

Mass measurements of neutron-rich nuclides for the astrophysical r process using the Canadian Penning trap mass spectrometer

by

Dwaipayan Ray

October 25, 2022

A thesis submitted to the Faculty of Graduate Studies, University of Manitoba in partial
fulfillment of the requirements of the degree of

Doctorate of Philosophy

Department of Physics and Astronomy

University of Manitoba

Winnipeg, Manitoba, Canada

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Abstract

About half of the elements heavier than iron ($Z = 26$) are believed to be formed via the r process (rapid neutron capture process), of which our understanding is limited. Running theoretical simulations to gain insight into this r process rely on the availability of nuclear data (like nuclear masses, beta decay lifetimes, neutron capture cross sections and others) with reduced uncertainties for nuclides near the expected r -process path. Such data is available in limited capacity due to the challenges in producing those exotic nuclides. The astrophysical conditions required for the r process, and the events that generate them are still open questions. The CALifornium Rare Isotope Breeder Upgrade (CARIBU) at Argonne National Laboratory (ANL) uses the spontaneous fission from a ^{252}Cf source and is one of the few facilities world-wide to produce beams of such neutron-rich isotopes around the mass numbers $A = 105$ and $A = 140$. The extracted ions are subject to multiple stages of cooling and purification, before they are sent to the Canadian Penning trap (CPT) mass spectrometer, where their masses are measured using the Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique. The main focus of this work was to provide data on atomic masses that would allow simulations to replicate the rare-earth peak in the observed r -process abundance pattern. Masses of a total of 34 nuclides are presented in this thesis, of which seven were measured for the first time, including the first-time measurements of isotopes of Ce ($Z = 58$): $^{152-155}\text{Ce}$. The results of these Ce isotopes, with their uncertainties reduced from the extrapolated values of $\geq 200 \text{ keV}/c^2$ in the latest Atomic Mass Evaluation (AME2020), to $\leq 6 \text{ keV}/c^2$, agree with mass predictions from a reverse engineered Markov Chain Monte Carlo technique for a hot r -process environment, hence validating this “hot” r -process model for this mass region.

Preface

I started my PhD research in 2017 with the Canadian Penning trap (CPT) project at the CALifornium Rare Isotope Breeder Upgrade (CARIBU) facility at the Argonne Tandem Linac Accelerator System, Argonne National Laboratory. Between 2017 and 2018, I acquainted myself with the project, gaining valuable hands-on experience with the complex apparatus and the Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) mass measurement technique. During this time, I was involved in three mass measurement campaigns, the results of which were published in multiple publications, including in two PhD theses, and two journals. In the middle of 2018, I became the sole PhD student with the CPT group upon the graduation of my predecessor. Soon after, a quench of our superconducting magnet occurred. It took us almost two months to re-energize the magnet, and return to operational ultra-high vacuum conditions. After that, I calibrated the Penning trap for the new magnetic field, which required another three months. Since then, I have organized over 10 successful mass measurement campaigns, leading to 2 publications to date as co-authors and three more, currently in preparation. The measurements in this thesis were conducted in June, July and October 2019, and in August, October and December of 2021. The COVID-19 enforced lockdowns in 2020-2021 introduced a series of complications in the experiment including but not limited to instabilities and break-down of multiple devices. These, coupled with an overall drop in the extraction of exotic nuclides from CARIBU by 70% of the expected numbers, have severely affected the mass measurement program and limited the range of accessible nuclei. In spite of these challenges, it was possible to measure the masses presented here. During the measurement campaigns, my first responsibility was optimizing beam production from the CARIBU facility. This included extraction of rare isotope beam (RIB) from the CARIBU gas-catcher, and tuning all the different beam-line components to ensure that a RIB with optimal transmission and mass resolution is being transported to the CPT. Once in the CPT, the ion species in the RIB would have to be identified before starting precision measurement of their masses. I developed a data analysis code for online and offline analysis, including particle identification and mass measurements. Finally, I have spent a considerable time understanding and quantifying the systematic effects, and exploring ways to minimize

them in our measurements. In between runs, I was in charge of maintaining, troubleshooting and optimizing the whole system. This included performing upgrades like installation of offline sources, ensuring the stability of the system, and identifying and repairing or replacing faulty devices.

List of acronyms

AC : Alternating current

ADC : Analog-to-digital converter

AME : Atomic Mass Evaluation

AMP : Amplifier

ANL : Argonne National Laboratory

ATLAS : Argonne Tandem Linac Accelerator System

BNG : Bradbury Nielsen gate

CAMAC : Computer-aided measurement and control

CARIBU : CALifornium Rare Isotope Breeder Upgrade

CFD : Constant fraction discriminator

CPT : Canadian Penning trap mass spectrometer

DAC : Digital-to-analog converter

DAN : Data analysis

DAQ : Data acquisition

DC : Direct current

DZ : Duffo Zuker mass model

EM : Electro-magnetic

FAMP : Fast amplifier

FRDM : Finite Range Droplet Model

FWHM : Full width half maximum

GPB : General purpose interface bus

HFB : Hartree-Fock-Bogoliubov mass model

HV : High voltage

KTUY : Koura-Tachibana-Uno-Yamada mass model

LMF : List mode file

LT : Linear trap

MCMC : Markov Chain Monte Carlo

MCP : Microchannel plate

MNT : Multi neutron transfer
 MR-TOF : Multi-Reflection Time-Of-Flight
 NIM : Nuclear instrumentation module
 NS : Neutron star
 NSM : Neutron star merger
 p process : Proton capture process
 PI-ICR : Phase-Imaging Ion-Cyclotron-Resonance
 PID : Particle identification
 PS-MCP : Position-sensitive microchannel plate
 r process : Rapid neutron capture process
 REP : Rare-earth peak
 RF : Radio-frequency
 RFQ : Radio-frequency quadrupole
 RMS : Root mean square
 rp process : Rapid proton capture process
 s process : Slow neutron capture process
 SHV : Safe high voltage
 Si : Silicon surface barrier semiconductor detector
 SNe : Core-collapse supernovae
 SRS : Stanford Research System
 TDC : Time-to-digital converter
 TOF : Time-of-flight
 TOF-ICR : Time-Of-Flight Ion-Cyclotron-Resonance
 TP : Turbo pump
 TRP : Trap raising pulse
 TTL : Transistor-transistor logic
 UHV : Ultra high vacuum
 WS : Weizsäcker-Skyrme mass model

1 Introduction

The work behind this thesis connects nuclear physics and astrophysics to shed light on the nucleosynthesis of heavier elements in the universe, by way of mass measurements of exotic nuclides using the Canadian Penning trap (CPT) mass spectrometer at the CALifornium Rare Isotope Breeder Upgrade (CARIBU) facility, Argonne National Laboratory (ANL). Mass is one of the most fundamental properties of an object and is defined as the total quantity of matter contained in the object irrespective of its size. The mass of an atom cannot be calculated by simply adding the masses of constituent neutrons (m_n), protons (m_p) and electrons (m_e). One has to consider the binding energies (BE), electron and nuclear, to accurately determine the mass. The electron binding energy is usually many orders of magnitude smaller than m_n , m_p , m_e and the nuclear BE , and will be ignored in the context of this thesis. The mass of an atom with Z protons and N neutrons is hence given as:

$$m(Z, N) = Z(m_p + m_e) + Nm_n - BE(Z, N)/c^2, \quad (1.1)$$

where c is the velocity of light in vacuum. Therefore mass measurements of nuclides, that is atoms characterized by their nucleons, can provide valuable insights into the forces binding the nucleus together. The most commonly used unit of atomic mass is the unified atomic mass unit (u): $1 \text{ u} = (1660539.0666 \pm 0.0005) \times 10^{-33} \text{ kg}$ [1]. It is defined as 1/12th of the mass of the ground state of a neutral carbon-12 (^{12}C) atom at rest [2]. Another way of expressing atomic masses is in terms of *mass excess* (ME), defined as difference between its actual mass (m) in unit of u and its mass number (A), that is $ME = (m - A) \times \mathcal{C}_m \text{ keV}$ where $\mathcal{C}_m$ is a u to keV conversion constant given as $\mathcal{C}_m = 931494.10242(28) \text{ keV/u}$ [1].

Figure 1.1 shows a nuclear chart, where the two axes X and Y represent the number of neutrons (N) and protons (Z) respectively. Each position on the chart shows a nuclide of a chemical element, naturally occurring on earth or synthetically created, color-coded on the basis of their half-lives ($t_{1/2}$). $t_{1/2}$ is a fundamental property of the nucleus, and is defined as the time taken by a certain quantity of the nucleus to decay to one-half of its initial amount. The black dots represent the stable nuclides with $t_{1/2} > 1 \text{ Gy}$, and this region is often referred

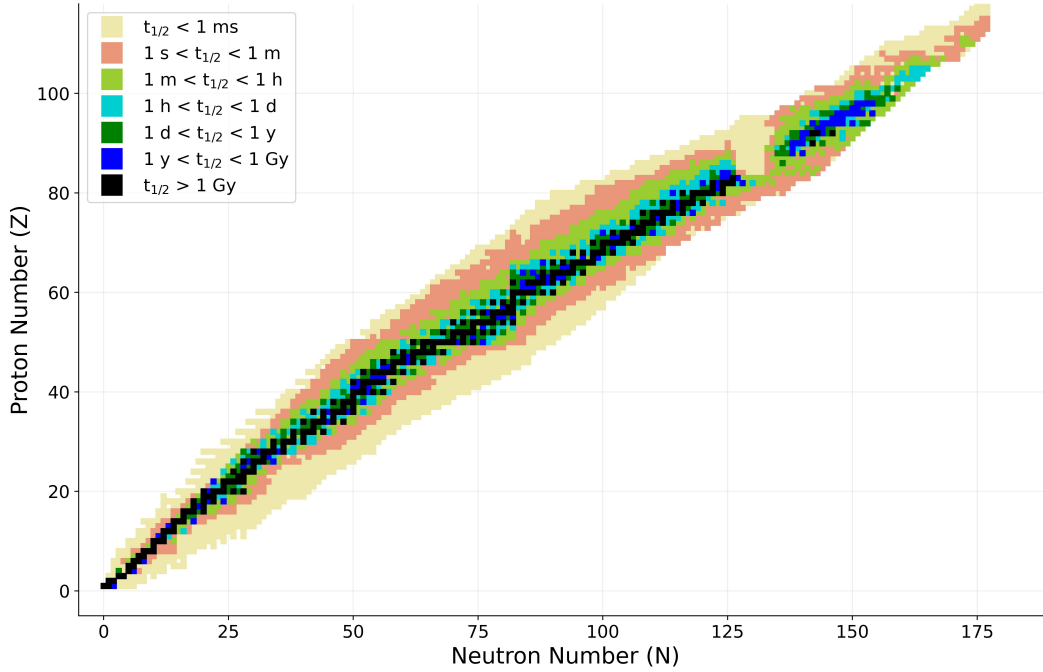


Figure 1.1 : Nuclear chart showing the half-lives ($t_{1/2}$) of all nuclei. The $t_{1/2}$ data has been taken from the NUBASE2020 [3].

to as the *valley of stability*. As one moves away from this *valley* in the chart to the proton-rich side (Y direction) or neutron-rich side (X direction), $t_{1/2}$ of the nuclides keep decreasing. These short-lived or unstable nuclides decay by different nuclear decay mechanisms to reach stable configurations. The most common decay mechanism in the context of this thesis is the β -decay (β^-). In β^- -decay, a neutron decays into a proton inside the nucleus, keeping the total number of nucleons (A) the same. This reaction is accompanied by the emission of an electron e^- and an electron anti-neutrino $\bar{\nu}_e$. As a result, a parent nucleus $^A_Z X_N$ decays into the daughter nucleus $^A_{Z+1} X_{N-1}$:

$$^A_Z X_N \rightarrow ^A_{Z+1} Y_{N-1} + e^- + \bar{\nu}_e, \quad (1.2)$$

1.1 Astrophysical motivation

The origin of the chemical elements in the universe has motivated multiple scientific studies in the fields of astrophysics and nuclear physics over the past several decades. The formation of lighter nuclides up to iron (atomic number $Z = 26$) have been well researched. Nuclides from ^1H to ^7Li were formed by the Big Bang Nucleosynthesis at the beginning

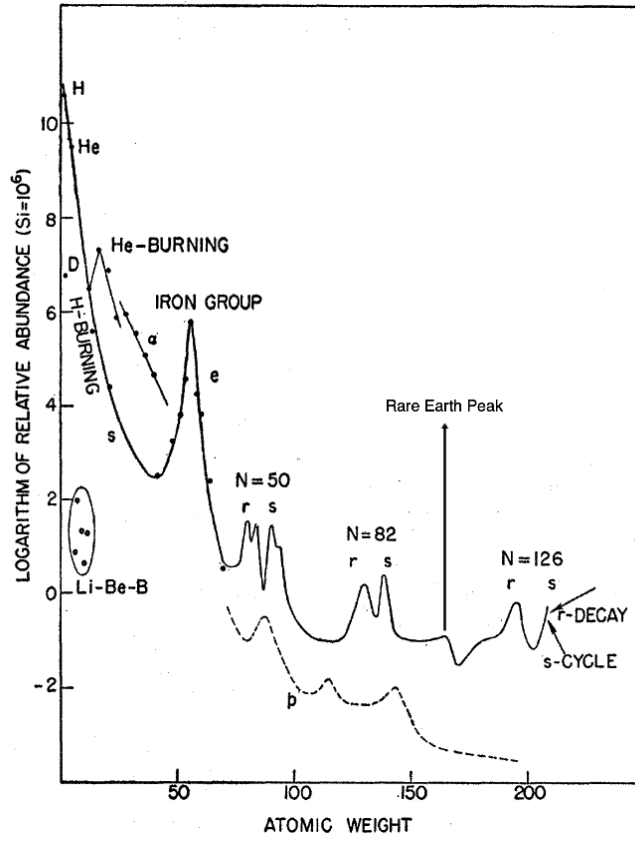


Figure 1.2 : Relative elemental abundance as a function of atomic weight. Figure adapted from Ref. [4] and the annotation “Rare Earth Peak” was added.

of the universe [5]. From ${}^7\text{Li}$ to ${}^{56}\text{Fe}$, the nuclides were formed by various stellar fusion processes [4, 6, 7]. Since ${}^{56}\text{Fe}$ has the maximum binding energy per nucleon, making it the most stable nuclide, spontaneous fusion to produce heavier nuclides is not energetically feasible. These nuclides are formed by capture processes: proton-capture processes (like the p and rp processes) for the proton-rich nuclides; and neutron-capture processes, slow (s) and rapid (r), for the neutron-rich nuclides. This is shown in Figure 1.2 [4], where the solar abundance pattern is plotted against the atomic weights of the nuclides, with their formation mechanisms annotated. Coulomb repulsion limits the formation of the proton-rich nuclides, and therefore the two neutron-capture processes can each account for nearly half of the nuclides with $Z > 26$ [4, 8–10]. Additional stability provided by the neutron shell closures mean that most of these neutron-rich nuclides cluster around the magic neutron numbers $N = 50, 82$, and 126 , as shown by the double peaked structures around mass number $A \sim 85, 135$, and 200 in Figure 1.2.

The mechanism governing the s and r processes is similar: an initial stable or long-lived seed nucleus captures one or more neutrons (n, γ) before it β -decays (Equation 1.2). In an (n, γ) reaction, the initial nucleus ${}^A_Z X_N$ captures a neutron, accompanied by the emission of γ rays:



The difference between the two processes lies in the competing rates of the two processes because of the environments where they occur. The s process takes place in environments of neutron concentration ($\mathcal{N}_n$) $\sim 10^8$ n/cm³ and temperatures (T) ~ 0.35 GK, typically found in quiescent stars. The moderate $\mathcal{N}_n$ means the seed nuclide captures neutrons at a time scale that is typically longer than that of the β -decay lifetimes for the nuclides involved, allowing the daughter nuclei to β -decay towards stability after every neutron capture. This results in the s -process path not venturing far from the region of stability in the chart of nuclides, where the naturally occurring nuclides lie. These s -process nuclides are easily producible for research and the process has been well studied and understood [11–15].

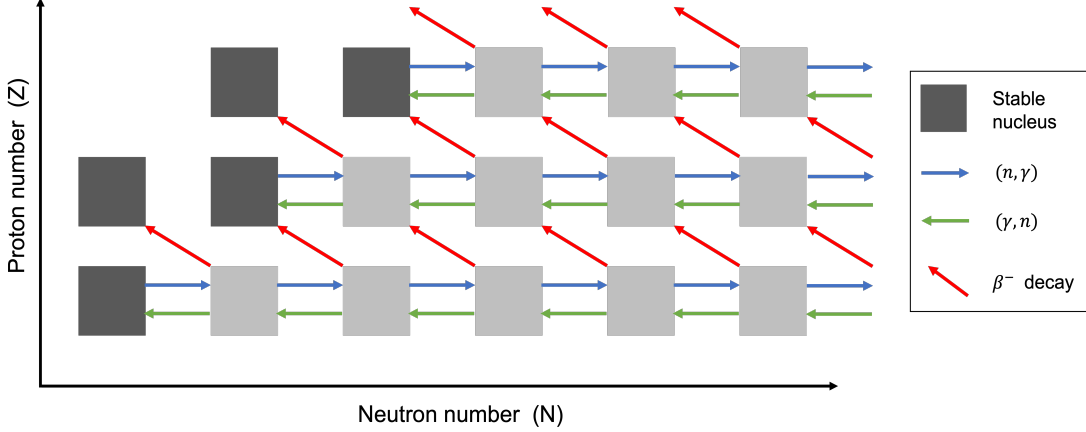


Figure 1.3 : Cartoon illustrating the r process. The blue, green, and red arrows indicate (n, γ) , (γ, n) , and β -decay respectively. The stable nuclei are shown with dark grey squares.

1.1.1 The r process

In this section, I will start by describing the r process, before discussing its sites, and finally I will connect it to mass measurements. The r process takes place in environments of high neutron concentrations ($\mathcal{N}_n \geq 10^{21} \text{ n/cm}^3$) and temperatures ($T \sim 1 \text{ GK}$) [15], typically found in explosive astrophysical phenomena. The process involves three main reactions: neutron capture (n, γ) (Equation 1.3), photodissociation (γ, n) (Equation 1.4), and β (β^-) decay (Equation 1.2). (γ, n) reactions take place as:

$${}^A_Z X_N + \gamma \rightarrow {}^{A-1}_Z X_{N-1} + n \quad (1.4)$$

The reactions are further illustrated in Figure 1.3, where a part of the r process is shown, with the dark and light grey boxes representing the participating nuclides and the blue, green, and red arrows depicting the three reactions. Due to the large density of neutrons in the neighbourhood, the stable seed nuclei (shown in dark grey squares) capture multiple neutrons (in blue arrows going left to right) as the process navigates through the neutron-rich part of the nuclear chart. On the other hand, as a result of the high temperature, the nuclides undergo simultaneous (γ, n) reactions (shown in green arrows pointing left), which lowers the number of neutrons in the nuclides. Finally, the unstable nature of the nuclides means they always tend to β decay towards stability. These are represented by the red diagonal arrows and take the process path to the next element, that is an increase in the

proton number Z by one.

Because of the extreme conditions necessary for the r process, which cannot be created in a laboratory, studies are conducted mainly using simulations, which utilize different astrophysical models and nuclear physics properties to output an elemental abundance prediction. The success of a simulation depends on how accurately it can reproduce the solar r -process abundance pattern. The temperatures used in these models can broadly characterize the r process into two types: the hot r process and the cold r process. The hot r process, as the name suggests, takes place in hotter environments ($T > 1$ GK). In such scenarios, (γ, n) plays a dominant role and the path of the process is guided by competing rates of the (n, γ) and (γ, n) reactions. Along an isotopic chain, the abundance distribution hence proceeds as [15]:

$$\frac{d\mathcal{N}_{Z,N}}{dt} = \underbrace{-\mathcal{N}_n \mathcal{N}_{Z,N} \langle \sigma v \rangle_{Z,N}}_{(n,\gamma)} + \underbrace{\mathcal{N}_{Z,N+1} \lambda_\gamma(Z, N+1)}_{(\gamma,n)} \quad (1.5)$$

where, $\mathcal{N}_{Z,N}$ is the number density of a nuclide A_ZX_N having a (n, γ) cross section of $\langle \sigma v \rangle_{Z,N}$ and (γ, n) decay constant of $\lambda_\gamma(Z, N)$. Under the influence of the high surrounding temperature and neutron density, an eventual equilibrium between (n, γ) and (γ, n) is reached, that is $\frac{d\mathcal{N}_Z(A)}{dt} \approx 0$. The abundances Y of two neighbouring species of the same element (Z) at such equilibrium can be deduced from the Saha equation, as [15]:

$$\begin{aligned} \frac{Y_{Z,N+1}}{Y_{Z,N}} &= \frac{\mathcal{N}_{Z,N+1}}{\mathcal{N}_{Z,N}} \\ &= \frac{(2j_{Z,N+1} + 1)}{(2j_{Z,N} + 1)(2j_n + 1)} \frac{G_{Z,N+1}^{\text{norm}}}{G_{Z,N}^{\text{norm}}} \left(\frac{h^2}{2\pi M_{A,n} k_B T} \right)^{3/2} \mathcal{N}_n \exp \left(\frac{S_n(Z, N+1)}{k_B T} \right) \end{aligned} \quad (1.6)$$

where, $j_{Z,N}$ and $G_{Z,N}^{\text{norm}}$, are the spin and normalized partition function of the species A_ZX_N , j_n is the neutron spin, h and k_B are the respective Planck's and Boltzmann's constants, $M_{A,n}$ is the reduced mass of A_ZX_N and a neutron, and $S_n(Z, N)$ is its neutron separation energy of A_ZX_N , given by:

$$S_n(Z, N) = \left[M(Z, N-1) + M_n - M(Z, N) \right] c^2 \quad (1.7)$$

where $M(Z, N)$ and M_n are the masses of nuclide A_ZX_N and a neutron, respectively. At this point of equilibrium, the nuclide cannot evolve any further unless it β decays to the next element $(Z + 1)$ and starts undergoing (n, γ) and (γ, n) processes again. Taking $G_{Z,N+1}^{\text{norm}} \approx G_{Z,N}^{\text{norm}}$, $j_{Z,N+1} = j_{Z,N} = 1$ and $M_{A,n} \approx M_n$, we can get:

$$\frac{Y_{Z,N+1}}{Y_{Z,N}} = \frac{\mathcal{N}_{Z,N+1}}{\mathcal{N}_{Z,N}} = \frac{\mathcal{N}_n}{1.188 \times 10^{34} T_9^{3/2}} \exp \left(\frac{11.605}{T_9} S_n(Z, N + 1) \right) \quad (1.8)$$

where T_9 is the temperature in GK. From equation 1.8, putting $T_9 = 1$ and $\mathcal{N}_n = 10^{22}$, it can be seen that if $\mathcal{N}_{Z,N+1} \approx \mathcal{N}_{Z,N}$, $S_n \approx 2$ MeV. In other words, in an atmosphere with the above mentioned temperature and neutron number density, the abundance maximum along any isotopic chain will occur at the nuclide with $S_n \approx 2$ MeV, regardless of the element. An increase in $\mathcal{N}_n$ will push the maximum towards the neutron rich side while a rise in T will shift it towards the valley of stability [15].

This explanation however ignores the odd-even effects in nuclear binding energy described by the pairing term in the semi-empirical mass formula [4]. Thus, it will be more appropriate to use the two neutron separation energy, S_{2n} , in the above expression in the place of S_n , as [9]:

$$S_{2n}(Z, N + 2) = S_n(Z, N + 2) + S_n(Z, N + 1) \approx 2S_n(Z, N + 1) \quad (1.9)$$

This creates significant isotopic abundance for all even N isotopes. The $(n, \gamma) \rightleftharpoons (\gamma, n)$ equilibrium is also known as the waiting point approximation, as the abundance evolution will “wait” at the nuclide with $S_{2n} \approx 4$ MeV till it β decays and moves one up in Z , before the whole process repeats again. On the other hand, a cold r process takes place in relatively lower temperature ($T_9 \sim 0.5$). Under such conditions, photodissociation’s role is reduced. Therefore the path is set by competition between (n, γ) and β decay [16, 17].

Although the main decay mode a nuclide involved in the r process undergoes is β decay, there are other decay modes that take place at different stages along the r -process path. Sometimes β decay populates an excited state in the daughter nuclide, which then decays by emission of one or more β -delayed neutrons (n_β -s) [9]. This phenomenon leads to a smoothing of the abundance curve by restricting the nuclear pairing effect due to odd-even number of

neutrons. With the formation of heavier nuclides ($Z > 80$), sometimes spontaneous fission also leads to a nuclide decaying back to a lighter nuclide in the r -process path. This is known as fission recycling and can have a significant impact in the final elemental abundance. For even heavier nuclides ($A \geq 210$), the path reaches nuclei susceptible to α decays. This leads to the α -decay chains which produce some species with long half-lives [15].

One thing that must be mentioned here is that, the waiting point approximation, where it is assumed that a nuclide will β decay once the $(n, \gamma) \rightleftharpoons (\gamma, n)$ equilibrium is attained, is a vast simplification. In practice, at every location, a nuclide is capable of undergoing all three reactions, and a nuclide's evolution can be determined from [9]:

$$\begin{aligned} \frac{d\mathcal{N}_{Z,N}}{dt} = & \underbrace{\mathcal{N}_n \left[\mathcal{N}_{Z,N-1} \langle \sigma v \rangle_{Z,N-1} - \mathcal{N}_{Z,N} \langle \sigma v \rangle_{Z,N} \right]}_{(n,\gamma)} \\ & + \underbrace{\mathcal{N}_{Z,N+1} \lambda_\gamma(Z, N+1) - \mathcal{N}_{Z,N} \lambda_\gamma(Z, N)}_{(\gamma,n)} \\ & + \underbrace{\mathcal{N}_{Z-1,N} \lambda_\beta(Z-1, N) - \mathcal{N}_{Z,N} \lambda_\beta(Z, N)}_{\beta \text{ decay}} \end{aligned} \quad (1.10)$$

Once the astrophysical phenomenon ends, that is the neutron source freezes out (time-scale $\sim$ s), the neutron concentration drops and the capture process terminates. The nuclides formed until this point then β decay towards stability, producing most of the r -process nuclides (that is, the nuclides assumed to be created from the r process). The exact location and time-scales of the cessation of the process depends on the fission cross section of these far-from-stability nuclei and the surrounding environment respectively [15, 18].

One of the characteristic features of the r process as can be seen from Figure 1.2 [4] is the formation of abundance peaks around magic neutron numbers $N_{\text{magic}} = 50, 82$, and 126. The neutron shell closures at these magic neutron numbers provide increased stability, and hence decreased (n, γ) cross section. A nuclide, (with $Z = Z_i$) at these locations cannot capture any more neutrons and undergoes one β decay instead, moving to an $N = N_{\text{magic}} - 1$ and $Z = Z_f = Z_i + 1$ location. However, the increased stability at $N = N_{\text{magic}}$ also makes the (n, γ) cross section for $N = N_{\text{magic}} - 1$ quite high. Therefore, after the β decay, the nuclide quickly captures a neutron to go back to closed shell configurations at this $Z = Z_f$.

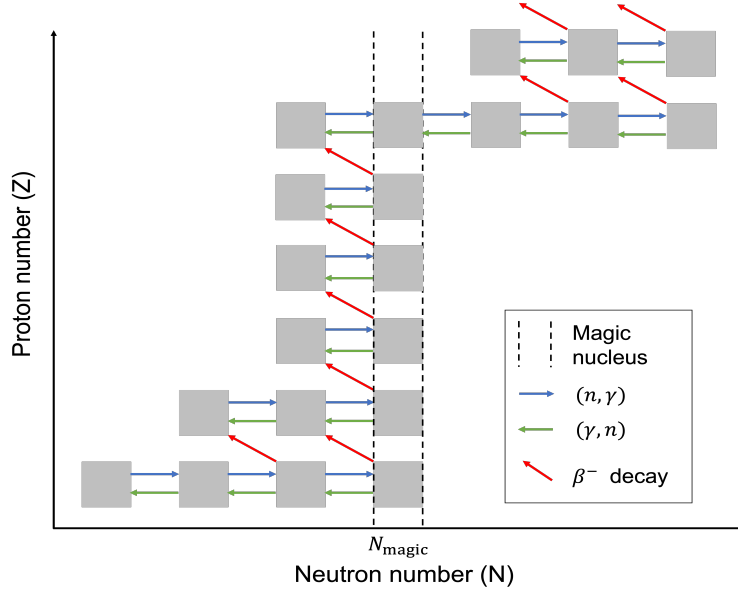


Figure 1.4 : A possible portion of the r -process path. As before, the blue, green, and red arrows indicate (n, γ) , (γ, n) , and β -decay respectively. A closed neutron shell is shown by the two dashed vertical lines.

This leads to the formation of a series of waiting points and the r -process path proceeds vertically along Z (Figure 1.4) [15]. After a few such successive β -decay — (n, γ) processes, the Coulomb repulsion, due to the increased number of protons resulting from the β decays, reaches its threshold and then the nuclide starts capturing multiple neutrons again, leading to a repeat of the process. This is shown in Figure 1.4, where a portion of a possible r -process path is shown. The blue, green, and red arrows indicate the (n, γ) , (γ, n) , and β decay reactions, while the dashed vertical lines represent a neutron magic number. This leads to the formation of peaks at these magic neutron number locations in the solar abundance pattern.

Circling back to Figure 1.2, a small peak can be seen around $A \sim 160$, in between the double peak structures due to the neutron shell closures at $A = 135$ and 200 ($N = 82$ and 126), known as the rare-earth peak (REP). This was first mentioned in the famous B²FH paper [4]. Simulations showed that REP formation can occur in both hot [17, 19] and cold r processes [17]. In the hot environment, nuclear deformation turns out to be a necessary condition. As a nuclide approaches the mass number $A = 160$, it starts to deform. This leads to a spike in its neutron separation energy leading to the nuclide capturing more

and more neutrons, till it reaches a deformation maximum. These additional neutrons make such nuclides β decay more than the nuclides “after” the spike, which are more likely to photodissociate. This leads to the formation of the REP once β -decay takes prominence. For a cold process however, the neutron capture (n, γ) rate becomes more important in the REP formation. Calculations show that, under the cold conditions, once the neutron concentration falls to $\sim 10^{18}$ n/cm³ closer to freeze out, the (n, γ) rates before ($A = 150-159$) and after ($A = 168 - 180$) the REP are much larger than the β -decay rate. In the REP region, a trough in the (n, γ) rates allows the nuclei here to β decay faster than they can capture more neutrons. This results in an accumulation of material in this region, forming the REP [17].

Over the years, numerous studies have been conducted to identify the r -process sites. Simulations using different astrophysical conditions have been able to reproduce different features, like the different peaks, of the r -process abundance pattern, meaning the r process could take place in multiple astrophysical conditions in one or more astrophysical sites [20]. The two most studied candidates of r -process sites are neutrino-driven winds from core-collapse supernovae (SNe) leading to the formation of a neutron star [9, 20, 21], and neutron star mergers (NSMs) [22–24]. Simulations have shown that even though SNe can produce the lighter r -process nuclides, they are not neutron-rich enough to produce the heavy r -process peak around $A \sim 195$ [15, 20, 22]. Most of the modern day simulations instead focus on NSMs [16]. When two neutron stars (NSs) come close to each other, mass exchange takes place in the form of long tidal arms around the NSs. This leads to the formation of a disc around the merging NSs. During this process, the nucleons in their crust combine, alongside undergoing radioactive decays, to generate heat and expand the material in the disc. This might lead to a portion of the arms being lost in the form of ejecta. Due to the high surrounding temperature and neutron concentration, NSMs are an ideal location for r processes.

A NSM is expected to release huge amounts of energy in the form of gravitational waves (GWs). However, these GWs should be accompanied by short gamma-ray bursts and electromagnetic radiation (EM) counterparts. If the NSMs are r process sites, the materials ejected during or immediately after an NSM would contain r -process elements, namely the

lanthanides and actinides, which would increase the opacity (or equivalently, decrease the luminosity) of the EM waves, shifting the detected spectra towards the infrared. Also, a short delay between the EM and GW waves and the isotropic nature of such an EM emission makes it a perfect candidate to prove the NSMs as r -process sites [23, 24]. On August 17, 2017, gravitational waves from the merging of two NSs were detected for the first time by the advanced LIGO and VIRGO collaboration [25]. A few hours after the merger (GW170817), the EM counterpart was detected by various telescopes including the Southern Astrophysical Research (SOAR) telescope (Cerro Pachón, Chile), Swope, du Pont and Magellan telescopes (Las Campanas Observatory, Chile), Keck-I telescopes (W. M. Keck Observatory, USA) and the Hubble Space Telescope (NASA, USA). The detected spectra showed an initial blue component evolving towards red and infrared over a span of ~ 10 days, as is expected for optical transients powered by radioactive decays of r -process nuclides in the NSM ejecta. Thus, this EM counterpart, variously named SSS17a (Swope Supernova Survey 2017a), DLT17ck, and AT2017gfo provided evidence, for the first time, of r process occurring in NSMs [26–28]. Additionally, upon reanalysis of the AT2017gfo spectra, the presence of the r -process element strontium had been identified [29], thereby making a stronger argument in favour of NSMs being an r -process site. But the presence of multiple environments inside the merging neutron stars means the exact conditions of the process still elude us. Furthermore, the infrequent nature of the occurrence of NSMs raises the question of whether NSMs alone can account for all r -process nuclides in the universe. Therefore we require more experimental, observational and simulation-based studies to completely understand the r process, its sites, and the exact conditions necessary for its occurrence.

1.1.2 r process and mass measurements

Nuclear physics plays a very important role in r -process simulations. As mentioned before, simulations use precise nuclear physics inputs in addition to astrophysical models. Masses are arguably one of the most important physical properties in r -process calculations. For a hot process, along an isotopic chain, the main reactions taking place are (n, γ) and (γ, n) , and the path in a hot r process is set by the $(n, \gamma) \rightleftharpoons (\gamma, n)$ equilibrium. The photodissociation rate can be found from [16]:

$$\lambda_\gamma(Z, N) \propto \exp\left(-\frac{S_n(Z, N)}{k_B T}\right) \quad (1.11)$$

with the symbols having the same meanings as Equations 1.5 and 1.6. The neutron separation energy is given by Equation 1.7. For a cold r process, masses can have slightly reduced importance because of the reduced role of photodissociation, but are critical nonetheless as they are used to calculate other properties. The β -decay rates can be determined using masses of the participating nuclides, from the Q -values of the reaction (Q_β):

$$Q_{\beta-}(Z, N) = \left[M(Z, N) - M(Z + 1, N - 1)\right] c^2 \quad (1.12)$$

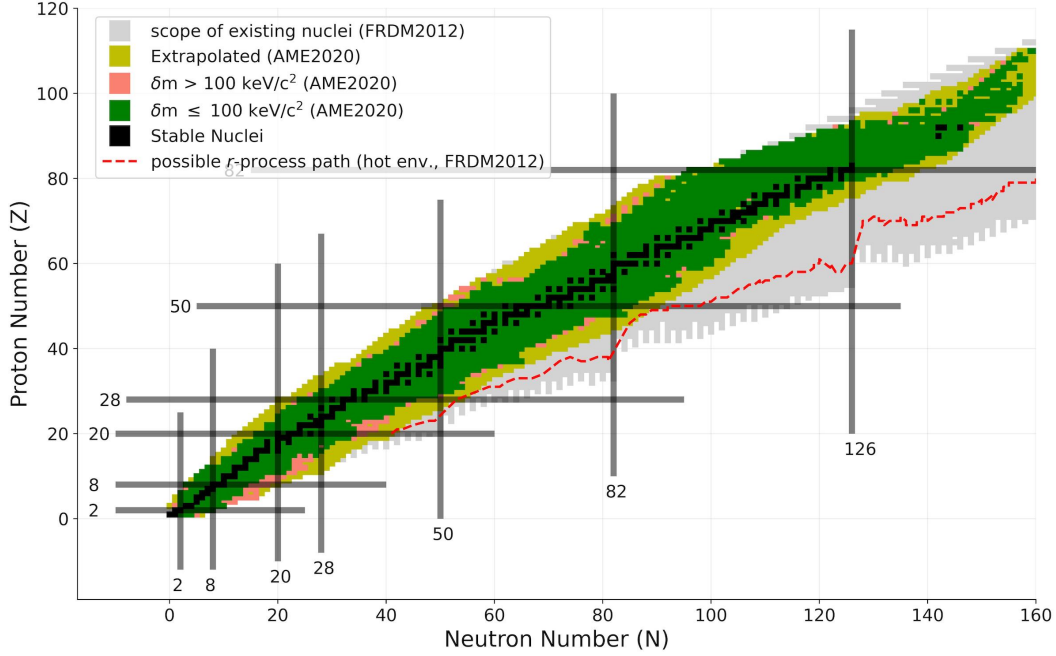
Additionally, masses are also essential in calculating the (n, γ) cross-sections [16]. Thus it is evident that the confidence of a simulation depends on the availability of precise mass data. Studies have shown that in order to constrain the r -process calculations and validate them by comparing with the solar abundance pattern, masses of the REP nuclides need to be known to a precision of ~ 100 keV/ c^2 [16].

Due to the challenges in producing the neutron-rich nuclides involved in the r process, the masses of a majority of them are not known to the required precision. This is demonstrated in Figure 1.5(a), where our knowledge of masses from the latest Atomic Mass Evaluation, AME2020 [30] is shown. The AME2020 provides a comprehensive summary of atomic masses of nuclides from measurements using different techniques, and extrapolations from the measured values. In Fig. 1.5(a), the nuclides whose masses are known within 100 keV/ c^2 are shown in dark green, while those whose mass uncertainties are > 100 keV/ c^2 or are extrapolated from the known masses are shown in salmon and light green. A hypothetical hot r -process path, calculated using the Finite Range Droplet Model (FRDM2012 [31]) mass model, is also shown as a red dashed line for reference. When mass data is not available, simulations rely on the different mass models. For example, the aforementioned FRDM2012, FRDM1995 [32], the Koura-Tachibana-Uno-Yamada (KTUY) [33], the Duflo-Zuker (DZ) [34], the Hartree-Fock-Bogoliubov (HFB-27) [35] and the Weizsäcker-Skyrme (WS4) [36] mass models are commonly used. The predictions from these models diverge considerably in regions where mass measurements are not available, as shown in Figures 1.5(b) and 1.5(c)

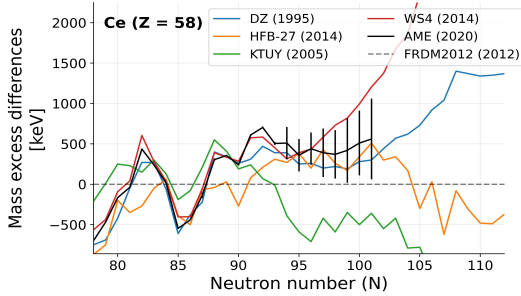
for different isotopes of the elements cerium and praseodymium. These deviations arise from the theoretical description of the nucleus, which uses a number of free parameters depending on the models. These parameters when fitted to measured masses allow the models to extrapolate better for unknown nuclides [37].

Current and upcoming rare isotope beam (RIB) facilities mainly use fragmentation (ISOLDE facility at CERN, Switzerland; the ISAC facility at TRIUMF, Canada; GSI and the future GSI-FAIR project in Germany; NSCL and the future FRIB at MSU, USA; the RIBF facility at RIKEN, Japan; and the RIBLL project in Lanzhou, China), fusion (the IGISOL facility at the University of Jyväskylä, Finland; and SPIRAL2 at GANIL, France; SHIP facility at GSI, Germany), spallation (ISOLDE facility at CERN, Switzerland; the ISAC facility at TRIUMF, Canada), nucleon induced fission (ISOLDE facility at CERN, Switzerland; the ISAC facility at TRIUMF, Canada), or spontaneous fission reactions (CARIBU at ANL, USA) to produce these nuclides [16]. Additionally, a new facility is currently under construction at the Argonne Tandem Linac Accelerator System (ATLAS) facility at ANL. This facility, named “ $N = 126$ factory” will populate isotopes closer to the $N=126$ shell closure through multi-nucleon transfer by deep inelastic scattering of a high energy stable ion beam (eg ^{136}Xe) from ATLAS on a heavy target (eg ^{195}Pt) [38, 39], and is scheduled to come on-line later in 2022. It should be mentioned here that all these facilities have different reaches and focus on populating different regions in the nuclear landscape, although some overlap could be found between different facilities.

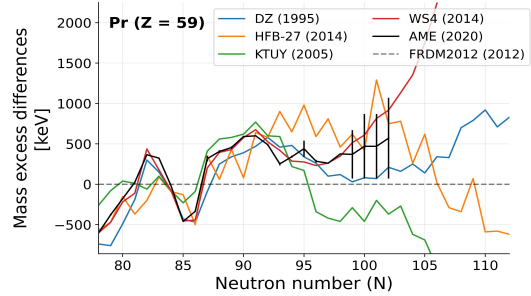
The main instruments used for mass measurements are Penning traps [37, 40], storage rings [37, 41], and Multi-Reflection Time-Of-Flight (MR-TOF) mass spectrometers [42, 43]. Penning traps have been involved in the majority of the most accurate and precise mass measurements in the latest Atomic Mass Evaluations AME2016 [44] and AME2020 [16, 30]. In a Penning trap, the atomic mass is calculated by measuring the cyclotron frequency of an ion trapped in a homogeneous magnetic field. Some of the Penning traps dedicated to measuring masses of r -process nuclides include the ISOLTRAP at ISOLDE (CERN), the CPT at CARIBU (ANL), the JYFLTRAP at IGISOL (University of Jyväskylä), and TITAN at ISAC (TRIUMF) [40]. The traditional Penning-trap-mass-measurement technique for short-lived nuclides is the time-of-flight ion-cyclotron-resonance (TOF-ICR) technique, where



(a) Uncertainties in masses reported in the AME2020 [30].



(b) Masses of isotopes of Ce ($Z = 58$), predicted and measured.



(c) Masses of isotopes of Pr ($Z = 59$), predicted and measured.

Figure 1.5 : Panel (a) shows the uncertainties in known masses from AME2020 [30]. The masses in dark green are known to within $100 \text{ keV}/c^2$, while the uncertainties in the salmon ones exceed $100 \text{ keV}/c^2$. The extrapolated masses are shown in light green. A scope of existing nuclides from FRDM2012 [31] is shown in grey, while a hypothetical r -process path calculated for a hot environment using the FRDM2012 model is depicted using a red dashed line. The stable nuclides are shown in black squares, while the magic neutron and proton numbers are shown in grey vertical and horizontal lines respectively. Panel (b) shows comparison between mass excess values from different mass models (namely, DZ [34], HFB-27 [35], KTUY [33], and WS-4 [36]) and the Atomic Mass Evaluation 2020 (AME2020) [30], all with respect to the FRDM2012 [31] for two elements Ce and Pr.

the ion's cyclotron frequency is determined by measuring its time-of-flight (TOF) from the trap to a detector resulting from the energy gained from a resonant excitation [40]. The TOF-ICR technique has been successfully used world-wide in Penning trap mass spectrometry for decades and relative mass precisions of $\sim 10^{-8}$ have been achieved for nuclides with $t_{1/2} \sim \text{s}$, and $\sim 10^{-7}$ for nuclides with shorter half-lives ($\sim 100\text{s}$ of ms). A relatively novel detection technique, phase-imaging ion-cyclotron-resonance (PI-ICR), first devised by the SHIPTRAP group [45], is now used with some of the traps, namely the CPT [46], ISOLTRAP [47], and JYFLTRAP [48]. In PI-ICR, the ion's cyclotron frequency is evaluated by projecting and measuring the orbital phase the ion accumulates over a period of time. Using PI-ICR, masses of nuclides with half-lives $\sim 100\text{s}$ of ms can be measured to relative precisions $\sim 10^{-8}$ with comparative ease. Storage rings rely on measuring masses of charged particles from the momentum of their revolution around a ring. Relative uncertainties of $\sim 10^{-6}$ can be achieved using storage rings [41], limiting their usefulness in mass measurements for r -process studies. MR-TOFs are a relatively newer addition to the list of instruments of mass measurements [42]. The principle of MR-TOF mass spectrometry is measuring the masses of nuclides from the time they take to travel a certain distance. This distance can be set by controlling the number of times the nuclides get reflected and travel back-and-forth within a tube of fixed length. Mass measurements of short-lived nuclides, with $t_{1/2} < 100 \text{ ms}$, can be done with MR-TOFs, while achieving relative precisions of $\sim 10^{-7}$ [43]. Since this thesis involves mass measurements conducted with a Penning trap using PI-ICR, details of PI-ICR along with its advantages over TOF-ICR will be discussed, while storage ring mass spectrometry will not be discussed any further. Additionally, an MR-TOF was used for solely beam purification purposes in this work, and not for mass measurements. Therefore details of the use of MR-TOF for mass separation are included here, instead of their use in mass measurements.

In the subsequent chapters, I will first give a general theoretical description of the instruments and experimental techniques used to trap ions and measure their masses (Chapter 2), before discussing CARIBU, including the MR-TOF (Chapter 3), the CPT experimental system (Chapter 4) and PI-ICR (Chapter 5) as the facility, instrument and technique of mass measurement used for this thesis in detail. Finally I will present the measured masses

in Chapter 6 and provide an outlook for the CPT project (Chapter 7).

2 The theory

Over the years, scientists have devised different ways of producing ions and studying them using time-of-flight spectroscopy, mass spectroscopy and other techniques. Then in the 1980s, W. Paul and H.G. Dehmelt developed two new techniques of trapping ions using dynamic and static fields respectively [49], for which they shared one half of the 1989 Nobel prize in physics [50, 51]. This opened new horizons in ion-research. By thermally isolating the ions, all types of extraneous perturbations could be avoided enabling reaching unprecedented levels in precision of measurements. Currently, the Paul and the Penning traps are the most commonly used ion traps in different branches of atomic, nuclear and particle physics [52]. In the context of this thesis, the traps used are linear Paul traps and a Penning trap, which I will discuss in the subsequent sections.

2.1 Radio frequency quadrupole (RFQ) or Paul trap

Trapping an ion requires a restoring force which can bring the ion back to its original position in the event that it moves away from that space. In the simplest case of one dimension (1D), a restoring force of the form $F_x = -k_x(x - x_0)$ serves the purpose, where k_x is the force constant, and $(x - x_0)$ is the ion's displacement from some initial position x_0 . Such a force on an ion can be applied by an electric field E_x :

$$E_x = \frac{F_x}{q} = -\frac{k_x}{q}(x - x_0) \quad (2.1)$$

where q is the charge of the ion. Now, the electric field can easily be created by a potential (Φ), which from $\vec{E} = -\vec{\nabla}\Phi$, can be found as:

$$\Phi = \Phi_x = \lambda_x(x - x_0)^2 + \varphi_0, \quad (2.2)$$

with $\lambda_x = \frac{k_x}{2q}$ and φ_0 being an arbitrary constant potential at the position x_0 . To simplify matters, I will set x_0 and φ_0 to zero in the subsequent derivations without loss of any generality. Physically, this only means that the ion is being trapped around the origin in the x -dimension where the electric potential is zero. Therefore, equation 2.2 becomes:

$$\Phi = \Phi_x = \lambda_x x^2. \quad (2.3)$$

Similarly, confinement in two dimensions (2D) can be achieved by a potential:

$$\Phi = \Phi_x + \Phi_y = \lambda_x x^2 + \lambda_y y^2. \quad (2.4)$$

Being in a source-free region, the potential must satisfy Laplace's equation:

$$\nabla^2 \Phi = 0. \quad (2.5)$$

From equation 2.4 and 2.5, we get:

$$\lambda_x + \lambda_y = 0. \quad (2.6)$$

Not considering the trivial solution $\lambda_x = \lambda_y = 0$ as that will not enable confinement, we get:

$$\lambda_x = -\lambda_y = \lambda. \quad (2.7)$$

Using 2.7, equation 2.4 becomes:

$$\Phi = \lambda (x^2 - y^2). \quad (2.8)$$

One way of creating such a potential is by having two pairs of equipotential surfaces given by:

$$\frac{x^2}{r_0^2} - \frac{y^2}{r_0^2} = 1, \quad \text{at } \Phi = \frac{\Phi_0}{2} \quad \text{and} \quad \frac{x^2}{r_0^2} - \frac{y^2}{r_0^2} = -1, \quad \text{at } \Phi = -\frac{\Phi_0}{2}. \quad (2.9)$$

Equation 2.9 represents a pair of hyperbolic electrodes, centered at $(0,0)$, with vertices at $(\pm r_0, 0)$ and $(0, \pm r_0)$, and having respective electric potential of $\pm \Phi_0/2$. This constitutes a “quadrupole” and is shown in Figure 2.1. This would make the λ of equation 2.8, $\lambda = \Phi_0/(2r_0^2)$, and equation 2.8 would become:

$$\Phi = \frac{\Phi_0}{2r_0^2} (x^2 - y^2). \quad (2.10)$$

With such an arrangement, the force on a trapped ion would be:

$$\begin{aligned} \vec{F} &= q \vec{E} \\ &= -q \vec{\nabla} \Phi \\ \text{or, } m(\ddot{x}\hat{x} + \ddot{y}\hat{y}) &= -\frac{q\Phi_0}{2r_0^2} (2x\hat{x} - 2y\hat{y}), \end{aligned} \quad (2.11)$$

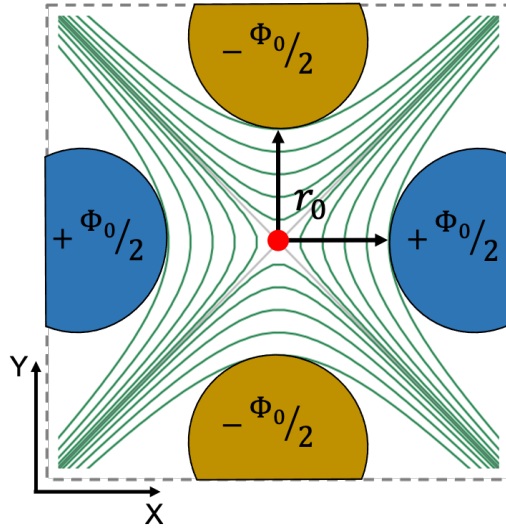


Figure 2.1 : Two dimensional quadrupole electrodes from equation 2.9, producing a potential of the form of equation 2.8. The electrodes of same color (blue or gold) have the same potentials ($\frac{\Phi_0}{2}$ and $-\frac{\Phi_0}{2}$ respectively) creating equipotential lines (shown in green). The red dot in the middle represents the center of the trapping volume.

where $\hat{x}$ and $\hat{y}$ are unit vectors along the x and y directions, and $\ddot{x}$ and $\ddot{y}$, the accelerations in the x and y directions due to the force $\vec{F}$. From equation 2.11, the motion of the trapped ion can be deduced as:

$$F_x = m\ddot{x} = -\frac{q\Phi_0}{r_0^2}x \quad (2.12a)$$

$$F_y = m\ddot{y} = \frac{q\Phi_0}{r_0^2}y \quad (2.12b)$$

It becomes clear immediately that such an ion will be trapped along the x -direction, undergoing a simple harmonic motion with an angular frequency $\omega_x = \sqrt{\frac{q\Phi_0}{mr_0^2}}$, but free to drift away along the y -direction. Such a situation is called a saddle point, as shown in Figure 2.2.

Therefore, to achieve confinement along both the directions, the solution is to use a periodic potential, in the radio-frequency (RF) range, in the following form:

$$\Phi_0 = V_{dc} + V_{rf} \cos(\omega_{rf}t) \quad (2.13)$$

where V_{dc} is a direct current (DC) offset, V_{rf} and ω_{rf} are the amplitude and frequency of the applied RF. This confines the ion along one direction for half a period of excitation

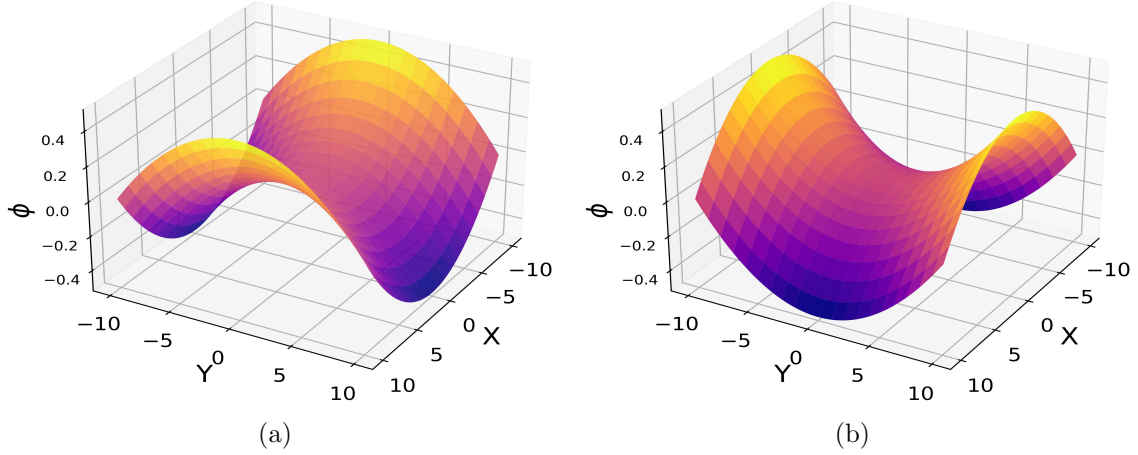


Figure 2.2 : Saddle point potential in two dimensions. In (a), the ions are confined along x but can move away along y . The opposite takes place in (b)

($\tau_{rf} = \frac{2\pi}{\omega_{rf}}$) and the other direction for the other half. The combination of RF excitations on quadrupole electrodes gives it the name RFQ. Using the above form of Φ_0 from 2.13 in equations 2.12, the equations of motion become:

$$\ddot{x} + \frac{q}{mr_0^2} \left[V_{dc} + V_{rf} \cos(\omega_{rf}t) \right] x = 0 \quad (2.14a)$$

$$\ddot{y} - \frac{q}{mr_0^2} \left[V_{dc} + V_{rf} \cos(\omega_{rf}t) \right] y = 0 \quad (2.14b)$$

Now by defining the following dimensionless parameters:

$$a = \frac{4qV_{dc}}{m\omega_{rf}^2 r_0^2}, \quad Q = \frac{2qV_{rf}}{m\omega_{rf}^2 r_0^2} \text{ and } \epsilon = \frac{\omega_{rf} t}{2}, \quad (2.15)$$

equations 2.14 can be re-written in the form of Mathieu's equations:

$$\frac{d^2x}{d\epsilon^2} + \left[a + 2Q \cos(2\epsilon) \right] x = 0 \quad (2.16a)$$

$$\frac{d^2y}{d\epsilon^2} - \left[a + 2Q \cos(2\epsilon) \right] y = 0 \quad (2.16b)$$

Stable solutions can be obtained for specific values of a and q . This is illustrated in Figure 2.3. That means, to successfully confine a charged particle with a certain q/m ratio simultaneously in both x and y dimensions at a fixed r_0 , the values of V_{dc} , V_{rf} and ω_{rf} must

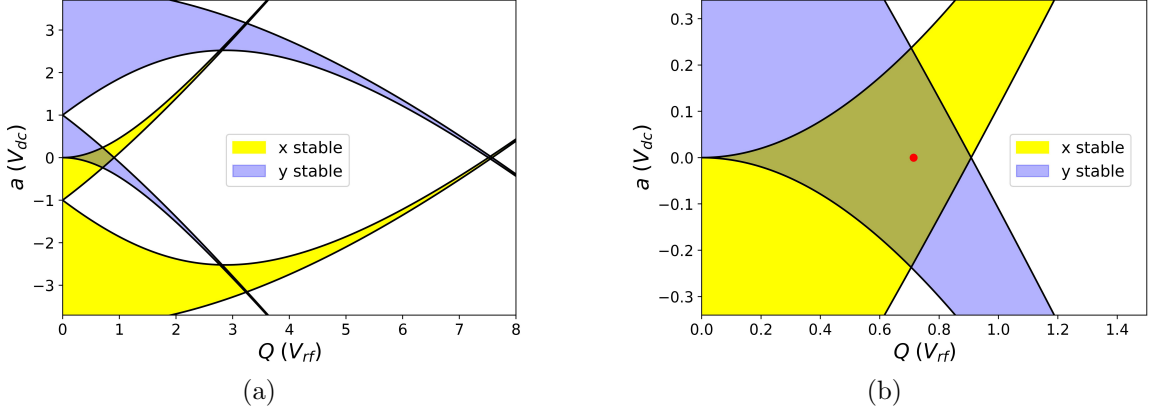


Figure 2.3 : Solutions to Mathieu's equation in an $a - Q$ parameter space. (b) is a zoomed-in look into one region of overlapping stability in (a). The yellow and blue bands signify regions of x and y stability respectively. The red dot in (b) is a typical parameter set that can be for $^{133}\text{Cs}^+$ ions in a linear Paul trap, details in text.

be set such that the a and Q parameters lie within one of the overlapping regions in Figure 2.3. For example, in a linear Paul trap with $r_0 = 0.5$ mm, to trap a $^{133}\text{Cs}^+$ ion in two dimensions, a typical set of values could be $V_{dc} = 0$ V, $V_{rf} = 300$ V_{pp} and $\omega_{rf}/2\pi = 800$ kHz, shown with the red spot in Figure 2.3(b). Once tuned to one ion species of charge to mass ratio of q_0/m_0 , the frequency of the RF can be scaled to switch to another species of charge to mass ratio of q_1/m_1 without changing other settings, as [53]:

$$\frac{\omega_{rf1}}{\omega_{rf0}} = \sqrt{\frac{q_1/m_1}{q_0/m_0}}. \quad (2.17)$$

Extending the potential in equation 2.4 to three dimensions (3D), we can write:

$$\Phi = \Phi_x + \Phi_y + \Phi_z = \lambda_x x^2 + \lambda_y y^2 + \lambda_z z^2. \quad (2.18)$$

As before, applying Laplace's equation ($\nabla^2 \Phi = 0$), we obtain:

$$\lambda_x + \lambda_y + \lambda_z = 0. \quad (2.19)$$

Again, not considering the trivial solution $\lambda_x = \lambda_y = \lambda_z = 0$, we get:

$$\lambda_x = \lambda_y = -\frac{\lambda_z}{2} = -\lambda. \quad (2.20)$$

Using 2.20, equation 2.18 becomes:

$$\Phi = \lambda (2z^2 - x^2 - y^2). \quad (2.21)$$

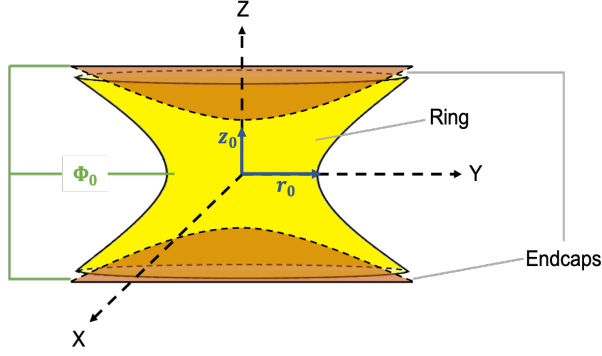


Figure 2.4 : Ring and endcap electrodes enabling 3D confinement in a Paul trap

Converting 2.21 to cylindrical coordinates, this becomes:

$$\Phi = \lambda (2z^2 - r^2), \quad (2.22)$$

where $r^2 = x^2 + y^2$. To create such a potential, let us consider a system of a 2-sheet hyperboloidal endcap electrodes and an intermediate 1-sheet hyperboloid ring electrode, as described below [49]:

$$\text{Ring: } \frac{r^2}{r_0^2} - \frac{2z^2}{r_0^2} = 1 \quad (2.23a)$$

$$\text{Endcaps: } \frac{r^2}{z_0^2} - \frac{2z^2}{z_0^2} = -1 \quad (2.23b)$$

where, r_0 is the radius at the central bore of the ring electrode while $2z_0$ is the distance between the vertices of the endcaps, as shown in Figure 2.4. Inside such a symmetric electrode arrangement, the potential minimum across both z and r has to be at the origin to trap an ion stably across that dimension. Therefore, if we use a dimensional constraint $r_0/z_0 = \sqrt{2}$, we can rewrite equation 2.22 as:

$$\Phi = -\frac{\Phi_0}{r_0^2 + 2z_0^2} (2z^2 - r^2). \quad (2.24)$$

Then, the potentials at $(r, z) = (\pm r_0, 0)$ and $(r, z) = (0, \pm z_0)$ would be $\Phi = \frac{\Phi_0}{2}$ and $\Phi = -\frac{\Phi_0}{2}$ respectively, creating a saddle point at the origin [49]. Then, similar to the 2D scenerio, using a dynamic periodic potential for Φ_0 (similar to equation 2.13), an ion can be trapped in such a trap in all three dimensions. This system of hyperboloidal ring-endcap electrodes is the original design of a Paul trap [49].

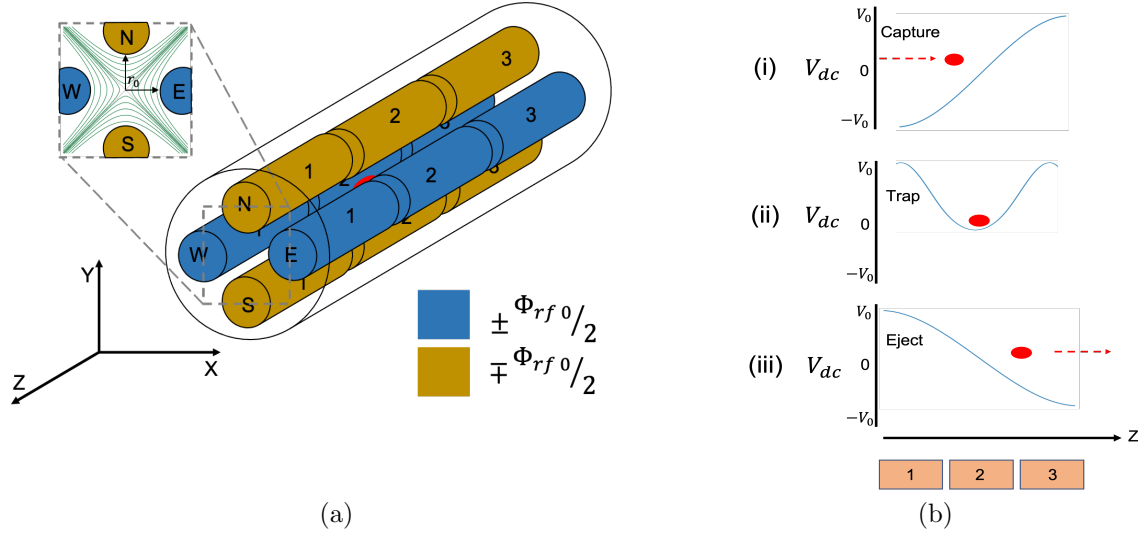


Figure 2.5 : Schematic diagram of a linear Paul trap. (a) represents a system of four cylindrical segmented rod-electrodes. The same colored electrodes (blue or gold) signify electrodes having the same phase, creating the equipotential lines (shown in green on the inset) responsible for $x - y$ confinement. (b) depicts three such V_{dc} settings enabling capture (panel b(i)), trapping (panel b(ii)) and ejection (panel b(iii)) of the ion applied across three segments (shown in different shades of orange). The red ellipse in both the pictures (partially visible in (a)) symbolizes the ion cloud.

The Paul traps used in the work presented in this thesis are slightly different to the one just described (Figure 2.4). The arrangement of hyperbolic electrodes, as shown in Figure 2.1 to achieve 2D confinement, can be part of a set of four cylindrical rod-electrodes, labelled North (N), East (E), West (W) and South (S). To ensure confinement in all three dimensions, these cylindrical electrodes can be segmented into three sections (labelled 1, 2, and 3). This is shown in Figure 2.5(a). The same RF potential (Φ_{rf0}) is applied on the same-colored electrode (gold and blue) creating the equipotential lines (shown in green), allowing confinement in the $x - y$ plane, shown in the inset.

Different values of V_{dc} (in equation 2.13) can be applied to the three sections in the form of a potential well with a minimum at the central segment for trapping the ion along the z -direction or a potential gradient to enable capture or ejection of the ions. This is shown in Figure 2.5(b), with the three segmented electrodes along the z -direction shown in orange. On the top (panel b(i)), V_{dc} is set across the three electrodes in a way that it creates a potential gradient to enable capture of the ions. In the middle (panel b(ii)), electrode 1 potential is raised to create the potential well and enable trapping. In the bottom (panel

b(iii)), electrode 3 potential is lowered to eject the ions. The red ellipse in both the pictures in Figure 2.5 represents the ion cloud. Such a trap is called a linear Paul trap.

Some of the most common uses of such a linear arrangement of RFQs include construction of ion guides, coolers and bunchers. They differ on the basis of the DC voltages applied to them, whether they are of the form of a potential gradient to facilitate ion transport or a potential well to trap them. In the presence of buffer gas, an ion travelling through or trapped in an RFQ will undergo multiple collisions with the gas atoms and lose energy, thereby getting cooled. This is known as buffer gas cooling. With full valence shells making them the least reactive, noble gases like He are the best choice of gases for such type of cooling. For the work presented in this thesis, linear RFQs are used in the RFQ buncher (Section 3.3) in the CARIBU facility, to convert a DC beam into a bunched beam and a linear Paul trap (Section 4.3) in the CPT system to cool the ions and prepare them for measurement in the CPT (Section 4.4).

2.2 Penning trap

Hans G. Dehmelt first came up with the idea of a Penning trap from a Penning ion gauge, where the pressure in a discharge tube is measured as proportional to the current generated by electrons in a magnetic field [54, 55], to which Albert Einstein was quoted saying “You know, it would be sufficient to really understand the electron” [56]. It was a revolutionary device which opened new avenues of physics research.

The basic idea of a Penning trap is fairly simple. A charged particle moving through a magnetic field $\vec{B}$ with a velocity $\vec{v}$ experiences Lorentz force, given by $\vec{F} = q \vec{v} \times \vec{B}$, causing it to undergo cyclotron motion with frequency:

$$\omega_c = \frac{q}{m} B_0 \quad (2.25)$$

where q and m are the charge and mass of the particle, while B_0 is the magnitude of the magnetic field. In principle, measuring this frequency allows one to measure the mass of the ion very precisely. If $\vec{B} = B_0 \hat{z}$, this cyclotron motion essentially restricts the particle in the $x-y$ plane. Simultaneous confinement in all three dimensions can be achieved by applying a harmonic electric potential well along this axial or z direction, in the form of equation 2.22.

To characterize λ here, let us define a potential difference V_0 such that:

$$V_0 = \Phi(r = 0, z = z_0) - \Phi(r = r_0, z = 0) = \lambda(2z_0^2 + r_0^2). \quad (2.26)$$

This makes:

$$\lambda = \frac{V_0}{2z_0^2 + r_0^2}. \quad (2.27)$$

Now, defining a parameter d_0 , commonly known as the characteristic trap dimension, as:

$$d_0^2 = \frac{1}{2} \left(z_0^2 + \frac{r_0^2}{2} \right), \quad (2.28)$$

we can rewrite equations 2.22 as:

$$\Phi = \frac{V_0}{2d_0^2} \left(z^2 - \frac{x^2 + y^2}{2} \right). \quad (2.29)$$

Such a potential can be created by a hyperbolic set of electrodes [57]:

$$\text{Ring: } \frac{r^2}{r_0^2} - \frac{2z^2}{r_0^2} = 1 \quad (2.30a)$$

$$\text{Endcaps: } \frac{r^2}{2z_0^2} - \frac{z^2}{z_0^2} = -1 \quad (2.30b)$$

similar to Figure 2.4, without the dimensional constraint of $r_0/z_0 = \sqrt{2}$. Since V_0 , being the potential difference between the trapping electrodes (endcaps and the ring), represents the depth of the potential well, it is known as the trap depth. In other words, with the presence of $\vec{B}$, the periodic potential Φ_0 as in the case of the Paul trap, can be replaced with a fixed DC potential difference between the endcap and the ring electrodes (V_0), also known as the trap depth, allowing confinement across all three dimensions. Such an arrangement of fields is shown in Figure 2.6. For precision measurements, we need to first understand an ion's motion inside such a combination of $\vec{B}$ and $\vec{E}$ fields.

With the electric potential well, the Lorentz force becomes:

$$\vec{F} = q (\vec{E} + \vec{v} \times \vec{B}) , \quad (2.31)$$

where, $\vec{E} = -\vec{\nabla}\Phi$. We can, hence, deduce the equations of motion as follows:

$$\begin{aligned} \vec{F} &= q (-\vec{\nabla}\Phi + \vec{v} \times \vec{B}) \\ \text{or, } m(\ddot{x}\hat{x} + \ddot{y}\hat{y} + \ddot{z}\hat{z}) &= -\frac{qV_0}{2d_0^2} (2z\hat{z} - x\hat{x} - y\hat{y}) + qB_0 (\dot{y}\hat{x} - \dot{x}\hat{y}), \end{aligned} \quad (2.32)$$

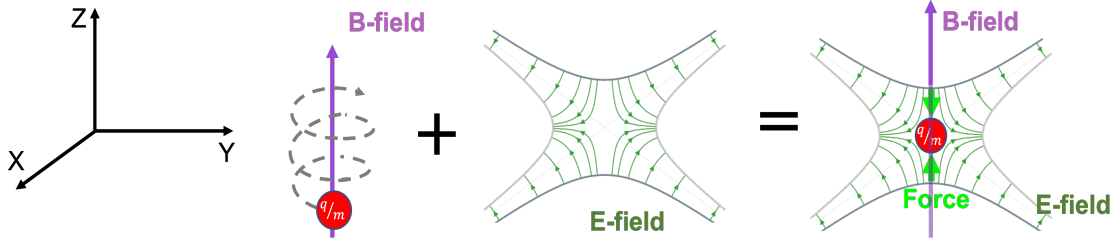


Figure 2.6 : Penning trap: magnetic and electric fields required for ion confinement.

Separating the three simultaneous differential solutions in equation 2.32:

$$m\ddot{x} = \frac{qV_0}{2d_0^2} x + qB_0 \dot{y} \quad (2.33a)$$

$$m\ddot{y} = \frac{qV_0}{2d_0^2} y - qB_0 \dot{x} \quad (2.33b)$$

$$m\ddot{z} = -\frac{qV_0}{d_0^2} z \quad (2.33c)$$

Along the z -direction (equation 2.33c), the motion is independent of the other two as it is dependent solely on electric potential V_0 . As a result, the particle undergoes a simple harmonic motion with frequency (known as the axial frequency in this context) given as:

$$\omega_z = \sqrt{\frac{qV_0}{md_0^2}} \quad (2.34)$$

Rewriting equations 2.33a and 2.33b:

$$\ddot{x} - \omega_c \dot{y} - \frac{\omega_z^2}{2} x = 0 \quad (2.35a)$$

$$\ddot{y} + \omega_c \dot{x} - \frac{\omega_z^2}{2} y = 0 \quad (2.35b)$$

One way to approach equations 2.35 is by defining a complex variable:

$$u = x + iy, \quad (2.36)$$

thereby reducing it to a single equation:

$$\ddot{u} + i\omega_c \dot{u} - \frac{\omega_z^2}{2} u = 0. \quad (2.37)$$

From this, using the ansatz:

$$u = e^{-i\omega t}, \quad (2.38)$$

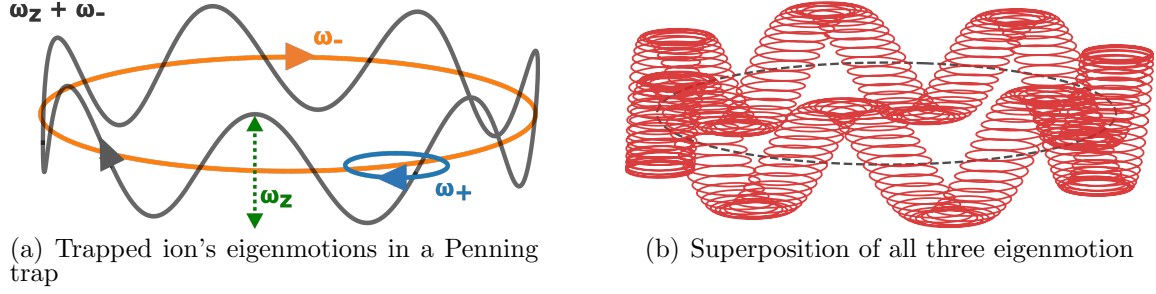


Figure 2.7 : Motion of a trapped ion in a Penning trap. Figure courtesy: R. Orford.

an equation for the frequency of the radial motions can be obtained as:

$$\omega^2 - \omega_c \omega + \frac{\omega_z^2}{2} = 0. \quad (2.39)$$

This equation has two solutions, which give us the two radial eigenfrequencies:

$$\omega_{\pm} = \frac{1}{2} \left(\omega_c \pm \sqrt{\omega_c^2 - 2\omega_z^2} \right) \quad (2.40)$$

which are known as the reduced cyclotron (ω_+) and magnetron (ω_-) frequencies. As a result, a trapped particle's motion in a Penning trap is a superposition of all three eigenmotions (ω_+ , ω_- and ω_z), shown in Figure 2.7

From equation 2.40, one thing becomes clear immediately that in order to have a stable trapping condition:

$$\omega_c^2 > 2\omega_z^2. \quad (2.41)$$

Following up on equation 2.41, from the expressions for ω_c (2.25), ω_z (2.34) and d_0 (2.28), we get:

$$B_0 > \sqrt{\frac{2mV_0}{q d_0^2}}. \quad (2.42)$$

With the characteristic trap dimension of the CPT, $d_0 = 0.9145$ cm (from $r_0 = 1.16$ cm and $z_0 = 1.0$ cm) and a typical trap depth $V_0 \approx 10$ V, to trap an ion of the typical tuning beam of $^{133}\text{Cs}^+$ (mass number $A = 133$ and $q = 1e^-$), the smallest magnetic field required is ~ 0.57 T. For the masses presented in this thesis, the isotopes are mostly around $A = 160$ and $q = 2e^-$, and the minimum B required to trap them is ~ 0.44 T. The field of the CPT magnet is 5.8 T, which is well above the requirement.

As I have mentioned, to measure the mass of an ion, one needs to measure the ω_c . But since ω_c is not an eigenfrequency of an ion inside the trap, its determination relies on

its relationships with the eigenfrequencies. Equations 2.34 and 2.40 point us towards the following frequency relations:

$$\omega_c = \omega_+ + \omega_- \quad \text{and} \quad (2.43a)$$

$$\omega_c^2 = \omega_+^2 + \omega_-^2 + \omega_z^2. \quad (2.43b)$$

Equation 2.43a allows determination of the cyclotron frequency from measuring the two radial eigenfrequencies. This is the base of the mass measurement technique implemented at the CPT for the work presented in this thesis and will be discussed in further detail in Chapter 5.

All the discussion till now has been done assuming a perfectly harmonic potential well. In a real Penning trap, practicalities dictate certain moderations to the trap electrodes, like having finite sized electrodes, presence of apertures to facilitate the entry and exit of the ions into and from the trap, and partitioning the ring electrode to enable application of RF excitations (described in the next Section 2.3). These lead to anharmonicities in the trapping potential, which is usually improved by adding correction electrodes. Equation 2.43b, known as the Gabrielse-Brown invariance theorem [58] also helps in that regard. It is independent of the field misalignments and potential anharmonicities and its sum-of-squares form enables cancellation of the first order frequency shifts [58]. It is therefore used to determine ω_c from the radial and axial eigenfrequencies when the mass precision is required to be around 10^{-9} . Details of higher order anharmonicities and required corrections can be found in reference [58] and will not be discussed here. In general, the frequencies follow the hierarchy:

$$\omega_- \ll \omega_z \ll \omega_+ \approx \omega_c \quad (2.44)$$

At this point, let us look into ω_- in a little more detail. Rewriting 2.40, we get:

$$\omega_- = \frac{\omega_c}{2} \left(1 - \sqrt{1 - 2\frac{\omega_z^2}{\omega_c^2}} \right). \quad (2.45)$$

Using the CPT values ($d_0 = 0.9145$ cm , $V_0 \approx 10$ V, and $B_0 = 5.8$ T), for both species having A/q around 160 / 2+ or 133 / 1+, we get:

$$\frac{2\omega_z^2}{\omega_c^2} = \frac{2\frac{qV_0}{md_0^2}}{\frac{q}{m}B_0} \approx 0.01 \quad (2.46)$$

While equation 2.46 reaffirms the trapping capability at the CPT (from 2.41 and 2.42), it helps us simplify the the expression within the square-roots in 2.45. Expanding the said expression as a binomial series:

$$\begin{aligned} \left(1 - 2\frac{\omega_z^2}{\omega_c^2}\right)^{1/2} &= 1 - \frac{\omega_z^2}{\omega_c^2} - \frac{1}{2} \frac{\omega_z^4}{\omega_c^4} + \dots \\ &= 1 - \frac{\omega_z^2}{\omega_c^2} \text{ (ignoring the higher order terms since, } \frac{2\omega_z^2}{\omega_c^2} \ll 1) \end{aligned} \quad (2.47)$$

Therefore, from 2.45 and 2.47:

$$\omega_- = \frac{\omega_c}{2} - \frac{\omega_c}{2} \left(1 - \frac{\omega_z^2}{\omega_c^2}\right) = \frac{\omega_z^2}{2\omega_c} = \frac{V_0}{2B_0d_0^2} \quad (2.48)$$

This shows that ω_- is only weakly mass dependant and depends largely on the field, the characteristic trap dimension and the trap depth. So once a Penning trap magnet is energized to a certain field, the ω_- stays more-or-less constant for a fixed V_0 , subject to the temporal instabilities of the B -field and the DC power supplies used for the trap electrodes.

2.3 Manipulating the ions motions inside a Penning trap

Measuring the motional frequencies of an ion trapped inside a Penning trap requires one to manipulate the ion's motion in different ways, by applying different types of excitations. For this purpose, the ring electrode is divided into four quadrants, and the excitations are applied as RF electric pulses with differing phases on those quadrants, as shown in Figure 2.8.

2.3.1 Dipole excitation

A time-varying radial potential with amplitude, frequency and phase of V_D , ω_D and ϕ_D respectively given by:

$$\Phi_D = V_D \cos(\omega_D t + \phi_D) \quad (2.49)$$

applied to the ring electrodes such that the opposite quadrants are 180° out-of-phase, provides a dipole field:

$$\vec{E}_D = -\vec{\nabla}\Phi_D = -V_D \cos(\omega_D t + \phi_D) \hat{x} \quad (2.50)$$

During the application of such an excitation, this field (2.50) modifies the ion's equation of motions (2.37):

$$\ddot{u} + i\omega_c \dot{u} - \frac{\omega_z^2}{2} u = -\frac{qV_D}{m} \cos(\omega_D t + \phi_D). \quad (2.51)$$

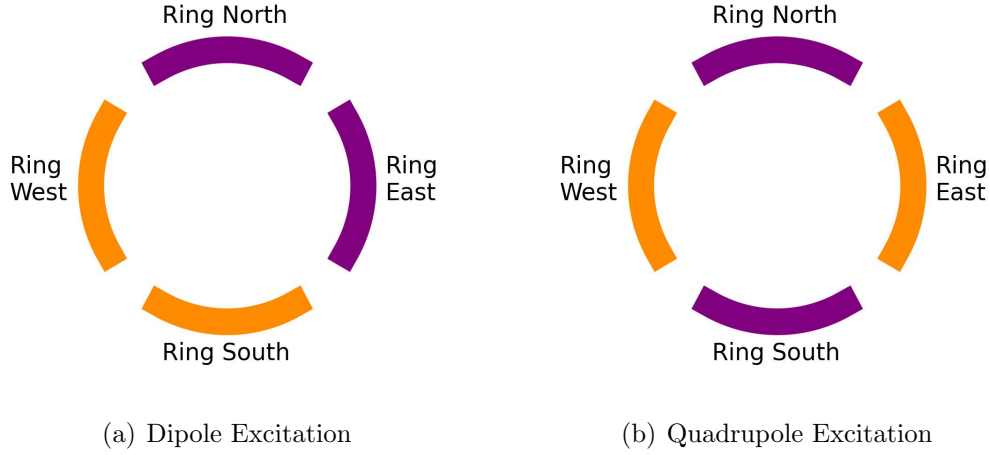


Figure 2.8 : A schematic view of the ring quadrants during dipole and quadrupole excitations. The violet and orange colors represent 180° out-of-phase excitations.

If $\omega_D = \omega_+$ or ω_- , the subjected ions will absorb energy resonantly. Therefore, proceeding with a similar ansatz (2.38) as before [53]:

$$u = r_+(t) e^{+i(\omega_+ t + \phi_+)} + r_-(t) e^{-i(\omega_- t + \phi_-)} , \quad (2.52)$$

where $r_\pm(t)$ are the ion's $\omega_\pm$ radii and $\phi_\pm$ are the ion's initial $\omega_\pm$ phases, the ion's locus can be described by [53]:

$$\begin{aligned} x(t) &= r_+(t) \cos(\omega_+ t + \phi_+) + r_-(t) \cos(\omega_- t + \phi_-) \\ y(t) &= r_+(t) \sin(\omega_+ t + \phi_+) - r_-(t) \sin(\omega_- t + \phi_-) \end{aligned} \quad (2.53)$$

Using a velocity vector method, an expression for the time-dependent radii $r_\pm(t)$ can be found via Laplace transformation, as [59]:

$$r_\pm(t) = \frac{1}{\omega_+ - \omega_-} \sqrt{A_\pm^2(0) + \frac{k_D^2}{4} t^2 \mp A_\pm(0) k_D t \sin(\Delta\phi_{D\pm})} \quad (2.54)$$

where:

$$A_\pm(0) = (\omega_+ - \omega_-) r_\pm(0) \quad (2.55a)$$

$$k_D = qV_D/m \quad (2.55b)$$

$$\Delta\phi_{D\pm} = \phi_D - \phi_\pm , \quad (2.55c)$$

$r_\pm(0)$ being the ion's initial $\omega_\pm$ radii. Upon careful examination, the following phase dependence can be immediately deduced [59].

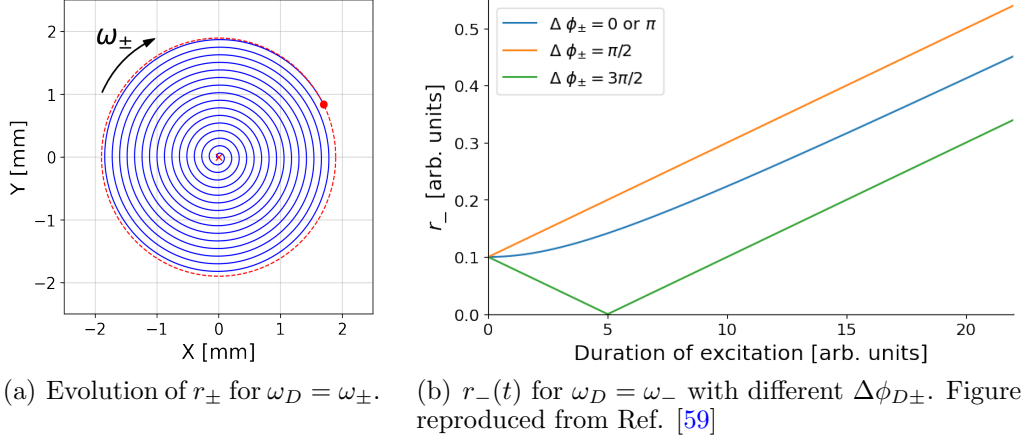


Figure 2.9 : In (a) an ion's path during dipole excitation, leading to an increase of its $r_{\pm}(t)$ starting from $r_{\pm}(0) = 0$ for $\omega_D = \omega_{\pm}$ respectively is shown. The red cross at the center signifies the center of the trap, the red dot symbolizes the ion while the blue line spiralling outwards is the ion's path. In (b), an ion's $r_{-}(t)$ for $\omega_D = \omega_{-}$ is computed and shown for $\Delta\phi_{D\pm} = 0$, or π (blue), $\pi/2$ (orange) and $3\pi/2$ (green).

1. $\Delta\phi_{D\pm} = 0$ or π : In this case, the sine-term in equation 2.54 vanishes resulting in $r_{\pm}(t)$ increasing first slowly and then almost linearly with the duration of the excitation.
2. $\Delta\phi_{D\pm} = \pi/2$: Here, the sine-term becomes $+1$, which makes:

$$r_{\pm}(t) = \left| r_{\pm}(0) \mp \frac{k_D t}{2(\omega_{+} - \omega_{-})} \right|. \quad (2.56)$$

Thus $r_{+}(t)$ first diminishes to zero and then increases linearly with time, while $r_{-}(t)$ keeps increasing linearly with time from the beginning.

3. $\Delta\phi_{D\pm} = 3\pi/2$: This time, the sine-term becomes -1 , leading to:

$$r_{\pm}(t) = \left| r_{\pm}(0) \pm \frac{k_D t}{2(\omega_{+} - \omega_{-})} \right|. \quad (2.57)$$

Therefore in this case $r_{+}(t)$ keeps increasing linearly with time from the beginning, but $r_{-}(t)$ first diminishes to zero and then linearly increases with time.

Since ω_{+} is a mass-dependent eigenmotion, the case of $\omega_D = \omega_{+}$ with $\Delta\phi_{+} = 0$, $\pi/2$ or π is a common technique used to increase the radius of a selected ion species for a mass-selective cleaning in case of time-of-flight ion-cyclotron-resonance (TOF-ICR) or to excite and park the ions in some orbit for phase-imaging ion-cyclotron-resonance (PI-ICR). A dipole excitation with $\omega_D = \omega_{-}$ is used in TOF-ICR to set a non-zero ω_{-} radius for all trapped

species independent of their masses. The special case of $\omega_D = \omega_-$ with $\Delta\phi_- = 3\pi/2$ is used to diminish the ions' initial magnetron radius before applying the necessary PI-ICR RF excitations. These are shown in Figure 2.9. These will be discussed in Section 2.4.

2.3.2 Quadrupole excitation

An RF electric potential, with amplitude, frequency and phase of V_Q , ω_Q and ϕ_Q , of the form:

$$\Phi_Q = -V_Q \cos(\omega_Q t + \phi_Q) xy \quad (2.58)$$

applied such that the adjacent ring electrodes will have a 180° phase difference will create an electric field:

$$\vec{E}_Q = -\vec{\nabla} \Phi_Q = V_Q \cos(\omega_Q t + \phi_Q) (y\hat{x} + x\hat{y}) \quad (2.59)$$

providing a quadrupole excitation. Detailed derivations relating to quadrupole excitation can be found in Ref [60, 61]. Using the same ansatz as 2.52, the ion's equations of motion would have a similar form as 2.53. But in this case, the time-dependent radii ($r_\pm(t)$) will be given by [53, 60]:

$$r_\pm(t) = \left[r_\pm(0) \cos(\omega_B t) \mp \frac{1}{2} \frac{i(\omega_Q - \omega_c) r_\pm(0) + k_Q^\pm r_\mp(0)}{\omega_B} \sin(\omega_B t) \right] e^{i(\omega_Q - \omega_c) t/2} \quad (2.60)$$

with ω_B and $k_Q^\pm$ defined as:

$$\begin{aligned} \omega_B &= \frac{1}{2} \sqrt{(\omega_Q - \omega_c)^2 + k_0^2} \quad \text{and} \quad k_Q^\pm = k_0 e^{\pm i \Delta\phi_Q}, \quad \text{with} \\ k_0 &= \frac{qV_Q}{m(\omega_+ - \omega_-)} \quad \text{and} \quad \Delta\phi_Q = \phi_Q - (\phi_+ + \phi_-). \end{aligned} \quad (2.61)$$

where $r_\pm(0)$ and $\phi_\pm$ are the ion's initial $\omega_\pm$ radii and phase. Now, if $\omega_Q = \omega_c$, 2.60 reduces to:

$$r_\pm(t) = \left| r_\pm(0) \cos\left(\frac{qV_Q}{2m(\omega_+ - \omega_-)} t\right) \mp r_\mp(0) \sin\left(\frac{qV_Q}{2m(\omega_+ - \omega_-)} t\right) e^{\pm i \Delta\phi_Q} \right| \quad (2.62)$$

In the special case where the duration of the excitation is:

$$t = t_{\text{conv}} = \frac{\pi}{k_0} = \pi \frac{m}{q} \frac{\omega_+ - \omega_-}{V_Q} \approx \frac{\pi B_0}{V_Q} \quad (\because \omega_- \ll \omega_+ \approx \omega_c) \quad (2.63)$$

will lead to the cosine term in equation 2.60 vanishing and the sine-term reducing to unity, thereby coupling the initial radius of one motion to the final radius of the other, as shown below:

$$r_{\pm}(t_{\text{conv}}) = r_{\mp}(0) e^{\pm i \Delta\phi_Q} \quad (2.64)$$

Rewriting the ansatz in equation 2.52 [53]:

$$\begin{aligned} u &= r_+(t) e^{+i(\omega_+ t + \phi_+)} + r_-(t) e^{-i(\omega_- t + \phi_-)} \\ &= R_+(t) + R_-(t) \end{aligned} \quad (2.65)$$

where $R_{\pm}(t) = r_{\pm}(t) e^{\pm i \phi_{\pm}(t)}$, with $\phi_{\pm}(t) = \omega_{\pm} t + \phi_{\pm}$, being the $\omega_{\pm}$ phase progression starting from $\phi_{\pm}$. Now, multiplying $e^{\pm i \phi_{\pm}(t_{\text{conv}})}$ to both sides of equation 2.64, we get:

$$\begin{aligned} r_{\pm}(t_{\text{conv}}) e^{\pm i \phi_{\pm}(t_{\text{conv}})} &= r_{\mp}(0) e^{\pm i [\Delta\phi_Q + \phi_{\pm}(t_{\text{conv}})]} \\ &= r_{\mp}(0) e^{\pm i [(\phi_Q - \phi_+ - \phi_-) + (\omega_{\pm} t_{\text{conv}} + \phi_{\pm})]} \\ &= r_{\mp}(0) e^{\pm i (\phi_Q + \omega_{\pm} t_{\text{conv}} - \phi_{\mp})} \end{aligned} \quad (2.66)$$

Therefore, equating the real and imaginary parts of equation 2.66, we get two important results:

$$r_{\pm}(t_{\text{conv}}) = r_{\mp}(0) \quad (2.67a)$$

$$\text{and, } \phi_{\pm}(t_{\text{conv}}) = \phi_Q + \omega_{\pm} t_{\text{conv}} - \phi_{\mp} . \quad (2.67b)$$

Equation 2.67a means that after undergoing quadrupole excitation for a time $t = t_{\text{conv}}$, an ion's radial eigenmotions are converted completely, that is, its purely reduced cyclotron motion will be converted to a purely magnetron motion or the other way round. This is shown in Figure 2.10. On the other hand, equation 2.67b shows how the phase of a certain eigenmotion of the ion changes due to the application of such an excitation.

Quadrupole excitation is used to impart maximum axial energy to the ions in TOF-ICR [57, 60] (discussed in Section 2.4.1), and to extract the exact location of the spots in PI-ICR [45, 53], with the phase change equation essential in understanding the PI-ICR implementation at the CPT (described in sections 2.4.2 and 5).

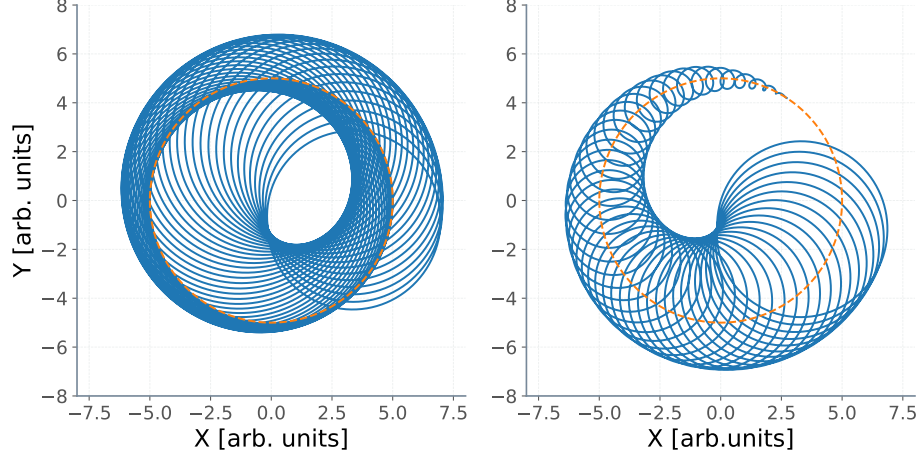


Figure 2.10 : Conversion of the ion's initial pure ω_+ to pure ω_- motion due to a quadrupole pulse at ω_c . The left and right panel denotes the motion for $0 \leq t \leq t_{\text{conv}}/2$ and $t_{\text{conv}}/2 \leq t \leq t_{\text{conv}}$ respectively. Figure taken, with permission, from Ref. [53]

2.4 Measurement techniques

As mentioned in Chapter 1, the necessary precision for mass measurements for astrophysics is within $100 \text{ keV}/c^2$, and for that, the two main techniques currently implemented by Penning trap groups around the world for ω_c measurement are TOF-ICR [57, 60] and PI-ICR [45, 53]. Till 2015, the CPT has used the TOF-ICR technique, and numerous papers [62–68] and Ph.D. theses [57, 69–71] have been published on the basis of those measurements. Since January 2016, CPT has implemented the PI-ICR technique [46, 53, 72–74]. In this section, I will discuss the basic theory behind both the techniques. The mass measurements presented in this thesis are conducted using PI-ICR. So PI-ICR is discussed in more detail and a separate chapter (5) is dedicated to that.

2.4.1 Time-of-flight ion-cyclotron-resonance (TOF-ICR)

TOF-ICR was first implemented at the ISOLTRAP group at the ISOLDE facility in CERN in the 1980s [75, 76] to conduct mass measurements of short lived ions with precision better than 0.5 ppm. The main principle governing TOF-ICR is that when a charged particle like an ion with a radial motion passes through a region of magnetic field gradient, they get accelerated. This is the situation when an ion is ejected from the Penning trap.

At such a point, the force on the particle would be:

$$\vec{F} = \vec{\nabla}(\vec{\mu} \cdot \vec{B}), \quad (2.68)$$

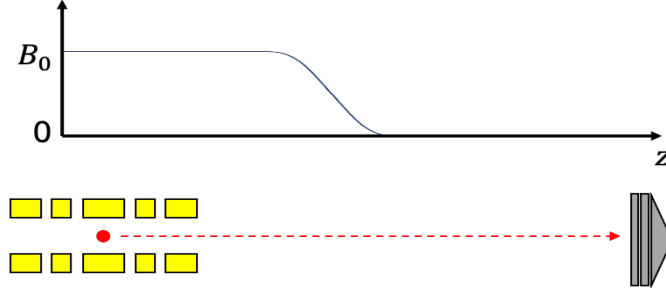


Figure 2.11 : Basic idea of TOF-ICR operation. On the top, the magnetic field is shown along the z -direction. At the bottom, the Penning trap electrodes are shown in yellow with the ion represented by the red dot. The dashed red arrow is the ion's path to the detector. Figure reproduced from Ref. [45].

where $\vec{\mu}$ is the magnetic moment of the ion. On the other hand, if U_r is the initial radial energy, we get:

$$\vec{F} = -\vec{\nabla} U_r. \quad (2.69)$$

Comparing equations 2.68 and 2.69:

$$U_r = -\vec{\mu} \cdot \vec{B}. \quad (2.70)$$

Now if the field inside the trap is $\vec{B} = B_0 \hat{z}$ and μ is given as $\vec{\mu} = -\mu_0 \hat{z}$, we get:

$$\mu_0 = \frac{U_r}{B_0}, \quad (2.71)$$

which makes equation 2.68:

$$\vec{F} = -\mu_0 \frac{\partial B}{\partial z} \hat{z} = -\frac{U_r}{B_0} \frac{\partial B}{\partial z} \hat{z}. \quad (2.72)$$

In other words, an ion exiting a Penning trap would feel a force along the z -direction. This force will be proportional to the ion's radial energy at that point. Inside the trap, the ion, undergoing ω_+ and ω_- motion, (Figure 2.7(b)) will have a total radial energy:

$$U_r = \frac{1}{2} m [(r_+ \omega_+)^2 + (r_- \omega_-)^2]. \quad (2.73)$$

If an ion has a purely ω_- motion at time $t = 0$ before a quadrupole excitation is applied on it, that is $r_+(0) = 0$, equation 2.60 reduces to:

$$r_+^2(t) = \frac{1}{4} \frac{k_0^2}{\omega_B^2} r_-^2(0) \sin^2(\omega_B t) \quad (2.74a)$$

$$r_-^2(t) = r_-^2(0) \left[\cos^2(\omega_B t) + \frac{1}{4} \frac{(\omega_Q - \omega_c)^2}{\omega_B^2} \sin^2(\omega_B t) \right]. \quad (2.74b)$$

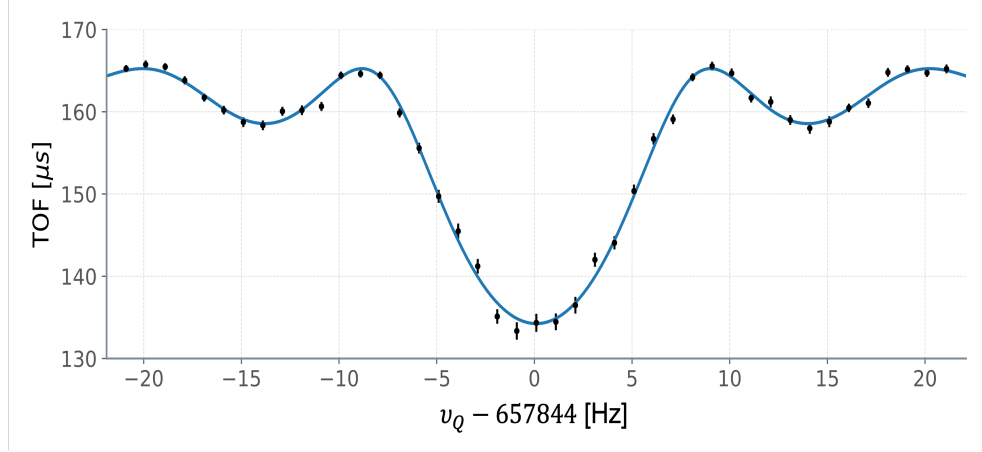


Figure 2.12 : A TOF-ICR spectrum of $^{133}\text{Cs}^+$ at the CPT using a $t_Q = 100$ ms, where $\nu_Q = \omega_Q/2\pi$. Figure taken, with permission, from Ref. [53].

Therefore, after some duration t of quadrupole excitation, using 2.74, the ion will gain a radial energy given, from equation 2.73, as:

$$U_r = \frac{1}{2} m r_-^2(0) \left[\frac{1}{4} \frac{k_0^2}{\omega_B^2} \omega_+^2 \sin^2(\omega_B t) + \omega_-^2 \left(\cos^2(\omega_B t) + \frac{1}{4} \frac{(\omega_Q - \omega_c)^2}{\omega_B^2} \sin^2(\omega_B t) \right) \right]. \quad (2.75)$$

Now, if $\omega_Q = \omega_c$, $\omega_B = k_0/2$ and then, this becomes:

$$U_r = \frac{1}{2} m r_-^2(0) \left[\omega_+^2 \sin^2\left(\frac{k_0}{2}t\right) + \omega_-^2 \cos^2\left(\frac{k_0}{2}t\right) \right]. \quad (2.76)$$

Since $\omega_+ \gg \omega_-$, from equation 2.76, we see that U_r will be the maximum if $\sin^2(k_0 t/2) = 1$, or $t = \pi/k_0 \approx \pi B_0/V_Q$. Therefore, to summarize, if an ion trapped inside a Penning trap, with a negligible ω_+ radius and a certain non-zero ω_- radius, undergoes quadrupole excitation at its cyclotron frequency ($\omega_Q = \omega_c$) and hence a complete conversion of its radial eigenmotions, it will gain a maximum radial energy. This energy will lead to it experiencing the maximum force upon ejection from the trap, meaning it would take the minimum time to reach a detector, placed a certain distance away from the trap (Figure 2.11).

In practice, once an ion is captured, it is excited to a radius by dipole excitation at the $\omega_D = \omega_-$ inducing a certain ω_- radius in its motion. Next, a quadrupole excitation is applied for time t_Q at some frequency ω_Q , slightly different from the ion's true cyclotron frequency, and then it is ejected. Post ejection, its TOF to the detector is measured. For the next ion batch, the same procedure is followed, except the quadrupole frequency is again different

to the last one. This is repeated and in the process, a scan of ω_Q is performed such that, $\omega_c - \Delta\omega < \omega_Q < \omega_c + \Delta\omega$, where $2\Delta\omega$ is the range of the scan. The result would be a time-of-flight spectrum plotting the TOF against the ω_Q , as shown in Figure 2.12, where $\nu_Q = \omega_Q/2\pi$. In this figure, each data point represents the mean of several TOFs obtained from the procedure being repeated several times at a particular ω_Q , while the errorbar is the standard deviation of those measurements [53]. The final ω_c is extracted from the minimum TOF position in the modified sinc fit function. The uncertainty and mass resolving power of this technique can be obtained from the Full Width Half Maximum (FWHM) as [53, 60, 77]:

$$\delta\omega_c = 2\pi\delta\nu_c \propto \frac{1}{t_Q \sqrt{n}} \quad (2.77a)$$

$$R = \frac{m}{\Delta m} = \frac{\omega_c}{\Delta\omega_c} \approx 0.2 \omega_c t_Q. \quad (2.77b)$$

where n is the number of detected ions. To give an idea, a measurement using a pure beam consisting of only $^{133}\text{Cs}^+$ ions for a time $t_Q = 100$ ms is shown in Figure 2.12. This measurement produced a statistical uncertainty of 0.07 Hz [53] in the ω_c measurement, and a mass uncertainty of ~ 13 keV.

Apart from the ISOLTRAP [78] and the CPT [79], TOF-ICR has been successfully used for many years by several Penning trap groups including the JYFLTRAP [80], the Titan Penning trap [81] and the LEBIT Penning trap [82]. But it has some limitations, especially when it comes to measuring weakly produced nuclides. Additionally, as can be seen from 2.77, the Fourier-limited mass resolution of TOF-ICR makes it difficult to conduct mass measurements when molecular, isomeric or other isobaric close-massed contaminants are present in the beam, as is usually the case for beams of weakly produced neutron-rich isotopes. Finally, to reduce uncertainty, longer t_Q is needed which again makes it challenging to study the short lived isotopes [53, 57].

2.4.2 Phase-Imaging Ion-Cyclotron Resonance (PI-ICR)

PI-ICR was first devised at the SHIPTRAP group [45, 83] at GSI in 2014 to overcome the limitations of TOF-ICR. It relies on measuring the frequency of the trapped ion by measuring the phases of its radial motions, accumulated over a period of no excitation, by

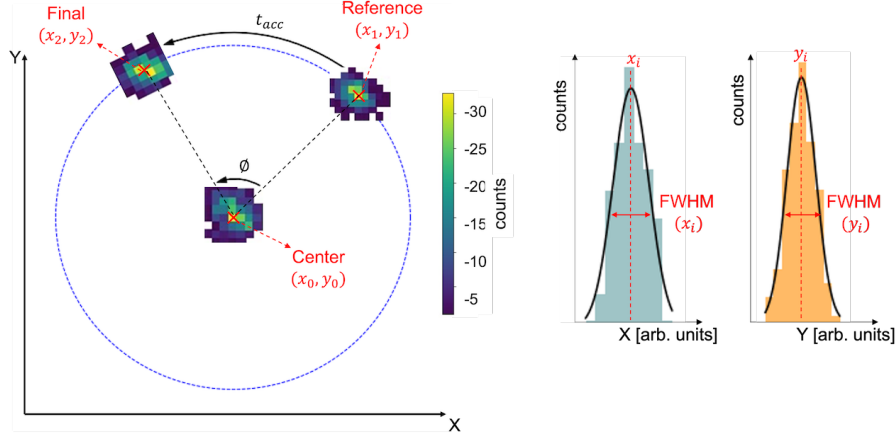


Figure 2.13 : Basic concept behind PI-ICR. On the left, the three positions center (0), reference (1) and final (2), are plotted on the same spectrum to clarify the concept. The surplus acquired phase ϕ over the accumulation time t_{acc} is also shown. On the right, the X and Y projections of the i -th spot (i being 0, 1 and 2), fitted to a Gaussian profile are shown along with the extracted x and y coordinates and FWHM from the fits.

projecting them onto a position sensitive detector and obtaining three positions:

1. The center (position ‘0’): This is essentially probing the center of the trap. It is obtained by capturing a batch of ions, applying a *re-centering* dipole excitation such that $\omega_D = \omega_-$ with $\Delta\phi_{D-} = 3\pi/2$ (see equation 2.55c), and finally ejecting them.
2. The reference (position ‘1’): For the reference position, the ion batch is captured and re-centered like the ”Center” spot. But then, they are excited to some large radius via a continuation of the re-centering excitation, or dipole excitation with $\omega_D = \omega_+$ or ω_- , depending on the frequency being measured, before they are ejected.
3. The final (position ‘2’): For the final position, the ions are captured, re-centered and excited to a radius similar to the reference position. But this time, they are allowed to acquire some phase ϕ_{total} over some time of accumulation t_{acc} before being ejected.

If the procedures to obtain the center, reference and final positions are repeated with multiple batches of ions, we get the corresponding spots. From such spots, we can project the X s and Y s, fit them to a Gaussian profile and extract the positions (x_i, y_i) , i representing all three positions and given as $i = (0, 1 \text{ and } 2)$, along with their standard deviations (σ_{xi} and σ_{yi}). This is shown in Figure 2.13, where all three positions are shown on the same

XY-plot to clarify the concept. Now, to simplify matters, let us define a new co-ordinate system, shifting the origin from $(0, 0)$ to the trap center (x_0, y_0) :

$$x'_j = x_j - x_0 \quad ; \quad \delta x'_j = \sqrt{(\delta x_j)^2 + (\delta x_0)^2} \quad (2.78a)$$

$$y'_j = y_j - y_0 \quad ; \quad \delta y'_j = \sqrt{(\delta y_j)^2 + (\delta y_0)^2}. \quad (2.78b)$$

j here represents the excited positions, reference and final, that is $j = 1$ and 2 , while δA_i is the positional uncertainty and given as $\delta A_i = \sigma_{Ai}/\sqrt{n_i}$ ($A = x, y$), and n_i being the number of ion hits for the i^{th} spot (as before, i represents all three positions). With this, we can find the surplus acquired phase ϕ , as follows:

$$\phi = \begin{cases} \left| \tan^{-1} \left(\frac{y'_2}{x'_2} \right) - \tan^{-1} \left(\frac{y'_1}{x'_1} \right) \right|, & \text{for } 0 \leq \phi \leq \pi \\ 2\pi - \left| \tan^{-1} \left(\frac{y'_2}{x'_2} \right) - \tan^{-1} \left(\frac{y'_1}{x'_1} \right) \right|, & \text{for } \pi < \phi < 2\pi \end{cases} \quad (2.79a)$$

$$\delta\phi = \sqrt{\frac{x_1'^2 (\delta y_1')^2 + y_1'^2 (\delta x_1')^2}{(x_1'^2 + y_1'^2)^2} + \frac{x_2'^2 (\delta y_2')^2 + y_2'^2 (\delta x_2')^2}{(x_2'^2 + y_2'^2)^2}} \quad (2.79b)$$

From ϕ , one can find the frequency as:

$$\nu = \frac{\omega}{2\pi} = \frac{\phi_{total}}{2\pi t_{acc}} = \frac{\phi + 2\pi N}{2\pi t_{acc}} \quad (2.80a)$$

$$\delta\nu = \frac{\delta\omega}{2\pi} = \frac{\delta\phi}{2\pi t_{acc}}. \quad (2.80b)$$

If we consider a slightly exaggerated $\text{FWHM}(x_i) = \text{FWHM}(y_i) \approx 2$ mm, a typical reference and final position radius $r_j = \sqrt{x_j'^2 + y_j'^2} \approx 4$ mm, a $t_{acc} = 100$ ms (comparable to the TOF-ICR measurement shown in Figure 2.12), we get $\delta\nu \sim 0.007$ Hz with 100 ions per spot, which is an order of magnitude improvement on the TOF-ICR result. For the mass resolving power between the two techniques, a PI-ICR measurement of ^{106}Nb at the CPT at $t_{acc} = 1500.03$ ms has helped resolve masses of ^{106}Nb from its isomer ^{106m}Nb , thereby setting a record mass resolving power of $R \approx 2.5 \times 10^7$ [53]. From equation 2.77b, a similar measurement time (t_Q) will help achieve a mass resolving power of $\sim 0.4 \times 10^7$, that is, PI-ICR could provide a six times improvement in mass resolution. Details of mass resolution in PI-ICR at the CPT will be discussed in section 5.4.2.

Using PI-ICR, both the radial eigenfrequencies (ω_+ and ω_-), as well as ω_c directly can be measured. One needs to drive the ions such that they undergo the relevant motions while

accumulating the phases. For example, if the dipole excitation to increase the ions' radius is applied at $\omega_D = \omega_{\pm}$, at the end of it, the ions will have $\omega_{\pm}$ motion, and therefore the phases acquired at that point will be of the corresponding eigenmotion. Finally, since it is a fast motion, ions undergoing ω_+ when they are ejected will be imaged as a ring on the detector, rather than a spot, making it impossible to extract the positional information. Thus at ejection, the ions must be undergoing the slow ω_- motion, and so, if necessary, a quadrupole $\omega_Q = \omega_c$ excitation must be applied before ejection. These will be described in further detail in Chapter 5.

2.5 Mass determination in a Penning trap

As mentioned before, Penning trap mass spectrometry relies on measuring the cyclotron frequency of the trapped ion (equation 2.25). Once ω_c of the isotope of interest (denoted by “ I ”) is measured precisely, one needs to know the magnetic field to the same, if not better, precision. This is done by measuring the frequency of a second isotope, a calibrant species (denoted by “ cal ”), usually a stable isotope or molecule, whose mass is already known precisely. Therefore, the mass of the isotope I will be found as:

$$m_I = \frac{q_I}{q_{cal}} \frac{\omega_{c,cal}}{\omega_{c,I}} (m_{cal} - q_{cal}m_e) + q_I m_e \quad (2.81a)$$

$$\begin{aligned} \delta m_I = & \left(\frac{q_I}{q_{cal}} \frac{m_{cal} - q_{cal}m_e}{\omega_{c,I}} \right)^2 (\delta\omega_{c,cal})^2 + \left(\frac{q_I}{q_{cal}} \frac{\omega_{c,cal}(m_{cal} - q_{cal}m_e)}{\omega_{c,I}^2} \right)^2 (\delta\omega_{c,I})^2 \\ & + \left(\frac{q_I}{q_{cal}} \frac{\omega_{c,cal}}{\omega_{c,I}} \right)^2 (\delta m_{cal})^2. \end{aligned} \quad (2.81b)$$

where m_e is the mass of an electron. Now if we define a ratio parameter R_c as $R_c = \omega_{c,cal}/\omega_{c,I}$, any shift in the measured frequency ω_c , due to systematic effects from misalignment of the trap's electrodes, deviations or fluctuations in the B -field, or a practical trap being different from an ideal trap, will lead to a slightly different ratio $R_{c, meas.}$, given as:

$$R_{c, meas.} = \frac{\omega_{c,cal} + \Delta\omega_{c,cal}}{\omega_{c,I} + \Delta\omega_{c,I}} \quad (2.82)$$

If $\Delta R_c = R_{c, meas.} - R_c$, we see that:

$$\frac{\Delta R_c}{R_c} = \frac{R_{c,meas.} - R_c}{R_c} = \frac{1 + \frac{\Delta\omega_{c,cal}}{\omega_{c,cal}}}{1 + \frac{\Delta\omega_{c,I}}{\omega_{c,I}}} - 1 \approx \frac{\Delta\omega_{c,cal}}{\omega_{c,cal}} - \frac{\Delta\omega_{c,I}}{\omega_{c,I}} \quad (2.83)$$

Usually $\Delta\omega_{c,cal} \approx \Delta\omega_{c,I}$. Therefore, for $\frac{\Delta R_c}{R_c} \approx 0$, we need $\omega_{c,cal} \approx \omega_{c,I}$. In other words, to minimize systematic effects, and hence maximize accuracy, the ratio q/m or equivalently A/q (A being the mass number) of the calibrant (“*cal*”) and the species of interest (“*I*”) is usually kept very similar, that is R is kept as close to 1 as possible. As mentioned before, the electron ionization energy or other molecular binding energies applicable here are usually of the order of eV, many orders of magnitude smaller than the masses measured [39]. Therefore, they will be ignored in the context of this thesis.

3 The facility : CARIBU

All the nuclides measured as part of this thesis were produced from CARIBU [84], located in the ATLAS facility [85, 86] at ANL. CARIBU was commissioned in 2009 and came online in 2011 [70, 71]. It is a unique facility, which produces neutron-rich nuclides by collecting the spontaneous fission fragments from a ^{252}Cf source. Currently, the facility consists of two levels of beamlines. The first or lower level is used for beam production, optimizations, and delivery to the CARIBU low-energy experimental area, where the CPT resides, while the upper level is used to transport beam to either a second low-energy experimental area (known as Area 1), or the Electron Beam Ion Source (EBIS) [87] for charge-stripping before being injected into ATLAS for other experiments [88]. A schematic plan of the lower level is shown in Figure 3.1. The major components here are the ^{252}Cf source and the gas catcher (for beam generation and extraction, see Section 3.1), the isobar separator (as a first stage of purification, see Section 3.2), the RFQ buncher and elevator (to convert the DC beam to a bunched beam and to lower the ions' energies, see Section 3.3) and the Multi-Reflection Time-Of-Flight (MR-TOF) mass separator and a Bradbury Nielsen Gate (BNG) (as the second and final stage of purification, see Section 3.4). Before proceeding any further, I would like to define the term “isobars”. In physics, an atomic or a nuclear species having the same mass number A , that is same total number of protons and neutrons, are called isobars. However in the context of this thesis, the word isobar has been used, in a broader sense, to mean ionic species with the same A/q .

There are four diagnostic stations to optimize the settings of the components and tune the beam. These consist of different combinations of silicon (Si) surface barrier and microchannel plate (MCP) detectors, and are located between the isobar separator and the deflectors (location name: *deflector*), the deflectors and the buncher (location name: *buncher-up*), the buncher and the MR-TOF (location name: *buncher-down*), and downstream from the MR-TOF (location name: *MR-TOF*). Additionally, a BNG is included at the post-MR-TOF location. The Si detectors have an aluminium foil mounted in front of a reverse biased Si semiconductor diode. When the foil is placed in the path of the beam, ions get implanted on it. If these ions are radioactive, they emit β particles, which pass through the foil and the

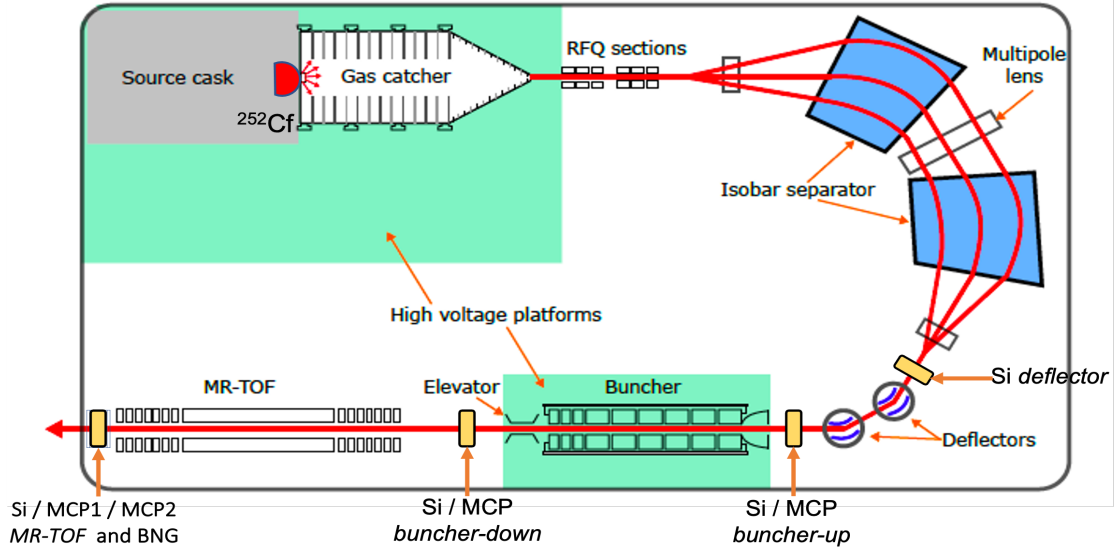


Figure 3.1 : Schematic diagram of the lower floor of CARIBU, with labelled major components. Figure originally drawn by R. Orford, with the ^{252}Cf source, and the diagnostic detectors added by this author.

depletion zone of the Si diode. As the particles pass through the diode, they create electron-hole pairs generating a current which is detected, amplified and sorted using a Single Channel Analyzer (SCA), and used to trigger a counter. This measured rate of “radioactivity” gives one a measure of intensity of the radioactive beam. The MCP detectors consist of two plates with multiple channels, acting as continuous dynodes, stacked in a chevron configuration. When a potential difference is applied across the two plates, each ion hit on the front plate triggers a cascade of electrons, which are detected at the back plate (anode). MCP detectors can be used to determine the time of flight of ions with respect to some trigger. For stable beams, the beam intensity is determined using the MCP detectors. The diagnostic station before the deflector consists of a Si-detector (*Si deflector*), and the ones before and after the buncher (that is, *buncher-up* and *buncher-down*) include a Si and an MCP detector (*Si / MCP buncher-up* and *buncher-down*). At the end of the MR-TOF, timing becomes of paramount importance as the MR-TOF resolution can only be tuned with an MCP. So at this location, there are one Si and two MCP detectors (*Si / MCP1 / MCP2 MR-TOF*). These are shown in Figure 3.1.

3.1 Ion source and gas catcher

The root of CARIBU is a ^{252}Cf source decaying with a half-life of 2.645(8) years and having a spontaneous fission branch of 3.092(8)% [89], the rest decaying via α -decay [90]. The fission fragments consist of neutron-rich isotopes, distributed around two broad peaks around $A = 105$ (*light peak*) and $A = 140$ (*heavy peak*) as shown in Figure 3.2. The current ^{252}Cf source was obtained from the Oak Ridge National Laboratory and installed in March 2018, and after extensive tests and optimization, it delivered beam to the first successful experiment in June 2018 [91]. The source had a total strength of ~ 0.5 Ci upon delivery, but due to its thickness, this was reduced to an effective strength of ~ 150 mCi upon installation [92], with a rate of ~ 22 kHz of $^{150}\text{Ce}^{2+}$ detected on the Si-*deflector*. The fission yield data in Figure 3.2 has been taken from Ref. [93], then normalized to the rate of $^{150}\text{Ce}^{2+}$, detected at ~ 17 kHz on Si-*deflector* in 2019. $^{150}\text{Ce}^{2+}$ belongs to a fission branch of 0.9% [93] and has a half-life of 6 s [3], making it a good candidate for optimizing the CARIBU beam for most of the masses presented in this thesis.

The source is hosted on a mount, and shielded by a movable cask at the back. A gas filled cell, also called gas catcher [84, 90], sits at the front of the source, allowing the fission fragments to enter it. It consists of a cylindrical shaped section (*body*) and a cone shaped section (*cone*). The fission fragments are emitted with average energies of 80 MeV for the *heavy peak* and 104 MeV for the *light peak* [94]. In order to stop and cool these energized fragments, the gas catcher is filled with He gas, supplied from the ATLAS cryogenic system. To reduce the number of contaminants in the beam coming out from the gas catcher and minimize charge exchange, the purity of this He gas needs to be of the highest order. This is ensured by first having it pass through a cold trap, maintained at liquid nitrogen (LN_2) temperature, and then filtering it through a commercial purifier. Since He cannot be frozen above 270 K, the cold trap freezes and eliminates most of the contaminants, while letting He gas pass through. In principle, a denser He gas in the gas catcher means better stopping and cooling of the fission products. On the other hand, a high pressure in this section could affect the pressures in the subsequent sections, and hence efficient transport of ions from the gas catcher [53]. One way to tackle this would be to increase the He pressure (P) as much

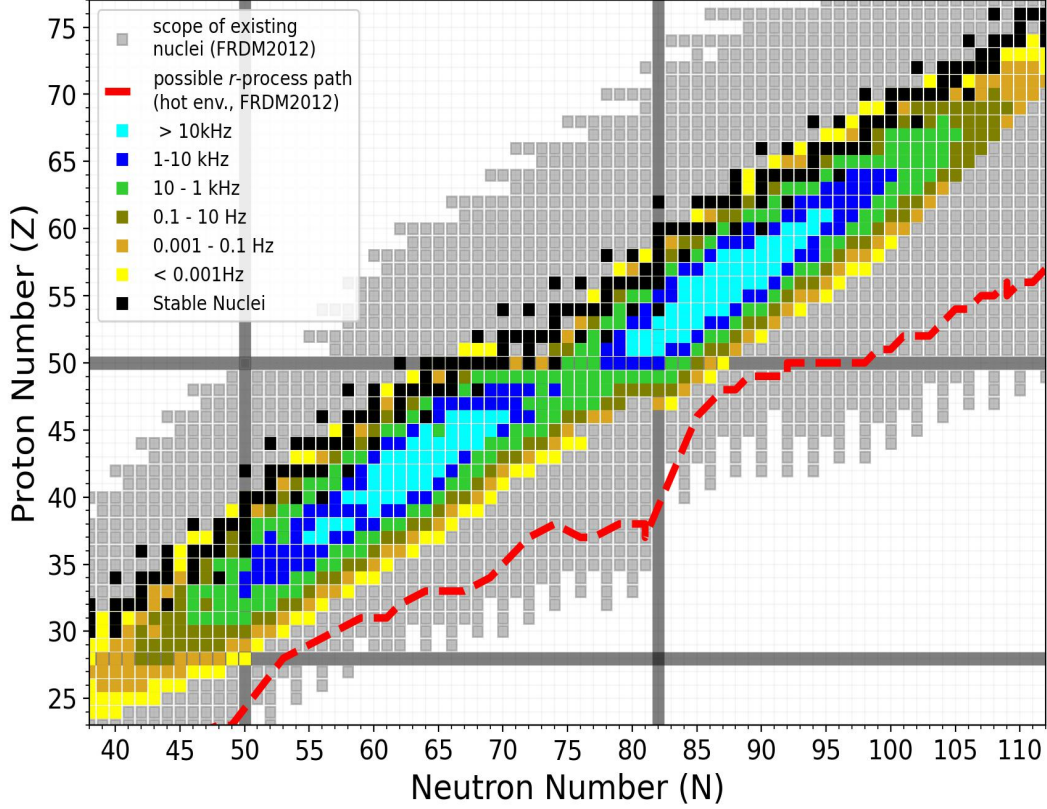


Figure 3.2 : Distribution of ^{252}Cf [93] fission fragments normalized to the rate of $^{150}\text{Ce}^{2+}$ on Si *deflector*. The black squares represent the stable nuclei and the gray bands at $Z = 28, 50$ and $N = 50, 82$ represent the magic proton and neutron numbers respectively. The grey squares shows the scope of existing nuclei from the Finite Range Droplet Model (FRDM2012) mass model [31]. A projected r -process path for a hot environment is also shown in red.

as possible while lowering the temperature (T) to ensure a maximum possible density (ρ), from the ideal gas law equation:

$$P = \rho RT \quad (3.1)$$

where R is the ideal gas constant. Keeping this in mind, the gas catcher is typically operated at $35 - 40$ torr and -35° C [95]. The pressure is set by the He gas flow, controlled from a Type 247 mass flow controller from MKS Instruments. According to calculations done in the freeware Stopping and Range of Ions in Matter (SRIM2013) [96], the range of ^{150}Ce with energy 80 MeV within a He gas column at 40 torr and -35° C is around 2 m . The CARIBU gas catcher is 1.2 m in length and 50 cm in diameter [90]. To collect and thermalize the fragments inside this volume, a thin gold degrader foil is placed between the source and the gas catcher to lower the energies of the fragments to a few MeV [90, 97], such that they are stopped within $\sim 60 \text{ cm}$ [98] in the gas catcher. Sometimes, the presence of water molecules in the gas catcher leads to formation of contaminant isobar molecules. The -35° C temperature also freezes out these water molecules on the surface of the chamber and helps to minimize such contaminants.

When the fission fragments first come out from the source, they possess a wide range of charge states. As they travel through the gas catcher and undergo collisions with the residual He gas, they accept electrons. The charge states of the fission fragments after they undergo these collisions, depend on the fragment's ionization energies. The first, second, and third ionization energies (E_n , $n = 1, 2$ and 3) of elements from Mn ($Z = 25$) to Re ($Z = 75$), along with the first ionization energy of He gas (24.6 eV) and its metastable 2^3S_1 and 2^1S_1 states (19.8 eV and 20.6 eV , respectively) [99], are shown in Figure 3.3. The yield per 100 fissions of ^{252}Cf [93], summed over Z , is depicted as vertical bands in grey gradient to illustrate the positions of the light and heavy peaks. If the ionization energy E_n ($n = 1, 2$ or 3) of an element is less than that of the He metastable states, all isotopes of that element will be extracted primarily at charge state $q = n+$.

For the elements of the heavy peak, Cs denotes a well defined boundary, such that all elements from indium to caesium are produced primarily in the $q = 1+$ charge state, while everything from barium and heavier ($Z \geq 56$) are extracted mostly in the $q = 2+$ state. For

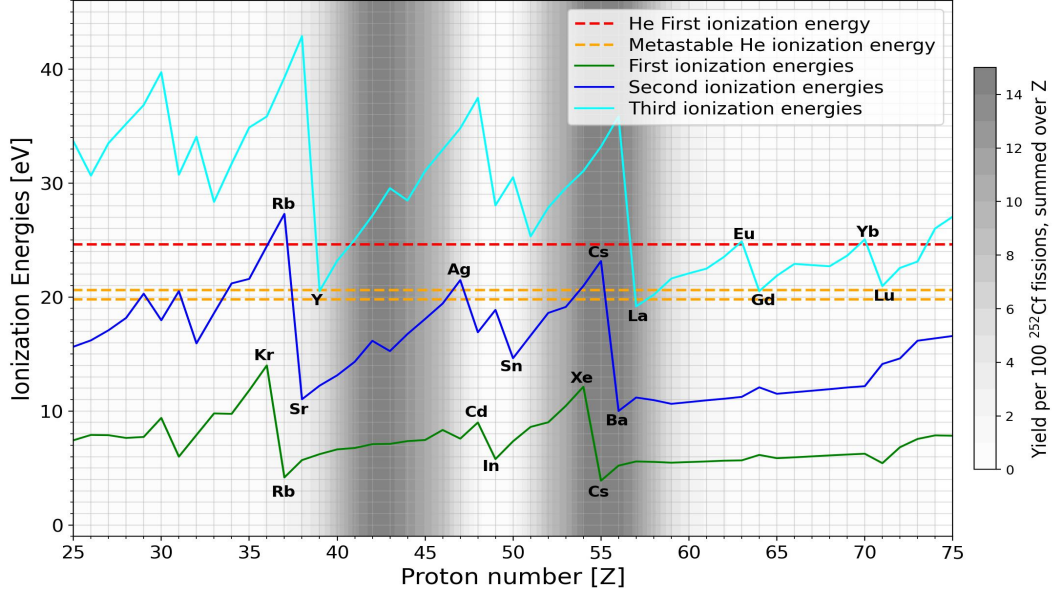
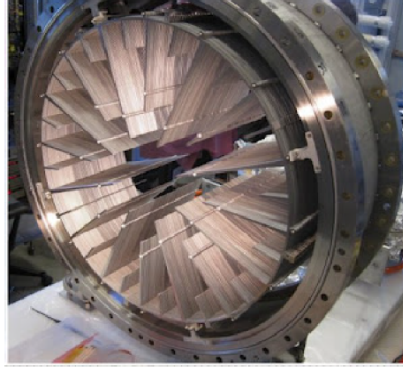


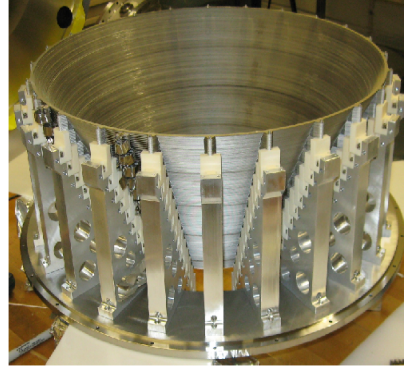
Figure 3.3 : First, second and third ionization energies of elements from Mn ($Z=25$) to Re ($Z=75$) are plotted in green, blue, and aqua, respectively. The first ionization energy of He (24.6 eV) is shown with the red dashed line. The orange dashed lines represent the ionization energies of the metastable 2^3S_1 and 2^1S_1 He states (19.8 eV and 20.6 eV, respectively) [99]. The yield per 100 fissions of ^{252}Cf [93], summed over Z , is also shown as vertical bands in grey gradient to illustrate the light and heavy peaks.

the lighter elements however, it is a little tricky. Rubidium, like Cs, and Ag are produced mainly in the $1+$ state, while Sr comes out as $2+$ ions. The other lighter nuclides, having intermediate second ionization energies between Sr and Ag or Cs, could be found at both $1+$ and $2+$ states. It should be mentioned that, being a noble gas, with minimum chemical reactivity and maximum first ionization energy, He is the best choice of medium for stopping, thermalizing, and ionizing the fission fragments.

The gas flow alone is not sufficient to efficiently transport the ions through the *body* section of the gas catcher to the exit nozzle at the end of the *cone*. To facilitate the ions' movement towards the cone, a DC gradient is applied along the length of the *body*, with a series of ring-shaped electrodes connected together through a resistor chain. Because of the large diameter of this section, these electrodes are actually spoked rings, so that ions, irrespective of their positions, whether near the center or the wall, experience a similar force from the potential gradient. One such spoked ring section is shown in Figure 3.4(a). Additionally, to prevent the ions from hitting the rings and getting lost, an RF oscillating voltage is coupled to the DC of each ring, such that the adjacent rings are 180° out-of-phase,

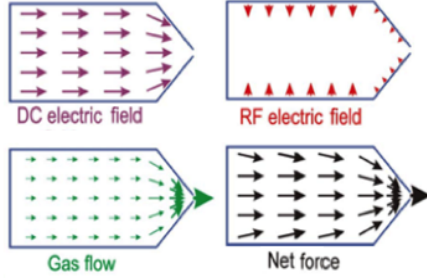


(a) The ring electrodes on the body section during assembly

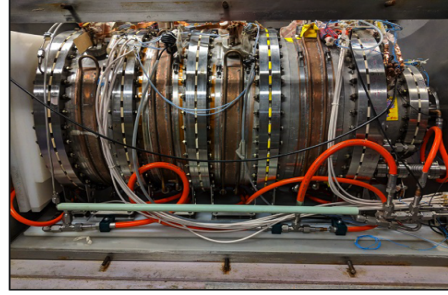


(b) The cone section during assembly

Figure 3.4 : The CARIBU gas catcher body and cone sections showing spoked and concentric ring electrodes respectively.



(a) Combination of gas flow, DC and RF field enabling transport of ions through the gas catcher.



(b) Picture of the gas catcher in place during a period of maintenance

Figure 3.5 : The CARIBU gas catcher

which pushes the ions towards the center when they come close to the outer diameter of the rings. A similar principle is used for the cone, except here the spokes are replaced by concentric rings of gradually tapering radius to focus the ions towards the exit nozzle. This is shown in Figure 3.4(b). This combination of gas flow, and DC and RF fields enable an efficient extraction, which typically takes around 20 – 30 ms from fission production to the exit from the gas catcher [90]. A cartoon showing the combination of these forces, and a picture of the gas catcher in place, is shown in Figure 3.5. The entire gas catcher is kept within an insulated vacuum chamber so that it can be isolated from the room temperature and the atmosphere, allowing us to heat or cool it without condensation or ice formation. This keeps contaminants from getting inside the gas catcher.

A beam consisting of multiple isobars exits the gas catcher and undergoes isobaric sep-

aration in the first stage of its path while passing through a pair of dipole magnets. Upon their exit from the gas catcher, the ions undergo cooling by passing through a series of RFQ ion guides, and are separated from the residual gas by differential pumping. The CARIBU gas catcher and associated RFQ ion guides are located on a high voltage (HV) platform, that is biased to voltages up to 36 kV. As they emerge from the HV deck, they are accelerated to a kinetic energy of qV , and hence a speed of:

$$v = \sqrt{2V(q/m)} , \quad (3.2)$$

V being the deck voltage and q/m being the charge-to-mass ratio of the ions, before entering the isobar separator section [53]. Typical settings for the gas catcher are shown in Table 3.1.

Table 3.1 : Typical CARIBU gas catcher settings

DC settings	Typical values	Notes
Extraction voltage	36 kV	Supplied by a high voltage power supply from FuG Elektronik GmbH with 65 kV and 5 mA limits.
Pressure	~ 35 torr	Set by the He gas flow, controlled from a Type 247 mass flow controller from MKS Instruments.
Temperature	-35°C	Chiller range: -60°C to $+50^\circ\text{C}$.
DC settings	Typical values	Notes
Body section start	1200 V	Supplied by an ISEG MPOD HV card with 2kV and 2mA limits.
Body section end	540 V	
Cone section start	540 V	
Cone section end	440 V	
RF settings	Typical values	Notes
Body section RF frequency	1550 kHz	Supplied by two AG Series Amplifier from T&C Power Conversion Inc.
Body section amplifier power	100 W	
Body section RF voltage [98]	150 V _{pp}	
Cone section RF frequency	3035 kHz	
Cone section amplifier power	50 W	
Cone section RF voltage [98]	150 V _{pp}	

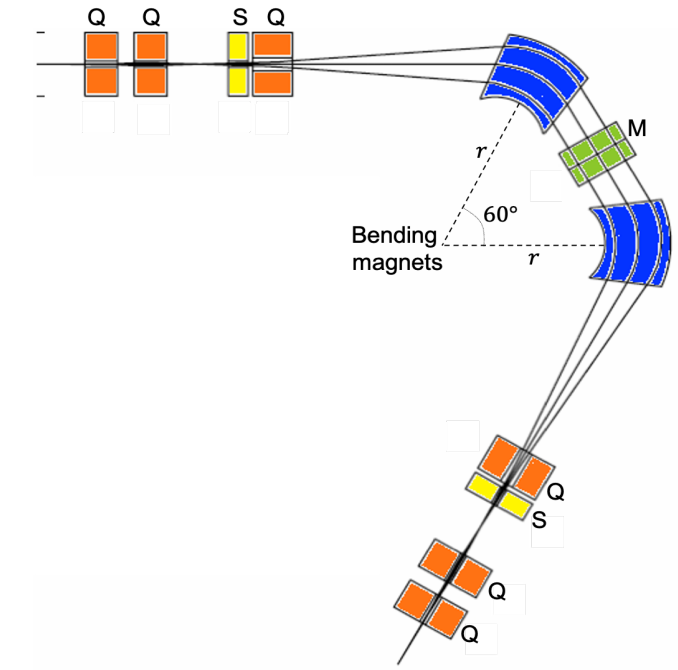


Figure 3.6 : A schematic diagram of the CARIBU isobar separator design, consisting of two 60° bending magnets and a system of six quadrupoles (Q), two sextupoles (S), and one multipole (M) lenses. Figure taken from Ref. [100] and colors added for ease of understanding.

3.2 Isobar separator

The ions, as mentioned, undergo a first stage of selection and purification at the isobar separator [100] after exiting the gas catcher. The device consists of a pair of 60° bending dipole magnets with a radius (r) of 50 cm and a series of focussing and aberration-correction elements. These elements had to be solely electrostatic to make them mass independent. The focussing is done with a pair of quadrupole doublets and singlets, one on either side of the magnet pair. A sextupole singlet, next to the quadrupole singlet, and a 48-element multipole lens, with a radius of 20 cm, comprising of sextupole, octupole, decapole and dodecapole components, in between the magnets, allows for aberration corrections up to the 5th-order [100]. A schematic diagram of the isobar separator showing all the components is shown in Figure 3.6.

As the ion beam approaches the isobar separator, it is flattened in the vertical or y -direction while diverging in the horizontal or x -direction. This allows for the beam to be maximally spread as it passes through the bending magnets and gets mass resolved on the

basis of their mass-to-charge (m/q) ratio. The ions accelerated off the deck with velocity, given by Equation 3.2, will experience a centripetal force (mv^2/r), equal to the Lorentz force ($q \vec{v} \times \vec{B}$), inside the magnet:

$$\begin{aligned} \frac{mv^2}{r} &= q \vec{v} \times \vec{B} = qvB \quad (\because \vec{v} \perp \vec{B}) \\ \therefore B &= \frac{\sqrt{2V(m/q)}}{r} . \end{aligned} \tag{3.3}$$

For a particular set of B and V , only ions of a particular m/q will pass through. The CARIBU isobar separator magnets are capable of fields up to 1 T. For a beam of $A = 170$ and $q = 2+$ ions, one of the heaviest 2+ beam coming out from the deck at 36 kV, $B \approx 0.5$ T, while for $A = 149$ and $q = 1+$, the heaviest 1+ beam, $B \approx 0.68$ T. Additionally, a feedthrough with four slits of sizes 0.1 mm, 0.3 mm, 0.5 mm and 4 mm is placed after the magnets, between the quadrupole-sextuple singlet and the quadrupole doublets. These further help in removing isobaric contaminants, far away in mass from the species of interest, that are still present in the beam. B is optimized by checking the ion-rate at Si *deflector*. Typically one starts with a wider slit and moves to the narrow slits as B gets tuned. Once a field is set, it usually takes both the magnets some time to lock on to that value and stabilize. So for faster tuning, one would adjust the deck voltage to maximize the ion rate, then set the field once a *good* voltage setting has been obtained from Equation 3.3, and finally return the deck to the original 36 kV bias. After the isobar separator, the ions still retain the same 36 keV energy as they had at the exit of the gas catcher, but at this point the beam only consists of ions of a selected m/q or A/q , as they are deflected by a couple of electrostatic deflectors and sent to the RFQ buncher. The isobar separator was designed to achieve a theoretical mass resolution of $R = m/\Delta m = 22,400$ with a transmission of 95% [100], assuming a deck voltage of 50 kV. With the deck being operated at 36 kV, the best resolution observed to date is $R \sim 14,000$ while the device is routinely operated at $R \sim 5,000$ [53]. The presence of the MR-TOF downstream deems this resolution less critical. This pre-selection by the isobar separator plays an important role in preventing saturation in the next element of the system, the buncher, allowing for efficient bunching.

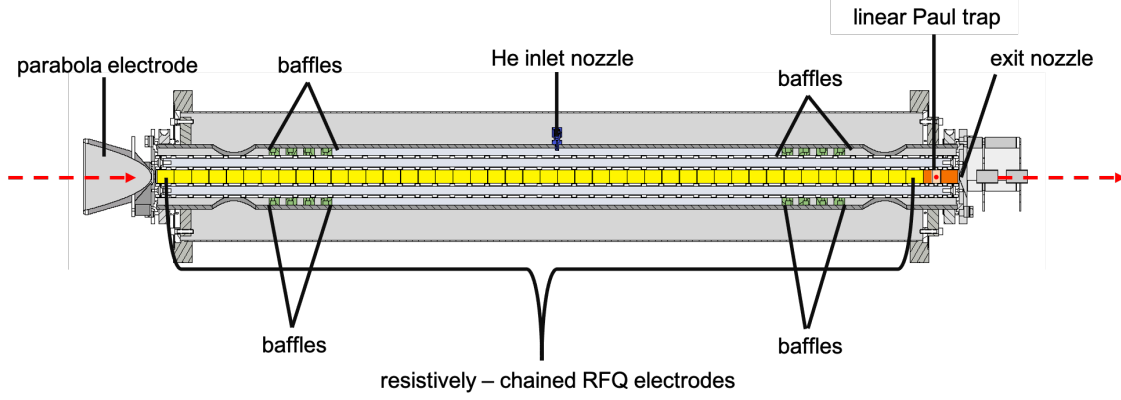


Figure 3.7 : Schematic diagram of the CARIBU buncher with labelled parts and a cartoon of the ion cloud (red circle in the linear RFQ trap). Figure courtesy: Nathan Callahan.

3.3 RFQ buncher and elevator

The ions of a selected A/q ratio, at this point, are in the form of a continuous or DC beam with energies 36 keV. Since the MR-TOF and the CPT (further downstream) require ions in bunches, the DC beam needs to be converted into a bunched beam. This is done at the RFQ buncher. A schematic picture is shown in Figure 3.7. The buncher consists of 4 stainless steel rods, similar to Figure 2.5(a) with 47 segments. RF potentials are applied to the four rods forming an RFQ. The first 44 segments are resistively chained together to form a potential gradient and create an RFQ ion guide, while the last three (45 – 47) create a potential well to form an RFQ linear Paul trap. The *ion-guide* electrodes and the last, 47th one, are each 20 mm long while the 45th and 46th electrodes are both 10 mm in length. These electrodes are enclosed within a 1 m long tube, with a He inlet nozzle at the middle. Two sets of 4 baffles are placed in the ion guide section (as shown in Figure 3.7) to create a high pressure region in the ion-guide section where the He gas and the RFQ cool the ions while pushing them towards the center of the RFQ. The baffles also reduce the amount of He going into the trapping region and the nozzle, thereby preventing collision-induced heating and ensuring a low pressure in the subsequent sections, the elevator and the MR-TOF. The device is placed on a HV platform maintained at the same potential as the gas catcher, to effectively stop and bunch the ions.

The ions enter the buncher through an aperture, focussed by a parabola shaped electrode.

As they are cooled and reach the final 3 electrodes, they are trapped at the bottom of the electrostatic potential well created by the linear Paul trap. An RF, set at around 1.5 MHz with an amplitude of $0.2 V_{pp}$ from a Stanford Research System (SRS), model DS335 function generator, and amplified to $600 V_{pp}$ [98] with an AG Series Amplifier from T&C Conversion Inc., is applied to the buncher. The exit nozzle is maintained at ground potential at all times to stop the ions from passing through or escaping, except during ejection. After accumulating ions for a user defined time (typically between 50 ms to 100 ms), the ions are ejected as a bunched beam by pulsing electrode 45 to +100 V and electrodes 47 and the exit nozzle to -100 V [53].

The bunched beam passes through an elevator, an MR-TOF, and a BNG before entering the CPT system. To effectively catch and release the bunched beam, precise timing is required. For all measurements presented in this thesis, the buncher ejection rate was set to 20 Hz. All the aforementioned post-buncher components are triggered by the signal which controls the ions' ejection from the buncher. These will be discussed in more detail in Section 3.5.

The beam upon exit from the buncher consists of isobars of a particular A/q at energy 36 keV. But the MR-TOF requires ions of lower energies. So the next step is to reduce the beam energy. This is done using an elevator, situated right after the buncher. This device consists of a 30 cm long drift tube, pulsed between HV (~ 33 kV) and ground while the bunched ions are inside. The energy of the ions is therefore ~ 3 keV upon exit [53], and they are prepared for injection into the MR-TOF.

3.4 MR-TOF and BNG

The CARIBU multi-reflection time-of-flight (MR-TOF) [101] was installed downstream from the buncher-elevator section in the CARIBU beamline in late 2014 and first commissioned in early 2015. It is modelled on the ISOLDE MR-ToF-MS [102] and consists of two 30 cm long electrostatic mirrors comprising of six independently controlled electrodes made from stainless-steel, separated by a 75 cm long central lift electrode which is basically a drift tube made out of stainless steel mesh (Figure 3.8). The electrodes that make the mirrors on either end are referred to as the mirror electrodes, labelled 1 to 6 from the inside going

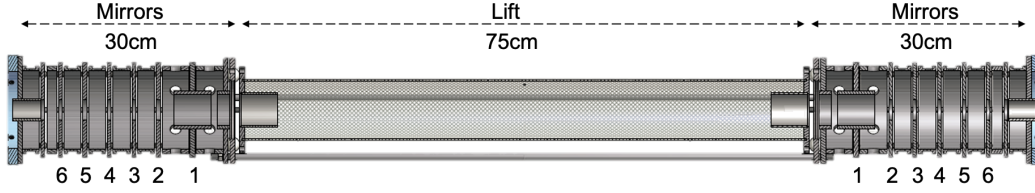


Figure 3.8 : Drawing of the CARIBU multi-reflection time-of-flight (MR-TOF). The mirror electrodes 1 to 6 on both sides are labelled along with their length and that of the central lift electrode. Figure courtesy: R. Orford

outwards, and are separated by shielding electrodes maintained at intermediate voltages to provide field uniformity and a smooth potential distribution. Of the mirror electrodes, 2 to 6 are identical coaxial rings with an inner diameter of 2.6 cm and outer diameter of 9.4 cm. Electrode 1, being the longest of all the mirror electrodes with a length of 5.0 cm and having a slightly larger inner diameter of 4.0 cm [101], acts as the focussing element. The MR-TOF in conjunction with the BNG, is operated as a mass selector to limit the number of species in the beam sent to the CPT in a fast and efficient manner. This is of paramount importance for the accuracy and precision of CPT measurements, as will be discussed in section 3.4.1, and a considerable time, online and offline, is spent to ensure the stability and high efficiency of the MR-TOF. In this section, I will discuss the MR-TOF in detail, its necessity, the technique implemented, its control, and operations.

3.4.1 The necessity

The mass separated beam from the isobar separator consists of multiple isobars of radioactive ^{252}Cf fission fragments, stable isotopes from He impurities in the gas catcher that cannot be removed by the cold trap, and other hydrocarbons, water, and hydroxide molecules. One main factor affecting the sensitivity of mass measurements in a Penning trap is the purity of the beam. In TOF-ICR, the presence of multiple species would lead to a shallow TOF spectrum. Figure 3.9 shows a preliminary $^{156}\text{Pr}^{2+}$ frequency scan done at the CPT in 2014 [53] with $t_Q = 50$ ms. This was done before the installation of the MR-TOF, meaning all species in the beam post the isobar separator were sent to the CPT. During that time, an effective, but time-consuming ω_+ cleaning cycle was employed at the CPT. At the time of this measurement, the $t_{1/2}$ of ^{156}Pr was not known. So special care was taken to keep the total time of cleaning and excitation to a minimum of 150 ms and 50 ms, respectively. This

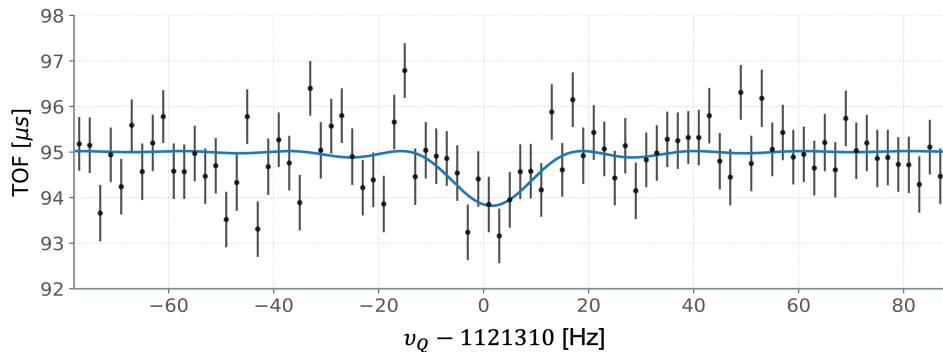


Figure 3.9 : A TOF-ICR spectrum of $^{156}\text{Pr}^{2+}$ with a contaminated beam at the CPT with $t_Q = 50$ ms. Figure taken, with permission, from Ref. [53].

resulted in a mass excess uncertainty of 258 keV [53]. The isotope was later measured using PI-ICR with an uncertainty of 1.0 keV [103]. Even though this example highlights the limitations of TOF-ICR, it also shows the necessity of having a beam as pure as possible. A longer ω_+ cleaning cycle, in principle, could clean the contaminants in the beam, but that would limit the scope of measuring short lived isotopes.

On the other hand, with PI-ICR, different species in the beam will acquire different final phases depending on the accumulation time (t_{acc}), leading to increased resolution with larger t_{acc} . A busy PI-ICR final phase spectrum from a CPT measurement in 2019 is shown in Figure 3.10, where the system was being set up for a measurement of ^{168}Ho and its isomer ^{168m}Ho . During this campaign, even with the MR-TOF and BNG operating as per usual, a number of close-massed contaminants still got through. At the beginning of the measurement, a considerable amount of allotted beam-time was spent identifying the ω_c of each species in the beam, corresponding to each spot. The need for this and the technique used will be discussed at length in Chapter 5. The cyclotron frequencies of five of the spots corresponded to that of $^{168}\text{Ho}^{2+}$, $^{168m}\text{Ho}^{2+}$, $^{168}\text{Dy}^{2+}$, and two molecules $^{150}\text{PrH}_2\text{O}^+$ and $^{151}\text{CeOH}^+$. Three spots could not be identified from their frequencies and are annotated as X_i ($i = 1, 2$ and 3) in Figure 3.10. Half lives greater than 1 s [104] for both the Ho states allowed us to use a $t_{acc} = 369.275$ ms, where all the beam components are resolved albeit on different turns. This measurement yielded an uncertainty of 3.2 keV. While it paints a “Hurray!” for PI-ICR, it also points to a dire picture of what it could be without the MR-TOF. In that case, there would have been many more species in the beam, leading to a

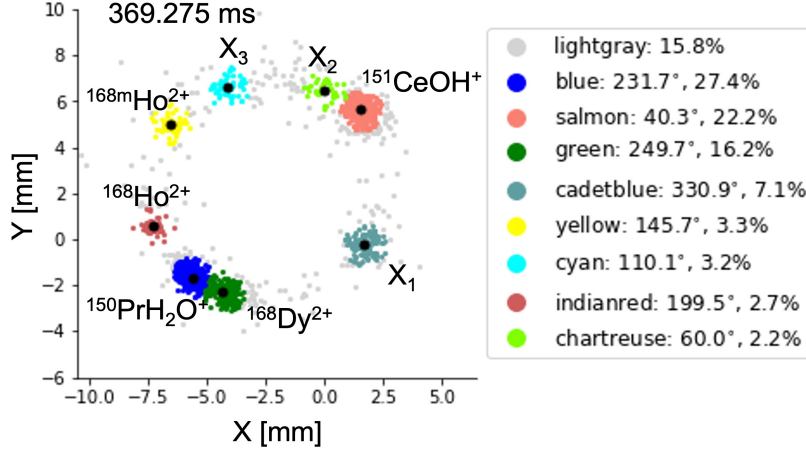


Figure 3.10 : A PI-ICR final phase spectrum of $^{168}\text{Ho}^{2+}$ and $^{168m}\text{Ho}^{2+}$ with multiple contaminants. The different colored clusters denote the different beam species.

saturation of the spectrum, overlapping of spots, and potentially faulty results.

3.4.2 MR-TOF theory, controls, operation and performance

The theory behind the MR-TOF operation is straightforward: ions of different species (denoted by i) passing through some potential difference (V) are accelerated to different velocities (v_i) depending on their q_i/m_i ratio:

$$v_i = \sqrt{2V (q_i/m_i)} , \quad (3.4)$$

As a result, over a fixed time t , they would travel a slightly different distance d_i :

$$d_i = \sqrt{2V (q_i/m_i)} t . \quad (3.5)$$

Therefore, increasing the flight time t would increase the separation between the different species over their traversed distances, and hence resolve them. This can be done by having a beam path, hundreds of metres long. An alternate and efficient solution is to reflect the ions back and forth within a fixed space, multiple times, to effectively provide a much longer path, which is what happens inside an MR-TOF. The technique implemented is illustrated with a cartoon in Figure 3.11. An ion bunch consisting of multiple species exits the elevator (post RFQ buncher) with energy ~ 3 keV and enters the MR-TOF (Step 1). The six mirror electrodes at the two ends are set in a symmetric configuration. Electrode

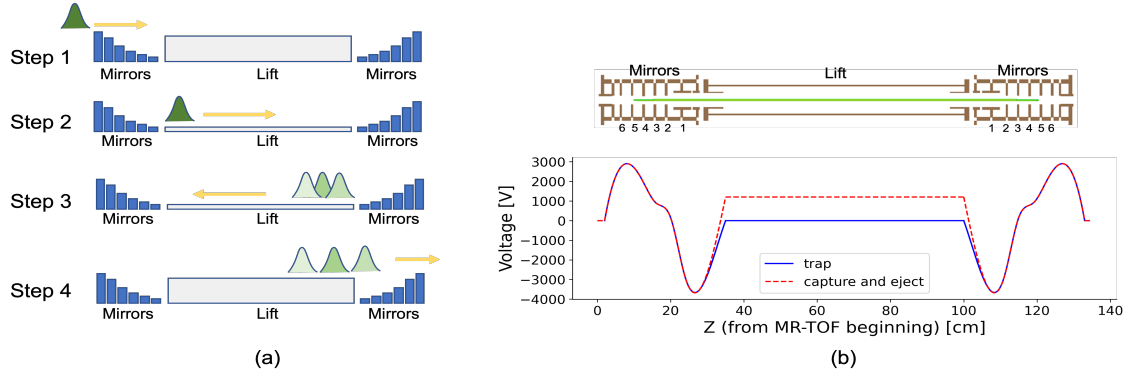


Figure 3.11 : In (a), a cartoon illustrating the lift capture method implemented at the CARIBU MR-TOF is presented. For description, see text. Panel (b) demonstrates a typical voltage setting for the different electrodes. The lift electrode's voltages are in red (during capture and ejection of the ion bunch) and blue (during trapping of the ion bunch), while the mirrors' unchanged potential is shown in purple (superposition of blue and red). A simplified schematic diagram of the MR-TOF, along with the path of an ion bunch in green, is also shown for reference.

6 is typically biased at ~ 2.9 kV, which is slightly below the ions' energies, and has the maximum potential of all the mirror electrodes. This allows the ions to clear the first set of mirror electrodes and reach the central lift electrode. At this point, the lift is pulsed typically from ~ 1100 V to ground, lowering the ions' energies and hence imparting slightly different velocities based on their charge-to-mass ratio (Equation 3.4). This is shown in Step 2. The lift is kept at 0 V as the ions go back-and-forth, getting reflected multiple times from the mirror electrodes (Step 3). This continues for a user-defined time, during which the ions traverse slightly different distances (Equation 3.5) and get resolved on the basis of their q/m . Once the necessary resolution is achieved, the lift electrode is raised back to ~ 1100 V at an appropriate time and the ions exit the MR-TOF as separated species, with the same energies with which they entered the MR-TOF (~ 3 keV). This is shown in Step 4. This technique of capturing, trapping and ejection using just the lift electrode is known as the lift-capture method. One big advantage of this method is that once the MR-TOF electrodes are tuned at the beginning of a measurement campaign, all that needs to be scaled with change of masses are the lowering and raising times of the lift electrode. Typical voltages used for the CARIBU MR-TOF are shown in Table 3.2.

One ISEG DPS series and six ISEG DPS-mini series power supplies are used to bias the seven MR-TOF electrodes. Electrodes 1 and 2 use two DPS-mini 4 kV, 2 mA power

Table 3.2 : Typical settings for the CARIBU MR-TOF used in this work

Electrodes	Biases [V]	Source
1	-3693.0	ISEG DPS-mini with 4 kV, 2 mA, and negative polarity
2	-2836.0	
3	194.0	ISEG DPS series with 500 V, 10 mA, and reversible polarity
4	866.25	
5	2179.2	ISEG DPS-mini with 4 kV, 2 mA, and positive polarity
6	2920.3	
Lift (high)	1107.0	ISEG DPS-mini with 4 kV, 2 mA, and positive polarity, through a 4 kV push-pull Behlke switch
Lift (low)	0	Grounded terminal in the Behlke switch

supplies with negative polarity, electrode 3 gets biased by the DPS series 500 V, 10 mA with reversible polarity, and electrodes 4, 5 and 6 are biased by four DPS-mini 4 kV, 2 mA power supplies with positive polarity. The lift electrode is pulsed using a 4 kV push-pull MOSFET switch, manufactured by Behlke, and equipped with three inputs: a “high” state provided by the final DPS-mini 4 kV, 2 mA positive polarity power supply; a “low” state provided by a grounded terminal; and a “control” provided by a TTL signal which commands the switching between the “high” and “low” state. An external 24 V power supply is used to provide power to all seven supplies. and a Hytec IP-DAC-8402 digital-to-analog converter (DAC), with 16 independently programmable channels, and 16-bit resolution, is used to control their output voltages. Signals between 0 to 5 V (V_{DAC}) are sent from the DAC to each supply, which get translated linearly to the output voltage of the power supplies (V_{out}) as [53]:

$$V_{out} = \frac{V_{DAC}}{5} V_{max} \quad (3.6)$$

where V_{max} is the maximum voltage output of each power supply. A Hytec IP-ADC-8411 analog-to-digital converter (ADC), with 16 channels and 16-bit resolution, is used for reading the output voltages, and accordingly sending a 0 - 5 V signal back to the CARIBU control

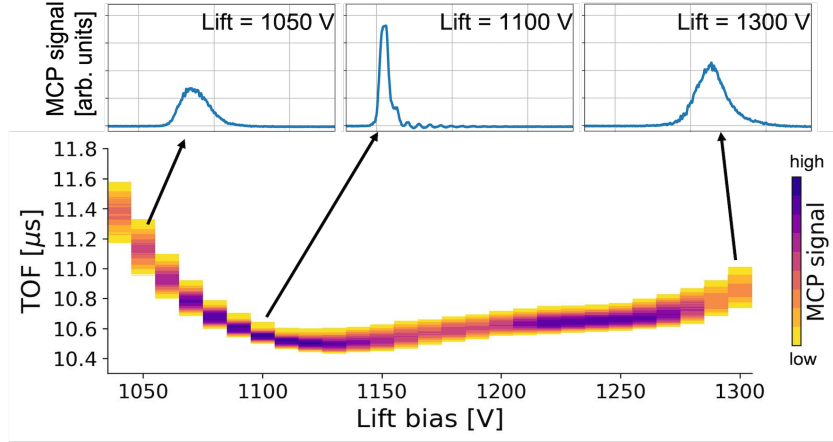


Figure 3.12 : Effect of the lift voltage on the mass resolution of the MR-TOF. The TOF of the ions at the MCP *MR-TOF* against the lift voltage is shown on the lower panel, as a heat map of the beam intensity. On the top, the MCP spectrum corresponding to three such cases are shown. The best resolution without compromising the signal strength is achieved around lift voltage 1100V, shown in the central spectrum of the top panel. Figure reproduced from Ref. [53].

system [53].

From 3.5, the mass resolving power of an MR-TOF can be derived as:

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t} \quad (3.7)$$

where t is the MR-TOF cycle duration and Δt is the FWHM time spread of each ion species upon exit from the MR-TOF. Δt and hence R depends on minimizing the spread in the kinetic energies of the ions, and the mirrors do not contribute to that spread. With the pulsed lift electrode in the lift capture method, we have the ability to precisely control the kinetic energy of the ions inside the MR-TOF, such that their temporal and spatial spread converge on the BNG (described in Section 3.4.3), which maximises the resolving power [53, 101]. Figure 3.12 shows the effect of the lift voltage on Δt for a beam of $A = 150$, $q = 2+$ ions. At the bottom of Fig. 3.12, a colormap of the intensity of the signal at the MCP *MR-TOF* is shown, with the peak's TOF plotted against the lift voltage. On the top of Fig. 3.12, the MCP spectrum corresponding to three such cases are shown. As can be seen in this case, the best resolution without compromising the signal strength is achieved around lift voltage 1100 V, shown on the central spectrum of the top panel of Fig. 3.12.

An optimized lift voltage can minimize the Δt and focus the beam which, once fixed, makes R , more or less, linearly dependant on the cycle time (t). Figure 3.13 illustrates the

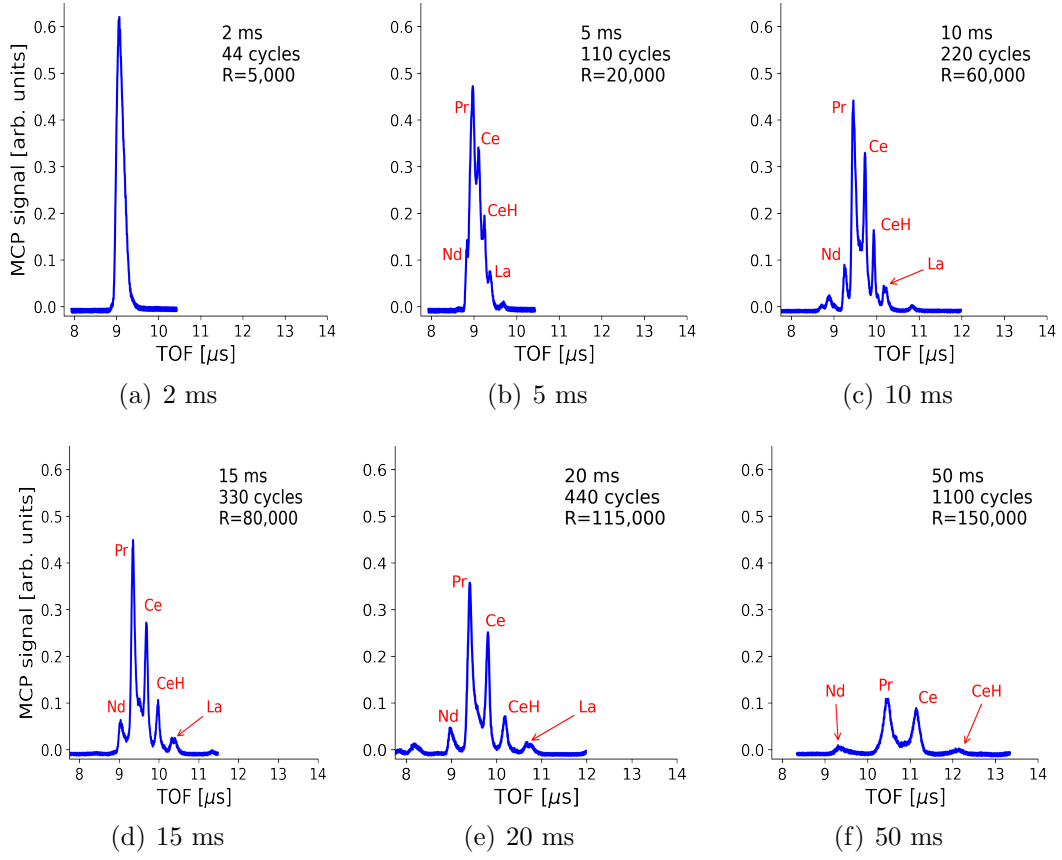


Figure 3.13 : Mass separation by the CARIBU MR-TOF for a beam of $A = 150$, $q = 2+$ ions with increasing cycle time.

operation of the MR-TOF for a beam of $A = 150$, $q = 2+$ ions. The different subfigures indicate spectra recorded at the MCP *MR-TOF* for six different cycle times inside the MR-TOF. Fig. 3.13(a) is the MCP spectrum after the ions spend 2 ms inside the MR-TOF, when the beam components are not separated and hence manifest as one peak. With 5 ms inside the MR-TOF, the beam species could be seen starting to separate from each other (Fig. 3.13(b)). The subsequent subfigures (Fig. 3.13(c) to 3.13(f)) show the beam species getting separated with increasing t , as expected. The peaks were identified first by comparing their masses to their TOFs and later using PI-ICR at the CPT (described in Chapter 5). However, increasing the cycle time t means a higher chance of space-charge effects disrupting beam transmission, or the ions colliding with the residual gas particles inside the MR-TOF and getting lost, or both. This can be seen in Fig. 3.13, where the loss in transmission with increasing t can be seen with the decrease in the signal strength. In other words, obtaining a better resolution with increased t can come at the cost of a loss in transmission. The pressure inside the MR-TOF plays an important role in this regard and is hence kept low at $\sim 10^{-9}$ Torr. To compare the MR-TOF's resolution and transmission for increasing cycle time, the beam intensity was measured at the Si *buncher-down* and Si *MR-TOF* as the MCP spectra of Fig. 3.13 were acquired [53]. This is shown in Fig. 3.14. A mass resolving power of 150,000 can be achieved with the CARIBU MR-TOF with a cycle time of 50 ms, the price of that being a transmission efficiency of only 15%. This significant loss of transmission can also be seen in Fig. 3.13(f) where the La is barely visible. For most CPT measurement campaigns, the cycle time is kept between 10–20 ms, which allows for the realization of a mass resolving power of $\sim 100,000$ and a transmission efficiency of 30-40%.

3.4.3 Bradbury-Nielsen gate

A mass resolved beam, similar to Fig. 3.13(c) to 3.13(e) exits the MR-TOF. In order to send a clean beam to the CPT, the desired species needs to be selected and the unwanted species blocked need to be removed. This is done by a Bradbury Nielson Gate (BNG)[105], located at the focal plane of the beam exiting the MR-TOF, in the same diagnostic cross feedthrough as the Si and the two MCP *MR-TOFs*. The BNG consists of wires in a grid like arrangement, perpendicular to the direction of the beam. The BNG can be operated in two

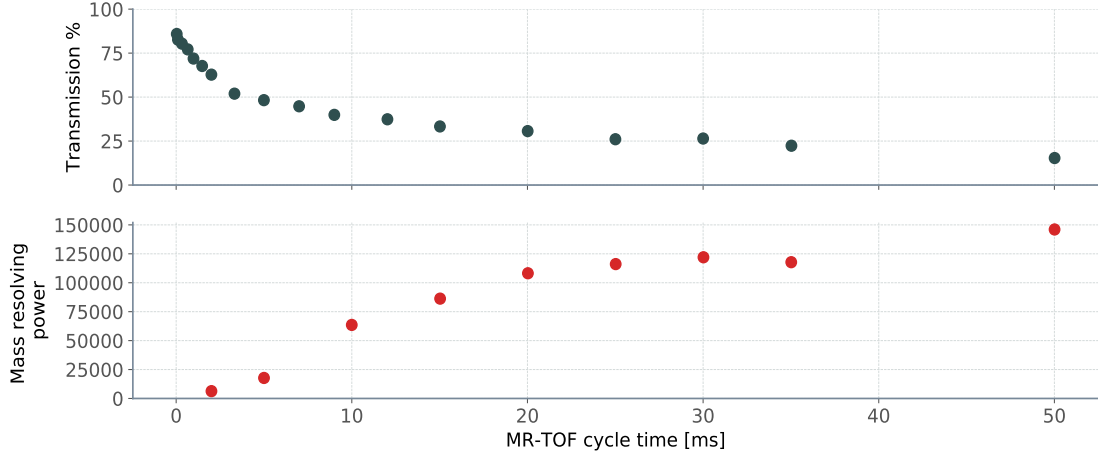


Figure 3.14 : Resolution and transmission of the CARIBU MR-TOF with an $A=150$, $q = 2+$ beam for different cycle times. Figure taken, with permission, from Ref. [53]

states: “open” and “closed” as shown in Figure 3.15(a). In the “open” configuration, the wires are grounded and the beam passes uninterrupted through the BNG. In the “closed” state, alternate wires are biased with oppositely phased voltages ($\pm V_b$) such that they create a strong localized electric field which deflects the ions in the beam. This deflection angle (θ) from the original beam direction can be found from [106]:

$$\tan \theta = \frac{\pi}{2 \ln[\cot(\pi R/2d)]} \frac{V_b}{V_0} , \quad (3.8)$$

where R is the radius of the wires, d is the distance between them and V_0 is the electric potential of the ions as they pass through the BNG.

The precise timings to induce the “open” and “closed” states are critical to the BNG operation. From Equation 3.4, ions having different m/q ratios will take different times to travel fixed distances:

$$t_i = \sqrt{m_i/(2Vq_i)} d , \quad (3.9)$$

where t_i is the time taken by the i -th beam species to travel a fixed distance d . If d is thought to be the sum of the total flight path of the ions inside the MR-TOF as they undergo multiple reflections and the distance between the MR-TOF and the BNG, the ions will pass through the BNG at slightly different times, as given by Equation 3.9. Therefore, if an “open” state

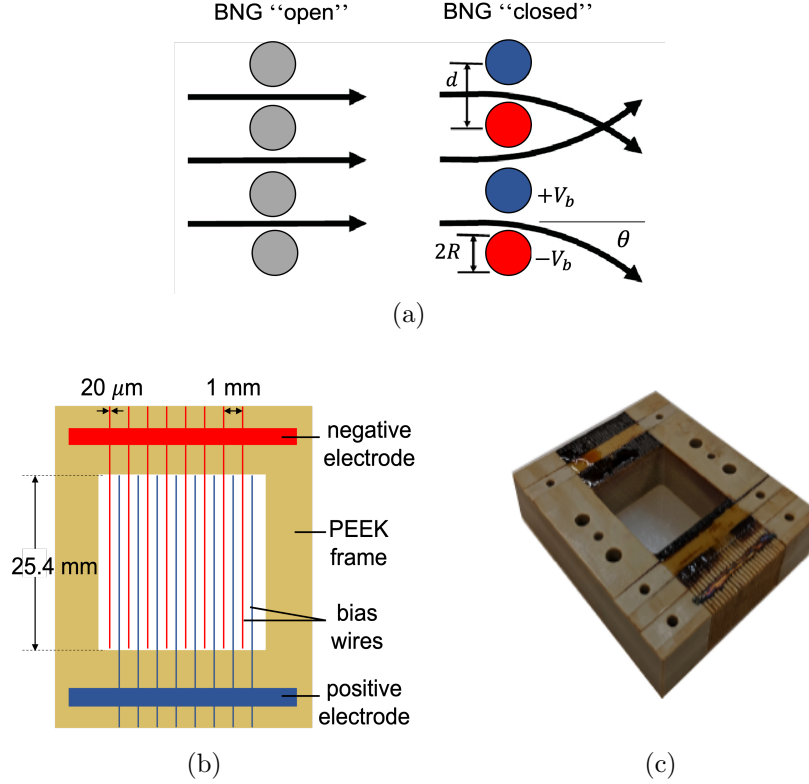


Figure 3.15 : Panel (a) shows the basic theory behind the BNG. The wires are denoted as the colored circles: grey symbolizing grounded and blue and red representing biases of $+V_b$ and $-V_b$ respectively. The wires have a diameter of $2R$ and d distance apart from each other. The black arrows represent the ions' path, straight or deflected by θ . Figure reproduced from [106]. In panel (b), a schematic diagram of the CARIBU BNG, with all the necessary dimensions, is shown. Panel (c) is a picture of the CARIBU BNG.

is induced on the BNG for the precise period when the ions of interest pass through it, and the gate is kept at a “closed” state at all other times, most isobaric contaminants in the beam would be removed.

The CARIBU BNG consists of gold-plated tungsten wires $20\ \mu\text{m}$ wide and $0.5\ \text{mm}$ apart, held in a polyether ether ketone (PEEK) frame with a $25.4\ \text{mm}$ square gap in the middle, as shown in Figure 3.15(b). The wires are connected to two pulser modules, similar to the MR-TOF lift switch, except these modules are designed to operate at comparatively lower voltages of $\leq 400\ \text{V}$. The “low” input of one module and the “high” input of the second module are grounded. $+300\ \text{V}$ is applied to the “high” input of the first module, while $-300\ \text{V}$ is applied to the “low” input of the second module. Both modules accept a common TTL control input. The outputs of these pulser modules are connected to the positive and negative electrodes of the BNG respectively. The modules are operated in *inverse* and *normal* mode, such that the wires stay biased at $\pm 300\ \text{V}$ and get pulsed to ground during the high TTL pulse, which is tuned to the time of passing of the species of interest through the BNG, thereby allowing the ions of interest to pass through while gating out the contaminants. Putting $d = 1\ \text{mm}$, $2R = 20\ \mu\text{m}$, $V_b = 300\ \text{V}$ and $V_0 = 3\ \text{kV}$ into Equation 3.8, $\theta \approx 2^\circ$, which is enough to induce BNG “closed”.

Once the voltages of the MR-TOF lift and mirror electrodes, and the cycle time are optimized, the location of the different peaks corresponding to the different isobars in the beam are identified at the MCP *MR-TOF* on the basis of their TOF (Fig. 3.13(c) to Fig. 3.13(e)). The BNG is then timed to allow the species of interest to pass through to the CPT system (Chapter 4), while the rest of the isobars are gated out. The gate-open period at the BNG is usually set with an initial $\sim 1\ \mu\text{s}$ and then fine tuned. The minimum width that can be set is $250\ \text{ns}$, limited by the stability of the pulser modules. It is typically set at $300\ \text{ns}$.

3.4.4 MR-TOF stability

Once the timing and width of the BNG “open” mode, which are independent of any drift inside the MR-TOF, are set, the stability of MR-TOF becomes of paramount importance. This is illustrated in Figure 3.16. A beam of $A = 150$, $q = 2+$ ions, after spending 10

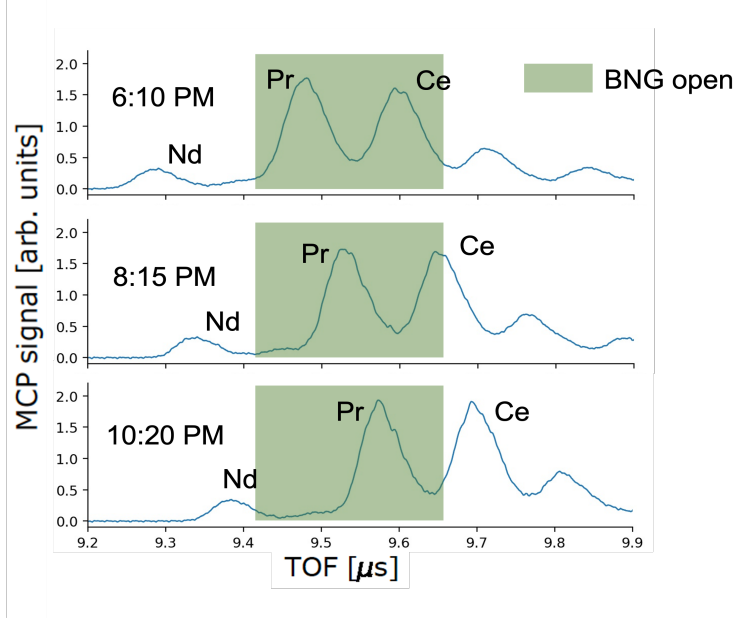


Figure 3.16 : Signal from the MCP *MR-TOF* for an $A = 150$, $q = 2+$ ion beam at three different times on the night of October 23, 2019. The green rectangle illustrates a theoretical BNG *open* state.

ms in the MR-TOF, was monitored over 15 hours at the MCP *MR-TOF* on October 23, 2019, with the spectra being recorded every 3 s. Three such spectra are shown in the Figure 3.16, with the Nd, Pr and Ce peaks annotated. At 6:10 PM, if the BNG was positioned to allow the Pr and the Ce peaks to go through (shown in green), by 10:20 PM, it would be letting mainly the ^{150}Pr . This means during a measurement campaign, an experimenter will have to monitor the beam occasionally and, if the TOFs of the different beam constituents drift, move the BNG accordingly. This could be difficult and at times impossible as checking the beam on the MCP *MR-TOF* would mean interrupting the ongoing data acquisition. Sometimes it could take hours to even notice this drift.

A study was conducted after the commissioning of the MR-TOF, where the effects of the stabilities of individual mirror electrodes on the TOF of the ions were investigated. It was observed that, while the TOF of the ions showed almost no dependence on the voltage-changes of electrodes 1-4, they were the most sensitive to the changes in the voltages applied to the electrodes 5 and 6. For electrode 5, a relative change of ± 100 ppm voltage would amount to ~ 250 ns TOF change, while the same change in 6 would result in a ~ 350 ns change in TOF [53]. The typical 300 ns BNG width means that a stability of ≤ 50 ppm would

be adequate for CPT campaigns. Another additional advantage of the lift-capture method can be seen there. Keeping the mirror electrodes fixed would ensure that this stability only depends on the stability of the corresponding power supplies. If the mirror electrodes were pulsed instead for capture and ejection, it would be difficult to maintain the same level of stability, as the stability of the high voltage (HV) switch would also come into the equation. Even if it were possible, it would be an expensive endeavour to obtain electronic modules with minimal noise, in addition to the stable HV switches.

The baseline stability of all seven power supplies is dictated by the stability of the DAC. During the commissioning of the MR-TOF, this stability was found to be around 3.5 ppm [53]. This stability, which translates to the same parts-per-million stability for the electrodes (from equation 3.6) is well within the required 50 ppm stability requirement for CPT experiments. The next parameter that could cause instabilities in the power supply outputs is temperature. The ISEG DPS and DPS mini series power supplies have a temperature coefficient of 50 ppm/K or equivalently 50 ppm/°C [107], meaning a temperature change of even 1°C could cause a significant drift in the TOF. Therefore a temperature stabilization system was set up to keep the stabilities of the outputs of the power supplies to within the required limit. The power supplies are mounted on an aluminium plate. The other side of this plate is thermally connected to a pair of Peltier elements. The Peltier elements are in thermal contact with a second aluminium plate, supported by copper pipes through which water is circulated from a water chiller. The temperature is read using a thermistor and a Newport temperature controller, model LDT-5980 [108]. If the temperature of the power supply plate deviates from a certain set value, the controller module supplies the Peltier elements with the necessary current to reverse the change and bring the temperature back to the preset point. The power supplies along with the plates and the Peltier elements sit inside a thermally insulated box, while the water chiller and the temperature controller module are located right next to the box. The thermistor probes, the copper pipes, and the control and biasing cables for the power supplies enter the box through three small holes on the side. The set-point in the temperature controller box is usually set a few degrees Celsius above the room temperature, to ensure that the temperature stabilization is done primarily by heating from the Peltier effect to minimize excess heat inside the box. Usually, the room

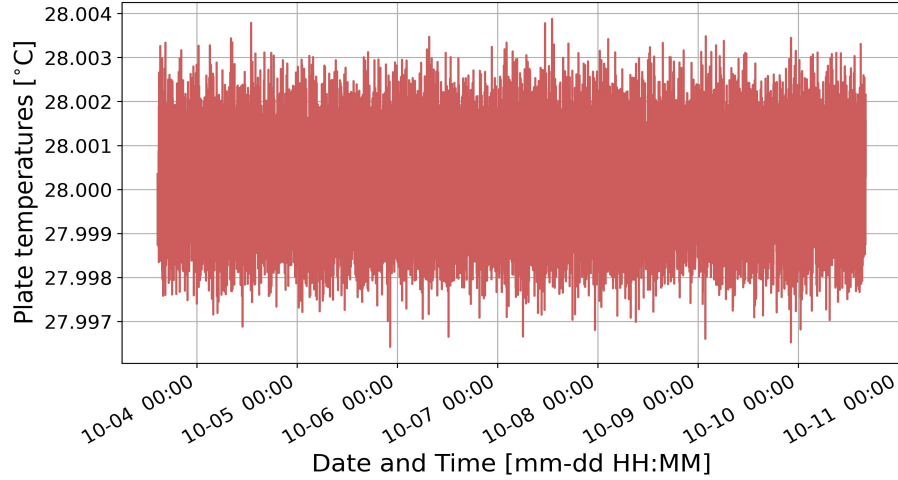


Figure 3.17 : Stability of the temperature of the plate hosting MR-TOF electrode power supplies, regulated by the temperature controller box, monitored over 6 and half days.

temperature inside CARIBU is kept at around 16-18°C, while this temperature controller box is typically set to maintain the temperature of the plates at $\sim 28^\circ\text{C}$. Any additional heat generated from the Peltier effect is removed by the cold water circulating through the copper pipes. Figure 3.17 shows the temperatures of the power supply plate over 6 and half days. Average peak to peak temperature variations of $\sim 0.005^\circ\text{C}$ around the set 28°C can be seen on the electrodes' plate. This means the temperature stabilization set-up is good enough for the power supplies to provide stable voltage to the electrodes.

In principle, the DAC supply is stable and the temperature can be regulated to ensure the power supplies' stabilities within the required limit of 50 ppm. In spite of this, if there is a drift in the TOF of the ions, the cause could be the condition of the actual power supplies. On the night of October 23, 2019 when Figure 3.16 was taken, electrodes 5 and 6 power supplies were also monitored. The outputs of the two power supplies were connected to the HV inputs of two voltage dividers of the ratio $\sim 1000 : 1$ with a temperature stability of 5 ppm/K themselves. These voltage dividers have two outputs, a HV and a low voltage. The HV outputs, which mirror the HV inputs, were connected to the corresponding electrodes so the MR-TOF would be operational. The low voltage outputs were connected to two 34401A 6.5 digit precision multimeters from Agilent and HP. These are shown in Figure 3.18, along with the drift of the TOF of the Pr peak.

Over the 15 hours, electrode 6 power supply was fairly stable to around ~ 14 ppm. For

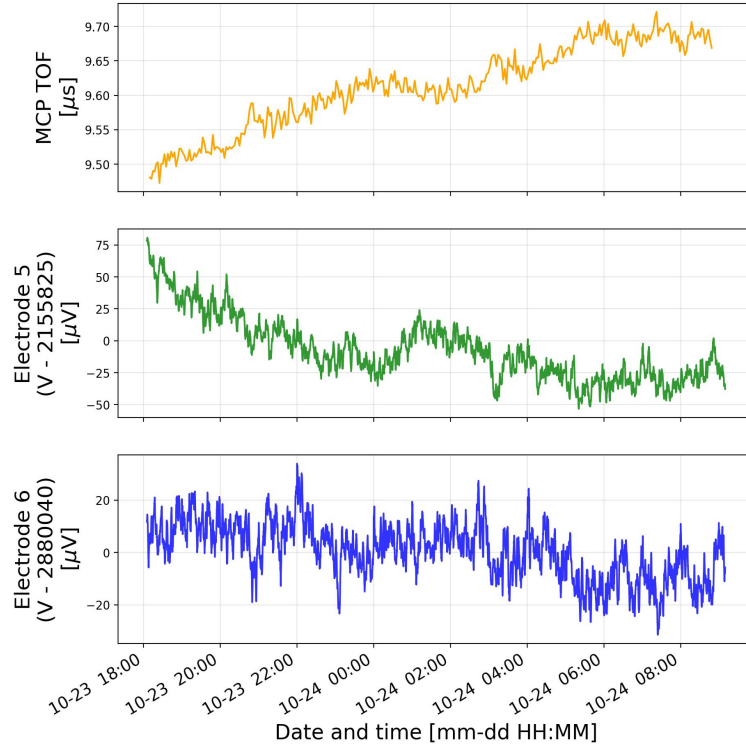


Figure 3.18 : Overnight stability monitored on the night of October 23, 2019 as Figure 3.16, showing the drifts in the TOF of the Pr peak and the biases of electrodes 5 and 6, with the temperature stabilization system in operation. The voltages on the electrodes were monitored using a voltage divider, and the applied voltage is stated in the legends.

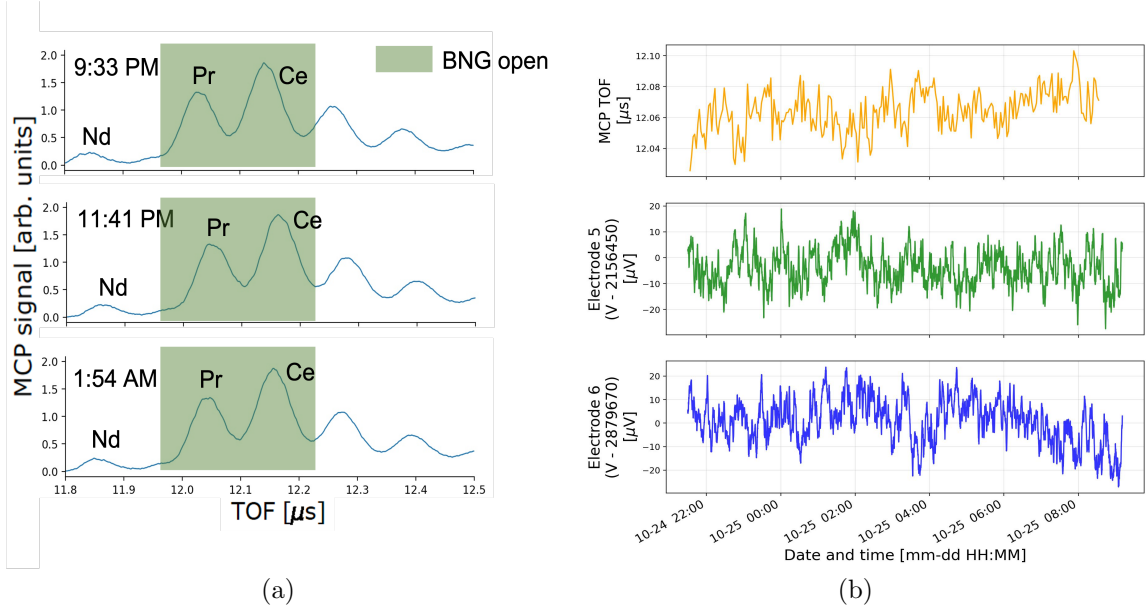


Figure 3.19 : Panel (a) shows the signal from the MCP *MR-TOF* for an $A = 150$, $q = 2+$ ion beam at three different times on the night of October 24, 2019. As in Figure 3.16, the green rectangle illustrates a theoretical BNG *open* state. Panel (b) shows the drifts in the TOF of the Pr peak and the biases of electrodes 5 and 6 on the same night. Like Fig. 3.18, the voltages were monitored using a voltage divider, and the applied voltage is stated in the legends.

electrode 5, on the other hand, a drift of around $\sim 100 \mu\text{V}$ can be seen, amounting to a stability of ~ 60 ppm. That would explain the 200 ns drift in the TOF of the Pr peak. Later that day, October 24, 2019, the electrode 5 power supply was replaced with a new one and the test was repeated the next night. This is shown in Figure 3.19. Over the ~ 12 hours, both electrodes 6 power supply and the new power supply for electrode 5 were fairly stable with fluctuations of $\sim 40 \mu\text{V}$ amounting to around ~ 14 ppm and ~ 18 ppm stability respectively. The TOF drift can be seen to be around 40 ns.

3.5 CARIBU operation and timings

A typical CARIBU experiment would start with extracting one of the tuning beams from the gas catcher. The mass number is selected on the basis of the experiment. For most of the masses presented in this thesis, which are extracted in the $2+$ state, the tuning beam used was $^{150}\text{Ce}^{2+}$. For species that exit the gas catcher with charge $1+$, a beam of $^{142}\text{Cs}^+$ was used. Usually, the last saved settings for that isotope ($^{150}\text{Ce}^{2+}$ or $^{142}\text{Cs}^+$) are loaded. Then the beam steerers and lenses are tuned to maximize transmission.

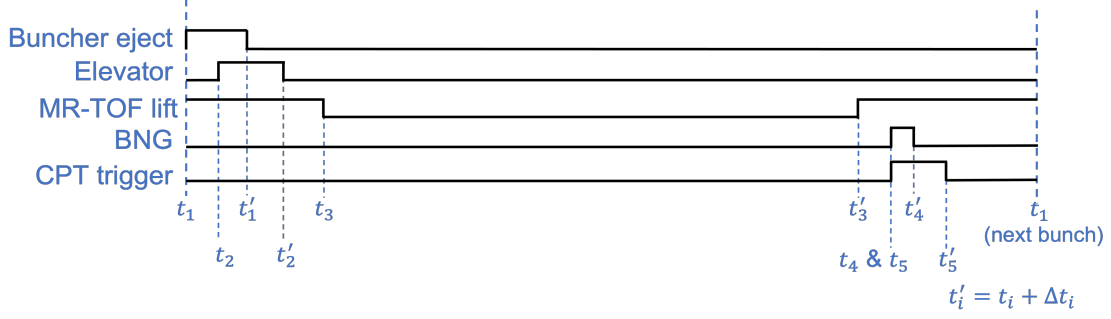


Figure 3.20 : Summary of CARIBU timings. The start and end of different functions are denoted by t_i and t_i' , ($i = 1$ to 5), such that $t_i' = t_i + \Delta t_i$. t_i and Δt_i are set by the user from the control system. The timings are not to scale.

Downstream from the buncher, along with steering and bunching, precision timings become an absolute necessity and need to be tuned. All the timings are set by an 8-channel digital delay generator from Quantum Composers (model 9518) with 1 ns timing resolution, 0.1 ps precision, and jitter < 400 ps [109]. All the 8 channels can be controlled independently from the CARIBU control system. A timing diagram is shown in Figure 3.20. The start and end of each function trigger are denoted by t_i and t_i' . In reality, the user sets the delays t_i s and the widths Δt_i s such that $t_i' = t_i + \Delta t_i$.

The buncher ejection (t_1) is the trigger for all CARIBU signals. As mentioned earlier, it is repeated every 50 or 100 ms as set by the user. After time t_2 from the buncher ejection, when the ion bunch has entered the elevator, the elevator pulse is triggered. About time t_3 post the buncher ejection trigger, the ions are ready for injection into the MR-TOF and have cleared the first set of MR-TOF mirror electrodes. At this time, the Behlke HV switch controlling the MR-TOF lift is triggered. This pulse is inverted and applied to the switch such that it lowers the lift potential for the width of the pulse (Δt_3). In other words Δt_3 determines how long the ions stay inside the MR-TOF, and hence the achievable resolution. At the start of an experiment, an approximate rough time is input for Δt_3 (e.g 20 ms). After that, it is slightly changed in fine steps of ~ 500 ns looking at one of the two MCP *MR-TOF*s until the ions' peaks can be seen, i.e. the MR-TOF timing is tuned such that the lift electrode is pulsed up when the ions are on their way out of the MR-TOF after an integer number of cycles. In principle, a range of time for Δt_3 , equal to half of a cycle time will ensure that the lift electrode is raised when the ions are travelling downstream in the

right direction. However, it is usually fine-tuned such that the ions are at the middle of the lift when the lift voltage is raised. The BNG timing t_4 is tuned to let the isotope of interest go through. The duration of this gate-open period (Δt_3) is usually set on the basis of the resolution achieved and the separation of adjacent peaks at the MCP *MR-TOF*. As mentioned in Section 3.4.3, the minimum Δt_3 is 250 ns. Finally, a separate signal mirroring the trigger of the BNG is sent to the CPT system as a trigger. A typical set of timings for a beam of $^{150}\text{Ce}^{2+}$, with a 20 ms MR-TOF cycle, is shown in Table 3.3.

Table 3.3 : Typical CARIBU timings for a $^{150}\text{Ce}^{2+}$ beam with a 20 ms MR-TOF cycle.

i	Labels	Pulse triggers (t_i)	Pulse widths (Δt_i)
1	Buncher eject	0.00 μs	15.00 μs
2	Elevator	6.70 μs	14.00 μs
3	MR-TOF lift	28.00 μs	20.01750 ms
4	BNG	20.05150 ms	0.30 μs
5	CPT trigger	20.05150 ms	5 μs

Once a considerable amount of ion-beam with good mass resolution can be seen at the Si and MCP *MR-TOF* respectively, it is sent to the CPT system. The timings, and the voltages to the steerers and lenses in the CPT system are tuned to maximize transmission and quality of PI-ICR spots. Then, the beam species are all identified by their cyclotron frequencies or equivalently their masses. These will be discussed in the next two chapters and 4 and 5. Next, the magnetic field of the CARIBU isobar separator and all the timings for the beam-line elements after the buncher are scaled from the tuning mass to the target mass. The scaling is done by calculating a scaling factor equal to the ratio of the m/q of the tuning isotope to the target isotope. Since the ions' energies, set by the deck voltage and the elevator stay unchanged, the magnetic field (B) and the timings (t_i and Δt_i) can be scaled as:

$$\frac{B_a}{B_b} = \sqrt{\frac{m_a/q_a}{m_b/q_b}} \quad (3.10a)$$

$$\frac{(t_i)_a}{(t_i)_b} = \sqrt{\frac{m_a/q_a}{m_b/q_b}} \quad (3.10b)$$

where a and b are the beam species after and before the scaling. Of the timings (triggers and widths), the buncher ejection trigger and width (t_1 and Δt_1), elevator width (Δt_2) and the CPT trigger width (Δt_5) are not scaled and kept fixed at the values given in Table 3.3. Once the mass of an isotope has been measured, the system is scaled again to the next target nuclide using equations 3.10a and 3.10b.

4 The Canadian Penning trap system

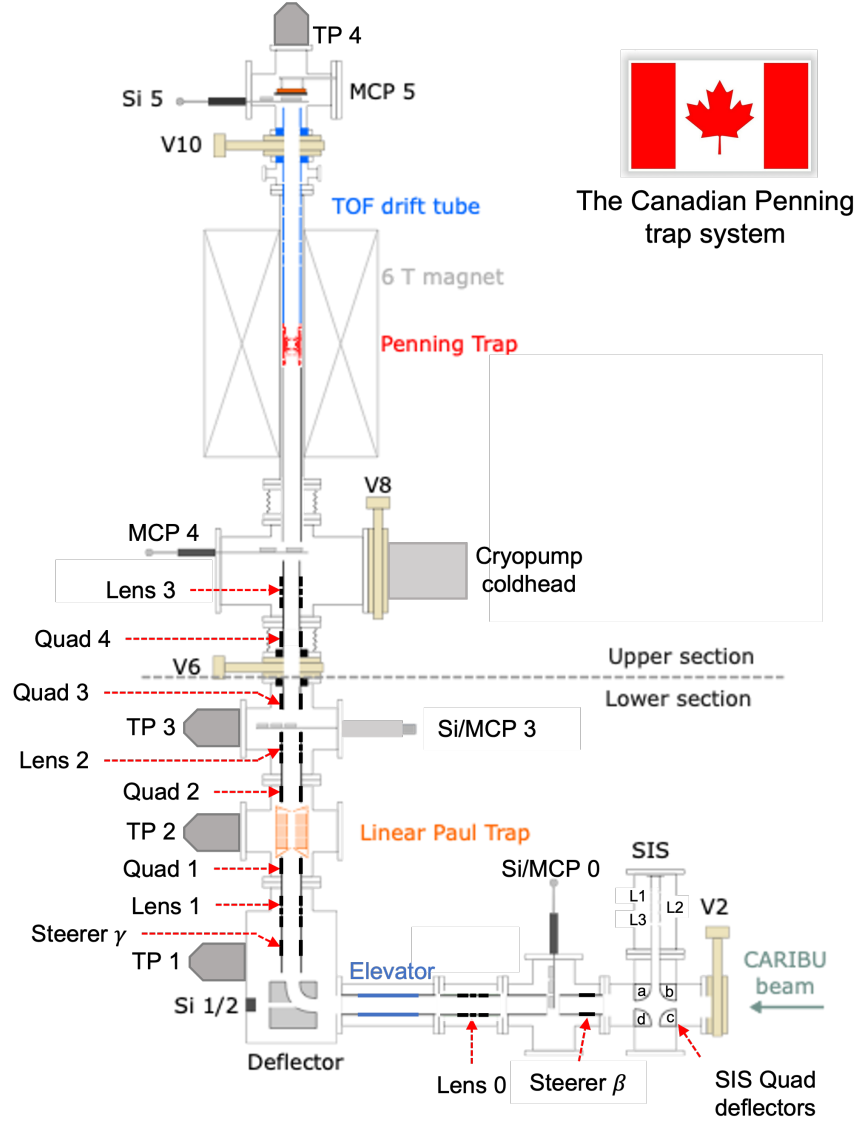


Figure 4.1 : Schematic diagram of the CPT tower (not to scale), showing the major components. Figure originally drawn by R. Orford, and the different beamline components like steerers and lenses, and vacuum pumps were added by this author.

The CPT system was designed and commissioned at the Tandem Accelerator Superconducting Cyclotron (TASCC) facility at Chalk River Laboratories in Ontario, Canada [79] in the late 90s. Upon the decommissioning of TASCC, the CPT was moved to the ANL and was set up in the “Triangle Room” experimental area in the ATLAS facility [110] in 2001. Upon the completion of CARIBU in 2009, it was moved there and took the first beam in 2011 [68]. After undergoing numerous upgrades since then, currently the CPT system mainly consists

of an elevator and a linear Paul trap used as a buffer-trap, constituting the lower section, and the Penning trap (CPT), used as a precision-measurement-trap, equipped with a position sensitive MCP detector (PS-MCP, also known as MCP 5) for PI-ICR measurements, making up the upper section, in a ~ 4.5 m *tower* structure as shown in Figure 4.1. The system also includes an offline source, Stable Ion Source (SIS) located at the bottom of the tower, and a host of electrostatic beam-line components like steerers (β , γ , Quad 1–4), einzel lenses (Lens 0-3) and drift tubes to guide the ions along the tower from the end of CARIBU to the PS-MCP. The drift tubes are not shown in figure 4.1 to avoid clutter.

There are five diagnostic stations located at strategic places in the tower. Each CPT diagnostic station consists of some combination of Si and MCP detectors, and a drift tube for the beam to pass through. The two most commonly used ones are Si / MCP 0 and Si / MCP 3, located at the bottom of the tower just before the elevator and downstream from the linear Paul trap respectively. Si / MCP 0 gives us an indication about beam transmission from CARIBU. MCP 0 is also used to get a starting point in tuning the elevator voltage and timing. Si / MCP 3 is primarily used to optimize capture in the linear trap. Si 1/2, located at the 90° deflector, can be used to test the tuning of the elevator by allowing the beam to pass through the deflector. MCP4 can be used to check the time-spread of the ion bunches before they enter the Penning trap. Further upstream, Si 5 can be used to check transmission efficiency between the two traps. Si 1/2, MCP 4 and Si 5 are rarely used during routine operations, but are useful for diagnosing and troubleshooting problems.

Three turbo molecular pumps (TP 1-3), located at the deflector, the linear Paul trap and the Si/MCP 3 cross, and backed by two roughing pumps, are used in the lower section to maintain pressures ranging from ~ 1 μ Torr to ~ 100 nTorr. For precision measurements, the upper section needs to be kept at ultra-high vacuum (UHV, < 1 nTorr). This is achieved by one turbo molecular pump (TP4), located at the top of the tower above MCP 5, backed by a dedicated roughing pump, and a cryopump attached to a coldhead next to the valve V8. V8 lets us isolate the cryopump during out-gassing of the cold head. The upper and lower sections are separated by valve V6, allowing one to install components and do other maintenance work in each section, without breaking vacuum in the other section. The upper section, because of the UHV conditions, takes at least 2-3 days to return to operational

mode once exposed to air. Therefore, it is rarely vented, other than to change TP4 or do maintenance works on the PS-MCP. V10 separates the Penning trap from the PS-MCP and TP4, meaning the Penning trap is almost always under vacuum. The whole system can be isolated from CARIBU by closing valve V2.

The main power supply used to bias most beam line components, except the Penning trap, is a CAEN SY-2527 universal multichannel power supply system [111] with four negative 4 kV cards, one positive 4 kV card and one low volt (< 100 V) card with user-defined polarity for each channel. Each card has 12 independently programmable channels. Quad steerers and focussing elements are controlled by two custom made low voltage supplies capable of providing voltages up to ± 200 V on top of an externally applied float HV. This external HV is provided from the CAEN. Other low voltages like heating the SIS source, supplying power to the elevator HV switch and running a liquid nitrogen (LN_2) pump to keep the Paul trap cold are applied using three low voltage supplies from BK Precision (model 1550). The critical trapping potentials required by the Penning trap are provided by a dedicated ultra high precision voltage supply from Stahl Electronics (4 channel, ± 14 V BS Series) [112], referred to as the Stahl for the rest of this thesis. The Stahl will be discussed in further detail in Section 4.4.

4.1 Offline source (SIS)

The Stable Ion Source (SIS) is located at the bottom of the tower between the valve V2 and the Si / MCP 0 diagnostic station. It consists of a mixed-alkaline, surface-ionization ion source (Model 101139) from Heat Wave Labs [113] containing salts of elements Cs, Rb and K in roughly equal proportions. The source is attached to a pair of coaxial heating elements and three grounding pins, as shown in Figure 4.2(a). Currents of $\sim 1.10 - 1.30$ A is applied, from one of the mentioned BK Precision power supplies, to the heating elements. The SIS produces continuous ion streams of $^{133}\text{Cs}^+$, $^{85}\text{Rb}^+$, $^{87}\text{Rb}^+$ and $^{39}\text{K}^+$. The mass range of these three beams help to tune and optimize the CPT system for different mass-regions. Additionally, the masses of these isotopes are known to 0.1 ppb [30], and are used as reference species to calibrate the Penning trap magnetic field during a mass measurement campaign.

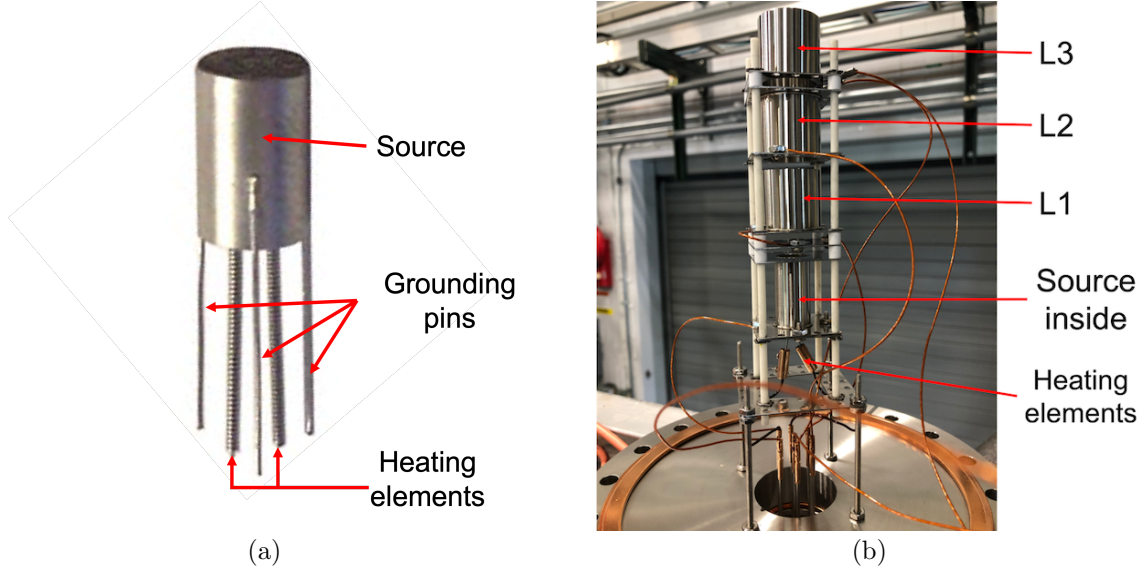


Figure 4.2 : Stable Ion Source (SIS). Panel (a) shows a picture of the Stable Ion Source with the grounding pins and heating element. Picture taken from [113]. Panel (b) shows a picture of the SIS assembly at the CPT along with the extraction electrodes.

The ions from the source are extracted by the extraction electrodes L1 and L2 maintained at -100 V and -350 V respectively, and then accelerated to 1.49 keV by L3. The assembly of the source and electrodes L1, L2 and L3 is shown in picture 4.2(b). This structure is rotated upside down and put in place, as can be seen in Figure 4.1. The ions next approach the SIS Quad deflectors, consisting of four electrodes (“a”, “b”, “c” and “d”), where electrodes “a” and “c” are biased at -2400 V, while -600 V is applied to “b” and “d”. These electrodes deflect the ions into the main beam line as a DC beam.

4.2 CPT elevator

Ion bunches from CARIBU have a kinetic energy of ~ 3 keV when they enter the CPT system. The linear Paul trap and the CPT are designed to operate at low voltages and to trap and manipulate ions with near-ground energies, so the CARIBU-beam energy needs to be lowered first, which is done with the CPT elevator. The SIS Quad deflector electrodes, along with steerer β , and lens 0 electrodes are maintained at ground potential to guide the ions from CARIBU towards the CPT elevator unperturbed. The elevator tube consists of a 30 cm long tube, flanked by two shorter drift tubes, the one upstream maintained at ground, while the one downstream maintained at -1490 V. When the ion bunches enter the elevator,

its voltage is pulsed from ground to ~ -3 kV. Therefore, as the ions leave the elevator, they pass through the -1490 V drift tube, which accelerates them to 1.49 keV. The pulsing is done with another 4 kV MOSFET switch from Behlke, similar to the one used for the MR-TOF lift.

The voltage ($V_{\text{Elev.}}$) and trigger timing ($t_{\text{Elev.}}$) of the CPT elevator are tuned at the beginning of every campaign. The velocity of the ions coming out from CARIBU can be calculated by measuring the TOF of the ions on the MCP *MR-TOF* ($\text{TOF}_{\text{MR-TOF}}$) and MCP 0 (TOF_0) with respect to some common trigger like the MR-TOF lift or the BNG:

$$v_{\text{ions}} = \frac{D_1}{\text{TOF}_0 - \text{TOF}_{\text{MR-TOF}}} , \quad (4.1)$$

D_1 being the distance between the MCP *MR-TOF* and MCP 0, and measured to be around 5.26 m. From v_{ions} , the ions' energy (E_{ions}) is calculated by:

$$E_{\text{ions}} = \frac{1}{2} m_{\text{ions}} v_{\text{ions}}^2 \quad (4.2)$$

where m_{ions} is the ions' mass and can be approximated to the mass number A for this purpose. Once E_{ions} has been calculated, the elevator voltage will be:

$$V_{\text{Elev.}} = E_{\text{ions}}/q . \quad (4.3)$$

Since the trigger for all the components in the CPT tower starting with the CPT elevator is initiated by the “CPT trigger” signal which mirrors the BNG signal, $t_{\text{Elev.}}$ can be found as:

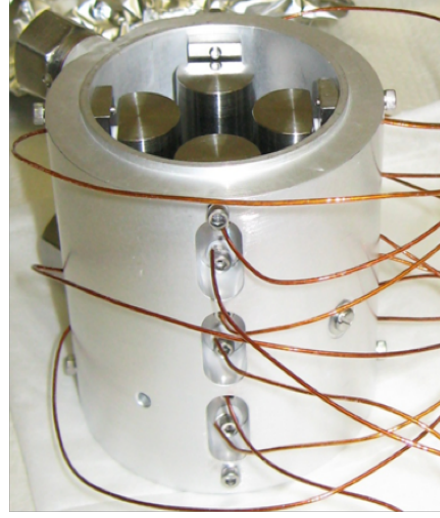
$$t_{\text{Elev.}} = \text{TOF}_{\text{MCP } 0}^{\text{BNG}} + \frac{D_2}{v_{\text{ions}}} \quad (4.4)$$

where D_2 is the distance between MCP 0 and the elevator, measured to be around around 0.4 m, and $\text{TOF}_{\text{MCP } 0}^{\text{BNG}}$ is the TOF of the ions at MCP 0 with respect to the BNG or equivalently the CPT trigger.

$V_{\text{Elev.}}$ and $t_{\text{Elev.}}$ gives one a good starting point to tune the elevator, which is then fine-tuned looking at the ion rate at Si / MCP 3 with the linear trap being operated in not-capturing or fly-thorough mode. This will be discussed in the next section (Section 4.3).



(a) Top view of the linear Paul trap. Picture taken from [57].



(b) Side view of the linear Paul trap. Picture taken from [71].

Figure 4.3 : The linear Paul trap in the CPT system.

Post the elevator, the ions are deflected by 90° upwards towards the Paul trap by a pair of spherical, electrostatic deflector electrodes biased at ~ -1800 V and ~ -1200 V.

Ions from the SIS are extracted at an energy of 1490 eV and do not require the implementation of the elevator. For these ions, the electrodes of the elevator are set to a constant value of -1490 V, and operated as a drift tube.

4.3 Linear Paul trap

After the 90° deflector, the ions pass through steerers γ and Quad 1, Lens 1, and a number of drift tubes before entering the linear Paul trap (LT). Tuning the Penning trap can be a time-consuming affair, and is done offline using the SIS before any measurement campaign. To make the Penning trap settings robust, and independent of the source and beam-extraction settings, the LT is operated as a buffer trap. It consists of four segmented rods and two end cap electrodes (top and bottom). Each rod has three segments, named S1, S2 and S3. The rods are 19 mm in diameter. Each segment is 20.25 mm in length. There is space of 16 mm between the opposite rods. The entire structure including the end caps is 96.5 mm long and 60 mm wide, with two 6 mm apertures on either side for the entry and exit of the ions. The bottom end-cap electrode also acts as a deceleration electrode to slow

down the ions before they are captured. Both the bottom and the top end caps ensure that the ions with slightly higher energies do not escape from the trap and enable us to capture multiple ion bunches from CARIBU.

Table 4.1 : Typical DC biases applied to the linear Paul trap electrodes

Electrodes	Flythrough mode voltages	Trap mode voltages		
		Capture	Trap	Eject
Bottom end cap	−100	−10.7	19.3	19.3
S1	−100	−3.7	8.3	8.3
S2	−100	−4.76	−4.76	−4.76
S3	−100	8.3	8.3	−46.7
Top end cap	−100	19.3	19.3	−35.7

The LT can be operated in two modes: trap and flythrough. In the “trap” mode, the LT captures, accumulates and cools ions. In other words, it prepares the ions for precision mass measurement with nearly identical initial conditions, making the ions’ energies from upstream of the LT unimportant for the accuracy of mass measurement at the Penning trap. The operation consists of three main steps: capture, trap (and cool), and eject. As the ions approach the LT, the bottom end cap and S1 are lowered for a user-defined duration to let the ions in, and hence capture them. The lowering and raising of the end cap and S1 is done using a 30 V amplifier (which amplifies a negative TTL pulse and uses it to lower the end-cap by −30 V during capture of ions), and a pulser module named “S1 capture”. Once the bottom end-cap and S1 electrodes’ potentials are raised, the ions are trapped radially using RF excitations applied to the four rods, and axially using a DC potential well, applied to the segments of each rod, similar to Figure 2.5. The RF excitations are applied from a Stanford Research System (SRS), model DS345 function generator, through an amplifier and an LCR network. For masses of around $A = 133$, $q = 1$, the RF applied is around 800 kHz, with approximately 300 V_{pp}. For a different mass, the frequency can be scaled using equation 2.17. Once the ions are trapped, they are cooled via collisions with He gas. For that, the trapping volume is filled with ~ 1 μ Torr of He gas. In addition, the whole structure sits inside a container with walls through which liquid nitrogen (LN₂) is circulated using a

custom made LN₂ pump [114] to keep the trap cold at -170°C and further facilitate cooling of the ions. The cooling of the ions continues until a user defined number of ion bunches from CARIBU are captured. 30 ms after the last bunch is captured and once the Penning trap is ready to accept ions, the potentials of S3 and the top end cap are lowered using another pulser module named “S3+top end cap eject”. This forces the trapped ions to exit through the upper aperture and move towards the Penning trap. Typical values of the DC potentials applied to the electrodes are shown in Table 4.1.

Beside the trap mode, the LT can also be operated in flythrough mode. As mentioned earlier, at the beginning of a measurement campaign, the elevator is tuned looking at the Si/MCP 3 with the linear trap at flythrough. This makes it easy to tune the elevator, without having to worry about the LT capture parameters. In this configuration, S1, S2, S3 and the two end caps are biased at -100 V and the beam just passes from above the trap potential without getting captured. Once the elevator is tuned, the linear trap is put back to trapping mode and the deceleration and capture pulse timings are tuned. The TOF of the ions at MCP 3 with the LT at flythrough gives one a good starting point for tuning these timings, which are then fine tuned to maximize transmission looking at Si / MCP 3. Typically, from the Si MR-TOF to Si 3, transmission efficiency is $\sim 50\%$.

4.4 The Canadian Penning trap (CPT)

As the ions approach the CPT, they first encounter a pair of deceleration electrodes (top and bottom), just below the trap. These electrodes are added to slow down the ions before their capture. The CPT is a hyperboloidal structure consisting of ten electrodes: pairs (top and bottom) of correction tubes, end caps and correction rings, on either side of a 4-segment ring electrode, see Figures 4.4(a) and 4.4(b). The ring electrode is segmented to apply the RF excitations required for mass measurement. The trap has characteristic dimensions of $r_0 = 11.6\text{ mm}$ and $z_0 = 10\text{ mm}$, with 5 mm apertures on either side, and sits in the bore of a 5.84 T superconducting solenoidal magnet. The correction ring and correction tube electrodes compensate for the non-ideal or *practical* aspects in the trap design, namely by the segmented ring, the finite size of the electrodes and the apertures, and create a harmonic electric potential. Downstream from the Penning trap, a series of 9

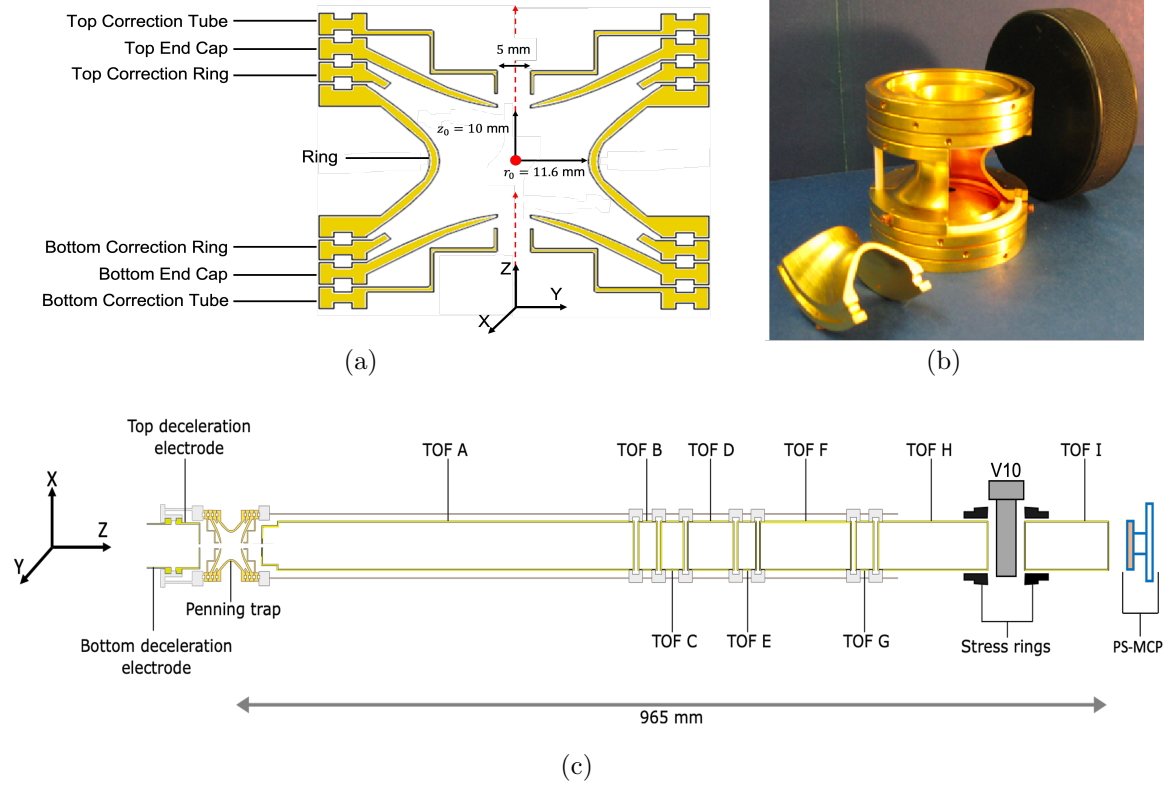


Figure 4.4 : The Canadian Penning trap (CPT) and CPT electrodes assembly. In panel (a), a schematic diagram with the electrodes labelled and the dimensions annotated is shown. The red dot at the center symbolizes a trapped ion cloud, while the red dashed arrows represent the path of the ions into and out from the CPT. A small inset of the coordinate axes system is also included for reference. Panel (b) shows a picture of the CPT next to a hockey puck for scale. Picture taken from [115]. In panel (c), the assembly of the trap along with the deceleration and extraction electrodes is shown. Figure originally drawn by R. Orford, and the V10 and the PS-MCP were added by this author.

cylindrical drift tubes, named TOF A to I, collectively called the “TOF drift tubes”, make up the extraction electrodes, leading the ions away from the trap and the magnetic field to the PS-MCP. Since the gate valve V10, which separates the Penning trap from the PS-MCP, is located between TOF H and I, thick ring shaped electrodes, called stress rings are placed on either side of the valve to ensure continuity of the electric field. From the trap center to the PS-MCP, the TOF drift tubes including the stress rings extend around 965 mm. The CPT along with the deceleration electrodes and the TOF drift tubes form one electrode assembly, as shown in Figure 4.4(c).

The whole trap and extraction region is maintained at UHV of $\sim 0.1 - 1$ nTorr. Therefore the choice of material for the trap and other components in this region had to be carefully considered given the electrical, magnetic and UHV requirements. All the electrodes are made of oxygen-free high purity copper (OFHC), and isolated from each other with insulators made from Macor[®]. OFHC having a high electrical and thermal conductivity and low magnetic susceptibility, is one of the best materials for the purpose. In addition, the electrodes are plated with gold to improve conductivity and prevent building up of static charge which could otherwise distort the electric field. The assembly is supported by a structure made of beryllium copper and molybdenum screws [57].

The bottom deceleration electrode is biased at -1300 V. As the ions exit from the linear trap and approach the CPT, the top deceleration electrode is pulsed from -20 V to -150 V. These voltages are applied from the CAEN, while the pulsing is done with a pulser module named “CPT deceleration”. A few 10s of micro-seconds after the deceleration pulse, the bottom correction tube and end cap electrodes are lowered for $\sim 10 - 25$ μ s, depending on the m/q or equivalently the A/q of the captured ions. The capture pulse is a negative square wave generated from an SRS DS345 function generator (named “CPT Capture”) and applied to the electrodes. The potentials required by the trap are provided by the Stahl. Three channels of the Stahl are used for the correction tubes, end-caps and correction rings, one for each top-bottom electrode pair. This arrangement ensures the application of a symmetric potential. The four segments of the ring electrode are biased from the fourth channel of the Stahl power supply. Once the ions are captured, the high energy ions are removed by using a combination of evaporation pulse and trap raising pulse (TRP)

waveforms, generated from two SRS DS345 function generators (named “Evaporation” and “TRP”), applied to the bottom correction tube and the ring electrodes respectively. The details of the evaporation and the TRP will be discussed in Section 4.4.1. Next, the PI-ICR RF excitations are applied to the ring segments from a four channel arbitrary waveform generator from Tabor Electronics (model WW1074) [116]. Once the ions have gained the required phases, they are ejected. The exact forms, schemes and timing of the RF excitations will be discussed in Chapter 5. The ejection pulse, similar to the capture pulse, is a negative square wave generated from another SRS DS345 function generator (named “CPT Eject”) and applied to the top end cap and correction tube electrodes. The time-dependent voltages and PI-ICR RF waveforms are capacitively coupled with the DC biases and then applied to the corresponding trap electrodes. The voltages applied to the CPT electrodes during capture, trapping and ejection of ions is shown in Table 4.2.

Table 4.2 : Typical DC biases applied to the CPT electrodes

Electrodes	Voltages		
	Capture	Trap	Eject
Bottom correction tube	−17.5	1.2668	1.2668
Bottom end cap	−13.5	−3.0836	−3.0836
Bottom correction ring	−8.9622	−8.9622	−8.9622
Ring	−12.7206	−12.7206	−12.7206
Top correction ring	−8.9622	−8.9622	−8.9622
Top end cap	−3.0836	−3.0836	−13.5
Top correction tube	1.2668	1.2668	−17.5

In PI-ICR, the ions phases are calculated by mapping their in-trap position onto a position sensitive detector. Therefore, to minimize distortions and optimize the quality of the projection, the ions’ TOF through the magnetic field gradient from the trap to the detector has to be minimized. After the CPT, as illustrated in Figure 4.4(c), the first electrode the ions encounter is TOF A, the longest of all the TOF drift tubes, and which sits in a region of large magnetic field gradient. To minimize the effect of the magnetic field on the ion trajectories, they are accelerated to a substantial energy by biasing the TOF A at a much

negative voltage compared to the trapping potential in the CPT. On the other hand, the proximity of TOF A to the CPT means, biasing it at a large negative voltage might disturb the harmonicity of the trapping electric field in the CPT. In order to avoid perturbing the trapping field in the Penning trap during excitation, the electrode is maintained at -20 V and pulsed to -300 V during the ejection process, using a pulser module, named “TOF A pulser”. The rest of the TOF electrodes and the stress ring, except TOF F, are maintained at -400 V. TOF F is biased at -850 V, making it act like an einzel lens to focus the ejected ions towards the center of the detector. The biases of the TOF drift tubes and the stress rings are shown in Table 4.3. This set of TOF voltages is just one of many combinations tried offline with $^{133}\text{Cs}^+$ when PI-ICR was commissioned at the CPT and has worked best to minimize the size of the projected spots on the detector [53]. After passing through the TOF drift tubes, the ions are detected at the position-sensitive MCP (PS-MCP). The details of the PS-MCP and the data acquisition will be discussed in Chapter 5.

Table 4.3 : Typical DC biases applied to the TOF electrodes and the stress rings

Electrodes	Voltages
TOF A	-20^{a}
TOF B	-400
TOF C	-400
TOF D	-400
TOF E	-400
TOF F	-850
TOF G	-400
TOF H	-400
Stress rings	-400
TOF I	-400

^aPulsed to -300 V during ejection

Similar to the linear trap, the Penning trap can also be operated in the flythrough mode. This is only done to diagnose problems, including but not limited to matching the Penning trap potential to the linear trap potential, or investigating transmission issues through the

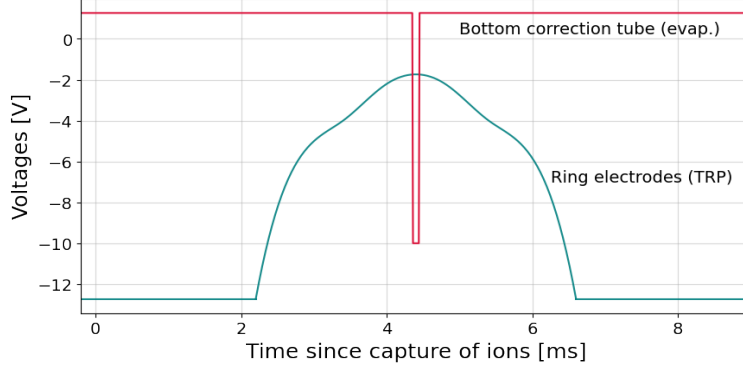


Figure 4.5 : TRP and evaporation pulses.

Penning trap. The Stahl power supply is equipped with a ± 30 V float input. The CPT electrodes' potential is lowered by applying -25 V into this float input to the Stahl, allowing the ions to pass above the trap potential without getting captured. The flythrough mode is only included here for completeness, and since it is never used for actual measurements, will not be discussed any further.

4.4.1 Removal of high energy ions

The magnetic field of ~ 5.84 T is measured at the center of the trap along all three dimensions. As the ions move away from the center, there is a field gradient. The ion's measured frequencies depend on the trap's magnetic field. Therefore, during a precision measurement, all the ions in an ion cloud must probe approximately the same magnetic field in every measurement cycle. As an ion cloud is transferred from the Paul trap to the Penning trap, some energy spread is imparted in it. In the current set-up, due to the UHV conditions in the Penning trap, gas cooling cannot be implemented. Instead, once an ion batch is captured, the ions with high energy are simply removed from the trap. This is done by applying a trap raising pulse (TRP) to the ring electrode which raises the ring adiabatically by ~ 10 V over ~ 3 ms, and an evaporation pulse to the bottom correction tube electrode which lowers the correction tube by ~ 10 V for $10 \mu\text{s}$. This is shown in Figure 4.5.

The adiabatic increase of the ring electrode potential raises all the trapped ions' potential. Next, the lowering of the bottom correction tube bias forces the ions, with potential greater than the lowered correction tube to escape from the trap. This is shown in Figure 4.6. Recall

from equation 2.34, the ions' axial frequency in a Penning trap is given as:

$$\omega_z = \sqrt{\frac{q}{md_0} V_0} , \quad (4.5)$$

where q/m is the charge to mass ratio of the trapped ions, d_0 is the characteristic trap dimension and V_0 is the trap depth, defined by the voltage difference between the endcap and the ring electrodes. Most of the ion species trapped in the CPT have period of axial motion between $10 - 30 \mu\text{s}$. The TRP plateaus for $\sim 400 \mu\text{s}$ plateau at the high voltage. This reduces V_0 in such a way that it causes a fractional shift in the ions' axial motion [53]. As a result, the low energy ions stay minimally perturbed and stay trapped, while the high energy ones escape. Along with the TRP and the evaporation pulses, the CPT capture pulse is also critical in this aspect. A mistimed capture pulse can also impart some additional energy to the ions. So the capture pulse needs to be tuned carefully such that the ions are in the $+z$ direction in their ω_z motion when the capture pulse is applied. A combination of well-tuned capture, TRP and evaporation pulses typically removes $\sim 50\%$ of the initially trapped ions.

As mentioned earlier, the TRP and the evaporation pulses are programmed into two SRS DS345 function generators, named "TRP" and "Evaporation" respectively. The TRP is coupled to the DC voltage from the Stahl "ring" channel via a pulser module named "TRP pulser". The TRP function generator output and the Stahl "ring" channel output are applied to the "high" and "low" inputs of the TRP pulser respectively, and the output of the pulser is applied to the ring electrode through the CPT coupling box. The Evaporation function generator output is sent to a $\pm 30 \text{ V}$ summing driver input. The capture pulse is sent to a second input of the same summing driver. The output from the summing driver is applied to the bottom correction tube. Since the two pulses (capture and evaporation) are applied at different times and the respective function generators are programmed to output a 0 V DC at all other times except during the respective pulses, summing the two keeps the individual waveforms unchanged.

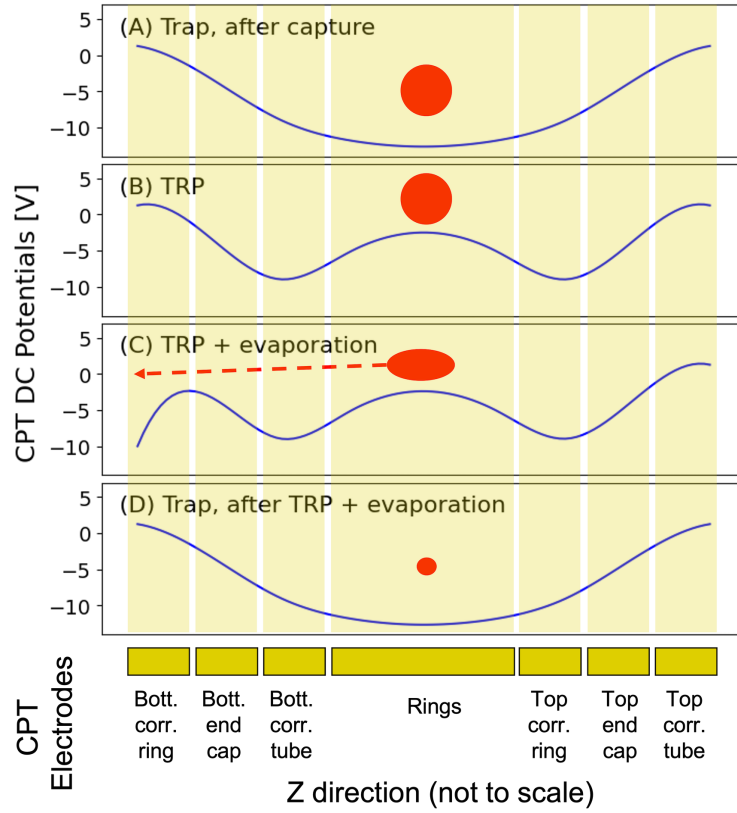


Figure 4.6 : An illustration of the principle behind removal of high energy ions in four stages shown in the four panels. The y axis represents the voltages applied to the seven trap electrodes, shown in golden rectangles at the bottom along the x -axis representing the z direction, and the seven vertical golden bands on all four plots. The blue line in every plot shows the electric potential as seen by the ion cloud, depicted by the red circle (or oval in panel C). Panel A shows an ion cloud, just after its capture, trapped in the CPT electric potential well. In panel B, the ring electrode's voltage is raised, which also increases the potential of the ion cloud. In panel C, the bottom correction tube voltage is lowered and the high energy ions from the cloud escape from the trap, as shown with the red dashed arrow. Panel D shows all electrodes back to their "trapping" setting and a low-energy ion cloud trapped.

4.5 CPT operation and timings

The main components of the CPT control system are housed in a Computer-Aided Measurement And Control (CAMAC) crate, which communicates with the CPT computer through a General Purpose Interface Bus (GPIB) interface. The timing parameters are programmed into four digital delay generators DG11A (named A, B, C and E) from Data Design Corporation [117] and a Timing and Sequence Module (Model 221A) from Jorway Corporation [118]. Each DG11A module has four outputs which can be independently programmed with respect to some common trigger. These modules have a time resolution of 10 ns and a maximum delay of 167.77216 ms [117]. The Jorway 221A has 12 output channels which are programmed to produce the same output after some delay with respect to some trigger. It has a timing resolution of 1 μ s and a maximum delay of 16.777215 s [118]. For the sequence of logic flow, two *coincidence* modules, a *rate divider* module and a *set/reset* module are used. The *coincidence* modules produce a true output when two true inputs are recorded. The *rate divider* module produces multiple true outputs once it registers a user-defined number of true inputs. This user-defined number of inputs can be set between 1 and 999. The *set/reset* module starts with a true output until it records a true signal at its “reset” input, at which point it outputs a false logic signal. Its output stays false until it registers a true logic at this “set” input. A map of all these modules is shown in Figure 4.7. Intermediate modulations of the logic shapes, that is NIM (Nuclear Instrumentation Modules) to TTL (Transistor-Transistor Logic) and vice versa in normal as well as complimentary mode, as required by the different devices are done by NIM / TTL converter modules from Teledyne LeCroy (NIM model 688AL Level Adapter). A number of Quad Gate / Delay Generators from Phillips Scientific (model PS794) are also used to transmit logic signals with fixed delays or fixed widths or to create simple flip flops to create user-defined pulses. The level adapters and the PS794s are not shown in Figure 4.7 for a cleaner representation. The DG11A, Jorway and one of the *coincidence* modules are powered by the CAMAC crate while all the other modules are powered from Ortec 4001 NIM BIN crates.

A pulse scheme timing diagram of a CPT measurement cycle is shown in Figures 4.8 and 4.9. For simplicity, the entire cycle is divided into two pictures: first one (Figure 4.8) shows

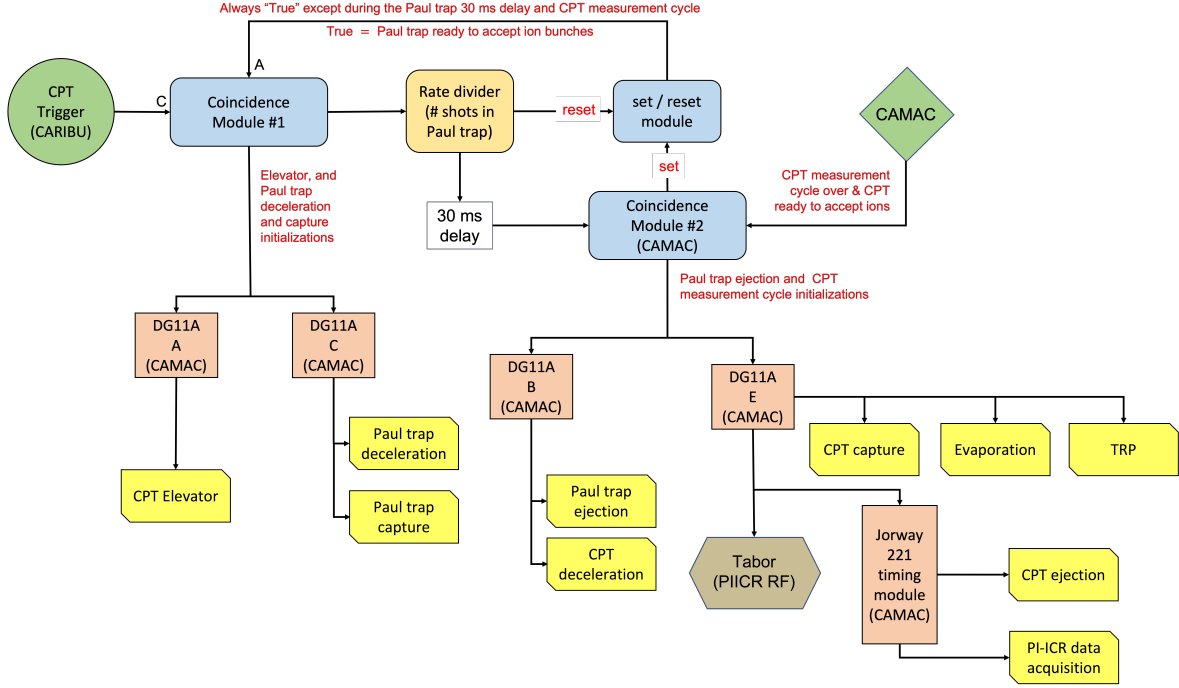


Figure 4.7 : CPT electronic modules map and logic flow

the transfer of ions from CARIBU to the linear trap and then from the linear trap to the Penning trap, the second one (Figure 4.9) repeats the transfer of ions from the linear trap to the Penning trap for reference and also shows the CPT cycle and data acquisition (DAQ). The details of the measurement procedures will be shown and discussed in Chapter 5.

The whole process is started by loading a parameter file with all the delays and the waveforms onto a *cycling* program and by starting “cycling”. The program in turn loads the delays into the delay generators through the CAMAC crate and the wave functions into the SRS DS345 function generators. The output of the *set/reset* module is by default set to 1, that is, the Paul trap is ready to accept ions. This makes the “A” input to the *coincidence* module (named “#1”) true. The other input (“C”) comes from the “CPT Trigger (t_5)” signal mentioned in Section 3.5. As the “CPT Trigger” registers a true signal, the *coincidence* module “#1” produces two outputs. The first output triggers two DG11A modules, A and C. After a user-defined delay (t_6), the DG11A A then sends a signal to trigger the CPT elevator HV switch through a PS794 module. The width of the elevator pulse is set to $\sim 15 \mu\text{s}$ by the PS794. All four outputs of module DG11A C are used. Channels 1 and 3 set the start and stop time of the linear Paul trap deceleration pulse. These outputs are fed

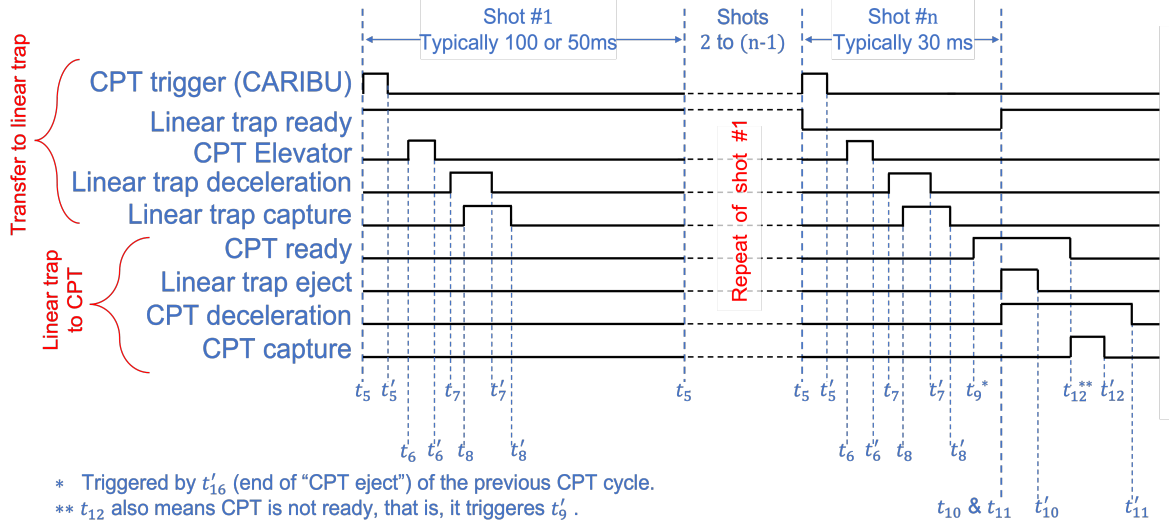


Figure 4.8 : CPT timings (part 1): Paul cycle completion and Penning cycle initiation. As in Figure 3.20, the start and end of different functions are denoted by t_i and t'_i , ($i = 7$ to 12). The timings are not to scale.

into the trigger and reset inputs of a PS794 module, operated in flip-flop mode. As a result, the PS794 outputs a gate signal starting at t_7 and ending at t'_7 , which is then amplified and combined with the LT bottom endcap DC biases in a simple summing driver circuit and applied to the electrode. Channels 2 and 4 of DG11A C are used to create the start and stop of the LT capture. Similar to LT deceleration pulse, these are connected to the trigger and reset inputs of a PS794 in flip-flop mode creating a gate signal between t_8 and t'_8 . This gated signal is then applied to the "S1 Capture" pulser module, which lowers the S1 electrode bias to enable capture of the ions. The second output from the *coincidence* module "#1" goes to the input of the *rate divider* and is registered as 1 count. The *rate divider* controls the number of ion bunches or shots from the CARIBU buncher, that will be collected for cooling in the linear trap, before they are all sent to the Penning trap for measurement. After a user-defined number of input counts (n) are registered, the rate divider in turn produces two outputs. The first one sends a true signal to the "reset" input of the *set/reset* module, which changes its output to false. This false signal is sent to the *coincidence* module "#1" input "A" which signifies that the LT is not ready to accept further ions. The second output from the *set/reset* module is used to trigger a PS794 module, which outputs a hard-coded 30 ms delayed output. This ensures that the last captured ion bunch has enough time to cool, and the LT does not accept any more ions bunches till all the captured and cooled ions are sent to

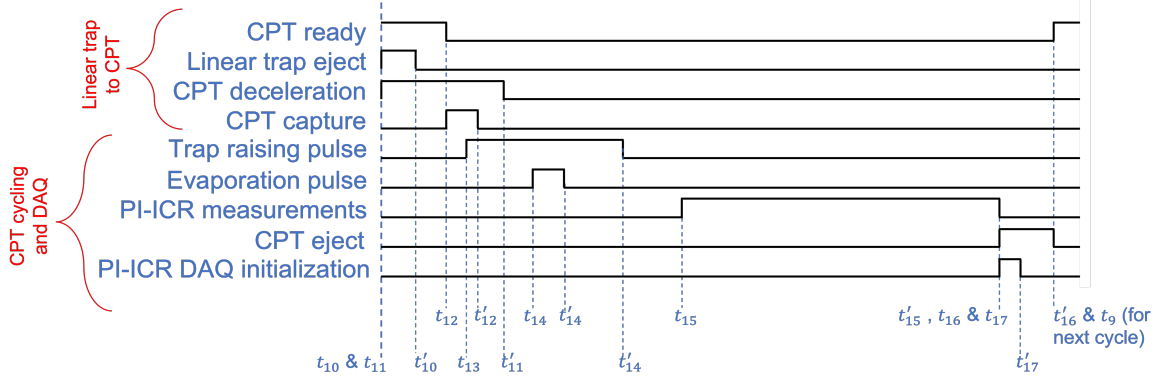


Figure 4.9 : CPT timings (part 2): Transfer to Penning trap, Penning cycle and PI-ICR DAQ initiation. As before, the start and end of different functions are denoted by t_i and t'_i , ($i = 10$ to 16). The timings are not to scale.

the CPT. After this 30 ms, the PS794 output signal is sent to a second *coincidence* module (named “#2”), which keeps track of whether the CPT is in the middle of a measurement cycle or if it is ready to accept ions. If the CPT is empty and ready to accept ions (time t_9), the *coincidence* module “#2” produces two outputs. The first one is fed into the “set” input of the *set/reset* module which sets its output back to true. This true output in turn is sent to the “A” input of *coincidence* module “#1”, meaning the LT is again ready to accept ions. The second output from *coincidence* module “#2” is used to trigger two DG11A modules, B and E. After a short delay, of the order of a few μs (t_{10}), the first output (channel 1) from DG11A B is produced and it is sent to the “S3 + top end cap eject” pulser modules via a PS794. Since the width of the LT ejection pulse is not as critical as the deceleration and the capture pulse, it is set to $\sim 100 \mu\text{s}$ by the PS794 module. The “S3 + top end cap eject” pulser lowers the S3 and end cap electrode biases to facilitate ejection of the ions from the LT. Output channels 3 and 4 from DG11A B are sent to the trigger and reset inputs of a PS794 operated in flip-flop mode, resulting in a gate pulse with a user defined width starting at t_{11} and ending at t'_{11} . This pulse is sent to the “CPT deceleration” pulser module, which slows down the ions and prepares them for capture by the CPT. In practice, t_{10} and t_{11} are kept the same.

The DG11A E module is used to initiate the CPT cycle. After a few 10s of μs of the trigger, channel 1 from the DG11A E is used to trigger the “CPT Capture”, “Evaporation” and “TRP” function generators. The start-times (t_{12} , t_{13} and t_{14}), stop-times (t'_{12} , t'_{13} and

t'_{14}) and the exact wave-forms are programmed and loaded into the function generators from the “cycling” program. The start of the CPT Capture pulse also symbolizes that the CPT is in the middle of a measurement cycle and is not ready to accept ions. This information is fed into the coincidence module “#2” by the CAMAC. This means for the next Paul trap cycle, even if the user defined number of shots have been captured and cooled, they will not be sent to the CPT. About ~ 7.9 ms (t_{15}) after the DG11A E module is triggered, it produces an output from its channel 2, which is used to trigger the Tabor function generator (for PI-ICR RF excitation, see Chapter 5) and the Jorway 221 timing module. Another user-defined time later (t_{16} and t_{17}), the Jorway emits two outputs. The first one (after t_{16}) is used to trigger the “CPT Ejection” function generator, the “TOF A pulser” and as an input to the time-to-digital converter (TDC). Similar to the CPT capture, evaporation and TRP waveforms, the ejection waveform, and its start and stop time are also programmed and loaded from the “cycling” program. The end of of the ejection pulse (t'_{16}) also signifies that the CPT is ready to accept ions and the CAMAC crate in turn triggers the time t_9 . To avoid skipping ion shots from CARIBU, care is taken such that the CPT cycle time is always shorter than the LT time. In practice, this is done by setting the CPT time as dictated by the required precision and mass resolution (see Chapter 5) and then increasing the LT time by increasing the number of shots to be captured (n). The TDC input is used to calculate the TOF of the ions. The other Jorway output (after t_{17}) is used to create a veto for the constant fraction discriminator (CFD). This is labelled in Figure 4.9 as PI-ICR DAQ initialization. These will be discussed in Chapter 5. A summary of all CPT timings for a beam of $^{150}\text{Ce}^{2+}$ for a ~ 270 ms CPT cycle is shown in Table 4.4. If a signal is triggered from one source and programmed from another, both are mentioned in the “Source” column with the source of the trigger indicated as “(trig.)”.

Once transmission of ions through the CPT system is maximized at the beginning of a measurement campaign using a tuning beam, or the mass measurement of a certain nuclide has concluded, the timings of the CPT system, except t_{15} , t_{16} and t_{17} , are scaled to the A/q of the next isotope-of-interest using equation 3.10b, similar to scaling of the CARIBU timings. The reason behind keeping t_{15} , t_{16} and t_{17} unchanged will be explained in Section 5.2.

Table 4.4 : Typical CPT timings for a $^{150}\text{Ce}^{2+}$ beam from CARIBU for a ~ 270 ms CPT cycle

i	Labels	Triggers (t_i)	Widths (Δt_i)	Source
Transfer to LT: after CPT trigger (t_5)				
5	CPT trigger	0	$5 \mu\text{s}$	CARIBU delay generator
6	CPT elevator	$61.51 \mu\text{s}$	$5 \mu\text{s}$	DG11A A Ch. 4
7	LT deceleration	$73.91 \mu\text{s}$	$6.36 \mu\text{s}$	DG11A C Ch. 1 and 2
8	LT capture	$76.60 \mu\text{s}$	$13.02 \mu\text{s}$	DG11A C Ch. 3 and 4
LT to CPT: 30 ms after last captured shot in the LT				
9	CPT ready			CAMAC
10	LT eject	$8.03 \mu\text{s}$	$5 \mu\text{s}$	DG11A B Ch. 1
11	CPT deceleration	$8.03 \mu\text{s}$	$147.01 \mu\text{s}$	DG11A B Ch. 3 and 4
12	CPT Capture	$37.18 \mu\text{s}$	$11.30 \mu\text{s}$	DG11A E Ch. 1 (trig.) and SRS DS345 (“CPT Capture”)
CPT cycling and DAQ (~ 270 ms CPT cycle): after LT eject (t_{10})				
13	TRP	$2235.18 \mu\text{s}$	$4097.82 \mu\text{s}$	DG11A E Ch. 1 (trig.) and SRS DS345 (“TRP”)
14	Evaporation	$4035.18 \mu\text{s}$	$100 \mu\text{s}$	DG11A E Ch. 1 (trig.) and SRS DS345 (“Evaporation”)
15	PI-ICR measurements (trig.)	$7908.28 \mu\text{s}$	$5 \mu\text{s}$	DG11A E Ch. 2
16	CPT eject	272.40 ms	$1950.2 \mu\text{s}$	Jorway (trig.) and SRS DS345 (“CPT Ejection”)
17	PI-ICR DAQ initialization	272.40 ms	$5 \mu\text{s}$	Jorway

5 PI-ICR at the CPT

The basic concept of PI-ICR has been discussed in Section 2.4.2. Here I will describe how the trapped ions' radial eigenfrequencies (ν_- and ν_+), as well as ν_c are measured in the CPT. Since PI-ICR relies on projecting the in-trap locations of the ions onto a position sensitive detector, I will first describe the detection system in place to acquire the information from the detector, followed by the details of the technique as implemented at the CPT.

5.1 Detection system : hardware, software and data acquisition

The PI-ICR detector and data acquisition set-up at the CPT is a commercial package from RoentDek Handels GmbH, which includes the PS-MCP (DLD40), the associated electronics comprising of a signal de-coupling feedthrough (FT12TP), a fast signal amplifier (FAMP6), a constant fraction discriminator (CFD4c), a high resolution time-to-digital converter (TDC8HP), and read-out software (CoboldPC). The set up provides a position resolution of < 0.1 mm, temporal resolution of < 0.2 ns, rate capability of 1 MHz and multi-hit dead time of 10 – 20 ns for each ion-hit [119, 120]. An overview of the data acquisition system, with the major components, is shown in Figure 5.1.

The PS-MCP consists of a pair of matched image quality MCP plates, from Hamamatsu Photonics K.K., stacked in a chevron configuration, with a delay line anode. Each plate has an active diameter of 45 mm, thickness of 1.5 mm and provides an electron gain of 10^7 with a bias of -2400 V [119, 120]. The two plates, front and back, are stacked together using a pair of ceramic rings, held together by four spring clamps. The ceramic rings have an inner metallic ring, which makes contact with the plates and through which the biases, as required by the plates, are applied. The delay line anode sits in a “holder”. The MCP plates and support structure are held by the holder with holder rods, which also isolates them from the delay lines. The delay line anode is made of two sets of copper wires, wrapped around a ceramic support, to independently provide precise positional information in both X and Y dimension. For each dimension, there exist a pair of differential wire combination, *collection* and *reference* wires, in two orthogonal directions: X_1 and X_2 , and Y_1 and Y_2 . The *collection* and *reference* wires are connected, by a twisted-pair configuration, to the FT12TP feedthrough equipped with a decoupling plug. The difference between the *collection* and

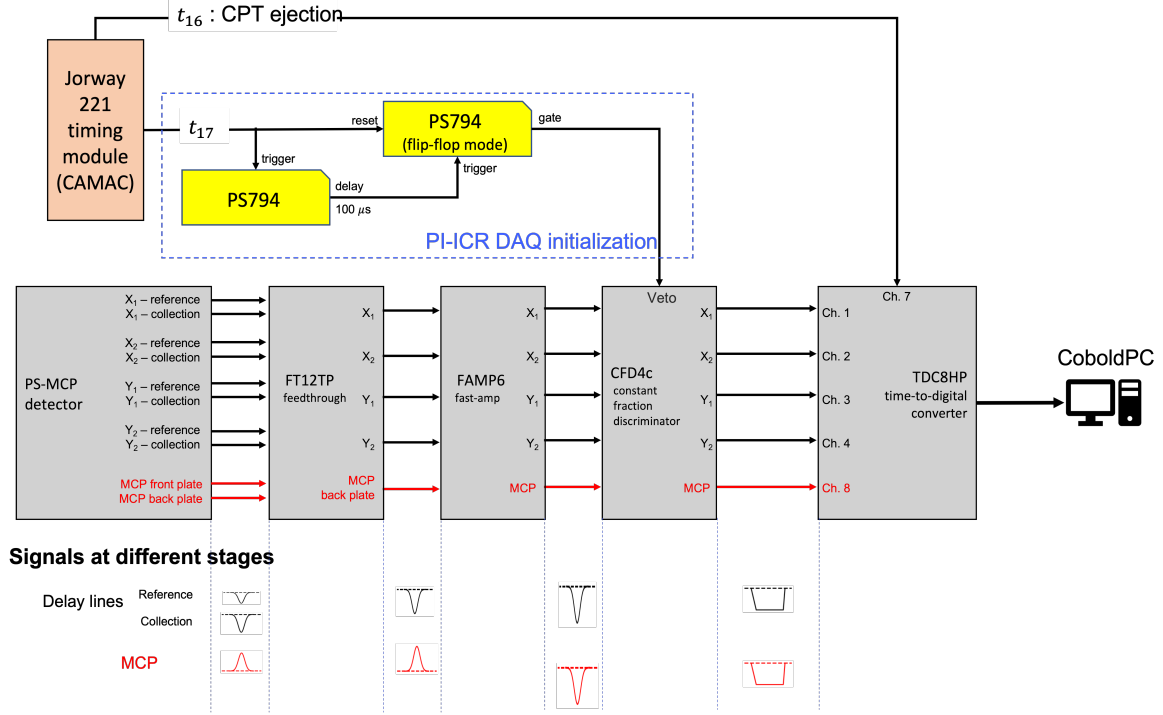


Figure 5.1 : Overview of the PI-ICR data acquisition with the major hardware components labelled. Also shown are the MCP and delay line signals at different stages of processing.

reference wires lies in their biases. Two pictures of the PS-MCP, one focussing on the MCP front plate and the other on the delay lines, are shown in Figure 5.2.

All the components of the PS-MCP are biased from a dual channel ISEG NHQ 214M [121] HV power supply. A voltage of -2300 V is applied to the front MCP plate directly from channel A of the power supply. Channel B, set at $+300$ V, is connected to the input of a RoentDek HVZ voltage divider, which has four outputs. These four outputs are used to bias the back plate, the holder, and the *reference* and *collection* wires of the delay lines. All the biases are applied via SHV (safe high voltage) sockets on the FT12TP feedthrough. The typical applied voltages are shown in Table 5.1.

The two MCP-plates are resistively matched to create a smooth potential gradient between the two. Each plate consists of a number of capillary tubes, $25 \mu\text{m}$ in diameter and inclined at $8^\circ \pm 1^\circ$ [119, 120], which act as continuous dynode or channeltron electron multipliers. When a positively charged ion hits the high-negatively biased front plate of the MCP, a cascade of electrons is initiated, which further gets amplified through collisions with the channel walls. However, this amplified electron cloud potentially ionizes the residual gas

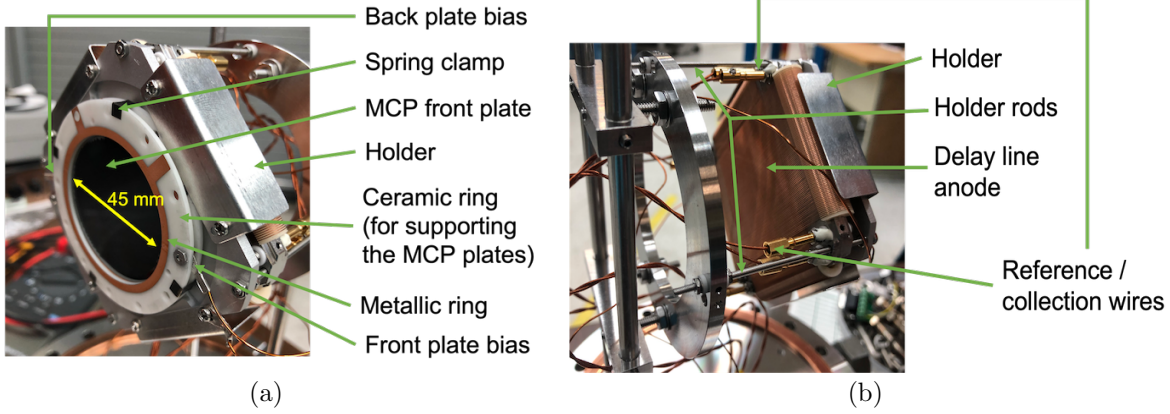


Figure 5.2 : Picture of the PS-MCP with the plate and the delay lines. Panel (a) is taken from the side of the MCP front plate, while panel (b) is taken from the side of the delay lines.

Table 5.1 : Typical voltages applied to the PS-MCP components

PS-MCP Part	Voltage [V]	Applied from
MCP front plate	−2300	Power supply channel A (directly)
MCP back plate	33	Power supply channel B (through the HVZ voltage divider)
Holder	86	
Reference wires	260	
Collection/signal wires	300	

molecules within the MCP, which causes an ion-feedback noise effect. The chevron configuration minimizes this effect, as these molecular ions get absorbed at the junction between the plates [122]. The result is a clean and amplified signal at the back plate. The intermediate bias of the holder, between that of the back plate and the anode wires, ensures the electron cloud travels from the back plate to the anode wires with the minimal spatial distortion. In order to further minimize image distortion, the PS-MCP is situated outside any magnetic field > 5 mT. A potential difference of ~ 40 V between the *collection* and the *reference* wires, with the *collection* being more positive, means the majority of the electron cloud is collected by the *collection* wires. This creates four pairs of differential signals to propagate through the corresponding wires, *collection* and *reference*, in the respective four directions X_1 , X_2 , Y_1 , and Y_2 . A schematic diagram of the PS-MCP set-up with the delay line anode, and the propagation times of the eight signals from the location of the ion hit to the end of

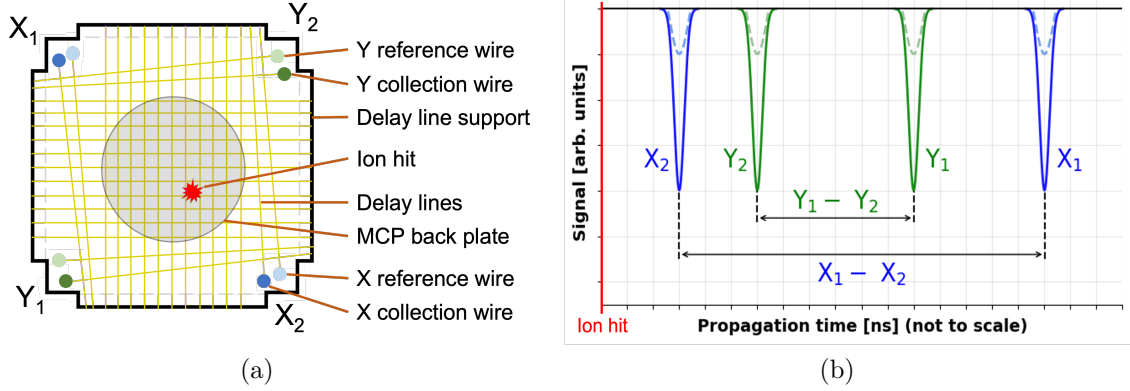


Figure 5.3 : Illustration of the delay line anode operation. Panel (a) shows a schematic rear view of the PS-MCP with the delay lines and the back MCP plate visible, along with a cartoon of an ion hit. Figure reproduced from Ref. [53]. Panel (b) shows the propagation time taken by the four *reference* (small peaks in dashed lines) and *collection* (big peaks in continuous lines) signals corresponding to X_1 , X_2 , Y_1 , and Y_2 from the ion hit to the end of the corresponding delay lines.

the corresponding delay lines, are shown in Figure 5.3. These propagation times, along with two signals from the time of the ions hitting the MCP front and back plate (to calculate the TOF of the ions from the CPT to the detector), are processed by the FT12TP decoupling feedthrough. Pictures of the feedthrough along with the plug and its airside pin map is shown in Figure 5.4, while the pin assignment is shown in Table 5.2

Table 5.2 : Pin assignment of the FT12TP feedthrough

Pin #	Connection	Pin #	Connection	Pin #	Connection
1	Not used	5	X_1 - reference	9	Y_1 - reference
2	MCP front plate	6	X_2 - collection	10	Y_1 - collection
3	MCP back plate	7	X_2 - reference	11	Y_2 - reference
4	Holder	8	X_1 - collection	12	Y_2 - collection

The resulting outputs from the FT12TP are four negative signals corresponding to the four directions X_1 , X_2 , Y_1 , and Y_2 , and one positive signal for the MCP hit, which the user can select to be either the front or back plate via a toggle switch. The back plate produces a larger signal [53] and is therefore used for all the measurements presented here. The difference of the propagation time in each dimension that the signals take to reach the end of the wires gives the exact coordinate in that dimension. For simplicity, from here on, I

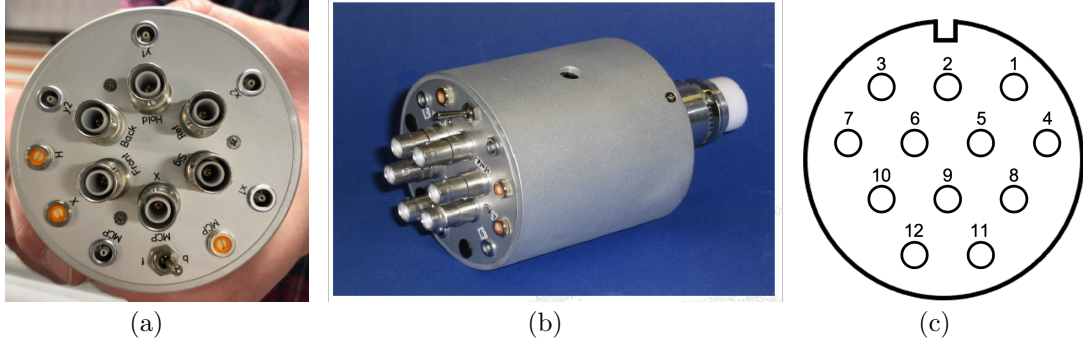


Figure 5.4 : FT12TP feedthrough. Panel (a) shows the outside of the FT12TP plug. The SHV sockets used for biasing the different PS-MCP components can be seen towards the center, while LEMO output sockets to transmit the signals X_1 , X_2 , Y_1 , Y_2 , and MCP are also visible. The toggle switch next to the MCP LEMO output port with labels “f” and “b” decides which plate signal is to be transmitted. Panel (b) shows a picture of the whole FT12TP plug. Panel (b) is taken from Ref. [120]. Panel (c) is a pin map of the FT12TP airside feedthrough.

will refer to the four propagation times by their directions, that is X_1 , X_2 , Y_1 , and Y_2 , which gives the position of the ion hit as:

$$X = k_x (X_1 - X_2) \quad (5.1a)$$

$$Y = k_y (Y_1 - Y_2) \quad (5.1b)$$

where k_x and k_y are the signal propagation coefficients in units of mm/ns and obtained from calibrating the detector during commissioning [53]:

$$k_x = 0.645 \quad \text{and} \quad k_y = 0.655 . \quad (5.2)$$

The details of the calibration procedure can be found in reference [53]. There are three steps before the location (X, Y) can be decrypted, the first of which is signal amplification. These five signals are sent to a 6-channel fast amplifier (FAMP6) [123]. Each channel of the FAMP6 has one non-inverted and one inverted output, and the gain on each channel can be independently tuned up to a maximum of $200\times$.

Since the precise location of an ion hit depends on the timing signals, which can vary widely in their heights and can also include low amplitude noise, the signals need to go through a discriminator circuit before the positions (X, Y) can be calculated from the relative timings $(X_1 - X_2)$ and $(Y_1 - Y_2)$ [124]. Because of the walk and jitter effects associated with a leading edge discriminator[124], a constant fraction discriminator (CFD) is used. The

function of the CFD is to produce a NIM output once the input signal is greater than a constant fraction of the pulse height. The CFD module from RoentDek, CFD4c [125] has four independent channels and requires a negative pulse input, with amplitude ≤ 2 V and has a bandwidth of 300 MHz. Five negative signals from the FAMP6, four from the non-inverted outputs corresponding to the four delay line signals X_1 , X_2 , Y_1 , Y_2 and one from the inverted output for the MCP hit, are sent to the inputs of five independent channels of two CFD4c modules. The gain of each FAMP6 channel is carefully tuned so that their output amplitude never exceeds 2 V. Typically these amplitudes vary between 200 – 500 mV.

In principle, a number of events can get registered as an ion hit on the MCP. Other than hits of *interesting* ions ejected from the CPT, these could include dark counts, large amplitude noise pulses, and stray ions. Including the position information for these events along with the *interesting* ion hits could lead to noisy data. A number of measures are followed to minimize this. The first one is enforcing a veto on the input signals read by the CFD4c. These modules are equipped with a Veto feature, wherein their outputs are disabled when a False signal is present on the Veto input. As illustrated in Section 4.5 (shown in Figure 4.7), two outputs from the Jorway 221 timing module are used to control ejection of ions from the CPT and subsequent data acquisition, denoted by the respective timings t_{16} and t_{17} in Figure 4.9. The second output (t_{17}), along with a delayed signal from one PS794 module, is used to reset and set, respectively, a second PS794 module operated as a flip-flop. As a result, the second PS794 outputs a signal which is always False, except for a certain duration after the CPT ejection, and this output is sent to the CFD4c Veto input. The duration is decided by the delay set in the first PS794 and is typically fixed at 100 μ s. This “PI-ICR DAQ initialization” set-up is shown by the blue dotted-lined rectangle in Figure 5.1, and makes sure that the detector is muted when ejected ions are not expected to be present.

In case of an electrical discharge in the upper section of the CPT tower, a large number of ions would follow the magnetic field, hit the MCP, and would damage the plates. In order to avoid that, a protection circuit is set up to remove the voltages applied to the plates if this should happen. The ISEG NHQ 214M modules have an inhibit option, where in, a TTL signal at their inhibit input would trip the power supplies. An output from the CFD

channel “MCP” is sent to a ratemeter. The ratemeter is set to output a 10 V signal if the input signal exceeds a rate of 1 kHz. This signal is converted to a TTL pulse by sending it through a LeCroy (NIM model 688AL Level Adapter) before it is fed into the power supply inhibit input. Additionally, each channel of the power supply is current limited to 300 μA as an additional protection.

The final step in the DAQ system is digitizing the five NIM signals from the CFD corresponding to the five accurate timings. This is done with a time-to-digital converter (TDC) from RoentDek, model TDC8HP, which communicates with the CPT computer through a PCIe card. The TDC8HP module has 8 NIM compatible high resolution input channels, numbered 1 – 8, with bin sizes of 25 ps [126]. Channels 1 – 4 are used for X_1 , X_2 , Y_1 , and Y_2 respectively, while the MCP signal is fed into channel 8. The CPT ejection signal (t_{16}) from the Jorway is connected to channel 7 of the TDC, and, together with the MCP signal (TDC channel 8), it is used to obtain the TOF information of the ions. The delay line signals range between 0 to 45 ns depending on the position of the ion hit on the MCP, while the TOF of the ions could vary from $\sim 5 \mu\text{s}$ to $\sim 200 \mu\text{s}$, depending on the mass of the ions and how quickly they are ejected from the CPT (controlled by the amplitude of the TOF A pulse) [53]. The TDC8HP also has a low resolution *trigger* input with a binsize of 12.8 ns, and a *clock* input to sync multiple TDCs [126]. None of these two are used in the CPT set up. We operate the TDC in continuous mode wherein it collects data continually.

The signals from the TDC are interpreted and analyzed using a software package, obtained from RoentDek, known as “Computer Based Online-offline Listmode Dataanalyser” or CoboldPC [127], which works hand-in-hand with the TDC8HP. CoboldPC is written in C++ and can be operated in two main modes: Online (or read from hardware) and Offline (or read from file). In the online mode, the experimenter can choose whether to save the data to a file or not. The program is initiated by executing a *startup* batch file, before one would select the mode of operation. The *startup* file uses four classes of definitions to instruct CoboldPC how to decode and process the TDC signals. The classes are:

1. Parameters: These are used to assign the different TDC channel numbers to the different physical quantities (shown in Table 5.3) and instruct the program how to classify the different delay-line signals into groups of events for DAQ, as well as declaring

different constants for online data analysis (DAN) of the signals.

2. Coordinates: These are used to declare all the data variables, either directly acquired (namely any trigger event, the delays X_1 , X_2 , Y_1 , Y_2 , and the ions' TOF) or calculated (like absolute time of an event, time difference between two successive events, sum of the delay line signals, namely $\text{sum}X = X_1 + X_2$ and $\text{sum}Y = Y_1 + Y_2$, absolute positions X and Y , radius of a hit from a center, phase acquired by an ion with respect to a center, and others).
3. Conditions: Data-cut definitions, that can be applied to any one or multiple coordinates to differentiate between *good* data and noise.
4. Spectra: These, as the name suggests, are used to define different spectra to visualize the different coordinates in 1-D or 2-D histograms, with one or multiple Conditions imposed.

Table 5.3 : CoboldPC parameters set from the TDC channels

Parameter #	Name	TDC channel	Original source
64	Trigger	8	MCP back plate
1026	X_1	1	Delay line X_1
1027	X_2	2	Delay line X_2
1028	Y_1	3	Delay line Y_1
1029	Y_2	4	Delay line Y_2
1032	TOF	7	CPT eject (Jorway)

During online operation, the *trigger* parameter (# 64) dictates how the delay line signals from a particular hit will be grouped as one event. It defines the start of an event, as well as the end of the previous event. The MCP hit channel of the TDC (channel 8) is set as the trigger. A signal on this channel is first timestamped. Next, the program waits for signals from the other channels (1 – 4) till the next MCP hit on channel 8 is registered. This is set from the *startup* file by a parameter # 1040 (Reset Event Counter). If there is at least one signal on any of the delay lines, it is stored as the value for the corresponding parameter or

equivalently the physical quantity in a data array. In other words, parameter 1026 (named X_1), for example, is set by the *startup* file to take the value of the TDC channel 1, and is therefore interpreted and stored as X_1 . Additionally, the trigger signal considers the last signal registered on channel 7 from the last CPT eject before the trigger, to record the TOF as a negative value. When operated in saved-to-file online mode, the data is then saved to a file with the extension *.lmf*, which stands for *list mode file*. In the LMF format, the data is stored on an event-by-event basis in a data-list [127]. While reading a previously saved LMF file in offline mode, one has to be careful to use a *startup* file with the same parameters as the one used to create that LMF file, so that the data is read correctly. The main DAN parameters include the signal propagation coefficients k_x and k_y to obtain the position of the ion hit, and the (X, Y) of the center to calculate the radius as well as angle of the ion hit. Every spectrum typically updates every 1 s.

A condition is defined by stating its name and specifying two limits on a particular coordinate. A spectrum is defined by specifying 1-D (or 2-D) histogram, the coordinate(s) one wants to visualize, the upper and lower limits on that (those) coordinate(s), and finally the condition(s) one wants to impose. Any number of conditions and spectra can be defined in the *startup* file. The conditions do not delete any data or control what data is written to the LMF files, rather they only enable one to visualize a subset of data for any coordinate.

The first and foremost coordinate is called *four*, which makes sure that only events with all four delay line signals are considered. It is set using a coordinate Consistency Indicator (C), defined as:

$$C = \sum_i u \cdot 2^{(i-1)}, \quad (5.3)$$

where, i is the TDC channel number for the delay lines (see Table 5.3) and u is given by:

$$u = \begin{cases} 1, & \text{if at least one hit in the TDC channel } i \text{ was registered.} \\ 0, & \text{otherwise.} \end{cases}$$

Every time there is a signal on the MCP channel of the TDC, CoboldPC starts populating the Consistency Indicator spectrum (spectrum 0), plotting histograms of counts for the different C values. If this signal is NOT followed by any signal on the delay lines channels,

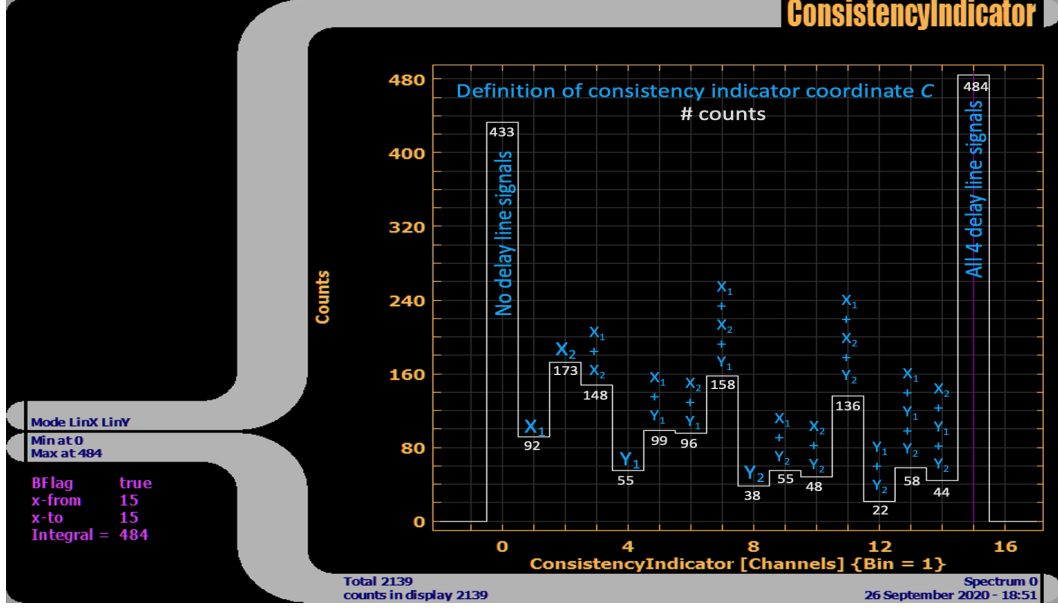


Figure 5.5 : Screenshot of CoboldPC spectrum 0 with the definitions of each value of C shown in blue and the number of counts for each value of C labelled in grey.

u in Equation 5.3 becomes 0, resulting in non-zero counts in $C = 0$ channel. This is often caused by the electrical noise generated during the ejection of the ions from the trap and the subsequent pulsing of the TOF A electrode. If the MCP signal is followed by hits on the delay lines, u in Equation 5.3 becomes 1, and $C = 1$ to 15 starts getting populated depending on which delay lines have registered the hits. $C = 1$ to 14 show the number of times when any one, two or three, (but not all four) of the delay lines have signals, and these can be used to set the thresholds of the CFD. A *clean* hit is defined when there are signals on all four delay lines, X_1 , X_2 , Y_1 , and Y_2 , that is a non-zero number of counts in $C = 15$. This is the definition of the condition *four* wherein the C must have a value between 14.5 and 15.5. It should be noted that if a high threshold is set at the CFD on the MCP signal, nothing will show up on this spectrum at all. High thresholds on the individual delay lines could also result in throwing away real data. On the other hand, a low threshold will render a high number of counts on this spectrum but will not necessarily pose a problem as these can be gated by the different conditions, discussed later in this section. The main use of this spectrum is to tune the CFD thresholds of the MCP and delay line signals in order to maximize the number of counts in $C = 15$. A sample spectrum 0 of CoboldPC is shown in Figure 5.5. The non-zero counts in all of $C = 1$ to 14 indicates that the thresholds of the

Table 5.4 : CoboldPC Spectrum 0: Consistency Indicator

Delay line signals	i	$C = \sum_i u \cdot 2^{(i-1)}$	Counts in Fig. 5.5
None	—	0 ($u = 0$)	433
X_1	1	1 ($= 2^0$)	92
X_2	2	2 ($= 2^1$)	173
$X_1 + X_2$	1 & 2	3 ($= 2^0 + 2^1$)	148
Y_1	3	4 ($= 2^2$)	55
$X_1 + Y_1$	1 & 3	5 ($= 2^0 + 2^2$)	99
$X_2 + Y_1$	2 & 3	6 ($= 2^1 + 2^2$)	96
$X_1 + X_2 + Y_1$	1, 2 & 3	7 ($= 2^0 + 2^1 + 2^2$)	158
Y_2	4	8 ($= 2^3$)	38
$X_1 + Y_2$	1 & 4	9 ($= 2^0 + 2^3$)	55
$X_2 + Y_2$	2 & 4	10 ($= 2^1 + 2^3$)	48
$X_1 + X_2 + Y_2$	1, 2 & 4	11 ($= 2^0 + 2^1 + 2^3$)	136
$Y_1 + Y_2$	3 & 4	12 ($= 2^2 + 2^3$)	22
$X_1 + Y_1 + Y_2$	1, 3 & 4	13 ($= 2^0 + 2^2 + 2^3$)	58
$X_2 + Y_1 + Y_2$	2, 3 & 4	14 ($= 2^1 + 2^2 + 2^3$)	44
$X_1 + X_2 + Y_1 + Y_2$	1, 2, 3 & 4	15 ($= 2^0 + 2^1 + 2^2 + 2^3$)	484

CFD are set in a way that no real signal is screened out. The counts in $C = 15$ means there are 484 times when all 4 delay lines had hits. These can still include some noise and/or dark counts, but they do not pose a problem as those can be gated out by implementing different conditions or data-cuts. The spectrum is further explained in Table 5.4.

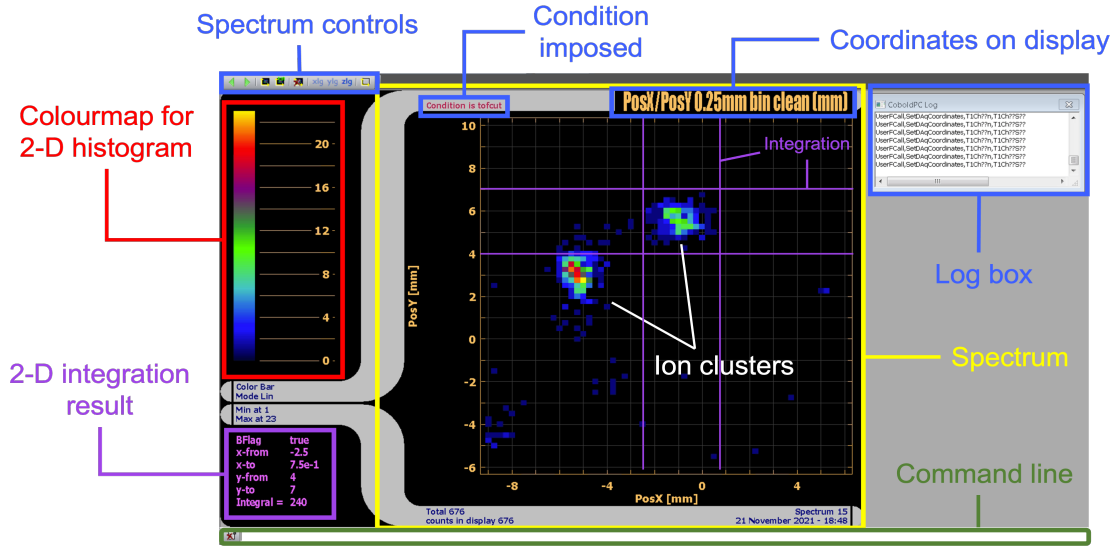
Since the delay lines have a fixed length, the total propagation time along any dimension ($sumX$ and $sumY$) should be independent of the location of the ion hit and should be more-or-less constant. For the CPT PS-MCP detector, $sumX \approx 46.5$ ns and $sumY \approx 45.5$ ns, with FWHM of 0.25 ns per peak, and the slightly larger $sumX$ value is due to the wires between the detector and the FT12TP being of slightly different lengths [53]. Multiple conditions can be combined to create new conditions in the *startup* script. For example, $sumX$ and $sumY$ cuts can be combined to create a $sumXY$ -cut. If some noise signal would make it through

the CFD veto, the CFD thresholds and hence the condition *four*, and the *sumXY*-cut is used to gate them out. Other useful conditions are *RAD-cut* (to select ions excited to a certain orbit) and *TOF-cut* (for ions with TOF within a certain interval).

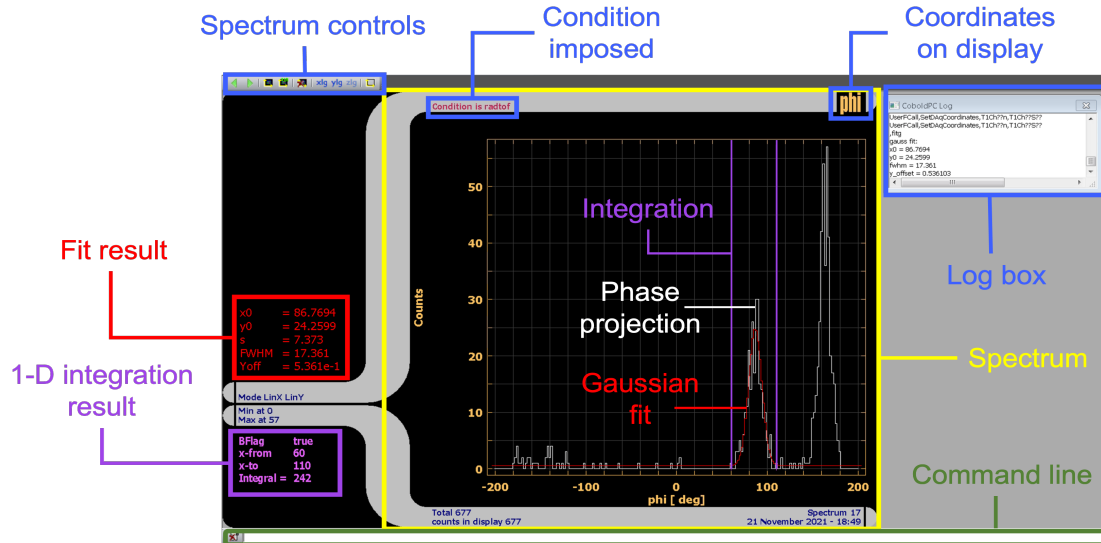
The two most commonly used spectra are a 2-D histogram showing the position of the ion hits (spectrum 15) and a 1-D projection of the angular position of the ions in their orbit in the trap (spectrum 17). Screenshots of these two spectra are shown in Figure 5.6, along with the key features labelled. On the top left corner, a panel labelled “Spectrum controls” allows the experimenter to navigate through the different spectra, zoom out of a spectrum, change to log scales, and finally reset all the spectra. The top right shows the coordinates on display in that spectrum, while the imposed conditions are shown in the top middle. On the left of spectrum 15, a heat map or colormap of the number of ion hits is displayed. At the bottom, the command line allows one to execute different commands to extract information. The log box on the right keeps a log of all the commands. CoboldPC has several commands built in, and allows the addition of user-defined commands. Some of the most commonly used commands are:

- **irish:** Calculates the total rate of ion hits on the spectrum on display since the beginning of DAQ and displays it on the CoboldPC log text-box.
- **fitg:** Allow to fit a 1-D histogram with a Gaussian distribution and show the result on the left-center, as shown by the red box in Figure 5.6(b).
- **int:** Used to integrate both 1-D and 2-D spectra, to calculate the number of ion-hits in an ion cluster. The user sets the boundaries of the integration, as shown by the purple lines on the spectra in Figures 5.6(a) and 5.6(b), and the result is displayed to the lower left, shown in purple box.
- **exp:** Used to zoom into a spectrum.

These functions can be used in both the online and offline modes. Other spectra like 1-D projections of absolute time of an event since the beginning of DAQ (spectrum 1), X_1 (spectrum 3), X_2 (spectrum 4), Y_1 (spectrum 5), Y_2 (spectrum 6), *sumX* (spectrum 7), *sumY*



(a) Screenshot of CoboldPC spectrum 15



(b) Screenshot of CoboldPC spectrum 17

Figure 5.6 : Screenshot of CoboldPC spectra 15 and 17 with the different features labelled. Spectrum 15 is a 2-D histogram showing the XY positions of two ion clusters, while spectrum 17 is a 1-D histogram showing the phase projection of the same two ion clusters. Also shown are integration results for the 87 degree cluster in both spectra, with a Gaussian fit for the 1-D phase projection.

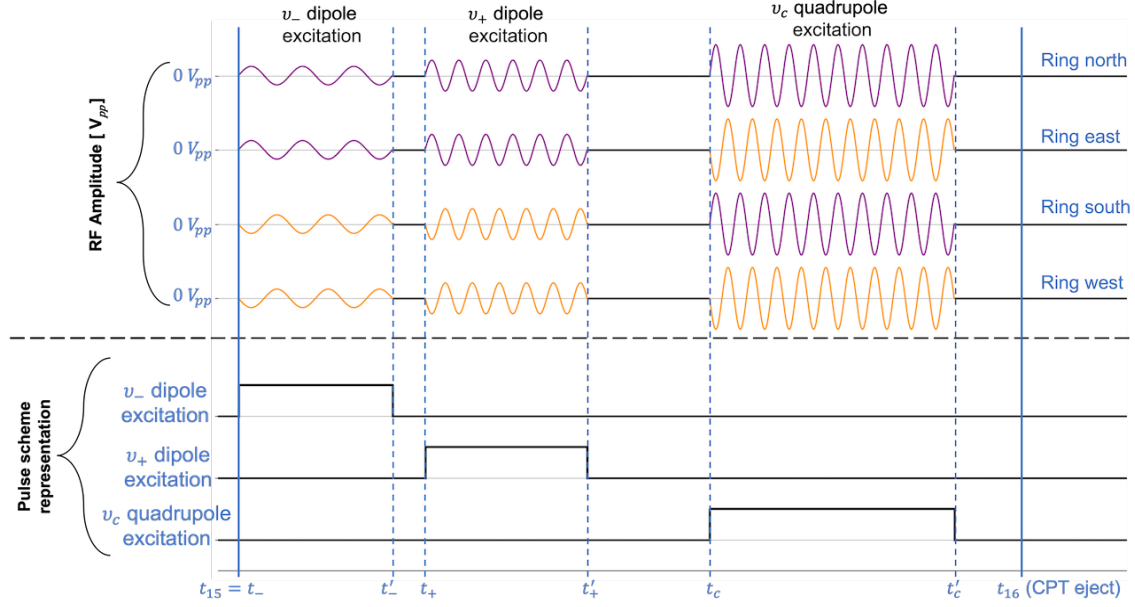


Figure 5.7 : Pulse scheme representation of RF excitations applied to the different quadrants of the ring electrode. The durations of the excitations are not to scale. Description in text.

(spectrum 9), position X (spectrum 11), position Y (spectrum 12), ions' TOF (spectrum 8) and radius of the ion hits (spectrum 18) are useful for tuning and troubleshooting.

When the CPT is operated in flythrough mode for troubleshooting, the CFD veto and the TDC channel # 7 signal are taken from the linear trap eject instead of the CPT ejection signal from the Jorway timing module. This has no direct relevance to PI-ICR measurements, and is only included to complete the picture of the detection system.

5.2 Measuring the ions' radial frequencies

Once the ions are trapped in the CPT and the high energy ions have been removed (see sections 4.4 and 4.4.1), the RF excitations required for PI-ICR measurements are initiated (t_{15} in Figure 4.9). These include dipole excitations at the ions' ν_- and ν_+ frequencies and quadrupole excitation at their ν_c frequency, the effects of which are illustrated in Section 2.3. Here I will discuss how they are used for PI-ICR measurements at the CPT. A four channel arbitrary waveform generator from Tabor Electronics (model WW1074) [116] is used to program four waveforms for the four ring quadrants. In the subsequent sections, I will present the RF excitation using a pulse scheme representation, as shown in Figure 5.7.

Figure 5.7 can be seen as made of two parts, separated by a black horizontal dashed

line. The first or top part shows the waveforms of the RF excitations as applied to the ring quadrants. Recall from Section 2.3 that dipole excitations are applied such that opposite ring electrodes are 180° out-of-phase, while in quadrupole excitations, the adjacent electrodes are 180° out-of-phase. This is shown with the purple and orange waveforms, where the same color indicates the same phases of the applied excitations. The black horizontal lines in between the excitations or after the ν_c excitation symbolize periods when no RF excitations are applied. A PI-ICR measurement starts with the start of PI-ICR excitations (t_{15}) and stop with the ejection of the ions (t_{16}). These are shown with the two blue continuous vertical lines. t_{15} is also when the first RF excitation (ν_- dipole) is applied, that is $t_{15} = t_-$. The blue dashed vertical lines depict the end of the ν_- dipole excitation (t'_-), and the start and stop of the ν_+ dipole (t_+ and t'_+) and ν_c (t_c and t'_c) quadrupole excitations. As before, $t'_i = t_i + \Delta t_i$, where $i = -, +, c$. The bottom part of Figure 5.7 shows the same three excitations in pulse scheme representation.

As mentioned in Section 2.4.2, PI-ICR relies on measuring the phase acquired by the ions while undergoing one of its eigenmotions, ν_- or ν_+ . This acquired phase is determined from two positions, the reference and the final. That means measuring any frequency would require at least a reference and a final file. Mass measurements require knowing ν_c precisely, which can be determined by measuring ν_- and ν_+ individually, and using equation 2.43a. However, that would require two sets of reference-final datasets. That could be challenging given most of the isotopes measured as part of this thesis are some of the most weakly produced isotopes from ^{252}Cf fission, sometimes being recorded at 10 – 20 ions per hour. However, in the scheme of the direct measurement of ν_c , as described in Section 5.2.1, this can be achieved with just two datasets. Additionally from equation 2.80b, the uncertainty in determining ν_c from measuring ν_- and ν_+ will be $\sim \sqrt{2}$ times the uncertainty from measuring ν_c directly. For these reasons, all the masses presented in this thesis are measured using the direct ν_c measurement technique and I will describe that technique first, before discussing the measurement procedures of the individual eigenfrequencies.

Before I explain the measurement procedures, I would like to clarify the sign convention used. The projection of the eigenmotions of the ions on the detector is clockwise. For the derivations here, I will take the counter-clockwise direction as positive, as per the normal

convention. Therefore, the phases acquired by the eigenmotions in this convention will be negative. One other thing should be mentioned here that the catch-center-release technique to determine the trap center, mentioned in Section 2.4.2 is not followed at the CPT. The reason for that and the center measurement technique will be described in Section 5.5.6.

5.2.1 Direct measurement of cyclotron frequency

As mentioned, through PI-ICR, one can measure a certain frequency of the ions from the phase they accumulate while undergoing that motion. But since ν_c is not an eigenmotion of the ions, this is not possible to do for ν_c measurement. Recall from equation 2.43a that the ions' ν_c is related to their eigenmotions (ν_- and ν_+) as $\nu_c = \nu_- + \nu_+$. Therefore, the ions' ν_- and ν_+ motions are manipulated such that they accumulate the corresponding phases and then equation 2.43a is used to find ν_c . The excitation scheme for the reference and final spots for such a measurement is shown in Figure 5.8(a), with a horizontal black dashed line separating the two.

Once the PI-ICR measurements are initiated, the first excitation applied is a ν_- dipole excitation, starting at time $t_{15} = t_-$ and ending at time t'_- . The purpose of this excitation is to center the ions. For that, the phase, peak-to-peak amplitude and duration of the excitation are carefully tuned (see equation 2.57 and Figure 2.9(b)), such that the ions' $r_- \approx 0$. Next, the ions are excited to a radius by a ν_+ dipole excitation (see Figure 2.9(a)), starting at time t_+ and ending at time t'_+ . At the end of this excitation, the ions have a ν_+ motion. As ν_+ is a fast motion, its projection on the detector would be imaged as a ring instead of a spot. Therefore to get an image spot on the detector, the ν_+ motion has to be converted to the slow ν_- motion. This is done by applying a ν_c quadrupole excitation (see Figure 2.10). The durations of all three excitations and ejection time (t_{16}) are kept the same for both the reference and the final spot. The exact time of application of the ν_c excitation differentiates the two spots. For the reference spot, the ν_c excitation is applied right after the end of the ν_+ excitation, where as for the final spot, there is a delay (t_{acc}) between the two.

Since the centering pulse reduces the magnetron radius to ≈ 0 , the ions' phase at the end of the ν_+ excitation (at time t'_+), when they have only ν_+ motion, for both the reference and final spots, is given by:

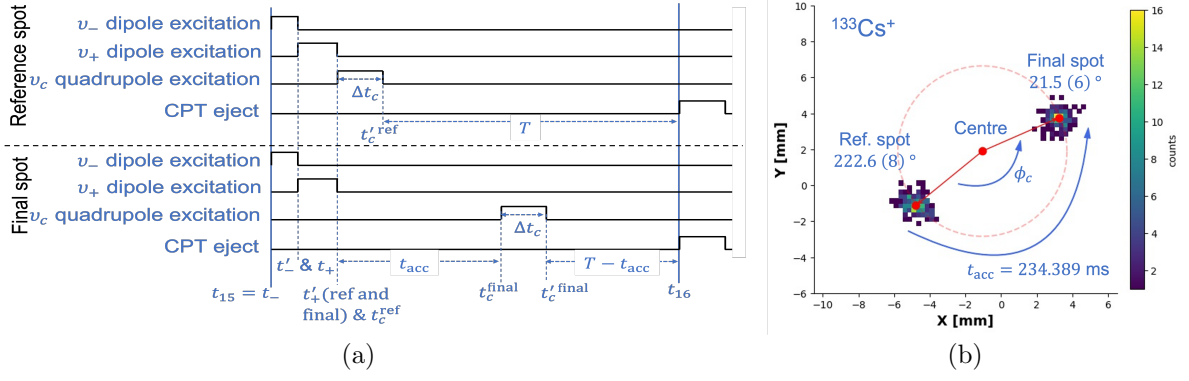


Figure 5.8 : ν_c measurement using PI-ICR. Panel (a) shows the RF excitations for ν_c measurement. Panel (b) is an illustration of the reference and final spots, superimposed on the same spectrum, along with the phase ϕ_c acquired over a time t_{acc} .

$$\phi_{t'_+}^{\text{ref}+} = \phi_{t'_+}^{\text{final}+} = \phi_0^+ \text{ (say), and} \quad (5.4a)$$

$$\phi_{t'_+}^{\text{ref}-} = \phi_{t'_+}^{\text{final}-} = 0. \quad (5.4b)$$

For the reference spot, the conversion ν_c excitation is applied at this point. Recall from equation 2.67b, the phase evolution during the ν_c quadrupole excitation is $\phi_{\pm}(t_{\text{conv}}) = \phi_Q + 2\pi\nu_{\pm}t_{\text{conv}} - \phi_{\mp}(0)$. Here, $t_{\text{conv}} = \Delta t_c$. Therefore, at the end of the conversion pulse, the ions' phase is:

$$\phi_{t'_c}^{\text{ref}-} = \phi_Q + 2\pi\nu_- \Delta t_c - \phi_0^+. \quad (5.5)$$

Between the ν_c excitation and their ejection from the trap, the ions acquire ν_- phase over a time T . Therefore, the total reference phase of the ions at ejection, as imaged on the PS-MCP, is:

$$\begin{aligned} \phi^{\text{ref}} &= \phi_{t'_c}^{\text{ref}-} - \left(2\pi\nu_- T - 2\pi N_-(T)\right) \\ &= \phi_Q + 2\pi\nu_- \Delta t_c - \phi_0^+ - \left(2\pi\nu_- T - 2\pi N_-(T)\right), \end{aligned} \quad (5.6)$$

where $N_-(T)$ is the number of complete revolutions the ions ν_- motion has undergone in time T . As it can be seen, the reference phase is essentially a ν_- phase, which is a mass-independent motion. Therefore, both for pure beam with one species, or impure beam with multiple species, the reference spot spectrum will always consist of one spot.

For the final spot, between the end of the ν_+ pulse and the start of the ν_c pulse, the ions acquire a ν_+ phase over time t_{acc} . Therefore, the ions' phase at time t_c^{final} is given by:

$$\phi_{t_c}^{\text{final}} = \phi_0^+ - \left(2\pi\nu_+ t_{\text{acc}} - 2\pi N_+(t_{\text{acc}}) \right). \quad (5.7)$$

$N_+(t_{\text{acc}})$ is the number of complete turns the ions ν_+ motion has undergone in time t_{acc} . After the conversion pulse, the ions' phase will be:

$$\phi_{t_c'}^{\text{final-}} = \phi_Q + 2\pi\nu_- \Delta t_c - \phi_0^+ + \left(2\pi\nu_+ t_{\text{acc}} - 2\pi N_+(t_{\text{acc}}) \right). \quad (5.8)$$

As with the reference spot, between the conversion excitation and their ejection from the trap, the ions acquire ν_- phase, but this time the time of accumulation is reduced by t_{acc} , that is they accumulate the ν_- phase over time $T - t_{\text{acc}}$. This makes the ions' final phase:

$$\begin{aligned} \phi^{\text{final}} &= \phi_{t_c'}^{\text{final-}} - \left(2\pi\nu_-(T - t_{\text{acc}}) - 2\pi N_-(T - t_{\text{acc}}) \right) \\ &= \phi_Q + 2\pi\nu_- \Delta t_c - \phi_0^+ + \left(2\pi\nu_+ t_{\text{acc}} - 2\pi N_+(t_{\text{acc}}) \right) \\ &\quad - \left(2\pi\nu_-(T - t_{\text{acc}}) - 2\pi N_-(T - t_{\text{acc}}) \right). \end{aligned} \quad (5.9)$$

The final phase is a mixture of the mass-independent ν_- phase and a mass-dependent ν_+ phase. This ν_+ phase is acquired over time t_{acc} . Therefore, for an impure beam with multiple species, a final spot spectrum will consist of multiple spots as the different species with different masses will acquire different ν_+ phases. As t_{acc} is increased, the separation (in other words, resolution) between such close-massed species will also increase. This will be discussed more in Section 5.4.2.

Between the reference and the final spot, the difference in phase acquired by the ions is:

$$\begin{aligned} \phi^{\text{final}} - \phi^{\text{ref}} &= \left(2\pi\nu_+ t_{\text{acc}} - 2\pi N_+(t_{\text{acc}}) \right) - \left(2\pi\nu_-(T - t_{\text{acc}}) - 2\pi N_-(T - t_{\text{acc}}) \right) \\ &\quad + \left(2\pi\nu_- T - 2\pi N_-(T) \right) \\ &= 2\pi(\nu_+ + \nu_-)t_{\text{acc}} - 2\pi \left(N_+(t_{\text{acc}}) + N_-(t_{\text{acc}}) \right). \end{aligned} \quad (5.10)$$

Since $N_-(T - t_{\text{acc}})$ is the number of complete turns the ν_- motion completes in time $(T - t_{\text{acc}})$, one can write $N_-(T - t_{\text{acc}}) = N_-(T) - N_-(t_{\text{acc}})$. Again, from $\nu_c = \nu_+ + \nu_-$ (ν_+ and ν_- being

in the same direction and both motions being at the same radius), $N_+(t_{\text{acc}}) + N_-(t_{\text{acc}}) = N_c(t_{\text{acc}})$. Therefore equation 5.10 can be rewritten as:

$$\phi^{\text{final}} - \phi^{\text{ref}} = 2\pi\nu_c t_{\text{acc}} - 2\pi N_c(t_{\text{acc}}) = \phi_c . \quad (5.11)$$

In other words, $\phi^{\text{final}} - \phi^{\text{ref}}$ can be thought of the excess phase (ϕ_c) acquired by the ions' ν_c motion in the counter-clockwise direction in time t_{acc} after completing $N_c(t_{\text{acc}})$ number of complete turns. This is shown in Figure 5.8(b). From equation 5.11, one can find ν_c as:

$$\nu_c = \frac{\phi_c + 2\pi N_c(t_{\text{acc}})}{2\pi t_{\text{acc}}} . \quad (5.12)$$

Equation 5.12 shows how ν_c can be measured directly from the reference and final spots using the excitation scheme shown in Figure 5.8(a). $N_c(t_{\text{acc}})$ can be found from a guess ν_c . For isotopes whose masses have been previously measured, this ν_c -guess can be calculated easily from equation 2.25. For unknown species or isotopes whose masses are not known precisely, ν_c -guess has to be determined first. The technique will be discussed in Section 5.4.1. Figure 5.8(b) illustrates a ν_c measurement with a reference and a final spot with $t_{\text{acc}} = 234.389$ ms, using a pure beam of $^{133}\text{Cs}^+$ ions, superimposed on the same spectrum. The number of complete turns N_c , calculated with a guess ν_c of 674999.6 Hz, is 158212, with a phase excess of $159 (1)^\circ$ and the ν_c can be found as 674999.43 (1) Hz. The uncertainty quoted here is only statistical from a single set of measurements, a detailed discussion of the systematic contributions to the uncertainty in measurements at the CPT is included in Section 5.5.

The beauty of this method lies in its simplicity. The timing of the ν_- pulse ($t_{15} = t_-$) sets the ions' phase when the excitation is applied. This timing can be tuned so that the phase difference between that of the excitation and the ions' motions $\phi_{D-} \approx 3\pi/2$. ν_- being an almost mass-independent frequency, the ν_- pulse timing, amplitude, and width can be tuned beforehand using the SIS. The injection settings into the CPT (electrostatics beam-line components like lenses and steerers) are set in a way that the ν_- pulse stays the same for all the masses and the calibrants measured during a campaign. For this reason, t_{15} is not changed every time the A/q of the beam is changed. Typical duration of the ν_- pulse was kept at ~ 4.4 ms with an amplitude of $0.0015 V_{pp}$ for all the measured masses presented

here. The phases of all three excitations (ν_- , ν_+ and ν_c) are kept at 0° . The amplitudes and durations of the ν_+ and ν_c are tuned offline before a campaign. The ν_+ pulse is tuned such that the ions are excited to a considerable orbit without hitting the trap electrodes or the 5 mm aperture on their way out. An optimized ν_c pulse ensures that a complete conversion between the ν_+ and ν_- motions of the ions has taken place before their ejection. During online measurements with the beam from CARIBU comprising of multiple isobaric species, they all need to be excited to the same radius, and total conversion between their ν_+ and ν_- needs to happen. This is accomplished by applying excitations with a wide bandwidth, which is attained by keeping the amplitudes of the excitations comparatively large (limited by the $10 V_{pp}$ limit of the Tabor function generator), and their durations very small. Typical amplitude and duration for the ν_+ and ν_c pulses are $5 V_{pp}$ and $153 \mu s$, and $10 V_{pp}$ and $338 \mu s$ respectively, setting bandwidths of ~ 10 kHz for the ν_+ pulse and ~ 2 kHz for the ν_c pulse, which correspond to mass ranges of $\sim 1800 \text{ MeV}/c^2$ and $\sim 300 \text{ MeV}/c^2$ respectively. Once these settings are optimized offline, they are maintained for every measurement irrespective of the A/q of the trapped ions. During a measurement campaign, after fixing the ν_+ and ν_c durations to the pre-set optimized values, the experimenter would first set an ejection time t_{16} , that is the total time the ions would spend in the trap, which stays constant throughout the measurement. Next, one would change t_{acc} from 0 for reference spots to some non-zero time for final spots within the limits imposed by the set t_{16} , so that the ν_c pulse is completed before ejection of the ions ($t'_c \leq t_{16}$). If some measurement requires one to set a large t_{acc} , such that the ions would get ejected before the ν_c excitation can be completed ($t'_c \geq t_{16}$), the ejection trigger time (t_{16}) is increased first ensuring $t'_c \leq t_{16}$ is maintained. Since t_{16} is dictated by the requirements of a measurement, and t_{17} essentially follows t_{16} , these two are not scaled with the A/q the beam.

5.2.2 Magnetron frequency measurement

Measuring ν_- using PI-ICR is straightforward. Recall from Figure 2.9(b), a ν_- dipole excitation, irrespective of the phase of the excitation, if applied for a long enough duration, will lead to an increase of the ions' r_- . At this radius, the ions can acquire phases corresponding to two accumulation times, and then ν_- can be determined from the difference in

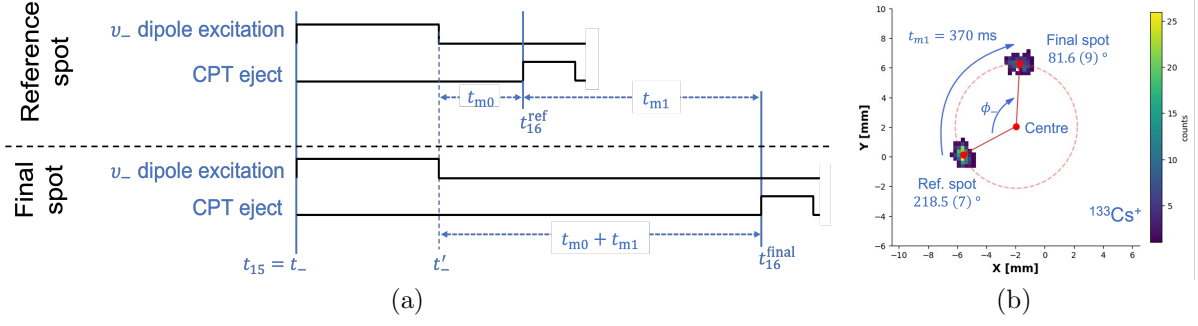


Figure 5.9 : ν_- measurement using PI-ICR. Panel (a) shows the excitation scheme for ν_- measurement. Panel (b) is an illustration of the reference and final spots, superimposed on the same spectrum, along with the phase ϕ_- acquired over a time t_{m1} .

the acquired phases and the times. This is shown in Figure 5.9(a).

A long ν_- excitation is applied first, for both the reference and final spots. For the reference spot, the ions are ejected from the trap after time t_{m0} from the end of the ν_- excitation, making the reference phase:

$$\phi^{\text{ref}} = -2\pi \left(\nu_- t_{m0} - N_-(t_{m0}) \right). \quad (5.13)$$

For the final spot, the ejection of the ions was delayed by an additional time t_{m1} , that is the ions acquire a ν_- phase over time $(t_{m0} + t_{m1})$. Therefore:

$$\phi^{\text{final}} = -2\pi \left(\nu_- (t_{m0} + t_{m1}) - N_-(t_{m0} + t_{m1}) \right). \quad (5.14)$$

The acquired phase is:

$$\phi^{\text{final}} - \phi^{\text{ref}} = -2\pi \left(\nu_- t_{m1} - N_-(t_{m1}) \right) = -\phi_-. \quad (5.15)$$

ϕ_- is the excess phase in clockwise direction after the ions have completed $N_-(t_{m1})$ complete turns. This is shown in Figure 5.9(b). Therefore:

$$\nu_- = \frac{\phi_- + 2\pi N_-(t_{m1})}{2\pi t_{m1}}. \quad (5.16)$$

The example shown in Figure 5.9(b) was done with $t_{m1} = 370$ ms, right after the CPT magnet was energised in 2018. The magnetic field was measured to be around 5.84 T using

an NMR probe. Using equation 2.48, that is

$$\nu_- = \frac{\omega_-}{2\pi} = \frac{V_0}{4\pi B_0 d_0^2} ,$$

with a trap depth $V_0 = V_{\text{endcap}} - V_{\text{ring}} = 9.637$ V, $B_0 = 5.84$ T and a characteristic trap dimension $d_0 = 0.9145$ cm, ν_- was calculated to be 1570 Hz. Using this guess ν_- of 1570 Hz, $N_-(t_{m1})$ was found to be 580. Therefore, with a phase excess of $136.9(14)^\circ$, ν_- was measured to be 1571.32 (1) Hz.

5.2.3 Reduced cyclotron frequency measurement

At the CPT, ν_+ is not measured using PI-ICR. Knowledge of ν_+ of the ions is only required for application of the ν_+ RF excitation. Because of the ~ 10 kHz bandwidth of the ν_+ excitation, calculating it from the relationship $\nu_+ = \nu_c - \nu_-$ is adequate. Therefore a detailed discussion of PI-ICR ν_+ measurement is not included.

5.3 Analysis

All the data analyses for this thesis have been done in Python using mainly the standard libraries *struct* [128] (to read the data from the LMF files), *numpy* [129], *scipy* [130], *math* [131] and *pandas* [132] (for calculations and sorting data), *lmfit* [133] (for all fitting done as part of the analysis), and *matplotlib* [134] (for generating plots).

Offline analysis starts with reading the LMF file and extracting the data. In the past, a C++ -based program *lmf2txt.exe* was used which would convert the LMF files to a *.txt* file containing a header with the DAQ start and stop times, and a timestamped event-by-event list of all delay-line signals including the TOF calculated between the MCP hit and the last CPT eject [53]. A Python function called *read_lmf* was later written by this author in collaboration with post-doctoral associate D. Gorelov, which first extracts the data from the LMF files, calculates the positional coordinates using equations 5.1 and 5.2, imposes different user-defined data cuts (namely *ion-cut*, *sumXY-cut*, *TOF-cut* and *RAD-cut*), displays the data, and finally outputs arrays of sorted data as per the user's instructions for subsequent calculations and analysis. *ion-cut* is an offline condition imposed to limit ion-ion interaction, by discarding data when more than a user-defined number of ions were in the trap at the same time. This is done by grouping all the ion hits with timestamps within a certain

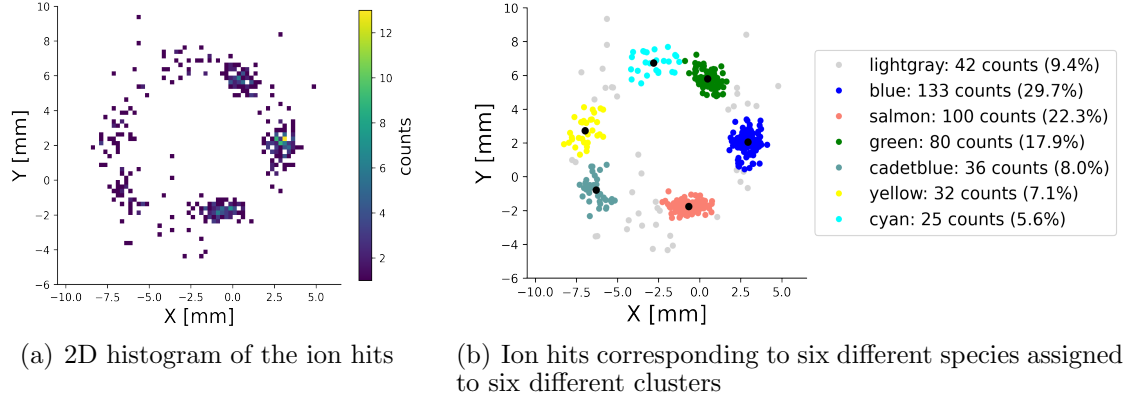


Figure 5.10 : Assignment of ion hits to clusters using *meanshift* algorithm, and finding the cluster centers from the Gaussian fits of the x and y projections.

interval depending on the cycle of measurement. The *read_lmf* provided a more general and user-friendly alternative to the original *lmf2txt.exe* program as it could be used across any computer operating system, and easily tweaked by users according to their needs by changing certain input limits for the different datacuts.

The next step is identifying ion groups, assigning them into spots or “clusters”, and finding the centroids of such clusters. This procedure is called “clustering”. For reference spots, or pure beams with one species, clustering is trivial. However for a beam consisting of multiple species (see Figure 3.10), this can get complicated. One option is individually selecting a lower and upper limit for x and y for each cluster and treating the ion hits within those limits as a single cluster. But this could be time consuming and subjective of the user’s choice of assigning limits to the clusters. Therefore a simple clustering algorithm was used. A function *cluster_spots* was written by R. Orford for the purpose [53]. *Cluster_spots* uses the *mean shift* clustering algorithm [135], part of the Python machine learning library *scikit-learn* [136]. Meanshift assigns different data points to different clusters by iteratively shifting individual cluster centroids towards the modes, or points of highest densities in the dataset [136, 137], depending on the optional maximum distance between adjacent modes and the minimum density of each such cluster. This is shown in Figure 5.10, where a beam of $A/q = 168/2+$ consisting of multiple species are clustered.

An array of the (x, y) coordinates of the individual ion hits, extracted from an LMF file, are fed into the *cluster_spots* function along with the maximum radius of each cluster, and

the minimum spot density. Figure 5.10(a) shows a 2D histogram of the coordinates of such individual ion hits. The *meanshift* first assigns individual ion hits to the different clusters and stores the corresponding x and y coordinates in arrays. Next the individual coordinates are projected onto the respective axes, binned into histograms using the Freedman-Diaconic rule as:

$$\text{bin width} = \frac{2 \text{ IQR}}{\sqrt[3]{n}} , \quad (5.17)$$

(IQR being the interquartile range, defined as the difference between the third and the first quartile), and fitted using a Gaussian distribution. The cluster centers are then obtained from the fits. This is shown in Figure 5.10(b), where the clusters assigned by the *meanshift* algorithm using a cluster-radius of 1.5 mm and a minimum density of 5 % are shown encoded in different colors. The cluster centers are shown with black spots in every cluster. Once the cluster center is found, the angle of the corresponding cluster with respect to the trap center can be easily calculated. From the angles, the phase acquired by the ions between the reference and the final spots can be determined and hence the ions' frequencies can be measured.

5.4 Measurement procedures and features

5.4.1 Particle identification (PID) : finding frequencies of unknown species

The first steps to ν_c measurement is finding and confirming the identity of the target nuclide, and obtaining a ν_c -guess. From equation 2.80b, we can see that increasing the accumulation time will lead to lower uncertainty and hence higher precision in measurement. On the other hand, from equation 5.12, the shift in the determined frequency from the number of turns is:

$$\Delta\nu_c = \frac{\Delta N_c}{t_{\text{acc}}} . \quad (5.18)$$

Therefore, 1 less or more turn would lead to a frequency shift of $1/t_{\text{acc}}$ Hz. For example, if $t_{\text{acc}} = 200$ ms, $\Delta\nu_c \approx 5$ Hz, which would correspond to a mass shift of ~ 600 keV/c². So without having a precise ν_c -guess, one cannot unequivocally determine N_c , especially at large t_{acc} , which could compromise the accuracy of measurement. In other words, to make precise and accurate ν_c measurements, one has to have a relatively accurate knowledge of

the ν_c beforehand. The trick is to start with a small t_{acc} and gradually increase it as one gains more and more precise and accurate values for ν_c . This is particularly necessary when measuring nuclides for which no prior experimental mass-data are available. For an impure beam, this method is also used to identify the frequencies of every species present in the beam, which is required for a phase correction to correct for a systematic effect, discussed in Section 5.5.7. For example, let us consider the case of the measurement of the mass of ^{152}Ce , as illustrated thorough Figure 5.11, and Tables 5.5 and 5.6.

Table 5.5 : Identification of ν_c of all species present in a beam of $152/2+$ ions. The frequencies, in Hz, represent the different spots for different numbers of turns around a nominal guess N_0 obtained from the AME2016 extrapolated mass.

t_{acc}	5 ms	35.007 ms		73.038 ms	83.010 ms	
Angles	92.3°	155.1°	322.1°	151.5°	141.9°	286.7°
$N_0 - 3$	1180341.4(8)	1180818.2(1)	1180830.2(1)	1180863.28(6)	1180868.15(5)	1180872.68(6)
$N_0 - 2$	1180541.4(8)	1180846.8(1)	1180858.9(1)	1180876.98(6)	1180880.19(5)	1180884.72(6)
$N_0 - 1$	1180741.4(8)	1180875.4(1)	1180887.4(1)	1180890.67(6)	1180892.24(5)	1180896.77(6)
N_0	1180941.4(8)	1180904.0(1)	1180915.9(1)	1180904.36(6)	1180904.29(5)	1180908.82(6)
$N_0 + 1$	1181141.4(8)	1180932.5(1)	1180944.9(1)	1180918.05(6)	1180916.33(5)	1180920.86(6)
$N_0 + 2$	1181341.4(8)	1180961.1(1)	1180973.1(1)	1180931.75(6)	1180928.38(5)	1180932.91(6)
$N_0 + 3$	1181541.4(8)	1180989.6(1)	1181001.6(1)	1180945.23(6)	1180940.43(5)	1180944.95(6)

Table 5.6 : Validation of $^{152}\text{Ce}^{2+}$ and $^{152}\text{Pr}^{2+}$. As before, the frequencies, in Hz, corresponding to the different spots at different angles for different number of turns.

t_{acc}	110.727 ms		237.656 ms	
Angles	189.0°	21.5°	196.1°	102.1°
N_0	1180904.27(5)	...	1180904.35(4)	...
...	...	...	...	...
$N_0 + 5$	...	1180944.79(3)	...	...
...	...	...	...	...
$N_0 + 10$	...	...	...	1180945.26(4)

The BNG was centered on the $^{152}\text{Ce}^{2+}$ ions with a 300 ns width, meaning any species with mass within $\sim 30 \text{ MeV}/c^2$ of the Ce was let through. The mass of ^{152}Ce was never measured before and the AME2016 [44] reported an extrapolated uncertainty of 200 keV/ c^2 .

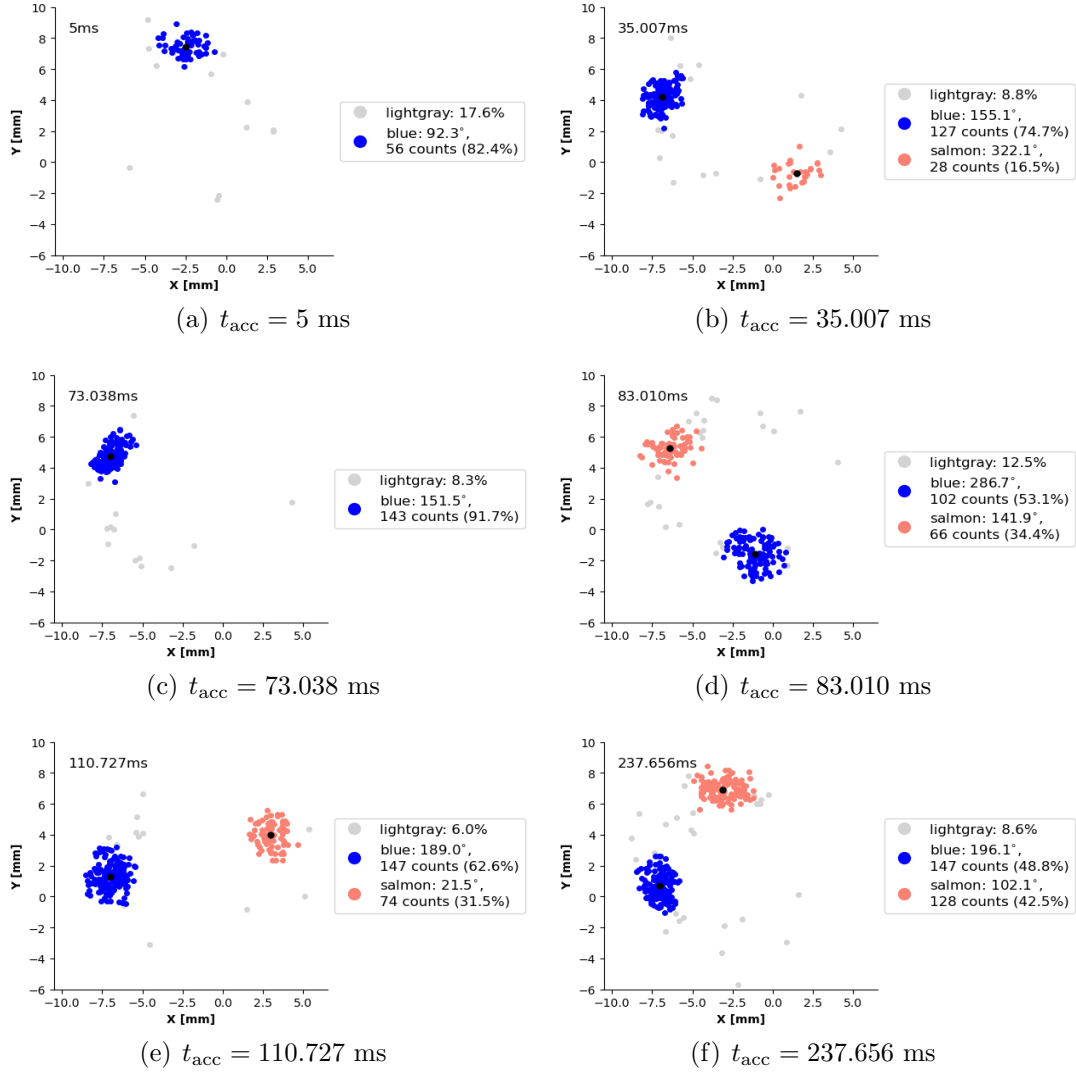


Figure 5.11 : Particle identification in a beam of $152/2+$. The target nuclide was $^{152}\text{Ce}^{2+}$. Its identity was confirmed and some $^{152}\text{Pr}^{2+}$ was also found to be present. It should be noted that the color of the clusters represent the relative population in the spots, and not the ion species. For example, blue represents the cluster with the maximum population followed by salmon, while grey represents the unclustered ion hits.

So, the ν_c from the AME2016, 1180906.1 Hz, was an unreliable choice of ν_c -guess to start a precision measurement using large values t_{acc} -s. The measurement began with a small t_{acc} of 5 ms. At 5 ms, $\Delta\nu_c$ between successive turns is 200 Hz, which corresponds to a mass of ~ 48 MeV/c². Therefore, at 5 ms, the ν_c -guess from AME2016 can be used to get an accurate number for N_c . N_0 in the first column of Table 5.5 represents the number of turns calculated with the guess frequency. The frequency values that result from varying this guess by ± 3 turns are shown in subsequent rows.

At 5 ms, only a single ion spot was observed. The corresponding frequency was close to $^{152}\text{Pr}^{2+}$, measured at the CPT using TOF-ICR [64]. Next, a number of slightly larger values of t_{acc} were tried. At 35.007 ms, we see the single spot, observed originally with a t_{acc} of 5 ms, splitting into two. For this and for longer values of t_{acc} we see two frequencies appearing: ~ 1180904.3 Hz and ~ 1180945.0 Hz, corresponding to $^{152}\text{Ce}^{2+}$ and $^{152}\text{Pr}^{2+}$. These are highlighted in Tables 5.5 and 5.6. Note that at $t_{\text{acc}} = 73.038$ ms (Figure 5.11(c)), only one spot was observed because at this t_{acc} , the Ce and Pr end up at the same spot, but on different turns. This can be further seen from the two highlighted frequencies in the 73.038 ms column of Table 5.5. To validate, we tried two more larger values of t_{acc} , and using the Ce ν_c from the 83.010 ms file, observed the same two species, as shown in Table 5.6. To minimize cluttering the Table with numbers, I have entered only the frequencies of $^{152}\text{Ce}^{2+}$ and $^{152}\text{Pr}^{2+}$. Note that all the frequencies shown in Tables 5.5 and 5.6 are raw values from the files with the statistical uncertainties, without any systematic corrections. Therefore the ν_c values of the same species from different values of t_{acc} do not always agree. This is not a problem as the purpose of this exercise was to unambiguously identify the beam constituents and not intended for a precise measurement of the mass of any particular species.

Identifying the ν_c of each species present in the beam is the most obvious and definite test of the identities of the beam constituents. Once the frequencies are determined, a few other tests are done to confirm the findings. This is particularly important for isotopes whose masses have never been measured. Firstly, we compare the observed rate of any isotope against the expected rate based on ^{252}Cf fission. For example, we intended to measure ^{166}Eu , which belongs to a $6 \times 10^{-5}\%$ fission branch and has an extrapolated uncertainty of 100 keV/c² in the AME2020 [30]. In the $A = 166$ mass region, the most abundant isotope

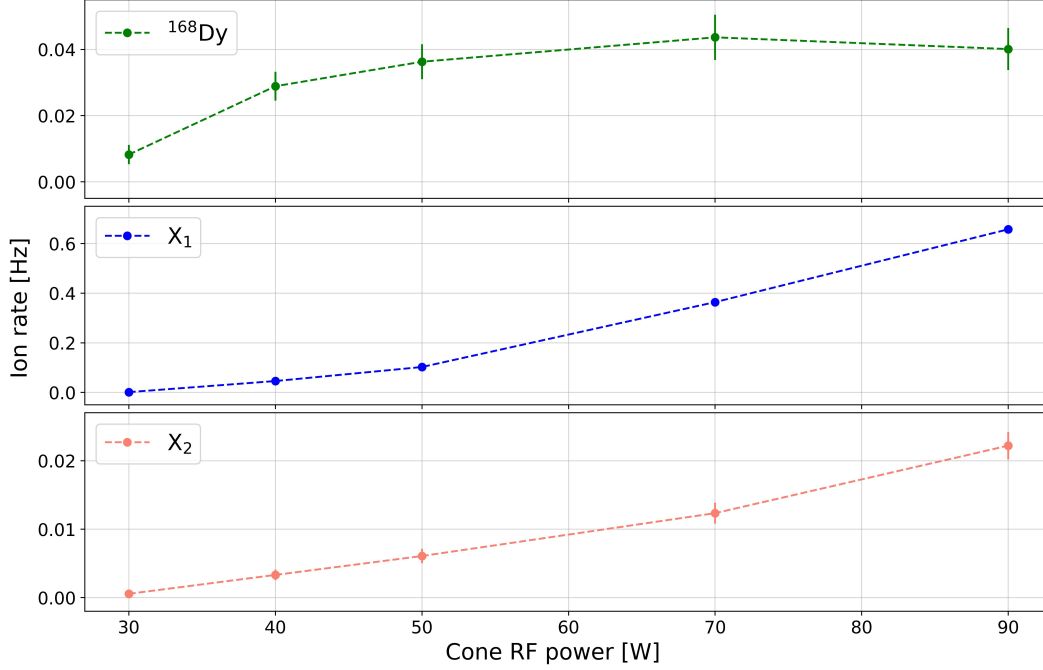


Figure 5.12 : Rate of ^{168}Dy and two molecular contaminants (X_1 and X_2) ions against cone RF power.

is ^{166}Tb belonging to a 0.01% branch. So after tuning the system with $^{150}\text{Ce}^{2+}$, we scaled to $^{166}\text{Tb}^{2+}$. ^{166}Tb was measured in the past at the CPT [53], and we positively identified it. The ion rate of $^{166}\text{Tb}^{2+}$ at the PS-MCP was observed to be around 0.4 Hz. Next we rescaled the system to move to $^{166}\text{Eu}^{2+}$, and observed a species with ν_c within 1 Hz of the expected Eu ν_c from AME2020, which agrees with the extrapolated mass uncertainty. But the ion-rate of this species was around ~ 0.008 Hz, almost four times the expected rate of ~ 0.002 Hz, which was a little suspicious. It required us to further confirm the identity of the species with a second test using the RF power applied to the cone section of the CARIBU gas catcher.

Recall from Figure 3.5(a) that RF fields are used in the gas catcher body and cone to push the ions towards the center. It can be shown that [138]:

$$E_{\text{eff}} \propto V_{\text{RF}}^2 k_0^2 \frac{m}{q} \quad (5.19)$$

where E_{eff} is the repelling field, V_{RF} is the amplitude of the RF voltage applied and k_0 is the ion mobility. In the presence of buffer gas, molecular ions, due to larger size, have

lower mobility compared to atomic ions of the same m/q or equivalently A/q ratio. To compensate for that, a higher V_{RF} is required to extract molecules out of the gas catcher as efficiently as the fission fragments [138, 139]. In other words, after a certain minimum V_{RF} , or equivalently applied RF power, increasing it further will not increase the extracted amount of the fission fragments, while the quantity of molecular contaminants will keep increasing. At the CARIBU gas catcher, because of the spoked ring structure of the body section, this becomes more noticeable with the cone, which is typically operated at a power of 50 W. This test can be used to determine if a certain species is a fission fragment or some molecular contaminant, as shown in Figure 5.12 where the variation in the rates of $^{168}\text{Dy}^{2+}$ and two molecular contaminants (X_1 and X_2) ions are plotted against cone RF power. The $^{168}\text{Dy}^{2+}$ rate gradually plateaued after 50 W on the cone RF, while the rates of X_1 and X_2 kept increasing. When we did this test on the $^{166}\text{Eu}^{2+}$ —candidate increasing the cone power from 30 W to 90 W, its rate kept increasing which confirmed that it was not $^{166}\text{Eu}^{2+}$, but a contaminant with a close mass.

The frequencies are also matched against that of known or possible isobars or molecules from databases like the SCM software (version 3.2.1) [140] made by the LEBIT group (Penning trap mass spectrometry at the NSCL) at Michigan State University and a list of contaminants seen in CARIBU over the years (and similar patterns). For example, a number of water and hydroxide molecules like $^{150}\text{PrH}_2\text{O}^{2+}$ and $^{151}\text{CeOH}^{2+}$ have been observed in the beam of $A/q = 168/2+$, as can be seen in Figure 3.10, and are expected to be present in the mass region $A \sim 168$. So during any measurement done around this mass region, we would always check for the presence of such molecules. This has proven to be very useful in case of a ^{171}Er measurement. A species with ν_c similar to $^{171}\text{Er}^{2+}$ — was observed during the ν_c identification procedure. The ion rate (1.6 Hz) was too high for $^{171}\text{Er}^{2+}$, produced from a $1.6 \times 10^{-4}\%$ branch. We matched this frequency to possible CARIBU contaminants database and found it to be $^{153}\text{NdH}_2\text{O}^{2+}$. If a species is confirmed to be not a fission fragment and no match is found from the databases, we usually designate those as species X (see Figure 3.10).

Finally, for isotopes with half lives $< 1 - 2$ s [53], one can do another test. Increasing the number of shots acquired at the linear trap would mean more bunches are captured at

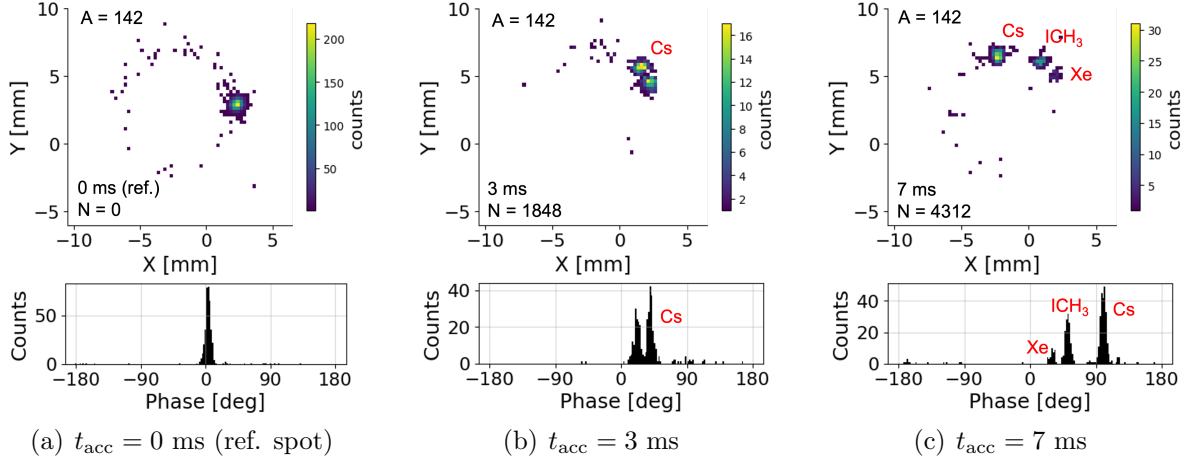


Figure 5.13 : PI-ICR mass resolution with a beam of $m/q = 142/1+$.

the linear trap, and hence the ion rate at the Penning trap should increase. However, it also means that the first few bunches of ions captured would spend a longer time at the linear trap, and decay. So for short lived species, the ion rate will not increase as much. This test is not without faults as increasing the linear trap cycle time would also mean a drop in trapping efficiency, and hence the observed ion rate could also be skewed. This method was not effective for many of the measurements presented in this thesis, and is only included as a possible test to complete the picture of particle identification.

5.4.2 Separating isobars in beam

Typically, a beam from CARIBU consists of multiple isobaric molecular ions along with the fission fragments. The MR-TOF in combination with the BNG selectively removes many of those contaminants. But, as can be seen in Figure 3.10, sometimes it is not able to remove all of the contaminants. The high resolution achieved with the PI-ICR technique is critical for separating these components. From equation 5.12, one can deduce the resolving power as:

$$R = \frac{m}{\Delta m} = \frac{\nu_c}{\Delta \nu_c} = \frac{2\pi \nu_c t_{acc}}{\Delta \phi_c} \quad (5.20)$$

where $\Delta \phi_c$ is the angular spread of the spots. What this means is that, the resolving power increases with t_{acc} . This is demonstrated in Figure 5.13, where the XY -positions of the ion hits is shown on the top, along with the phase projections on the bottom, for a beam of

142/1+ with three different values of t_{acc} .

Figure 5.13(a) is a reference file, that is, with $t_{\text{acc}} = 0$. As can be seen, the spectrum shows only one spot. In Figure 5.13(b), we can see that for $t_{\text{acc}} = 3$ ms, the spot from the reference file is starting to split, with $^{142}\text{Cs}^+$ ions separating from the other beam constituents. In the right panel (figure 5.13(c)), at $t_{\text{acc}} = 7$ ms, the three main beam species, $^{142}\text{Cs}^+$, $^{127}\text{ICH}_3^+$ and $^{142}\text{Xe}^+$, are all separated. The exact identities of the spot were determined using the procedures described in Section 5.4.1.

While increasing t_{acc} will mean better mass resolution, there is a possibility that spots belonging to two or more species may overlap on different turns. So during a measurement campaign, as one identifies the ν_c of all the species present in the beam, care is taken to make sure the spots do not overlap. During the identification procedure, this can be less critical as the frequencies of all overlapping species will be known from the list of ν_c values corresponding to different turns. This can be seen in Table 5.5 and Figure 5.11(c), where overlapping species were still identified. However during precision measurement, this becomes of paramount importance to preserve the accuracy of the measurement and considerable time is spent finding a good t_{acc} where all the species are separated and clearly visible (see Figure 3.10).

5.5 Systematic effects and corrections

A number of systematic effects have been observed in TOF-ICR Penning trap mass spectrometry over the years and these effects can also be present in PI-ICR measurements. In addition, there are some systematic effects that are specific to PI-ICR at the CPT. In this section, I will discuss these effects, and the procedures we follow during measurement campaigns and data analysis to address the effects.

5.5.1 Magnetic field drifts

From Equation 2.25, ν_c has a direct relationship with the magnetic field B and any drift in the field can have a significant effect in the accuracy of the measurement. Spatial variations in B are addressed by making sure that the ions always probe the same B field in all three dimensions. Along the axial direction, this is ensured by trapping the ions at the center of the trap in the z direction before starting the PI-ICR measurements (see Section 4.4.1), while in the radial direction, the ions are always excited to the same radius using the

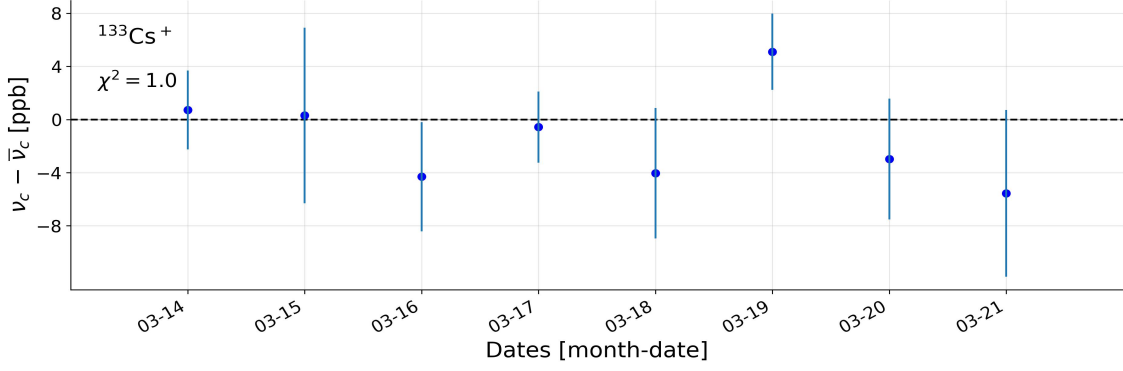


Figure 5.14 : Temporal drift of the magnetic field over 8 days shown as a difference in the measured cyclotron frequencies from a weighted average value in ppb.

dipole ν_+ excitation (described sections 2.3.1 and 5.2.1). Temporal drifts in B are handled by doing a field-calibration measurement as close in time as possible to an isotope-of-interest measurement. Typically, mass measurement campaigns at the CPT last for about a week and several calibrant measurements, with ions from CARIBU and SIS, are interleaved in between the isotope-of-interest measurements, to monitor the temporal drift of the B field. Figure 5.14 shows the ppb variation of ν_c of $^{133}\text{Cs}^+$ over a course of 8 days, as a difference from a weighted average value. This shows a net variation of 1.2 ppb on the weighted average over this period of time, smaller than the uncertainties in the mass excesses determined in this work.

5.5.2 Mass dependent systematic shift

Other systematic errors could originate from the choice of calibrant species, or presence of many ions in the trap at the same time leading to ion-ion interactions. As mentioned in Section 2.5, the ratio between the ν_c values of the calibrant species and the species of interest is kept as close to 1 as possible. This is done by selecting calibrants having the similar A/q as the species of interest. Mass measurements of C_6H_6 (calibrant: ^{85}Rb) ^{84}Kr (calibrant: ^{86}Kr), ^{85}Rb (calibrant: ^{133}Cs), ^{87}Rb (calibrants: ^{85}Rb and ^{133}Cs), and ^{133}Cs (calibrants: ^{85}Rb and ^{87}Rb) were conducted. All of these isotopes are known to ~ 0.1 ppb [30] and the deviations of these measured mass excess values from the values given in the AME2020 [30] are shown in Figure 5.15 as a function of the difference in mass numbers (A) of the isotope-of-interest and the calibrant species. From this, a mass dependent systematic effect of 2.5(9) ppb/u

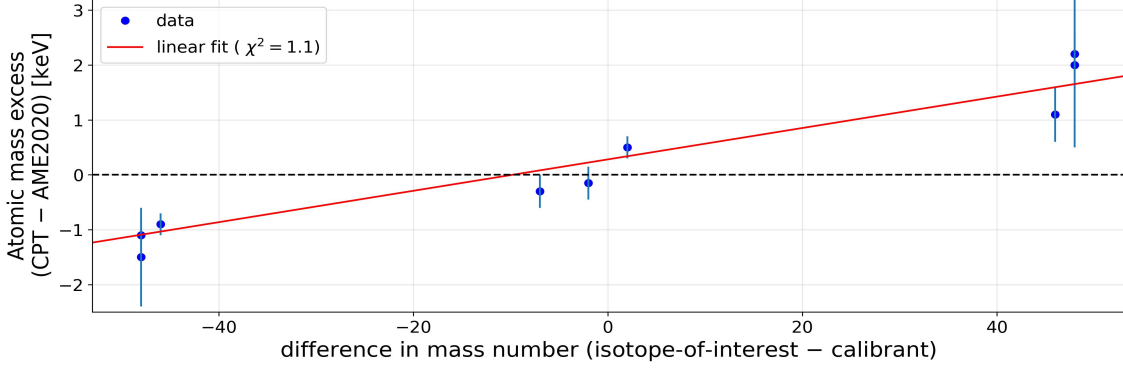


Figure 5.15 : Mass dependent systematic effect at the CPT. See text for details.

was determined, and applied to the results presented in this thesis.

5.5.3 Ion-ion interaction

The ion-ion interaction effect is kept to a minimum by keeping the ion rate low and imposing an *ion-cut* condition in the offline data analysis. Recall that *ion-cut* enables one to analyse data with only a specific number or specific range of ions detected at the same time. The clustering of spots has a significant dependence on the number of ions in that spot. Therefore, five sets of high statistics measurements were done, using a beam of $^{133}\text{Cs}^+$ ions, in 5 hours total, and with varying ion rates between 0.5 Hz to 10 Hz, keeping all other conditions identical. The high statistics and varying rates meant different *ion-cuts* could be imposed. Figure 5.16 shows the result with imposed *ion-cuts* of 1 – 8, as a difference from the ν_c calculated with one detected ion, in ppb, along with a linear fit. The slope of this line is $-1.6(4)$ ppb / detected ion. The ion rates during measurements are carefully maintained at $\sim 1 - 1.5$ Hz. For most fission-fragment measurements presented here, this was not a challenge, as they were detected at the CPT with rates ≤ 1 Hz. For more abundant species like the stable calibrant ions used ($^{84}\text{Kr}^+$, $^{86}\text{Kr}^+$, $^{85}\text{Rb}^+$, $^{133}\text{Cs}^+$, and C_6H_6^+), this would be done by detuning one or two steerers before the linear trap. All the results presented here were analysed with an *ion-cut* of 1 – 4, limiting this effect to a maximum of 6 ppb in the ν_c measurements of each species. Since masses are determined from the ratio between the cyclotron frequencies of the species-of-interest and a calibrant, having the same number of detected ions for both ν_c measurements further minimized this effect to within 1 ppb in mass measurements, smaller than the uncertainties reported here.

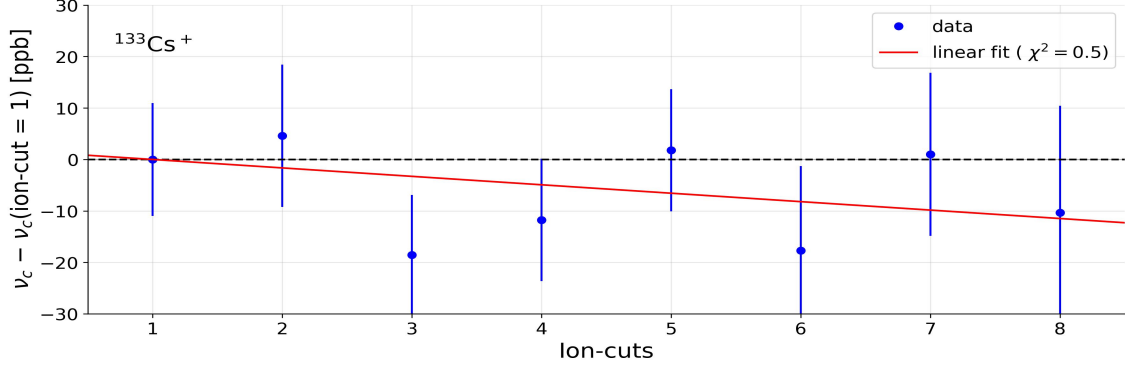


Figure 5.16 : Count class analysis using a beam of $^{133}\text{Cs}^+$ ions, shown as a difference from the ν_c calculated with one detected ion, in ppb. Also shown in red line is a linear fit to the data.

5.5.4 Non-circular projection on PS-MCP

The projected image of the ions in the Penning trap onto the PS-MCP is not circular, but elliptical instead. This projection can be tested by performing a ν_c *phase walk*, as shown in Figure 5.17(a) with a beam of $^{133}\text{Cs}^+$. For $^{133}\text{Cs}^+$, an increment of $6\ \mu\text{s}$ in t_{acc} corresponds to $\sim 18^\circ$. The ν_c *phase walk* is done by increasing t_{acc} from 0 to $114\ \mu\text{s}$ in steps of $6\ \mu\text{s}$, so that the spots are resolved from their adjacent ones. Then all the spots are clustered and the cluster centers are fitted to a circle (Figure 5.17(b)) and an ellipse (Figure 5.17(c)). It is clear from Figure 5.17 that an elliptical fit best represents the data. Since PI-ICR relies on measuring the phase difference between two spots, the location of the spots around this ellipse would have a significant effect on the accuracy of the measurement, leading to shifts up to ~ 0.2 ppm in the measured masses depending on the location of the two spots.

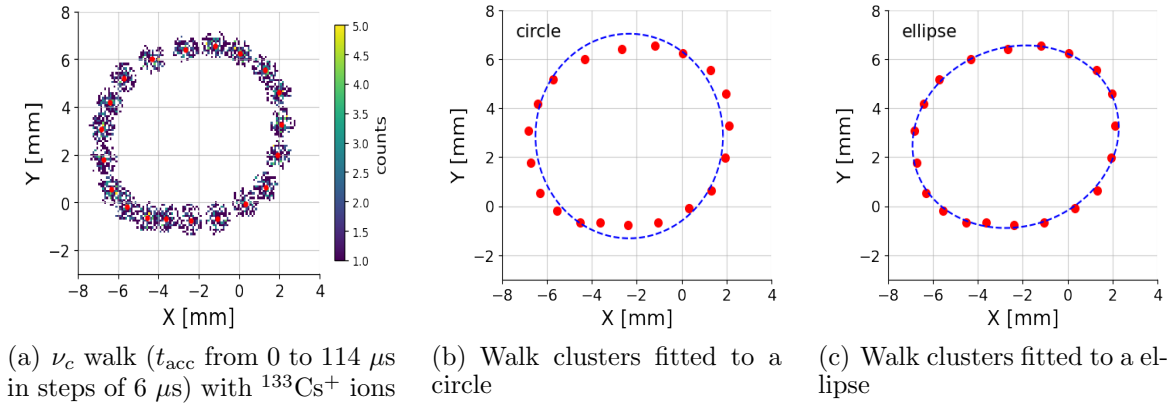


Figure 5.17 : Illustration of the elliptical projection from the trap onto the PS-MCP.

This elliptical mapping is due to a slight misalignment between the magnetic field axis and the TOF drift tube (TOF A to I). The shape can be reproduced in SIMION by tilting the magnetic field by $\sim 1^\circ$ [53]. Recall from Figure 4.4(c), the Penning trap is part of the deceleration and the TOF drift tube assembly. So in the current arrangement, this misalignment issue could only be solved in two ways: re-shimming the magnet such that the field axis coincides with the axis of the TOF tube, or bringing the trap out, and realigning the trap with the field axis. Re-shimming the magnet would need to be accompanied with a recalibration of the whole system as all the frequencies will change. In addition, getting the trap back to operational pressure after being exposed to air also requires a significant time. After the last quench of the magnet, it took around 5 months to prepare the system for measurement campaigns.

The problem, however, can be minimized by limiting the angular difference between the reference and final phases to small values [53]. This is addressed by carefully selecting the accumulation times such that the reference and the final spots are within $\pm 10^\circ$ of each other. This is not a trivial procedure as this requires somewhat precise knowledge of the ν_c of the measured species. So, for unknown species, identifying the ν_c as the first step is critical. The procedure described in Section 5.4.1 is adequate. With such knowledge about ν_c , and an approximate choice of t_{acc} (~ 100 s of ms, as dictated by the required precision of measurement), the accumulation time is gradually adjusted by typically a few μs until the final spot lands within $\pm 10^\circ$ of the reference spot. This brings the effect to within 2 ppb in mass measurements. If there are multiple isobaric species in the beam that one wishes to measure, the procedure is repeated for each one of the isobars. If for some reason this is not possible, an angular systematic correction is applied. For example, consider the campaign for measuring the masses of the ground and isomeric states of ^{119}Ag . Once the isomeric state measurement was completed, a series of electrical discharges near the trap led to the trapping potential being unstable, and forced the measurements to be stopped. While the spots belonging to the isomeric state were within 10° of the reference spot during this measurement, the ground state was 64° away. Once the discharge problem was dealt with, a study was made with $^{133}\text{Cs}^+$ to quantify this and a frequency shift of 0.11 Hz was found. A systematic correction of 15 keV was applied to the mass excess of the ground state.

5.5.5 Electric field drifts

Although ν_c does not depend on the electric field of the trap, ν_+ and ν_- do. ν_- in particular has a strong dependence on the trap depth $V_0 = V_{\text{endcap}} - V_{\text{ring}}$ (see equation 2.48). Therefore, a drift in V_0 would lead to a drift in ν_- , leading to a shift in any phase acquired by the motion in time T :

$$\Delta\phi_- = 2\pi T \Delta\nu_- \quad (5.21)$$

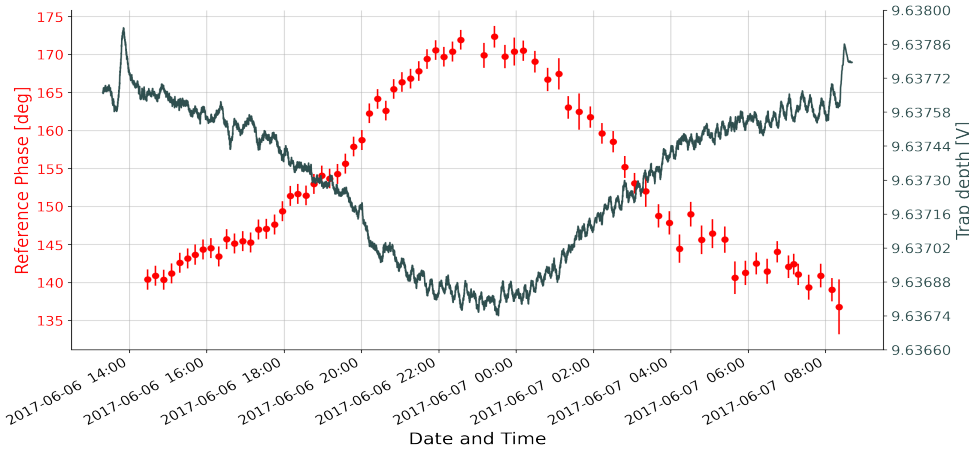


Figure 5.18 : Stability of trap depth and reference phase over a period of time, done with the previous CPT power supply showing a ~ 1 mV drift in the trap depth leading to $\sim 25^\circ$ shift in the reference phase. Figure reproduced from Ref. [53].

Recall from Equation 5.6, the reference phase is essentially a ν_- -phase. Therefore the stability of the trap power supply is of paramount importance. Formerly, the trap voltages were set by adjusting potentiometers which control the input voltage to an op-amp which supplied the required voltage. These input voltages were taken from two precision Kenwood PD56-6AD DC power supplies [53, 57]. Figure 5.18, reproduced from [53], shows the stability of trap depth and the corresponding shift in the reference phase over a period of ~ 20 hours. A drift of ~ 1 mV leading to a phase shift of $\sim 25^\circ$ can be seen. This could lead to a mass shift of $\sim 15 \text{ keV}/c^2$ ($\sim 0.1 \text{ ppm}$).

In 2018, the current Penning trap power supply from Stahl electronics was installed. Similar stability tests have been performed many times, and one such test is shown in Figure 5.19. Over ~ 23 hours, the power supply and the reference phases were more or less

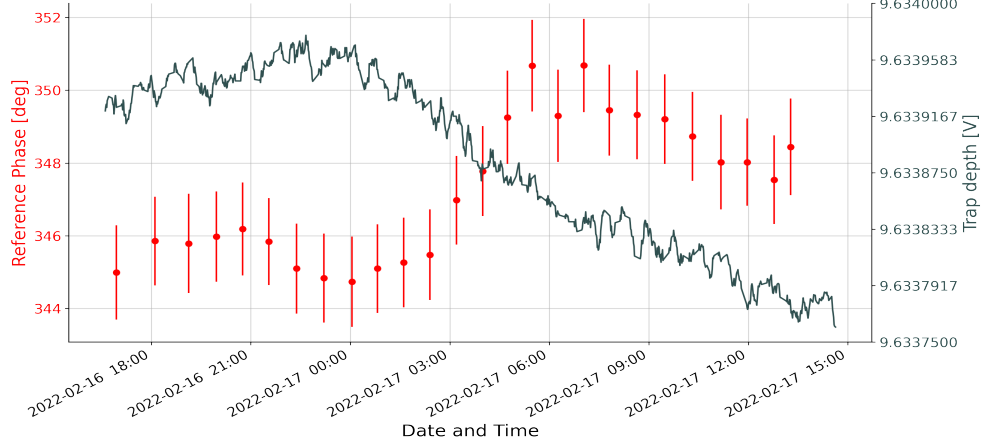


Figure 5.19 : Stability of trap depth and reference phase over a period of time, done with the current CPT power supply showing a much improved stability compared to Figure 5.18. Over ~ 23 hours, the depth has only drifted by $\sim 60 \mu\text{V}$, resulting in a reference phase shift of $\sim 4^\circ$.

stable, except for a period of 5 hours between 4 AM and 9 AM, when a shift of $\sim 60 \mu\text{V}$ was observed, which manifested in a reference phase shift of $\sim 4^\circ$. This corresponds to a mass shift of $\sim 1 \text{ keV}/c^2$ ($\sim 8 \text{ ppb}$). As an additional precaution, reference files are done every few minutes or once per final file, and then the reference closest in time to a final file is the one considered for that final file. As a result, mass variation due to this effect is kept to within 2 ppb.

5.5.6 Sinewave

When ν_c was measured carefully with these precautions, it was observed that measurements with different values of t_{acc} do not agree with each other. Repeated measurements revealed that ν_c shows a sinusoidal dependence on t_{acc} . Moreover, a similar dependence was seen between the radius of the spots and the t_{acc} . In an ideal world, the centering ν_- applied at the beginning of a PI-ICR measurement should center the ions, that is the ions should have a zero ν_- motion after this pulse. At this point when the ν_+ excitation is applied, the ions gradually spiral out and get excited to a larger radius. This is shown in Figure 5.20(a), where the blue lines indicate the ions' path as they get excited from the trap center marked with a red cross. At the end of the ν_+ excitation, the ions keep moving in a ν_+ motion along the orbit to which they are excited, as shown by the red dashed circle. In reality, that is not the case, as shown in Figure 5.20(b). Some ν_- motion still remains at the

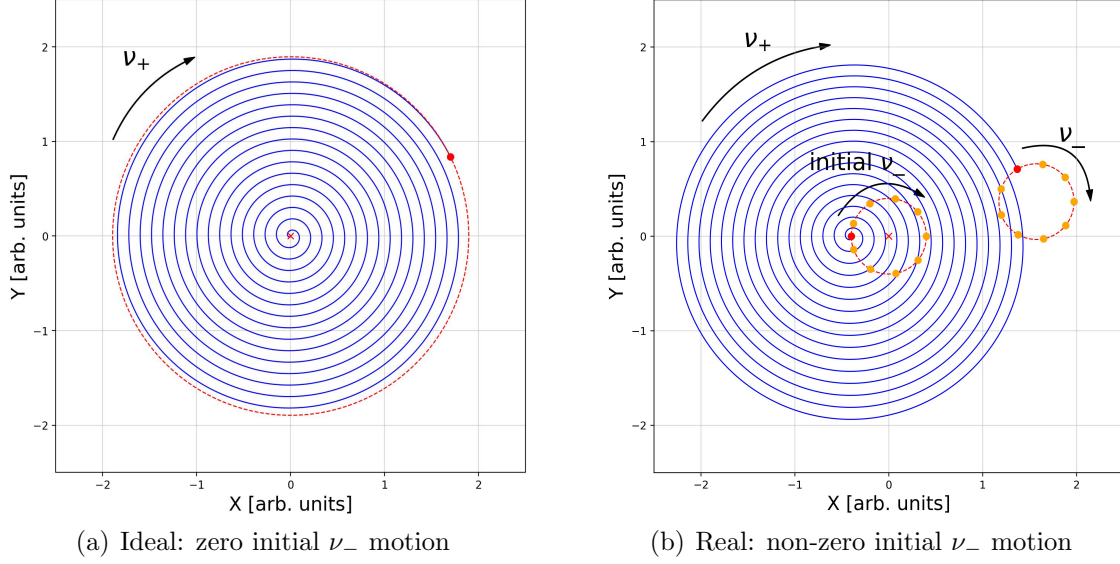


Figure 5.20 : Zero vs non-zero initial ν_- motion.

end of the centering excitation, as shown by the red dashed circle around the trap center, marked by the red cross and annotated “initial ν_- ”. The orange dots on this circle represent the ions’ positions at different times in their ν_- motion. The red dot in this circle marks the ions’ position when the ν_+ excitation is applied. This initial ν_- motion remains even at the end of the ν_+ excitation as shown by the red dashed circle on the outer orbit and annotated “ ν_- ”.

The connection between the sinusoidal dependence and this carried-over initial ν_- motion is shown in Figure 5.21. On the left, the sine fits of measured ν_c and radius against the t_{acc} is shown. On the right, a cartoon illustration of a PI-ICR ν_c measurement with a reference spot and four final spots on this ν_- motion is shown. Recall from Equation 5.12:

$$\nu_c = \frac{\phi_c + 2\pi N_c(t_{acc})}{2\pi t_{acc}},$$

that ν_c measurement relies on the angle (ϕ_c) between the reference and the final spot, that is the difference between the reference and the final phases. The carried-over ν_- motion leads to an oscillation in this phase difference, leading to the ν_c oscillation. The two extreme values of ϕ_c (final spots 1 and 3) leads to the measured ν_c sitting at a minimum and a maximum of the sine wave. ϕ_c would be the same for the two intermediate positions 2 and 4. Hence the corresponding ν_c values would be on the mean of the sine wave. On the other hand,

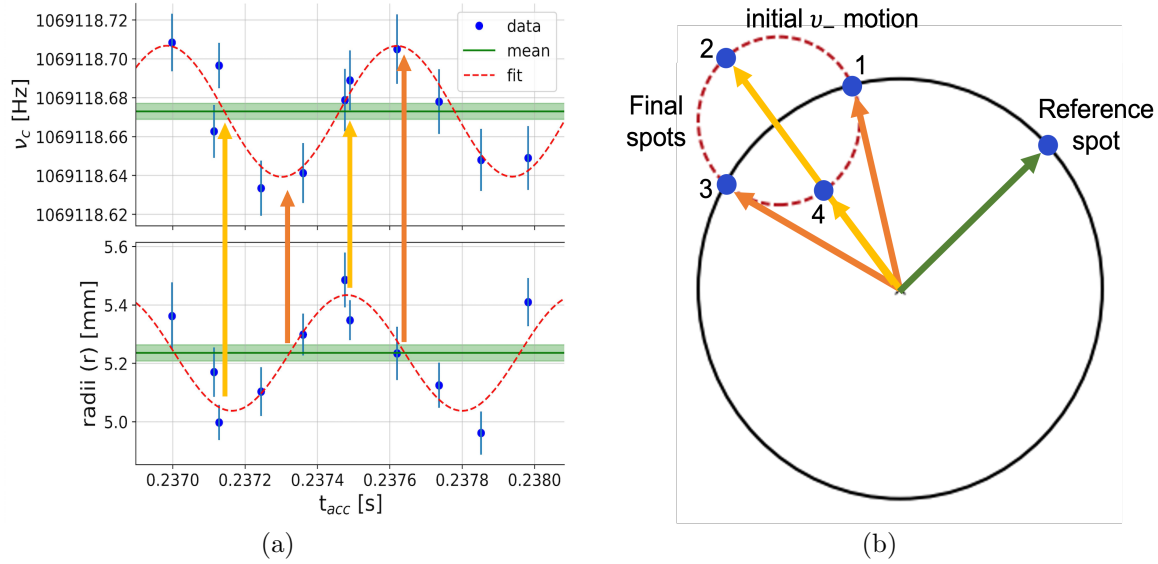


Figure 5.21 : Explanation of sine fits of ν_c and radius

the radius would be the same for final spots 1 and 3, while it would be maximally different between spots 2 and 4. These are shown by the two orange and gold arrows on both the panels. Finally, since this oscillation is due to the ν_- motion, the frequency of both the sine waves is also ν_- . This ν_c oscillation could lead to a mass oscillation of $\sim 10 \text{ keV}/c^2$ peak-to-peak (0.06 ppm). To accurately fit this phenomenon and make precision measurements, we use the following models:

$$\nu_c = \frac{A}{t_{\text{acc}}} \sin(2\pi\nu_- t_{\text{acc}} + \phi^{\text{initial}}) + \nu_c^{\text{mean}} \quad \text{and} \quad (5.22a)$$

$$r = B \cos(2\pi\nu_- t_{\text{acc}} + \phi^{\text{initial}}) + r^{\text{mean}} \quad (5.22b)$$

Here A and B are the amplitudes of the two sine waves and ϕ^{initial} is some initial phase. Evidently, there is a phase difference of $\pi/2$ between the two sine waves. In Figure 5.21(a), this difference is observed as $\sim 81^\circ$. This discrepancy can be attributed to the elliptical mapping of the ions' in-trap position onto the PS-MCP. The $1/t_{\text{acc}}$ component in the amplitude of the ν_c model comes from the $1/t_{\text{acc}}$ in Equation 5.12. The red dashed lines in Figure 5.21(a) demonstrate fitting done using these models. We usually make a number of measurements probing at least one ν_- period, and take the ν_c^{mean} and r^{mean} from the fits as the actual ν_c and radius, shown with the green horizontal line. The green bands signify

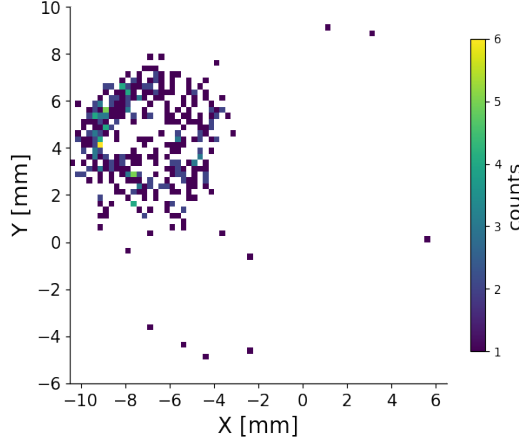


Figure 5.22 : Ring-like spot due to an untuned ν_- pulse

the resulting uncertainties from the fits. This uncertainty in the ν_c fit contributes to the statistical uncertainties in mass measurements.

The centering pulse is applied from the Tabor waveform generator. With an untuned ν_- pulse, the ions will be off-center by a large amount at the start of the ν_+ excitation, and it will be obvious from the spot quality. In the worst case scenario, one would see a ring instead of a spot, as shown in Figure 5.22. This is because after the ν_c conversion pulse, the carried-over initial ν_- motion around some initial non-zero $r_-(0)$ will be converted to a fast ν_+ motion around the same radius. Recall from Figure 2.9(b), only a perfectly tuned ν_- pulse would be able to center the ions. At the CPT, it has not been possible to tune the amplitude, duration, phase and frequency of ν_- pulse precisely enough to center the ions perfectly. Therefore, a well tuned ν_- pulse would only bring the ions closer to the trap center ($r_-(0) \rightarrow 0$), either undershooting the centering or overshooting. Such small $r_-(0)$ does not manifest in the spot quality, but affects the ν_c measurement.

This initial ν_- motion is the reason why we cannot determine the trap center using the catch-center-release technique. A non-zero initial ν_- phase means the exact location of the ions upon ejection, with just the centering pulse applied, depends on the time of ejection, and therefore it is not an accurate measure of the trap center. One way to get around this is applying the centering pulse in a detuned way, similar to the ν_- measurement scheme. As a result, the ions get excited to some orbit while undergoing ν_- motion, before they are ejected. Next the process would be repeated many times while scanning the ejection time

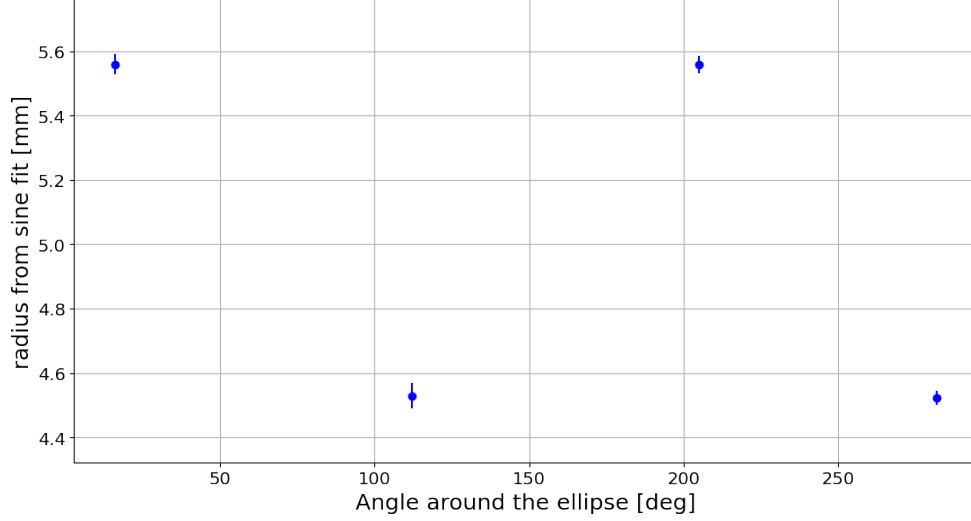


Figure 5.23 : center measurement with C_6H_6^+ ions resulting in a center location of $(-2.21, 2.82)$.

over a ν_- period, just like performing a ν_- walk. The data could be fitted to an ellipse and the center of the ellipse would be the center of the trap. Since the ejection pulse is generated by the low resolution Jorway delay generator, this procedure could result in a high uncertainty and questionable accuracy in the center determination. Instead a different technique using the radius from the radius fit (Equation 5.22b) is followed. Figure 5.23 shows such a measurement using C_6H_6^+ ions. For C_6H_6^+ ions, an accumulation time of $5\ \mu\text{s}$ accounts for a ν_c phase of $\sim 90^\circ$. So four datasets were acquired on four corners of the ellipse using four sets of t_{acc} -ranges, each spanning over $\sim 1\ \text{ms}$ and differing from each other by $5\ \mu\text{s}$. Individual sine fits were performed on each dataset, using an approximate trap center. Then the r^{mean} from fits on the opposite sides of the ellipse were compared. Next the center was changed slightly by hand in the analysis, and it was repeated. The goal was to guess a center for which the opposite radii would be equal. After a few trial and errors, a center location of $(-2.21, 2.82)\ \text{mm}$ was obtained, with an uncertainty of $0.05\ \text{mm}$ in both dimensions. The uncertainty was assigned from the range of guess initial center-coordinates which produced the same radii on opposite sides of the ellipse [53].

The uncertainty in mass determination originating from this non-zero initial ν_- motion can be minimized by collecting more data points. Since the model in Equation 5.22 has three variable parameters, we always aim to complete at least four final t_{acc} in every measurement.

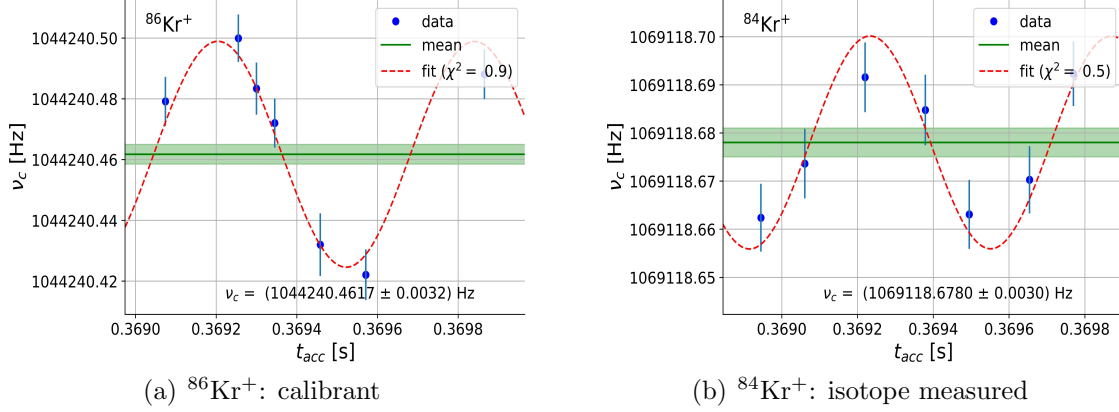


Figure 5.24 : ν_c measurement of $^{84}\text{Kr}^+$ with respect to $^{86}\text{Kr}^+$

However for some rare isotopes, this has proven to be challenging. In those cases, one of the isobaric molecular contaminants is used to do a sine fit with 7-8 observations, and the sine-fit parameters from that measurement are used and fixed to fit the rare isotope. This does not compromise the accuracy of measurement or introduce any additional systematic effects as the injection settings into the Penning trap and the RF excitations are kept exactly the same between the isotope of interest and the isobaric contaminant. As a result, it was possible to keep this uncertainty to within 4 ppb for all the masses presented here.

The judge of any experimental technique or systematic correction is accuracy. Figure 5.24 shows a measurement of $^{84}\text{Kr}^+$ with respect to $^{86}\text{Kr}^+$ done in 2019 with a series of accumulation times between 369 ms and 370 ms. The masses of these isotopes have been measured with the Penning trap at Florida State University [141] and are known to 4 eV [44], and can be used to test the accuracy of our system and study any additional systematic effects. Our measured mass excess of ^{84}Kr , -82439.5 (7) keV, is in good agreement with the literature value of -82439.343 (4) keV, giving confidence in our technique, corrections and model.

5.5.7 Reference phase correction

The ^{86}Kr measurement shows the accuracy of the system for a pure beam. For beams with multiple species, another additional correction needs to be made. Two ^{78}Se measurements were made in April and May 2017. ^{84}Kr was used as a calibrant for both the measurements and the results of the measurements are shown in Figure 5.25. In April, the beam consisted

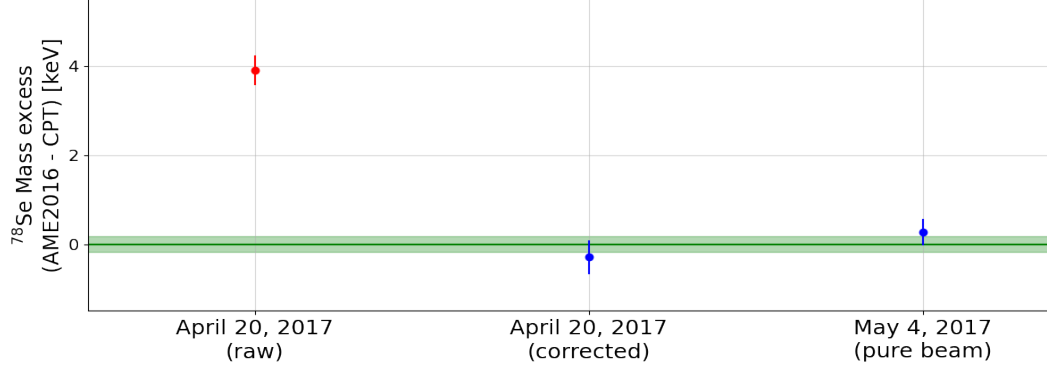
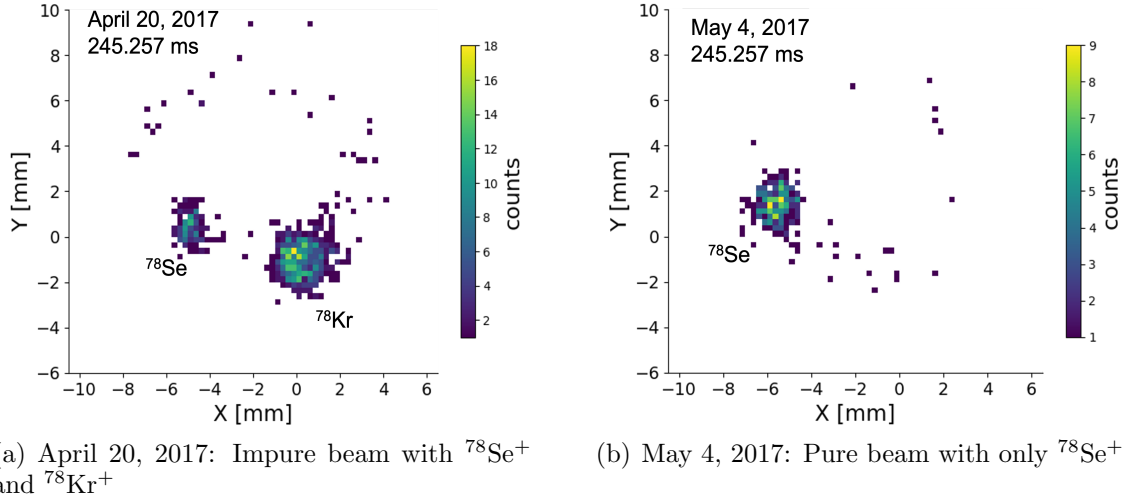


Figure 5.25 : ^{78}Se results for impure (raw and corrected) and pure beam with respect to AME2016 [44].



(a) April 20, 2017: Impure beam with $^{78}\text{Se}^+$ and $^{78}\text{Kr}^+$

(b) May 4, 2017: Pure beam with only $^{78}\text{Se}^+$

Figure 5.26 : Typical final spots of $^{78}\text{Se}^+$ measurements done in April and May, 2017. The April dataset shows a large spot of $^{78}\text{Kr}^+$ and a small spot of $^{78}\text{Se}^+$. The May dataset was acquired with a pure beam of $^{78}\text{Se}^+$.

of nearly $\sim 80\%$ of ^{78}Kr and $\sim 20\%$ of ^{78}Se , as shown by the two spots in Figure 5.26(a). The raw result from this measurement is shown by the red data point in Figure 5.25. As can be seen, the result has a ~ 4 keV difference from the AME2016 value [44]. When the measurement was repeated about a fortnight later, it was with a pure beam and the result agreed with the AME2016 value.

The only difference between the two measurements was the presence of ^{78}Kr in the beam in April. Upon further investigation, it was realized that this mass shift is due to a shifted reference phase. As mentioned in Section 5.2.1, the reference phase is essentially a ν_- phase acquired between the ν_c conversion pulse and the ejection, that is, it is mass independent.

However, as it turns out, it is not entirely true. The finite durations of the ν_+ and ν_c excitations mean the ions gain a mass dependent phase during these excitations. Therefore, the reference phase as seen on the PS-MCP for an impure beam is actually an average of all the reference phases of all the beam species, biased towards the more abundant ones. So in order to get a “true” reference phase for the species of interest, a phase correction needs to be applied to the reference phase, given by:

$$\phi_{\text{corr}}^{\text{ref}} = 2\pi t_{\text{exc}} \sum_k \chi_k (\nu_c^k - \nu_c^I) \quad (5.23)$$

where t_{exc} is the sum of the durations of the ν_+ and ν_c excitations, χ_k is the relative population of the k -th species in the beam, and I is the species of interest. This correction, amounting to $\sim 5.3^\circ$ for the ^{78}Se April measurements, is subtracted from the reference phase. The resulting mass excess value, as can be seen in Figure 5.25, is in good agreement with the AME2016. Due to this effect, one has to identify the ν_c of every species in the beam to < 1 Hz accuracy [53] before starting a sine-wave measurement. For the measurements presented in this thesis, the maximum uncertainty due to this effect has been observed to be ~ 2 ppb.

5.5.8 Summary of systematics

A summary of the systematic effects, with their sources, methods implemented to address them, and their maximum final effect on mass measurements, is shown in Table 5.7. Usually, most of these effects are either already incorporated in the statistical uncertainties of measurements or are small that they produce no significant change in the assigned uncertainties when added in quadrature. In some cases, if some effect could not be addressed during measurement (for example, the effect due to non-circular projection on the PS-MCP during the measurement of ^{119}Ag), it would be added in quadrature to the statistical uncertainties. The mass dependent shift has been used to correct all the determined mass excesses on the basis of the difference in mass numbers between the isotope of interest and the calibrant species, and the corresponding uncertainties have always been added in quadrature to the statistical uncertainties.

Table 5.7 : Summary of systematic effects with their sources, method implemented to address them and the final effect on mass measurements.

Source	Methods to address	Maximum final effect
Temporal instabilities of the B-field	Calibration done several times over the course of week-long measurement campaigns to monitor B-field and the one closest in time to the isotope-of-interest measurement is taken.	< 1.2 ppb
Mass dependent shift	Mass excess corrected and uncertainties inflated.	Mass shift of $\sim 2.5(9)$ ppb/u
Ion-ion interaction	Ion-rate kept low during measurement and <i>ion-cut</i> of 1 – 4 applied during analysis of both the calibrant species and the isotope-of-interest.	< 1 ppb
Non-circular projection on the PS-MCP	Final spots placed within 10° of reference spots.	< 2 ppb
Electric field instabilities	Reference spots measured every 10–15 minutes or once every final file and the reference closest in time to a final file is considered for that final file.	< 2 ppb
Non-zero initial ν_- motion	Enough final point measurements done to probe at least one ν_- period, and ν_c obtained from sine-fitting these measurements against t_{acc} .	< 4 ppb
Reference phase shift for impure beam	All beam species identified before starting measurement, and a phase correction applied on the basis of the populations and cyclotron frequencies of the contaminants.	< 2 ppb

6 Results and discussions

The masses of a total of 34 nuclides are presented in this thesis. These results include the masses of six isomers, and seven nuclides measured for the first time. The measurements were conducted mainly in June, July, and October of 2019 and August, October, and December of 2021 (after the restart of laboratory operations following the COVID-19 enforced shutdown in 2020-2021). The main focus of this work has been to measure neutron-rich nuclides produced in measurable quantities at CARIBU to probe the REP in the r -process abundance pattern. Two of the nuclides (^{155}Ce and ^{168}Gd) are produced from $10^{-5}\%$ branch of ^{252}Cf fission, and five (^{152}La , ^{154}Ce , ^{169}Dy , ^{168}Ho , and ^{169}Ho) are populated from $10^{-4}\%$ branch [93], constituting some of lowest ion-rate measurements done at CPT-CARIBU, while ^{152}La and ^{155}Ce , with $t_{1/2}$ of 287 (16) ms and 313 (7) ms respectively [3], make for two of the most short-lived nuclides measured at the CPT. The measured nuclides are shown in Figure 6.1, along with past CPT measurements using the TOF-ICR and PI-ICR techniques at two different locations in ATLAS (Triangle room and CARIBU). Also shown for reference are the stable nuclides and a portion of a possible r -process path, calculated for a hot environment.

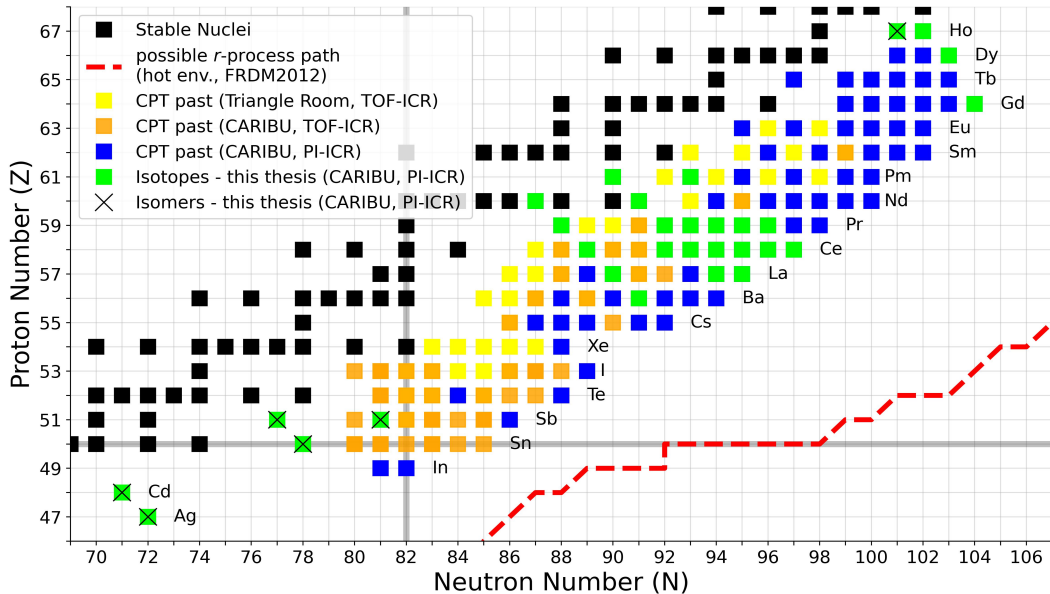


Figure 6.1 : Summary of nuclides measured as part of this thesis. Also shown are the nuclides measured at the CPT using TOF-ICR and PI-ICR, as well as the stable isotopes and a portion of the r -process path calculated for a hot environment using FRDM2012 mass model [31].

In this chapter, I will present the mass excess of all the nuclides, compare the results against the AME2020 [30] and the different mass models, and discuss their astrophysical relevance and implications.

6.1 Mass results

The mass excess of the nuclides is shown in Table 6.1. The measured nuclides (Meas.) and the calibrants (Cal.) are denoted by their corresponding ion species (singly or doubly charged). The ratio of the cyclotron frequencies of the two ion species, $r = \nu_c^{\text{Cal.}}/\nu_c$, along with the mass excess values from the latest Atomic Mass Evaluation, AME2020 [3, 30] and the deviations from the mass excesses reported here, are also shown. Additionally, a summary of the statistical and systematic uncertainties in measurements of the ν_c ratios between the calibrant and the species of interest is provided in Appendix I.

6.2 Discussions and comparisons

6.2.1 Discussions of the results of individual nuclides

$A = 119$: $^{119,119m}\text{Ag}$ and $^{119,119m}\text{Cd}$

The ^{119}Ag was first measured by ISOLTRAP [142], which is adopted by the AME2020 [30]. Our measured mass agrees with the result of Ref. [142]. Due to a series of electrical discharges near the trap resulting in unstable trapping potentials, it was not possible to place the spots of the ground state within 10° of the reference spots, as described in Section 5.5.4. Therefore, a systematic correction needed to be added to the mass excess of the ground state, and it led to the significant inflation in the uncertainty of the measured ground state mass. The AME also predicts an isomeric state with an extrapolated excitation energy of 20(20)# keV. The authors of [142] did not observe this ^{119m}Ag , and its mass was not measured until the CPT measurement in December 2021. The masses of both states agree with the AME results, measured and extrapolated. While the ground state uncertainty is large due to the reason stated above, the isomeric state measurement reduces the extrapolated uncertainty by a factor of 7.5. The excitation energy of 20(16) keV calculated from the CPT measurements agrees completely to the AME value.

The ^{119}Cd and ^{119m}Cd masses reported in the AME2020 [30] are derived from β -endpoint measurements from Ref. [143], with an excitation energy of 146.54 (11) keV. The CPT

Table 6.1 : A summary of the masses measured as part of this thesis. The corresponding calibrant (Cal.) ion species and the ν_c ratio between the measured (Meas.) and the calibrant species are also presented. Finally a comparison with the latest Atomic Mass Evaluation (AME2020[3, 30]) is given in the form of a difference in mass excess between CPT and AME2020, $\Delta(\text{CPT} - \text{AME2020})$. The # depicts a previously unmeasured isotope with the AME2020 publishing an extrapolated mass excess.

Meas.	Cal.	$r = \frac{\nu_c^{[\text{Cal.}]}}{\nu_c}$	Atomic mass excess [keV]		
			CPT	AME2020 [3, 30]	$\Delta(\text{CPT} - \text{AME2020})$
$^{119}\text{Ag}^+$	$^{133}\text{Cs}^+$	0.894 737 70(14)	-78 644(16)	-78 646(15)	2(22)
$^{119m}\text{Ag}^+$	$^{133}\text{Cs}^+$	0.894 737 861(31)	-78 623.5(33)	-78 626(25)#	2.5(25)#
$^{119}\text{Cd}^+$	$^{133}\text{Cs}^+$	0.894 693 875(31)	-84 068.8(41)	-83 980(40)	89(40)
$^{119m}\text{Cd}^+$	$^{133}\text{Cs}^+$	0.894 695 043(31)	-83 924.8(48)	-83 830(40)	95(40)
$^{128}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.962 407 088(20)	-84 609.2(21)	-84 630(19)	21(19)
$^{128m}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.962 407 438(21)	-84 565.8(23)	-84 620(18)	54(18)
$^{132}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.992 543 951(12)	-79 636.4(10)	-79635.3(25)	1.1 (27)
$^{132m}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.992 545 123(12)	-79 490.9(11)	-79 490(50)#	1(50)#
$^{147}\text{Ba}^{2+}$	C_6H_6^+	0.941 325 707 4(97)	-60 300.7(11)	-60 264(20)	37(20)
$^{147}\text{La}^{2+}$	C_6H_6^+	0.941 281 923 3(97)	-66 666.9(11)	-66 678(11)	11(11)
$^{151}\text{La}^{2+}$	C_6H_6^+	0.966 998 063 8(91)	-53 528.3(11)	-53 310(440)	218(440)
$^{152}\text{La}^{2+}$	C_6H_6^+	0.973 434 322(41)	-49 193.4(58)	-49 290(300)#	97(300)#
$^{147}\text{Ce}^{2+}$	C_6H_6^+	0.941 245 013 9(97)	-72 033.5(11)	-72 014(9)	19.5 (91)
$^{150}\text{Ce}^{2+}$	C_6H_6^+	0.960 513 659(21)	-64 863.7(28)	-64 847(12)	17(12)
$^{151}\text{Ce}^{2+}$	C_6H_6^+	0.966 945 175 4(77)	-61 218.18(83)	-61225(18)	7(18)

$^{152}\text{Ce}^{2+}$	C_6H_6^+	0.973 367 710(16)	-58 878.3(23)	-58980(200)#	102(200)#
$^{153}\text{Ce}^{2+}$	C_6H_6^+	0.979 802 813(17)	-54 711.5(24)	-54 910(200)#	198(200)#
$^{154}\text{Ce}^{2+}$	C_6H_6^+	0.986 227 433(18)	-52 068.9(24)	-52 220(200)#	151(200)#
$^{155}\text{Ce}^{2+}$	C_6H_6^+	0.992 664 512(52)	-47 614.7(60)	-47 780(300)#	165 (300)#
$^{147}\text{Pr}^{2+}$	C_6H_6^+	0.941 221 330 2(99)	-75 477.1(11)	-75 444(16)	33(16)
$^{151}\text{Pr}^{2+}$	C_6H_6^+	0.966 906 942 3(77)	-66 777.28(83)	-66 780(12)	3(12)
$^{152}\text{Pr}^{2+}$	C_6H_6^+	0.973 333 983(69)	-63 782.2(98)	-63 758(19)	24(21)
$^{153}\text{Pr}^{2+}$	C_6H_6^+	0.979 755 797(17)	-61 547.5(24)	-61 568(12)	20(12)
$^{154}\text{Pr}^{2+}$	C_6H_6^+	0.986 186 636(18)	-58 000.7(25)	-57 860(100)	140(100)
$^{155}\text{Pr}^{2+}$	C_6H_6^+	0.992 610 782(35)	-55 427.0(21)	-55 415(17)	12(17)
$^{147}\text{Nd}^{2+}$	C_6H_6^+	0.941 202 971 6(99)	-78 146.4(11)	-78 146.8(13)	0.4(17)
$^{151}\text{Nd}^{2+}$	C_6H_6^+	0.966 878 267 3(78)	-70 946.68(83)	-70 943.2(11)	3.5 (14)
$^{151}\text{Pm}^{2+}$	C_6H_6^+	0.966 861 402 3(82)	-73 398.78(91)	-73 386(5)	12.8 (51)
$^{154}\text{Pm}^{2+}$	$^{154}\text{Nd}^{2+}$	0.999 980 959(71)	-68 310(10)	-68 267(25)	43(27)
$^{168}\text{Gd}^{2+}$	$^{84}\text{Kr}^+$	1.000 746 569(36)	-48 171.2(56)	-48 150(300)#	21(300)#
$^{169}\text{Dy}^{2+}$	$^{84}\text{Kr}^+$	1.006 658 279(10)	-55 516.5(14)	-55 600(300)	84(300)
$^{168}\text{Ho}^{2+}$	$^{84}\text{Kr}^+$	1.000 670 824(20)	-60 012.0(32)	-60 060(30)	48(30)
$^{168m}\text{Ho}^{2+}$	$^{84}\text{Kr}^+$	1.000 671 198(20)	-59 953.5(30)	-60 000(30)	46(30)
$^{169}\text{Ho}^{2+}$	$^{85}\text{Rb}^+$	0.994 778 712(14)	-58 789.1(21)	-58 796(20)	7(20)

measurements, ground and isomer, are 2.2σ and 2.4σ heavier than the respective masses reported in the AME2020, but agree on the excitation energy, calculated as 144.0(47) keV. In addition, the uncertainties in the masses for both the states have been reduced by a factor of 12.

$A = 128 : {}^{128,128m}\text{Sb}$

No direct measurement existed for either of the ${}^{128}\text{Sb}$ states prior to the measurements reported in this thesis using the CPT. The AME ground state mass was taken from a β -endpoint measurement [144], while the reported isomer mass was the average of three β -endpoint measurements [145–147]. The masses of the ground and the isomer reported here are 1.1σ and 3σ heavier from the AME2020 results, and almost 13 and 11 times more precise. In addition, the AME2020 [3] reports an excitation energy of 10(6) keV between the two states, while that calculated with the CPT is 43.4(31) keV.

$A = 132 : {}^{132,132m}\text{Sb}$

The mass of ${}^{132}\text{Sb}$ was measured by the JYFLTRAP [148] and the CPT [68] using TOF-ICR. The AME2020 [30] mass is a weighted combination of the two with 83% taken from Ref. [148] and 17% from Ref. [68]. The CPT PI-ICR result reported here agrees with both the JYFLTRAP and the CPT TOF-ICR results, while reducing the uncertainties by factors of 2.7 and 6.1 respectively. The AME2020 reports an extrapolated mass excess of -79490(50)# keV for ${}^{132m}\text{Sb}$. The mass excess of this nuclide was measured using TOF-ICR and reported in Ref. [70] as -79495(17) keV, and the current CPT (PI-ICR) result is in agreement with it, while establishing a 15 times reduction in the uncertainty.

$A = 147 : {}^{147}\text{Ba}, {}^{147}\text{La}, {}^{147}\text{Ce}, {}^{147}\text{Pr}$ and ${}^{147}\text{Nd}$

The AME2020 [30] mass excess values for ${}^{147}\text{Ba}$, and ${}^{147}\text{Ce}$ are mainly based on CPT TOF-ICR measurements from Ref. [64]. The CPT PI-ICR measurements are 1.8σ , and 2.1σ lighter, respectively and lower the uncertainties by factors of 18 and 8. The ${}^{147}\text{La}$ AME2020 mass excess value, also adapted from Ref. [64] is in agreement with results here, while ensuring a 10 times reduction in its uncertainty. The AME2020 [30] ${}^{147}\text{Pr}$ mass excess is an average of multiple β -endpoint measurements [149–151] and the CPT measurements

reported here are 2.1σ lighter, and 14 times more precise. The ^{147}Nd AME2020 [30] value is a combination of two (n, γ) [152, 153] and a (d, p) [154] measurements and agrees with the CPT results presented here. This also supports the reliability of the (n, γ) and (d, p) techniques.

$A = 150 : ^{150}\text{Ce}$

The ^{150}Ce in the AME2020 [30] is also taken mainly from Ref. [64] and agrees within 1.4σ of the result reported here. The current result also lowers the uncertainty by a factor of 4.3.

$A = 151 : ^{151}\text{La}, ^{151}\text{Ce}, ^{151}\text{Pr}, ^{151}\text{Nd}, \text{ and } ^{151}\text{Pm}$

The ^{151}La was measured in the FRS-ESR, GSI [155, 156] and the AME2020 [30] adopted their result. The CPT results here agrees with the AME2020, and reduces the uncertainty by a factor of 412. The ^{151}Ce and ^{151}Pr values in the AME2020 [30] were taken mainly from the CPT TOF-ICR results in Ref. [64]. The latest PI-ICR results agree with both, while lowering the uncertainties by factors of 22 and 14. The AME2020 [30] ^{151}Nd is an average of (n, γ) measurements [153, 157] and agrees within 1.5σ of the CPT result here. The ^{151}Pm in the AME2020 [30] is a combination of a $(^3\text{He}, d)$ [158] and a β -endpoint [159] measurement and is 2.5σ heavier from the result reported here. The precision has again been improved, this time, by a factor of 6.

$A = 152 : ^{152}\text{La}, ^{152}\text{Ce} \text{ and } ^{152}\text{Pr}$

This is the first measurement of the masses of ^{152}La and ^{152}Ce . The AME2020 [30] reports extrapolated values for both. They both agree with the results in this work, while their uncertainties have been reduced by factors of 52 and 87 respectively. The ^{152}Pr comes from Ref. [64] and agrees within 1.1σ of this work's result.

$A = 153 : ^{153}\text{Ce} \text{ and } ^{153}\text{Pr}$

This is again the first time the mass of ^{153}Ce has been measured. The AME2020 [30] has an extrapolated mass excess for ^{153}Ce , which agrees with the result here, while the uncertainty has been reduced from 200 keV to 2.4 keV. The ^{153}Pr is a combination of two

past CPT publications (Ref. [64] and [67]). The reported result in Ref. [67] (-61576(14) keV) is actually a weighted average of their raw result (-61540(36) keV) and the result from Ref. [64] (-61582(14) keV). The CPT result reported here agrees with the raw result of Ref. [67], and is 2.4σ heavier from the reported result in Ref. [64].

$A = 154 : {}^{154}\text{Ce}, {}^{154}\text{Pr}$ and ${}^{154}\text{Pm}$

The mass excess of ${}^{154}\text{Ce}$ was measured in the FRS-ESR, GSI [156] as -52540 (577) keV. This was not included in the AME2020 because the expected mass from the local trends in the mass surfaces was 320 keV less bound. Therefore, the AME2020 reports an extrapolated mass excess of -52220 (200) keV [30]. The current CPT value of -52068.9 (24) keV is in agreement with this extrapolation, as well as the FRS-ESR measurement while reporting 83 and 240 times respective reduction in its uncertainty.

The AME2020 ${}^{154}\text{Pr}$ mass was adopted from a β -endpoint measurement [160] with respect to ${}^{154}\text{Nd}$. This value shifted by 240 keV from AME2016 [44] to AME2020 [30], because of the mass of ${}^{154}\text{Nd}$ being measured at the CPT [72] and the JYFLTRAP [161]. The new mass excess of ${}^{154}\text{Pr}$ agrees within 1.4σ of the CPT measurement reported here, while its precision has been improved by a factor of 40.

The ${}^{154}\text{Pm}$ mass in the AME2020 [30] comes mainly from a β -endpoint measurement with respect to ${}^{154}\text{Nd}$ [162] and agrees within 1.6σ of the CPT PI-ICR measurement here. The AME2020 [3] also mentions an isomeric state for the ${}^{154}\text{Pm}$, with an excitation energy of -230(50) keV. Now, the AME2016 [104] mentions two internal transition measurements between the two states with the energies reported as -210(70) keV [163] and 20(12) keV [164], and adopts the later value. Between the two AMEs (2016 and 2020), the ${}^{154}\text{Pm}$ ground state mass has shifted by 243(55) keV due to the CPT [72] and JYFLTRAP [161] measurements of ${}^{154}\text{Nd}$, leading to the 20(12) keV (from 2016) to -230(50) keV (in 2020) excitation energy shift. However, the authors of AME2020 state that due to limited experimental information to conclusively assign the two states, there is a possibility of the states being misidentified. Although this would not affect the masses, it could explain the negative excitation energy. During the CPT measurement, no isomeric state was observed at the PS-MCP, after going to a t_{acc} of 1 s which, conservatively speaking, should have been able to resolve any isomer within

~ 10 keV of the ground state. This measurement was done after the restart of the facility post COVID-19-enforced shutdown, and the overall yield and extraction from CARIBU was ~ 70 % below the expected levels. Although the ground state, which belongs to a 0.7 % branch of 252 fission [93], was observed at ~ 0.3 Hz, that is around 20 ions in an hour, the isomeric state, belonging to a 0.2% branch [93], was not detected.

$A = 155 : ^{155}\text{Ce}$ and ^{155}Pr

The AME2020 [30] reports an extrapolated mass for ^{155}Ce , which agrees with the CPT result here, while the uncertainty has been reduced by a factor of 50. The ^{155}Pr AME2020 [30] mass was taken from a CPT TOF-ICR measurement [67], which is in agreement with the current PI-ICR result. The current measurements have also resulted in an 8 times improvement in its precision.

$A = 168 : ^{168}\text{Gd}$ and $^{168,168m}\text{Ho}$

This is the first measurement of the ^{168}Gd and it agrees with the extrapolated mass excess reported in AME2020 [30], and its uncertainty has been reduced from 300 keV to 5.6 keV. The ^{168}Ho in AME2020 [30] is adapted from a β -endpoint measurement [165], while its isomer mass was calculated from the excitation energy of 59(1) keV obtained from decay studies [166]. The $^{168,168m}\text{Ho}$ CPT masses here are 1.6σ and 1.5σ heavier than the AME2020 masses, respectively, but the excitation energy agrees with the excitation energy of 58.5(44) calculated with the CPT masses [3]. The CPT measurements have reduced the uncertainties in the masses of both states by a factor of 10.

$A = 169 : ^{169}\text{Dy}$ and ^{169}Ho

The ^{169}Dy and ^{169}Ho masses in the AME2020 [30] are taken from β -endpoint measurements from Ref. [167] and [168], respectively, and both are in agreement with the CPT result reported here. The precision have been improved by factors of 214 and 10 respectively for the Dy and Ho.

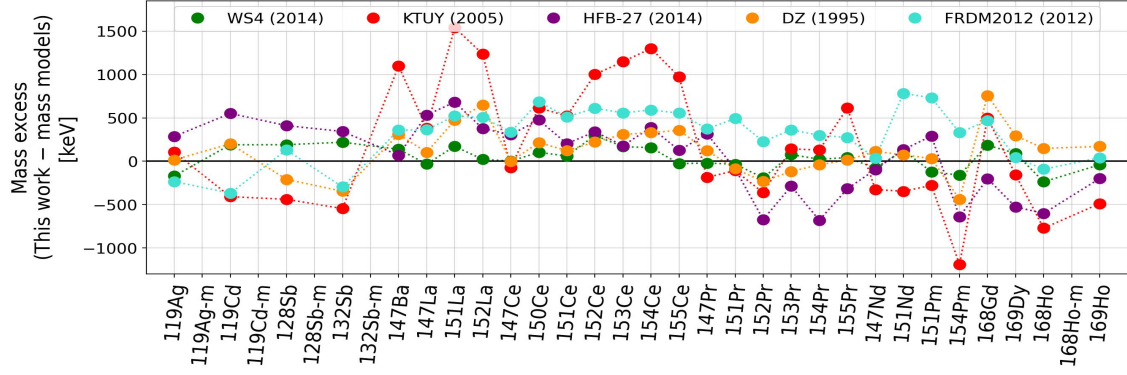


Figure 6.2 : Comparison between mass excess from this thesis and five mass models, namely WS4 [36] (green circles), KTUY [33] (red circles), HFB-27 [35] (purple circles), DZ [169] (orange circles), and FRDM2012 [31] (aqua circles).

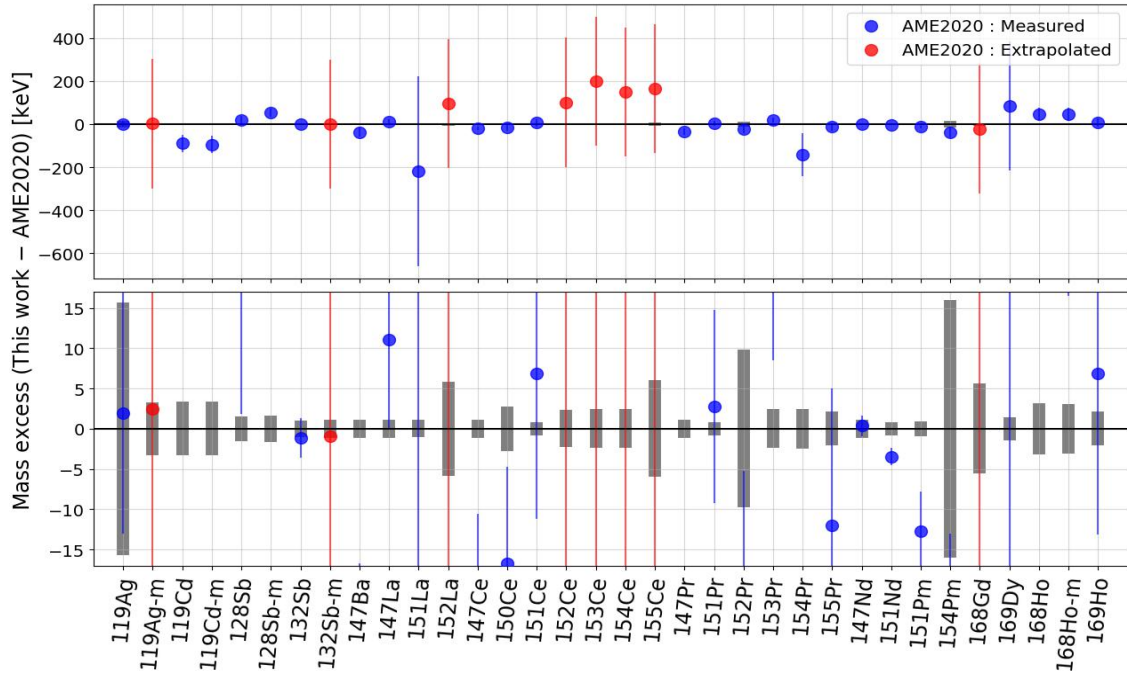


Figure 6.3 : Comparison between mass excess from this thesis and the AME2020 [30]. The lower panel shows the comparisons, zoomed in on the results from this work to highlight the precisions presented here.

6.2.2 Comparison of results with mass models and past measurements

A comparison between the measurements presented here, and predictions from five mass models, namely WS4 [36] (green circles), KTUY [33] (red circles), HFB-27 [35] (purple circles), DZ [169] (orange circles), and FRDM2012 [31] (aqua circles) is presented in Figure 6.2. The dotted lines are added to guide the eye. Additionally, the measured mass excesses are compared to the measured and extrapolated values reported in the AME2020 [30] and shown in Figure 6.3. The grey bands around the 0 keV line in Fig. 6.3 represent the uncertainties reported in this work, to highlight how the precision in our knowledge of masses has been improved by this thesis.

Of the masses presented here, 12 have been previously measured with Penning traps, namely ISOLTRAP, JYFLTRAP, and the CPT, using the TOF-ICR technique. These are shown in Figure 6.4. All the nuclides from Figures 6.2 and 6.3 are kept here to keep the two figures similar. As with Figure 6.3, the grey bands around the 0 keV represent the uncertainties in the results here. The masses of this thesis agree with the ^{119}Ag ISOLTRAP [142] and ^{132}Sb JYFLTRAP measurements [148]. Fairly reasonable agreement can be found with the old CPT measurements of Ref. [67, 68, 70]. Some disagreements arise from a few of the masses reported in Ref. [64] (see Section 6.2.1), the biggest of which is 2.4σ in the mass of ^{153}Pr . This can be attributed to possible frequency shifts in the TOF-ICR data due to the presence of contaminant species among the trapped ions during the measurements [53]. Additionally, the uncertainties quoted for these measurements may have been underestimated [170]. Furthermore, the mass uncertainties for all the isotopes in this thesis have been reduced considerably from the other Penning trap measurements, except for the ^{119}Ag , the reason for which has been explained in the individual nuclide discussion in Section 6.2.1.

The differences are further quantified in Table 6.2. The mean of the absolute mass excess differences ($|\overline{\Delta ME}|$) and the root mean square (RMS) mass excess differences (σ), between the results of this thesis, and the AME2020 [3, 30] and different mass models namely WS4 [36], KTUY [33], HFB-27 [35], DZ [169], and FRDM2012 [31] are shown. Similar differences between all the measured masses in the AME2020 and the same mass models

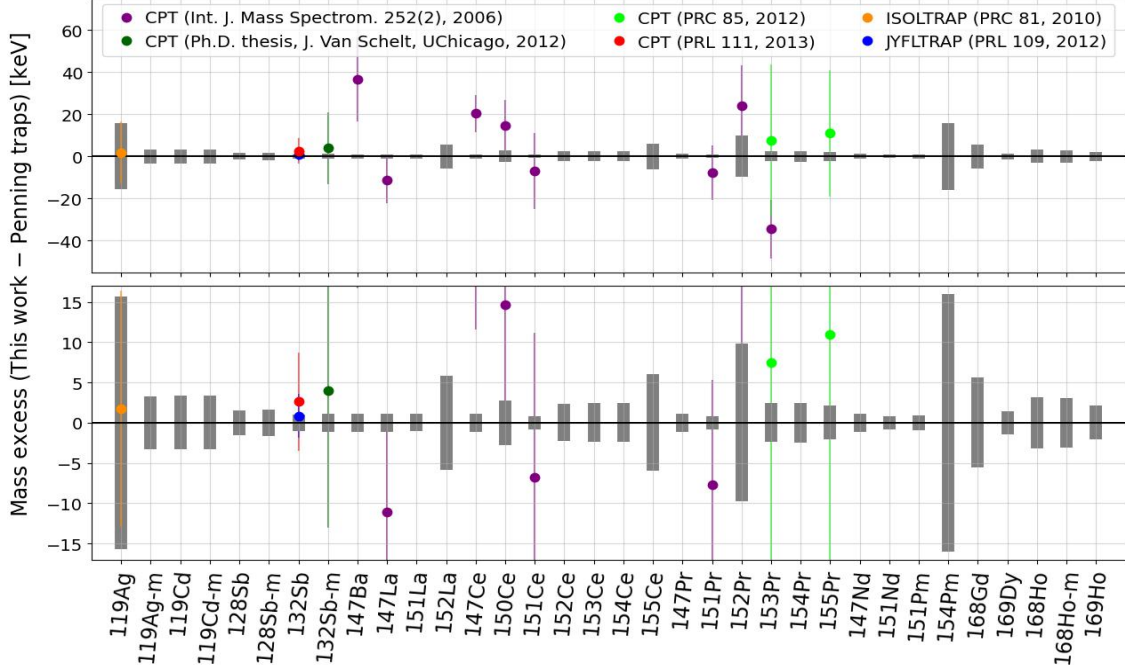


Figure 6.4 : Comparison between mass excesses reported in this thesis and past measurements done with Penning traps: CPT [64, 67, 68, 70] , ISOLTRAP [142], and JYFLTRAP [148] using the TOF-ICR technique.

are also presented. Finally, the comparisons between this thesis' results and past results from Penning traps and storage rings are also shown. As expected, the mass models do not compare well against the current measurements or the previously measured data from the AME2020. Best agreement is found in the case of the WS4 while the KTUY has the worst agreement with both, the current measurements (Figure 6.2) as well as measured masses in the AME2020. More precisely measured masses would mean better predictions from the models. This further reiterates the need for precision measurements of more neutron-rich nuclides. As can be seen from Figure 6.4 and Table 6.2, the results here agree well with other Penning trap measurements.

6.2.3 Two neutron separation energy trends

The two-neutron separation energy is defined as:

$$S_{2n}(Z, N) = M(Z, N - 2) - M(Z, N) + 2m_n \quad (6.1)$$

where $M(Z, N)$ is the mass of a nuclide with Z protons and N neutrons, and m_n is the mass

Table 6.2 : Mass comparison between the masses from this thesis and the AME2020 and different mass models. Also shown are the comparison between the mass models and measured masses in the AME2020. Finally a comparison is shown between this thesis’ measurements and past data from Penning traps and storage rings. The comparison is presented in the form of the mean of the absolute mass excess differences ($|\Delta ME|$) and the RMS mass excess differences (σ).

Models / Devices	Comparison with this thesis		Comparison with AME2020	
	$ \Delta ME $ [keV]	σ [keV]	$ \Delta ME $ [keV]	σ [keV]
AME2020 [3, 30]	55	79	-	-
WS4 (2014) [36]	98	127	136	185
KTUY (2005) [33]	482	647	575	742
HFB-27 (2014) [35]	308	382	366	508
DZ (1995) [34]	194	271	293	420
FRDM2012 (2012) [31]	314	391	407	595
Penning traps	13	17		
Storage rings	143	162		

of a neutron. The two-neutron separation energies of all the elements whose isotopes’ masses are presented here are calculated using the AME2020 [30] values and the masses reported here. These are shown in Figure 6.5.

The S_{2n} curves can provide information about evolution of nuclear structure along an isotopic chain. A steep dip in the S_{2n} curve would mean a shell closure, as can be seen around $N \sim 82$ corresponding to the N_{magic} number. On the other hand, a slight positive kink in the curves could indicate the presence of some nuclear deformation [53, 171]. Such kinks can be found over the region $N \sim 89 - 94$ in elements Ba – Dy with the AME2020 [30] masses. Calculations made with masses of isotopes of Ba – Pm from this work also agree fairly well with this trend.

6.3 Astrophysical implications

6.3.1 Reverse engineered Markov chain Monte Carlo studies

Typically r -process simulations use different astrophysical conditions in combination with nuclear physics properties, from experiments or from models, to attempt the reproduction of the variety of features in the solar r -process abundance pattern. A relatively new avenue is to

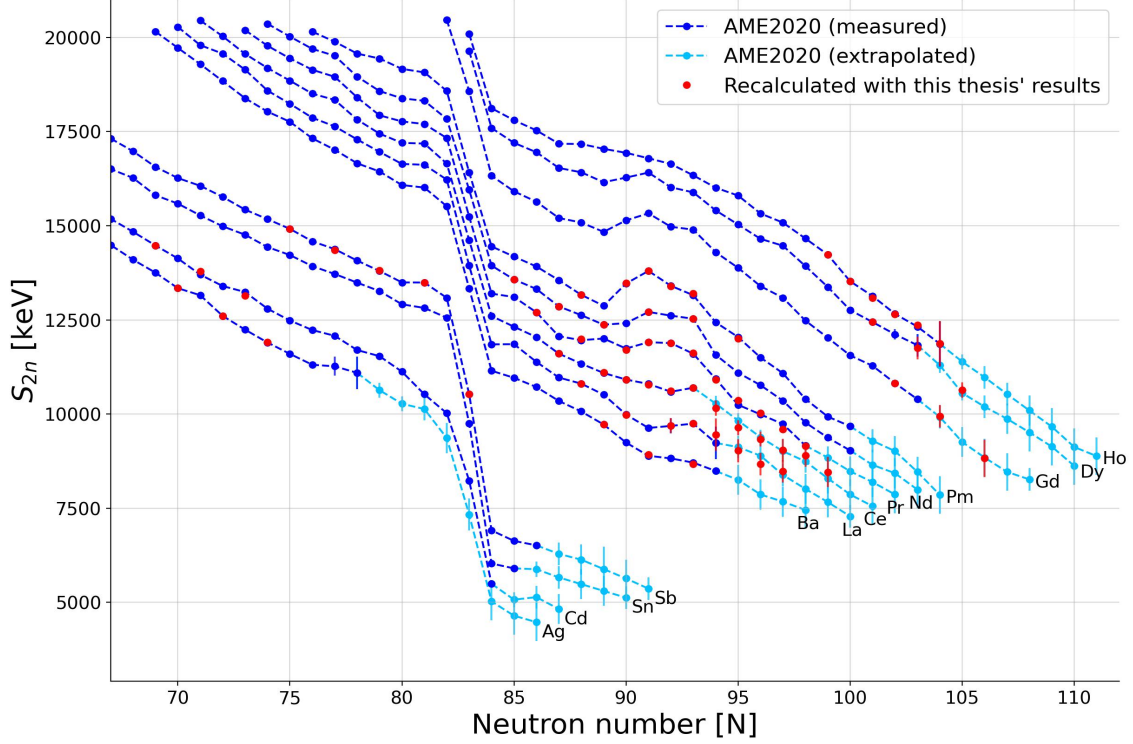


Figure 6.5 : Two neutron separation energy curves calculated with mass data from the AME2020 [30] and masses presented in this thesis.

approach the problem from an inverse perspective [172–174] by using a particular astrophysical condition and applying the Markov Chain Monte Carlo (MCMC) method to determine the masses, that would reproduce the solar r process abundance pattern for that particular condition. This technique was pioneered by the authors of [172–174], who conducted such studies with around 300 neutron-rich lanthanides, with $Z = 58 - 63$, $N = 93 - 111$, to reproduce the REP. Like traditional simulations, this method also starts with inputs of astrophysical conditions and nuclear properties, like masses from AME2012 [175] and half-lives from NUBASE2016 [104]. In the absence of available data, the authors chose to use the DZ mass model [34] as a starting point. Nuclear properties, like separation energies, Q values, β -decay, and (n, γ) rates [72], are calculated using these masses. The astrophysical conditions are characterized by the parameters: entropy (s) for the density of the expanding wind at a particular temperature, a dynamic expansion time-scale (τ), and electron fraction (Y_e) to describe the free neutron concentration (Y_n) as $Y_n = 1 - Y_p$ and $Y_p = Y_e$, Y_p being the proton fraction. Using these, an abundance pattern is derived and compared to the solar abundance

pattern. However, unlike traditional simulations, this “reverse engineering” framework does not stop here. Instead, the DZ masses are then adjusted using a mass parametrization:

$$M(Z, N) = M_{DZ}(Z, N) + a_N e^{-(Z-C)^2/2f}, \quad (6.2)$$

where $M_{DZ}(Z, N)$ are the masses from the DZ mass model, a_N is a set of 28 coefficients, one per neutron number considered in the calculations, f constrains the simulation to a certain mass region, and C sets the central proton number of that mass region. All the nuclear properties are recalculated for the changed masses and the simulation is repeated (with the astrophysical conditions unchanged), leading to an abundance pattern, which is then compared to the solar r -process abundance pattern. This is continued until a convergence with the solar abundance is achieved, thus concluding one simulation chain. Next, a new set of a_N is determined through MCMC and the whole procedure is repeated. This results in a band of predicted masses which would reproduce the solar REP for that astrophysical scenario. The predictions are then compared to experimentally measured masses, to determine if that scenario is a potential r -process site.

The method was first illustrated in Ref. [72]. The average and standard deviations of the reverse engineered mass predictions from 50 parallel and independent chains for a *hot* outflow scenario with $s = 30 \text{ } k_B/\text{baryon}$, $\tau = 70 \text{ ms}$, and $Y_e = 0.2$, were evaluated. Such conditions can be found in the accretion disc of merging neutron stars. The simulations were kept localized with $f = 10$, and solutions with the lowest χ^2 were obtained with a center C around Nd ($Z = 60$) [174]. The mass predictions agreed with masses of Nd and Sm isotopes measured independently at the CPT [72]. Further motivated by GW170817 [26–28], two more astrophysical scenarios, potentially found in a merging neutron star environment in accretion discs as well as the dynamical ejecta from the merger [103, 176–181], were considered: a *cold* outflow with $s = 10 \text{ } k_B/\text{baryon}$ and $\tau = 3 \text{ ms}$, and a *hot/cold* outflow with $s = 20 \text{ } k_B/\text{baryon}$ and $\tau = 10 \text{ ms}$. Just like the *hot* scenario of Ref. [72], the neutron concentration is kept fairly high for both scenarios with $Y_e = 0.2$. In the cold case, low- χ^2 solutions were obtained with center around Ce ($Z = 58$). This was reasonable, because in a cold scenario, photodissociation plays a diminished role and the r -process path ventures through the neutron-rich regions closer to the drip-line at early stages. The *hot/cold* scenario

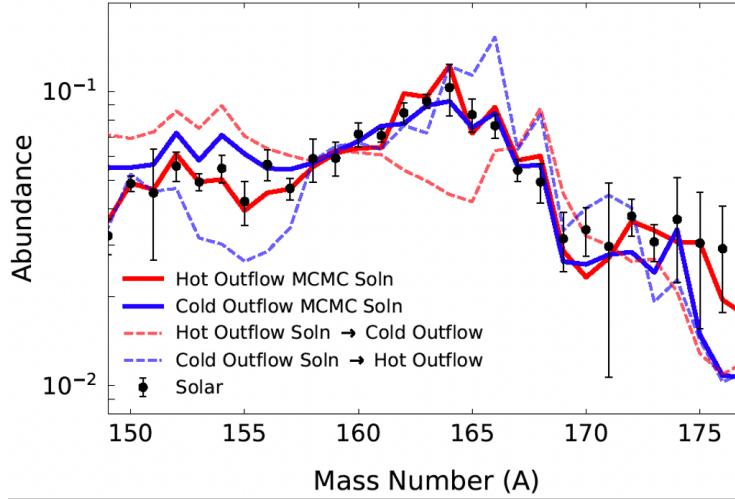


Figure 6.6 : REP abundances from reverse engineering framework for two distinct scenarios (*hot* in red and *cold* in blue) along with the solar abundances. The continuous or solid lines represent the calculated abundances when the solutions for one scenario is applied to the same scenario, while the dashed lines represent abundance calculated using solutions from one scenario applied to the other. Figure taken from Ref. [103].

has characteristics in between the hot and cold processes, starting like a hot r process with (γ, n) playing an important role, and behaving like a cold process at later times of the process. In this case, initial tests conducted with $C = 58$ and 60 both successfully resulted in solutions with low χ^2 , and finally $C = 60$ was used [174]. Like the *hot* scenario, the *cold* and the *hot/cold* were also conducted with $f = 10$ [174]. The results are shown in Figures 6.6 and 6.7 [103].

Figure 6.6 shows the abundance prediction for *hot* and *cold* scenarios in red and blue continuous lines respectively, along with the solar REP in black dots. In both cases, a mass solution was found which led to the REP formation. However, applying the solution from one scenario to the other and vice versa, do not lead to peak formation as shown by the red and blue dashed lines. This proves the existence of distinct solutions for each scenario.

Figure 6.7 shows the mass predictions for the three astrophysical conditions (*hot* in red, *cold* in blue, and *hot/cold* in green) for six elements. The central line and the band in each color represent the average and the standard deviation of 50 independent MCMC runs. Masses from AME2012 [175], past CPT publications [53, 72–74] (“CPT past”), and this thesis are shown. Masses of $^{151,155}\text{Ce}$, ^{155}Pr , and ^{154}Pm were measured after the manuscript of Ref. [103] was prepared and not included in the paper, but are included here. The measured

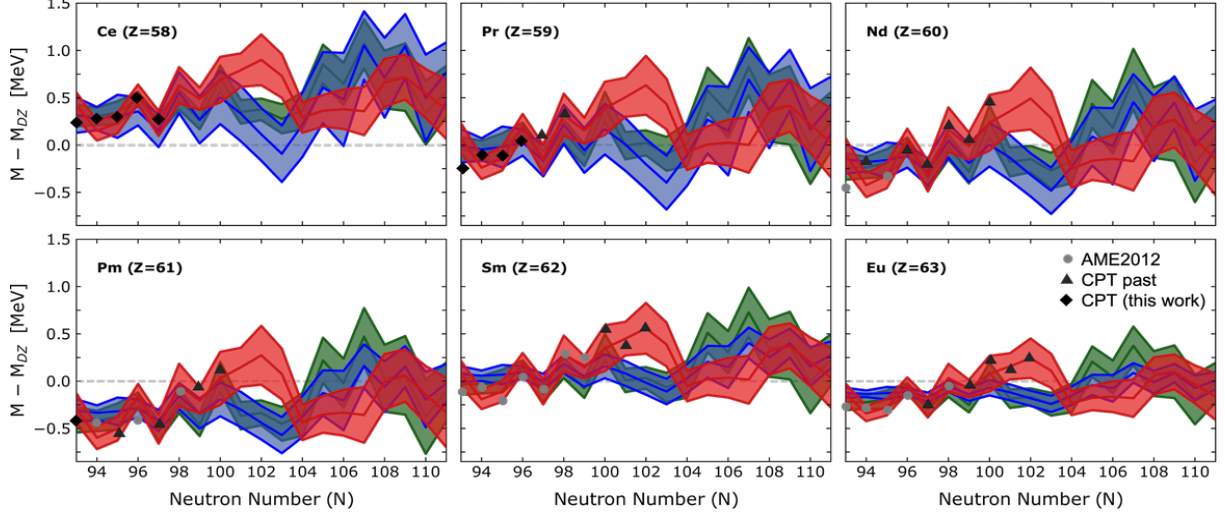


Figure 6.7 : Measured masses from the CPT compared to predictions from reverse engineering framework for three astrophysical scenarios (*hot* in red, *cold* in blue and *hot/cold* in green) for isotopes of elements Ce – Eu. Figure adapted from Ref. [103] and the markers, representing the measured masses, have been altered to differentiate between results from this work and CPT past.

masses consistently follow the predictions for the *hot* environment for all six elements. Furthermore, the predicted mass bands for the *cold* and the *hot/cold* conditions gradually fall out of agreement with the measured masses, as one moves to the more neutron-rich isotopes. This is an encouraging result as it points to a very specific set of conditions for the *r* process to take place. But there could be many such scenarios, and more simulations using those scenarios will need to be made and compared with experimental data. Since nuclides around $Z = 58$ were observed to be more influential for the peak formation in the cold scenario [174], a significant number of focussed measurement campaigns were conducted to measure masses of cerium isotopes to test the viability of the model. Even though in this region, the red bands (for the *hot* scenario), the narrowest of all three bands, always overlap with the blue (*cold* scenario) and the green (*hot/cold* scenario) bands, there are regions where the blue and green bands are distinguishable from the red one. The AME2020 [30] reports extrapolated masses for nuclides $^{152-155}\text{Ce}$, with large uncertainties of $\geq 200 \text{ keV}/c^2$. The left subfigure in Figure 6.8 shows the mass predictions from the reverse engineering framework for isotopes of Ce compared against the AME2020 values. For example, the AME2020 value of ^{154}Ce is actually on the blue band, and only agrees, barely, with the *hot* *r*-process model due to its large uncertainty of 200 keV. The current CPT results, for the isotopes of Ce ($Z = 58$) in

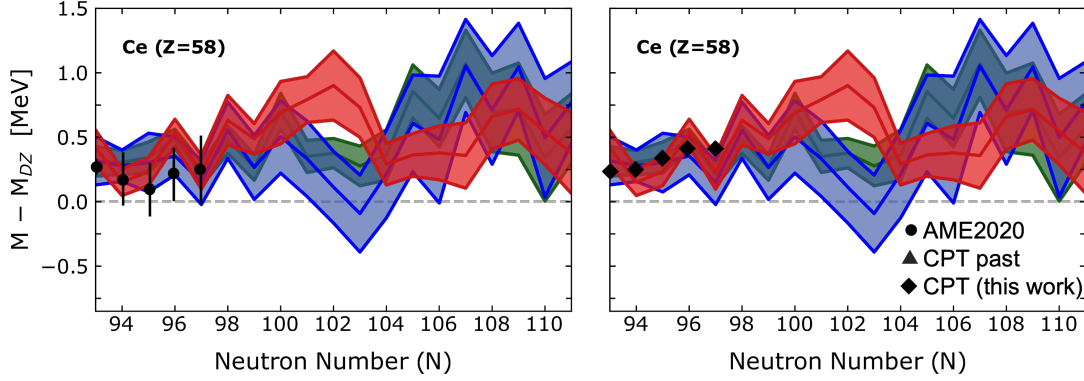


Figure 6.8 : Reported mass excesses from the AME2020 [30] and this work, compared to predictions from the reverse engineering framework for the three astrophysical scenarios (*hot* in red, *cold* in blue and *hot/cold* in green) for isotopes of Ce.

Figure 6.7, are shown for a second time in the right panel in Figure 6.8 to clarify the impact of the CPT results from this work. The agreement of these measured values, shown by the black diamonds in Figure 6.8, with predictions for the *hot* scenario show that the model still holds in this region.

The mass predictions for the *hot* process has a distinct drop between $N = 102 - 104$ for all the elements shown in Figure 6.7. The nuclides residing in this region are, therefore, important to confidently and completely validate these conditions as responsible for REP formation. Of the nuclides produced at CARIBU in this region, ^{166}Eu , produced from the $6.3 \times 10^{-5}\%$ branch of ^{252}Cf fission, presents the best opportunity to probing this drop. In the course of this thesis, multiple attempts were made to measure the mass of ^{166}Eu . For different reasons, including instabilities of the MR-TOF due to a deteriorating power supply used to bias the mirror electrode 5, the presence of a close contaminant molecule (mentioned in Section 5.4.1), pressure related discharge issues leading to an unstable electric field at the CPT, and an overall decrease in extracted fission fragments from CARIBU by almost 70% for the 2+ species after the facility restarted from the COVID-19 enforced shutdown, it was not possible to measure it. The upcoming $N = 126$ Factory, discussed in Chapter 7, however offers a promising path to measuring the masses of nuclides, and points to an exciting future.

6.3.2 Impact on abundance pattern calculations

The impact of the masses presented here on the r -process abundance pattern are being investigated currently by N. Vassh and M. Mumpower [182]. Preliminary results of these

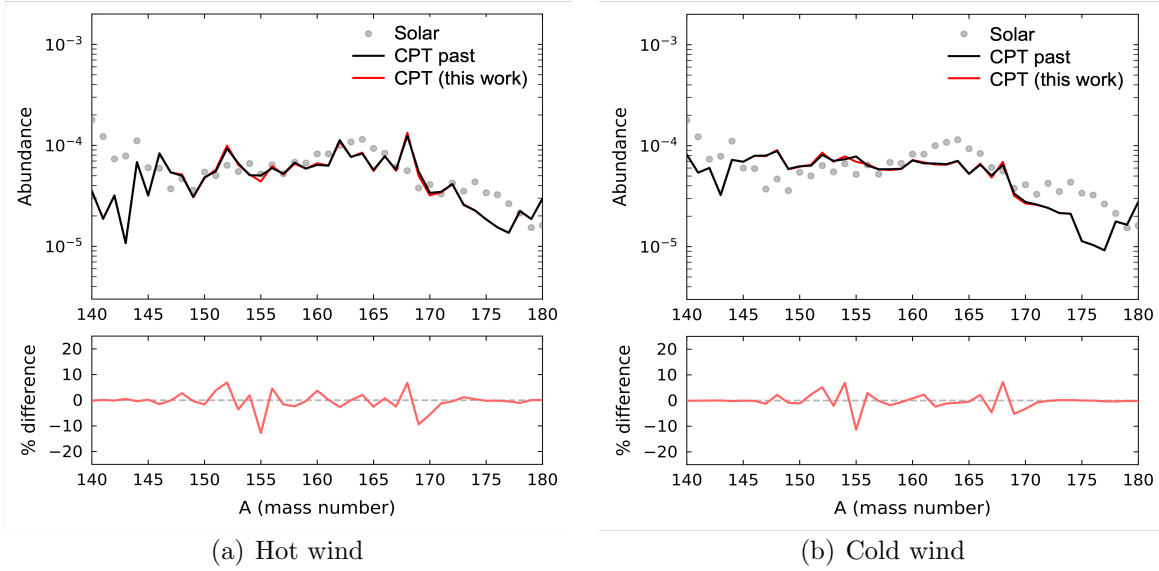


Figure 6.9 : Impact of the measured masses on abundance pattern with β -decay rates and n_β emission probabilities (P_n) being adapted from NUBASE2020 [3], for a hot and a cold wind scenario. The top panels in both subfigures show the abundances calculated using masses from this thesis, masses from past CPT publications, and the solar abundance pattern. The bottom panels show the percentage difference between the abundance calculations from masses from this thesis and past CPT publications.

calculations are presented here. The nuclear properties, namely the neutron separation energies and neutron capture rates were updated based on the masses from this work. β -decay rates and β delayed neutron (n_β) emission probabilities (P_n) were taken from the NUBASE2020 [3]. Using these nuclear properties, nucleosynthesis was computed for the *hot* and *cold* wind scenarios from Section 6.3.1. These were compared to similar calculations using past CPT TOF-ICR [67, 68, 70] and PI-ICR [53, 72–74] measurements. In absence of CPT data, the DZ masses were used. The results are shown in Figure 6.9. The black lines in the top panels of both subfigures (6.9(a) for *hot* and 6.9(b) for *cold* r -process) show the abundance calculations using masses measured in the past using the CPT [53, 67, 68, 70, 72–74], and red lines reflect the masses reported here, along with the solar abundance patterns (in grey dots). On the bottom panels, the differences (in %) between the calculated abundances from this work and past CPT results are shown. The main changes can be seen on the lighter side of the REP around mass numbers $A \sim 150 - 156$ from masses of nuclides $^{151,152}\text{La}$, $^{150-155}\text{Ce}$, and $^{151-155}\text{Pr}$, and after the peak around $A \sim 168$ due to the measurements of ^{168}Gd , ^{169}Dy , and $^{168,169}\text{Ho}$.

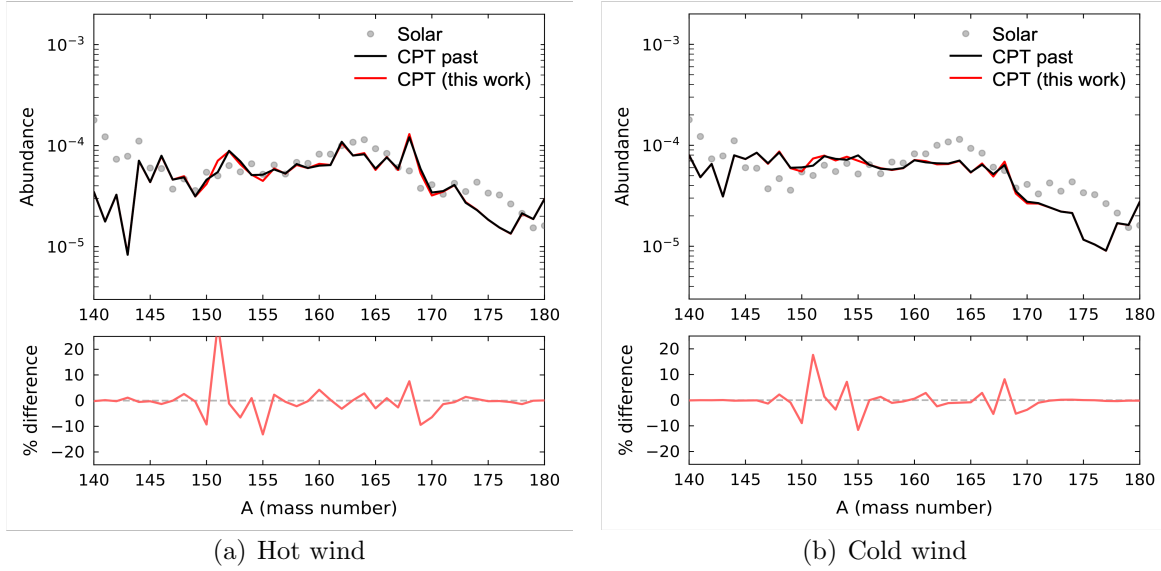


Figure 6.10 : Impact of the measured masses on abundance pattern with n_β emission probabilities (P_n) being calculated using the measured masses and overwritten on the NUBASE2020 [3] values, for a hot and a cold wind scenario. As before, the top panels in both subfigures show the abundances calculated using masses from this thesis, masses from past CPT publications, and the solar abundance pattern. The bottom panels show the percentage difference between the abundance calculations from masses from this thesis and past CPT publications.

The calculations were repeated, but this time the measured masses from this work and past CPT measurements [53, 67, 68, 70, 72–74] were used to also calculate the β delayed neutron (n_β) emission probabilities (P_n) using code BeoH [183], in addition to the neutron separation energies, and neutron capture rates. These calculated P_n values were overwritten on those available from the NUBASE2020 [3]. The results are shown in Figure 6.10. As with Figure 6.9, the top and bottom panels signify the abundance calculations and deviations due to masses from this thesis and past CPT measurements. The main differences are again observed around mass numbers $A \sim 150 - 156$ and $A \sim 168$. However, the magnitude of this deviation is bigger in this case. This is due to a considerable change in the calculated P_n values caused by the nuclides $^{151,152}\text{La}$ [184]. There are plans to undertake additional simulations wherein the high precision measurements of this thesis will be used to reduce the uncertainties in the reverse engineering framework. These detailed calculations require time and substantial computational power to conduct, and they were not possible to complete in the time-frame of this work. These will be completed in the upcoming months prior to any publication.

6.3.3 Astromers

Certain nuclides have been observed to have long-lived (> 1 ns [185]) excited states. The presence of these states, also known as nuclear isomers, was first theorized by F. Soddy in 1917 [186] and have been studied over the last century [187]. Over the last few decades, research into the importance of isomers in explaining the solar elemental abundance has gained traction [188]. Some of this research has been indirect, like nuclear structure studies investigating presence of nuclear deformation which could lead to the formation of the REP [73, 74]. Studies to explore the direct effects of these isomers on the abundances have also been performed [185, 187, 189]. These astrophysically important isomers, also known as “astromers”, have been seen to affect the r -process abundance curve. In hot environments, the ground state of a nuclide can get excited to its isomeric state, depending on the excitation energy between the two states. Furthermore, sometimes in extreme conditions like NSM, the isomer can get preferentially populated over the ground state. Studies have shown the presence of a thermalization temperature above which a nucleus behaves like a single species, while below it, the ground and isomeric states of the nucleus must be treated as different, as internal transitions will dominate [187]. Therefore, depending on the surrounding temperature, the dynamics of the process would be influenced by the nuclear properties (excitation energies, β -decay branching ratios, and half-lives) of the two states. If the two states differ in their $t_{1/2}$ significantly (factor of ≥ 10), the decay towards stability can be delayed or accelerated during nucleosynthesis depending on which state is considered [187]. In order to compute accurate r -process simulations, precise nuclear mass data for these astromers is necessary to calculate their excitation energies and hence ground-state to isomeric-state transition rates. Sensitivity studies of these astromers [185, 187, 189] have revealed the nuclei with $A \sim 120 - 140$ to be crucial. Masses of ground-astromer pairs of ^{119}Ag , ^{119}Cd , ^{128}Sb , and ^{132}Sb were measured and presented in this thesis. The implications of these measured masses and excitation energies on r -process calculations are currently being investigated by our theorist collaborators, W. Misch and M. Mumpower at Los Alamos National Laboratory, USA.

The nuclide ^{128}Sb and its isomer are particularly important. The significant difference

in $t_{1/2}$ between the two states, 9.05 (0.04) h for the ground state and 10.41 (0.18) min for the isomeric state, mean the isomer could greatly accelerate the decay, consequently pushing the process path towards stability faster and affecting production of subsequent nuclides. The level scheme of the ^{128}Sb was deduced from β -decay studies of ^{128}Sn [145, 190] and there are some inconsistencies in these results. Taking into account the systematics in these experiments, the ground state of the ^{128}Sb was assigned a spin-parity of $J^\pi = 8^-$, while for its isomer, $J^\pi = 5^+$ [3]. The high spin of the ground state means that it is not fed by the decay of the ^{128}Sn , which hence populates mainly the isomer [145]. Additionally, an upper limit of 20 keV on its excitation was placed [3] on the basis of calculations of the internal conversion coefficients [190]. But the 43.4(23) keV measured excitation energy at the CPT changes our understanding of this isomer, and therefore, a re-measurement of the structure of the ^{128}Sb and ^{128}Sn nuclides is warranted. Calculations are currently being conducted at the Los Alamos National Laboratory to realize the effect of this measured excitation energy on astrophysical simulations. Additionally, a proposal [191] has been approved at the May 2022 meeting of the ATLAS Program Advisory Committee (PAC) to conduct these measurements. This research aims to understand the level schemes in these two nuclides through decay spectroscopy measurements using the HEART (HEXagonal ARray for Triggering) detector array, in conjunction with Gammasphere [192, 193].

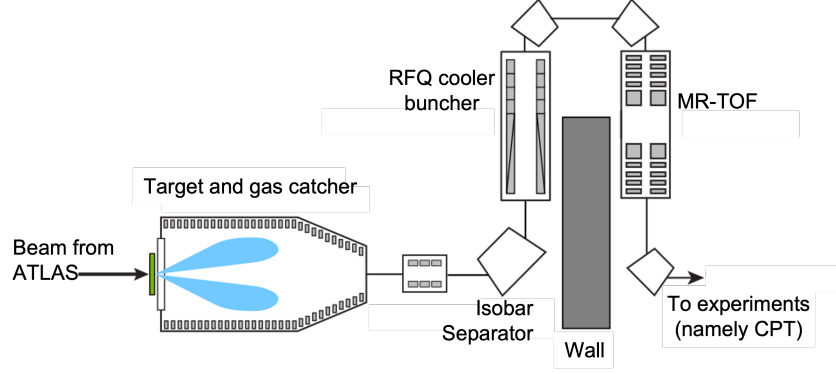


Figure 7.1 : A schematic view of the “N = 126 factory” with the main components labelled. Figure courtesy: A.A. Valverde.

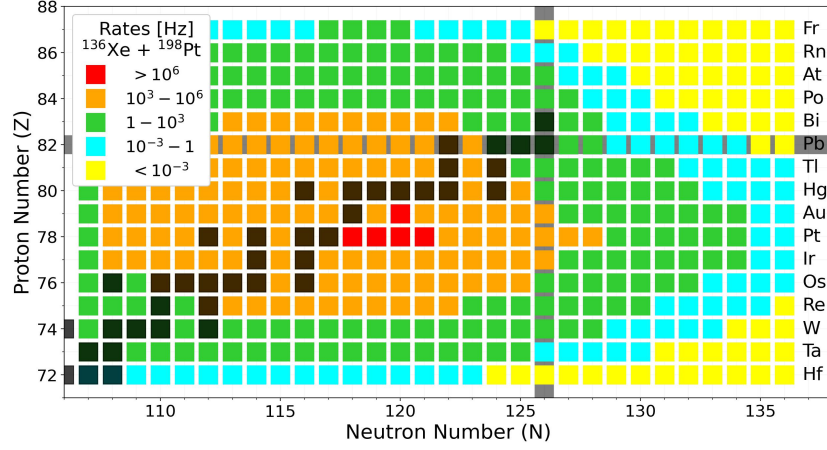
7 Future

The CPT has reached the limit of its capabilities with CARIBU. Despite gains in the efficiency of ion transport, better purification of the beams, and increased stabilities of the devices, dwindling activity because of the decay of the source, and reduced yield as we move into the wings of the fission yield, contributes to this. The next step in *r*-process research is probing the heavy peak in the *r*-process abundance pattern. As mentioned in Chapter 1, the “N = 126 factory” under construction at the ATLAS facility will enable us to probe this heavy region. Isotopes around $Z = 55 - 90$ will be produced by Multi-Nucleon Transfer (MNT) of a high intensity ($\sim 5 \text{ p}\mu\text{A}$) heavy-ion beam (eg ^{136}Xe) on targets made from neutron-rich stable isotopes of heavy elements (eg ^{198}Pt , ^{164}Dy , or ^{154}Sm) [38]. A schematic diagram of the facility is shown in Figure 7.1. The MNT recoil products will be collected using a gas catcher, and will undergo purification by passing through a dipole isobar separator, an RFQ cooler-buncher [194], and an MR-TOF [195] in combination with a BNG, before they are sent to different experiments, including the CPT.

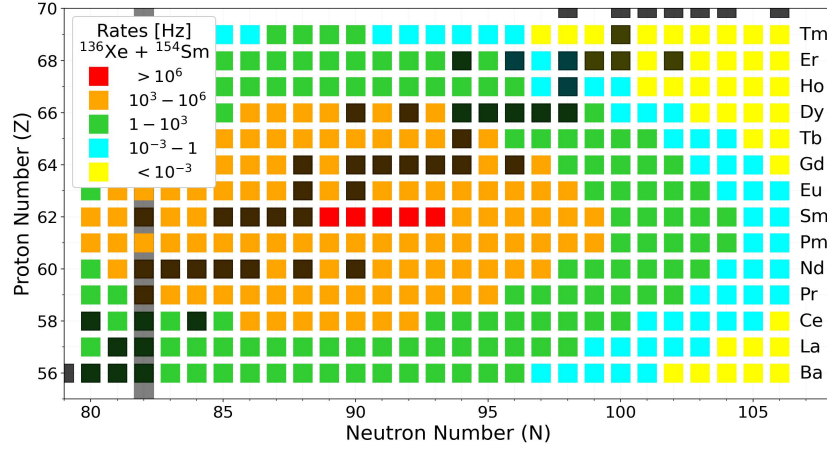
The main goal of the “N=126 factory” will be to help probe the $A \sim 195$ mass region. Historically, these nuclides have proven difficult to access, leading to little to no data currently existing for them. MNT reactions have not been explored as a means of producing these isotopes because of the challenges in collecting the products after such peripheral reactions occurring very close to the grazing angles [38]. However, by using a large volume gas catcher, these difficulties can be addressed, and with an extraction and beam manipulation set up

similar to CARIBU, a significant amount of mass-separated beam is expected to be obtained. Figure 7.2(a) shows expected ion rates, extracted from the gas catcher with 15% efficiency, using GRAZING calculations of ^{136}Xe beam at energies of $\sim 5 - 10$ MeV/u on ^{198}Pt target [196, 197], done by graduate student B. Liu. In addition to probing the $A \sim 195$ peak, the “ $N = 126$ factory” can be operated as a “Rare Earth Peak (REP) factory” by using MNT reactions to produce nuclides around the rare earth peak with ^{154}Sm and ^{164}Dy targets, shown in Figures 7.2(b) and 7.2(c). The facility is nearing completion and is expected to be operational by the end of 2022.

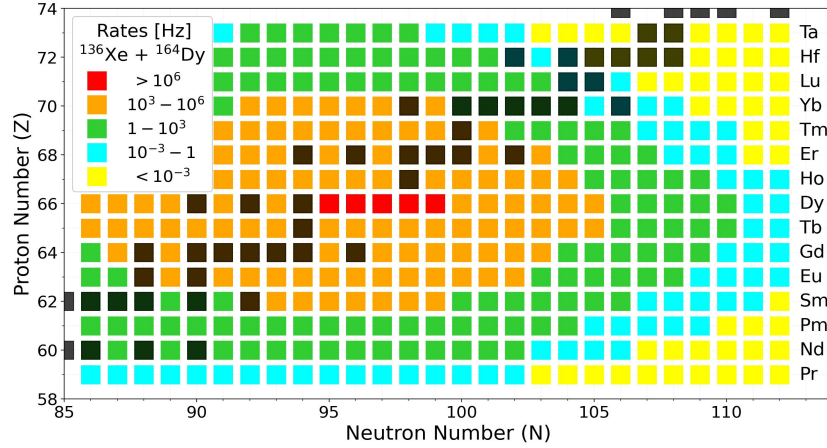
The CPT is currently being prepared to be moved to its new home at the “ $N = 126$ factory”. Over the next few years, the CPT will measure masses of some of the most neutron-rich nuclides: around $A \sim 165$ to explore the drop in MCMC-predicted mass bands around $N \sim 102 - 104$, and nuclides in the $A = 195$ peak in the r -process abundance pattern. Three proposals [198–200] have recently been approved for mass measurements using the CPT at this “ $N=126$ Factory”. While this marks the end of the CPT at CARIBU project, it points to an exciting future in studying neutron-rich nuclides far from stability. The expected r -process path still lies further out than the reach of this facility. Mass models will continue to be needed. The extension of the range of “measured” nuclei should allow testing and re-normalizations of these models and lead to an improvement in the reliability of the data that they provide. This, along with better computational resources, data (both nuclear and astronomical) and the evolution of the astrophysical models should contribute to a more robust understanding of the r process.



(a) “N=126 factory” MNT reaction: $^{136}\text{Xe} + ^{198}\text{Pt}$



(b) “REP factory” MNT reaction: $^{136}\text{Xe} + ^{154}\text{Sm}$



(c) “REP factory” MNT reaction: $^{136}\text{Xe} + ^{164}\text{Dy}$

Figure 7.2 : Projected production rates at the “N = 126 factory” upon extraction from the gas catcher using ^{136}Xe beam on three different targets: ^{198}Pt target (Panel (a)), ^{154}Sm (Panel (b)) and ^{164}Dy (Panel (c)). Also shown are the stable isotopes in black squares in all three panels and the magic N and Z numbers in gray bands (panel (a)).

Appendix I : Uncertainties in measurements

Table A1 : A summary of the statistical, systematic and reported uncertainties in measurements of the frequency ratios between the cyclotron frequencies of the calibrant and the species-of-interest.

Species - of - interest	Calibrant species [Cal.]	$r = \frac{\nu_c^{[\text{Cal.}]}}{\nu_c}$	$\sigma_r \times 10^9$	
			Statistical	Systematic
$^{119}\text{Ag}^+$	$^{133}\text{Cs}^+$	0.894 737 70(14)	17	140
$^{119m}\text{Ag}^+$	$^{133}\text{Cs}^+$	0.894 737 861(31)	27	13
$^{119}\text{Cd}^+$	$^{133}\text{Cs}^+$	0.894 693 875(31)	28	13
$^{119m}\text{Cd}^+$	$^{133}\text{Cs}^+$	0.894 695 043(31)	28	13
$^{128}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.962 407 088(20)	19	5.2
$^{128m}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.962 407 438(21)	20	5.4
$^{132}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.992 543 951(12)	12	2.8
$^{132m}\text{Sb}^+$	$^{133}\text{Cs}^+$	0.992 545 123(12)	12	2.8
$^{147}\text{Ba}^{2+}$	C_6H_6^+	0.941 325 707 4(97)	8.4	4.8
$^{147}\text{La}^{2+}$	C_6H_6^+	0.941 281 923 3(97)	8.4	4.7
$^{151}\text{La}^{2+}$	C_6H_6^+	0.966 998 063 8(91)	8.5	3.2
$^{152}\text{La}^{2+}$	C_6H_6^+	0.973 434 322(41)	41	3.5
$^{147}\text{Ce}^{2+}$	C_6H_6^+	0.941 245 013 9(97)	8.5	4.7
$^{150}\text{Ce}^{2+}$	C_6H_6^+	0.960 513 659(21)	21	3.8
$^{151}\text{Ce}^{2+}$	C_6H_6^+	0.966 945 175 4(77)	6.9	3.5
$^{152}\text{Ce}^{2+}$	C_6H_6^+	0.973 367 710(16)	16	3.2
$^{153}\text{Ce}^{2+}$	C_6H_6^+	0.979 802 813(17)	17	3.0
$^{154}\text{Ce}^{2+}$	C_6H_6^+	0.986 227 433(18)	18	2.8
$^{155}\text{Ce}^{2+}$	C_6H_6^+	0.992 664 512(52)	52	3.2
$^{147}\text{Pr}^{2+}$	C_6H_6^+	0.941 221 330 2(99)	8.7	4.7
$^{151}\text{Pr}^{2+}$	C_6H_6^+	0.966 906 942 3(77)	7.0	3.2
$^{152}\text{Pr}^{2+}$	C_6H_6^+	0.973 333 983(69)	69	3.2

$^{153}\text{Pr}^{2+}$	C_6H_6^+	0.979 755 797(17)	17	2.7
$^{154}\text{Pr}^{2+}$	C_6H_6^+	0.986 186 636(18)	18	2.5
$^{155}\text{Pr}^{2+}$	C_6H_6^+	0.992 610 782(35)	35	2.4
$^{147}\text{Nd}^{2+}$	C_6H_6^+	0.941 202 971 6(99)	8.7	4.8
$^{151}\text{Nd}^{2+}$	C_6H_6^+	0.966 878 267 3(78)	7.1	3.2
$^{151}\text{Pm}^{2+}$	C_6H_6^+	0.966 861 402 3(82)	7.3	3.7
$^{154}\text{Pm}^{2+}$	$^{154}\text{Nd}^{2+}$	0.999 980 959(71)	71	2.6
$^{168}\text{Gd}^{2+}$	$^{84}\text{Kr}^+$	1.000 746 569(36)	36	3.1
$^{169}\text{Dy}^{2+}$	$^{84}\text{Kr}^+$	1.006 658 279(10)	9.5	3.0
$^{168}\text{Ho}^{2+}$	$^{84}\text{Kr}^+$	1.000 670 824(20)	20	3.1
$^{168m}\text{Ho}^{2+}$	$^{84}\text{Kr}^+$	1.000 671 198(20)	20	3.1
$^{169}\text{Ho}^{2+}$	$^{85}\text{Rb}^+$	0.994 778 712(14)	14	3.2

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