

Charge States of  $^{229\text{m}}\text{Th}$ : Path to Finding the Half-Life

Molly A. Wakeling

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Dr. Jason T. Burke (LLNL) and Dr. Steven L. Tomsovic (WSU)

Department of Physics and Astronomy

College of Arts and Sciences

## Précis

The Universe is governed by the laws of physics and fundamental constants that are assumed to be unchanging and homogeneous throughout all of space and time. However, there is some physical evidence from cosmology and radioactive decay of elements here on Earth that suggests that the fine structure constant, referred to simply as  $\alpha$  by physicists, may not actually be constant in time. This would have far-reaching implications throughout science. A way to determine whether the fine structure constant is changing in time is to construct a nuclear clock, which is similar in principle to the current atomic clock, but much more accurate. The reason it is more accurate is because a nuclear clock would be based on the transitions of an atomic nucleus between energy levels, rather than the transitions of electrons between energy levels; nuclear transition levels are much finer than electron transitions. One element in particular is a good candidate for building a nuclear clock: the isomer of thorium-229, written as  $^{229\text{m}}\text{Th}$ . This isomer is special because it has an unusually low first excited nuclear state, the lowest of any nuclide known, at only 7.6 eV (electron volts). Typically, nuclear energies are measured in millions of electron volts. This low energy would allow  $^{229}\text{Th}$  nuclei to be excited to the isomeric state using a table-top laser because of its low energy. In order to use it for a nuclear clock, however, the half-life of the isomer must be known in order to know which frequency to tune the lasers. However, in the 35 years that this state has been known, no one has yet been able to observe its half-life. Before the half-life can be observed, it is necessary to know what kind of radiation  $^{229\text{m}}\text{Th}$  emits when it decays to the ground state (its lowest energy state). Based on the electron configuration of  $^{229\text{m}}\text{Th}$ , there are two ways that it can decay: by internal conversion, which would release low-energy electrons, or by bound internal conversion, which would release photons in the ultraviolet-optical range, ideal for detection.  $^{229}\text{Th}$  decays from

uranium-233 (written as  $^{233}\text{U}$ ) by alpha decay, and it may be emitted either in a charged state or electrically neutral, depending on whether it loses electrons in the process. If  $^{229}\text{Th}$  is emitted in an electrically neutral state (meaning it has all of its electrons), then the isomer will decay to the ground state by internal conversion, which will emit low-energy electrons. However,  $^{229}\text{Th}$  is emitted in a positively charged state (meaning it is missing electrons), then it will decay by bound internal conversion, which will emit light. In order to determine then whether  $^{229}\text{Th}$  is charged or neutral after decaying from  $^{233}\text{U}$ , I performed a time-of-flight experiment. This type of experiment measures the time it takes particles to fly from one point to another, and then the times are compared for different environments to see how those environments affect the time-of-flight. In my experiment, the  $^{229}\text{Th}$  nuclei flew through an electric field with different voltages. If the nuclei were electrically neutral, they would not be affected by changes in the electric field. However, if they were positively charged, then they should have different times-of-flight for different voltages; they would either be sped up or slowed down, and those times can be compared. After conducting my experiment, I was able to determine that the  $^{229}\text{Th}$  nuclei were indeed positively charged, which means that the isomers should decay to the ground state by bound internal conversion, emitting light in the process. By observing the emitted light, we should be able to measure the half-life of the isomer, which is the next step. After the half-life is observed, a nuclear clock can be built, and when compared with the atomic clock, we will know whether the fine structure constant is, indeed, a constant. Knowing the half-life of  $^{229\text{m}}\text{Th}$  will also allow scientists to probe relationships between electrons and their nuclei, an area of physics that is typically difficult to do because of the large differences in energy between electrons and nuclei. This research could potentially open up an area of physics that is not well understood, and could also eventually show whether some fundamental constants are indeed constant.

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## I. Introduction

It is taken for granted in the sciences that the fundamental constants are indeed unchanging in time. However, data from the study of cosmology exist that show that the fine structure constant may actually be changing with time. The fine structure constant, simply known as  $\alpha$  among physicists, is a universal fundamental constant that determines the values of other fundamental constants, such as the charge of an electron, the strength of the forces of gravity and electromagnetism, etc. Hints of this variation have been seen in quasar absorption spectra, Big Bang nucleosynthesis, and Oklo natural nuclear reactor data.<sup>1</sup> If  $\alpha$  were indeed varying in time, this would have far-reaching implications in many areas of science. This temporal variation could be measured with a nuclear clock, which is similar in concept to the familiar atomic clock. A nuclear clock would rely on the much-narrower energy transitions of the nuclei of atoms, as opposed to the current atomic clock, which relies on wider transitions of electron energies.<sup>2</sup> A good candidate element for use in a nuclear clock is the thorium-229 isomer (abbreviated as  $^{229m}\text{Th}$ , where the  $m$  stands for “metastable”), which has the lowest-lying first excited nuclear state of any nuclide known, at  $7.6 \pm 0.5$  eV (electron volt, a unit of energy based on the energy of a single electron).<sup>3</sup> An isomer is a metastable nuclear state that is created when one or more of the nucleons of an atom (protons or neutrons) is boosted into a higher-energy or “excited” state; the lifetime of isomers is typically many times longer than ordinary “prompt” half-lives, which can be as short as  $10^{-12}$  seconds. This 7.6 eV energy level is anomalously low compared to typical nuclear energies that are in the MeV range, or millions of electron volts. The energy of this level was determined by measuring the energies of the gamma

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<sup>1</sup> Uzan, “The Fundamental Constants and Their Variation.”

<sup>2</sup> Peik and Tamm, “Nuclear Laser Spectroscopy of the 3.5 eV Transition in Th-229.”

<sup>3</sup> Beck et al., “Energy Splitting of the Ground-State Doublet in the Nucleus  $^{229}\text{Th}$ .”

rays, a type of radiation from nuclear decay, emitted as the  $^{229}\text{Th}$  nucleus decays to the ground state (see Fig. 1) and subtracting the differences in the energy levels, which led to the theorization of a low-lying isomeric state.<sup>4</sup> Subsequent studies of gamma-ray emissions from  $^{229}\text{Th}$  continued to improve the accuracy of the value of this excited isomeric state, until its current accepted value was calculated to be  $7.6 \pm 0.5$  eV in 2007.<sup>5</sup>

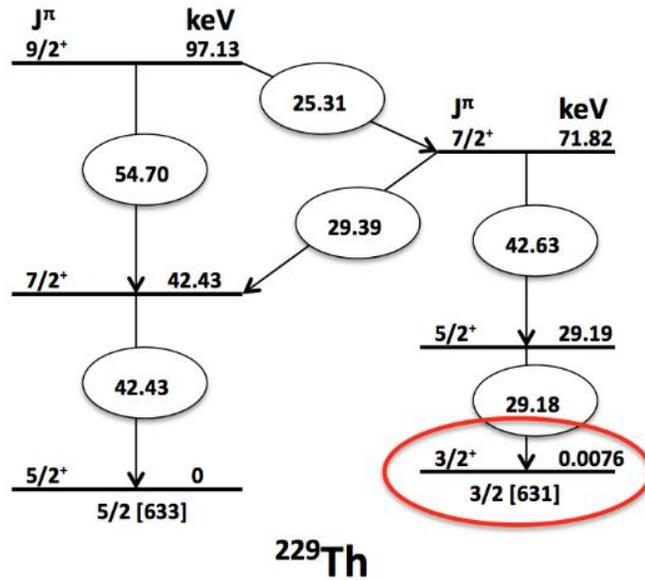


Figure 1: Partial level scheme for  $^{229}\text{Th}$ , with the isomer circled (based on Fig. 1 in Beck et al.)

$^{229}\text{Th}$  is created when uranium-233 (abbreviated  $^{233}\text{U}$ ) undergoes alpha decay, in which a  $^{233}\text{U}$  atom decays into  $^{229}\text{Th}$  and an  $\alpha$  particle (a helium atom, not to be confused with the fine structure constant symbol  $\alpha$ ), and the isomer has a probability of approximately 2% of being populated from the  $\alpha$ -decay of  $^{233}\text{U}$ .<sup>6</sup> Because of the isomer's unusually low nuclear energy, it is possible to excite  $^{229}\text{Th}$  into its isomeric state using an optical excitation source, such as the laser that would be used in a nuclear clock.<sup>7</sup>

<sup>4</sup> Kroger and Reich, "Features of the Low-energy Level Scheme of  $^{229}\text{Th}$  as Observed in the  $\alpha$ -decay of  $^{233}\text{U}$ ."

<sup>5</sup> Beck et al., "Energy Splitting of the Ground-State Doublet in the Nucleus  $^{229}\text{Th}$ ."

<sup>6</sup> Browne et al., "Search for Decay of the 3.5-eV Level in  $^{229}\text{Th}$ ."

<sup>7</sup> Burke et al., "Additional Evidence for the Proposed Excited State at  $\leq 5$  eV in  $^{229}\text{Th}$ ."

In order to observe the temporal variation of  $\alpha$ , one would measure the ratio of the  $^{229\text{m}}\text{Th}$  transition frequency to another narrow optical or microwave transition, such as the current standard of the cesium atomic clock. A  $^{229\text{m}}\text{Th}$  clock would rely on a nuclear transition, which means that the frequency of that transition would be dominated by what is known as the strong nuclear force (the force that holds protons and neutrons together). Comparing a nuclear clock with the current cesium atomic clock standard, which is based on the atomic transition and thus is dominated by the electromagnetic force, would allow measurement of variations of the ratio between the electromagnetic and strong-coupling constants,<sup>8</sup> and thus the temporal variation of  $\alpha$ . It would be possible to increase the sensitivity of the current atomic clock limits of the variation of fundamental constants like  $\alpha$  by several orders of magnitude (from  $10^{-15}$  per year to  $10^{-20}$  per year) using a nuclear clock.<sup>9</sup> This is possible because of the extremely narrow frequency bandwidth of the  $^{229\text{m}}\text{Th}$  transition, possibly on the order of  $10^{-4}$  Hz<sup>10</sup> -  $10^{-6}$  Hz<sup>11</sup> (Hertz, unit of frequency measurement, in this case transitions per second), as opposed to the width of atomic clock transitions, which are typically 1-100 Hz.<sup>12</sup> The much-narrower range of frequencies in a nuclear clock is what allows it to be more accurate than the atomic clock. In order to make a nuclear clock, however, the half-life of  $^{229\text{m}}\text{Th}$  deexcitation, or transition from the excited state of the nucleus to the ground (lowest energy) state, must be known. Although this low-lying nuclear state was predicted in the 1970s, no one has yet been able to directly observe the half-life of the isomer. It is necessary to know the half-life because according to the

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<sup>8</sup> Peik and Tamm, "Nuclear Laser Spectroscopy of the 3.5 eV Transition in Th-229."

<sup>9</sup> Flambaum, "Enhanced Effect of Temporal Variation of the Fine Structure Constant and the Strong Interaction in  $^{229}\text{Th}$ ."

<sup>10</sup> Tkalya, Zherikhin, and Zhudov, "Decay of the Low-energy Nuclear Isomer  $^{229}\text{Th}^{\text{m}}$  ( $3/2^+$ ,  $3.5\pm 1.0$  eV) in Solids (dielectrics and Metals)."

<sup>11</sup> Peik and Tamm, "Nuclear Laser Spectroscopy of the 3.5 eV Transition in Th-229."

<sup>12</sup> Flambaum, "Enhanced Effect of Temporal Variation of the Fine Structure Constant and the Strong Interaction in  $^{229}\text{Th}$ ."

Heisenberg uncertainty principle, the half-life of a state is inversely proportional to its energy width, by  $\Delta E \Delta t = \hbar/2$  (where  $\hbar$  is the Planck constant divided by  $2\pi$ , by convention). Nuclear isomers have half-lives that are much longer than other excited states, on the order of nanoseconds or longer, as opposed to the prompt gamma decay that occurs in the other states. If  $^{229\text{m}}\text{Th}$  has a half-life on the order of microseconds or longer, then it will have an extremely narrow energy width, making it usable as a new frequency standard to replace the current cesium atomic clock standard of measuring time. Because the nuclear transition is so narrow, the half-life (and thus the energy width) must be known in order to tune the excitation laser frequency to the exact value of the nuclear transition frequency.

In order to observe the deexcitation of the  $^{229\text{m}}\text{Th}$  isomer, it is necessary to know whether it decays by internal conversion or bound internal conversion. In the case of internal conversion, the isomer will relax by ejecting an outer electron. Since the energy needed to eject the first electron out of  $^{229}\text{Th}$  (the first ionization energy) is 6.3 eV,<sup>13</sup> the ejected electron will be very low-energy, at only 1.3 eV, and thus difficult to detect. Internal conversion will occur if the isomer is emitted from  $^{233}\text{U}$  decay in a neutral charge state, meaning that it has an equal number of electrons and protons. If the isomer is emitted in a 1+ or higher charge state, however, meaning that it is missing one or more of its electrons, the second ionization energy is 11.5 eV, which is more than the energy of the isomer (7.6 eV). Because of this, internal conversion is energetically forbidden for any positively charged state. Instead, bound internal conversion will occur, where a lower-lying electron will jump up to a higher-energy (or “excited”) state, but not make it all the way out of the shell, and then other electrons will cascade down to fill the

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<sup>13</sup> Köhler et al., “Determination of the First Ionization Potential of Actinide Elements by Resonance Ionization Mass Spectroscopy.”

vacancy. This will release a photon in the UV (ultraviolet)-optical range,<sup>14</sup> which is arguably easier to observe than a low-energy electron. In order to observe whether  $^{229}\text{Th}$  decays from  $^{233}\text{U}$  in a positively charged or neutral charge state, a time-of-flight (TOF) experiment was performed. By varying the electric field inside of a vacuum chamber, it can be observed that the  $^{229}\text{Th}$  nuclei will either have varying TOFs in the case of being charged, or will always have the same TOF in the case of being electrically neutral. This experiment found  $^{229}\text{Th}$  to be in 1+ and higher charge states after  $\alpha$ -decay from  $^{233}\text{U}$ .

## II. Methods

An electroplated 0.2  $\mu\text{Ci}$  (micro-Curie, unit of radioactivity) sample of  $^{233}\text{U}$  was placed inside of a vacuum chamber and connected to a high-voltage power supply. The  $^{233}\text{U}$  sample is a very pure 99.9999%  $^{233}\text{U}$  with 0.0001%  $^{232}\text{U}$ , and it was chemically purified to remove daughter nuclei (atoms from farther down the nuclear decay chain). A silicon  $\alpha$ -particle detector was placed 15 cm behind the source; a fine mesh grid that was connected to either ground or high voltage was placed in front of the source; and a microchannel plate detector (MCP) was placed 15.5 cm in front of the source with another fine mesh grounded grid placed directly in front of it. MCPs are used to detect either radiation (such as ultraviolet light and X-rays) or particles such as electrons and ions. The purpose of the shutter was to protect the MCP from being bombarded with the nuclei while the experiment was not running.

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<sup>14</sup> Irwin and Kim, "Observation of Electromagnetic Radiation from Deexcitation of the  $^{229}\text{Th}$  Isomer."

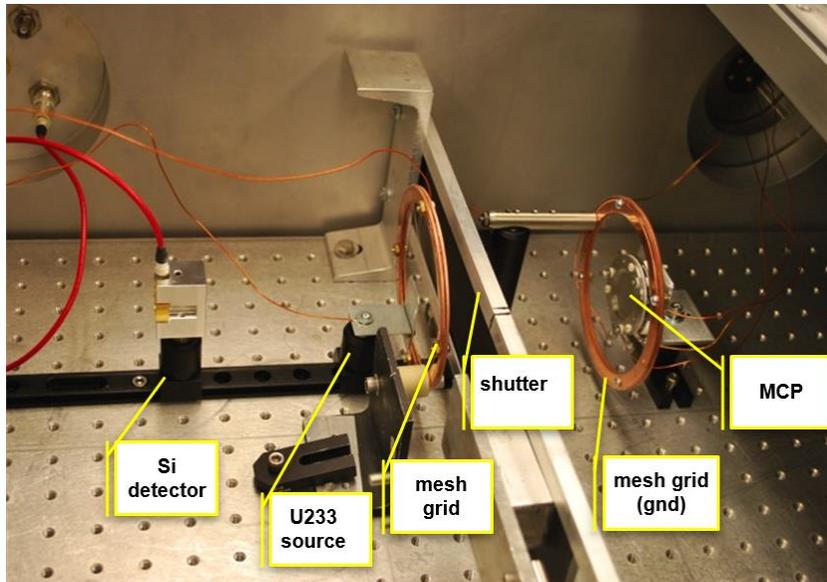


Figure 2: Experimental setup

The vacuum chamber was around  $8 \times 10^{-6}$  Torr (or  $1.5 \times 10^{-7}$  psi), the alpha detector was biased to 155 V (volts), and the MCP was biased to 3550 V. Because the  $^{229}\text{Th}$  nuclei from  $^{233}\text{U}$  decay have an energy of 84 keV (kilo-electron volts, or 1000 eV), the grounded grids would not stop the nuclei from hitting the MCP. The silicon detector signal was the “start” signal for a time-to-amplitude converter (TAC), and the MCP was the “stop” signal. The TAC measures the time difference between the two signals and outputs a voltage that corresponds to that time difference; the scale was set to be 10 V for 2  $\mu\text{s}$  (micro-seconds) full scale (FS). The TAC was connected to a pulse shaper (this changes the shape of the pulse to a Gaussian “bell” curve that the computer can read) and then a multi-channel analyzer (MCA), which was connected to a computer, which showed a plot of channel versus number of counts. Channel numbers could then be converted back to TOFs using a calibration table that was created.

#### a) Calculations

Before performing the experiment, calculations were made to predict the TOF for several different charge states, as well as determine how many of the  $^{229}\text{Th}$  nuclei would escape the  $^{233}\text{U}$

source and how much energy those nuclei might lose. The calculation of the TOF for  $^{229}\text{Th}$  nuclei was based on the non-relativistic kinetic energy equation  $E = \frac{1}{2}mv^2$ , where the energy of the nuclei is 84 keV. This calculation was also performed for no source bias and for -4 kV source bias. The kinetic energy of the recoil nuclei was calculated based on the known mass and energy of the  $\alpha$  particle from  $^{233}\text{U}$  decay, using the principle of conservation of momentum.

	+4 kV	0 V	-4 kV
q = 0	584 ns	584 ns	584 ns
q = +1	571 ns	584 ns	599 ns
q = +2	558 ns	584 ns	614 ns
q = +3	547 ns	584 ns	631 ns
q = +4	536 ns	584 ns	649 ns

Table 1: Predicted TOFs

Because the recoils have energy from the nuclear decay, it is possible for them to be highly ionized; the trend seen in the table continues up to higher charges.

Calculations for how many of the  $^{229}\text{Th}$  nuclei would escape the  $^{233}\text{U}$  source were also performed, using a program called SRIM (Stopping and Range of Ions in Matter). Based on the area of the source ( $1.27 \text{ cm}^2$ ) and its activity ( $0.2 \text{ }\mu\text{Ci}$ ), the source thickness was calculated to be  $259 \text{ \AA}$  (angstroms,  $1 \text{ \AA} = 10^{-10}$  meters). For this setup, the widest angle of recoil nuclei that would hit the MCP and also have an alpha particle hit the silicon detector was about  $5^\circ$ , so this angle of incidence was used for the calculation. Taking ten different starting points along the thickness of the source ( $\text{UO}_2$ , uranium dioxide, was used as the source material formula) and averaging them for 5,000 recoils per run, it was found that 80.65% of the recoils would escape the source. This number indicated that enough thorium recoils would escape to provide meaningful data.

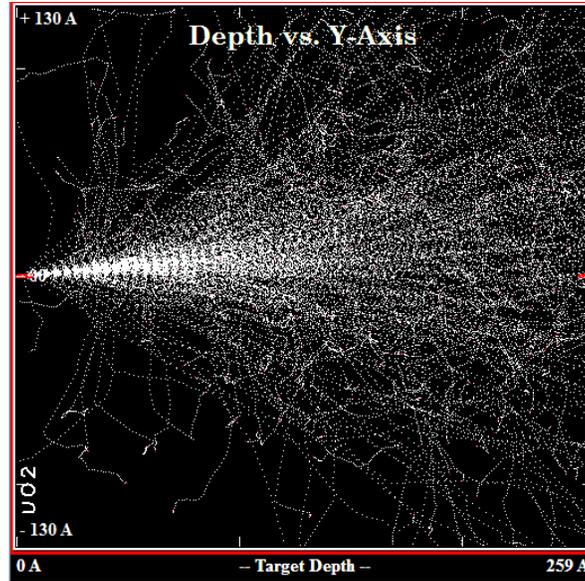


Figure 3: Graphical SRIM output for 1000 recoils

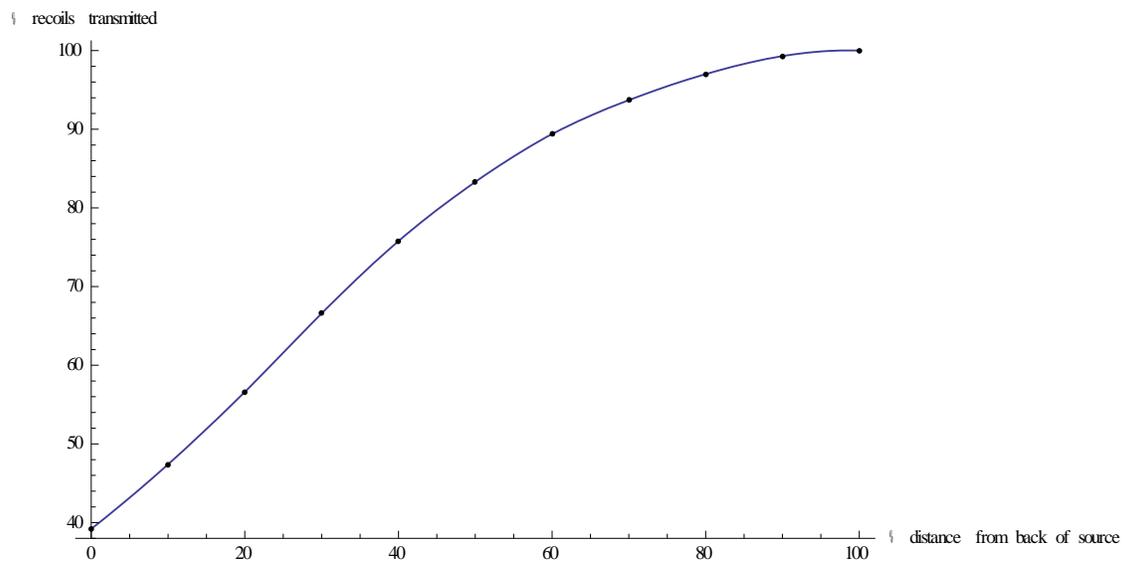


Figure 4: Percent of transmitted recoils through  $^{233}\text{U}$  as a function of decay position within the source

Figure 3 depicts what the paths for 1,000 thorium recoils would look like, if they came from a single point at the “back” of the source, with reference to the MCP. Figure 4 shows the approximate percentage of thorium recoils that are expected to exit the source as a function of how “deep” they are in the source; closer to the back (0%) or closer to the front (100%).

Because the source is not infinitely thin, the  $^{229}\text{Th}$  nuclei lose energy as they escape the source, which affects their TOF. Another calculation was performed, also using SRIM, to observe the magnitude of this effect. The average energy of an emitted recoil varied from 20.91 keV out of the back of the source to the full 84 keV out of the front. The distribution of the straggling energy follows an exponential curve.

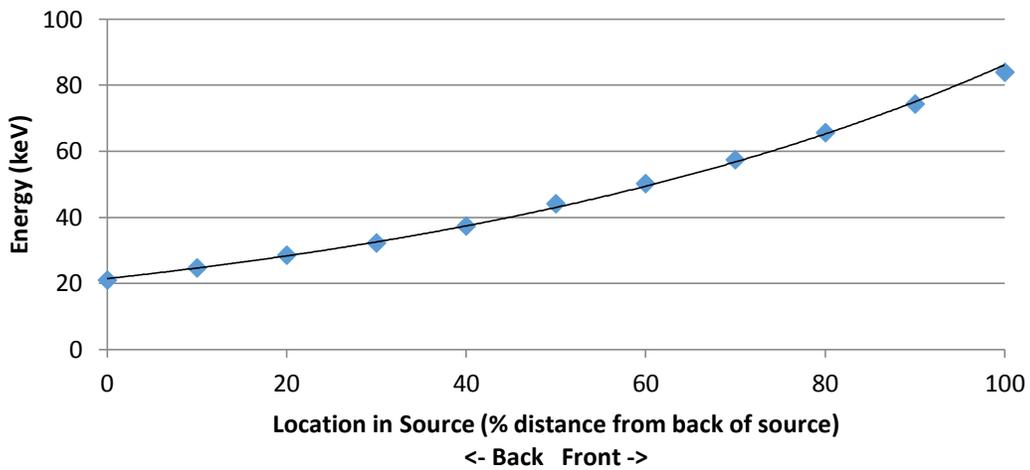


Figure 5: Graph of average straggling energy, as a function of decay position within the source

By multiplying the escape energy of the recoils by their probability of escaping the source with respect to decay position within the source, the shape of the TOF curve (for neutral thorium recoils, in this case) can be graphed:

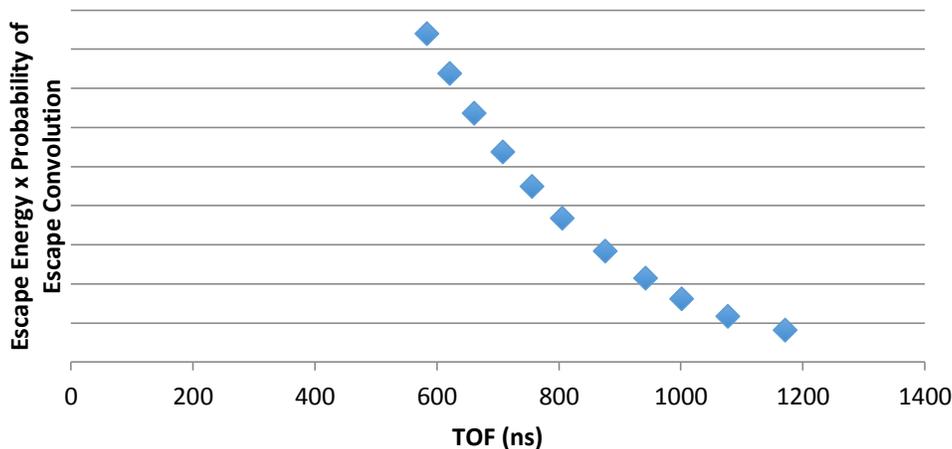


Figure 6: Convolution of transmitted recoils and average escape energy

The shape matches very well with what was seen in the data, which will be shown later.

*b) Experiment*

As mentioned previously, the “start” signal for the TAC was the  $\alpha$ -particle from  $^{233}\text{U}$   $\alpha$ -decay hitting the silicon detector behind the source. The signal from the  $\alpha$ -detector was pre-amplified, sent through a fast amp, then through a discriminator set at 1 V, and then into the “start” side of the TAC. A discriminator cuts out signals below a certain set threshold to reduce noise counts. The “stop” signal was from the  $^{229}\text{Th}$  recoil nuclei hitting the MCP, the signal from which went through a pre-amp, a fast amp, a discriminator set at 260 mV (milli-volts, thousandths of a volt) to reduce noise, a delay generator set to 180 ns (nano-seconds, billionths of a second), and finally into the “stop” side of the TAC. The purpose of the delay generator was to include a set and accounted-for delay in the time between the “start” and “stop” signals because there was an inherent electronics delay in the system. This set delay time was subtracted from the TAC TOF data. The TOF for the  $\alpha$ -particles from the source to the silicon detector was calculated to be 10 ns and was also subtracted from the TOF data. The output of the TAC was a signal on a 10 V, 2  $\mu\text{s}$  FS with voltage corresponding to TOF. These signals were sent through a

pulse shaper set with a peaking time of 3  $\mu\text{s}$  (sufficient for detection by the MCA, and also allowing plenty of electronics recovery time) and then sent through the MCA, which produced the results on the computer. The MCA was set to the maximum 16,324 channels, which allowed a resolution of 0.12 ns per channel, or 8.19 channels per ns, for 2  $\mu\text{s}$  FS on the TAC. This resolution was more than sufficient for this experiment. The TAC was calibrated by replacing the signals from the silicon detector and the MCP with a function generator, and setting the difference between the “start” and “stop” signals to be specific values by using the delay generator. The sharp peaks that were observed at a given channel range from the MCA on the computer corresponded with specific voltages from the TAC, which corresponded to specific times between the “start” and “stop” signals. This calibration was used to convert the channel numbers in the TAC output back to TOFs. The signal count rate for the experiment was 1-2 Hz or counts per second, so runs lasted several hours, and 22 runs total were performed at various configurations. Four main configurations of the electric field were performed: 4 kV source bias and grounded mesh grid, a positive potential (voltage) hill with 2 kV source bias and 4 kV mesh grid bias, -2 kV source bias and grounded mesh grid, and a negative potential hill with -2 kV source bias and -4 kV mesh grid bias. The potential hill configurations were used to block electrons, protons, and higher-mass particles that did not have enough energy to make it over a 2 kV potential hill from adding noise to the results.

### **III. Results and Discussion**

#### *a) Data*

The first several runs of the experiment were used to work out the kinks, such as adjusting the delay signal and the electronics settings, adding the grounded mesh grids, reducing noise, etc.

The first run for collecting meaningful data was Run09, which was 4 kV on the source, with no grid yet in front of the source, but a grounded grid just in front of the MCP.

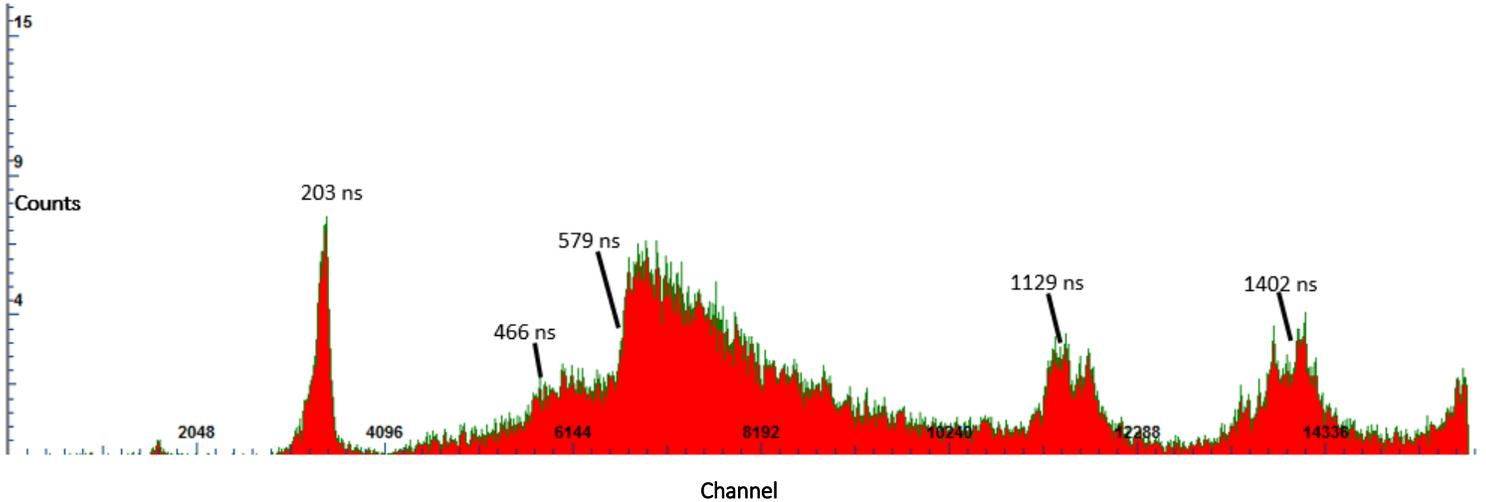


Figure 7: Run09, 4 kV on source, no mesh grid in front of source, ~15 hours

Comparing the values of the peaks to the calculated TOF table, it is easy to see that the third element, the 579 ns peak, is very close to both the  $q=0$  and  $q=+1$  TOFs. There is a long sloped curve after the 579 ns peak – this is the straggling of the recoil nuclei, as predicted by calculation. The next question related to this data set is what the first, very sharp peak is, and what the later peaks are. This will be addressed later.

The next significant data set is Run11, which was a positive potential hill, with 2 kV on the source and 4 kV on the mesh grid in front of the source. A potential hill means a difference in voltage that the particles must overcome in order to make it to the detector. Since the potential hill is 2 kV high, any charged particles with less than 2 keV energy will not have sufficient energy to make it over the hill and hit the MCP detector. The purpose of the potential hill was to see if the kinetic energy of the particles that cause the early, sharp peak and the later peaks was low or high.

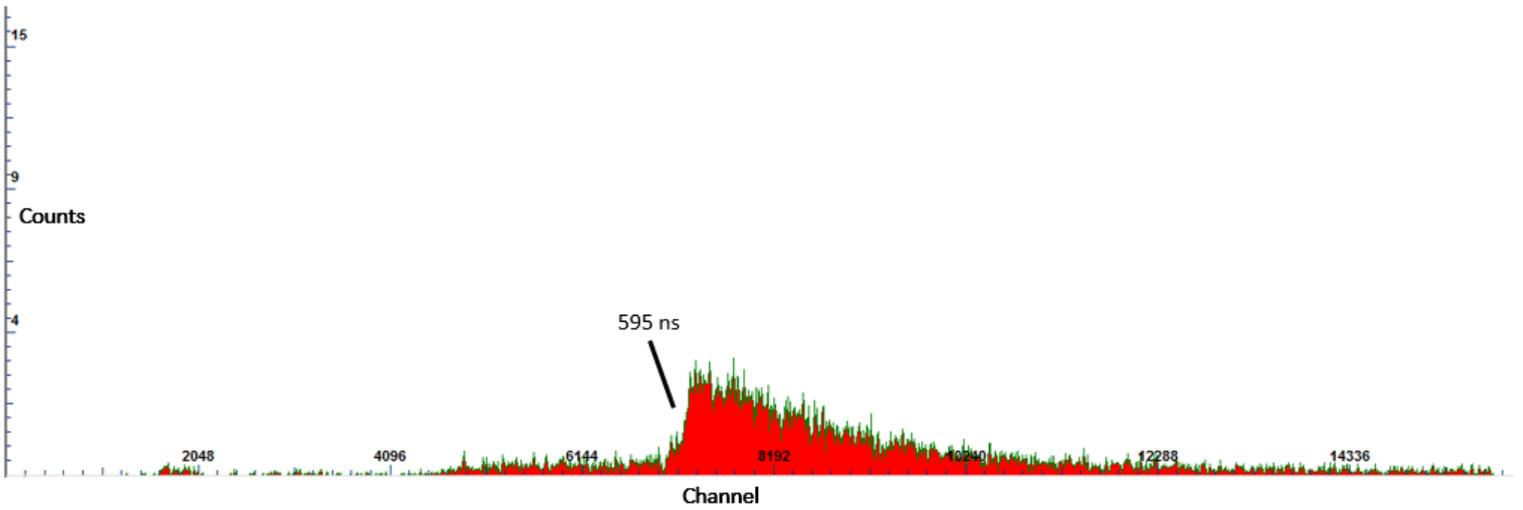


Figure 8: Run11, 2 kV on source, 4 kV on mesh grid

Here, it was observed that adding the potential hill eliminated both the early and later peaks, indicating the particles that cause those peaks are low-energy. The suspected  $^{229}\text{Th}$  peak came a little later than before as well.

The next significant run is Run13. For this run, a negative potential hill was made, with the source bias at -2 kV and the mesh grid at -4 kV. This configuration will block any negatively charged particles that have an energy of less than 2 keV, for the same reason that the positive potential hill blocks low-energy positively charged particles. This negative run reveals that the peak moves to a longer TOF of 601 ns for negative voltages as opposed to the +4 kV bias voltage in Run09. This was the first indicator that the  $^{229}\text{Th}$  recoils might be positively charged.

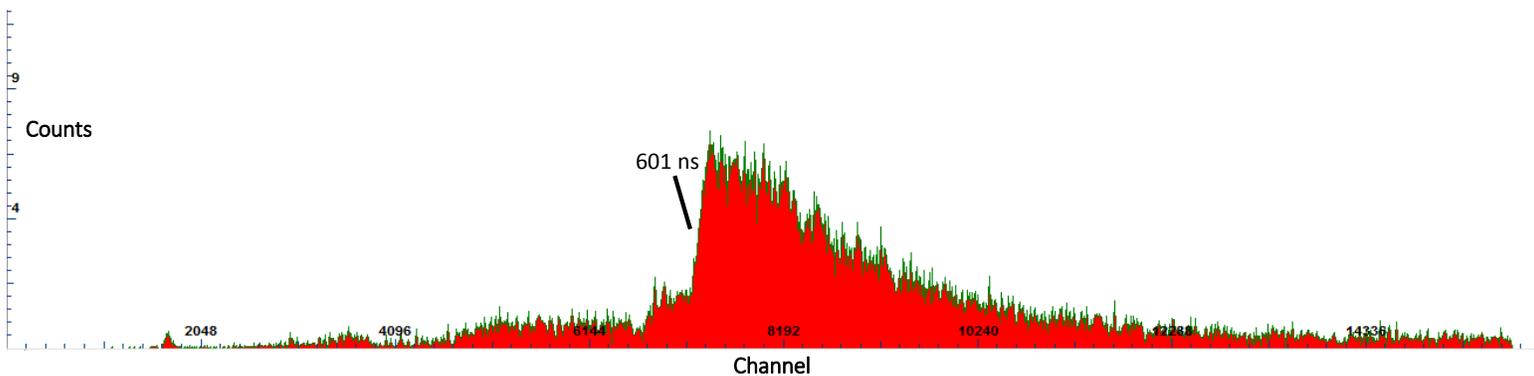


Figure 9: Run13, -2 kV on source, -4 kV on mesh grid

Run14 showed another interesting feature – a very sharp spike at a very fast TOF. This run was done with -2 kV on the source, and the mesh grid was grounded.

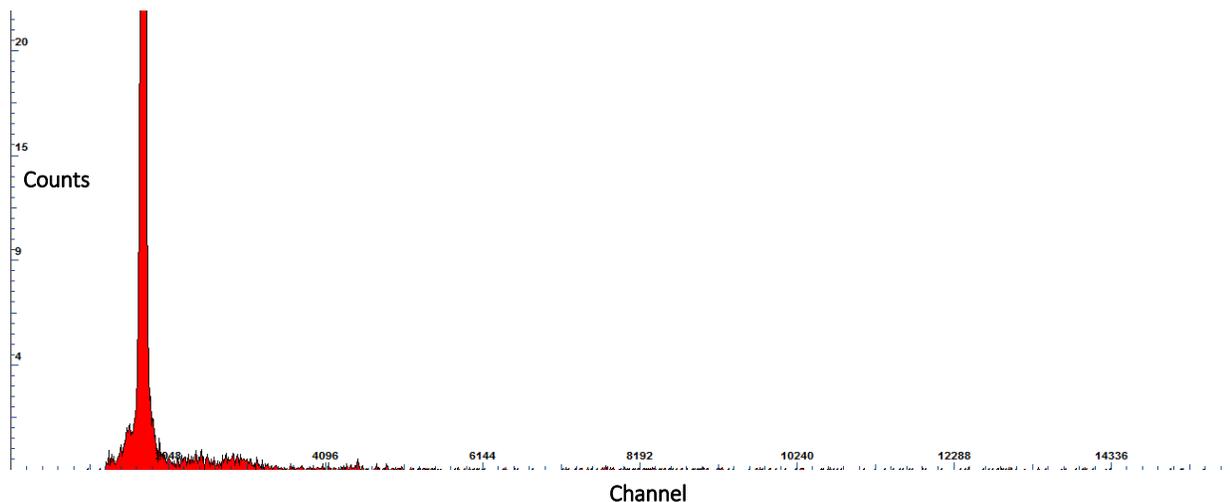


Figure 10: Run14, -2 kV on source, 0 V (gnd) on mesh grid

The particles in the sharp peak above have a TOF of ~2 ns. Suspecting that the particles were electrons, the TOF for an electron in the -2 kV field was calculated to be 5.8 ns, so the peak was assumed to be electrons. Because the electrons “steal” the “stop” counts on the MCP from the later-arriving  $^{229}\text{Th}$  recoils, a negative potential hill had to be used for subsequent negative voltage runs.

In order to better see the TOF differences between different configurations, two spectra were overlaid on the same screen. For instance, the plot below shows Run13 (red), which is for the negative potential hill (-2 kV on the source and -4 kV on the mesh grid), and Run17 (green), which is for +4 kV on the source. Run17 is similar to Run09, but was done after added the mesh grid was added in front of the source in order to reduce the acceleration time of the  $^{229}\text{Th}$  recoils. It is easy to see the differences in the TOFs between the negative and positive electric fields.

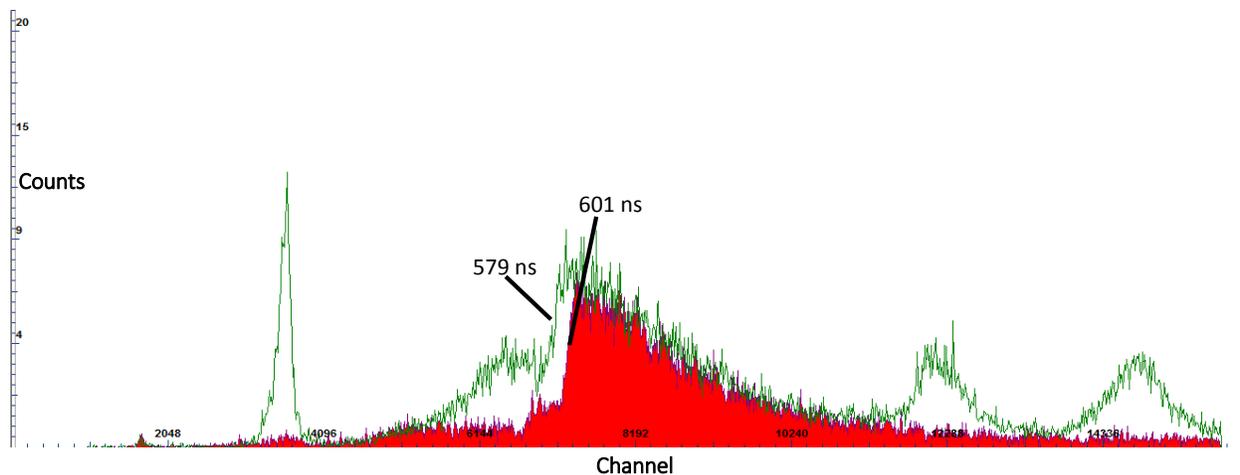


Figure 11: Run13 (red) vs Run17 (green)

The TOF values from these two runs are also very close to those predicted by calculation – for a thorium nucleus of 1+ charge in a +4 kV field, the calculated TOF is 571 ns, and in a -4 kV field, the calculated TOF is 599 ns. This makes for a calculated TOF difference of 28 ns, as compared to the measured TOF difference of 22 ns; these differences are reasonably close to each other.

Based on the data, it is clear that at least a majority of the thorium recoils are positively charged. This is because if the recoils were neutral, they would not be affected whatsoever by differences in electric field – only charged particles are affected by electric fields. The largest peak, at 579 ns, is closest to the predicted value for 1+ charged recoils, but there could also be higher-charged recoils as well. These would arrive sooner than the 1+ recoils, since a higher charge would cause them to fly faster across the positive potential field. A smaller peak can be seen before the 579 ns peak in the positive potential runs, such as Run17.

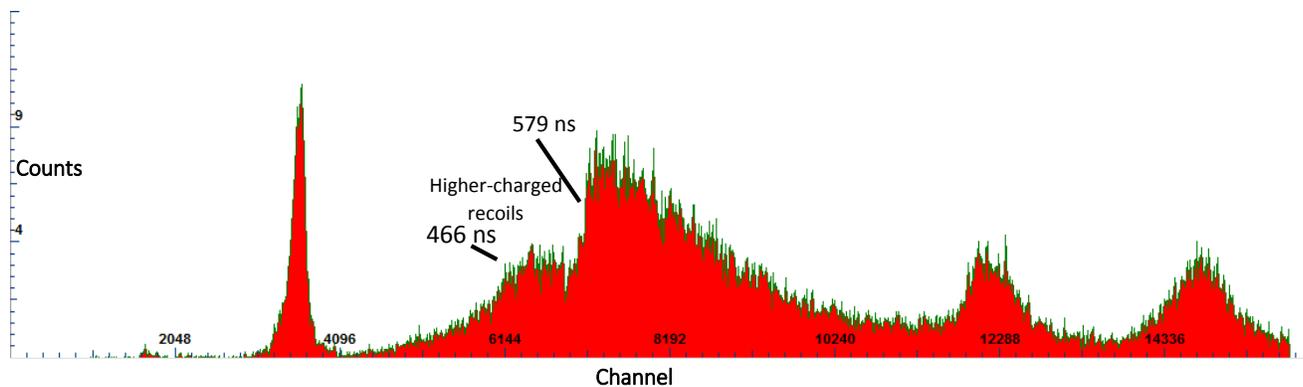


Figure 12: Run17, +4 kV on source, 0 V on mesh grid

The faster end of the peak seen here corresponds with charge states as high as 12+. Highly charged states are possible because of the relatively high energies from nuclear decay.

There are other interesting features of the spectra, most notably in the positive electric field runs. Referring back to Run09 (Fig. 7), there is a sharp peak centered on 203 ns. The peak could be hydrogen from the electroplate process, which uses an acid. For  $H^+$  with no initial kinetic energy, the TOF for 4 kV on the source would be 177 ns. For  $H_2^+$ , which is also possible, the TOF would be 250 ns. However, the peak lies at 203 ns, which is between these two values. Another possibility is that the  $H_2^+$  ions are ejected with some kinetic energy, perhaps from being kicked out by the thorium recoils or  $\alpha$ -particles. These ions would have to have an energy of 2.1 keV in order to match the TOF of 203 ns, or in the case of non-ionized, electrically neutral  $H_2$ , 6 keV. This peak does not appear in the negative potential hill runs, suggesting that the particles causing the peak must either be positively charged, have an initial kinetic energy of less than 2 keV (in order to make it over the potential hill, the particles would have to have more than 2 keV, since it is a 2 kV hill), or both.

As far as the two later peaks go, these are assumed to be either molecules or higher-Z (higher mass) elements. Assuming they come off the source with no initial kinetic energy, and are positively charged in order to be accelerated by the electric field, the mass range of the

particles causing the peaks can be calculated. The predicted atomic masses of the particles based on the observed TOFs is summarized below, for the first three charge states:

<b>TOFs</b>	Lower boundary	Center	Upper boundary
First peak	1078 ns	1129 ns	1172 ns
Second peak	1347 ns	1402 ns	1455 ns
<b>First peak</b>	Lower boundary	Center	Upper boundary
+1	37 amu	41 amu	44 amu
+2	74 amu	81 amu	88 amu
+3	111 amu	122 amu	132 amu
<b>Second peak</b>	Lower boundary	Center	Upper boundary
+1	58 amu	63 amu	68 amu
+2	115 amu	126 amu	135 amu
+3	174 amu	188 amu	203 amu

Table 2: Predicted atomic masses for later peaks, based on TOF

Another feature prominent in both the positive and the negative spectra is a long drop-off after the main thorium recoil signal. This is taken to be the straggling curve, as was modeled in Figure 6. In the model, re-calculated for 1+ thorium recoils, the straggling curve runs from 571 ns for 84 keV recoils off the surface of the source to 1073 ns for an average energy from the back of the source of 20.9 keV. This matches up very well with the curve shown in the spectra.

#### *b) Limitations*

There were a few limitations and difficulties that could potentially have some impact on the data that were seen. First, there was some difficulty in achieving a good vacuum pressure inside the chamber. The chamber was large, and most of the ports used rubber rather than copper gaskets. Even with a turbo pump, the average pressure that a given run was performed at was  $8 \times 10^{-6}$  Torr. The MCP has a maximum operating pressure of  $1 \times 10^{-5}$  Torr, with less than  $1 \times 10^{-6}$  being ideal, according to the manual. Having lower pressures reduces noise and will prevent the high voltage MCP from discharging. Fortunately, the MCP did not have a massive discharge during the experiment, but not operating at a low enough pressure can damage some of the channels and potentially limit the resolution of the MCP.

Another limitation has to do calculating the TOF from the TAC data. An AmpTek MCA was used to create spectra for the TOF data on the computer, and it came with its own software. To find the TOF of a given feature, one clicks on the visual location of the feature, records the channel number, and then converts the channel number to a TOF by a calibration table that was made. However, there is some variability in locating the exact TOF of a given feature. For instance, the peak for the thorium recoils does not rise up completely vertically – there is a difference in TOF between the bottom and the top of the peak. An example of this is Run13, a negative potential hill run, where if the cursor is placed at where the peak begins to rise, the channel number corresponds with a TOF of 592 ns, while at the crest of the peak, the corresponding TOF is 608 ns, for a difference of 16 ns. However, even the low side is still above the high side of the positive runs, such as Run17, which has a low side of about 571 ns and a high side of about 589 ns. The two do not overlap, and still display a difference in TOF (see Fig. 11). For the purpose of labeling the TOFs on the figures, the middle of the rise of the peaks was chosen.

#### **IV. Conclusion**

This research aimed to show whether  $^{229}\text{Th}$  decays from  $^{233}\text{U}$  in a neutral or positively charged state, and analysis of the data shows that the recoils are in 1+ or higher charge states. This is shown by the differences in times-of-flight between positive and negative electric fields. If the recoils were electrically neutral, then the TOFs would be the same every time, no matter what the electric field configuration was. The positive voltage runs, such as Run17, and the negative potential hill runs, such as Run13, have an easily discernable difference in TOF of the

recoils, of about 28 ns. This is in close agreement with the value that was calculated, about 22 ns, and the TOFs for each configuration are close to their predicted values.

It is important to know the charge states of  $^{229}\text{Th}$  when it alpha decays from  $^{233}\text{U}$  because it will determine how the next step of observing the half-life of the decay of the isomer can be achieved – since the  $^{229}\text{Th}$  recoils are positively charged, then the isomer will decay by bound internal conversion, which should emit light in the optical range. Once the half-life is known, then  $^{229\text{m}}\text{Th}$  can be used to make a nuclear clock, which is much more accurate than the atomic clock, and when the two clocks are compared, scientists will be able to observe whether the fine structure constant  $\alpha$  is changing with time. Beyond constructing a nuclear clock, knowing the half-life of the isomer will allow scientists to bridge the gap between atomic and nuclear physics. This is because atomic and nuclear energies are normally different by several orders of magnitude; but in the case of  $^{229\text{m}}\text{Th}$ , this anomalously low energy will allow scientists to probe atomic-nuclear coupling effects, which are poorly understood. Knowing the charge state of  $^{229}\text{Th}$  after  $\alpha$ -decay from  $^{233}\text{U}$  sets the stage for discovering the half-life of its isomer, which will lead to new insights on atomic-nuclear coupling, determining whether the fine structure constant is indeed constant, and in general improve understanding of fundamental physics.

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