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Author
Yee, Ryan Matthew

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Beta-delayed neutron spectroscopy using trapped radioactive ions

by

Ryan Matthew Yee

A dissertation submitted in partial satisfaction of the requirements for the degree of
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in

Engineering - Nuclear Engineering

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor Eric B. Norman, Chair
Professor Jasmina Vujic
Professor Joseph Cerny
Dr. Nicholas Scielzo

Fall 2013
Beta-delayed neutron spectroscopy using trapped radioactive ions

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Ryan Matthew Yee
Abstract

Beta-delayed neutron spectroscopy using trapped radioactive ions

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Ryan Matthew Yee

Doctor of Philosophy in Engineering - Nuclear Engineering

University of California, Berkeley

Professor Eric B. Norman, Chair

A novel technique for $\beta$-delayed neutron ($\beta n$) spectroscopy has been developed using trapped radioactive ions. The neutron energy spectrum was reconstructed by measuring the time of flight (TOF) of the nuclear recoil following neutron emission, thereby avoiding all the challenges associated with neutron detection such as backgrounds from scattered neutrons and $\gamma$ rays and complicated detector-response functions. A proof-of-principle measurement was conducted on $^{137}\text{I}^+$ by delivering ions from a $^{252}\text{Cf}$ source, confining them in a linear Paul trap surrounded by radiation detectors, and measuring the $\beta n$ energy spectrum and branching ratio by detecting the $\beta^-$ and recoil ions in coincidence. Systematic effects were explored by determining the branching ratio three ways. Improvements to achieve higher detection efficiency, better energy resolution, and a lower neutron energy threshold were implemented by upgrading the detectors and optimizing the trapping apparatus. These improvements were demonstrated in a campaign of measurements including $^{138}\text{I}$, which is a standard $\beta n$ precursor outlined by a recent IAEA report on delayed neutron measurements.
To my wife Sonya,

and my family,

for their love and support.
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Chapter 1

Introduction

The $\beta^-$ decay of neutron-rich nuclei often populates excited states in daughter nuclei, and when these states are above the neutron-binding energy they can de-excite by $\gamma$-ray or neutron emission, with the latter process identified as $\beta$-delayed neutron ($\beta n$) emission. This is illustrated in Fig. 1.1.

The properties of $\beta n$ emission are important to both the pure and applied nuclear physics communities [23]. Neutron-emission branching ratios are needed to determine how short-lived neutron-rich isotopes synthesized in the astrophysical rapid-neutron capture process (r-process) decay back to stability [42, 55, 56, 96, 50, 47]. Both neutron branching ratios and energy spectra are required for nuclear reactor kinetics calculations for reactor safety studies [24, 22, 23, 64], and are especially important given the recent nuclear crisis in Japan. Delayed-neutron measurements aid in the understanding of the nuclear structure of neutron-

![Figure 1.1: Process of $\beta n$ emission.](image-url)
rich nuclei [100, 68, 49, 39] and constrain the decay properties for nuclei for which no data exist [45]. High quality data are also important for the study of neutron-rich isotopes in high energy-density environments needed to understand stellar neutron-capture rates [51, 57] and support the stockpile stewardship mission [88, 89]. The following three sections highlight the importance of $\beta n$ in the aforementioned areas.

### 1.1 The r-process

Almost all the elements in nature were formed by nuclear reactions inside stars. Fusion reactions that power stars are exothermic, and burn hydrogen to form helium. However, the binding energy per nucleon, shown in Fig. 1.2, increases only up to $^{56}\text{Fe}$, and any direct fusion reaction (i.e. $^{56}\text{Fe} + \alpha$) thereafter is endothermic. The Coulomb barrier increases with increasing proton number, also rendering fusion unfavorable. Therefore, almost all elements beyond iron are formed in neutron-capture processes. There are two processes identified as the slow and rapid neutron capture processes. They are distinguished by the time scale it takes to capture a neutron, $\tau_n$, compared to typical beta-decay lifetimes, $\tau_\beta$. In the slow-neutron capture process (s-process), $\tau_n$ is much longer than $\tau_\beta$, and therefore almost all neutron captures are followed by a $\beta^-$ decay. This leads to a path along the nuclear chart that follows the valley of stability very closely, where the $\beta$ decay lifetimes can range from minutes to hours. As radioactive nuclides found close to stability have longer lifetimes and are relatively easy to produce, they have been much easier to study in the laboratory. However, in the case of the r-process, extremely high transient neutron fluxes give rise to very short $\tau_n$, compared to $\tau_\beta$. This produces a nucleosynthesis path that leads to very neutron-rich regions of the nuclear chart, as shown in Fig. 1.3, where the $\beta$ decay lifetimes can be fractions of a second. As the neutron flux subsides, these nuclei $\beta^-$ decay back to stability, sometimes proceeding by $\beta n$ emission, to finally form the isotopic distribution observed today.

Type II supernovae have long been suspected to be the site of the r-process, as these explosive environments may have the required density of free neutrons. However, confirmation of this has been tentative as many of the details such as the explosion mechanism and the role of neutrino interactions in the explosion are still unanswered [21]. It is, however, well accepted that the basic nuclear properties, such as the lifetimes, masses, and $\beta n$ branching ratios of the very neutron-rich matter that participate in the r process need to be determined. Thankfully, increasingly reliable data are becoming available, and the proposed technique in this work aims to facilitate these measurements using a combination of ion trapping techniques and modern radiation detection systems.

The neutron fluxes combined with high temperatures in the r process site lead to the production of high-energy gamma rays which can induce $(\gamma,n)$ reactions. This in turn becomes a competing reaction with neutron capture, as the time scale for these events is again much less than $\tau_\beta$. An equilibrium is therefore established for a given $Z$ between these two reactions, where $n + (Z,A) \leftrightarrow (Z,A + 1) + \gamma$. The maximum abundance along an isotopic chain is then determined by the neutron separation energy, $S_n$, for a given
Figure 1.2: Binding energy curve.

Figure 1.3: Classic picture of the s process (represented by the single line) and r process (represented by the band) paths [80].
temperature and neutron density. However, during an r-process event the temperature and neutron density can change, altering the path for these nuclei. It is therefore important to determine the values of $S_n$, which can vary from $\approx 4$ MeV to zero (the neutron drip line), from mass measurements as in Ref. [98], as it defines the r-process path in the N/Z plane. The heaviest nuclei produced in the r-process are then determined by fission, where the neutron-rich nuclei produced are above the fission barrier, resulting in fission “recycling”. This ultimately contributes to the distribution of lighter nuclei.

Once the intense neutron flux ends, and the so-called “freeze-out” period begins, the newly-created neutron-rich nuclei $\beta^-$ decay back to stability. For nuclei created far from stability, the small neutron separation energies and large Q values often lead to large probabilities of $\beta n$ emission. As $\beta n$ emission occurs during the decay back to stability, it can affect the abundance distribution of stable nuclei. It is instructive to consider Fig. 1.4 where the $\beta^-$ decay of $^{140}$I is considered. Here, $\approx 9.3\%$ of these decays result in the production of $^{139}$Xe instead of $^{140}$Xe. A possible final abundance distribution is shown in Fig. 1.5. The prominent peaks located at N = 50, 82, and 126 correspond to the closed neutron shells, where beta-decay half lives are particularly long (on the order of $\approx 0.3\text{–}0.4$ s). Here, the r-process path follows a series of single neutron capture and $\beta^-$ decays, as $\tau_\beta$ becomes more comparable to $\tau_n$, and moves closer to the valley of stability. Between these abundance peaks, $\tau_\beta$ is typically one to two orders of magnitude shorter.

Both the s process and the r process are believed to contribute roughly equally to the production of elements beyond iron, and some elements are in fact produced by both. Fig. 1.6 shows the relative contributions of the s and r processes, where the s process peaks appear systematically shifted to higher A as compared to the r process peaks. This is due to the s-process following a path much closer to stability, encountering the closed neutron shells at higher values of Z.

The r process follows a path in the N/Z plane that builds up nuclei by successive neutron captures, and moves from one isotopic chain to the next by $\beta^-$ decay. As this process involves the production of nuclei very far from stability, it underscores the need for the measurement of basic nuclear properties which are currently unknown. While $S_n$ determines the r-process path, and $\tau_\beta$ determines the shape of the abundance distribution, it remains important to determine $\beta n$ branching ratios as this can affect the final abundance distribution of the elements we observe today.

### 1.2 Nuclear energy

The control and safe operation of nuclear reactors depend on $\beta n$ emission. Since criticality in a reactor is not achieved solely on prompt neutrons from fission (so called prompt criticality), the inclusion of delayed neutrons emitted by the fission fragments allows a power reactor to sustain the multiplicative process under delayed criticality. It is instructive to consider a simple model of the time-behavior of a reactor, where the power of the reactor $P$ at some time $t$ is related to the initial power at $t = 0$ by the relation $P = P_0 exp(\frac{t}{\tau})$. The
Figure 1.4: Illustration of the possible “skewing” of the abundance distribution from the $\beta n$ decay of $^{140}$I, as a portion of the A=140 mass chain shifts over to the A=139 mass chain during the decay back to stability.

Figure 1.5: r-process abundances [4].
Figure 1.6: s and r-process abundances [84].

reactor period, $\tau$, is typically defined as $\tau = \frac{l}{\rho}$, where $l$ is the neutron lifetime, and $\rho$ is the reactivity. If we assume a thermal reactor, where $l$ is typically $10^{-4}$ s and $\rho$ reflects a 0.1% change in reactivity, the reactor period is $\approx 0.1$ s. Therefore, in a $t = 1$ s reactor transient, the power will increase by a factor of $\approx 20,000$, which is far too difficult to control by any mechanical apparatus. However, if delayed neutrons are accounted for, the neutron lifetime will increase in accordance with the lifetimes of the fission fragment precursors. This will increase the reactor period, tempering the effect of small changes in reactivity which can then be controlled by mechanical means such as neutron absorbing control rods.

Nuclear reactor analysis requires a wealth of data, including those on delayed neutrons, to evaluate their performance and safety. These data can be obtained experimentally or estimated theoretically. The body of nuclear data is then evaluated and included into nuclear data files such as the Evaluated Nuclear Data File (ENDF), the Japanese Nuclear Data Committee Nuclear Data Library of Fission Products (JNDC), etc. These data files can include the identification of delayed neutron precursors, their absolute branching ratios, the total delayed neutron yields, the delayed neutron energy spectra, the precursor half-lives, and the average delayed neutron energies. Most important to reactor analysis and safety calculations are the absolute yield ($\bar{\nu}_d$) of delayed neutrons following fission, their energy distribution ($\chi_d$), and their division into groups based on their decay constants ($\lambda_d$). In practice, however, it is the total delayed neutron fraction, $\beta$, that is of greatest concern, where $\beta = \frac{\bar{\nu}_d}{\bar{\nu}_p}$. It is defined as the ratio of the absolute delayed neutron yield to the average number of neutrons emitted per fission, $\bar{\nu}_p$. This parameter must then be weighted by a term, $\bar{\gamma}$, described as the neutron effectiveness factor, which takes into account the effectiveness of this neutron to cause a subsequent fission. $\beta_{eff}$, defined as $\beta_{eff} = \bar{\gamma}\beta$, is then the final term used in calculations. The calculation of this quantity depends on accurate knowledge of the
energy spectra of the delayed neutrons following fission, the neutron population, as well as
the composition, and design of the reactor. The target uncertainty in $\beta_{\text{eff}}$ is on the order
of $\approx 3\%$. The uncertainty in $\bar{\gamma}$ is dominated by incomplete knowledge of the delayed-neutron
spectra, while uncertainties in the absolute yields required at the $\approx 1.5\%$ level are lacking
for the evaluated data.

The absolute delayed-neutron yield, $\bar{\nu}_d$, is the product of the of fission product yield and
the absolute delayed-neutron branching ratio, $P_n$. As such, $\bar{\nu}_d$ can be calculated precursor-
by-precursor; however, many of the existing data have been determined from measurements
on critical assemblies and integral measurements, where empirical relations have been derived
depending on the composite mass and charge of the fissioning material. One such relation
is $\bar{\nu}_d = \exp(16.698 - 1.144Z_c + 0.377A_c)$, where $Z_c$ and $A_c$ are the effective atomic number
and mass, respectively, of the fissioning material, and $\bar{\nu}_d$ is the delayed-neutron yield per
one hundred fissions. As isotopically pure samples have been difficult to obtain, many
empirical relations such as these have been determined. This will be addressed with upcoming
fission fragment beams, where isotopically pure samples can be provided for detailed $\beta$-decay
spectroscopy studies.

The poorest known parameter is the energy spectrum of delayed neutrons for each delayed
group, $\chi_d$, leading to unwanted uncertainties in the neutron effectiveness factor. This has
largely been due to the complexity of the measurements, arising from the short lifetimes of
the precursors and the techniques used to achieve isotopic purity. One area of particular
concern has been the lack of adequate delayed neutron energy data for fast reactors. In a
$^{235}\text{U}$ thermal reactor, the $\beta$ is $\approx 0.0065$. However, a fast breeder reactor may contain large
quantities of $^{238}\text{U}$ and $^{232}\text{Th}$. These isotopes have a $\beta$ of 0.0148 and 0.0203, respectively, and
require more energetic neutrons to induce fission. Therefore, depending on the energies with
which the delayed neutrons are born, they can induce additional fissions in those isotopes,
modifying the dynamic behavior of the reactor. Delayed neutrons may also be more effective
in inducing fission in these fast reactor systems than prompt neutrons ($\approx 2$ MeV in energy)
as they are born with lower average energies ($\approx 100$–$1000$ keV), and therefore suffer smaller
leakage. In contrast to thermal reactors, neutron interactions occur in the keV range in a
fast reactor, which is the typical energy scale for delayed neutron emission, highlighting the
importance of determining their energy spectra.

1.3 Nuclear structure

The investigation of beta-decay properties far from the valley of stability provides valuable
information on the nuclear structure of neutron-rich nuclei. As the available beta-decay
energy is large in these nuclei, a very large number of excited states can be populated. It
is therefore practical to deal with these decays using a statistical model, and to discuss the
rates of these decays in terms of a $\beta$ strength function, $S_\beta$, to obtain an average over a
number of highly excited states rather than treating individual transitions. This can be
expressed as $S_\beta(E) = \frac{b(E)}{f(Q_{\beta-E, Z})t_{1/2}}$, where $b(E)$ is the relative beta intensity per unit energy,
Figure 1.7: The $\beta^-$ decay intensity, $I_\beta$, is determined by the phase space and Coulomb corrections (which are both folded into the statistical rate function $f$), and the beta strength, $S_\beta$. $S_\beta$ is strongly influenced by giant resonances outside the $Q_\beta$ window: the isobaric analogue state and the Gamow-Teller giant resonance. The large beta intensity that goes to the ground state of the daughter nucleus is dominated by the $\approx (Q_\beta - E_{ex})^5$ behavior of the phase space.

$f(Q_\beta - E, Z)$ is the statistical rate function, and $t_{1/2}$ is the half life. To determine the beta strength function, the beta branch intensities, $I_\beta$, to the highly excited states in the daughter nucleus must be obtained, where the phase space for the $\beta$ decay to a particular excitation energy $E_{ex}$ in the daughter nucleus goes as $\approx (Q_\beta - E_{ex})^5$. This leads to the population of many low-lying structures with large $I_\beta$. A cartoon of these features is shown in Fig. 1.7.

The beta-strength function is an important parameter in predicting $\beta$-decay $t_{1/2}$ and $P_n$ values by obtaining the beta feeding to the excited states in the daughter nucleus over the entire $Q_\beta$, and it is directly related to the reduced beta-transition probability. Early theories, such as the “gross theory of beta decay” by Takahashi [93] suggested that $S_\beta$ varied smoothly with $Z$ and $N$, independent of any nuclear structure. This was further supported by an experiment using low-resolution total absorption spectroscopy [40], leading to an extrapolation of beta-decay properties (i.e. half lives) to more neutron-rich nuclei. In this technique, the output signal of the spectrometer was proportional to the full energy of the gamma cascade depopulating the level fed by $\beta^-$ decay, yielding the beta intensity [25, 44, 3]. These measurements were however unreliable as it was acknowledged that the unfolding
process was difficult [25, 44], and corrections needed to be applied to the energy region above the neutron binding energy to account for neutron emission. Furthermore, the poor energy resolution of this technique only allowed the gross properties of $S_\beta$ to be studied.

More recent experiments have found nuclear structure considerations to be important, refuting trends of an approximately constant $S_\beta$ with Z and N. High-resolution beta-decay experiments as described in Ref. [52, 62] have revealed more structure within the $Q_\beta$ window. In this technique, a combination of gamma-ray and neutron spectroscopy was used to reveal structure above and below the neutron binding energy, and a detailed study of the A= 85 to 97 region was carried out in Refs. [53, 49]. These findings have generally motivated further experiments into the unexplored regions of the nuclear chart. Therefore, the measurement of neutrons in coincidence with gamma rays, is essential to determining the decay of excited levels in the daughter nucleus following $\beta$ decay.

Chapter 2 will discuss some of the existing techniques used for $\beta n$ spectroscopy, and the potential advantages of the recoil-ion technique described in this thesis. Chapter 3 summarizes a proof-of-principle measurement using the recoil-ion technique for $^{137}$I, a well-studied $\beta n$ precursor. Chapter 4 describes an improved experimental apparatus and measurement campaign, using a modified ion trap geometry and improved radiation detectors. Finally, a conclusion which provides an outlook for this technique at future radioactive beam facilities for the study of exotic neutron-rich nuclei.
Chapter 2

Existing detection techniques

There are numerous reasons why direct neutron detection can be challenging. One problem which is prevalent among all existing techniques is the ability to measure low-energy neutrons ($\lesssim 100$ keV) reliably. For example, neutron down scattering in the vicinity of the measurement apparatus can contaminate the low-energy part of the neutron spectrum, resulting in substantial corrections that need to be made. Measuring neutron energy spectra is also not straightforward, as the measured spectrum must be corrected for a complicated detector response. The detector response is often energy dependent, as the efficiency of the detector will vary as a function of neutron energy (as is the case in $^3$He spectrometry where the $^3$He($n,p)^3$H cross section varies as $1/v$). Intrinsic detection efficiencies can also be very low, on the order of $\approx 10^{-5}$, and must be compensated for by detector solid angle. In some techniques such as using plastic scintillators for time-of-flight (TOF) measurements, these potential increases in detector efficiency come at the cost of energy resolution, forcing unwanted experimental compromises. The aforementioned challenges in fast neutron measurements certainly motivate the development of new techniques to improve the state-of-the-art.

2.1 $^3$He ionization chambers

$^3$He ionization chambers are commonly used in neutron detection, and are therefore widely used in $\beta n$ spectroscopy. As neutral particles are difficult to detect, these detectors rely on the $^3He(n,p)^3H$ reaction (Q-value = 0.764 MeV) whereby the light ion reaction products are easily stopped in the detector volume, and an electrical signal can be recorded following ionization of the gas by the charged particles. The cross section for this reaction is 5330 b for thermal neutrons and follows a $1/v$ dependence for higher energy neutrons - it is therefore very dependent on the incident neutron energy. Two configurations are typical for $\beta n$ spectroscopy, where either $P_n$ or $E_n$ can be measured, but not both. In a typical setup to measure $P_n$, the $^3He$ detectors are surrounded by low-Z moderating material such as polyethylene to thermalize the incident neutrons, producing a "count" for every event. This
Figure 2.1: Differential energy spectrum for a $^3$He detector. A full-energy peak appears at a value corresponding to the energy of the incident neutron, $E_n$, plus the Q-value of the (n, p) reaction.

setup however does not allow a measurement of the energy of the incident neutrons, as this information is lost in the moderation process. Alternatively, the detectors can be surrounded by cadmium and/or boron to minimize the detection of thermal neutrons, preserving the energy information of the incoming “fast” neutrons to measure their energy. In this scenario, in addition to the reaction Q value, the neutron transfers it’s kinetic energy to the product proton and triton, which then deposit energy in the detector. A typical differential pulse height distribution resulting from these measurements is shown in Fig. 2.1. The first feature is the full-energy peak located at $E_n + Q$, where $E_n$ is the incident neutron energy and $Q$ is the Q-value of the ($n$, p) reaction. At lower energies, one sees a recoil continuum resulting from neutrons that scatter elastically from $^3$He, where the maximum is located at $\approx 0.75E_n$. The prominent feature located at the Q-value for the $^3He(n, p)^3H$ reaction arises from the detection of epithermal neutrons.

Physically, these detectors typically consist of long cylindrical aluminum tubes containing $^3$He gas, a heavy noble gas to help stop the reaction products (to avoid depositing energy into the wall of the detector), and a charge collection circuit. Intrinsic detection efficiencies for a $\approx 2$-3 cm diameter tube are on the order of 77% for thermal neutrons, decreasing to 0.002% for MeV neutrons.
2.2 Proton recoil

Proton recoil in methane gas-filled detectors has been widely used in $\beta n$ spectroscopy to obtain neutron energy spectra. Neutrons are indirectly detected by means of elastic scattering on the hydrogenous gas target. As the Q-value of elastic scattering is zero, the sum of the kinetic energies of the recoil proton and the scattered neutron is equal to that of the incident neutron. For a single scattering event in hydrogen, the fraction of the incident neutron energy transferred to the recoil proton can range anywhere from zero to the full neutron energy. As the range of the recoil protons is generally much smaller than the detector volume, their full energy is deposited yielding a pulse height distribution that is approximately rectangular, as shown in Fig. 2.2, with an endpoint corresponding to the energy of the incident neutron. A detector response with multiple rectangular pulse height distributions (representative of the different neutron energies) can be differentiated to obtain a neutron energy spectrum. The signal generated by the detector is proportional to the energy of the recoil proton, where electrons produced by ionizing the gas are typically collected on a central anode wire [14, 13].

2.3 Neutron time-of-flight measurements

Plastic and liquid scintillators have been used to determine $\beta n$ energy spectra by measuring the time of flight of the neutron. The neutron energy, $E_n$, is determined by $E_n = \frac{1}{2}m_n \frac{L^2}{\tau}$,
where $m_n$ is the mass of the neutron, $L$ is the distance travelled by the neutron, and $t$ is the time of flight. The measurement consists of a $\beta$-neutron TOF coincidence, where the beta is used as the start and the neutron as the stop. The beta particle is detected in an implantation detector (silicon or plastic scintillator) within the ion beam pipe, which is surrounded by a large array of plastic or liquid scintillator detectors to detect the neutrons. The uncertainty in the energy resolution that can be achieved in the measurement is limited by the uncertainties in the distance and time, given by $\frac{\Delta E}{E} = 2 \sqrt{\left(\frac{\Delta L}{L}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}$. Typical TOF distances are on the order of 100 cm, and timing resolutions $< 1$ ns can be achieved. Energy resolutions on the order of 10% full width at half maximum (FWHM) are common. The major disadvantage of organic scintillators is their sensitivity to gamma rays. This can pose a challenge in attributing an event in the detector to either a neutron or a gamma ray, as their detection probabilities are comparable, resulting in pulse height spectra that are similar and overlapping. However, most gamma-ray and neutron events can be separated by the TOF, as $\beta$-$\gamma$ coincidences will appear at much shorter TOFs than $\beta$-neutron coincidences. The remaining events can be sorted by using techniques such as pulse-shape discrimination [46], relying on the differences in the time distribution of scintillator light decay from recoil protons or electrons. With the appropriate electronics and analysis, the decay light can be decomposed into a fast and slow component, with the former decaying on the order of a few nanoseconds, and the latter in several hundred nanoseconds. Recoil protons, which have a higher specific ionization than gamma rays, will produce more delayed fluorescence, yielding a larger fraction of light observed in the slow component. Based on this physics, pulse-shape discrimination can be used as a technique to reject gamma ray events in neutron spectroscopy. Liquid scintillators have been the popular choice for neutron TOF spectroscopy as pulse-shape discrimination is easily implemented in these detectors, but recent developments in plastic scintillators have demonstrated promise to take advantage of such techniques [103].

### 2.4 Recoil-ion technique

Ion traps have revolutionized mass spectrometry, and have the potential to do so for $\beta n$ spectroscopy as well. These devices can confine cooled radioactive ions to a $\approx 1 \text{ mm}^3$ volume in vacuum, where they can be allowed to decay nearly at rest. The emitted radiation emerges from the trap with negligible scattering, and therefore the nuclear recoil can be studied. Recent measurements using atom traps [97, 35, 78] and ion traps [28] have inferred the neutrino momentum from $\beta$–recoil ion coincidence measurements to determine the $\beta$–$\nu$ correlation coefficient with a precision of $\approx 1\%$. A similar approach can be applied to
perform $\beta n$ spectroscopy from the $\beta$–recoil ion coincidence TOF. Here, neutron emission leads to high energy recoils having short TOFs, with the lower-energy recoil imparted by the leptons being a small perturbation to the measurement.

The recoil-ion technique offers several promising advantages over conventional neutron-detection techniques. It yields TOF spectra with a near Gaussian response, avoiding the complicated spectral unfolding techniques typically required to extract the neutron energy spectrum from a complicated detector response [30, 14]. Neutron energy resolutions approaching 3% FWHM are achievable using position-sensitive ion detectors and electronics with $\approx 1$ ns timing resolution. Total intrinsic detection efficiencies of $\gtrsim 60\%$ are achievable as this value approaches the open-area ratio of the microchannel plate ion detectors. Backgrounds from scattered neutrons and $\gamma$ rays, a challenge in traditional neutron detection, are avoided entirely because they have significantly shorter TOFs than the nuclear recoils. Low neutron detection thresholds of $\approx 25$-50 keV are achievable, ultimately limited at an energy where the neutron and lepton recoils produced are comparable. To verify the control of systematic effects, $P_n$ can be obtained by comparing the higher-energy recoil ions characteristic of neutron emission to: (1) the lower-energy recoil ions following $\beta$ decay, (2) $\beta$-delayed $\gamma$ rays emitted by the isotope being studied, and (3) $\beta$ singles.
Chapter 3

Proof-of-principle measurement

A proof-of-principle experiment using the recoil-ion technique was conducted at the Argonne Tandem Linac Accelerator System (ATLAS) facility located at Argonne National Laboratory (ANL) by studying a standard, well-known $\beta n$ precursor, $^{137}$I ($t_{1/2} = 24.5 \pm 0.2$ s [17], $Q_\beta = 6027 \pm 8$ keV [8], $P_n = 7.33 \pm 0.38\%$), where $P_n$ is a weighted average of four independent measurements evaluated in a recent review [43]. A partial decay scheme is shown in Fig. 3.1, with the nuclear data relevant to this experiment annotated. It was chosen for the demonstration measurement as the expected intensity of the ion beam would provide sufficient statistics to utilize an existing but unoptimized ion trap and radiation detector system. The previous four independent measurements of the neutron energy spectrum using two different techniques also provide an opportunity to verify the consistency of the results.

3.1 Ion beam preparation

Fission fragments from an approximately 1-mCi $^{252}$Cf spontaneous fission source were thermalized in a large-volume gas catcher [74], extracted, then bunched and further cooled using a radio-frequency (RF) quadrupole ion guide [73]. Singly-charged ions with a mass of 137 atomic mass units were selected using a timed deflection pulse and a He buffer gas-filled Penning trap [72] and delivered to the Beta-decay Paul Trap (BPT), where recoil-ion decay spectroscopy was performed.

3.2 The Beta-decay Paul Trap

The BPT, shown in Fig. 3.2 and Fig. 3.3, is an open-geometry linear radiofrequency-quadrupole ion trap which was built to perform precision $\beta$-decay studies. Four sets of 1.6-mm thick, stainless-steel electrodes plates are segmented into three pieces so that a DC potential well can be applied to confine the ions axially. Ions are confined radially by applying time-varying voltages of the form $V \cos(\frac{2\pi t}{f})$ to one pair of oppositely placed plates and $-V \cos(\frac{2\pi t}{f})$ to the other pair. The edge of these electrodes were located at a distance of $R_0 =$
17 mm from the trap center. Near the trap center, the electric field is well approximated by a quadrupole field equivalent to that produced by hyperbolic surfaces characterized by a radius of about $R_{\text{eff}} = 1.3R_0$. The RF is applied to the plates using a tuned circuit that consists of three center-tapped pancake wound coils, which are flux coupled with a common primary turn and three parallel variable capacitors to adjust the RF voltage between the three trap segments. The stability condition of the trap is given by $q_{\text{stability}} = \frac{qV}{\pi^2 m r_f^2}$, where $q$ is the charge state of the ion, $V$ is the RF amplitude, $m$ is the mass of the ion, $R_{\text{eff}}$ is the effective trap radius, and $f$ is the RF frequency. For an ideal trap, stable solutions exist for values of $q_{\text{stability}}$ between 0 and 0.908. In this experiment, $V = 200$ V and $f = 264$ kHz, where the RF amplitude was measured with a $\approx 10\%$ uncertainty using an oscilloscope via the tuned circuit. The stability condition of the trap was chosen such that $\geq 2+$ ions (and therefore all $\beta$ decay daughters) were not confined. The trap support structure allows the electrodes and detectors to be positioned to a precision of $\pm 0.1$ mm. Its open geometry allows four sets of detectors to be brought near the trapped ion cloud for the efficient detection of decay radiation. Further details on the trap can be found in Ref. [79].

3.3 Radiation detectors

The BPT was instrumented with a plastic scintillator $\Delta E$-$E$ telescope, a micro-channel plate (MCP), and two high-purity germanium (HPGe) detectors. The arrangement of the
Figure 3.2: Side view of the Beta-decay Paul Trap.

Figure 3.3: End-on view of the Beta-decay Paul Trap.
detectors relative to the trap electrodes is shown in Fig. 3.4. The $\Delta E$-E and MCP detectors were previously used for studies of the $\beta$-$\nu$ correlation [78], and characterized in detail in Ref. [77]. Recoil ions were detected by a metal-anode chevron MCP detector at $-2.65$ kV with an active diameter of $44.08 \pm 0.30$ mm, located at a distance of $61.3 \pm 0.2$ mm from the trap center. This detector was located $2.8 \pm 0.2$ mm behind a grounded $85\%$ transmission grid within a grounded aluminum shield with a window with a $95\%$ transmission grid. The average path length travelled by unperturbed recoil ions was $63.0 \pm 0.2$ mm. In this geometry, the $\beta n$ recoils have TOFs $> 440$ ns from the trap center to the MCP detector. The $\beta$-detector telescope, shown in Fig. 3.5, consisted of a 1-mm thick $\Delta E$ detector with a radius of $17.72 \pm 0.06$ mm optically isolated from a 15-mm thick $E$ detector with a radius of 22.5 mm and subtended $\approx 3\%$ of $4\pi$. The $\Delta E$ detector, which only has a small ($\approx 1\%$) detection efficiency for $\gamma$ rays and neutrons, was used to identify $\beta$ particles in coincidence with recoil ions. The telescope was separated from the vacuum by a 0.127-mm thick beryllium window providing a 150-keV threshold for $\beta$ detection. Re-entrant ports with 1.6-mm thick stainless steel windows allowed single-crystal 80% and 140% relative-efficiency HPGe detectors to be brought within 10 cm of the trapped ion cloud. The $\gamma$-ray energy and efficiency calibrations were determined using $^{137}$Cs, $^{60}$Co, and $^{152}$Eu sealed sources placed at the center of the trap. The total detection efficiency for $\beta$-recoil ion coincidences was $\approx 0.05\%$, given by the product of the $\beta$ detector solid angle ($\approx 3\%$), its efficiency for detecting electrons ($\approx 100\%$), the MCP solid angle ($\approx 3\%$), and its efficiency for detecting recoil ions ($\approx 60\%$).
Figure 3.5: ΔE-E detector from proof-of-principle measurement [76].
3.4 Trap cycle

A schematic of the trap cycle is shown in Fig. 3.6. Ions were captured in the BPT every 5 s, accumulated for 145 s, and then ejected toward a diagnostic silicon detector that monitored the contents of the trap. The trap was left empty for a period of 5, 25, or 40 s at the end of each cycle to assess backgrounds. Trap contents were also monitored by detecting a single peak from the 1218-keV ($I_\gamma = 12.8 \pm 1.3\%$) and 1220-keV ($I_\gamma = 3.5 \pm 0.4\%$) $\gamma$ rays [29] emitted following $^{137}$I $\beta$ decay and the 455-keV ($I_\gamma = 31 \pm 3\%$) $\gamma$ ray [17] emitted following $^{137}$Xe $\beta$ decay in coincidence with $\beta$ particles. The ratio of $^{137}$I to $^{137}$Xe was consistent with the independent yields (IY) from $^{252}$Cf fission [26]. Although no known $\gamma$ ray from $^{137}$Te $\beta$ decay ($t_{1/2} = 2.49 \pm 0.05$ s, $P_n = 2.99 \pm 0.16\%$ [17]) was observed, an amount consistent with the IY (after correction for decay losses during the ion preparation) was assumed to also be present. The buildup of activity in the trap was consistent with the $^{137}$I $t_{1/2}$, implying a trap storage $t_{1/2}$ of $> 220$ s, and therefore $\gtrsim 93\%$ of the $^{137}$I decays in the trap. A trapping efficiency of $\gtrsim 60\%$ was achieved for ions entering the BPT.

3.5 Electronics

A schematic of the electronics used for the proof-of-principle measurement is shown in Fig. 3.7. The $\Delta$E, E, and MCP signals were split into two separate signals that were amplified then sent to a constant fraction discriminator (CFD), or sent to a shaping amplifier to record the pulse height with an amplitude-to-digital converter (ADC). The $\Delta$E and E detector signals were delayed and sent to the stop of a time-to-amplitude converter (TAC). The MCP signal was sent to the start of the TAC. The $\Delta$E and E detector signals post-CFD were ORed together to start a 22 $\mu$s gate. This signal was also delayed to start the data acquisition trigger at the end of every ADC gate. The logic signals from the $\Delta$E, E, and MCP detectors were sent to two scalers to record singles as a function of the time in the trap cycle. The capture scaler was reset by every capture pulse, while the eject scaler was reset every eject pulse. The resistor network used to bias the MCP is shown in Fig. 3.8, where voltage is applied to the front and back of the chevron stack as the MCP plates are gain matched. The data acquisition system used was the Scarlet system [95] maintained by the ANL Physics...
Figure 3.7: Electronics used for proof-of-principle measurement.

Division.

The TOF calibrations for ∆E-MCP and E-MCP coincidences were obtained by sending a series of fixed delays to the MCP start input of the TAC. The $t = 0$ time was determined by coincident events where the MCP did not receive any additional fixed delay (only the delays in the circuit which are present throughout data collection). The TOF response to coincident events was determined by placing a $^{60}$Co source near the radiation detectors, and fitting these events to a Gaussian.

3.6 Data analysis

Gamma-ray calibration

The HPGe detectors were calibrated using a NIST calibrated mixed γ-ray source and an uncalibrated $^{152}$Eu γ-ray source. A table of the γ-ray lines with their respective calibrated activities from the mixed γ-ray source is shown in Table 3.1, and a table with the lines used from the uncalibrated $^{152}$Eu source with their absolute intensities is shown in Table 3.2. First, the ∆E-E telescope was removed to calibrate both the top and right HPGe detectors. Then, the top HPGe detector was removed to obtain a second calibration on the right HPGe detector. Finally, the top HPGe detector was repositioned and calibrated with the right HPGe detector removed. This procedure allowed multiple calibrations on each detector to
Figure 3.8: MCP voltage biasing network used for proof-of-principle measurement.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>γ-ray energy (keV)</th>
<th>Activity (γ-rays per second)</th>
<th>Uncertainty (95% C.L.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs</td>
<td>661.7</td>
<td>1.665E+03</td>
<td>2.8%</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1173.2</td>
<td>2.095E+03</td>
<td>2.7%</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1332.5</td>
<td>2.099E+03</td>
<td>2.8%</td>
</tr>
</tbody>
</table>

Table 3.1: γ-ray lines used from the NIST calibrated source, with the activities on the date of the detector calibration.

serve as redundancy checks. The radioactive sources were placed within $\approx 0.5$ mm of the trap center using a jig. The efficiencies for the top and right HPGe detectors are shown as a function of γ-ray energy in Figs. 3.9 and 3.10. The fits shown are of the form $A + \frac{B}{E}$, where $E$ is the γ-ray energy.

**Monte Carlo Simulations**

The $\beta^-$ decay of $^{137}$I yields $^{137}$Xe ($t_{1/2} = 229.08 \pm 0.78$ s, $S_n = 4025.56 \pm 0.10$ keV [8]) ions with recoil energies $< 170$ eV unless a neutron is emitted. Emission of a neutron with energy $E_n$ following $\beta$ decay yields $^{136}$Xe ions with recoil energies of $\frac{E_n m_n}{m_{^{136}}}$, where $m_n$ and $m_{^{136}}$ are the masses of the neutron and $^{136}$Xe ion, respectively. As $E_n$ can extend to
<table>
<thead>
<tr>
<th>$\gamma$-ray energy (keV)</th>
<th>Absolute intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>411.1</td>
<td>0.02238 (10)</td>
</tr>
<tr>
<td>444.0</td>
<td>0.03125 (14)</td>
</tr>
<tr>
<td>778.9</td>
<td>0.1297 (6)</td>
</tr>
<tr>
<td>867.4</td>
<td>0.04214 (25)</td>
</tr>
<tr>
<td>964.1</td>
<td>0.1463 (6)</td>
</tr>
<tr>
<td>1085.8</td>
<td>0.1013 (5)</td>
</tr>
<tr>
<td>1112.1</td>
<td>0.1354 (6)</td>
</tr>
<tr>
<td>1408.0</td>
<td>0.2085 (9)</td>
</tr>
</tbody>
</table>

Table 3.2: $\gamma$-ray lines used from $^{152}$Eu source.

Figure 3.9: Top HPGe efficiency calibration.
$1987 \pm 8$ keV [99, 8], $^{136}$Xe ions are expected to have energies up to 14.6 keV. However, the average neutron energy is $530 \pm 50$ keV [17], yielding an average $\beta n$ recoil-ion energy closer to 3.9 keV. Most of the daughter ions emerging from the trap are expected to have charge state $2^+$ as $\beta$-decay studies have shown that $\approx 77\%$ of the daughter ions retain all the orbital electrons and the emission of a neutron is expected to result only in limited additional ionization (see [77] and references therein). Simulations indicate that for recoil ions characteristic of neutron emission, the differences between charge states $2^+$ through $5^+$ can be neglected as they have a $<1\%$ effect on both the fraction of ions that reach the MCP detector and the TOF. Internal conversion could result in ions having higher charge states [85], but no significant conversion has been observed in $^{137}$I $\beta^-$ decay [17].

The analysis of the data required the development of detailed Monte Carlo simulations to understand both the $\beta^-$ and recoil-ion detection efficiencies for $^{137}$I, $^{137}$Xe, and $^{137}$Te decays, and to interpret their time-of-flight data. Although the $^{137}$I $\beta$ decay to the ground state ($45.2 \pm 0.5\%$ of total [17]) and many of the hundreds of transitions to excited states at energies below $S_n$ are likely first forbidden, there is essentially no data to determine potential deviations from allowed spectra and the complicated decay scheme is likely incomplete. For these decays, the lepton momenta were generated from an allowed distribution and $\gamma$-ray cascades were approximated as consisting of 1 or 2 isotropically emitted $\gamma$ rays. For decays to excited states above the neutron-separation energy of $^{137}$Xe, prior experimental results [63] are consistent with calculations based on the gross theory of $\beta$ decay [92] that indicate that for $^{137}$I, $\approx 75$-$80\%$ are expected to be allowed. For these allowed decays, no correlation between the $\beta$ particle and neutron momentum is expected. Any potential
anisotropic neutron emission in the remainder of the decays is anticipated to be an effect smaller than the ∼10% experimental uncertainty in the proof-of-principle measurement.

The $\beta^-$ detection efficiency was studied by propagating $\beta^-$ particles through a detailed model of the trap structure and associated detectors using GEANT4 [2]. Similarly, the recoil ions were studied by propagating them through the electric fields of the trap using SimIon 3D Version 8.0 [59]. A beta decay code, adapted from Ref. [76], was used to generate $\beta^-$ spectra and recoil-ion distributions. The beta energy, $E_\beta$, was uniformly generated over the range $0 \leq E_\beta \leq Q_\beta$ and the unit momentum vectors of the $\beta$ and $\nu$ were uniformly generated over the unit sphere. The neutrino energy was then given by $E_\nu = \frac{Q_\beta - E_\beta - Q_\nu^2 \cos^2 \theta_{\beta\nu}}{1 - E_\beta - \frac{|\vec{p}_\beta| \cos \theta_{\beta\nu}}{M}}$, and the recoil energy, $E_r$, and momentum, $\vec{p}_r$, were calculated from conservation of momentum. Each decay was randomly assigned a location with a 1-mm FWHM Gaussian distribution in x, y, and z directions. The location and size of the $\Delta E$-E detector was specified in the code, with events recorded only if the $\beta$ propagated along its momentum vector hit the detector. For valid events, the recoil ion’s position, energy, velocity vector, charge state, and mass were recorded. For the $\beta^-$, the initial position, energy, and direction were recorded.

The electric field in the trap was calculated using SimIon with a 1-mm grid spacing. Details in the simulation included the trap structure (electrodes, endplates, frame, RF shields, detector mounting plates), the $\beta$ detector housing, and the MCP (including the grid and frame holding the plates). Each of these objects was assigned a potential (for example, the RF shields were grounded). An end-on view of the simulation geometry is shown in Fig. 3.11. In addition to the DC potentials, the main electrodes were assigned an RF potential of a specified frequency and amplitude in the SimIon program file. The program file also included a buffer gas scattering code, using a temperature of 293 K consistent with the experimental conditions. The initial position, energy, velocity vector, charge state, and mass of the recoil ion generated in the Monte Carlo were used as inputs to SimIon, which then calculated the trajectories for the ions, and recorded their energy, position, and time-of-flight when they struck a material.

The fraction of $^{136}$Xe ions that hit the MCP detector is shown in Fig. 3.12 as a function of neutron energy. The simulations are performed for a range of neutron energies between 100 keV and 1300 keV, where the upper limit is defined by the energy window available for $\beta n$ emission, $Q_\beta - S_n$. The different curves are representative of the uncertainties in the experimental conditions of the experiment, and used to assess the uncertainty of the $^{136}$Xe recoil ion detection efficiency. The potential uncertainties in the experimental conditions include: the size of the ion cloud, the RF voltage of the trap, and the presence of the buffer gas. As expected, the $^{136}$Xe ion detection efficiency is insensitive to these uncertainties as the recoil energies are large (∼keV). The recoil ion distributions are all calculated assuming an allowed decay to the neutron emitting state. A weighted average of each of the curves was determined, weighted by the known neutron energy spectrum in Ref. [52]. The average of these values is then taken to be the $^{136}$Xe recoil ion detection efficiency and is determined to be $2.96 \pm 0.04\%$. The uncertainty assessed is dominated by the systematic uncertainties.
Figure 3.11: Proof-of-principle SimIon model. In this cut-away view, only the tips of the hollowed-out electrodes are shown. The cross section shown is at half the length of the apparatus along the beam axis.

Recoil ions following $\beta$ decay typically have $\lesssim 500$ eV of energy and are especially susceptible to perturbations by the RF fields. Unperturbed $^{137}\text{Xe}^{2+}$ ions that would otherwise have a drift time of $\gtrsim 4.2$ $\mu$s can give rise to TOF events as short as $\approx 3.2$ $\mu$s. With the $\beta$-recoil ion detectors at right angles, the recoil-ion detection efficiency is only mildly dependent on the details of the $\beta$-decay kinematics. However, for this experiment, the sensitivity of recoil ions to the RF amplitude, ion cloud size, and details of the $\beta$ decay were investigated and folded into the uncertainty of the recoil-ion detection efficiency.

The fraction of $^{137}\text{Xe}$ recoil ions that hit the MCP detector was determined from SimIon, and the results are shown in Fig. 3.13 as a function of $\beta^-$ endpoint energy. A weighted average for each curve was determined, weighted by the known $^{137}\text{I}$ $\beta$-decay scheme [17], where $\approx 45.2\%$ of the decays are expected to proceed to the ground state. The “nominal” case assumes allowed decays within a 1-mm$^3$ ion cloud, no buffer gas present, and the nominal trap voltage of 400 V$_{pp}$ as measured during the experiment. The sensitivity to the ion charge state, the beta neutrino correlation, and the effect of a forbidden shape factor can be seen in the figure. The effect from the beta-neutrino correlation is expected to be minimized in this experiment due to the 90$^\circ$ geometry between the $\beta$ and recoil-ion detector. The results from the simulation indicate a 10% difference in efficiency between the $a = -1/3$ and $a = +1$ cases.

The modification of the beta spectrum shape factor from allowed to forbidden results in a
approximately 1% effect, and is therefore negligible given the current precision of this experiment. A first forbidden unique decay is assumed, with the shape factor of the form $(E_β^2 - 1) + (Q_β - E_β)^2$ in addition to the allowed statistical factor [101]. An additional uncertainty was tested, where some of the $β$ decay strength was redistributed to higher lying states, accounting for possible inaccuracies in the $β$ decay scheme in the published nuclear data due to the Pandemonium effect [41]. The absolute beta intensities, $I_β$, were redistributed linearly according to the prescription $(1 + (E_β/Q_β)\times 0.1)\times I_β$. The fraction of $^{137}$Xe recoil ions that hit the MCP detector is determined to be 1.39 ± 0.10%. The uncertainty assessed is determined by summing the uncertainties of each of the cases in quadrature, where each of these uncertainties are defined as the difference between the weighted average calculated for that particular case, and the “nominal” case.

The beta detection efficiency associated with $βn$ emission, $ε_{β136}$, is expected to be smaller than that for $β$-delayed $γ$-ray emission, $ε_{β137}$, as the beta energies are restricted to an endpoint above the neutron separation energy. On the other hand, the energy of betas associated with delayed gamma ray emission may span the entire $Q_β$. Therefore, on average, the betas associated with $βn$ emission are less energetic. The relevance of these quantities is deferred to Section 3.6.3, where $P_n$ is calculated. For now, a description of how these quantities are obtained via Monte Carlo methods follows.

The $β^-$ detection efficiencies were determined by propagating the particles through a detailed GEANT4 [2] simulation, and included all the features present in the SimIon model. A representative model in Fig. 3.14 is shown with some features removed for clarity. The $β^-$ spectra generated in the Monte Carlo were used as inputs to GEANT4 using a “particle gun”, specifying the $β^-$ energy, direction, and initial position. The beta detection efficiencies were plotted as a function of neutron energy for the events associated with $βn$ emission, and as a
Figure 3.13: $^{137}$Xe ion detection efficiency determined by SimIon.

<table>
<thead>
<tr>
<th></th>
<th>50 keV</th>
<th>80 keV</th>
<th>90 keV</th>
<th>110 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_{137}$</td>
<td>1.21</td>
<td>1.22</td>
<td>1.23</td>
<td>1.25</td>
</tr>
<tr>
<td>$\varepsilon_{136}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3.3: Ratio of beta detection efficiencies with varying detection threshold.

function of $\beta^-$ endpoint energy for $\beta$-delayed gamma ray events. These results are shown in Fig. 3.15 and Fig. 3.16, where the efficiencies at each energy interval are determined by the fraction of initial $\beta$ particles which deposit energy in the detector, above a given threshold. $\varepsilon_{136}$ was determined by taking a weighted average of each of the points in Fig. 3.15, weighted by the known neutron energy spectrum in Ref. [52]. Similarly, $\varepsilon_{137}$ was determined by taking a weighted average of each of the points in Fig. 3.16, weighted by the known $^{137}$I $\beta$ decay scheme in published nuclear data. The ratio $\frac{\varepsilon_{137}}{\varepsilon_{136}}$, which represents the efficiency of detecting a beta particle associated with gamma ray emission as opposed to neutron emission, was determined to be $1.24 \pm 0.02$. This ratio is expected to be greater than 1 as decays to the excited states that can lead to $\beta n$ emission yield lower energy $\beta$ particles. The uncertainty is based on the reliability of the GEANT4 model, the $\beta$ detection threshold, and the $^{137}$I decay scheme [17]. The ratios determined for the different beta detection thresholds are summarized in Table 3.3.

**Determining $P_n$**

The branching ratio $P_n$ is determined three ways. For decays of trapped $^{137}$I$^+$, $P_n = \frac{N_{136}}{N_{\beta}}$ where $N_{136}$ is the total number of decays resulting in $^{136}$Xe recoil ions, and $N_{\beta}$ is the total
Figure 3.14: Proof-of-principle GEANT4 model. The green cylinder is the $\beta$ detector.

Figure 3.15: Beta detection efficiency for neutron energies from 100 - 1300 keV.
Figure 3.16: Beta detection efficiency for single gamma ray emission spanning the Q-value of $^{137}$I beta decay.

number of $\beta$ decays. $N_{\beta 136}$ is determined from $\frac{n_{\beta 136}}{f\epsilon_{\beta 136}\Omega_{136}\epsilon_{136}}$, where $n_{\beta 136}$ is the number of recoil ions observed in the time window 0.44-1.38 $\mu$s (corresponding to 200-2000 keV neutrons), $\epsilon_{\beta 136}$ is the $\beta$ detection efficiency for these events, $\Omega_{136}$ is the fraction of $^{136}$Xe ions that hit the MCP detector active area, $\epsilon_{136}$ is the $^{136}$Xe intrinsic recoil-ion detection efficiency, and $f = 92.5 \pm 1.5\%$ is the fraction of the $\beta n$ spectrum expected to fall in this energy window based on previous studies of $^{137}$I [81, 52, 63, 38]. The 200-keV neutron detection threshold, limited here by the larger than necessary electric fields for this ion trap, was conservatively selected to ensure that the $\beta n$ spectrum was not contaminated with events from recoil ions from $\beta$ decay to the ground state or $\gamma$ ray emitting states. $N_{\beta}$ was determined three ways by measuring recoil ions ($N^r_{\beta}$), $\beta$-delayed $\gamma$ rays ($N^\gamma_{\beta}$), and $\beta$ singles ($N^\beta_{\beta}$).

$N^r_{\beta}$ is given by $\frac{n_{\beta 137}}{\epsilon_{\beta 137}\Omega_{137}\epsilon_{137}(1-P_n)}$, where $n_{\beta 137}$ is the number of $^{137}$Xe recoil ions observed, $\epsilon_{\beta 137}$ is the $\beta$ detection efficiency for these events, $\Omega_{137}$ is the fraction of $^{137}$Xe ions that hit the MCP detector active area, and $\epsilon_{137}$ is the $^{137}$Xe intrinsic recoil-ion detection efficiency. Corrections were applied for the recoil ions expected from the $\beta$ decay of $^{137}$Xe$^+$ and $^{137}$Te$^+$ ions in the trap, and the expected number of $\beta n$ recoils in this time window.

$N^\gamma_{\beta}$ is determined from $\frac{n_{\beta \gamma}}{\epsilon_{\beta \gamma}I_{\gamma}}$, where $n_{\beta \gamma}$ is the number of $\beta$-$\gamma$ coincidences from $\beta$-delayed $\gamma$ rays at 1218 and 1220-keV from $^{137}$I decay, $\epsilon_{\gamma}$ is the $\gamma$-ray detection efficiency, and $I_{\gamma}$ is the absolute $\gamma$-ray intensity.

$N^\beta_{\beta}$ is given by $\frac{n_{\beta}}{\epsilon_{\beta 137}}$, where $n_{\beta}$ is the number of observed $\beta$s from trapped $^{137}$I, accounting for backgrounds from $^{137}$Te and $^{137}$Xe $\beta$ decay, untrapped $^{137}$I $\beta$ decay, and radiation from the room.
It is obvious now that the ratio of the beta detection efficiencies, \( \frac{\varepsilon_{137}}{\varepsilon_{136}} \), come into play in all three branching ratio calculations. The values of \( \Omega_{137} \) and \( \Omega_{136} \) were determined to be \( 1.39 \pm 0.10\% \) and \( 2.39 \pm 0.04\% \), respectively, from the Monte Carlo simulations using SimIon described in the previous section.

The \( P_n \) values determined from the three approaches all share the measurement of the high-energy recoil ions and are summarized in Table 3.7 and Fig. 3.17. The largest source of uncertainty in Method (1) is from \( \Omega_{137} \), which is sensitive to the details of the \( \beta \) decay and the electric field in the trap. The largest uncertainty in Method (2) is the 10\% uncertainty in the \( \gamma \)-ray intensity [29], while \( P_n \) measured in Method (3) is limited by the \( \approx 7\% \) statistical uncertainty in \( n_{\beta136} \).

**Measured neutron energy spectrum**

The \( \beta n \)-energy spectrum, shown in Figure 3.18, was reconstructed from the velocity of \(^{136}\text{Xe}\) recoil ions using conservation of momentum. As the recoil ions from neutron emission are only minimally perturbed by the electric fields, the velocity can be determined simply...
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_{137}$</td>
<td>1.24</td>
<td>0.02</td>
<td>1.60%</td>
</tr>
<tr>
<td>$\varepsilon_{136}$</td>
<td>1.24</td>
<td>0.02</td>
<td>1.60%</td>
</tr>
<tr>
<td>$n_{\beta 136}$</td>
<td>468</td>
<td>23</td>
<td>4.88%</td>
</tr>
<tr>
<td>$(1 - P_n)$</td>
<td>0.9286</td>
<td>0.0038</td>
<td>0.41%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.925</td>
<td>0.015</td>
<td>1.62%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0239</td>
<td>0.0004</td>
<td>1.47%</td>
</tr>
<tr>
<td>$\Omega_{137}$</td>
<td>0.0139</td>
<td>0.0010</td>
<td>10.58%</td>
</tr>
<tr>
<td>$n_{\beta 137}$</td>
<td>4980</td>
<td>245</td>
<td>4.91%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>6.80</td>
<td>0.88</td>
<td>12.94%</td>
</tr>
</tbody>
</table>

Table 3.6: $^{137}$I $P_n$ from the low-energy recoil ions.

<table>
<thead>
<tr>
<th>Method</th>
<th>$P_n$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) Low-energy recoil ions</td>
<td>$6.80 \pm 0.78 \text{ (sys)} \pm 0.41 \text{ (stat)}$</td>
</tr>
<tr>
<td>(2) $\beta$-delayed $\gamma$ rays</td>
<td>$6.88 \pm 0.79 \text{ (sys)} \pm 0.69 \text{ (stat)}$</td>
</tr>
<tr>
<td>(3) $\beta$ singles</td>
<td>$6.95 \pm 0.40 \text{ (sys)} \pm 0.65 \text{ (stat)}$</td>
</tr>
<tr>
<td>2011 IAEA evaluation [43]</td>
<td>$7.33 \pm 0.38$</td>
</tr>
</tbody>
</table>

Table 3.7: Summary of $^{137}$I $\beta n$ branching ratios.

Figure 3.17: $P_n$ in proof-of-principle compared to the 2011 IAEA weighted average.
Figure 3.18: Comparison of $\beta n$-energy spectrum for $^{137}$I measured here with a known spectrum from Ref. [63] that has been convoluted with the energy resolution currently obtained from the recoil ions (shown by the solid line).

from the average distance to the MCP and the TOF. The broadened TOF response from the recoil imparted by the leptons, and the impact of the RF fields was determined for recoil ions from mono-energetic neutrons from 200 keV to 1500 keV using SimIon. The measured recoil-ion TOF spectrum was corrected for the $\beta$ detection efficiency determined from the GEANT4 simulations, after the flat background from accidentals is subtracted. The $^{137}$Te $\beta n$ energy spectrum is not known, but is expected to contribute only $\approx 3\%$ of the total $\beta n$ counts. In Fig. 3.18, the $\beta n$-energy spectrum of $^{137}$I determined here is in excellent agreement with the results of Refs. [52, 63, 81, 38], if convoluted with the energy resolution ($\approx 10\%$) of this measurement.
Chapter 4

Improved apparatus and measurement campaign

The potential of this technique to revolutionize $\beta n$ spectroscopy was revealed in the proof-of-principle measurement. Three improvements, however, were identified as being crucial to demonstrating the full potential of this technique: decreasing the neutron energy threshold, improving the neutron energy resolution, and demonstrating a precision measurement of $P_n$. As the long-term goal of optimizing this technique involves building a dedicated and optimized ion trap and detector system at the Californium Rare Ion Breeder Upgrade (CARIBU), this chapter focuses on how to incrementally improve upon the three aforementioned issues using the existing ion beam infrastructure and BPT.

The most precise measurement of $P_n$ in the proof-of-principle measurement was determined by comparing the number of delayed-neutron recoil ions to the number of beta singles, limited by the 6% statistical uncertainty in the number of $^{136}$Xe ions observed. This was improved upon by increasing the $\beta$-recoil ion detection efficiency, and collecting additional statistics over a longer period of time. The design of an optimized, more efficient detector array to increase the $\beta$-ion detection efficiency would not only improve the current experiment, but also guide the future implementation at CARIBU.

The neutron energy threshold of $\approx 200$ keV in the proof-of-principle measurement was limited by the ion trap configuration, where RF electric fields of $\approx 400 \text{ V}_{pp}$ were used to confine the ions. The large fields perturbed the energies and trajectories of the $\beta$-decay recoil ions resulting in a smearing of the leading edge of their TOFs. This resulted in some of these ions gaining enough energy from the fields to contaminate the $^{136}$Xe TOF region of interest. In order to ensure that the selected recoil ions were associated with neutron emission, a conservative neutron threshold was chosen. With a smaller perturbation, the threshold could be lowered closer to an energy where the neutron and lepton recoils are comparable. To lower the neutron threshold, the design of the trap electrodes was revised to reduce the amplitude of the RF voltage required to trap the precursor ions and therefore reduce the perturbations to the $\beta$ decay daughter recoil ions.

The neutron energy resolution in the proof-of-principle measurement was limited in part
by the determination of the $^{136}$Xe recoil ion trajectory to the MCP detector. For example, the difference in the path length for an ion that strikes the edge of the detector and the average path length leads to a fractional uncertainty in energy of $\approx 7\%$. This was a value determined from the dimensions characteristic of this experiment, and would be greater for detectors subtending larger solid angles. Several factors can contribute to the uncertainty in the ion path: the decay location within the trapped ion cloud, any perturbation to the ion trajectory, and the final impact position on the MCP surface. Any perturbation to the ion path is expected to be negligible for $\approx$ keV ions. However, recoils associated with the lowest energy neutrons can be perturbed by the RF fields of the trap, and therefore careful consideration in the design and operation of the trap is required. The knowledge of the decay location within the trapped ion cloud becomes less important if the ion cloud to MCP detector distance is large. Assume naively that the ion cloud has a Gaussian distribution with a 1.5 mm FWHM along the direction normal to the MCP surface, and is located at a distance of 60 mm from the MCP. This leads to a 2.5\% uncertainty in distance, and a 5\% uncertainty in energy. Lastly, timing-to-amplitude converters (TACs) were used to measure the TOF in the proof-of-principle experiment, with timing resolutions on the order of $\approx 17$ ns FWHM. This could be improved to $\approx 1$ ns FWHM with the use of time-to-digital electronics. The dominant uncertainty in the neutron energy resolution is the determination of the ion flight path length, which can be better determined with a position-sensitive MCP.

The newly designed detectors were placed around the trap as shown in Fig. 4.1. The position-sensitive MCPs which sit at right angles to one another are backed by two single-crystal 80\% relative-efficiency HPGe detectors. The two $\beta$ detectors are placed in the remaining two detector positions around the trap. The details of these experimental improvements follow.

The approaches discussed above to improve the experiment were implemented in 2012. An improved measurement on the $P_n$ of $^{137}$I was sought, in addition to a measurement of the $^{138}$I $P_n$ ($t_{1/2} = 6.23 \pm 0.03$ s, $Q_{\beta} = 7992 \pm 8$ keV [8]). A partial decay scheme for $^{138}$I is shown in Fig. 4.2. The measurement would resolve a discrepancy identified in a recent IAEA evaluation [43] between two groups of measurements. The $^{138}$I measurements are shown in Fig. 4.3, where it can be seen that more recent measurements of the $^{138}$I $P_n$ disagree with several earlier measurements [6, 5]. The two older measurements, with low $P_n$ values, are excluded from an IAEA weighted average of 5.43 $\pm$ 0.20\%, determined solely from two recent independent measurements in Ref. [34] and [69]. Therefore, this discrepancy in the data motivates an additional independent measurement. The recoil-ion spectroscopy technique is ideal for this, as it is subject to different systematic effects than the other measurements. In addition, the reliability of the measurement can be tested by measuring the $P_n$ multiple ways (as discussed earlier) and also comparing the result to the $^{137}$I result.
Figure 4.1: End-on view of BPT with detectors used in the 2012 measurement campaign.

Figure 4.2: Partial decay scheme for $^{138}$I [8, 69, 17].
4.1 Modified electrodes to BPT

The existing electrodes in the BPT were replaced with new electrodes yielding a smaller $R_0$ of 11 mm. This allowed the RF amplitude to be $\approx 2$ times smaller than the value used in the proof-of-principle, while maintaining the same trapping potential, and reducing the perturbations to the low-energy recoil ions. The new electrodes, shown in Fig. 4.4 fully assembled and attached to the trap structure, were also segmented into two pieces: an electrode which carries the RF and a grounded flat metal support connected to the trap frame. These two segments were bridged together with a ceramic insulator, covered by two 0.001” thick stainless steel covers to prevent the accumulation of charge on its surface. One cover was held at the RF voltage, while the other was grounded. The electrode was wired to carry the RF, while the flat metal support, now decoupled from the front end of the electrode, reduced the RF fields along the ion trajectory. The center electrode was reduced in length to reduce the spatial extent of trapped ions along the beam axis, as $R_0$ was decreased.

4.2 Plastic scintillator telescopes

The $\beta$ detector used for the proof-of-principle experiment was not designed for $\beta n$ measurements, but was optimized for $\beta-\nu$ studies of $^{21}\text{Na}$ in Ref. [76]. Although it is a $\Delta E-E$ plastic scintillator telescope, the E detector is only thick enough to stop $\beta^-$ particles up to $\approx 3$ MeV in energy, and is inadequate to study neutron-rich nuclei which can typically emit $\beta$s with up to 5–10 MeV in energy. Furthermore, the solid angle of the detector is not maximized given the open geometry design of the BPT, limiting the overall efficiency for detecting $\beta$-ion coincidences. Finally, the existing housing of the $\Delta E-E$ telescope, which
prevents contaminating the UHV of the ion trap chamber with detector outgassing, incorporates a 127-µm-thick beryllium window. This resulted in a $\beta$ energy threshold of $\approx 150$ keV, and dramatically decreased the detection efficiency for $\beta^-$s associated with high energy neutron emission as seen in Fig. 3.15. Therefore, these limitations motivated the design of an optimized $\beta$ detector for the $\beta n$ work. Two larger $\Delta E$-E plastic scintillators were designed to improve the detection efficiency of beta particles by maximizing the solid angle subtended by the detectors. Each detector was enclosed in a separate housing in their own vacuum, allowing the use of a thin window to decrease the $\beta^-$ detection threshold. The detector assemblies were designed to be compatible with the existing BPT vacuum chamber. The improvement in $\beta$ detector solid angle was approximately a factor of 3, from $\approx 3\%$ of $4\pi$ in the proof-of-principle to $\approx 10\%$ of $4\pi$ in the improved setup.

The $\Delta E$-E telescope consisted of a 0.039” (1-mm) thick EJ-204 fast plastic scintillator, backed by a 5.5” thick scintillator of the same material to stop the betas. As shown in Fig. 4.5, the $\Delta E$ detector is a circular disk with a 4.2” diameter, surrounded by a light guide around its circumference. The light is piped down two strips into two 1.5” PMTs. The E detector is a cylinder with a 5.25” diameter, and is coupled to a 5” PMT (Electron Tubes Enterprises Ltd. #9390KEB). The $\Delta E$ and E assemblies are fastened to two aluminum struts, which is in turn fastened to the end of a flange with SHV and BNC connectors. The bases of the PMTs were potted specifically to prevent discharges under vacuum. The PMTs were also individually wrapped in Metglas, with several layers of Mu Metal inside the circumference of the re-entrant ports, to shield against any magnetic fields from the nearby isobar separator magnet. As shown in Fig. 4.6, the re-entrant port housing the scintillators can be pumped out via a KF-25 connection. The re-entrant ports are secured to the main
Figure 4.5: Upgraded ΔE-E telescope.

The front of the re-entrant ports accommodate the mounting of a 8 μm thick Kapton window, as shown in Fig. 4.7. A ≈ 1 μm thin layer of aluminum was evaporated on both sides of the Kapton material to ensure that the detector housing was light tight. In this configuration, β−s greater than ≈ 30 keV could penetrate the window. The window was sandwiched between two circular disks with an inner aperture of 3.24” diameter. The disks are mated with screws, while compressing the edges of a tautly held thin foil. The disk which mounts to the front face of the re-entrant port includes a groove for the application of an indium wire seal. In the initial tests, it was found that the main UHV chamber could not reach typical 10^-7 torr base pressures, likely due to small pinholes in the Kapton foil leading to a small leak. To achieve the required 10^-8 base pressures in the main UHV chamber, an additional turbo pump was required to pump on the detector volumes.
Figure 4.6: Cross-sectional view of the $\Delta$E-E telescope. All dimensions are in inches.

Figure 4.7: View inside vacuum chamber with re-entrant ports and thin windows mounted.
4.3 Position-sensitive MCPs

One limitation to the achievable neutron energy resolution in the proof-of-principle measurement was the determination of the recoil-ion trajectory. In order to reduce this uncertainty, a position-sensitive MCP was designed to better determine the recoil-ion trajectory for each β-recoil ion coincidence. The final design, intended to maximize the solid angle coverage in the existing BPT, was a 50x50-mm\(^2\) resistive anode chevron MCP detector fabricated by Quantar Technology [1], shown in Fig. 4.8. The solid angle was increased by a factor of \(\approx 3\), from \(\approx 3\%\) of \(4\pi\) in the proof-of-principle to \(\approx 10\%\) of \(4\pi\) in the current experiment. The claimed spatial resolution achievable with this unit was on the order of 200 \(\mu\)m under normal operating conditions. Several masks were designed to test the spatial resolution of the two detector units constructed. These masks were placed to \(\approx 0.5\)-mm precision above the MCP stack, in the place of a 89% transmission grid. Each detector was illuminated with a 1 \(\mu\)Ci \(^{238}\)Pu α source, taped to a detector support bracket mounted to the trap frame, approximately 12 cm from the detector face. The measured spatial resolution was determined to be \(\approx 500\ \mu\)m, and leads to a \(\approx 1\%\) uncertainty in neutron energy in the worst case scenario where an ion strikes the corner of the MCP detector. The dominant uncertainty arises from the size of the trapped ion cloud at \(\approx 1.5\) mm in diameter, which leads to a \(\approx 3\%\) uncertainty in distance, and \(\approx 6\%\) uncertainty in energy. Therefore, a \(\approx 6-7\%\) FWHM neutron energy resolution was expected in this experiment due to the size of the trapped ion cloud and the position resolution of the MCP detectors.

4.4 Trap Operation

For the trapping of \(^{137}\)I\(^+\) ions, the RF amplitude applied during different measurements was 176 \(V_{pp}\) and 230 \(V_{pp}\), yielding a \(q_{stability}\) of \(\approx 0.43\) and \(\approx 0.57\), respectively. For each of these measurements, ions were captured every 5 s in the trap for a total of 145 s, then ejected, when a background measurement was taken for 55 s. Similarly, for the \(^{138}\)I trapping, the RF amplitude applied during different measurements was 176 \(V_{pp}\) and 182 \(V_{pp}\), yielding a \(q_{stability}\) of \(\approx 0.43\) and \(\approx 0.45\), respectively. Ions were captured every 3 s in the trap for a total of 36 s, then ejected, with a background measurement taken for 18 s. The cycles are shown in Fig. 4.9 and 4.10 for \(^{137}\)I and \(^{138}\)I, respectively.

4.5 Electronics

The significant electronics upgrade to the 2012 measurement was the implementation of time-to-digital converters (TDCs) as opposed to TACs. This allowed time resolutions on the order of \(\approx 1\) ns FWHM to be achieved, thereby improving the neutron energy resolution of the measurement. The overall layout of the electronics for each detector is shown in Fig. 4.11. The TDC used in the measurement was a LeCroy Model 4208, where the logic signals from the \(\Delta E\), \(E\), MCP, and HPGe detectors were ORed together and sent to the TDC.
Figure 4.8: Resistive anode chevron MCP detector with 89% transmission grid. The red boxes highlight the 4 readouts of the resistive anode, and the 4 bias connections.

Capture every 5 sec  Eject for 55 sec

\[\downarrow \downarrow \downarrow \downarrow \cdots \cdots \downarrow\]

Beginning of cycle  Time  End of cycle

Figure 4.9: Schematic of the trap cycle used for the $^{137}$I measurements in 2012.

Capture every 3 sec  Eject for 18 sec

\[\downarrow \downarrow \downarrow \downarrow \cdots \cdots \downarrow\]

Beginning of cycle  Time  End of cycle

Figure 4.10: Schematic of the trap cycle used for the $^{138}$I measurements in 2012.
“Common” input. These logic signals were also sent to individual TDC inputs. The TDC was operated in a common start mode, where input signals preceding the common input were recorded as negative times, and signals arriving after the common input were recorded as positive times. The \( \Delta E \) and \( E \) detector signals were split using Mini-Circuits SCP-2-1+ splitters into two pulse height signals, one of which was recorded using the ADC, and the other used to drive the logic after being sent through a constant fraction discriminator. The signals from the four corners of the MCP resistive anode were amplified using a Quantar pre-amplifier/shaping amplifier, and sent through a RF transformer (Mini-Circuits T4-1+) to eliminate a DC offset prior to being recorded by the ADC. Two scalers were used to record singles for each detector as a function of time in the trap cycle, with one scaler reset by every capture pulse, and the other reset by every eject pulse. The voltage divider used to bias each resistive anode MCP is shown in Fig. 4.12 with the capacitively decoupled timing pickoff.

### 4.6 Data Analysis

The data analysis to determine \( P_n \) and \( E_n \) for \(^{137}\text{I} \) and \(^{138}\text{I} \) built on the tools developed for the proof-of-principle experiment in Chapter 3. The SimIon and GEANT4 Monte Carlo simulations were updated to reflect the changes to the experimental apparatus in the 2012 measurement campaign. The GEANT4 geometry, shown in Fig. 4.13, was updated with
the new ΔE-E detector geometry and new trap electrodes to determine the ratio of beta detection efficiencies that are used in the $P_n$ calculation. Similarly, the SimIon simulation, shown in Fig. 4.14, was updated with the new position-sensitive MCP detectors, and the two-piece electrodes with the rear support grounded. SimIon was used to determine the intrinsic recoil ion detection efficiency of the MCP detector, the fraction of $^{130}$Xe and $^{137}$Xe ions that hit the MCP detector in the $\beta^-$ decay of $^{137}$I and $^{138}$I, and the trap center to MCP distance. The right MCP detector was excluded from the analysis as it was discovered that its grid was not properly grounded during the experiment.

As in the proof-of-principle measurement, the RF phase was recorded for every ΔE trigger providing a means to study the perturbation by time-varying electric fields on the slow recoil ions from $\beta^-$ decay. Fig. 4.15 shows the dependence on the RF phase of the number of slow recoil ions detected in each of the ΔE-MCP combinations. As expected, the number of ions detected in the 180° pairs of detectors is enhanced by the kinematic focusing. It is also clear that in addition to the kinematic focusing, the number of recoil ions detected in the MCP is further enhanced for ions born during a time when the RF is at or near a maximum in amplitude. The slow recoil ion data combined with SimIon simulations was used to determine the trapped ion to MCP distance and the MCP ion detection efficiency in the following sections. The uncertainty in the MCP ion detection efficiency is reduced by taking advantage of the RF phase information, as described previously.
Determining the trap center to MCP distance

The physical measurement of the center of the electrode structure to MCP distance was determined to be 52.5 ± 0.4 mm; however, the center of the electrode structure does not necessarily correspond to the center of the trapped ion cloud, which is the relevant quantity to use in the analysis. The leading edge of the slow recoil ions from $^{137}$I, $^{137}$Te and $^{137}$Xe $\beta^-$ decay in the data can provide a reliable means to determine the trapped ion cloud to MCP distance. The 180$^\circ$ $\beta$-ion detector geometry was chosen for this study as it had the highest recoil-ion statistics. A comparison was performed between the data, shown in Fig. 4.16, and a realistic SIMION simulation. From the known $Q_\beta$ of $^{137}$I, $^{137}$Te, and $^{137}$Xe and the mass of the $^{137}$Xe, $^{137}$I, and $^{137}$Cs recoil ions, the TOF determined in the simulation can be tuned to match the data by varying the trapped ion cloud to MCP distance. $\beta$ decays and corresponding $^{137}$Xe, $^{137}$I, and $^{137}$Cs recoil ions in the 2+, 3+, and 4+ charge states were generated using the previously mentioned $\beta$ decay code. For $\beta$ particles that strike the $\Delta$E detector, the simulation was used to determine which 2+, 3+, and 4+ recoil ions would strike the MCP detectors. The distribution of 2+ (77%), 3+ (18%), and 4+ (5%) ions, shown in Fig. 4.17 and 4.18 for each of the RF voltages, was selected based on the existing experimental Xe $\beta$ decay data [85, 87]. The sum of each of these TOF spectra, shown in Fig. 4.17 and 4.18, were fitted to a Gaussian and compared to the fits to the data in Fig. 4.19 and 4.20. The range of the Gaussian fits was limited to TOFs between 2500–3500 ns, with reduced $\chi^2$ values between 1.1 and 1.6. The half-height of each fit are compared between
Figure 4.14: SimIon model for the 2012 measurement with new trap electrodes and MCPs. This is a cross section of the full 3D simulation.

<table>
<thead>
<tr>
<th>Physical measurement</th>
<th>$230V_{pp}$ data</th>
<th>$176V_{pp}$ data</th>
<th>Weighted average</th>
</tr>
</thead>
<tbody>
<tr>
<td>52.5 ±0.4 mm</td>
<td>52.6 ± 1.1 mm</td>
<td>52.6 ± 1.8 mm</td>
<td>52.6 ± 0.9 mm</td>
</tr>
</tbody>
</table>

Table 4.1: Summary of trap center to MCP distance measurements. The weighted average was determined from the data at $230V_{pp}$ and $176V_{pp}$ and was the quantity used in the remainder of the analysis.

the simulation and data to infer the trapped ion cloud to MCP distance. The results are summarized in Table 4.1, where the average distance of 52.6 ± 0.9 mm was used for the remainder of the analysis.

**Ion detection efficiency**

The position sensitive MCP detectors used in the 2012 measurements have a 65% open area ratio and a 89% transmission grid, and the intrinsic detection efficiency is expected to be approximately the product of these quantities. To measure this efficiency, an analysis was
Figure 4.15: Slow recoil ion data from the $^{137}$I measurements at $230V_{pp}$ sorted on the RF phase.

Figure 4.16: Mass 137 recoil ion data at two different RF voltages. The $176V_{pp}$ data are normalized to the $230V_{pp}$ data by the area under the $^{136}$Xe recoil-ion feature.
Figure 4.17: 2+, 3+, and 4+ $^{137}\text{Xe}$, $^{137}\text{I}$, and $^{137}\text{Cs}$ ions from $\beta^-$ decay propagated through SIMION at $230V_{pp}$ with charge state distributions consistent with the experimental Xe beta decay data [85].

Figure 4.18: 2+, 3+, and 4+ $^{137}\text{Xe}$, $^{137}\text{I}$, and $^{137}\text{Cs}$ ions from $\beta^-$ decay propagated through SIMION at $176V_{pp}$ with charge state distributions consistent with the experimental Xe beta decay data [85].
Figure 4.19: Fitted leading edge of the $^{137}$Xe, $^{137}$Cs, and $^{137}$I ions from $\beta^-$ decay at 230$V_{pp}$.

Figure 4.20: Fitted leading edge of the $^{137}$Xe, $^{137}$Cs, and $^{137}$I ions from $\beta^-$ decay at 176$V_{pp}$.
conducted to determine this value from the data, supported by Monte Carlo simulations. The analysis was carried out for the top MCP detector (as the grid on the right MCP detector was floating for all data sets taken for this thesis) for the $^{137}$I data at 230$V_{pp}$ and 176$V_{pp}$, as well as the $^{138}$I data at 182$V_{pp}$. The ion detection efficiency with the smallest uncertainty determined from these three data sets was then used in the $P_n$ analysis. Unfortunately during the experiment, a turbo pump failed and caused the pressure in the chamber to increase dramatically, damaging the MCP detectors. The top MCP detector survived the incident, and was refurbished by interchanging the two channel plates forming the chevron stack. The ion detection efficiency was determined separately for the top MCP after this event, for the $^{138}$I data taken at 176$V_{pp}$.

The intrinsic ion detection efficiency, $\varepsilon_r$, can be determined from the relation 

$$
\varepsilon_r = \frac{N_{\beta r}}{N_{\beta} \Omega_r (1 - P_n)},
$$

where $N_{\beta r}$ is the number of $^{137/138}$Xe recoil ions, $N_{\beta}$ is the number of beta singles from $^{137/138}$I $\beta^-$ decay, $\Omega_r$ is the fraction of $^{137/138}$Xe ions expected to hit the MCP detector, and $P_n$ is the $\beta n$ branching ratio for $^{137}$I or $^{138}$I taken from Ref. [43]. The uncertainty in $\Omega_r$ includes uncertainties arising from the knowledge of $a_{\beta\nu}$, and the charge state distribution (CSD) of the recoil ions in the $\beta^-$ decay of $^{137}$I and $^{138}$I. The calculation of $\Omega_r$ assumes an allowed decay and $a_{\beta\nu} = -1/3$ in the $\beta$ decay of $^{137}$I and $^{138}$I. However, most of the decays to excited states below $S_{\text{sn}}$ in these isotopes are first forbidden where the beta neutrino correlation is unknown, and $a_{\beta\nu}$ can take on a value between -1 and +1. The value of $a_{\beta\nu}$, however, can be constrained by comparing the number of $^{137/138}$Xe recoil ions in the 90$^\circ$ versus 180$^\circ$ $\beta$-ion detector combinations in the data and the simulation. The $^{137}$I data at 230$V_{pp}$ indicates a ratio of $\approx 6.4$ for the number of slow ions in the 180$^\circ$ versus 90$^\circ$ $\beta$-ion detector combination, while a ratio of $\approx 6.1$ is observed for the data taken at 176$V_{pp}$. The simulation suggests that a value of $\approx 0$ for $a_{\beta\nu}$ for the beta decay of $^{137}$I, $^{137}$Xe, and $^{137}$Te with 2+, 3+, and 4+ charge states for the daughter ions. Furthermore, only the 90$^\circ$ $\beta$-MCP detector combination is considered in the determination of $\varepsilon_r$, as the results from this combination are less sensitive to the knowledge of $a_{\beta\nu}$. The calculation of $\Omega_r$ also assumes a simplified CSD of 2+ (77%), 3+ (18%), and 4+ (5%) ions in the $\beta$ decay of $^{137}$I and $^{138}$I, following a distribution consistent with the experimental data on $^{131}$Xe $\beta$ decay in Ref. [85, 87]. However, the CSD for the decays of interest are unknown. An uncertainty was therefore applied to $\Omega_r$ by assuming a CSD extending to higher charge states of up to 6+ ions using a distribution again consistent with Ref. [85, 87], with 2+ (60%), 3+ (25%), 4+ (10%), 5+ (4%), and 6+ (1%) ions. In addition, higher charge states of up to 15+ are observed in $^{133}$Xe $\beta$ decay due to internal conversion and Auger cascades. Very little internal conversion is expected to occur in the beta decays studied here, so this effect has been neglected. An uncertainty of 3.5% arising from the trap-to-MCP distance was folded into the overall $\Omega_r$ uncertainty. A correction for $^{137}$Cs and $^{137}$I ions from the $\beta^-$ decay of the $^{137}$Xe and $^{137}$Te contaminants was also included in the $\varepsilon_r$ calculation. Approximately 10% of the decays in the trap are attributed to $^{137}$Te and was determined from the beta singles fits. Approximately 16% of the decays in the trap are attributed to $^{137}$Xe, and was determined from $\beta-\gamma$ coincidences. The $^{137}$Cs and $^{137}$I ions were then flown through SimIon to account
for the different fractions of ions expected to hit the MCP detector. Similarly, a correction for $^{138}$Cs and $^{138}$I ions from the $\beta^-$ decay contaminants of the $^{138}$I isobars was determined. The dominant uncertainty in the $\varepsilon_r$ calculation is expected to come from $\Omega_r$, where the determination of this value is dependent on knowledge of the RF amplitude of the trap and details of the $\beta$ decay. SimIon simulations have shown that a 7% uncertainty in RF amplitude in this experiment leads to a 5-10% uncertainty in $\Omega_r$. Measurements of the RF were performed by placing a 10X oscilloscope probe on the electrodes directly, determining the RF voltage and frequency for each trapped species. These measurements were done in series for each of the RF settings, waiting 10 – 15 minutes in between each measurement for the tuned circuit to equilibrate to its new temperature. The 7% uncertainty in the RF measurement was dominated by the uncertainty in the reliability of the probe, which was assessed by using several different 10X probes for the measurements.

Several approaches to reduce the uncertainty in $\Omega_r$ from the RF were explored: (1) sorting the data based on the RF phase, and (2) only using $\beta$-ion coincidences gated on > 3 MeV $\beta^-$s in the 180° back-to-back detector combination. Option (1) seemed viable as during certain phases of the RF, for example near the zero crossing, one would naively expect the ion trajectories to be less sensitive to the RF fields. On the other hand, during the peaks of the RF phase, one would expect the ion trajectories to be very sensitive. Option (2) seemed viable as kinematics dictate that high energy $\beta^-$s tend to focus the $^{137}$Xe recoil ions in the opposite direction. This effect could dominate any perturbation caused by the RF fields, especially in decays where very high energy $\beta^-$s are emitted and the $Q_\beta$ value is large.

A simple approach to sort the data by RF phase was devised where the ion time of birth (also the time of the $\beta$ decay) can be sorted by time regions P1 through P5 as shown in Fig. 4.21, where these regions span an entire RF period. P2 and P4 define times where the $\beta$ decay occurs during a period of maximum RF amplitude. Similarly, P1, P3, and P5 define the regions of time where a $\beta$ decay occurs during a RF minimum. P2+P4 and P1+P3+P5 span equal time periods. Initial studies were performed by generating $^{137}$Xe recoil ions from $^{137}$I $\beta$ decays to excited states spanning the entire $Q_\beta$ window using the $\beta$ decay code, and propagating them through SimIon at the nominally measured RF voltage, and a RF voltage 7% higher which is representative of the uncertainty in the RF measurement during the experiment. $^{137}$Xe ions with 2+ and 3+ charge states were propagated through SimIon at trap voltages of 230 V$_{pp}$ and 176 V$_{pp}$, and a value 7% higher. To determine the effect of the RF sort for each $\beta$-ion detector geometry, several new ratios were introduced into the analysis. $\frac{\Omega_r|_{V_{RF}}}{\Omega_r|_{1.07 V_{RF}}} \times 1.07$ was defined as the ratio of $^{137}$Xe ions detected at the MCP at the expected voltage to those detected at the voltage 7% higher. This ratio is determined for ions which are born during an RF maximum (P2+P4), an RF minimum (P1+P3+P5), and no sort based on the RF phase. Results for the 90° $\beta$-ion detector combination for $^{137}$Xe$^{2+}$ ions propagated through SimIon at trap voltages of 230 V$_{pp}$ and 246 V$_{pp}$ are shown in Table. 4.2, with the last row being a weighed average of $\frac{\Omega_r|_{V_{RF}}}{\Omega_r|_{1.07 V_{RF}}} \times 1.07$ using the known beta intensity. The weighted averages are then summarized for the 180° and 90° $\beta$-ion detector
configurations, in Table 4.3 and Table 4.4, respectively.

The results from the preliminary study show that the sorting of ions born during a period of maximum RF amplitude (P2 and P4) minimizes the dependence of the ion detection efficiency on the RF voltage of the trap. This can be explained intuitively. Most $^{137}$Xe$^{2+}$ recoil ions that strike the MCP detector are expected to have TOFs of $\approx 3-4$ µs, which is approximately one full RF period. For these ions, it will take $\approx 1$ µs, or one quarter of a full RF period, to enter a region in the trap (between the electrodes) where the RF electric field amplitude is the greatest. However, by only selecting ions that are born at or near a period of maximum RF amplitude, these ions are expected to travel through the region of distortion at close to a minimum in RF amplitude, minimizing the perturbation to these ions, and therefore decreasing the dependence of the ion detection efficiency on the RF voltage of the trap. For the $^{137}$Xe$^{2+}$ ions at 230/246 V$_{pp}$ for the 90° β-ion detector combination, the $\frac{\Omega_{r|V_{RF}}}{\Omega_{r|V_{RF}*1.07}}$ value of 1.06 for ions born during a RF maximum suggests that the sort will reduce the impact of the uncertainty of the RF voltage on the ion detection efficiency from a $\approx 9\%$ effect to a $\approx 6\%$ effect.

The next step was to combine the RF sort with the ion position information from the MCP detector to see if the uncertainty in the ion detection efficiency could be further reduced. Table 4.5 shows the potential reduction in uncertainty of the ion detection efficiency with several different constraints on the area of the MCP surface for a 180° β-ion detector geometry. This geometry was chosen as initial comparisons between the 90° and 180° combinations revealed that the final ion position on the MCP detector for the 180° combination had a dependence on the ion time of birth during an RF maximum. The results are shown for $^{137}$Xe$^{2+}$ ions flown through the trap with RF voltages of 230 V$_{pp}$ and 246 V$_{pp}$. The β decays for these ions are generated by populating excited states spanning the entire $Q_\beta$
\[ E_0 \text{ (keV)} \quad \text{Beta intensity (\%) } \quad \frac{\Omega_r |_{V_{RF}^1}}{\Omega_r |_{V_{RF}^1 \times 1.07}} \]

| \( E_0 \text{ (keV)} \) | \( \text{Beta intensity (\%)} \) | \( \frac{\Omega_r |_{V_{RF}^1}}{\Omega_r |_{V_{RF}^1 \times 1.07}} \) |
|-----------------|------------------|------------------|
| 500             | 0.012            | 1.10 ± 0.02      | 1.22 ± 0.04 | 1.03 ± 0.03 |
| 1000            | 0.039            | 1.12 ± 0.02      | 1.25 ± 0.04 | 1.06 ± 0.03 |
| 1500            | 1.328            | 1.13 ± 0.02      | 1.27 ± 0.05 | 1.07 ± 0.03 |
| 2000            | 8.266            | 1.11 ± 0.03      | 1.17 ± 0.05 | 1.08 ± 0.03 |
| 2500            | 2.391            | 1.10 ± 0.03      | 1.17 ± 0.05 | 1.06 ± 0.03 |
| 3000            | 2.178            | 1.08 ± 0.03      | 1.14 ± 0.05 | 1.05 ± 0.04 |
| 3500            | 6.956            | 1.08 ± 0.03      | 1.11 ± 0.05 | 1.06 ± 0.04 |
| 4000            | 13.654           | 1.07 ± 0.03      | 1.12 ± 0.06 | 1.04 ± 0.04 |
| 4500            | 16.260           | 1.07 ± 0.04      | 1.07 ± 0.06 | 1.07 ± 0.05 |
| 5000            | 0.027            | 1.07 ± 0.04      | 1.09 ± 0.06 | 1.05 ± 0.04 |
| 5500            | 0.215            | 1.08 ± 0.03      | 1.14 ± 0.06 | 1.05 ± 0.04 |
| 6026            | 48.673           | 1.10 ± 0.03      | 1.19 ± 0.06 | 1.05 ± 0.04 |

Weighted average 1.09 ± 0.02 1.15 ± 0.03 1.06 ± 0.02

Table 4.2: Results for the 90° \( \beta \)-ion detector combination for \(^{137}\text{Xe}^{2+}\) ions at 230 \( V_{pp} \). \( \frac{\Omega_r |_{V_{RF}^1}}{\Omega_r |_{V_{RF}^1 \times 1.07}} \) is tabulated for the different RF sorting conditions.

| Recoil ion \( ^{137}\text{Xe}^{2+} \) | RF Voltages | \( \frac{\Omega_r |_{V_{RF}^1}}{\Omega_r |_{V_{RF}^1 \times 1.07}} \) |
|---------------------------------|-------------|------------------|
| \( 230/246 \text{ } V_{pp} \)   | 1.11 ± 0.01 | 1.25 ± 0.01      | 1.06 ± 0.01 |
| \( 176/188 \text{ } V_{pp} \)   | 1.06 ± 0.01 | 1.10 ± 0.01      | 1.04 ± 0.01 |

Table 4.3: Results of RF sorting on 180° \( \beta \)-ion detector configuration.

| Recoil ion \( ^{137}\text{Xe}^{2+} \) | RF Voltages | \( \frac{\Omega_r |_{V_{RF}^1}}{\Omega_r |_{V_{RF}^1 \times 1.07}} \) |
|---------------------------------|-------------|------------------|
| \( 230/246 \text{ } V_{pp} \)   | 1.09 ± 0.02 | 1.15 ± 0.03      | 1.06 ± 0.02 |
| \( 176/188 \text{ } V_{pp} \)   | 1.08 ± 0.02 | 1.10 ± 0.02      | 1.05 ± 0.02 |

Table 4.4: Results of RF sorting on 90° \( \beta \)-ion detector configuration.
Table 4.5: Results by gating only on ions born during an RF maximum and final ion position on the MCP face for a 180° β-ion detector geometry at 230/246 V\textsubscript{pp}.

<table>
<thead>
<tr>
<th>Detector pos. X &amp; Y</th>
<th>% of total area</th>
<th>Position sort only</th>
<th>RF phase and position sort</th>
</tr>
</thead>
<tbody>
<tr>
<td>± 15 mm</td>
<td>39</td>
<td>1.09 ± 0.01</td>
<td>1.04 ± 0.01</td>
</tr>
<tr>
<td>± 20 mm</td>
<td>69</td>
<td>1.11 ± 0.01</td>
<td>1.05 ± 0.01</td>
</tr>
<tr>
<td>± 24 mm</td>
<td>100</td>
<td>1.11 ± 0.01</td>
<td>1.06 ± 0.01</td>
</tr>
</tbody>
</table>

Table 4.6: Sensitivity of ion detection efficiency by gating on a high-energy β\textsuperscript{−}.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>(N_\beta)</td>
<td>128614 ± 3858</td>
<td>3%</td>
<td>-</td>
</tr>
<tr>
<td>(\Omega_r)</td>
<td>0.0276 ± 0.0026</td>
<td>-</td>
<td>9.6%</td>
</tr>
<tr>
<td>(1 - P_n)</td>
<td>0.927 ± 0.004</td>
<td>-</td>
<td>0.4%</td>
</tr>
<tr>
<td>(\varepsilon_r)</td>
<td>0.44 ± 0.02 (5.5%)</td>
<td>± 0.04 (9.6%)</td>
<td></td>
</tr>
</tbody>
</table>

Table 4.7: \(\varepsilon_r\) calculation for \(^{137}\)I data at 230V\textsubscript{pp}.

range, and the results weighted appropriately by the known decay branches. A cursory look at the data indicates that the RF sort is the most effective in reducing the uncertainty of the ion detection efficiency. Therefore, the position sort was not pursued any further.

Option (2) was studied by generating \(^{137}\)Xe\textsuperscript{2+} recoil ions corresponding to β\textsuperscript{−} decays above 3, 4, and 5 MeV thresholds. The ions are flown through SimIon at the 176/188 V\textsubscript{pp} and 230/246 V\textsubscript{pp} trapping voltages. The ratio \(\varepsilon_{176/188}\) is defined as the ratio of \(^{137}\)Xe\textsuperscript{2+} ions for the 176 V\textsubscript{pp} versus the 188 V\textsubscript{pp} trapping voltage that make it to the MCP opposite the β detector. Similarly, \(\varepsilon_{230/246}\) is defined as the ratio of \(^{137}\)Xe\textsuperscript{2+} ions for the 230 V\textsubscript{pp} versus the 246 V\textsubscript{pp} trapping voltage that make it to the MCP. Table 4.6 summarizes these results, and demonstrates that even gating on > 5 MeV β\textsuperscript{−}s does not result in any substantial reduction on the uncertainty from the RF compared to > 3 MeV β\textsuperscript{−}s. This cut, however, is not attractive because it will substantially reduce the \(^{137}\)Xe recoil ion statistics as very few β\textsuperscript{−}s have energies high enough to make it past such a high threshold.

A summary of the ion detection efficiencies determined in the 2012 measurement campaign is shown in Fig. 4.22. The ion detection efficiency of 0.44 ± 0.02 (stat) ± 0.04 (sys) determined using the \(^{137}\)I at 230 V\textsubscript{pp} data set was used for the \(P_n\) calculation as it had the
The first three measurements are for the top MCP detector only, as the right MCP grid was floating. The last measurement is from the $^{138}$I data set taken after the MCP plates were refurbished. The result is $\approx 25\%$ lower than the expected value, and will need to be further investigated. The values used for the $\varepsilon_r$ calculation with statistical and systematic uncertainties are summarized in Table 4.7. The uncertainty in $\Omega_r$ of 9.60% folds in the uncertainty from $\sigma_{\beta \nu}$ (2.83%), the SimIon grid units (3.5%), the CSD (5.4%), the RF (5.6%), and the trapped ion to MCP distance (3.5%). The correction due to the $^{137}$Cs and $^{137}$I recoil ions is $1.33 \pm 0.05$ (3.8%). The weighted average of the two measurements was not used to determine $\Omega_r$ as the systematic uncertainty is $\approx 1.3$ times larger in the data set taken with a lower voltage. This was attributed to the sensitivity of $\frac{\delta \Omega_r}{\delta V_{RF}}$ to the chosen cut using the RF phase at a particular RF voltage. Fig. 4.23 illustrates the voltage dependent effectiveness of this cut at reducing the uncertainty in $\Omega_r$, where the uncertainty in RF amplitude is a $\approx 4\%$ effect at 176/188 V$_{pp}$ but a $\approx 6\%$ effect at 230/246 V$_{pp}$. The plot seems to suggest that $\frac{\delta \Omega_r}{\delta V_{RF}}$ for a given RF phase cut will vary, perhaps sinusoidally, with the RF voltage.

**Gamma-ray calibration**

The HPGe detectors were calibrated using a NIST calibrated mixed $\gamma$-ray source and an uncalibrated $^{152}$Eu $\gamma$-ray source as in the proof-of-principle experiment. The details of the
mixed γ-ray source are in Table 3.1 in Chapter 3. Calibrations were obtained by taping the sources to a pair of electrodes facing the HPGe detector. It is estimated that following this procedure, the radioactive sources could be placed within ≈1 mm of the trap center. The efficiencies for the top and right HPGe detectors are shown as a function of γ-ray energy in Figs. 3.9 and 4.24, after correction for summing and source attenuation through the source holder. The fits shown are of the form \( A + B\ln(E) + Cln^2(E) + D\ln^3(E) + E\ln^4(E) \) (where \( E \) is the γ-ray energy), and are adapted from Ref. [37].

\[ ^{137}I \] \( P_n \)

The \( ^{137}I \) \( P_n \) was determined using beta singles and beta-delayed gamma rays as described in the proof-of-principle measurement. \( \frac{x_{^{137}I}}{x_{^{136}I}} \) was determined to be 1.075 ± 0.01 from the updated GEANT4 simulations. \( f \) was determined to be 0.96 ± 0.01, and was higher than the value used from the proof-of-principle as the neutron threshold was reduced from 200 keV to 100 keV. \( \Omega_{^{136}} \) was determined to be 0.0642 ± 0.0019 for the 180° β-ion detector pairs and 0.0622 ± 0.0019 for the 90° detector pairs, from the updated SimIon simulations, reflecting the changes in the trap, as well as the larger MCPs. The intrinsic detection efficiency of 0.44 ± 0.05 for recoil ions was determined from the previous section. Tables 4.8 and 4.9 summarize the quantities used for the beta singles measurements using the 180° and 90° β-ion detector pairs, respectively. These were combined into a single \( P_n \) value of...
Table 4.8: $^{137}$I $P_n$ from $\beta$ singles for 180° $\beta$-ion detector combinations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$I</td>
<td>1.075</td>
<td>0.010</td>
<td>0.93%</td>
</tr>
<tr>
<td>$^{136}$I</td>
<td>619</td>
<td>39</td>
<td>6.27%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.9619</td>
<td>0.0100</td>
<td>1.04%</td>
</tr>
<tr>
<td>$\varepsilon_{136}$</td>
<td>0.44</td>
<td>0.05</td>
<td>11.09%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0642</td>
<td>0.0019</td>
<td>2.91%</td>
</tr>
<tr>
<td>$n_\beta$</td>
<td>413345</td>
<td>9027</td>
<td>3.00%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>5.88</td>
<td>0.79</td>
<td>13.48%</td>
</tr>
</tbody>
</table>

Fig. 4.24: Right HPGe efficiency calibration.

5.86 ± 0.68 (sys) ± 0.28 (stat)% using beta singles. Similarly, Tables 4.10 and 4.11 summarize the quantities used for the beta-delayed gamma measurements using the 180° and 90° $\beta$-ion detector pairs, respectively, yielding a combined $P_n$ value of 6.18 ± 0.97 (sys) ± 0.57 (stat)%. Fig. 4.25 summarizes the results using the BPT with the IAEA weighted average [43].

$^{138}$I $P_n$

The $^{138}$I $P_n$ was determined using beta singles and beta-delayed gamma rays as in the $^{137}$I $P_n$ measurement. The notation previously used for the $^{137}$I $P_n$ calculations in chapter 3 are adopted for $^{138}$I. $\varepsilon_{138}$ was determined to be 1.069 ± 0.02 from GEANT4. $f$ was determined
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}\beta$</td>
<td>1.075</td>
<td>0.010</td>
<td>0.93%</td>
</tr>
<tr>
<td>$^{\beta}_{136}$</td>
<td>583</td>
<td>34</td>
<td>5.87%</td>
</tr>
<tr>
<td>$n_{\beta_{136}}$</td>
<td>0.9619</td>
<td>0.0100</td>
<td>1.04%</td>
</tr>
<tr>
<td>$\varepsilon_{136}$</td>
<td>0.44</td>
<td>0.05</td>
<td>11.09%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0622</td>
<td>0.0019</td>
<td>3.00%</td>
</tr>
<tr>
<td>$n_{\beta}$</td>
<td>404752</td>
<td>8861</td>
<td>3.00%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>5.84</td>
<td>0.78</td>
<td>13.32%</td>
</tr>
</tbody>
</table>

Table 4.9: $^{137}I P_n$ from $\beta$ singles for 90° $\beta$-ion detector combinations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$I_\gamma$</td>
<td>0.163</td>
<td>0.017</td>
<td>10.43%</td>
</tr>
<tr>
<td>$\varepsilon_\gamma$</td>
<td>0.00540</td>
<td>0.00008</td>
<td>1.56%</td>
</tr>
<tr>
<td>$I_{137}$</td>
<td>1.075</td>
<td>0.010</td>
<td>0.93%</td>
</tr>
<tr>
<td>$n_{\beta136}$</td>
<td>619</td>
<td>39</td>
<td>6.27%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.9619</td>
<td>0.0100</td>
<td>1.04%</td>
</tr>
<tr>
<td>$\varepsilon_{136}$</td>
<td>0.44</td>
<td>0.05</td>
<td>11.09%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0642</td>
<td>0.0019</td>
<td>2.91%</td>
</tr>
<tr>
<td>$n_{\beta\gamma}$</td>
<td>344</td>
<td>39</td>
<td>11.30%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>6.22</td>
<td>1.26</td>
<td>20.29%</td>
</tr>
</tbody>
</table>

Table 4.10: $^{137}I P_n$ from $\beta$-delayed $\gamma$ rays for 180° $\beta$-ion detector combinations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$I_\gamma$</td>
<td>0.163</td>
<td>0.017</td>
<td>10.43%</td>
</tr>
<tr>
<td>$\varepsilon_\gamma$</td>
<td>0.00540</td>
<td>0.00008</td>
<td>1.56%</td>
</tr>
<tr>
<td>$I_{137}$</td>
<td>1.075</td>
<td>0.010</td>
<td>0.93%</td>
</tr>
<tr>
<td>$n_{\beta136}$</td>
<td>583</td>
<td>34</td>
<td>5.87%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.9619</td>
<td>0.0100</td>
<td>1.04%</td>
</tr>
<tr>
<td>$\varepsilon_{136}$</td>
<td>0.44</td>
<td>0.05</td>
<td>11.09%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0622</td>
<td>0.0019</td>
<td>3.00%</td>
</tr>
<tr>
<td>$n_{\beta\gamma}$</td>
<td>338</td>
<td>40</td>
<td>11.70%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>6.15</td>
<td>1.25</td>
<td>20.41%</td>
</tr>
</tbody>
</table>

Table 4.11: $^{137}I P_n$ from $\beta$-delayed $\gamma$ rays for 90° $\beta$-ion detector combinations.
Figure 4.25: Comparison of BPT $^{137}$I $P_n$ measurements with the IAEA weighted average [43].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_{^{138}}$</td>
<td>1.069</td>
<td>0.020</td>
<td>1.87%</td>
</tr>
<tr>
<td>$n_{^{6137}}$</td>
<td>332</td>
<td>23</td>
<td>6.83%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.83</td>
<td>0.04</td>
<td>4.82%</td>
</tr>
<tr>
<td>$\varepsilon_{^{137}}$</td>
<td>0.44</td>
<td>0.07</td>
<td>15.92%</td>
</tr>
<tr>
<td>$\Omega_{^{136}}$</td>
<td>0.0685</td>
<td>0.0019</td>
<td>2.73%</td>
</tr>
<tr>
<td>$n_{^{\beta}}$</td>
<td>308565</td>
<td>7306</td>
<td>2.40%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>4.63</td>
<td>0.86</td>
<td>18.53%</td>
</tr>
</tbody>
</table>

Table 4.12: $^{138}$I $P_n$ from $\beta$ singles for $180^{\circ}$ $\beta$-ion detector combinations.

to be $0.83 \pm 0.04$ for a 100 keV neutron threshold from previously measured neutron spectra [38, 81]. $\Omega_r$ was determined to be $0.0685 \pm 0.0019$ for the $180^{\circ}$ $\beta$-ion detector pairs and $0.0641 \pm 0.0019$ for the $90^{\circ}$ $\beta$-ion detector pairs. The intrinsic ion detection efficiency, $\varepsilon_r$, was determined in the previous section to be $0.44 \pm 0.05$ for the data at $182V_{pp}$, and $0.44 \pm 0.07$ for the data at $176V_{pp}$ (both for the top MCP detector). Table 4.12 and 4.13 summarize the quantities used in the beta singles measurements. These data sets are combined to determine a $P_n$ of $4.69 \pm 0.80$ (sys) $\pm 0.24$ (stat)% using $\beta$ singles. Similarly, the $\beta$-delayed gamma ray measurements were combined, yielding a $P_n$ of $6.30 \pm 1.21$ (sys) $\pm 0.34$ (stat)%.

Fig. 4.26 compares the BPT $^{138}$I $P_n$ measurements with previous independent measurements, and the most recent results in Refs. [34, 69].
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_{138}$</td>
<td>1.069</td>
<td>0.020</td>
<td>1.87%</td>
</tr>
<tr>
<td>$n_{\beta137}$</td>
<td>339</td>
<td>21</td>
<td>6.17%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.83</td>
<td>0.04</td>
<td>4.82%</td>
</tr>
<tr>
<td>$\varepsilon_{137}$</td>
<td>0.44</td>
<td>0.07</td>
<td>15.92%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0641</td>
<td>0.0019</td>
<td>2.91%</td>
</tr>
<tr>
<td>$n_{\beta}$</td>
<td>328861</td>
<td>7878</td>
<td>3.00%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>4.74</td>
<td>0.87</td>
<td>18.32%</td>
</tr>
</tbody>
</table>

Table 4.13: $^{138}$I $P_n$ from $\beta$ singles for 90° $\beta$-ion detector combinations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$I_\gamma$</td>
<td>0.56</td>
<td>0.05</td>
<td>8.93%</td>
</tr>
<tr>
<td>$\varepsilon_\gamma$</td>
<td>0.0081</td>
<td>0.0001</td>
<td>1.23%</td>
</tr>
<tr>
<td>$\varepsilon_{\beta137}$</td>
<td>1.069</td>
<td>0.020</td>
<td>1.87%</td>
</tr>
<tr>
<td>$n_{\beta137}$</td>
<td>332</td>
<td>23</td>
<td>6.83%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.83</td>
<td>0.04</td>
<td>4.82%</td>
</tr>
<tr>
<td>$\varepsilon_{137}$</td>
<td>0.44</td>
<td>0.07</td>
<td>15.92%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0685</td>
<td>0.0019</td>
<td>2.73%</td>
</tr>
<tr>
<td>$n_{\beta\gamma}$</td>
<td>1083</td>
<td>46</td>
<td>4.25%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>5.99</td>
<td>1.25</td>
<td>20.82%</td>
</tr>
</tbody>
</table>

Table 4.14: $^{138}$I $P_n$ from $\beta$-delayed $\gamma$ rays for 180° $\beta$-ion detector combinations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Abs. Uncertainty</th>
<th>Frac. Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$I_\gamma$</td>
<td>0.56</td>
<td>0.05</td>
<td>8.93%</td>
</tr>
<tr>
<td>$\varepsilon_\gamma$</td>
<td>0.0081</td>
<td>0.0001</td>
<td>1.23%</td>
</tr>
<tr>
<td>$\varepsilon_{\beta137}$</td>
<td>1.069</td>
<td>0.020</td>
<td>1.87%</td>
</tr>
<tr>
<td>$n_{\beta137}$</td>
<td>339</td>
<td>21</td>
<td>6.17%</td>
</tr>
<tr>
<td>$f$</td>
<td>0.83</td>
<td>0.04</td>
<td>4.82%</td>
</tr>
<tr>
<td>$\varepsilon_{137}$</td>
<td>0.44</td>
<td>0.07</td>
<td>15.92%</td>
</tr>
<tr>
<td>$\Omega_{136}$</td>
<td>0.0641</td>
<td>0.0019</td>
<td>2.91%</td>
</tr>
<tr>
<td>$n_{\beta\gamma}$</td>
<td>1057</td>
<td>41</td>
<td>3.86%</td>
</tr>
<tr>
<td>$P_n$</td>
<td>6.61</td>
<td>1.36</td>
<td>20.56%</td>
</tr>
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</table>

Table 4.15: $^{138}$I $P_n$ from $\beta$-delayed $\gamma$ rays for 90° $\beta$-ion detector combinations.
The IAEA weighted average [43] discounts the measurements in Refs. [6, 5].

**Measured neutron energy spectra**

The neutron energy spectra of $^{137}$I and $^{138}$I were reconstructed from the velocity of $^{136}$Xe and $^{137}$Xe recoil ions, respectively, using conservation of momentum. The velocity was determined by the path length and measured TOF of the recoil ions. The TOF and $\frac{1}{v}$ spectra for 90° and 180° $\beta$-ion detector combinations for the $^{137}$I data taken at 230V$_{pp}$ are shown in Figs. 4.27-4.30.

Similarly, the TOF and $\frac{1}{v}$ spectra for 90° and 180° $\beta$-ion detector combinations for the $^{138}$I data taken at 182V$_{pp}$ are shown in Figs. 4.31-4.34. The path length, $p$, was determined by the distance from the trapped ion cloud to MCP distance, $h$, and the 2D spatial coordinates of the ion impact location on the MCP surface ($x$ and $y$). $p$ was calculated using the relation $p = \sqrt{h^2 + x^2 + y^2}$, where $h$ was 52.6 ± 0.9 mm as determined previously. The four signal posts of the MCP were denoted A through D in a clockwise direction as shown in Fig. 4.35. $x$ and $y$ were calculated by the relations $x = \frac{(B+C)-(A+D)}{A+B+C+D} \times s$ and $y = \frac{(A+B)-(C+D)}{A+B+C+D} \times s$, respectively. $s$ was a scaling factor determined from the MCP position calibration using the masks to scale the $x$ and $y$ coordinates to the physical dimensions of the MCP.

The measured neutron energy spectra of $^{137}$I and $^{138}$I are shown in Fig. 4.36 and Fig. 4.37, respectively. Corrections applied include the neutron energy dependent $\beta$ detection efficiency, and the acceleration of the ions in the region between the grid and MCP face. The latter correction was determined by propagating neutral and 2$^+$ $^{136}$Xe recoil ions corresponding to neutron energies between 100 and 1500 keV. The difference in the mean velocities for each charge state are then used to determine a neutron energy dependent correction, which
Figure 4.27: Measured $^{137}$I TOF spectrum for 90° $\beta$-ion detector combination at 230$V_{pp}$.

Figure 4.28: Measured $^{137}$I TOF spectrum for 180° $\beta$-ion detector combination at 230$V_{pp}$.
Figure 4.29: Measured $^{137}\text{I} \frac{1}{v}$ spectrum for $90^\circ$ $\beta$-ion detector combination at $230V_{pp}$.

Figure 4.30: Measured $^{137}\text{I} \frac{1}{v}$ spectrum for $180^\circ$ $\beta$-ion detector combination at $230V_{pp}$. 
Figure 4.31: Measured $^{138}$I TOF spectrum for $90^\circ$ $\beta$-ion detector combination at $182V_{pp}$.

Figure 4.32: Measured $^{138}$I TOF spectrum for $180^\circ$ $\beta$-ion detector combination at $182V_{pp}$. 
Figure 4.33: Measured $^{138}\text{I} \frac{1}{v}$ spectrum for $90^\circ$ $\beta$-ion detector combination at $182V_{pp}$.

Figure 4.34: Measured $^{138}\text{I} \frac{1}{v}$ spectrum for $180^\circ$ $\beta$-ion detector combination at $182V_{pp}$. 
is then applied to the measured neutron energy spectrum. The neutron energy resolution obtained in this experiment is on the order of $\approx 10\%$ FWHM.

The $^{137}$I neutron energy spectrum measured in this work was compared to previous measurements in Ref. [63], where the low-energy neutron component of our measurement below 200 keV is larger than in the previously published data. A similar comparison was made for the measured $^{138}$I spectrum to previous measurements by Greenwood [38] and Shalev [82]. The measured spectrum has a larger low-energy neutron component below 300 keV compared to the Greenwood data [38], but is consistent with the Shalev data [82]. The discrepancies may be due to highly-charged ions in the present measurements contaminating the low-energy part of the neutron spectrum, or the reliability of the $^3$He and proton recoil response functions used in the previous measurements (in the case of the $^{137}$I measurement by Ohm [63] and the $^{138}$I measurement by Greenwood [38]).
Figure 4.36: Measured $^{137}$I $\beta n$ energy spectrum.

Figure 4.37: Measured $^{138}$I $\beta n$ energy spectrum.
Chapter 5

Conclusions & Future work

A novel method for studying $\beta$-delayed neutron energy spectra and branching ratios has been demonstrated by measuring the large momentum kick imparted to the nucleus following the $\beta$ decay of trapped $^{137}$I, with the results of a proof-of-principle in 2011 consistent with existing measurements. An experimental campaign was conducted in 2012, working towards improving the technique in three areas: decreasing the neutron energy threshold, improving the neutron energy resolution, and demonstrating a better measurement of $P_n$. This was achieved by designing and implementing several upgrades to the apparatus as described in Chapter 4. Following these improvements, an improved measurement of the $^{137}$I $P_n$ was made, complemented by a measurement of its neutron energy spectrum with a $\approx 10\%$ FWHM resolution. In addition, $^{138}$I was studied to resolve a discrepancy between previous independent $P_n$ measurements. The result of our measurement is consistent with the IAEA weighted average, taken from the two most recent $P_n$ measurements. It resolves the discrepancy using a new technique with different systematic effects than previous measurements. An experimental campaign is currently being planned for the end of 2013, aimed at collecting significantly higher statistics at CARIBU using the ion trap and detector configuration from 2012. Isotopes with half-lives as short as $\approx 50$ ms can be studied, and $\beta n$ measurements relevant to the r-process, nuclear reactor design, and stockpile stewardship will be performed. Beyond 2013, a dedicated ion trap and detector system are being designed for this work. A summary of the two previously completed campaigns and the planned 2014 campaign is shown in Table. 5.1, highlighting the lower RF amplitude for a new trap design and much larger detector solid angles for $\beta$, ion, and $\gamma$-ray detection for the setup at CARIBU.

Currently, the uncertainties associated with the $P_n$ measurements are limited by a systematic uncertainty, namely the ion detection efficiency $\varepsilon_r$. The leading systematics in determining $\varepsilon_r$ are the trapped ion to MCP distance, knowledge of the charge state distribution of the $\beta$-decay recoil ions, and the correction due to contaminant recoil ions from the isobaric impurities loaded into the trap. The correction due to contaminant recoil ions can be eliminated if superior isobaric separation can be achieved prior to loading ions in the trap, by using a high-resolution double-focusing magnetic spectrometer, purifier Penning trap, or reflectron. The correction due to the charge state distribution can be mitigated by placing
a secondary grid between the trapped ion species and the primary grid in front of the MCP face. The secondary grid can be positively biased at a value which will create a potential hill insurmountable by $>3^+$ ions. Future measurements of the ion detector efficiency should consider using an isotope with a simple, well-known decay scheme such as $^{96}\text{Sr}$. This would limit the uncertainties in the simulations due to uncertainties in the decay scheme (i.e. knowledge of $a_{\beta\nu}$, $\beta$ decay branching ratios). This, in addition to constraining the charge state of the ions that hit the detector, will improve the determination of the trapped ion to MCP distance.

The neutron energy threshold was improved from $\approx 200$ keV to 100 keV between the proof-of-principle and 2012 measurements. It is anticipated that for the optimized ion trap with a 7 mm trap radius, the threshold can be lowered to a value of 25–50 keV, where the neutron and lepton recoils are comparable.

For the first time, delayed neutrons have been studied via the nuclear recoil, made possible by leveraging modern ion trapping techniques and radiation detectors. This thesis work is a first step towards precision measurements of $P_n$ and $E_n$ for $\beta\nu$ spectroscopy relevant to r-process nucleosynthesis, nuclear reactor design, and stockpile stewardship. The isotopes of interest to these research areas will be accessible in the near-term at fission-fragment beam facilities such as CARIBU, and in the near future at fragmentation facilities where a stopped radioactive beam infrastructure is available.

<table>
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<tr>
<th></th>
<th>2011 proof-of-principle</th>
<th>2012 campaign</th>
<th>Planned 2014 CARIBU</th>
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</thead>
<tbody>
<tr>
<td>$^{137}\text{I}$ rate</td>
<td>30 Hz</td>
<td>20 Hz</td>
<td>20,000 Hz</td>
</tr>
<tr>
<td>Trap radius</td>
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<td>11 mm</td>
<td>7 mm</td>
</tr>
<tr>
<td>$V_{RF}$</td>
<td>200 V</td>
<td>85 V</td>
<td>30 V</td>
</tr>
<tr>
<td>$\Omega_\beta$</td>
<td>3%</td>
<td>10%</td>
<td>12%</td>
</tr>
<tr>
<td>$\Omega_r$</td>
<td>3%</td>
<td>10%</td>
<td>26%</td>
</tr>
<tr>
<td>$\Omega_\gamma$</td>
<td>10%</td>
<td>10%</td>
<td>28%</td>
</tr>
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<td>1%</td>
<td>3%</td>
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<td>10%</td>
<td>10%</td>
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<tr>
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<td>150 keV</td>
<td>25 keV</td>
<td>25 keV</td>
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Table 5.1: Summary of progress in the generations of the technique.
Bibliography


