscopy [65; 66]. Some ion traps have been constructed to test the limits of capacity of a linear Paul ion trap to trap  $10^3$  ions [67]. The ion traps constructed for this work are further testing the capacity using a larger linear Paul ion trap design.

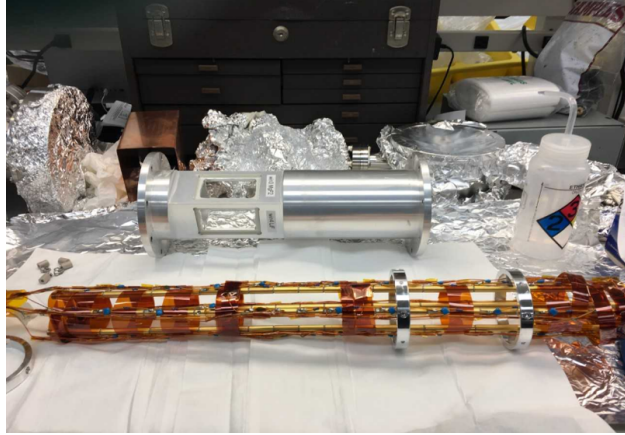


Figure 4.16: The first ion trap design completed before inserting it inside the housing (shown in the back). Mylar was wrapped around the resistors and capacitors to block a possible shorting route to the housing, which is grounded.

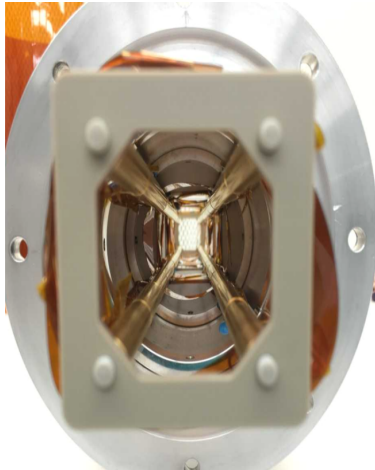


Figure 4.17: A view of the first ion trap design looking down the beam path. Ceramic rods are threaded through the center of the electrodes which hold them in place.

The first ion trap design utilized segmented, gold-coated, aluminum rods similar to both RFQs. This trap used two different lengths of segmented rods all with 4 mm radius. The 2-inch long pieces created a steeper voltage gradient which forced the ions toward the center of the trap. The middle of the trap consisted of five shorter pieces that were 1-inch in length. The smaller pieces help smooth out the voltage gradient which keep perturbations during the trapping process to a minimum. While using this trap design, a helium injection line was attached to the trapping region in order to operate the ion trap in helium pressures between 1 mTorr and 10 mTorr while maintaining the main vacuum chamber pressure between  $10^{-6}$  and  $10^{-5}$  Torr. Since cylindrical



quadrupole electrodes were used, quadrupole theory can still be applied and the experimental  $r/r_0$  value of the quadrupole can be calculated. The  $r/r_0$  value for this trap was calculated to be 0.352, where the optimum value is approximately 1.128. This led to weak RF fields near the center of the trap which consequently decreased the half-life of the trap considerably. Therefore it was concluded that a new ion trap design was necessary to progress.

The current trap design sways away from the typical cylindrical rod ion traps. Thin, rectangular electrodes called vanes are used in place of the cylindrical rods. Because vanes are so thin, they can be placed closer to the center of the ion trap without sacrificing viewing solid angle. This corrects the small  $r/r_0$  value that plagued the previous ion trap. However, elliptical electrode quadrupole theory no longer applies, so the experimental  $r/r_0$  values do not apply. Figure 4.18 shows a cross section of the model of the entire trap split down the beam path. This trap utilizes three different vane lengths. Each vane is 1 mm thick. The first four and last six electrodes are 50.8 mm in length. The longer vanes create a steeper voltage gradient that forces the ions toward the center of the trap. Instead of the middle having 5 smaller electrodes, this design utilizes only three middle electrodes. The two electrodes adjacent to the center electrode are 19 mm in length while the middle electrode is 4.3 mm in length. The smaller electrodes create a smoother voltage gradient that helps contain the ions near the center with minimum perturbations. The helium injection also changed with the design of the new trap. It was noted that the pressure gradient between the main vacuum chamber and the ion trapping region was creating a gas jet that was oriented toward the main vacuum chamber. Therefore the ion injection into the trapping region was removed and the pressure of the ion trap and the vacuum chamber was nearly equalized by replacing the orifice plate with the one with the largest hole of 8 mm diameter.

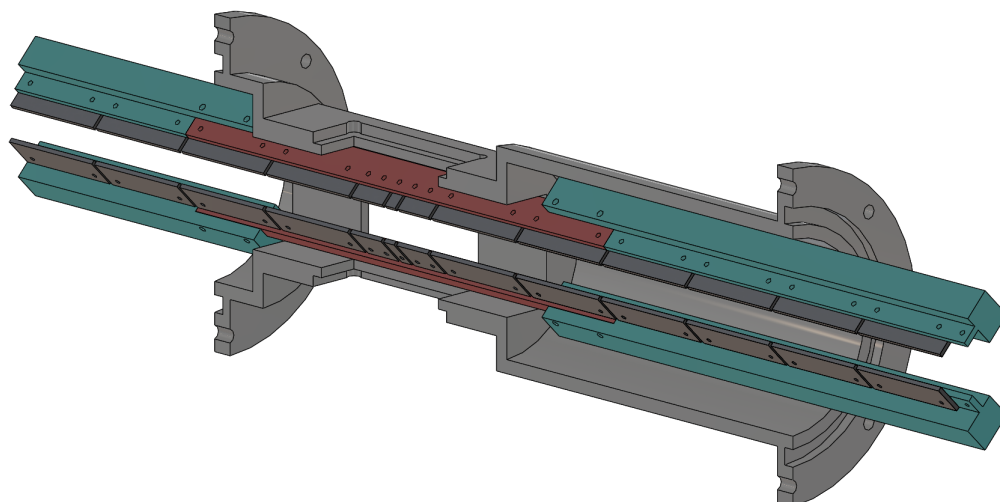


Figure 4.18: Cross section of the current ion trap design split down the beam path. Note the three different sizes of electrodes that allow the transition of a steeper voltage gradient to accelerate/decelerate the incoming  $^{229}\text{Th}^{3+}$  to a smoother gradient which traps the ions in the center.

Towards the end of the research that this thesis covers, a cryopump was attached to the end of the ion trapping region. This was due to a hypothesis that the half-life of the ion trap was suffering from an overabundance of water vapor outgassing inside the chamber which fails to be pumped out while the only operational pump is the 210 L/s small turbomolecular pump. This specific pump is a CTI Cryo-torr 4F cryopump that pumps out 1,100 L/s of water vapor while at a temperature of 20 K. A cryopump works by condensing gases into a separate chamber which then removes them from the ion trapping region. However, helium does not condense until approximately 4 K and will not be pumped out. Figure 4.19 shows a picture of the cryopump attached at the back end of the ion trapping region. The results of the addition of the cryopump will be described in the following chapter.



Figure 4.19: Picture of the cryopump attached to the end of the ion trapping region.



## 4.4 Helium Injection Manifold

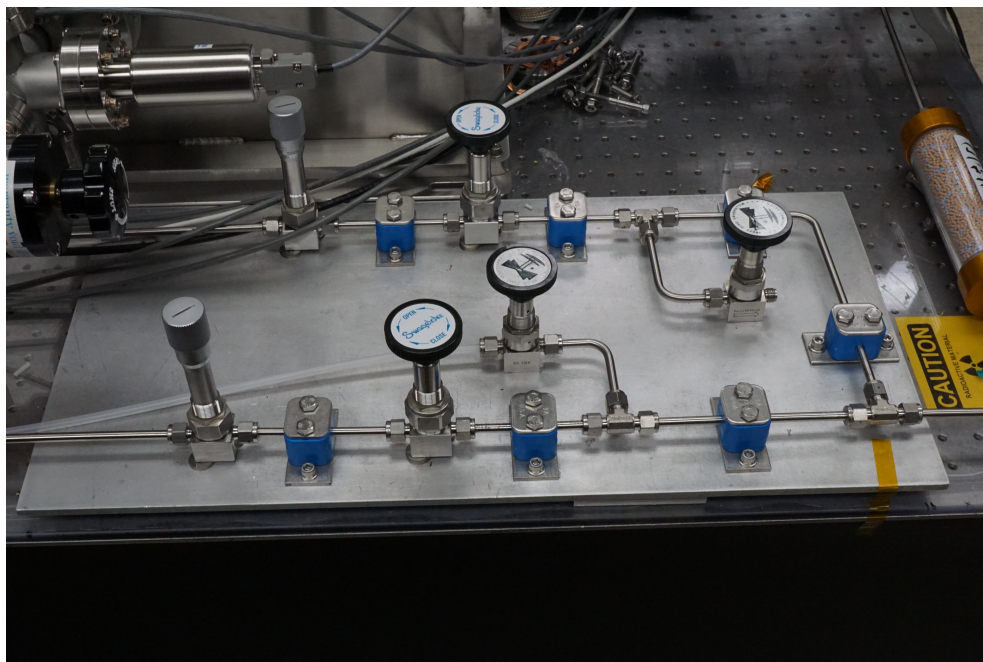


Figure 4.20: Photo of the helium gas injection system. The top of the injection system leads into the gas stopping cell. The bottom leads to the ion trapping region. The right valve is a bleed valve that is used to purify the helium injection line. The middle left valve is the leak checker line attached to flexible plastic tube which was used to check for leaks across the entire apparatus.

The helium injection line starts from a bottle of ultrapurity helium and enters into a LabClear Refillable Gas Filter RGF-250 molecular filter which further purifies the helium gas. From there, the gas line splits off into four different routes. Figure 4.20 shows the gas injection system. The top valve is attached to an on/off valve with another valve that permits adjustment of the amount of helium inserted into the gas stopping cell. The bottom gas line works identically, but leads to the ion trapping section. The right middle valve is used to purge the line of any air to ensure only helium gas is inserted. The left middle valve has a long plastic tube attached and is used as a vacuum leak-testing line.

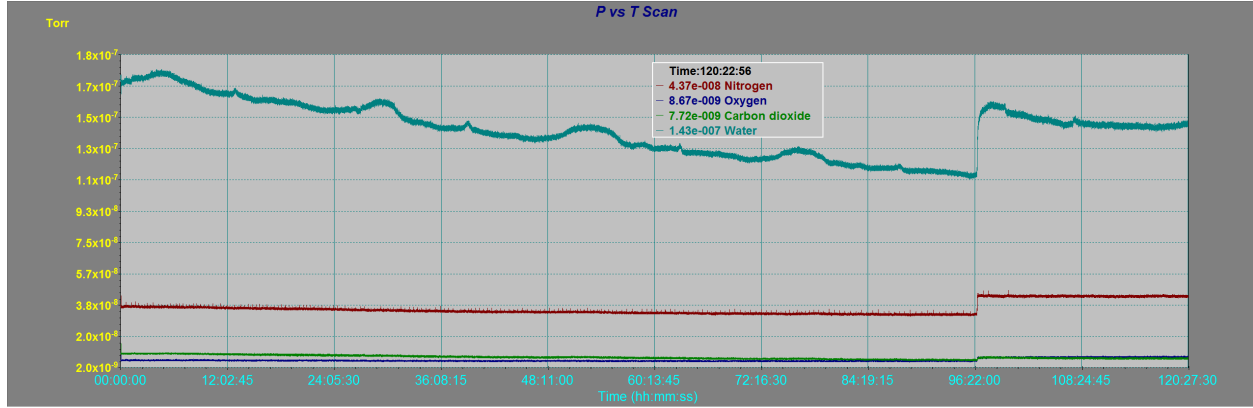


Figure 4.21: A 5-day RGA spectrum showcasing the gas response inside the main vacuum chamber as pumping and outgassing occurs. The increases of pressure are likely due to pockets of water vapor being pumped out. At hour 96, both side turbomolecular pumps were shut to view the response.

## 4.5 Experimentation

In this section, the physics of each section of the apparatus will be described. In addition, this section will entail each individual study performed that assisted in the understanding of the thorium ion trajectory as well as the goal of this research, which is to construct a linear Paul ion trap to aid with the detection of  $^{229m}\text{Th}$ .

### 4.5.1 The Main Vacuum Chamber

As previously stated, the main vacuum chamber houses the gas stopping cell, first RFQ, QMS, second RFQ, and the orifice plate. To keep the number of unwanted interactions to a minimum, the partial pressures of residual gases must be kept to a minimum. The goal is to pump the main vacuum chamber to a pressure of at least  $10^{-8}$  Torr while the experiment is inactive. Figure 4.21 shows a pressure versus time spectrum of five days worth of pumping with every turbomolecular pump operational. Pockets of water vapor outgas occasionally and attribute to increases in partial pressure. This plot also shows that water vapor is the major undesirable gas inside the main vacuum chamber. Figure 4.22 shows partial pressures of different masses of gases inside the main vacuum chamber. This is an uncalibrated spectrum that was taken at a pressure of  $7.8 \times 10^{-8}$  Torr. The partial pressures related to total pressure is of no consequence. The desired relation is

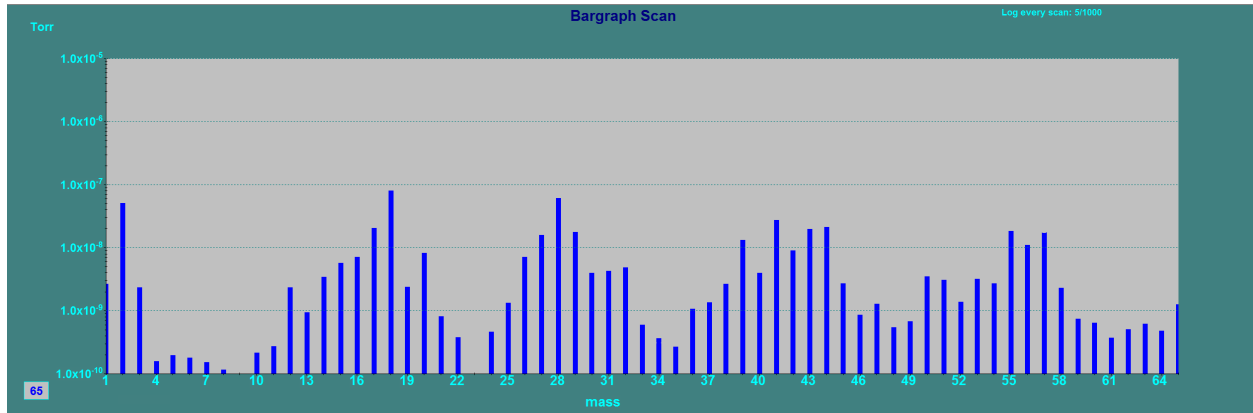


Figure 4.22: Uncalibrated RGA partial pressure spectrum taken at  $7.8 \times 10^{-8}$  Torr. The goal was to view  $N_2$  in relation to  $O_2$ . Since air is 78% nitrogen and 21% oxygen, if the partial pressures inside the chamber matched that ratio then an air leak may exist.

single partial pressures to other single partial pressures such as the partial pressures of  $N_2$  to  $O_2$ . Assuming standard temperature and pressure (STP), this yields a background volumetric particle quantity of  $2.76 \times 10^9$  particles per  $cm^3$ . Multiple background gases exist inside the chamber. The majority of the background gases consist of  $N_2$  (28 amu),  $O_2$  (32 amu),  $H_2O$  (18 amu), and  $CO_2$  (44 amu). The ionization potential of each of these gases are less than the ionization potential for  $^{229}Th^{3+}$ . When a  $^{229}Th^{3+}$  ion interacts with one of these gases, an electron will be transferred to the thorium ion and the resulting effect will decrease the charge state to  $^{229}Th^{2+}$ . This would be detrimental to the goal of this thesis as it would decrease the total amount of  $^{229}Th^{3+}$  ions trapped per second.

During helium injection into the gas stopping cell with all turbomolecular pumps operational, the pressure inside the main vacuum chamber will rise to  $10^{-5}$  Torr. It was learned that using a helium pressure of this magnitude was not attenuating the thorium beam enough as it traveled down the beam line. The ions were passing straight through the ion trap into the MCP, regardless of the DC potential gradients on the RFQs and ion trap. Therefore, it was hypothesized that the thorium ions would be attenuated properly if all turbomolecular pumps were shut during helium injection with only the small 210 L/s turbomolecular pump operational. This increased the pressure of the main chamber to  $10^{-3}$  Torr (more about this in Section 4.5.3). This is the current pressure that is used to attenuate the thorium ions. However this introduced another problem. With the larger turbomolecular pumps shut, the outgassing of water vapor exceeded

the amount of water vapor pumped out of the system. To correct this problem, a cryopump was purchased and attached to the ion trapping region, which will be explained in more detail in Section [4.5.4](#).

### 4.5.2 Gas Stopping Cell

The electroplated  $^{233}\text{U}$  source used inside the gas stopping cell has an activity of 10  $\mu\text{Ci}$ , which corresponds to 370,000 decays per second. This specific source has been previously characterized and measured to eject approximately 38% of decays forward [[60](#)]. Therefore, approximately 140,000 thorium ions of various charge states will be emitted into the gas stopping cell. In this same study, an approximate 0.05%  $^{232}\text{U}$  contamination from this source was measured which corresponds to approximately 200 decays per second into  $^{228}\text{Th}$ . When  $^{233}\text{U}$  decays, the resulting emissions are  $^{229}\text{Th}$  of various charge states with an energy of 84.5 keV as well as an  $\alpha$ -particle with an energy of 4800 keV. The decay scheme for  $^{233}\text{U}$  and  $^{232}\text{U}$  is shown in Figure [4.23](#).



mechanism for the ions during their trajectory towards the RF carpet.

| Location           | Potential(V) |
|--------------------|--------------|
| <b>Gas Stopper</b> |              |
| Source Plate       | 400          |
| Final Ring         | 175          |
| <b>RF Carpet</b>   |              |
| Outermost Ring     | 150          |
| Innermost Ring     | 75           |
| Extraction         | 50           |
| RF                 | 100-120      |

Figure 4.24: Bias potentials of the various electrodes inside the gas stopping cell separated by the gas stopping cell rings and RF carpet rings.

The voltage potentials of the gas stopping cell and RF carpet can be viewed in Figure 4.24. The electroplated  $^{233}\text{U}$  source is biased to a potential of +400 V. This is to immediately accelerate forward all positively charged ions while attracting all electrons and negatively charged particles that are produced during  $^{233}\text{U}$  decay to inhibit any charge recombination. Using SRIM, the projected range of  $^{229}\text{Th}$  and alpha particles in a pressure of 0.8 Torr, corresponding to the operating pressure of the gas stopping cell, were found as 401.98  $\mu\text{m}$  and 576.72  $\mu\text{m}$ . An accelerating voltage gradient of 5.6 V/cm is applied to the rings of the gas stopping cell to continue the ion trajectory forward during the loss of energy due to ion/helium interactions. The ion trajectory and ionization is shown in Figure 4.25 and was simulated using SRIM. This specific simulation used a gas stopping cell operating pressure of 0.8 Torr. The energy of the incoming ions corresponded to the energy of the  $^{229}\text{Th}$  ions after decay (84.5 keV). Without the voltage gradient on the rings of the gas stopping cell, the ions would travel less than 500  $\mu\text{m}$  before stopping in the helium buffer gas. Therefore, the major source of ion mobility inside the gas stopping cell is due to the electric fields.

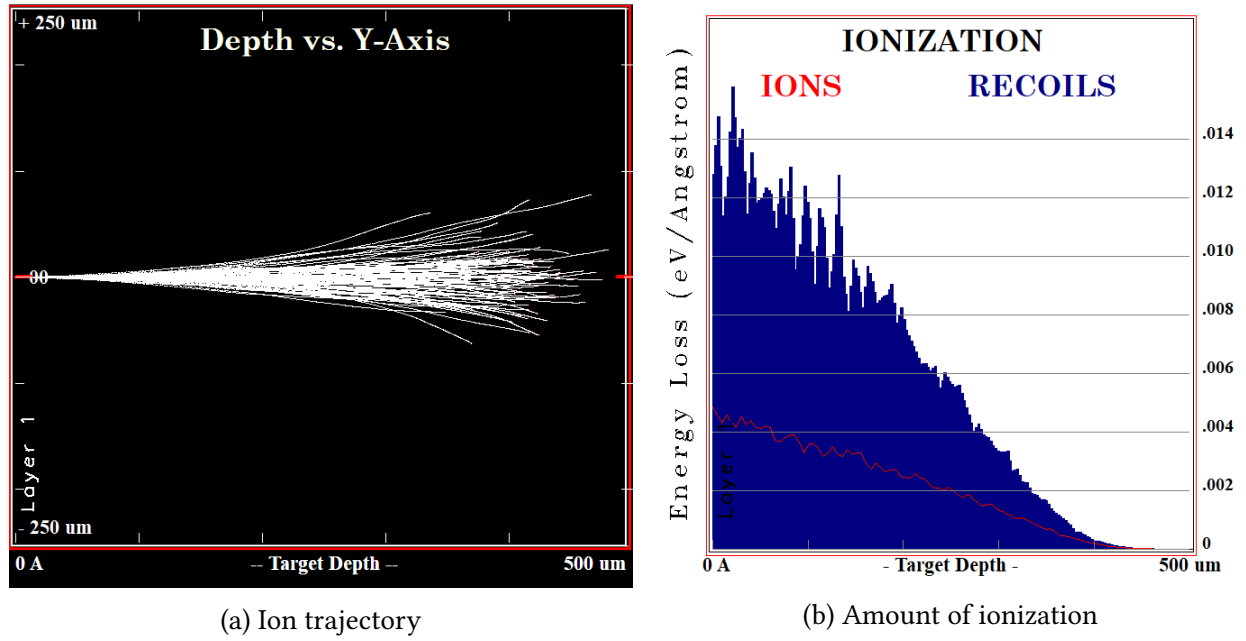


Figure 4.25:  $^{229}\text{Th}$  ion trajectories and amount of ionization modeled in SRIM. The initial energy of the  $^{229}\text{Th}$  is set to 84.5 keV which corresponds to its emission energy after decay from  $^{233}\text{U}$ . The pressure of the helium gas used is 0.8 Torr.

At the end of the gas stopping cell lies the RF carpet which guides the ions towards a 1 mm extraction hole in the center. This is done by using an applied RF voltage and DC voltage gradient to guide the ions toward an extraction hole in the center of the RF carpet. The applied 100-120 peak-to-peak RF voltage is used to inhibit the ions from colliding into the RF carpet during their arrival. The RF applied to the RF carpet works similarly to the quadrupoles. Adjacent rings are  $180^\circ$  out of sync not only to inhibit electrode/ion collisions, but also to assist the guidance toward the center. The 10.5 V/cm voltage gradient applied to the electrodes guides the ions toward the center of the RF carpet. The positive polarity on the electrodes also assists in stopping the ion/electrode collisions on the RF carpet. At the center exists an extraction hole that creates a gas jet that overcomes all forces closer to the center and ejects all positively charged particles that reach the end of the gas stopping cell. The ejected ions are picked up by the first RFQ. The extraction hole is biased to a voltage of +50 V, without applied RF, to stop the ions from colliding with the electrode during the extraction process. Although it is realized other ions are emitted and produced inside the gas stopping cell, the main focus is  $^{229}\text{Th}^{3+}$ . Information on other ions inside the gas stopping cell is out of the scope of this thesis.

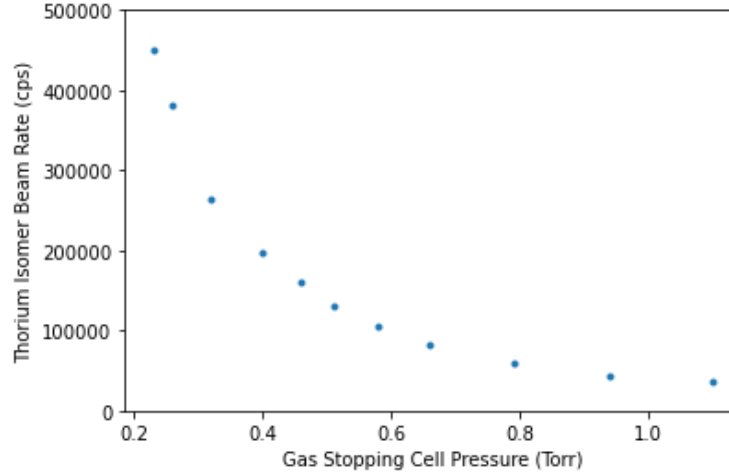


Figure 4.26: Plot of the  $^{229}\text{Th}^{3+}$  beam rate versus the gas stopping cell pressure. The lower gas stopping cell pressure corresponds to lower residual gas pressure inside the main vacuum chamber and ion trapping regions. The higher the gas stopping cell pressure, the greater the attenuation inside the main vacuum chamber and ion trapping regions. Associated uncertainties are represented, however, are unable to be viewed. A beam rate of 50,000 per second has an uncertainty of 223 per second. Likewise, a beam rate of 230,000 per second has an uncertainty of 479 per second.

The pressure of the gas stopping cell has an effect on the  $^{229}\text{Th}^{3+}$  beam rate. Figure 4.26 shows a plot of the beam rate as a function of the gas stopping cell helium pressure. This study was performed using a continuous accelerating potential gradient across the RFQs and the ion trap with the QMS being biased to only allow transmission of  $^{229}\text{Th}^{3+}$ . With the residual gas pressure inside the main vacuum chamber increasing up to 8 mTorr when the gas stopping cell pressure approaches upwards of 1 Torr, the applied accelerating gradient on the RFQs and ion trap will inhibit the ions from stopping before reaching the MCP. Intuitively, it was thought the beam rate should be equivalent for each pressure. There are two different explanations for the lack of this hypothesized response. As the gas stopping cell pressure increases, so does the residual gas inside the main vacuum chamber and the ion trapping region. This increases the chances for multiple interactions that would result in ejection from the beam line.

From Figure 4.26, using a typical operating pressure of 0.8 Torr inside the gas stopping cell, the  $^{229}\text{Th}^{3+}$  rate leaving the gas stopping cell is approximately 90,000 ions per second. Since the source emits  $^{229}\text{Th}$  ions at a rate of 137,000 per second, after traversing through the gas stopping cell, the percentage of  $^{229}\text{Th}^{3+}$  ions exiting the gas stopping cell is 65.7%.



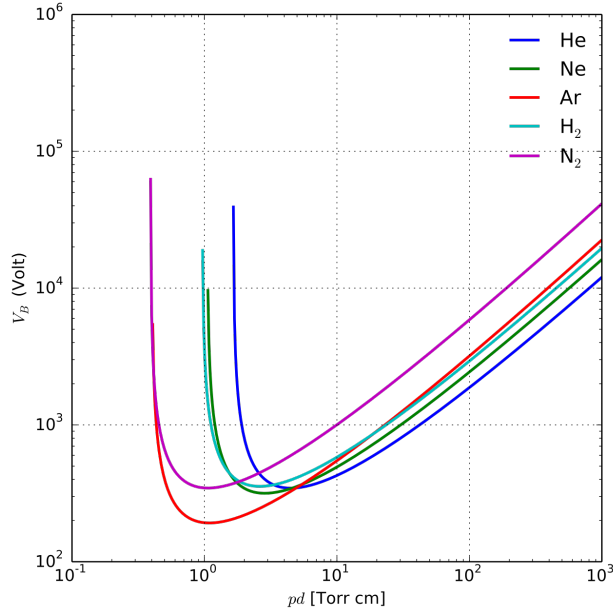


Figure 4.27: Plot of Paschen curves for different gases that show the breakdown voltage as a function of pressure and distance between electrodes. Helium gas is denoted by the blue line.

There also exists a law called Paschen's Law that describes the voltage necessary to create discharge between two electrodes in a variety of different pressures for gases. Figure 4.27 shows the breakdown voltage as a function of pressure and distance between electrodes. The distance between the aluminum electrodes of the gas stopping cell is 1 cm with the diameter of the electrodes being approximately 8 in. The aluminum rings are insulated from the chamber of the gas stopping cell by Kapton sheets which impedes the ability for discharge between the aluminum electrodes and the gas stopping cell chamber wall. At a maximum potential of +400 V with a decreasing potential gradient, sparking should occur between a value of approximately 2.5 - 10 Torr·cm. For example, at a pressure of 0.4 Torr, the distance becomes 6.25 - 25 cm for discharge to occur. For example, this can occur between the source plate and an aluminum ring closer to the RF carpet. Based on the values of Figure 4.26, operating the gas stopping cell at a pressure greater than 0.6 Torr would be ideal as the beam rates with pressures less than 0.6 Torr have an unrealistic magnitude based on the activity of the  $^{233}\text{U}$  source.

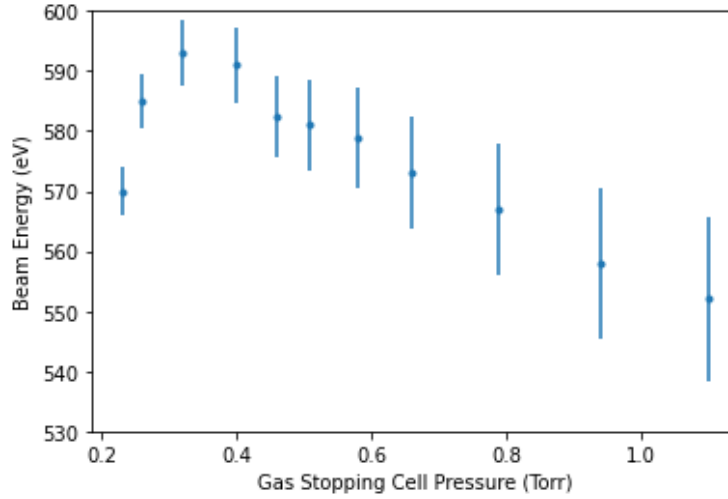


Figure 4.28: The mass separated velocity profile of  $^{229}\text{Th}^{3+}$  exiting the RF carpet extraction hole. The energy of the beam increases due to an increased pressure differential between the gas stopping cell and the main vacuum chamber. The decrease of the energy of  $^{229}\text{Th}^{3+}$  beam is due to the increase of residual gas inside the main vacuum chamber due to the increase of the gas stopping cell pressure. The increase of the associated uncertainty is due to a decreased thorium beam rate as the gas stopping cell pressure increases.

The energy of the ion beam exiting the gas stopping cell is dependent on the gas stopping cell pressure and can be visualized in Figure 4.28. During this study, the RFQs were biased to +50 volts with no voltage gradients to allow a constant velocity after extraction from the gas stopping cell. The QMS was biased to allow only transmission of  $^{229}\text{Th}^{3+}$ . With increasing pressure inside the gas stopping cell, residual pressure also increases in the main chamber. Therefore, the energy of the beam will be skewed slightly due to extra attenuation inside the chamber as the pressure increases. However, the pressure inside the main vacuum chamber never decreases below  $10^{-5}$  Torr. The beam energy approaches a maximum at approximately 0.35 Torr. This is because of the increasing pressure gradient between the main vacuum chamber and the gas stopping cell which creates a gas jet at the extraction nozzle of the RF carpet. The gas flow inside this nozzle will increase as the pressure gradient increases. The charged particles and ions that reach the gas jet created by this pressure gradient will follow the gas flow and are ejected into the main vacuum chamber. The decrease in energy after 0.4 Torr can be attributed to an increase in residual gas inside the main vacuum chamber and ion trapping region as the pressure inside the gas stopping cell increases.

### 4.5.3 The Beam Line

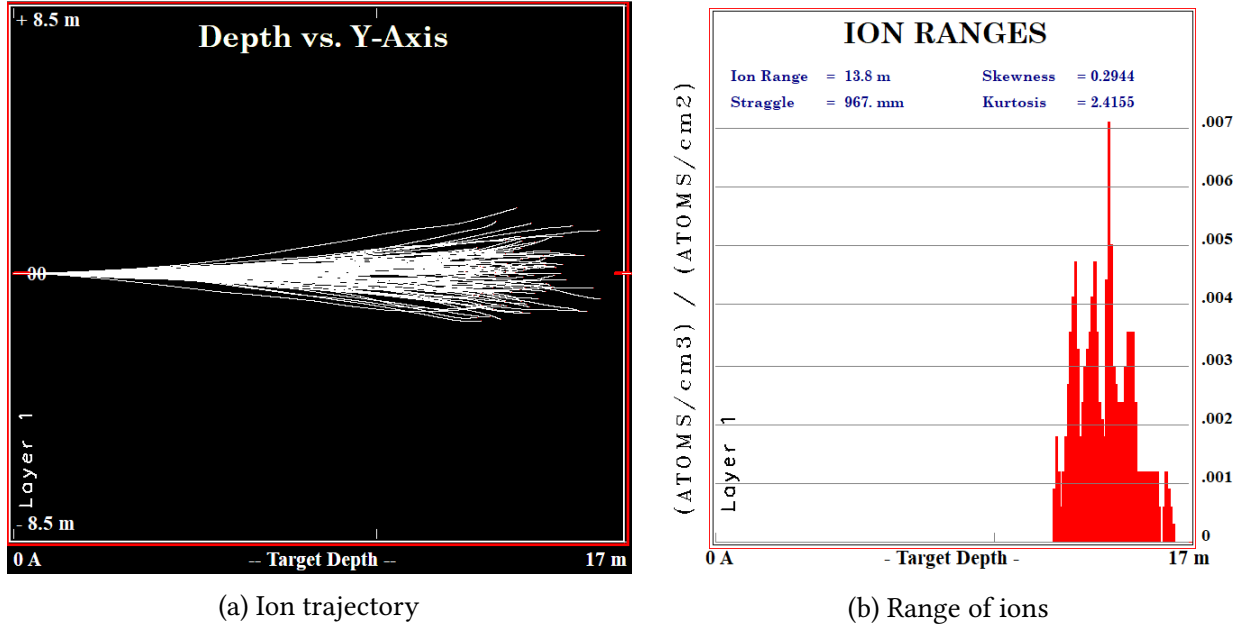


Figure 4.29: The range of ions simulated in SRIM using a helium gas pressure of  $5 \times 10^{-5}$  Torr. The incoming energy of thorium ions was chosen to be 580 eV which corresponds to a gas stopping cell operating pressure of 0.6 Torr.

The beam line, which lies inside the main vacuum chamber, was pressurized to approximately  $10^{-5}$  Torr while operating the cylindrical ion trap. This pressure corresponded to a beam energy of above 550 eV. The gradients of the RFQs were unable to slow the ions sufficiently before entering the ion trap. This was experimentally viewed by the inability to reduce the number of events collected by the MCP while raising the orifice plate voltage to stop the beam no matter the magnitude of the decelerating voltage gradients applied to the RFQs. Using SRIM, the range of  $^{229}\text{Th}$  was simulated and can be viewed in Figure 4.29. The length of the beam line after the RF carpet extraction is approximately 1.7 meters while the projected range of  $^{229}\text{Th}$  ions with an energy of 580 eV is greater than 12 meters. Consequently, it took greater pressures and gradients inside the cylindrical ion trap to slow the ions down. This will be explained in greater detail in Chapter 5. Once the small 210 L/s turbomolecular pump was attached to the chamber, the valves attached to the other three turbomolecular pumps could be shut and the helium pressure of the main vacuum chamber would rise to, at most, 10 mTorr. This allowed proper attenuation of the ion beam inside the main vacuum chamber. Figure 4.30 shows the ion trajectory and ionization of  $^{229}\text{Th}$  ions in a

helium gas pressure of 8 mTorr which corresponds to the operating pressure of the main vacuum chamber. The projected range of ions decreased from 13.8 m to 0.157 m, which is approximately the middle of the first RFQ. Ion mobility becomes dominated by the electric fields created by the beam line and minimal ionization occurs after the QMS.

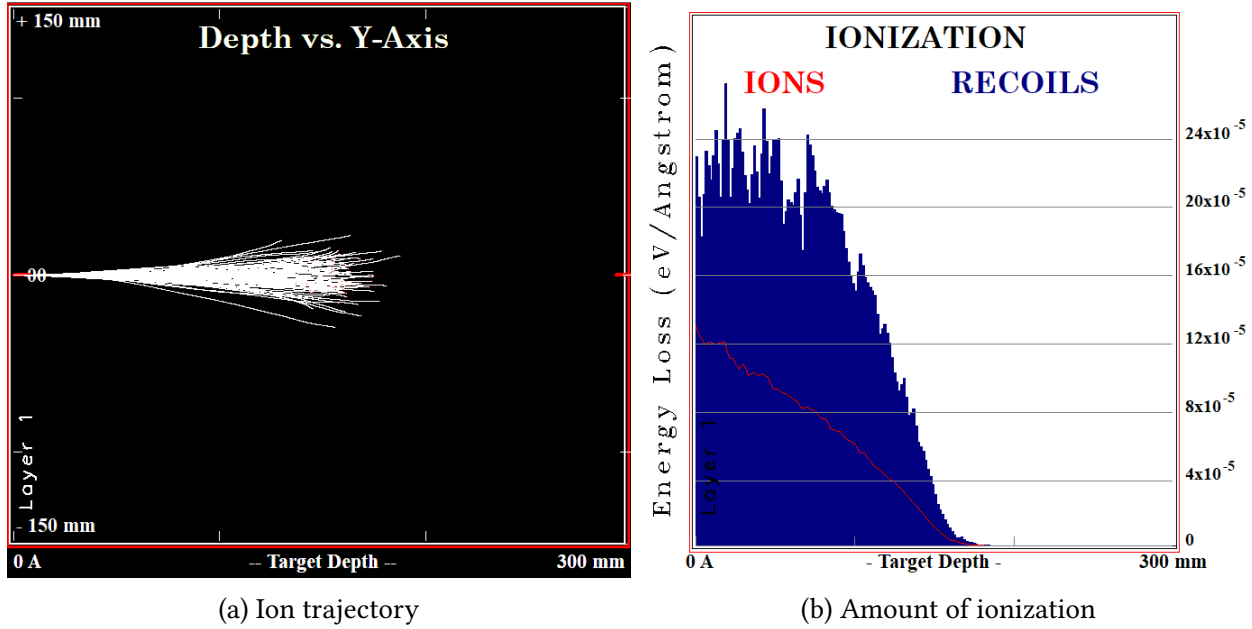


Figure 4.30: Ion trajectory range and ionization inside the main vacuum chamber after the increase of operating main vacuum chamber helium pressure to 8 mTorr.

Figure 4.31 describes how the pressures vary in the main vacuum chamber and the ion trapping region with gas stopping cell pressure while the three turbomolecular pumps that were previously described are closed. While describing the turbomolecular pumps and helium pressure, this orientation will now be known as "trapping operation". Although the velocity study from the previous section was performed with all turbomolecular pumps operational, the linearity of the pressures still exists.

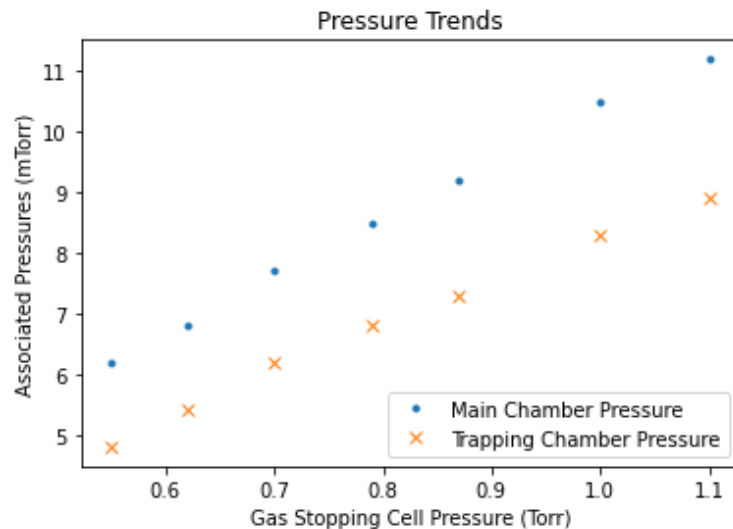


Figure 4.31: How the pressures in the main chamber and ion trapping region vary with gas stopping pressure during trapping operation. The only pump operational during this study is the small 210 L/s turbomolecular pump.

The beam line accomplishes two goals. The first goal is to eject all undesired mass and charge states of ions out of the beam using the QMS. The QMS efficiency increases as the pressure increases due to the ions spending more time inside the QMS. The next goal is to decrease the energy of the ions to less than 5 eV before entering the ion trapping region. The reason for this is to inhibit ionization after the QMS due to the kinetic energy of the  $^{229}\text{Th}^{3+}$  ions and to inhibit trajectory perturbations during the transition phase from the main vacuum chamber to the ion trapping region through the orifice plate. If ionization occurs after the QMS, the contents of the ion trap will be contaminated. This is done by operating the main vacuum chamber with a pressure of 7 - 8.5 mTorr while the voltage gradients in the RFQs have a measured voltage gradient of 0.43 V/cm that inhibit the ions from stopping inside the quadrupoles and can visually be seen in Figure 4.32.

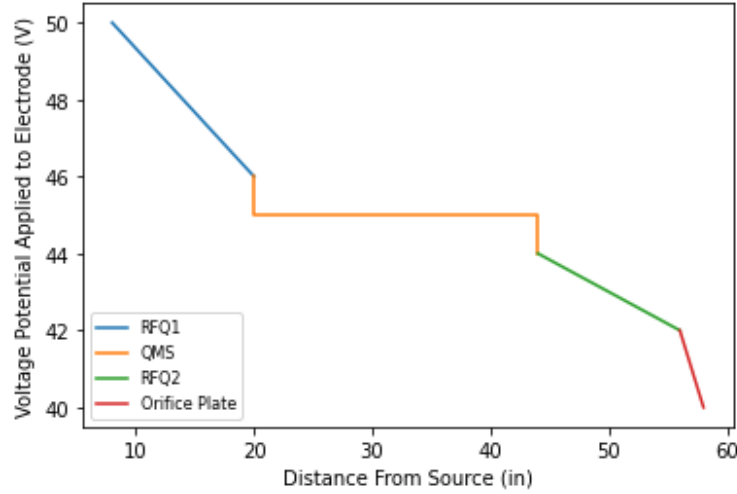


Figure 4.32: Potentials of the beam line starting with the first RFQ and ending at the orifice plate.

If it is assumed that the beam consists of  $^{229}\text{Th}^{3+}$  ions, the energy of the beam can be determined by raising the voltage of the orifice plate to stop approximately 90% of the beam. Once this is done, the voltage difference is multiplied by the charge state of the ion, 3+, which will yield the energy of the beam in eV. Figure 4.33 shows the response of increasing the orifice plate voltage to stop the beam. The corresponding converted beam energy plot is also given. This study was done using a 2 V accelerating gradient on all RFQs to ensure that the ions do not stop in the chamber due to interactions with the helium gas. Because the orifice plate is biased to 1 V below the last electrode of the adjacent RFQ, a stopping voltage of 1 V indicates that the orifice plate is biased to the same potential as the last electrode of the second RFQ and will not slow the beam until greater than 1 V is attained. When the valve that connects the main chamber to the ion trapping region is shut, the pressure inside the ion trap region increases due to less pumping. This also decreases the initial rate of the beam as a result of the higher ion trap pressure. The conclusion from this plot is as the pressure of the entire system increases, the energy of the beam decreases. However, the intensity of the beam also decreases. Therefore, an operating pressure must be chosen that slows the ions sufficiently to inhibit ionization without sacrificing a large portion of the beam. A gas stopping cell pressure of approximately 0.72 - 0.86 Torr is the operating pressure that was chosen based on this study. The corresponding main vacuum chamber pressure is 7.8 - 8.9 mTorr which corresponds to a beam energy of 5.4 - 3.3 eV, respectively. The converted energy plot of

the  $^{229}\text{Th}^{3+}$  ions entering the ion trapping region from Figure 4.33 can be seen in Figure 4.34.

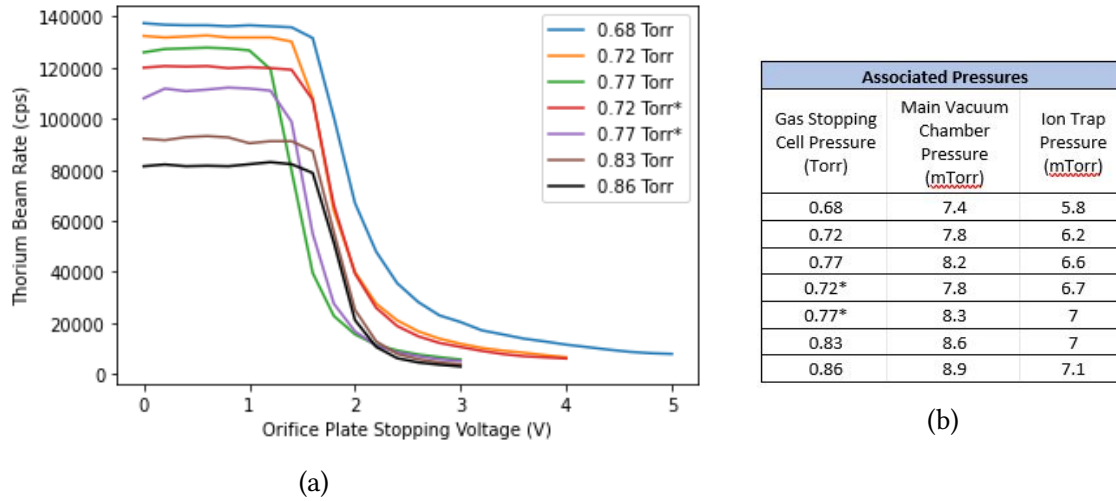


Figure 4.33: (a) Plot of differing ion stopping voltages for different gas stopping cell pressures. The stopping voltage is determined when approximately 90% of the beam rate is stopped at the orifice plate. This quantity can be used to determine the energy of the beam by multiplying the stopping voltage by the charge of the ion, 3+. Pressures marked with \* indicate that the valve attached to the vacuum line connecting the ion trapping region to the main vacuum chamber was shut. This limits the amount of pumping on the ion trapping region and raises the pressure. A stopping voltage of 1 indicates that the orifice plate is biased to the same voltage as the last electrode on the second RFQ and is where the difference begins. (b) The corresponding pressures of each separate vacuum chamber of the apparatus.

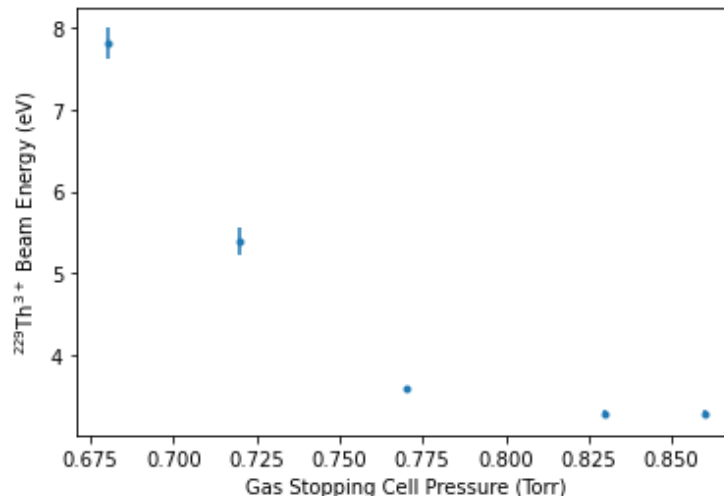


Figure 4.34: The beam energy of the  $^{229}\text{Th}^{3+}$  ions entering the ion trapping region. This plot was created using the data from the orifice plate stopping voltage study in Figure 4.33. The increase of residual helium gas inside the main vacuum chamber results in the decrease of the uncertainty of the energy due to ions arriving in a decreased energy spread in comparison to lower pressures where the energy spread is higher due to less interactions with the helium gas.

While the ion trap was not working properly, it was necessary to find a way to experimentally confirm that the QMS was working as expected. It was hypothesized that using the stability diagram from Figure 3.8 and the theory from Section 3.3.2, one could vary the potential difference between adjacent QMS rods and the RF voltage to confirm the contents of the beam. Based on this theory, the operating peak-to-peak RF voltage should be approximately 230 V and the potential difference between adjacent rods should be approximately 16 V. This would set the (q,a) values in the first stability region in the stability diagram for quadrupoles to be approximately (0.706, 0.246) with the peak of the first stability region at a (q,a) value of (0.706, 0.267). This value is set slightly under the peak to account for any voltage fluctuations that may occur. With these parameters,  $^{229}\text{Th}^{1+}$ ,  $^{229}\text{Th}^{2+}$ , and  $^{229}\text{Th}^{4+}$  will have (q,a) positions as shown in Figure 4.35. Thus, the values for 1+, 2+, and 4+ charged thorium are outside of the stability region.



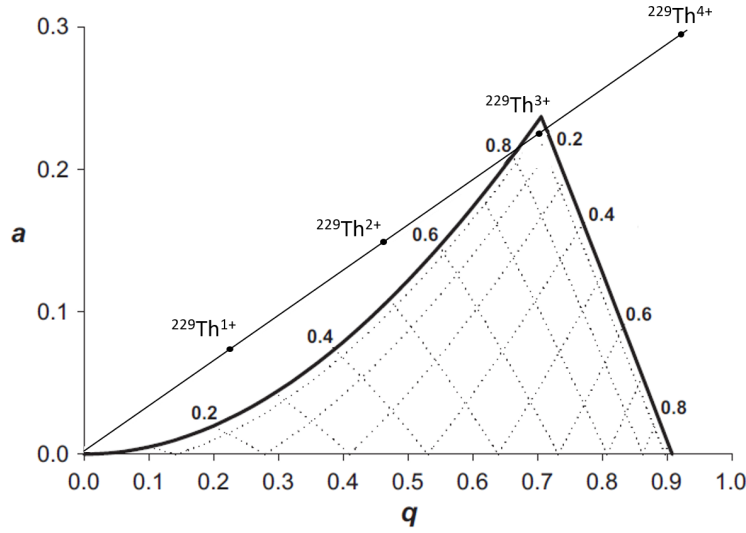


Figure 4.35

Figure 4.36 shows the mapping of the QMS to confirm this theory. The goal of this study was to map out the first stability region by increasing the DC offset voltage between adjacent rods ( $U$ ) with respect to increasing peak-to-peak RF voltage with parameters set to  $^{229}\text{Th}^{3+}$ . When the potential difference  $U$  is 0 V, the stability line in the stability diagram is equivalent to  $a = 0$  and every ion is transmitted through the QMS. However, when you increase the DC offset  $U$  to 10 V with RF voltage at 0 V, the starting point  $(q, a)$  has a value of  $(0, 0.085)$ . Increasing the RF voltage scans the region that  $^{229}\text{Th}^{3+}$  will be transmitted across a horizontal line intersecting the point  $(0, 0.085)$ . Increasing the value of  $U$  increases the value of  $a$  at which  $^{229}\text{Th}^{3+}$  starts. After  $a$  increases beyond 0.267, no ions will transmit through the QMS. The corresponding value of  $U$  that sets  $a = 0.267$  is approximately 22 V. Once this value is obtained,  $a$  is decreased until a count rate is seen. The RF voltage is then adjusted to allow the QMS to transmit the maximum amount of ions. This is visible in Figure 4.36. As theory suggests, a large peak clearly exists at an RF voltage of 230 V when  $U = 20$  V, which is where the QMS parameters are operated. Confidently, we can say that this is  $^{229}\text{Th}^{3+}$ . Contamination of the beam after the QMS is inhibited by reducing the energy at which the ions are traveling so ionization does not occur.  $^{229}\text{Th}$  has a half-life of  $4.16 \times 10^9$  min. This corresponds to a probability of decay of  $\lambda = 1.67 \times 10^{-10} \text{ min}^{-1}$  and is considered negligible.

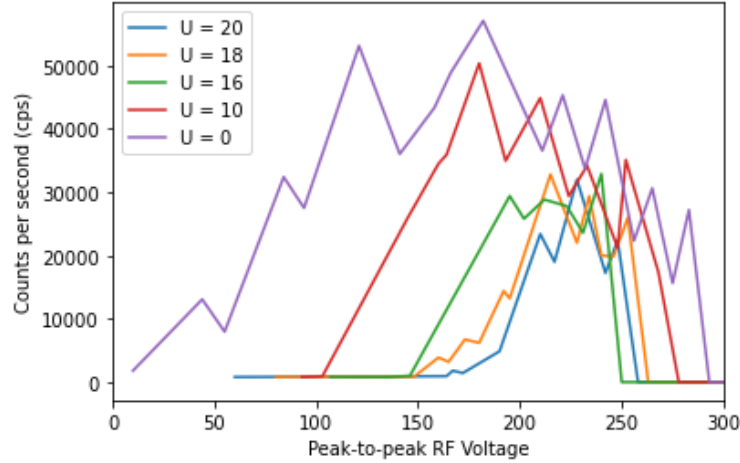


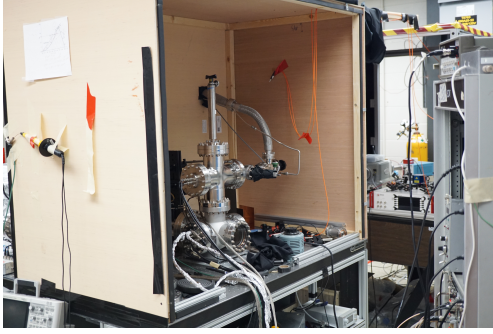
Figure 4.36: Plot of the beam rate while varying the potential difference  $U$  and the RF voltage  $V_{RF}$  of the QMS. Note that as  $U$  increases, the peak at which the maximum beam rate occurs approaches an RF voltage of 230 V suggesting that this is  $^{229}\text{Th}^{3+}$ .

#### 4.5.4 Linear Paul Ion Trap

The energy of the ion beam entering the ion trapping region is a very important quantity. If the ions enter at a high enough energy, two instances can occur. The first instance can lead to ionization of the helium gas. This will lead to false, increased beam rate results and trapping of ionized helium,  $^{229}\text{Th}^{2+}$ , and  $^{229}\text{Th}^{1+}$ . The second instance is the  $^{229}\text{Th}^{3+}$  can traverse through the ion trap without being slowed enough to initialize trapping. This can be shown by lowering the helium pressure inside the ion trapping region until an increased MCP response is shown. All data taken using the different ion traps will be shown in Chapter 5.

## 4.6 Laser Optics

In Section 3.4.2, the atomic excitation scheme of  $^{229}\text{Th}^{3+}$  ions was discussed. Essentially,  $^{229}\text{Th}^{3+}$  has two distinct atomic transitions that can be driven to confirm and quantify the contents of the ion trap. Figure 3.9 references the atomic transitions that will be used in this thesis to confirm and quantify the number of  $^{229}\text{Th}^{3+}$  ions trapped. The ground state,  $5F_{5/2}$ , must be driven with a 690 nm laser to the  $6D_{5/2}$  state. This state can either decay back to the ground state or to the  $5F_{7/2}$  state. Therefore, it is necessary to drive the  $5F_{7/2}$  state with a 984 nm laser back to the  $6D_{5/2}$



(a)



(b)

Figure 4.37: Pictures of the constructed dark box while open (a) and while closed (b). The black curtain serves to block photons from entering the dark box from the various drill holes and cable feed-thru holes.

state. This section will describe the different optical setups used to drive these two states.

#### 4.6.1 Optical Setup

The optical setup that this thesis utilizes requires two different optical tables. One optical table is located away from the experimental apparatus and is where the laser controllers and laser spectrometers are kept. The next optical table is located where the ion trapping chamber lies and is where the lasers will be directed for atomic excitation. The two optical tables are connected by a fiber optic cable to transport the laser. Figure 4.39 shows the optical setup before the lasers enter the fiber optic cable. Immediately after exiting the lasing mechanisms, the photons enter a window where 10% of the light is reflected toward a spectrometer that is used to constantly monitor the wavelength of the lasers during operation. The other 90% of the laser is then directed into a mirror which reflects the light into a prism that combines both lasers. This newly combined laser beam is directed into a single fiber optic port which collects the light and transports it to the other optical table. Once the laser exits the fiber optic cable, it enters a fiber optic collimator that manipulates the laser. The fiber optic collimator of choice was the ThorLabs ZC618FC-B. This specific collimator allows adjusting of the focal length of the laser exiting the fiber optic cable which can be used to adjust the diameter of the region of the ion trap that gets driven for optimal atomic excitation.

A large, wooden box was constructed over the ion trapping chamber optical table and used

as a dark box. This box has a removable front panel for easy access to the optical table. A black drape was attached to the front panel to inhibit photons from entering through the various drill holes and the holes created for cable feed-thrus. Two photomultiplier tubes are used to detect photons inside the dark box. One photomultiplier is an ET Enterprises model 9403B which is used to detect ultraviolet photons. It has a 25 mm active diameter with a spectral response between wavelengths of 110 nm to 230 nm. The background count rate for this phototube is 10 counts per second and has a quoted maximum efficiency of 10%. The next photomultiplier tube is an ET Enterprises model 9113B which is used to detect photons from the decay of the  $6D_{5/2}$  state from Figure 3.9. This phototube has an active diameter of 22 mm and has a spectral response between 280 nm to 850 nm. The background count rate for this phototube is 3,000 counts per second and has a maximum efficiency of 21%. The phototubes are placed out of the laser path and right against the windows to maximize solid angle from the center of the trap. Two orientations of laser paths were investigated in order to reduce the amount of background laser light detected. These two orientations include the laser being directed perpendicular to the  $^{229}\text{Th}$  beam path and parallel to the  $^{229}\text{Th}$  beam path.

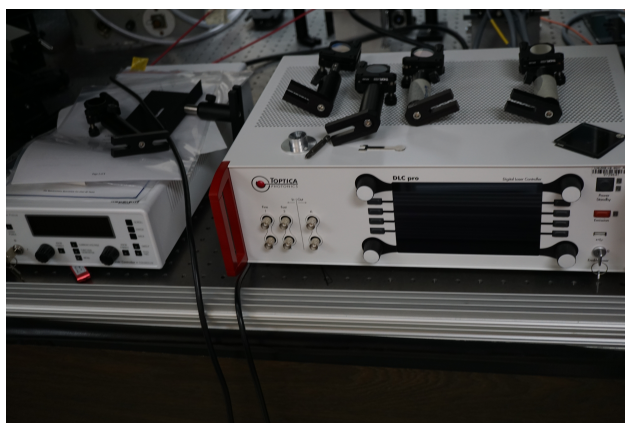


Figure 4.38: New Focus (left) and Toptica Photonics (right) laser controllers for the laser optics system. The New Focus controller runs the 984 nm laser while the Toptica Photonics controller runs the 690 nm laser.

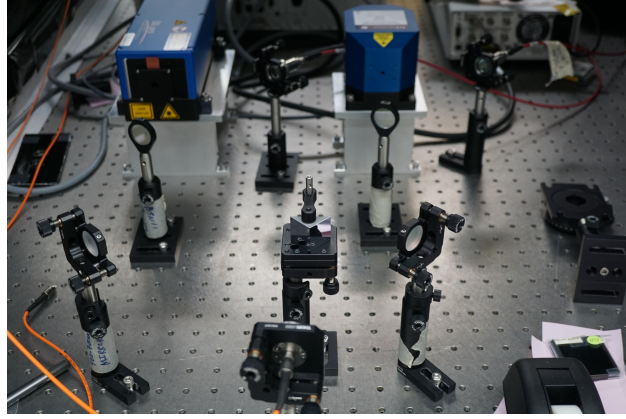


Figure 4.39: The current laser optics setup. The two lasers are directed into 90/10 splitting windows which are used to direct 10% of the light into a spectrometer for constant monitoring of the wavelength. After the 90/10 window is a mirror that directs the light into a prism. The prism combines the lasers and directs them into a fiber port coupler which carries the light onto the optical table where the ion trap sits.

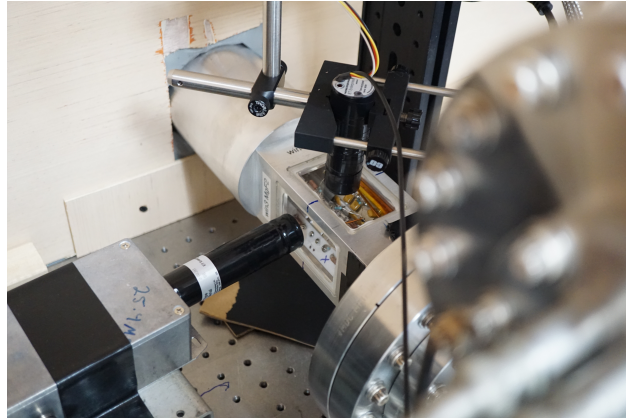


Figure 4.40: Picture of the center of the trapping region with the two different phototubes. On the left is the UV phototube while on top is the visible/infrared phototube. This setup is only used when the lasers are directed into the path of the thorium beam.

#### 4.6.2 Perpendicular to Beam Path

Because the windows on the ion trapping region allow transmission of the majority of wavelengths of photons, the lasers can be directed perpendicular to the beam path through the windows in the center of the ion trap. However, an issue arises from this. The windows used on the ion trapping region have a transmission rate of about 95% for 690 nm wavelength photons. A 5 mW power laser of this wavelength will emit approximately  $1.73 \times 10^{16}$  photons per second. If it

is assumed that the photons that are not transmitted through the window are scattered instead of absorbed, the number of scattered photons through the first window is approximately  $8.65 \times 10^{14}$  and the second window is approximately  $8.21 \times 10^{14}$  photons per second. This leads to a high phototube background while the lasers are operational. Therefore it is necessary to either decrease the amount of scattered laser light inside the ion trap or increase the number of  $^{229}\text{Th}^{3+}$  ions trapped so that the number of photons emitted from atomic de-excitation well exceeds the amount of scattered photons from the laser. Beam dumps and multiple photon shields were inserted onto the optical table in an attempt to reduce the amount of scattered light. This was an ineffective method to reduce stray light. Another method that was investigated was changing the direction of laser incident on the ion trap.

#### **4.6.3 Horizontal to Beam Path**

In an attempt to decrease the amount of scattered laser light, the laser path was directed down the path of the  $^{229}\text{Th}^{3+}$  beam. A viewing port with 95% transmission of 690 nm photons was inserted at the back end of the ion trapping region. With only one window to scatter the laser, the amount of scattered laser photons should decrease by approximately 50%. However, the MCP must be removed from operation during laser operation. Initially, the laser was directed with a focal point corresponding to the distance to the orifice plate from the edge of the fiber optic cable. This ensured minimum reflection by the shiny, aluminum surface of the orifice plate. To stop photons from scattering back into the ion trapping region after entering the main vacuum chamber, the inside of the QMS was coated with a carbon-based black, conductive coating that helps absorb photons.



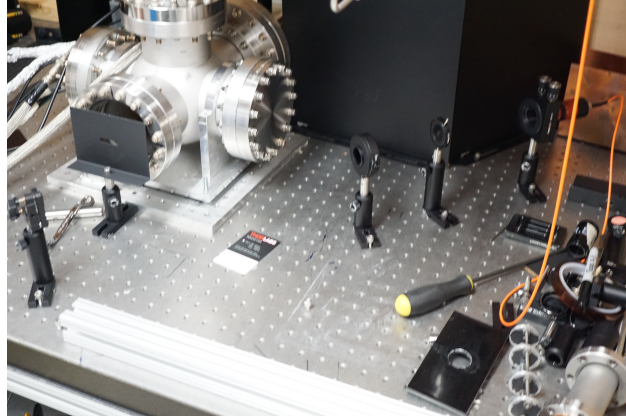


Figure 4.41: Shown is a typical collimated laser beam path which is directed down the thorium beam path line. This is used to excite the  $^{229}\text{Th}^{3+}$  ions that are trapped inside the linear Paul ion trap.

## 4.7 Microchannel Plate Detector

At the end of the ion trapping region is a microchannel plate detector (MCP) which is used to measure the  $^{229}\text{Th}^{3+}$  beam rate as well as measure the quantity of ions released from the trap by lowering the end of the potential well to accelerate ions toward the MCP. The type of MCP used is a Photonis MCP with a diameter of 1 inch. MCPs contain 4 different sections: the grid, front and black plates, and the anode.

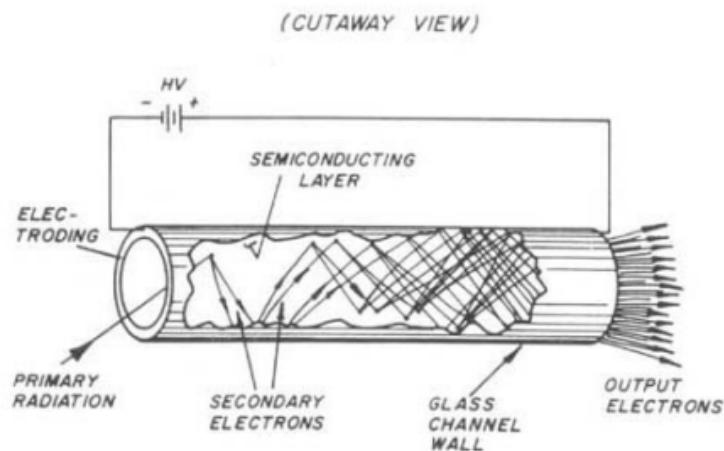


Figure 4.42: A single channel in the glass array of the MCP [22].

The grid attracts the  $^{229}\text{Th}^{3+}$  ions as they approach as well as insulates the front plate from

outside fields while also repelling any negatively charged particles approaching the MCP. The front and back plates contain an array of tiny lead glass tubes that are fused together which form channels for electron multiplication. A single channel of the glass array can be seen in Figure 4.42. Both faces of the front and back plates are coated with a nickel and chromium alloy that allows biasing of each individual glass tube simultaneously. When the potential gradient across the front and back plates is set to 800-1400 V, the glass channel becomes a continuous dynode electron multiplier. A conceptual example of this process can be seen in Figure 4.43.

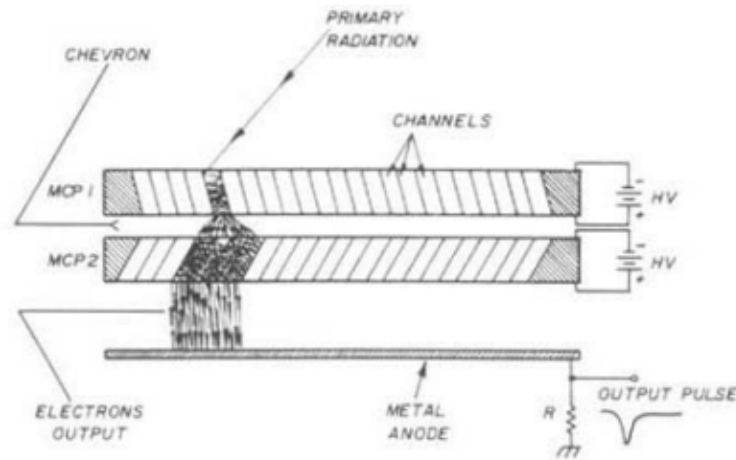


Figure 4.43: Conceptual view of the electron multiplication process inside the MCP [22].

When an event happens that triggers the release of an electron, i.e. when an ion collides with the MCP, one or more electrons will enter the glass tube array. Those electrons are forced through the glass tubes by the applied voltage gradient where each collision inside the tube produces secondary electrons. In vacuum, with a 1,400 V potential difference between the front and back plates, the gain of the MCP is approximately 1,000,000. These electrons are gathered by the anode plate on the MCP where the signal is output. The MCP that is used for this thesis uses a resistor network to set all the voltages appropriately. The potentials applied on the MCP for this thesis are shown in Table 4.1.



| Location    | Potential (V) |
|-------------|---------------|
| Grid        | -3300         |
| Front Plate | -2200         |
| Back Plate  | -800          |
| Anode       | -70           |

Table 4.1: The potentials specific to the Photonis MCP used for detection of  $^{229}\text{Th}^{3+}$  ions.

The MCP is attached to a plunging system in order to relocate the MCP during use of the laser optics system. However, the plunging system is only stable in the vertical direction. Once the vertical position is set, there is an uncertainty in the position along the other two axes. In this thesis, it is assumed that the MCP is 50% efficient at detecting charged particles and has a measured background of 10 counts per second. Because of the position of the MCP in the apparatus, it was difficult to insert an alpha source with a large enough solid angle for detection to characterize the MCP efficiency. Two alpha sources were used in an attempt to characterize the efficiency. The first source used was an  $^{241}\text{Am}$  source that had a quoted activity of 4181 Bq on July 1, 2010 which corresponds to an activity of 4085 Bq in 2020 at the time of the study. The Am source was placed on the ceramic material holding the ion trap inside the ion trapping region. The MCP was plunged to a distance of 1 - 1.5 inches away from the source directly in the center. This distance was unable to be measured precisely. The efficiency of the MCP in a vacuum pressure of  $2.3 \times 10^{-6}$  Torr ranged from 0.24 to 0.47. When the helium was added to the ion trapping region, the count rate on the MCP started to increase which can be viewed in Figure 4.45. It is believed that the alpha particles from the decay were ionizing the helium atoms which were then being detected by the MCP. Because of the range of the calculated MCP efficiency, it was discussed to assume the efficiency was 50% in vacuum.  $^{252}\text{Cf}$  was also used in an attempt to characterize the efficiency. The  $^{252}\text{Cf}$  used had an activity of 3841 on July 1, 2020. When the source was used, the activity of the californium had dropped to 267 Bq and the response was unable to be distinguished from the noise.

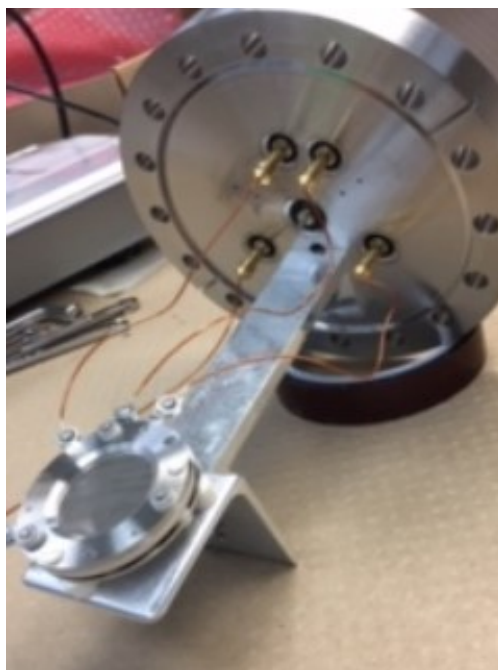


Figure 4.44: Photo of the MCP used in the ion detection process before the plunger was attached.

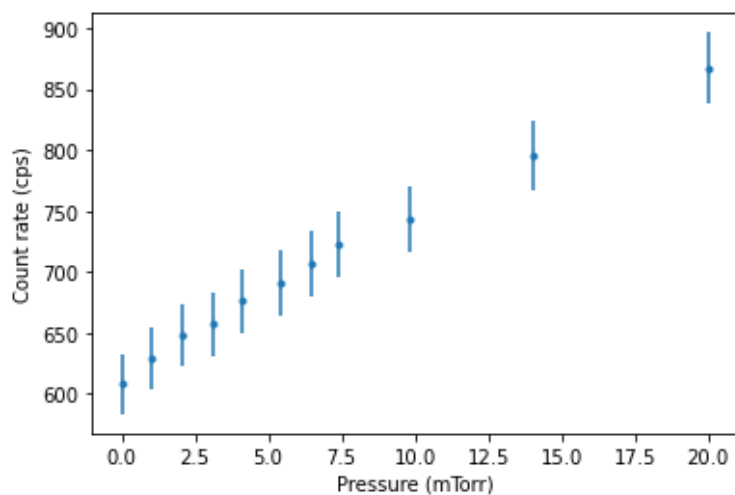


Figure 4.45: The count rate obtained from the decay of an  $^{241}\text{Am}$  alpha source in an effort to characterize the MCP efficiency. The Am source was positioned 1-1.5 inches away from the center of the MCP and had an activity of 4085 Bq at the time of this study.

## 4.8 Ion Counting Signal Processing

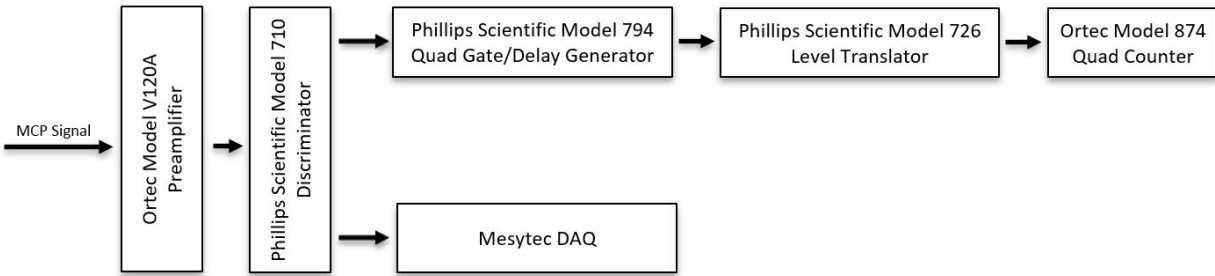


Figure 4.46: The electronics schematic for the detection system using the MCP.

Once the ion trap potential well is opened, the ions will be slowly guided to the MCP. Because the MCP is biased to a potential of -3,300 V, the  $^{229}\text{Th}^{3+}$  ions gain kinetic energy the closer the ions get to the MCP due to Coulombic attraction. Assuming the MCP attraction is the major force in the energy gain of the ions after the potential well release, the velocity of ions will increase to approximately 3,000 m/s before colliding with the MCP. Once the ions collide with the front plate of the MCP with force, electrons are released and enter the cascading which is described in Section 4.7.

The signal processing electronic schematic for the Photonis Chevron MCP with 1-inch diameter is shown in Figure 4.46. The raw MCP signal is fed through an Ortec Model VT120A preamplifier to amplify the raw signal to -3 to -5 V which can be viewed in Figure 4.47. The approximate 15 ns MCP pulse has a ringing effect which creates a response that is approximately 130 ns wide. This ringing can be described as an impedance mismatch between the preamplifier and the MCP circuit that applies the voltage potentials to the MCP electrodes. The preamplifier has an impedance of  $50\ \Omega$  and the MCP circuit has an impedance of  $12.6\ \text{M}\Omega$  which causes a signal reflection.

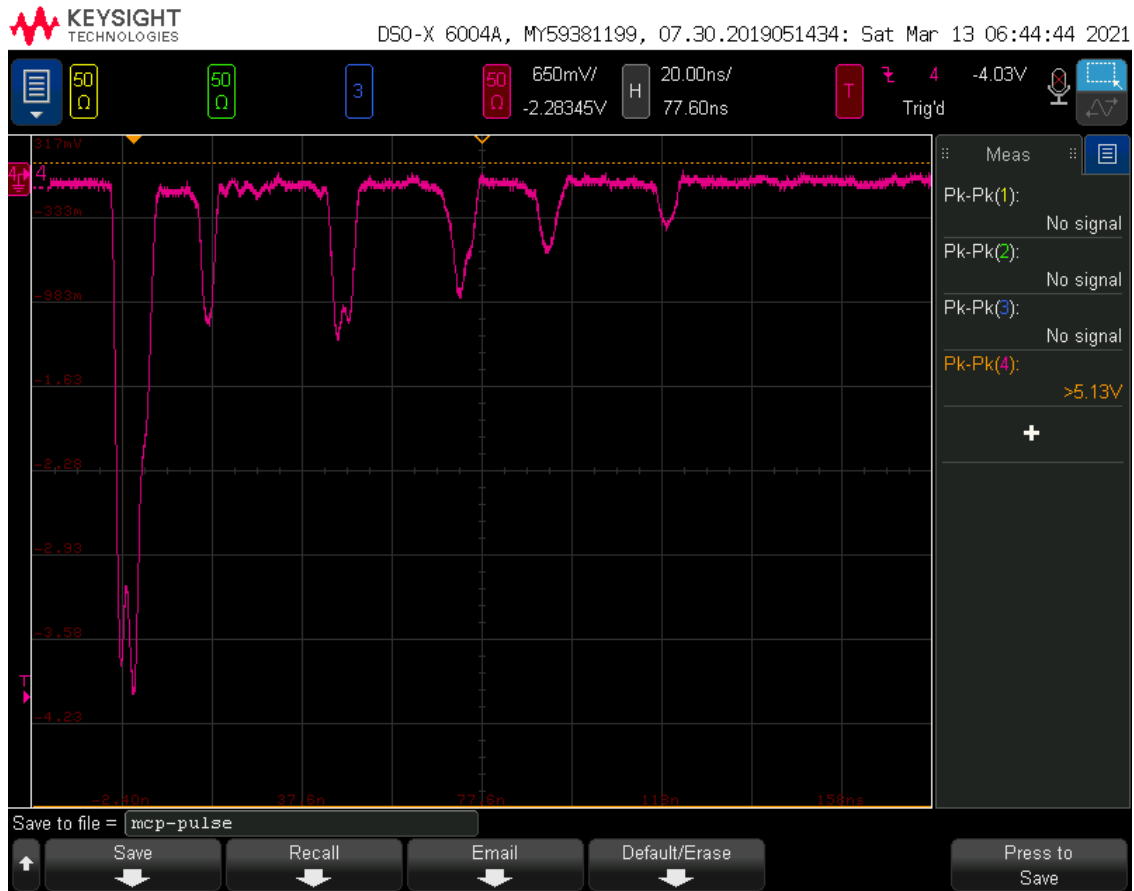


Figure 4.47: A typical MCP response that exhibits the ringing feature due to an impedance mismatch between the MCP and the preamplifier.

The signal is amplified to -3 to -5 V in order to easily distinguish between electronic noise and the MCP signal. This amplified signal is fed through a Phillips Scientific Model 710 octal discriminator. On the discriminator, a 700 mV threshold is set to eliminate electronic noise and possible RF leakage. This changes the raw, amplified MCP signal into a NIM pulse with a signal width of 10 ns which triggers approximately 15 ns after the initial MCP pulse. The output of the discriminator can either be directed toward the Mesytec data acquisition system where the data is plotted and stored digitally, or can be directed into a gate/delay generator, signal converter and into a counter. The Phillips Scientific Model 794 quad gate/delay generator is used to inhibit multiple event triggering during the ringing of the MCP. The gate/delay generator has a set output delay of 70 ns with a gate width of 120 ns. In a single event, only the initial MCP pulse will be accepted even if the discriminator were to trigger multiple times during the ringing of the MCP.

Because the Ortec Model 874 quad counter requires a TTL signal, the signal generated by the gate/delay generator is fed through a Phillips Scientific Model 726 level translator which is used to convert the NIM signal into a TTL signal. The final section of the detection schematic is the quad counter which is used to receive a quick quantification of the beam rate or the amount of ions released from the potential well of the ion trap.

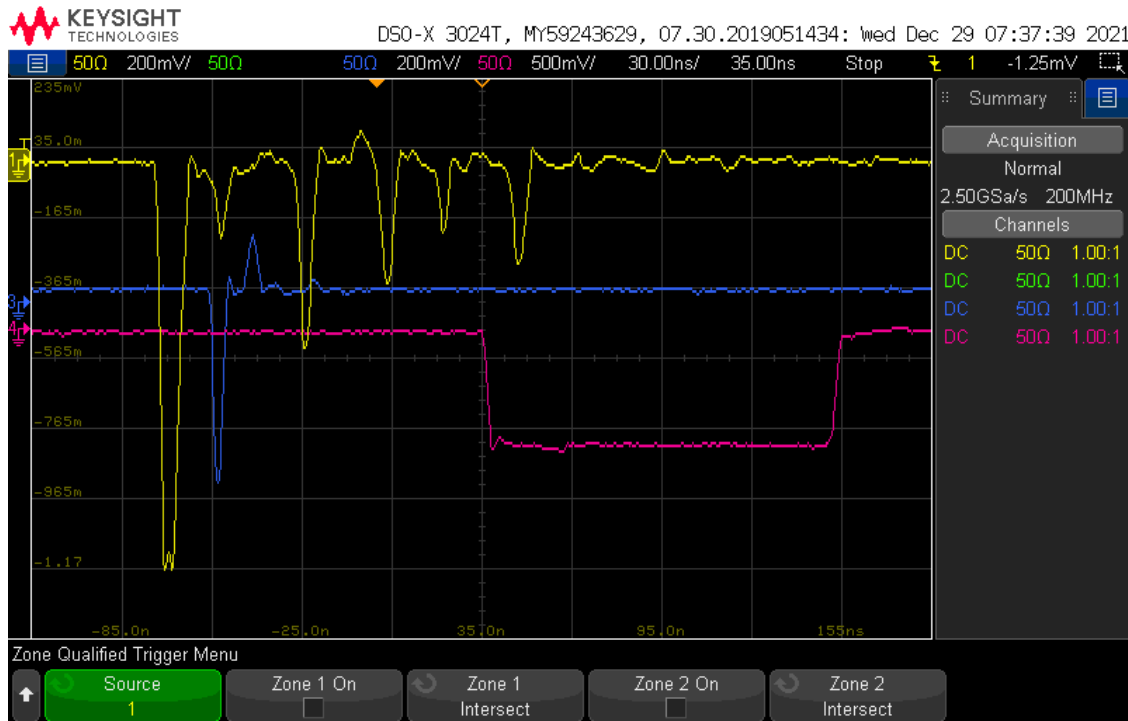


Figure 4.48: Oscilloscope signals of the MCP (yellow), the discriminator (blue) and the delay/gate generator (magenta).

Using data from Figure 5.9, the number of ions trapped is 350,666 ions which are recorded over 2 seconds. The probability an event is to be overlooked due to pulse-pileup can be viewed as  $1 - \exp(-rt)$  where  $r$  is the average rate of the response and  $t$  is the processing time of the system. This probability can be attributed to the dead time of the system and is calculated to be 2.4%. With the delay on the delay/gate generator, the total electronic response time takes approximately 260 ns from the beginning of the MCP pulse to the end of the gate. Therefore the maximum counting rate of the system, corrected for dead time, is approximately  $4 \times 10^6$  events per second.

# Chapter 5

## Results and Discussions

The results of the effort to construct and optimize a linear Paul ion trap for the direct detection of  $^{229m}\text{Th}$  will be covered in this chapter. These results will be separated into two different sections, ion trapping and atomic laser excitation. The ion trapping results will be separated into three different subsections. The first subsection will include results and further discussion of the first ion trap that was based on the cylindrical quadrupole design and why it was decided to redesign the ion trap. The next subsection will cover the vane ion trap which was constructed to correct the issues the first ion trap had while preserving viewing solid angle. The third subsection will cover the results obtained after the addition of a cryopump at the end of the trapping region to combat the water vapor outgassing issue which allowed for an increased ion trapping capacity.

### 5.1 Ion Trapping Results

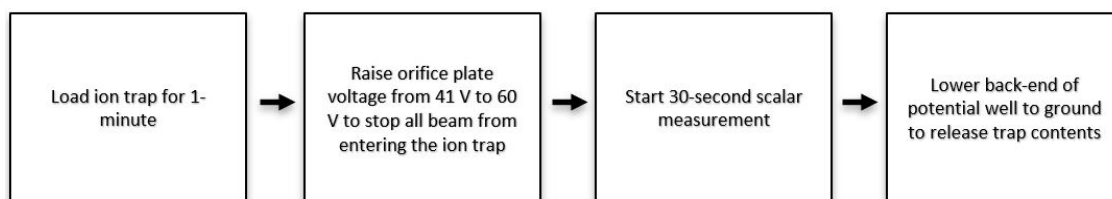


Figure 5.1: The procedure used to load and empty the ion trap to measure the quantity of ions trapped. This procedure starts after the optimization of the  $^{229}\text{Th}^{3+}$  ion beam.

The procedure for trapping and estimating the number of  $^{229}\text{Th}^{3+}$  ions trapped is as follows and can be viewed in Figure 5.1: the ion beam rate is first optimized which includes adjusting the RF voltages for the RF carpet (between 100 V and 120 V), the first RFQ (between 300 and 400 V), and the second RFQ (between 120 and 180 V) to allow the maximum  $^{229}\text{Th}^{3+}$  beam rate attainable (between 80,000 - 90,000 per second) as well as mapping out the QMS to ensure the ion beam contains only  $^{229}\text{Th}^{3+}$  ions.

The ion trap is loaded for 1-minute after optimization of the ion beam to allow as many  $^{229}\text{Th}^{3+}$  ions to be trapped as possible. The next three steps are performed in quick succession. A 30-second scalar measurement is started which allows ample time for the ions to escape into the MCP once the potential well on the back of the ion trap is lowered. Immediately following, the bias voltage on the orifice plate is risen to 60 V to stop all beam from entering the ion trap. Only background counts should be detected by the MCP at this point. The bias potential of the far end of the potential well of the ion trap is lowered slowly to ground which allows the trapped ions to escape the potential well and be guided into the MCP. The voltage is lowered slowly to ease the saturation effect of the MCP as the ions will arrive at the MCP within hundreds of microseconds of each other. Once the first 30 seconds concludes, a 30-second background measurement is taken and subtracted from the results obtained during the opening of the potential well. One issue with having a continually loading ion trap is the amount of time it takes to load and unload the trap, vary the parameters of the trap, let it stabilize and repeat. The time it takes to test one configuration of DC potentials, RF voltages, and gas pressures is heavily reliant on the large RC time constant due to having multiple capacitors and resistors on the ion trap.

### 5.1.1 Ion Trap Optimization

The procedure for finding the optimal configuration for the ion trap was as follows and can be viewed in Figure 5.2: the first and last electrodes on the ion trap were always biased to 40 V. The voltage gradients on the potential well were set to +1 V and -1 V, initially. The middle electrode was always initially set to 1 V beneath the adjacent electrodes and started at 38 V. This created a very slight potential well. The peak-to-peak RF voltage was set to a starting value of +360 volts.

The pressure of the ion trap was set to 5 mTorr, which was lower than the pressure of the gas stopping cell that provided the lowest energy  $^{229}\text{Th}^{3+}$  ions entering the trap with a beam intensity of 80,000 ions per second. To begin, a measurement was taken using the initial trapping parameters stated. The bias voltage of the middle electrode was lowered by 1 V and a measurement was taken. If an increase in ions trapped was measured, the voltage on the middle electrode was lowered again. This was repeated until a decrease in ions trapped was measured. The optimum parameters were recorded and the RF voltage was increased by 10 V. The process thus far was restarted and repeated until the MCP started picking up RF noise, which occurred around +450 V. The RF voltage was then reset to its initial value of +360 V. The potential well gradient on the ion trap was then increased by 1 V and the middle electrode was set to a value of 1 V beneath the adjacent electrodes and the entire process was repeated until the average number of  $^{229}\text{Th}^{3+}$  ions trapped per potential well gradient started to decrease. The helium pressure was then increased by 0.5 mTorr and the entire process repeated again. The entire optimization process was stopped when the gas stopping cell chamber pressure approached 1 Torr. This is because the  $^{229}\text{Th}^{3+}$  beam rate had decreased substantially as seen from Figure 4.33. The results of each ion trap will be discussed in the following sections.



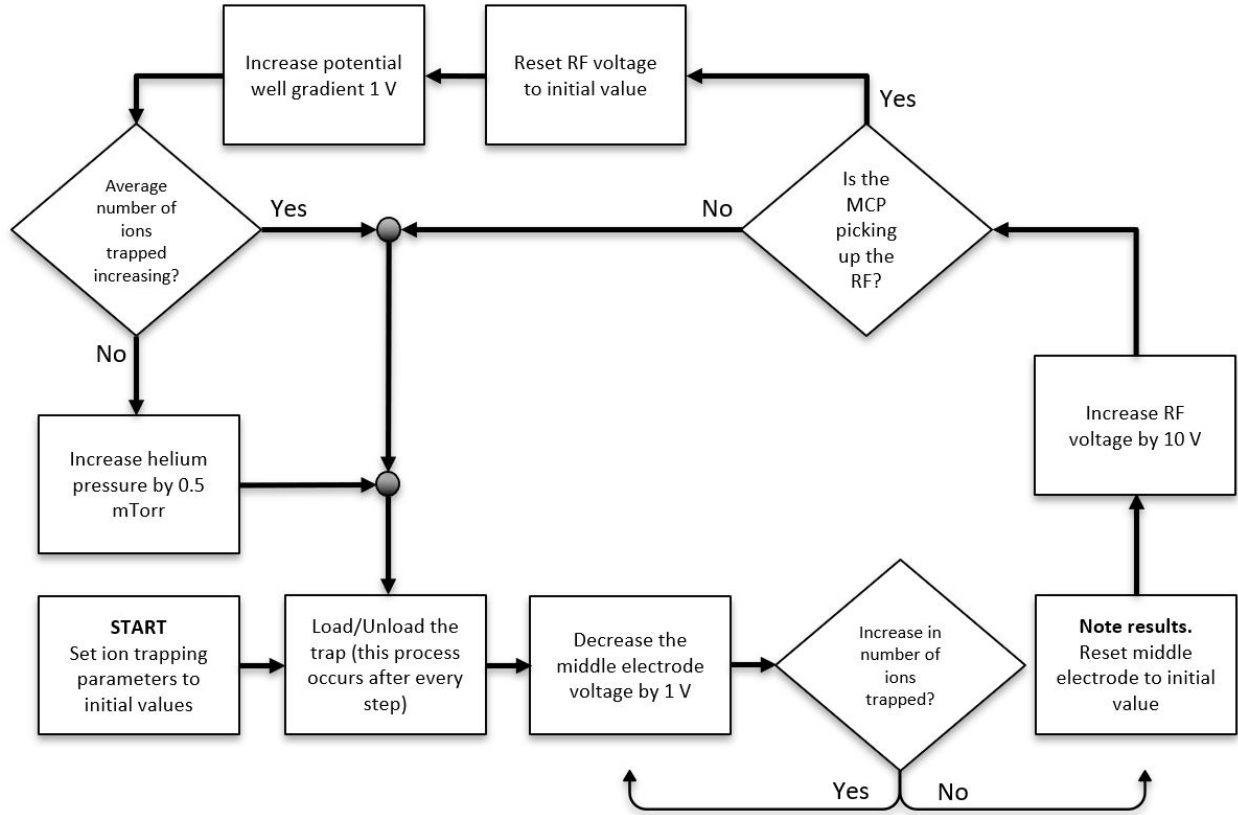


Figure 5.2: The algorithm used to optimize the ion trap. The initial values for this process are as follows: upstream and downstream potential well gradients on the ion trap are set to 1 V. The first and last electrodes on the ion trap are always biased to a potential of +40 V. The middle electrode starts at 38 V. The initial RF voltage for the ion trap is +360 V. Initial helium pressure starts at 5 mTorr. Please refer to thesis text for a detailed description of each process.

The optimum configuration of ion trap settings found to date is as follows: The first 7 electrodes of the ion trap have an 8 V accelerating gradient applied. The first electrode of the ion trap is biased to 40 V, 1 V below the operating voltage of the orifice plate. This is done to ensure the continuation of the beam through the orifice plate into the ion trap. The voltage of the middle electrode is biased to 3 V below each adjacent electrode. The following 5 electrodes have an 8 V decelerating gradient applied. The potentials of the beam line plus ion trap are shown in Figure 5.3 during trapping operation as well as during the release of the contents in the trap. Figure 5.3a are the potentials of the ion trap during trapping operation and will be constant. However, Figure 5.3b potentials are not constant. The potentials shown are the final potentials during the lowering of the back end of the potential well on the ion trap to guide the trapped ions into the MCP.

The peak-to-peak RF voltage applied to the ion trap is +410 V. This voltage will change slightly depending on the temperature of the room that day. The optimum trapping pressure found is 6.2 - 7.1 mTorr.

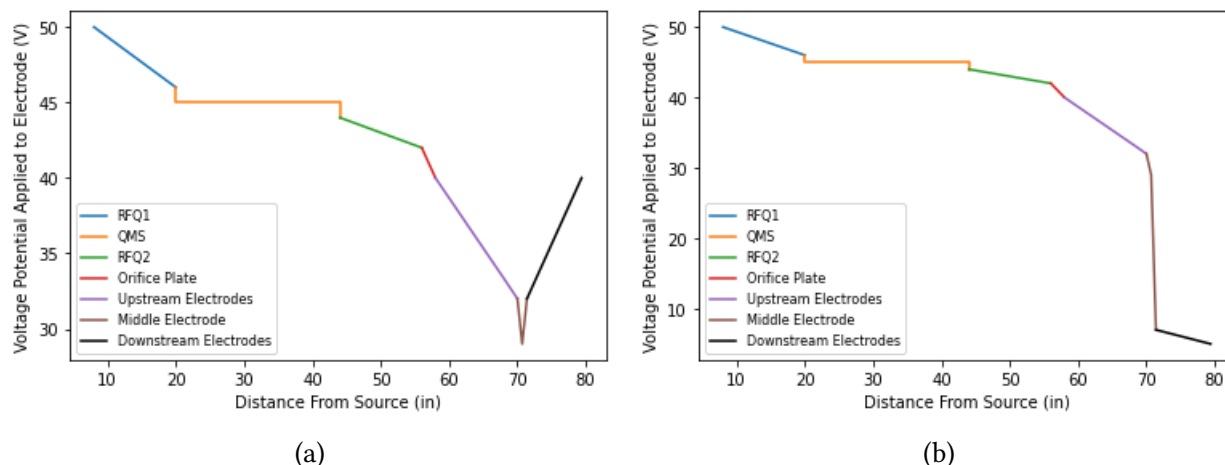


Figure 5.3: The voltage potentials of the beam line during (a) trapping operation and (b) unloading of the ion trap. Note that (b) are the final potentials of the ion trap during release. The ion trap is separated into upstream electrodes, middle electrode, and downstream electrodes

### 5.1.2 Cylindrical Ion Ion Trap

As mentioned previously, the cylindrical ion trap had a very small experimental  $r/r_0$  value, where  $r$  is the radius of a quadrupole electrode and  $r_0$  is the shortest distance from the center of the quadrupole to an electrode. This led to gaps in the applied RF fields that allowed  $^{229}\text{Th}^{3+}$  ions to escape during the trapping process and/or limit the containment of ions in the center which decreased the half-life of the ion trap considerably. Figure 5.4 shows a typical response received using the cylindrical ion trap. At 28 seconds is when the orifice plate voltage was raised above the stopping voltage to stop the beam from entering the ion trap. Before 28 seconds, the response includes ions that are energetic enough to escape the potential well. At approximately 30 seconds, the bias on the potential well is lowered to guide the ions toward the MCP. Two peaks are visible in the response. This is due to the variation of velocities of the ions when the potential well is dropped. The first, narrow peak is likely due to  $^{229}\text{Th}^{3+}$  ions that have yet to reach the end of the potential well. These ions have the greatest velocity and, therefore, will reach the MCP the quickest. The second peak is the typical response of trapped/slowed ions being released with a

large tail at the end that corresponds to ions that were released toward the front of the ion trap.

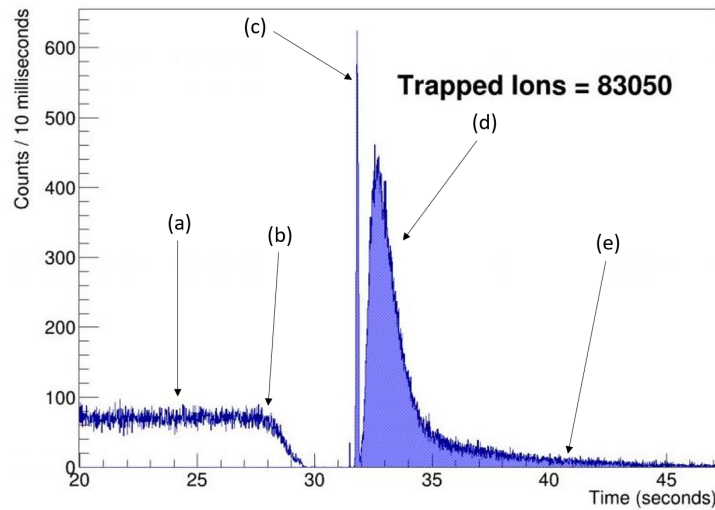


Figure 5.4: A typical response using the cylindrical ion trap design after attempting to trap  $^{229m}\text{Th}^{3+}$  for 60 seconds with a beam rate of approximately 80 kcps. Note the two different peaks and tail of the second peak. (a) is the response of the ions that escape the potential well into the MCP. (b) is the moment the orifice plate voltage is risen to inhibit the beam from entering the ion trapping region. The background rate should be zero after this happens. Immediately after the orifice plate voltage is increased, the potential well on the back of the ion trap is lowered to ground to guide the ions into the MCP. (c) is the immediate response of the ions releasing. The sharp peak indicates ions that were not trapped. (d) is the typical response of the unloading of an ion trap with (e) the long tail that is described in the text. This data is corrected for an assumed 50% MCP efficiency.

The  $^{229}\text{Th}^{3+}$  beam rate for this specific run had an intensity of 80,000 ions per second. The pressure inside the main vacuum chamber was approximately  $10^{-5}$  Torr for this run which leads to a large energy ( $>500$  eV) of incoming ions. To compensate, the pressure inside the ion trapping region was risen to 7-8 mTorr. This can lead to multiple problems. The first problem that arises is the pressure differential between the main vacuum chamber and the ion trapping region. This will cause a gas jet to occur in the orifice plate that will attempt to force ions into the main vacuum chamber. The second problem that arises is potential ionization due to the  $^{229}\text{Th}^{3+}$  ions having a large incoming energy. With such a large incoming energy, the incoming ions may strip electrons off of the helium buffer gas which will cause production of  $^{229}\text{Th}^{2+}$ . Therefore, a fraction of ions in the response shown in Figure 5.4 may potentially be  $^{229}\text{Th}^{2+}$ ,  $^{229}\text{Th}^{1+}$ , or even positively charged helium. Because all turbomolecular pumps were operational during the use of the cylindrical ion

trap, the water vapor outgassing issue should not occur due to the extra pumping, which will be explained in the next subsection.

### 5.1.3 Vane Ion Trap

The rectangular electrode design of the new vane ion trap allows the placement of electrodes closer to the center of the beam path without sacrificing viewing solid angle. If the electrodes of the cylindrical ion trap were constructed closer to the center, this would inhibit the solid angle of the design from the center of the trap to the viewing windows. A flat, rectangular electrode will not have this problem. This allows more confinement toward the center of the beam path as well as stopping ions from leaking out through potential RF field gaps in the ion trap. While using this trap design, the main vacuum chamber and ion trapping region pressures are nearly equalized and operated in the mTorr region (5-8 mTorr) in order to suppress the gas jet created by the orifice plate while operating at a pressure differential. Increasing the gas pressure inside the main vacuum chamber also decreases the kinetic energy of the thorium beam before entering the ion trap. With an incoming energy of approximately 4.5 eV, instead of approximately 500 eV when the main vacuum chamber pressure was operated at  $10^{-6}$  Torr, this allows application of more gradual gradients for the potential well which inhibits perturbations during the trapping process as well as eliminates all ionization from helium interactions. With an increased helium pressure inside the main vacuum chamber, a 4-volt accelerating gradient is applied to the first RFQ to assist the ions trajectory forward through the QMS. Since the QMS has no associated potential gradient, there is a chance the ions will stop inside. However, the Coulomb forces between the positive ions will slowly force the ions forward while the RF confines them inside the quadrupole. This also increases the separation efficiency of the QMS as the ion beam will spend increased time inside. The second RFQ has a 2-volt gradient applied in order to gently guide the ions into the ion trapping region. Due to the lower velocity of the  $^{229}\text{Th}^{3+}$  beam with respect to the cylindrical ion trap, a potential well of approximately 11 volts is sufficient to trap three times the beam rate without the use of the cryopump.

Because the filament inside an RGA burns out quickly during operation inside low vacuum,

the RGA was never operated during the increase of helium pressure inside the main vacuum chamber. However, it was hypothesized that the reason for an insufficient number of ions trapped and the half-life of the ion trap decreasing quickly was due to the presence of residual gas. With a 30-minute run of the RGA using the trapping configuration, it was quickly determined that a build-up of water vapor was occurring. The 210 L/s small turbomolecular pump could not pump the water out faster than it was outgassing. Figure 5.5 shows the pressure versus time overnight response of various background gases while the turbomolecular pumps were configured as if operating during trapping. A 100-fold increase of the partial pressure of water vapor happens in approximately 3.5 hours. The partial pressures of nitrogen and oxygen also increase, but come to equilibrium faster than the partial pressure of water does.

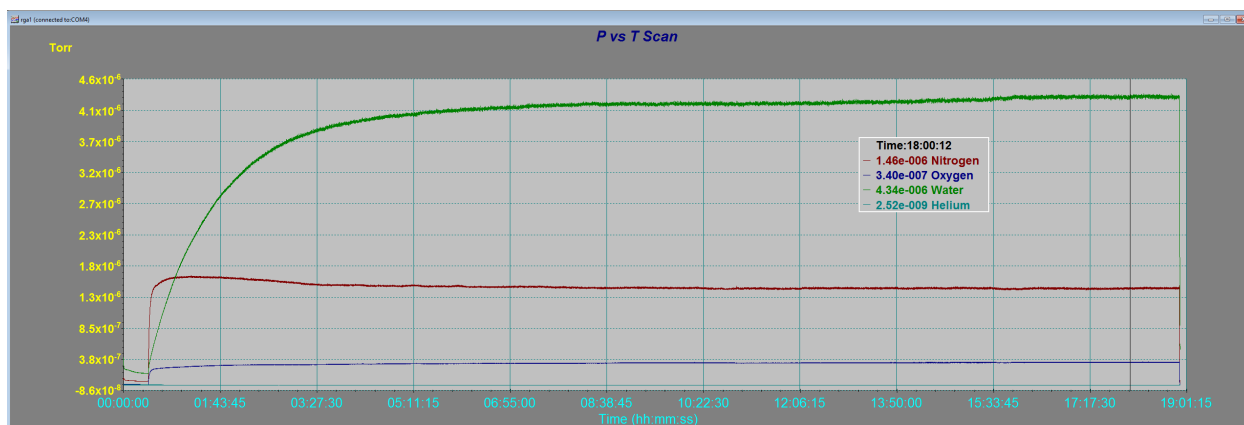


Figure 5.5: Response of nitrogen, oxygen, and water vapor inside the main vacuum chamber with only the small 210 L/s turbomolecular pump operational. Note that the amount of water vapor inside the main vacuum chamber increases 100 times in approximately 3 hours and continues to increase afterwards insinuating that the outgassing rate is exceeding the pumping rate.

Water vapor has an ionization potential of approximately 13 eV [68] and a bond dissociation energy of 5.15 eV. While the  $^{229}\text{Th}^{3+}$  ions are trapped, a water molecule can interact with the thorium ion which will cause the molecule to separate into  $\text{OH}^-$  and  $\text{H}^+$ . If this happens, the negatively charged hydroxide ion can bond with the positively charged  $^{229}\text{Th}^{3+}$  ion. This leads to the alteration of the contents of the ion trap and the half-life to decrease over time due to neutralization of the contents inside the trap. An excess of positively-charged hydrogen ions can also skew results as the hydrogen ions will likely be trapped with the  $^{229}\text{Th}^{3+}$  ions. Before this was learned, helium was introduced to the entire system approximately two hours before the

experiment was to begin. This was to allow time for temperatures and pressures of the system to come to equilibrium. Therefore, when attempting to trap, an overabundance of water vapor already existed in the system. Due to this water vapor issue, a cryopump was purchased and attached to the ion trapping region in an attempt to continuously pump out the water vapor. This will increase the number of ions trapped as well as the half-life of the ion trap. Figure 5.6 shows the percentage of ions lost inside the ion trap after delaying the opening of the potential well in an attempt to view the half-life of the ion trap. This specific run was performed after 4 hours of water vapor accumulation due to outgassing. This figure shows the ion trap having a half-life of  $10.30 \text{ seconds} \pm 0.70 \text{ seconds}$ . This uncertainty is attributed to the assumed flat  $\pm 0.4 \text{ second}$  uncertainty due to manual adjustments of the potentials. Due to the amount of interactions that is thought to be occurring after the amount of water vapor outgassing was discovered, the contents of the trap are not considered to be solely  $^{229}\text{Th}^{3+}$  ions. Figure 5.7 shows the response of the opening of the potential well in the ion trap allowing trapped ions to be guided toward the MCP after 2 hours of water vapor accumulation. In this specific run, if the MCP efficiency is assumed to be the quoted 50%, the number of ions trapped is  $303,652 \pm 779$  ions. The potential well is opened at approximately 9.1 seconds. The approximate 3-second wide response is likely due to the Coulombic repulsion forces happening inside the trap which are delaying the acceleration of ions during the potential well opening process. There are two separate peaks that are visible in the plot. The first small peak is due to the ions that are trapped closest to the MCP. These ions are uninhibitedly forced forward as soon as the potential well is opened by both Coulomb forces between the trapped ions and the accelerating gradient that is created as the back of the potential well approaches ground. The majority of trapped ions arrive at the MCP in approximately one second after the potential well is opened with a tail at the end that designates the ions trapped furthest from the MCP are decelerated by the Coulomb forces from the ions closer to the MCP while the gradient after opening the potential well accelerates the ions forward. Depending on the configuration of the settings on the ion trap, the number of ions inside the two peaks can change. For instance, if the gradient on the potential well is more subtle, the confinement along the beam path line decreases, therefore the ions will escape faster. This will increase the intensity of the first peak and reduce the intensity of the second peak.

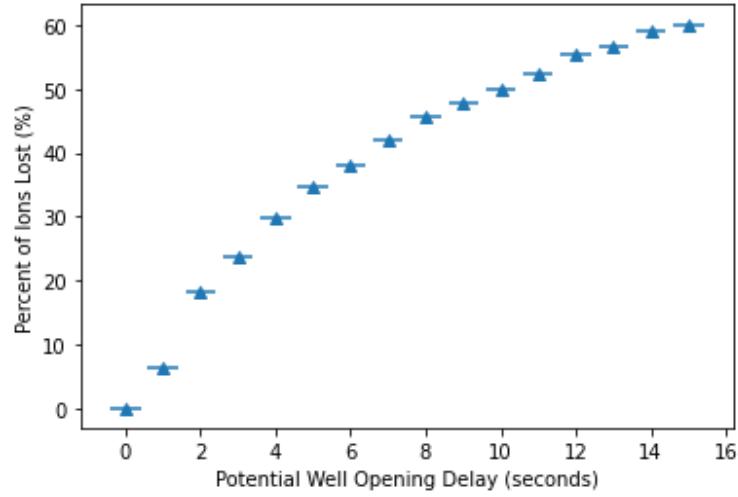


Figure 5.6: A plot of the percentage of ions lost inside the ion trap while delaying the opening of the potential well before pumping excess water vapor out. This dataset was taken with 4 hours of water accumulation. This plot also highlights the half-life of the ion trap which is associated with the amount of potential ions that can be trapped. A  $\pm 0.4$  second time uncertainty is associated due to manual operation of the potentials. The associated uncertainty in percentage of ions lost is not seen due to large counting statistics. At a delay of 15 seconds, the uncertainty is 0.29% while at 1 second the uncertainty is 0.03%.

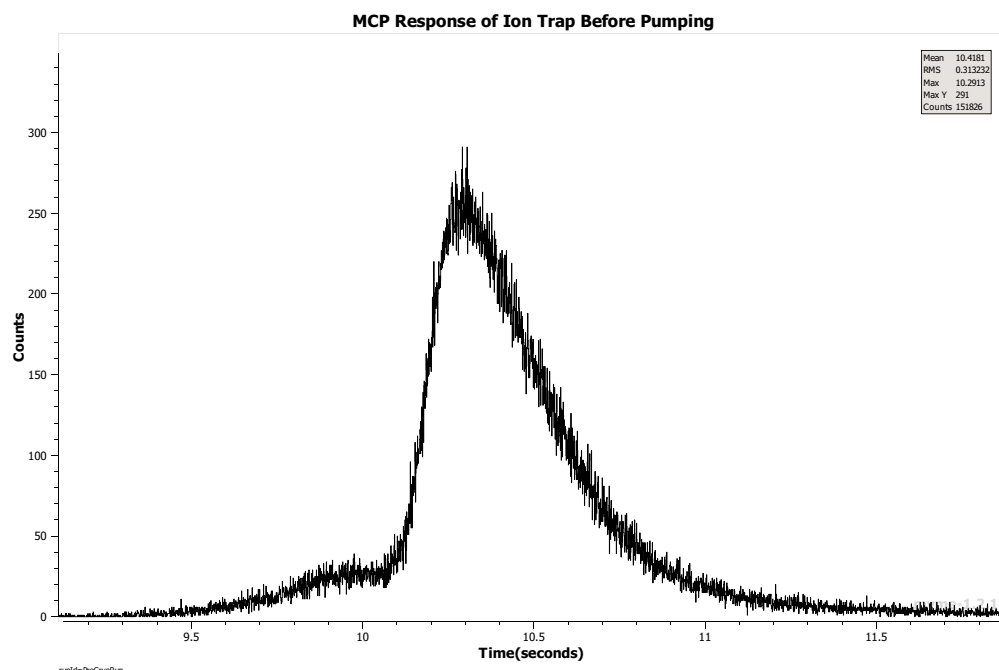


Figure 5.7: The MCP response after opening the potential well on the ion trap after 1-minute of loading time with 2-hours accumulation of water vapor. The entire response is approximately 3 seconds long with the large majority of ions arriving at the MCP after approximately 1.5 seconds after the potential well is opened. This data is considered raw data and is not corrected for the assumed 50% MCP detection efficiency.

#### 5.1.4 Results with Cryopump Addition

The cryopump was purchased in an effort to pump constantly during trapping operation in order to reduce the partial pressures of water vapor, nitrogen, and oxygen to a minimum. Cryopumps operate at a temperature of 20 K and condense gaseous ions that contact the surface. The lightest gases, such as helium, do not condense at 20 K. However, they do adsorb onto the side of the cryopump. This creates a slight problem. With an ion trapping region pressure of approximately 6-8 mTorr, a build-up of adsorbed helium onto the sides of the cryopump occurs after approximately 20 minutes. This causes the backing pressure of the cryopump to increase until it is unable to continue pumping efficiently. At this time, the outside of the cryopump becomes encased in frost due to the cooling of the adsorbed helium which also cools the casing of the cryopump. Therefore, the cryopump is unable to run continuously during trapping operation. However, the cryopump was able to serve another purpose. Since it was shown in Figure 5.5 that the pres-



sures of residual gases increase over time, all turbomolecular pumps and the cryopump were operated occasionally throughout trapping operation to pump out and minimize the amount of background gases throughout the vacuum chambers. This had a positive effect on the amount of  $^{229}\text{Th}^{3+}$  ions trapped as well as the half-life of the trap itself. Figure 5.8 shows a comparison between the half-life measurement of the ion trap between 4 hours of water vapor outgassing and having the water vapor pumped out for 5 minutes after each consecutive measurement. The half-life of the ion trap increases by approximately  $17.03\% \pm 2.47\%$  after pumping on the ion trap using the cryopump and the two 330 L/s side turbomolecular pumps to  $12.79 \text{ seconds} \pm 0.76 \text{ seconds}$ . The uncertainty associated with the half-life measurement has taken into account the flat assumed uncertainty of 0.4 seconds for timing as well as the uncertainty of the number of ions lost. Using the data from both half-life measurements, the amount of ions trapped also increased approximately  $36.50\% \pm 0.13\%$ , from 274,000 to 374,000 ions. The small error is associated with the counting statistics of the amount of ions trapped. An ion trap of  $374,000 \pm 612$   $^{229}\text{Th}^{3+}$  ions is now the largest ion trap created for the purpose of determining the transition energy of  $^{229\text{m}}\text{Th}$  [20; 36]. Figure 5.9 shows the MCP response of the release of the ion trap after pumping out the residual gases. The shape of the response is similar to that of Figure 5.7 where 2 hours of water vapor accumulation occurred, although the amount of ions trapped increased by approximately  $15.5\% \pm 0.06\%$ .

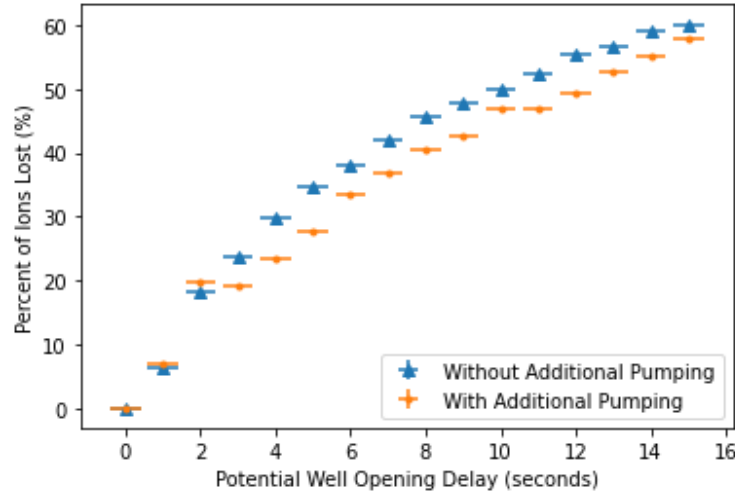


Figure 5.8: A comparison of the percentage of ions lost inside the ion trap while delaying the opening of the potential well. The "Without Additional Pumping" dataset comes from Figure 5.6. The "With Additional Pumping" dataset was taken after an initial 30 minutes of pumping with the cryopump and the two 330 L/s turbomolecular pumps. After that, 5 minutes of extra pumping was performed after each individual measurement to keep the water vapor inside the ion trap at a minimum. The trap half-life increased  $17.03\% \pm 2.47\%$  and amount of ions trapped increased  $36.50\% \pm 0.13\%$  after pumping out the residual gases. A  $\pm 0.4$  second time uncertainty is associated due to manual operation of the potentials. The associated uncertainty in percentage of ions lost is not seen due to large counting statistics. At a delay of 15 seconds, the uncertainty is 0.25% while at 1 second the uncertainty is 0.02%.

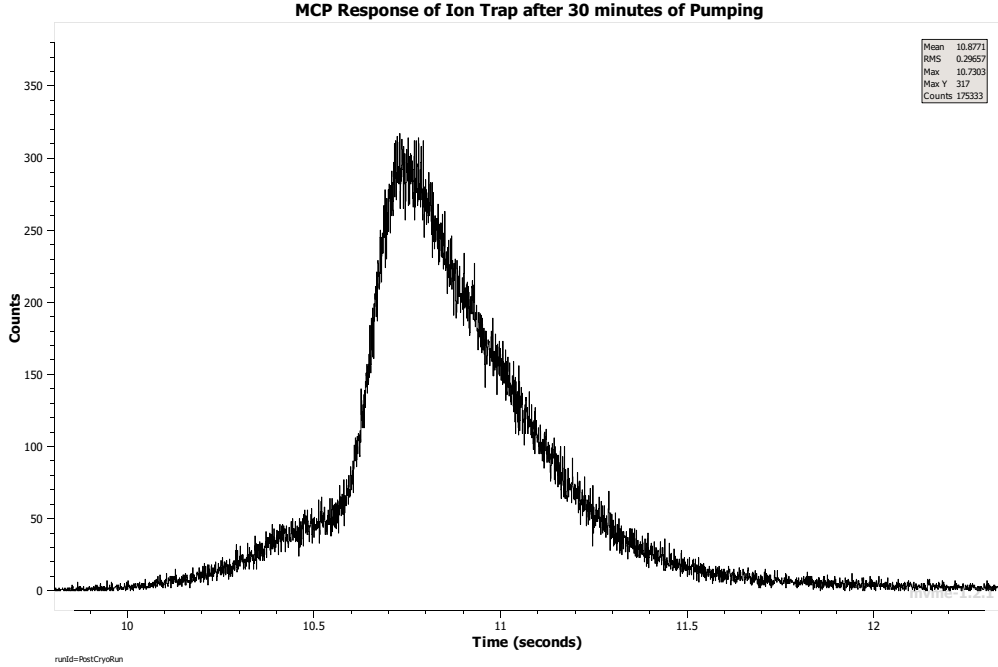


Figure 5.9: The similar MCP response after pumping on the entire chamber for 20 minutes and then loading the trap for 1 minute. Notice the peak is taller than the peak in Figure 5.7 with the same features. There is a  $15.5\% \pm 0.05\%$  increase in the number of ions trapped between this run and the run from Figure 5.7. This data is considered raw data and is not corrected for the assumed 50% MCP detection efficiency.

## 5.2 Laser Excitation Results

The results in this section will cover the ability to atomically excite the  $^{229}\text{Th}^{3+}$  transitions mentioned in Figure 3.9 in order to confirm and quantify the contents of the ion trap. It was shown in Section 4.6 that the amount of scattered light off of just one window from a 5 mW power laser with a wavelength of 690 nm is approximately  $8.65 \times 10^{14}$  photons per second. Due to this laser photon scattering issue, the phototube response while operating the lasers for atomic excitation was always saturated with a dead time of greater than 90%. The phototube signal was fed into the discriminator in Figure 4.46 which follows the schematic and ends with the counter. The gate generator used in this setup was set to 200 ns. Due to this, the maximum amount of signals the electronics can output is 50,000,000 events per second. Assuming a worst case scenario of 5% solid angle and 10% detection efficiency, the minimum number of photons that could be detected through the scattering of just one window would approximately be  $4.33 \times 10^{12}$ , which is on the

order of  $10^5$  times more than the maximum signal the discriminator can output. If the phototube is connected to the Mesytec data acquisition system, the maximum amount of pulses that can be processed is 1 in every 12.5 ns, or 80,000,000 events per second. In order to use the current laser optics system to confirm and quantify the contents of the ion trap, a new ion trap chamber will need to be designed to reduce the amount of scattered light by the windows. This new ion trapping chamber design will be discussed in the next chapter.

# Chapter 6

## Conclusions

The results that this thesis provides can be used to improve large ion traps that trap  $^{229}\text{Th}$  ions for laser interrogation purposes. Once a commercial UV laser becomes available at approximately 150 nm wavelength, this laser can be used to scan for the wavelength of the isomeric transition which will provide the energy of the transition to a much higher degree of accuracy than is currently known.

The goal of this thesis was to design and construct a linear Paul ion trap that can trap at least  $10^6$   $^{229}\text{Th}^{3+}$  ions. After correcting for the 2% branching ratio of  $^{233}\text{U}$  decaying to  $^{229m}\text{Th}$ , the 5% solid angle from the center of the ion trap to the phototube used for detection of UV photons, and the 10% efficiency of the phototube, trapping  $10^6$   $^{229}\text{Th}^{3+}$  ions would correspond to 100  $^{229m}\text{Th}$  decays detected per second and should be easily distinguishable from the MCP background response. In order to attain the goal, a custom-made linear ion guide system with mass separation capabilities and a custom-made linear Paul ion trap were created.

### 6.0.1 Custom Linear Ion Guide System

The linear ion guide system starts with a 10  $\mu\text{Ci}$   $^{233}\text{U}$  source that emits approximately 137,000 thorium ions of different charge states forward into a gas stopping cell that is filled with 99.9999% ultrahigh purity helium to favor creation of  $^{229}\text{Th}^{3+}$  ions (Section 4.5.2). The favorable creation of  $^{229}\text{Th}^{3+}$  as well as its inability to decay by internal conversion (Section 3.2.2) is the reason the 3+

charge state was chosen for the contents of the ion beam. These ions, as well as other positively charged ions and alpha particles, are initialized into a beam using a two-dimensional RF funnel called an RF carpet, located at the end of the gas stopping cell, and ejected into a quadrupole ion beam guide. This quadrupole ion beam guide contains a mass separation quadrupole that allows only transmission of  $^{229}\text{Th}^{3+}$  ions which are then guided into the linear Paul ion trap (Section 4.5.3). The intensity and energy of the  $^{229}\text{Th}^{3+}$  beam entering the ion trap varies on the gas pressures of the various sections of the apparatus. Different studies were performed to determine these operating pressures (Section 4.5). With the operating pressures shown in Figure 4.1, the intensity of the beam entering the ion trap was 80,000 - 90,000  $^{229}\text{Th}^{3+}$  ions per second and the energy of the incoming  $^{229}\text{Th}^{3+}$  ion beam was 3 - 4.5 eV.

## 6.0.2 Linear Paul Ion Trap

Two different linear Paul ion trap designs were created during the work of this thesis. The housing of the ion trap, which remains unchanged with both ion trap designs, was designed with LiF and  $\text{MgF}_2$  windows that transmit 95% of UV photons which will allow transmission of the gamma ray (approximately 150 nm) emitted during the decay of  $^{229\text{m}}\text{Th}$ . The first ion trap design was unable to contain the  $^{229}\text{Th}^{3+}$  ions during the trapping process (Section 5.1.2). This was due to an insignificant  $r/r_0$  value of 0.352. The optimum  $r/r_0$  value referenced by Douglas [18] was shown to be 1.128. The intensity of the ion beam entering the ion trap while using this design was approximately  $80,000 \pm 283$   $^{229}\text{Th}^{3+}$  ions per second. The energy of the beam was greater than 500 eV. This lead to ionization of the helium gas and a contamination of the contents inside the ion trap. Therefore, the contents of the ion trap will contain positively-charged helium as well as  $^{229}\text{Th}^{3+}$ ,  $^{229}\text{Th}^{2+}$ , and  $^{229}\text{Th}^{1+}$ .

The redesigned ion trap utilized thin, rectangular electrodes that decreased the distance from the center of the ion trap to the electrodes which increased the containment of ions inside the ion trap. A 1 mm thick rectangular electrode at  $45^\circ$  perpendicular to the beam path meant that the solid angle from the center of the ion trap to the windows was now limited by the width of the windows and not the placement of the cylindrical electrodes from the previous ion trap

design. The energy of the  $^{229}\text{Th}^{3+}$  ions while using this design was reduced to 3 - 4.5 eV to inhibit ionization of the helium gas after the mass separation quadrupole while maintaining a  $^{229}\text{Th}^{3+}$  ion beam intensity of  $90,000 \pm 300$  ions per second. This was done by limiting the amount of pumping in the main chamber. A near equalization of gas pressures between the main vacuum chamber, 7.8 - 8.9 mTorr, and the ion trapping region, 6.2 - 7.1 mTorr, was attained. However, a water vapor outgassing issue occurred due to the limited amount of pumping across the apparatus. Due to this outgassing issue, the maximum number of  $^{229}\text{Th}^{3+}$  ions trapped was  $274,000 \pm 523$  ions with a trap half-life of  $10.30 \pm 0.77$  seconds (Section 5.1.3). To combat this issue, a cryopump was purchased and the entire chamber was pumped out for 5-minutes before each measurement in order to remove the excess water vapor. The maximum number of  $^{229}\text{Th}^{3+}$  ions trapped using the redesigned ion trap with the extra pumping was  $374,000 \pm 612$  ions (Section 5.1.4). The half-life of the ion trap also increased to  $12.79 \text{ seconds} \pm 0.76 \text{ seconds}$ , which allows more ions inside the trap before an equilibrium is approached. This is the largest ion trap created specifically for the energy determination of  $^{229\text{m}}\text{Th}$  by a factor of 200 [20; 36]. One of the reasons this ion trap design traps 200 times more ions is due its continuous loading versus other ion traps that only trap small bunches of ions for spectroscopic purposes. The quick load and release method of ion trapping is used as a way to measure half-lives of isomeric states, lifetimes of atomic states, and hyperfine states. A continuously loading ion trap such as the ion trap constructed for this thesis is used to detect the lowest lying nuclear transition in  $^{229}\text{Th}$  without the use of lasers as well as increasing the statistics when laser interrogation is used due to more ions being excited.

### 6.0.3 Additional Improvements

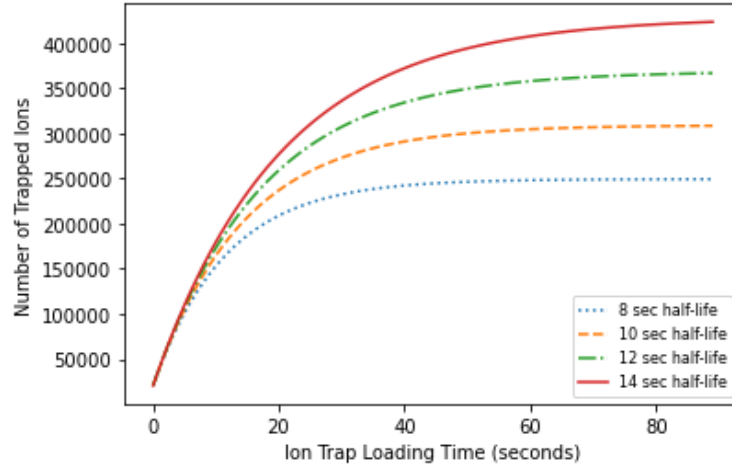


Figure 6.1: A plot showing the maximum estimation of the number of  $^{229}\text{Th}^{3+}$  ions trapped for various half-lives. The lines from top to bottom correspond to ion trap half-lives of 14, 12, 10, and 8 seconds, respectively. An increase of half-life from 12.79 seconds to 14 seconds will increase the number of ions trapped by 16% to 433,000 ions.

Further improvement to the half-life of the ion trap would increase the number of  $^{229}\text{Th}^{3+}$  ions trapped. Figure 6.1 was created using a model that estimates the number of  $^{229}\text{Th}^{3+}$  ions trapped using the redesigned vane ion trap. The number of ions being trapped can be expressed as a summation over time using the incoming  $90,000 \pm 300$   $^{229}\text{Th}^{3+}$  ion beam rate with various half-lives of the ion trap,  $\sum_{n=1}^{\infty} 90,000 \exp(-\lambda t)$ . A correction factor of 0.23 has been applied that corresponds to the trapping efficiency of the ion trap. This trapping efficiency was calculated by comparing the model with the experimental data. An increase of the ion trap half-life from 12.79 seconds to 14 seconds will increase the number of ions trapped by approximately 16% which would increase the number of ions trapped from approximately 374,000 ions to 433,000 ions. It has been proposed that in order to increase the half-life of the ion trap further, the materials inside the apparatus that attribute to the outgassing of water vapor must be removed. These materials include the polyetheretherketone (PEEK) material that holds the quadrupole rods, the Kapton sheets used to insulate the chamber of the gas stopping cell from the electrodes, Kapton tape used to hold wires in place, and various ceramics that insulate simultaneous electrodes.

The MCP used for charged particle detection in this thesis has an assumed 50% efficiency



(Section 4.3.5). An efficiency study was performed in vacuum and in various pressures. However, due to the inability to measure the distance from the MCP to the alpha source precisely, the measured efficiency of the MCP varied between 24% and 47%. Having a stable plunging system with the ability to measure the distance between the alpha source and the MCP would allow a more accurate efficiency characterization. Due to the conservative assumption of an efficiency of 50%, a precise efficiency characterization will likely entail a lower efficiency which will increase the calculation of the number of  $^{229}\text{Th}^{3+}$  ions trapped. For example, if the MCP efficiency was 25%, the total number of  $^{229}\text{Th}^{3+}$  ions trapped after correction for MCP efficiency would be 748,000 ions instead of 374,000 ions.

Additional improvements are in the design process but are beyond the scope of this thesis. A larger ion trapping chamber has been proposed and will allow use of the current atomic laser optics setup while reducing the number of scattered photons. The inside of the new ion trapping chamber will be coated with a photon absorbing substance to absorb scattered photons. The distance between the center of the ion trap and the LiF and  $\text{MgF}_2$  windows on the larger ion trapping chamber will be increased which will decrease the solid angle between the windows and the phototube. A laser beam dump will also be inserted inside the ion trapping chamber which can be used to decrease the amount of windows the laser will need to transmit through, further decreasing the potential background scattered laser photons. The current setup uses multiple inputs for DC bias and RF voltage potentials. In order to automate the ion trapping process and eliminate human uncertainty, the DC and RF voltages would need to be directed through a single programmable mechanism. An example of human uncertainty is the time it takes to manually raise the voltage potential of the orifice plate to stop the beam from entering the ion trap, start a scalar measurement, and then lower the back end of the potential well to ground on the ion trap, simultaneously.

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