

**Staking out the Tm Proton Drip-line
with Mass Spectrometry
and
Using Electrons to Sympathetically Cool Ions
Inside a Penning Trap**

by

Brian A. Kootte

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

Doctor of Philosophy

Department of Computer Science
The University of Manitoba
Winnipeg, Manitoba, Canada
April 2023

© Copyright 2023 by Brian A. Kootte

Thesis advisor
Gerald Gwinner

Author
Brian A. Kootte

**Staking out the Tm Proton Drip-line
with Mass Spectrometry
and
Using Electrons to Sympathetically Cool Ions Inside a
Penning Trap**

Abstract

Energy differences in nuclei are paramount to understanding the nuclear systems far from stability, their rare decay modes, and trends in their relative stability. Modern ion trapping techniques such as Penning Trap Mass Spectrometry (PTMS), and Multiple Reflection Time Of Flight Mass Spectrometry (MRTOF-MS) have enabled the precision measurement of energy trends in even very short-lived nuclei, by allowing their masses to be measured. Such nuclei can be generated in large quantities at TRIUMF's Isotope Separator and Accelerator (ISAC), and TRIUMF's Ion Trap for Atomic and Nuclear science (TITAN) specializes in measuring the masses of the short-lived species produced at ISAC using an array of ion traps.

This thesis describes measurements of the masses of neutron-deficient lanthanide elements produced at the ISAC facility at TRIUMF in Vancouver, Canada using the TITAN Multiple Reflection Time-Of-Flight (MR-TOF) mass spectrometer. The goal of these measurements was to extend the known existence of the N=82 shell closure up to Yb (Z=70), and to experimentally determine the location of the proton drip-line in Tm (Z=69). Mass measurements of ^{150}Yb , ^{151}Yb , and ^{153}Yb confirm the continued existence of the N=82 shell closure. Additionally, new mass measurements of ^{149}Tm and ^{150}Tm were performed, as well as a re-measurement of ^{149}Er that provides a correction to the literature value as published in the 2020 Atomic Mass Evaluation. Together, these mass data points provide the first experimental confirmation that

^{149}Tm is the first proton-unbound species in the Tm isotopic chain.

Furthermore, a new technique intended for rapidly cooling highly-charged ions (HCIs) produced via charge-breeding in TITAN's Electron Beam Ion Trap (EBIT) is experimentally tested. This technique involves sympathetically cooling the ions using free electrons, and holds the promise of cooling HCIs on a time-scale of seconds or less for studies with either short-lived or stable species. Technical challenges to implementing sympathetic cooling are discussed, and evidence for fast, electron-facilitated cooling of singly-charged species is presented.

Contents

Abstract	ii
Table of Contents	vi
List of Figures	vii
List of Tables	xi
Acknowledgments	xii
Dedication	xiv
1 Nuclear Physics Background	1
1.1 Introduction and Scientific Objectives	1
1.2 Binding Energy	3
1.3 The Nuclear Shell Model	5
1.3.1 Testing Models and Magic Numbers	8
1.4 Charged Particle Emission and the Drip line	10
1.4.1 Proton Emission	11
1.5 Summary	14
2 Ion Traps for Precision Mass Spectrometry at TITAN	15
2.1 Mass Spectrometry of Exotic Nuclei	15
2.1.1 Resolution, Resolving Power, and Precision	16
2.1.2 Facilities for Producing Exotic Isotopes	16
2.1.3 Producing Isotopes at TRIUMF's Isotope Separator and AC- celerator (ISAC)	17
Delivering Radioactive Beam to Experiments at ISAC	18
2.2 Ion Trap Mass Spectrometry of Radioactive Isotopes	20
2.2.1 An Overview of Penning Trap Mass Spectrometry	21
FT-ICR	24
TOF-ICR	24
PI-ICR	26
Penning Trap Precision	27
2.2.2 Brief Overview of Multiple Reflection Time-of-Flight (MRTOF) Mass Spectrometry	27
Time of Flight Mass Spectrometry	27

	The MRTOF Technique	28
	MRTOF Resolving Power	31
2.3	Mass Spectrometry Conclusion	32
2.4	TRIUMF's Ion Trap for Atomic and Nuclear Science (TITAN)	32
2.4.1	The TITAN RFQ Cooler Buncher	34
2.4.2	The TITAN MRTOF	36
	Limitations on the beam rate	40
	Mass-Selective Retrapping to Suppress Contaminants	41
	Mass Range Selector	44
	Systematic uncertainties in the MRTOF	46
2.4.3	The TITAN MPET	46
2.4.4	The TITAN EBIT	49
2.4.5	Motivation for a Cooler Penning Trap	52
3	MRTOF Mass Measurements of Neutron-Deficient Tm and Yb	56
3.1	Rare Isotope Beam Production	57
3.2	MRTOF Measurement and Analysis	58
3.2.1	First Retrapping Online	59
3.2.2	Time-Resolved Calibration	60
3.2.3	Peak Fitting Procedure	63
	Fit Function	63
	Peak Width	63
	Weighting the Fit and Assigning a Data Cut Threshold	64
	Fit Procedure Summary	65
3.2.4	Note on Fitting with Exponentially Modified Gaussian	66
3.2.5	Mass Calibration	67
3.2.6	Statistical Uncertainty Characterization	71
3.2.7	Systematic Characterization	73
4	Results of the Lanthanide Mass Measurements	75
4.1	Results and Discussion	76
4.2	Scientific Impact of Tm and Yb Mass Measurements	77
4.2.1	Comparison to Theoretical Models	78
4.2.2	Establishing the Limit of Proton-Bound Thulium	82
	The Impact of ^{149}Er and ^{149m}Er	83
4.3	Newly Determined Mass Values	90
5	The Cooler Penning Trap (CPET)	94
5.1	Sympathetic Cooling for Highly Charged Ions	96
5.2	The Electron Advantage: Benefits of Sympathetic Cooling Using Elec- trons	100
5.2.1	Timescale of Electron Self-Cooling	101

5.3	Design of CPET	103
5.3.1	Possibility of Electron Recombination in CPET	107
5.3.2	Vacuum Requirements in CPET	110
5.3.3	Status of Sympathetic Electron Cooling	110
5.4	Technical Challenges of Electron Cooling	111
5.4.1	Penning Discharge	111
5.4.2	Plasma Behaviour of Trapped Electrons	114
5.5	Hardware Improvements to CPET	116
5.5.1	An Improved Electron Source	117
5.5.2	A Modification for Detecting Trapped Electrons	119
5.5.3	Minimizing Penning Discharges	121
5.5.4	Slowing Electrode Switching	124
5.5.5	High Voltage DAC box	126
5.6	Developments Toward Achieving an Electron-Ion Interaction	127
5.6.1	Electron Injection Into CPET	128
5.6.2	Electron Trapping Lifetime	130
5.6.3	Evidence for Electrons Cooling	131
	Cooling Electrons Within a Shaped Potential Well	132
5.6.4	Co-Trapping of Ions with Electrons	133
5.6.5	Ion (Injection and) Extraction	134
5.7	Studies Performed on the Two Plasma Species	135
5.7.1	Ion Detection	137
	Further Discharge-Like Behaviour in CPET	137
	Penning Ion Accumulation	139
	Application of RF to Electron Trapping	141
5.7.2	Searches for an Electron-Ion Interaction	143
	Using Space-Charge to Test and Optimize Electron-Ion Inter- action	143
	Time Of Flight Approach	146
	Retarding Field Approach	148
5.8	Conclusion of Sympathetic Cooling Studies	152
6	Outlook	156
6.1	Sympathetic Cooling with Electrons	156
6.2	Scientific Outlook for Mass Measurements of Neutron-Deficient Lanthanides	158
A	Overview of Personal Contributions	159
A.1	Contributions to Lanthanide Mass Measurements	159
A.2	Contributions to Cooler Penning Trap	160
	Bibliography	182

List of Figures

1.1	Colour map of all nuclear binding energies taken from the 2020 Atomic Mass Evaluation	5
1.2	In the conventional nuclear shell model the coupling of the spin and orbital angular momentum of nucleons splits energy levels	7
1.3	In the shell model the neutron energy levels (left) and proton energy levels (right) fill independently	8
1.4	The shell closure at N=82 is apparent in the steep change in two-neutron separation energies between N=82 and N=84 when plotting the isotopic chains from Z=59 to Z=70	10
1.5	Diagram of a proton within a simplified nuclear potential	11
1.6	Theoretical relationship between the decay energy and half-life for the proton emitter ^{147}Tm	13
2.1	Exotic isotope production within the target is seen here as simulated in GEANT4 for a 480 MeV proton beam impinging on a Ta target . .	18
2.2	The ISAC Facility at TRIUMF	19
2.3	Schematic diagram of the geometry of an ideal hyperbolic Penning trap	22
2.4	Radial projection of ion motion inside a Penning trap	23
2.5	After undergoing a quadrupolar excitation, the final kinetic energy of an ion in a Penning trap depends on the deviation of the applied RF frequency from the reduced cyclotron frequency	26
2.6	An illustration showing how reflection from an electrostatic mirror can create a new time-focus.	30
2.7	The TITAN Facility	34
2.8	Schematic showing the basic operation of a radiofrequency quadrupole cooler-buncher (RFQ) trap	36
2.9	TITAN's MRTOF mass separator	37
2.10	A re-trapping cycle and a measurement cycle in the MRTOF analyzer	43
2.11	Analyzer showing the Mass Range Selector (MRS) used to remove ions from the trap which undergo a different number of reflections	45

2.12	A pulsed drift tube adjusts the ion beam energy from a transport energy of about 2 keV to match the potential of MPET	47
2.13	An example Time Of Flight Ion Cyclotron Resonance (TOF-ICR) spectrum showing the response in TOF to 41 different excitation frequencies	48
2.14	The magnetic field of an EBIT compresses the electron beam in order to create a deep potential well for ions using their space-charge	50
2.15	“Kicking” the ion bunches extracted from the EBIT can only achieve a compromise between a well-defined focus in position and a well-defined focus in energy	52
2.16	The emittance is determined by the area occupied by the beam in phase space	53
2.17	Possible flow of ions between the ion traps of TITAN	55
3.1	Comparison of raw vs drift-corrected MRTOF time-of-flight data . . .	62
3.2	This fit of the ^{151}Ho peak is an example of a peak that is believed to contain an unresolved overlap of the isomer and ground state	70
3.3	The ^{150}Er calibrant required caution due to its proximity to the tail of the $^{150}\text{Ho}/^{150m}\text{Ho}$ peak	71
3.4	A weighted fit of the ^{150}Er mass calibrant	72
4.1	Comparison of the Yb ($Z=70$) isotope masses measured at TITAN compared with the 2020 Atomic Mass Evaluation	78
4.2	Comparison of the Tm isotope masses measured at TITAN to the 2020 Atomic Mass Evaluation	79
4.3	The ratio of the $4^+/2^+$ excitation energies for $N=82$ nuclei approaching $Z=70$ begins to increase from a value of 1.33	80
4.4	DFT predictions of the two-neutron separation energy of Yb around the $N=82$ shell closure	81
4.5	The shell closure is apparent in the two-neutron separation energies (S_{2N}) given by the newly measured masses of the Yb ($Z=70$) isotopic chain	82
4.6	Theoretical (DFT) predictions of proton separation energy of Tm disagree upon whether the drip line occurs at $N=80$	83
4.7	Raw TOF data at $A=149$	84
4.8	Comparison of the Er isotope masses measured at TITAN in 2019 to the literature values of the 2020 Atomic Mass Evaluation	85
4.9	The 1-proton separation energy calculated with the newly measured Tm and Er masses	86
4.10	A weighted fit of the ^{150}Tm peak after a cut is made at 20% of the peak height	87
4.11	Gaussian fits of ^{149}Er and ^{149m}Er with the distance between them constrained by the known excitation energy	89

4.12	Log plot of the ^{149m}Er peak after a cut is made at 20% of the peak height	89
5.1	The energy spread of ions exiting the EBIT was previously probed using a <i>retarding field analyzer</i> to scan over a range of blocking potentials for the ions. Images courtesy of Ish Mukul.	97
5.2	Electrons orbit the magnetic field in accordance with the left-hand rule, and emit synchrotron radiation in the plane perpendicular to the acceleration vector.	98
5.3	A conceptual illustration of how cooling can be performed with oppositely-charged species by implementing a nested potential configuration . .	101
5.4	Predictions for the sympathetic cooling of ions with different ratios of ions to electrons. Image from [1] and reproduced with permission from Springer Nature.	103
5.5	An image of the CPET trap electrodes and drift tubes, which are housed within the superconducting magnet, and the feedthrough section which is located outside the magnet. From [131].	104
5.6	An 8-segmented electrode (octopole) was included with the intention of using it for rotating-wall compression of ions	105
5.7	The stable ion source used to deliver thermally ionized positive ions (including Na) for CPET tests.	105
5.8	The stable ion source used for CPET tests	106
5.9	Electrodes and detectors surrounding the Cooler Penning Trap (CPET)	106
5.10	Radiative recombination of electrons vs dielectronic recombination . .	107
5.11	Predictions for the sympathetic cooling of ions over time	109
5.12	The Paschen curve shows the expected electrical breakdown voltage (V_B) for various gasses	112
5.13	Mechanism of a Townsend discharge	115
5.14	Mechanism of a Penning discharge	116
5.15	Electrons from the off-axis electron source are steered onto the ion beam axis using the potentials applied to various ion-optical elements	119
5.16	A thermal camera image shows heating of the vacuum chamber due to poorly steered electrons	120
5.17	Images of the harp/mesh detector used to detect electrons within CPET	122
5.18	Electrodes within CPET were reconfigured and/or shorted to minimize the possibility of Penning discharges.	123
5.19	The previous CPET electrode configuration before improvements were made. Image from [131].	124
5.20	The new configuration of the CPET electrodes maximized the distance between higher-voltage, and switching electrodes	124
5.21	Old vs new wiring for biasing the CPET electrodes	125
5.22	Electrons were trapped by scattering them behind the gate electrodes	129
5.23	Number of electrons detected on the harp detector vs trapping time .	131

5.24	Simulation showing increased ion radius upon injection in CPET when the second steering element after the ion source is used for focusing .	136
5.25	Side view of SIMION simulations of the ion radius in CPET upon injection	137
5.26	An example potential configuration for combining ions with electrons	147
5.27	Sympathetic cooling of ions is demonstrated by observing the effect of trapping duration on the time-of-flight on extracted ions	149
5.28	On-axis potential with a triple electrode end-cap	150
5.29	The effect of electron cooling on scans of the number of detected on the final MCP detector versus the blocking voltage applied to the end-cap electrode. Image provided by Stefan Paul.	150
5.30	The energy of trapped $^{23}\text{Na}^+$ ions in the trap was measured for various trapping duration	152
5.31	Same as Figure 5.30, for the energy of trapped $^{39}\text{K}^+$ ions as a function of trapping time. Provided by Stefan Paul.	153
5.32	Same as Figure 5.30, for the energy of trapped $^{85}\text{Rb}^+$ ions as a function of trapping time. Provided by Stefan Paul.	153
5.33	Ion losses in the trapped $^{23}\text{Na}^+$	154
5.34	Ion losses in the trapped $^{39}\text{K}^+$	154
5.35	Ion losses in the trapped $^{85}\text{Rb}^+$	155

List of Tables

2.1	A summary comparing some of the relative strengths and limitations of the Penning trap and MRTOF mass spectrometry techniques at TITAN	31
4.1	A summary of the 2018 measurements of Yb atomic masses	91
4.2	A summary of the 2019 measurements primarily of Tm atomic masses	92
5.1	A comparison of ion cooling time constants when trapped with electrons to when they are trapped without introducing electrons into the trap.	152

Acknowledgments

Writing this acknowledgment is difficult because there have been so many amazing people who have played so many different roles in my journey up until this point; it is not possible to be comprehensive. I thank God for these people in my life and for the abundance of privilege that has enabled me to pursue this work and this degree.

Many thanks go to the people who gave me the opportunity to do this work. To my thesis advisor, Gerald Gwinner. You have forever impressed upon me the importance of computer backups, and modeled many aspects of the type of supervisor and scientist that I strive to be; thanks for your encouragement, support, and humour. Jens Dilling, for breathing life into TITAN and for providing your helpful advice when needed. Thanks to Ania Kwiatkowski for your scientific guidance, your amazing feedback on presentations, and for providing opportunities to take part in so many different aspects of both science and leadership.

Thanks to my committee members for your time.

I would like to thank the CPETeers I have had the opportunity to work with: Brad Schultz, Usman Chowdhury, Dan Lascar (who forever impressed upon me the value of precise language), Stefan Paul (whose drive for data collection was nothing short of inspirational), and Roshani Silwal. The results obtained from CPET hinged critically on the diligent work of every single one of these people. I would also like to thank our exceptional undergraduate co-op students, Rane Simpson, Solbee Seo, and Duane Byer.

Thanks to Soenke Beck, Marilena Lykiardopolou, and Pascal Reiter for useful discussions on the Yb data.

I feel I must acknowledge hard work of the many professional administrators at TRIUMF and the University of Manitoba, who keep these departments running; in particular, every step of this program could have been astronomically more difficult without Susan Beshta always providing a vital lifeline to the University of Manitoba.

I am grateful for the supportive environment of TITAN students and postdocs who I have had the pleasure of working with and learning from. Aaron, Renee, Erich, Annika, Julia, Eleanor, Tobias, Pascal for many great technical discussions, Carla, Chris, Marilena, James, Ruben, Sam, Andrew F., Andrew J., Brad, Yang, Kyle, Jeff...

Thanks to Mel Good for supporting the sometimes naive requests of physicists

with his professional know-how. The same goes for the many other talented people at TRIUMF including but not limited to: Daryl Bishop and the TRIUMF electronics group, the high-voltage engineering group, Jens Lassen, the many beam and accelerator experts, and Ben Smith for his hard work on the CPET controls in the final days.

Thanks to all the people who have shared their knowledge and encouragement with me. Bruno Olaizola Mampaso, Bruno Arderucio, and Jennifer Fallis Starhunter come to mind, but there have been many others.

Thank you Mom; more than all the practical support you gave, thank you for modeling the forging of relationships with people around me, openness, and concern for others, and for providing an amazingly supportive environment in which I could believe in myself and strive for anything I put my mind to. For this I am grateful. Thanks to Glenn for the patience and support. My brother Derek, for taking the time to provide feedback on my public speaking and for being a constant friend in my life. Laura, your understanding, your support, your feedback, and your sense of humour have brought me much encouragement and joy throughout this process. Thanks Dad, for the many breakfasts and lunches, home and car maintenance, and for sort of accepting that I haven't taken up a trade and started earning some real money...

*This thesis is dedicated to Grammy and Poppy. You truly went beyond
the call of duty of grandparents in my life, and I will always be grateful
for that.*

Chapter 1

Nuclear Physics Background

In the light of [current research on atomic structure] the physicists have, I think, some justification for their faith that they are building on the solid rock of fact, and not, as we are often so solemnly warned by some of our scientific brethren, on the shifting sands of imaginative hypothesis.
— Ernest Rutherford’s Opening Address to the British Association for the Advancement of Science, Winnipeg, 1909 [2]

1.1 Introduction and Scientific Objectives

Precision mass spectrometry is one of the foundations of modern nuclear science. Ever since the work of Rutherford and Brooks uncovered the first solid evidence that radioactive processes can transmute elements from one chemical species into another [3], scientists have sought to study the properties of the products of nuclear reactions. During the early explorations of the nature of the atomic nucleus, Francis William Aston observed that the mass of a nucleus is not precisely equal to the additive sum of the masses of its constituent nucleons, but rather, the composite system had slightly less mass than expected. He initially attributed this “mass defect” to the interference of electromagnetic fields in the densely packed region of the nucleus. He also inferred that the energy released would be proportional to the mass destroyed, and that a greater release of energy would result in a more stable nucleus [4]. While the true origin of this mass defect was, in fact, primarily due to the fields associated

with the *strong interaction* rather than those of electromagnetic fields, the predicted consequences of these nuclear masses proved to be key to understanding the physics of these systems. Through great labours, Aston was able to measure the charge-to-mass-ratio to much better than single mass-unit precision with his earliest mass spectrometer [5], which led to the first quantification of the “binding energy” of many nuclei (see Section 1.2) using a mass spectrometer claiming a mass accuracy of 1/1000 [4].

Nuclear masses are a fundamental observable in studying the atomic nucleus, and as such, their precise measurement is often required to test nuclear models, constrain astrophysical calculations, and test fundamental interactions. The content of this thesis revolves around the development and utilization of cutting edge mass spectrometry techniques for atomic and nuclear physics. Precision mass spectrometry techniques are applied to measure masses of exotic nuclei in order to address questions at the forefront of nuclear science.

Chapter 1 provides scientific motivation and background, by addressing in detail a few of the areas of fundamental research that can be addressed through atomic mass measurements, as well as some of the relevant historical context. This will lead to a more detailed discussion in Chapter 2 of modern mass spectrometry techniques and their relevance to enhancing our understanding of nuclei and their structure. Chapter 2 goes on to describe the TITAN facility and the experimental set-up as it pertains to mass spectrometry of increasingly exotic nuclear isotopes at TRIUMF, an accelerator laboratory located in Vancouver, Canada. Chapter 3 will provide a detailed account of the measurement and analysis procedures used to obtain the masses of neutron-deficient Yb and Tm isotope chains using the TITAN’s MRTOF mass spectrometer, and Chapter 4 will discuss the results and scientific consequences of this analysis, including the impact on the binding energies and resulting drip line of neutron-deficient Tm isotopes. Finally, chapter 5 will discuss technical developments made towards achieving the first cooling of short-lived, highly-charged ions using sympathetic electron cooling. Although the primary motivation of this research was to enhance the capabilities of Penning trap mass spectrometry of rare isotopes, the technique could be applied in several other fields of science.

1.2 Binding Energy

Building on the work of Aston, George Gamow first addressed the theoretical basis of the mass defect by modelling the nucleus as “*a collection of particles attracting one another with the forces very rapidly decreasing with distance*”. In this way, the nucleus behaves in a similar fashion to a droplet of liquid, with the outermost particles experiencing a *surface-tension* effect [6]. This simple model proves to be a very useful description of the nucleus when trying to predict the energetics of a particle bound within the nuclear potential. The change in the total binding energy of a system as a result of undergoing a nuclear reaction is a fundamentally important quantity for reaction rates and pathways known as the reaction *Q-value*, which is given by the difference between the total mass of all reactants and the total mass of all products, $Q = (\Sigma m_r - \Sigma m_p)c^2$.

Today, the enhancement in the nuclear mass observed in more weakly bound nuclei attributes it to the corresponding increase in the energy of the system via the famous mass-energy relation, $E = mc^2$. This energy results from the collective forces acting within the system of nucleons due to all interactions (e.g. the strong nuclear force, the electromagnetic interaction, etc.). As such, precise determination of the nuclear binding energy obtained through measurement of a nuclide’s mass can provide insight into its microscopic structure, as well as the nuclear processes in which it may take part.

Gamow’s liquid drop model led to the formulation of the *semi-empirical mass formula* by Von Weizsäcker [7]. It predicts the mass of an atomic nucleus using the following expression:

$$M(Z, A) = ZM_p + NM_n - E_B(Z, N)/c^2 \quad (1.1)$$

with the total mass number, A , of a nuclide, being the sum of the proton number Z and the neutron number N ($A = Z + N$). Furthermore, M_n is the mass of the neutron, and M_p the mass of the proton. The binding energy, E_B , is determined by the equation

$$E_B = a_V A - a_S A^{2/3} - a_C \frac{Z(Z-1)}{A^{1/3}} - a_A \frac{(A-2Z)^2}{A} \pm \delta, \quad (1.2)$$

where a greater binding energy implies that the nucleus is more strongly bound ¹.

The first term in the binding energy equation, $a_V A$, accounts for the finite-range, mutual attraction between nucleons, and therefore increases with A . The second term, $a_S A^{2/3}$, makes a correction to account for the reduced attractive forces at the surface of the nucleus. The third term, $a_C \frac{Z(Z-1)}{A^{1/3}}$, accounts for the long-range, mutually repulsive Coulomb force between the protons. The fourth term, $a_A \frac{(A-2Z)^2}{A}$, is an asymmetry term, which takes into account that protons and neutrons are fermions; as such, they independently fill up higher and higher energy levels, and therefore are in the lowest energy state when their numbers are balanced under the assumption of isospin (i.e. proton/neutron) symmetry. The final term, δ , is a pairing term which accounts for the odd-even staggering in the binding energy due to spin pairing of both types of nucleon.

If one considers a chain of isobars (momentarily ignoring the pairing term), the mass number, A , remains constant. Therefore, only the third and fourth terms in equation 1.2 remain to influence the trend in binding energies. The third term, $-a_C \frac{Z(Z-1)}{A^{1/3}}$, will become quadratically more negative with increasing Z , and the fourth term, $-a_A \frac{(A-2Z)^2}{A}$, will become quadratically more negative with increasing N . This produces an approximately parabolic decline in binding energy, which is centred on the most strongly bound nuclei. For even mass numbers, the pairing term will split the doubly-paired even-even nuclei and the unpaired odd-odd nuclei into two parabolas, offset from one another in energy.

This relatively simple model of the nucleus provides a useful approximation of the masses of nuclei both at, and close to, nuclear stability by fitting these constants to the known data. However, experimental measurements of nuclei reveal many often subtle, but important departures from the semi-empirical formula, which become more prominent far from stability. Figure 1.1 shows a colour map of the experimental *binding energy per nucleon* of the known nuclides using data from the 2020 Atomic Mass Evaluation, a compilation of experimentally measured masses of the known nuclides. The modern mass spectrometry techniques used to measure binding energies

¹Since this expression is for nuclear binding energies, it does not take into account the masses and binding energies of orbital electrons.

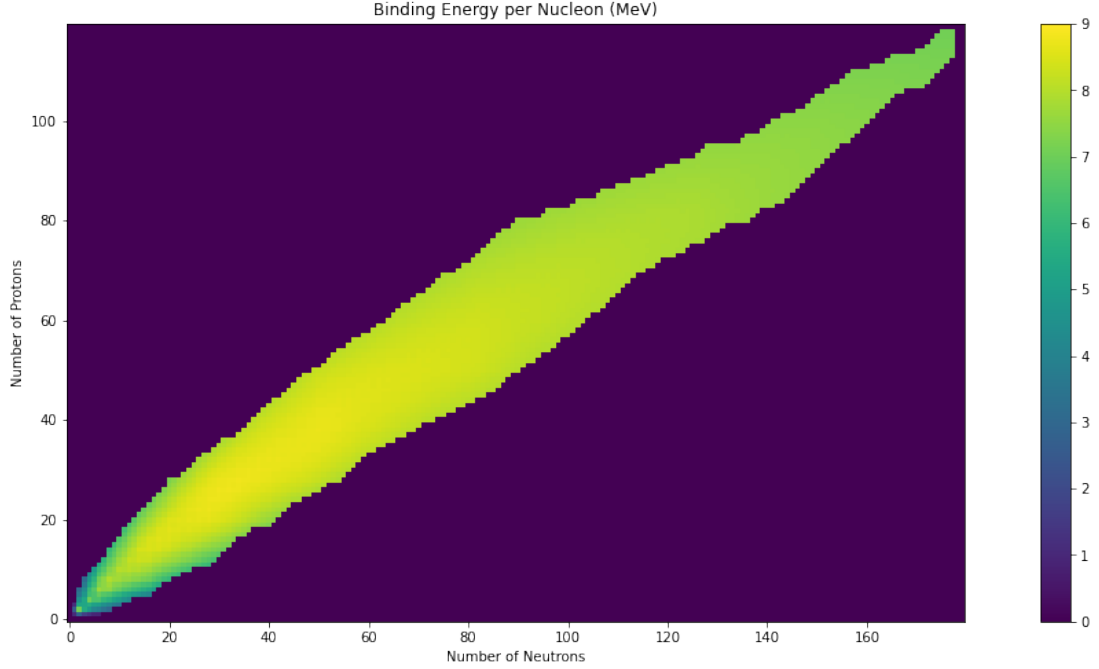


Figure 1.1: Colour map of all known nuclear binding energies on a scale of 0 to 9 MeV per nucleon. Each square represents a distinct species of a given atomic number, Z , and neutron number, N . The maximum binding of approximately 8.8 MeV/nucleon occurs at $Z = 28$ and $N = 34$ (^{62}Ni). Note the substantial increase in binding energy between ^1H and ^4He in the lower left corner. Data taken from the 2020 Atomic Mass Evaluation [8].

of the very unstable nuclei studied in this thesis are discussed in Section 2.2.

1.3 The Nuclear Shell Model

The so-called nuclear *magic numbers* were independently explained in the late 1940s in work by Maria Goeppert Mayer as well as Otto Haxel, J. Hans D. Jensen, and Hans E. Suess [9][10][11]. They realized that existing observational evidence could explain the magic numbers of nuclei to be a consequence of the sequential filling of all available angular momentum states for each energy level of an oscillator bound in a potential well, providing that a *spin-orbit* correction was included in

the potential. The nuclear spin-orbit interaction is a consequence of the alignment (or anti-alignment) of a nucleon's spin with its orbital angular momentum, and is analogous to the electromagnetic spin-orbit interaction of atomic physics.

Although the proton and neutron are composite particles, they have spin $1/2$ and follow Fermi-Dirac statistics similarly to electrons. They therefore are subject to the Pauli exclusion principle, allowing no two identical particles to occupy the same state, but the protons and neutrons fill states independently from one another (see Figure 1.3). The quantum number of the strong interaction which breaks the symmetry between protons and neutrons is known as isospin, where the projection onto the flavour eigenstates is given by $I_3 = \frac{1}{2}(n_d - n_u)$, where n_d and n_u are the number of 'down' and 'up' quarks. This means that protons have an isospin of $-1/2$ and neutrons an isospin of $+1/2$, and the ground state of a bound system of these two particles will independently occupy the lowest energy levels of each ². To a good approximation, protons and neutrons can often be treated as different states of the same particle, differing only in their isospin. Therefore, the ground state of a bound system of these two particles will independently occupy the lowest energy levels of each, up to the most energetic nucleon (which is at the *Fermi energy*). With all of the orbitals filled, the particles cannot exchange energy with one another.

Nucleons fill up nuclear *shells*, in a similar fashion to the electron shells of atomic theory, but there are several differences between nuclear and atomic shells. As mentioned above, since they are different particles, protons and neutrons sequentially fill up separate sets of orbitals in accordance with Pauli exclusion principle (mostly independently from one another). Secondly, the nuclear potential is more complicated than the central potential of an atom; the nucleus is bound by a short-range force which is distributed relatively uniformly throughout the nuclear matter. Thirdly, the energy gaps between nuclear shells are made less apparent by the strong spin-orbit induced splitting of energy levels in the nucleus. The energy levels which impact the shell closures are influenced by these differences. This results in shells that are consistent with a potential intermediate between a square well and a three-dimensional

²The sign convention is reversed in particle physics, with $I_3 = \frac{1}{2}(n_u - n_d)$.

harmonic oscillator [10]. Spin orbit effects then shift the energy levels, as shown in Figure 1.2, resulting in closed nuclear shells for both neutrons and protons at 2, 8, 20, 28, 50, and 82 nucleons, and at 126 neutrons for species relatively close to stability. However, these magic numbers may change for isotopes lying far from stability [12].

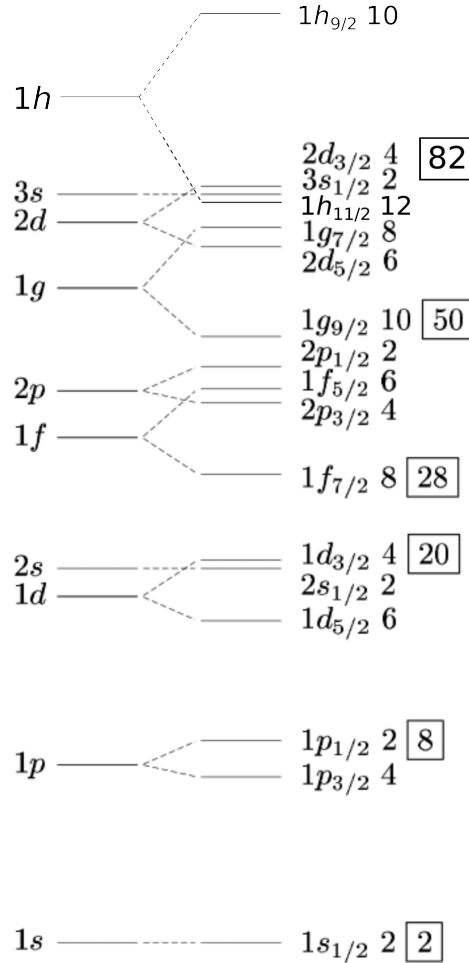


Figure 1.2: In the conventional nuclear shell model the coupling of the nuclear spin with the orbital angular momentum of nucleons splits energy levels, and their clustering results in the known nuclear magic numbers of 2, 8, 20, 28, 50, 82, 126, and 184. Modified from an image by Bakken under CC BY-SA 3.0.

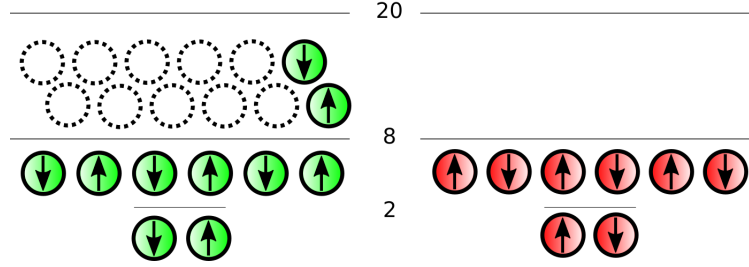


Figure 1.3: In the shell model the neutron energy levels (left) and proton energy levels (right) fill independently. Here, ^{18}O is given as an example. This is a simplified picture since the angular momentum states are not shown.

1.3.1 Testing Models and Magic Numbers

The fact that nuclear shell closures can weaken or disappear far from stability means they are of fundamental scientific interest in order to accurately describe the nucleus.

Since the identification of nuclear shell closures is often less obvious than for their atomic shell analogues, especially for very unstable isotopes, several different criteria are routinely used to identify their presence. The first nucleon added beyond a magic number will be less strongly bound than expected from those leading up to the magic number due to having crossed the shell closure. This change in the binding energy trend gives rise to a corresponding change in the mass trend, so mass measurements are valuable for characterizing shell closures. Also, since all the angular momentum levels are filled at a closed shell, any single-nucleon excitation will require a larger amount of energy than usual to enter the next available orbital. This results in an enhancement in the first excitation energy.

The presence of a nuclear shell closure can thus be tested through several related observables. Such effects include:

- The two-neutron or two-proton separation energy, S_{2N}/S_{2P} (see Equation 1.3).
- The neutron or two-proton pairing gap, Δ_{2N}/Δ_{2P} (see Equation 1.4).
- A higher excitation energy of the first excited state for even-even nuclei ($E2^+$).

- An increase in charge radius after crossing shell closure (seen through isotope shifts).
- An extended half-life at a shell closure.

The relative increase in binding energy that impacts the S_{2N} and Δ_{2N} values can be observed through direct measurements of the ground-state nuclear masses. Another way to think about this is that the mass will start to become lower than expected as the final nucleons are added to fill up a closed shell. Subsequent nucleons added beyond a shell closure must occupy an orbital of the next available shell, so low lying excited states of closed shell nuclei are likely to have an enhanced excitation energy from their ground state. The effect of the $N=82$ shell closure is clearly seen in the S_{2N} plots shown in Figure 1.4.

The neutron separation energy, S_N indicates how strongly a single neutron is bound to the rest of the nucleus. The two-neutron separation energy, S_{2N} , is a representation of the strength of neutron binding, while canceling out pairing effects. It is defined by the expression

$$\begin{aligned} S_{2n}(Z, N) &= B(Z, N) - B(Z, N - 2) \\ &= (-M(Z, N) + M(Z, N - 2) + 2m_n)c^2. \end{aligned} \tag{1.3}$$

The neutron pairing gap indicates how rapidly this binding is changing for a given nuclide with respect to the neutron number. This is essentially a discrete derivative of the two-neutron separation energy:

$$\begin{aligned} \Delta_{2n}(Z, N) &= S_{2n}(Z, N) - S_{2n}(Z, N + 2) \\ &= 2 \times B(Z, N) - B(Z, N + 2) - B(Z, N - 2) \\ &= (-2 \times M(Z, N) + M(Z, N + 2) + M(Z, N - 2))c^2. \end{aligned} \tag{1.4}$$

These values provide a quantifiable measure of how the nuclear binding energy evolves along an isotopic chain in order to identify the regions of stronger binding associated with a shell closure.

Therefore, nuclear masses are among the most important observables for understanding the properties of exotic nuclei. They provide a means of testing both the

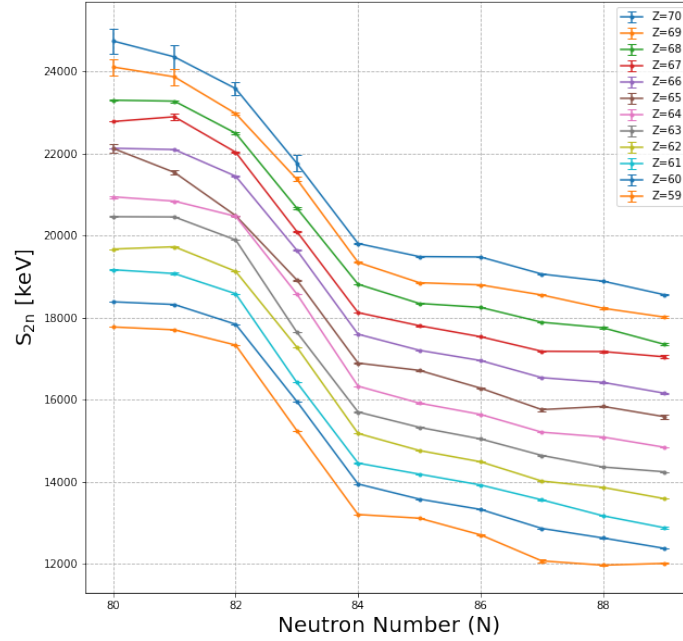


Figure 1.4: The shell closure at $N=82$ is apparent in the steep change in two-neutron separation energies between $N=82$ and $N=84$ when plotting the isotopic chains from $Z=59$ to $Z=70$.

simple nuclear models discussed above, as well as more sophisticated models. The masses provide a benchmark for comparison when modelling the trends in these many-body systems ever further from nuclear stability.

1.4 Charged Particle Emission and the Drip line

The proton drip line is defined as the point at which a chain of nuclei transition from all protons being bound in the ground state, to a nuclear species for which it becomes energetically favourable for the ground state to emit one or more protons [13][14].

The theory of charged particle emission has its origin in the development of the Geiger-Nuttall Law during the early days of nuclear physics. H. Geiger and J.M. Nuttall observed that the alpha decay rate depends exponentially on the energy of

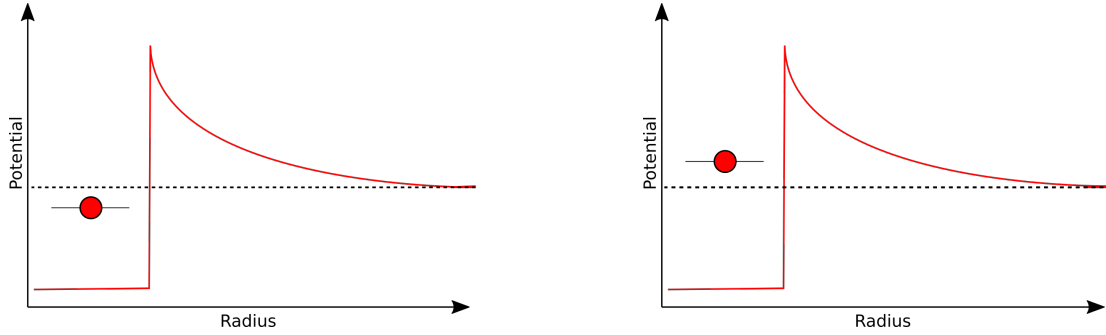


Figure 1.5: Diagram of a proton within a simplified nuclear potential. In the image on the left the proton is in a bound state and decay is energetically impossible, whereas in the image on the right the proton is unbound and will eventually escape the nucleus.

the emitted alpha particle [15][16], or equivalently, the decay Q-value [17],

$$\log(t_{1/2}) = aQ^{-1/2} + b, \quad (1.5)$$

where a , and b are constants, $t_{1/2}$ is the alpha emission half-life, and Q is the Q-value.

In 1928 George Gamow realized that this process can be thought of as an alpha particle quantum mechanically tunneling through the classically forbidden region just outside the relatively sharp cutoff of the nuclear surface [18].

Though we now understand that an alpha particle is a composite system, consisting of two protons and two neutrons, it is salient to remember that the theory of alpha decay was developed before Chadwick's 1932 discovery of the neutron.

1.4.1 Proton Emission

In a similar fashion to alpha decay, the repulsive Coulomb force can allow a single proton to tunnel out of the nuclear potential well and escape as a free proton. The difference between a proton which is bound within its potential well and one which is unbound is shown on the left of Figure 1.5. In this way, this exotic nuclear decay mode operates via a mechanism very similar to alpha decay. However, unlike alpha decay, *ground state proton emission* is energetically possible in only a minority of the known proton-rich nuclei, and there are no known long-lived proton emitters. Proton emission half-lives tend to be shorter than those of alpha decay, because the

process does not rely on the formation of a helium nucleus before tunneling through the potential barrier.

Proton emission from a parent nucleus, ${}^A_ZX_N^3$, is modelled by considering a proton residing within the potential of the daughter nucleus ${}^{A-1}_{Z-1}Y_N$. The proton occupies a local potential minimum, and after a very large number of reflections from the potential barrier it is confined within, it ultimately passes through the barrier via quantum tunneling to access the lower energy state of a free proton plus a separate daughter nucleus.

It is extremely useful to look at the energetics of nuclear decays in terms of the reaction Q-values. For the decay of a single particle, the Q-value is given by: $Q = (M_{parent} - \sum M_{decayproducts})c^2$. Any nuclide for which the Q-value for proton emission from the ground state is positive is considered to lie beyond the proton drip line. However, these nuclei represent only a small minority of the table of nuclides.

In contrast to protons, α -particles are not considered to have a drip line. Although many nuclei exist as metastable species, for which $Q_\alpha > 0$ (i.e. alpha decay is energetically possible), α -particles tend to be emitted more slowly than protons due to the finite probability of forming the alpha particle within the nucleus, and the need for twice the charge to tunnel through the Coulomb barrier, as can be seen in a generalized form of the Geiger-Nuttall law [19].

Additionally, proton emission provides a valuable, and unique window through which to study nuclear structure. In contrast to alpha decay, proton emission allows for a single-particle state to be directly probed, since only one nucleon is emitted. Proton emission is therefore an important nuclear process which will be addressed in the experimental work of this thesis. This decay mode defines one of the two drip lines for radioactive nuclei [20]. It is important to draw the distinction between proton emission from a ground state, versus that of an excited state. Many nuclei have excited states which can also emit protons, but the proton drip line is defined by the nuclides for which ground-state proton emission becomes a possible decay mode. Furthermore, this process is fundamentally different than electron capture or

³ A is the mass number of the nucleus, Z the atomic (i.e. proton) number, and N the neutron number.

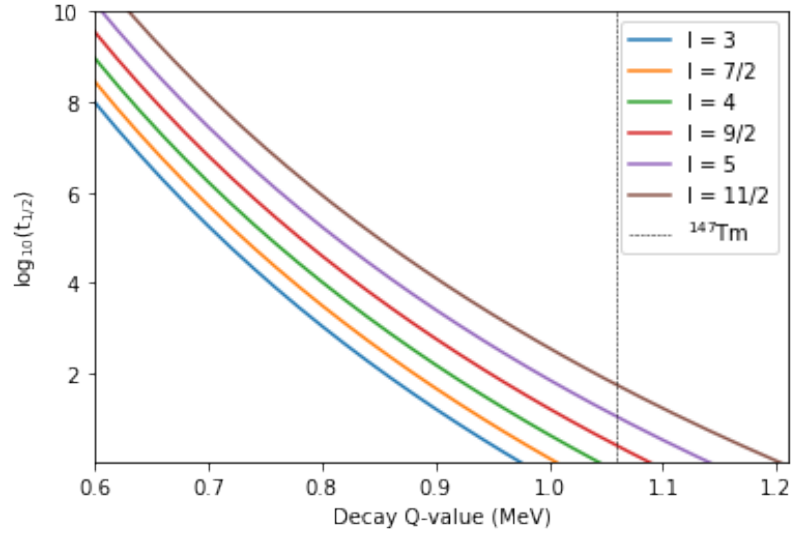


Figure 1.6: Dependence of the ^{147}Tm half-life on the decay energy using the model of Zhang et al. [21]. Each curve shows the theoretical relationship when assuming a different angular momentum change associated with the decay, and the dotted line shows the known proton emission decay energy of ^{147}Tm .

β^+ decay in that it does not rely on the flavour-changing weak interaction to facilitate it.

Furthermore, the single-particle character of the emitted proton allows a relationship to be established between the half-life, and the decay energy of the emitted proton. The half-life also depends sensitively on the angular momentum, l , carried away by the proton to allow the nucleus to transition from initial to final state. For example, using Formula 2 from reference [21], as well as the coefficients obtained from the fit performed in the same paper, the relationship between the proton emission half-life of ^{147}Tm and the energy of the decay process Q_p is shown graphically in Figure 1.6. It can be seen that the proton decay half-life can be very sensitive to even relatively modest changes in the Q_p -value in regions beyond the drip life.

Lastly, the proton drip line itself is of fundamental interest. The location of the proton drip line has been predicted through a Bayesian analysis of the results of applying various density functionals within density functional theory [14]. Mass and/or decay spectroscopy measurements are required to experimentally test such

predictions of the limits of proton binding.

1.5 Summary

Over the last century, nuclear physics has evolved from using simple models to describe obscure phenomena, into a richly detailed picture of the complexities of nuclear systems. The dynamics of these systems can be described through various theoretical frameworks with varying degrees of success. Some of the most exotic nuclei are often also the most challenging to describe, but are of fundamental scientific interest. To address this challenge, nuclear observables are needed to benchmark models and determine the properties of these nuclei. Nuclear masses provide a key observable which allows for the direct determination of nuclear Q-values and for probing trends in exotic nuclei.

Chapter 2

Ion Traps for Precision Mass Spectrometry at TITAN

2.1 Mass Spectrometry of Exotic Nuclei

Much of the work of this thesis revolves around utilizing and understanding the nuclear processes which transmute one species of nucleus into another. In order to understand the dynamics of nuclei that may decay by proton emission, state-of-the-art radioactive beam facilities are needed to produce the nuclei under study. Studies performed on these nuclei in turn, exploit modern techniques in atomic and nuclear physics in order to increase our knowledge of the fundamental properties of these species.

The mass spectrometry techniques discussed within this chapter enable valuable measurements of exotic isotopes for studies in nuclear physics. These measurements pose a unique set of challenges, but many opportunities exist to determine the masses of highly unstable species which approach the extremes of the mass parabola that results from the simple model of binding energy discussed in section 1.2. These masses can provide crucial insight by uncovering the nuclear binding energies which cannot be described by the classical shell model. This includes regions with high collectivity, emerging [22] or quenched [23] shell effects, or other exotic behaviour. Both proton and neutron separation energies can be important for understanding astrophysical

nucleosynthesis pathways and the locations of the drip lines are of fundamental interest. Furthermore, high precision mass measurements are essential to calculate the β -decay Q -values of superallowed beta emitters. These provide a crucial input for the precise determination of the V_{ud} component of the CKM matrix which contributes to tests of its unitarity [24].

2.1.1 Resolution, Resolving Power, and Precision

To meaningfully assess the capabilities of mass spectrometers, it is worthwhile to briefly define some terminology that will be used throughout this thesis.

The *resolving power* of a mass spectrometer is a measure of the closest peaks that can be distinguished from one another. It is defined by $R = m/\delta m$, where m is the mass measured, and δm is the *mass resolution*¹. Although other definitions exist, here resolution will refer to the width of a peak as measured at 50% of the maximum height (i.e. the Full Width Half at Maximum, or FWHM). However, the ratio of ions in two adjacent peaks will clearly also play a role in whether or not it is possible to resolve the weaker peak. Unless otherwise indicated, any stated resolving power will assume that the two species are measured with approximately a 1:1 ratio.

Precision refers to the repeatability of measurements which arises from the width of a mass peak, while *accuracy* reflects how close the peak lies to the true mass value [25]. Masses can be determined significantly more precisely than the resolution since the uncertainty in the location of the centroid of the the peak can be known much more precisely than its width, as long as the shape is reasonably well understood.

2.1.2 Facilities for Producing Exotic Isotopes

In order to address many of the pressing questions in nuclear physics, state-of-the-art experimental facilities are required to produce the exotic isotopes for study.

There are two primary categories of exotic isotope facilities. *In-Flight* facilities initiate the nuclear reactions that generate isotopes by impinging a relatively heavy

¹It is important to note that in some fields and textbooks, the definitions of resolving power and resolution are the reverse of the definition presented here

beam on a thin target, and *Isotope Separation On-Line (ISOL)* facilities usually produce isotopes by directing a low mass beam (such as protons) on a thick target. There are several differences between these isotope production methods. In-Flight facilities operate at higher beam energy and the products produced by the collisions within the thin target retain their forward momentum in the reference frame of the laboratory and therefore remain as a beam. In contrast, the reaction products will stop within an ISOL target, and must diffuse out before being re-accelerated to experiments.

These two isotope production methods are complementary to each other. While ISOL facilities are able to produce very rich beams of the highest intensities, In-Flight facilities can supply beams of isotopes with the shortest half-lives. A significant difference between these two isotope production mechanisms is also seen in the composition of the beams they produce. Radioactive ISOL beams depend heavily on the high-temperature chemical and physical processes occurring between the element being extracted and the chemical environment of the target itself. In contrast, In-Flight beams depend primarily on the nuclear reaction physics. The beam in an In-Flight facility is produced in a fully ionized state, whereas the target at an ISOL facility initially releases most atoms in a neutral state, and subsequently ionizes them.

2.1.3 Producing Isotopes at TRIUMF's Isotope Separator and ACcelerator (ISAC)

Presently, exotic isotopes are produced at TRIUMF using the Isotope Separator and ACcelerator (ISAC) facility [26]. ISAC is an ISOL-type facility as described in the previous section. In the future, exotic isotopes will also be supplied by the Advanced Rare Isotope Electron Linac (ARIEL) facility, which is expected to triple the production of these species at TRIUMF by adding an electron-induced photo-fission target and an additional proton target [27][28].

The isotope beam under study is produced by impinging the proton beam from TRIUMF's driver cyclotron upon a solid, heavy, target nucleus. Some typical materials used to withstand the high target temperatures include uranium carbide and elemental tantalum, and are selected based on the region of the nuclear chart under

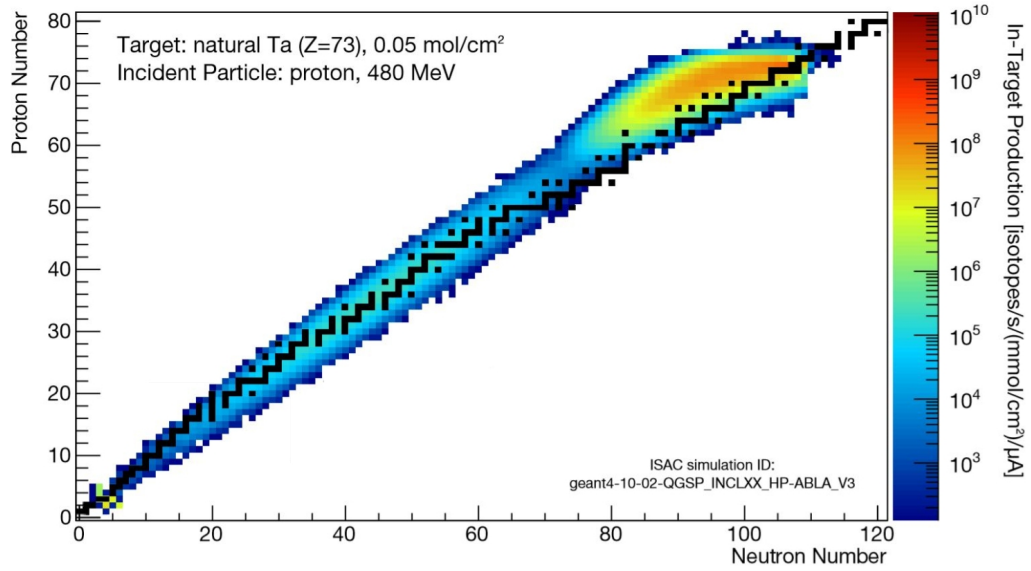


Figure 2.1: Exotic isotope production within the target is seen here as simulated in GEANT4 for a 480 MeV proton beam impinging on a Ta target. Data from <http://isys01.triumf.ca/search/simulation/data>. Image modified from [60] courtesy of Erich Leistenschneider.

study. For this work, a 480 MeV proton beam ranging between 25 and 50 μA , was used to initiate a variety of different nuclear reactions in a tantalum (Ta) target material to produce the isotopes under study. Figure 2.1 shows the simulated in-target production yields within the Ta target. The relevant beams of exotic nuclei were then extracted to study the nuclear binding energies, shell closures, and possible proton emission decay modes for these unstable nuclei. The reaction products escape the target at a rate that depends on the interaction of the element or molecule formed with the target material. This rate strongly depends on the chemical properties of the species in question, such as the vapour pressure, and the rate of the heavy lanthanide elements was observed to vary with the target temperature.

Delivering Radioactive Beam to Experiments at ISAC

Since the elements escaping the ISAC target are largely released in the neutral state, several techniques exist to ionize the atoms released by the target in order to

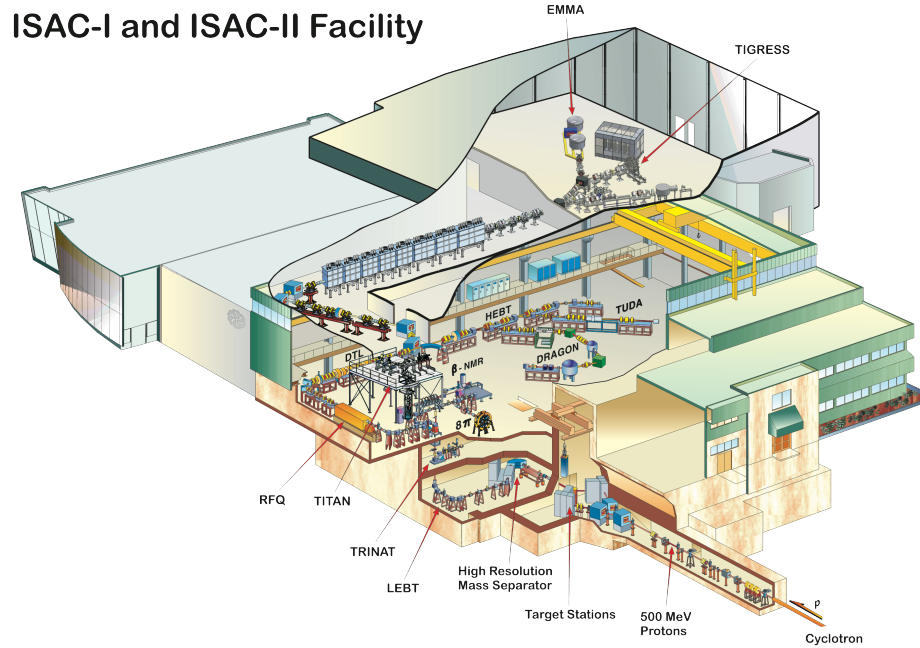


Figure 2.2: The ISAC Facility at TRIUMF from [29]. Reproduced with permission from Springer Nature.

produce an ion beam. A rhenium foil is often used to promote ionization within the ionizing tube due to its high electron work function, and atoms are surface ionized by maintaining the temperature of the tube at approximately 2000 °C.

Alternatively, when a resonant laser ionization scheme exists for a particular element, the yields of one species relative to the other species can be enhanced using the TRIUMF Resonant LASER Ion Source (TRILIS). Additionally, by employing a somewhat more sophisticated setup in the target area, the Ion Guide Laser Ion Source (IG-LIS) can provide the greatest reduction of contaminant elements by using a repelling potential to repel species that undergo surface ionization at or near the target. A LASER then resonantly ionizes the desired species, which escapes the target and enters the IG-LIS as a neutral atom. Once ionized, the ions are guided away from the target region using a linear radio-frequency quadrupole and a DC gradient.

Furthermore, a Forced Electron Beam Induced Arc Discharge (FEBIAD) ion source [30] can be used to ionize atoms with the highest ionization potentials using a plasma discharge. This is a very effective ionization method, however it is also

the least selective with regard to the ions it delivers. Molecules and multiply-charged ions will be included in the beam.

In order to extract the ion beam, the target region is biased with a floating voltage of 20 kV. After the ions are extracted, they are passed through a stage of electrostatic pre-separation before the beam is separated in a magnetic dipole mass separator to produce a beam of the desired mass unit. The Isotope Separator and ACcelerator (ISAC) facility at TRIUMF allows for mass separation of ions into beams of a single mass unit by achieving a mass resolving power of about 2500 [31], depending on the separator settings. Since the enhanced mass defect of exotic nuclei causes them to tend to have larger masses than stable isobars of the same mass number, this allows for adjusting the mass separator to provide a degree of mass selectivity within a given mass unit, A , by adjusting the magnet.

The beam is then transported from the ISAC separator to a low energy experimental facility, such as TITAN, or accelerated to higher energies using the ISAC accelerators (see Figure 2.2).

2.2 Ion Trap Mass Spectrometry of Radioactive Isotopes

In contrast to the measurements of stable nuclides upon which the field of mass spectrometry was founded, mass measurements of radioactive species pose several unique challenges. These include short half-lives, extremely small abundances which are limited by production cross-sections, extensive isobaric contamination, and facility-limited time frames within which these measurements must be completed.

Lower precision techniques such as time-of-flight mass spectrometry can be performed on extremely short-lived species, but higher precision techniques require more involved apparatuses. Today, two of the best techniques for studying the masses of exotic nuclei with high precision are *Penning Trap Mass Spectrometry* and *Multiple-Reflection Time Of Flight Mass Spectrometry (MRTOF-MS)*, the former being a very well established technique for high precision measurements. Their strength lies in

the fine balance of characteristics required to produce accurate, high-precision mass measurements while still accommodating the short half-lives, and minuscule abundances associated with many of these isotopes. Stable isobars will often outnumber the unstable species in a typical ISOL beam. A relevant advantage of MRTOF mass spectrometry is its ability to take a measurement in the presence of significant isobaric contamination. The relative strengths of these two techniques are summarized in Table 2.1.

2.2.1 An Overview of Penning Trap Mass Spectrometry

Penning trap mass spectrometry (PTMS) has become well-established as providing “the most accurate and precise mass measurements” [32] of radioactive ion beams. Penning Traps have been used to study radioactive isotopes since their introduction at CERN’s Isotope Mass Separator On-Line facility (ISOLDE) in 1987 [33]. Since then, similar facilities have proliferated worldwide, including the Canadian Penning Trap which was relocated to Argonne National Lab in 1998 [34][35], JYFLTRAP which was installed at the IGISOL facility at the University of Jyväskylä in 2003 [36], and LEBIT [37] located at Michigan State University’s Facility for Rare Isotope Beams (FRIB). These facilities have greatly expanded knowledge of masses across the nuclear chart due to their ability to obtain precise measurements from short-lived species [38].

A Penning trap is able to statically trap charged particles in three dimensions by combining a strong axial magnetic field, which provides radial confinement, with an axial quadrupolar electric field, which provides axial confinement. A harmonic potential well can be produced with a uniform magnetic field and hyperbolic electrodes [39] (see Figure 2.3). Alternatively, an equivalent field can be produced by applying the appropriate voltages to a series of simpler, cylindrically-symmetric ring electrodes.

The basic equation for the non-relativistic, cyclotron frequency of a particle of mass, m , and charge, q , situated in a magnetic field, B , is given by

$$\nu_c = \frac{1}{2\pi} \frac{qB}{m}. \quad (2.1)$$

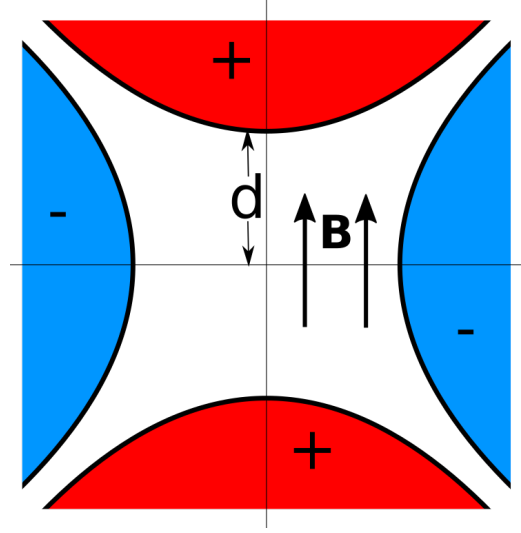


Figure 2.3: Schematic diagram of the geometry of an ideal hyperbolic Penning trap for positive ions shown in the longitudinal plane. A full rotation about the vertical axis gives the three-dimensional form of the trapping electrodes. Positive ions can be confined by the quadrupolar electrostatic potential when the red electrodes are biased with a higher positive voltage than the blue electrodes.

This relationship allows Penning traps to be superbly useful for precisely determining the mass of a trapped particle, because modern clocks allow for measurements up to extremely high frequencies.

Inside a Penning trap the ion motion is described by three independent eigenmodes. These include the reduced cyclotron frequency ν_+ , the magnetron frequency ν_- , and the axial frequency ν_z . They are related to the cyclotron frequency by

$$\begin{aligned}\nu_c &= \nu_+ + \nu_-, \\ \nu_c^2 &= \nu_+^2 + \nu_-^2 + \nu_z^2, \\ \text{and} \\ 2\nu_+\nu_- &= \nu_z^2.\end{aligned}\tag{2.2}$$

In practice, ν_z is typically significantly smaller than the other two frequencies [40][41] and can therefore often be neglected ². A projection of the radial motion of an ion in

²The rotational frequency is related to the angular frequency by $\nu = \frac{\omega}{2\pi}$

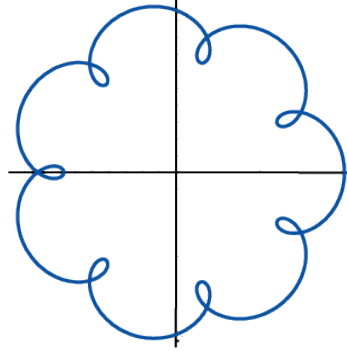


Figure 2.4: Radial projection of ion motion inside a Penning trap with a frequency ratio of $\omega_+/\omega_- = 8$.

the trap is shown in Figure 2.4.

The angular frequency inside an ideal Penning trap is given by $\omega_z = \sqrt{\frac{2qV_0}{md^2}}$, where V_0 is the depth of the attractive electrostatic potential at the centre of the trap, and d is the distance from the inner radius of the radial electrode of an ideal Penning trap to the minimum of the axial hyperbolic electrode. For example, within a Penning trap consisting of a 4 T magnet, a singly-charged ^{85}Rb ion will have a cyclotron frequency, ω_c , of about 4.545 MHz inside the potential well. On the other hand, given an energy of 1 eV above the floor of the trap and $d = 2$ cm, the axial frequency, ω_z , for ^{85}Rb will be only about 75 kHz.

When a Penning trap mass measurement is performed, it is always bracketed by at least two measurements of an isotope with a well-established mass. This is essential to correct for drifts in the magnetic field over time. An appropriate linear interpolation can then be made for the time at which the measurement was taken.

Several different methods are employed to perform Penning trap mass measurements at high-precision, in order to account for the needs of different experiments. These include Fourier-Transform Ion Cyclotron Resonance (FT-ICR), Time-of-Flight Ion Cyclotron Resonance (TOF-ICR), and Phase Imaging Ion Cyclotron Resonance (PI-ICR).

FT-ICR

Many of the highest precision measurements of the masses of stable isotopes are done with the Fourier-Transform Ion Cyclotron Resonance method. The frequency is measured by resonantly coupling the trapped ions to an external tank circuit. This technique can non-destructively monitor ions for as long as they can be trapped, and the precision improves with the duration of the measurement. However, the time needed to collect the ion signal in order to produce a high-precision mass measurement of a single trapped ion makes FT-ICR unsuited to studies of extremely short-lived species.

TOF-ICR

One method to rapidly perform mass measurements using a Penning trap is the Time-Of-Flight Ion-Cyclotron-Resonance (TOF-ICR) technique. This technique takes advantage of the fact that the ion motion can be converted between the ω_+ and ω_- eigenmodes by applying a quadrupolar radio-frequency excitation to the “guard” electrodes of TITAN’s Measurement PENning Trap (MPET), which also provide an additional static voltage correction in order to optimize the harmonic character of the trap.

By precisely tuning the ion injection trajectory using “Lorentz steerers”, ions are injected in the ω_- (magnetron) mode as purely as possible [42]. This makes it possible to determine the cyclotron frequency of a trapped ion by scanning over a range of excitation frequencies and searching for a resonance in the kinetic energy of the ion. When the ion is excited at the resonant frequency, the trapped ion is most effectively converted from the magnetron mode into the cyclotron mode, and therefore experiences the greatest increase in kinetic energy.

The degree of conversion from the magnetron motion into cyclotron motion is observed as a time-of-flight to a detector. The enhanced rotational velocity associated with the cyclotron mode is converted into a greater axial velocity of the ion upon extraction from the trap. As the ion passes through the strong magnetic field gradient, the Lorentz force begins to transition from radially confining the ion to accelerating it

out of the trap. This corresponds to the transition from the parallel field lines within the trap to the diverging dipole field lines outside of the solenoid. As the ion interacts with the transverse component of the magnetic field, energy is transferred from the azimuthal to the axial direction. Therefore, any radial energy imparted to the ion within the trap corresponds to an increase in axial kinetic energy upon exiting the trap.

The resonance can then be observed by scanning over a range of excitation frequencies and the reduced cyclotron frequency is indicated by a dip in the time-of-flight (TOF) of the ion (see figure 2.5). The TOF is measured between ejection of the ion from the trap and it striking a microchannel plate (MCP) detector. The MCP amplifies the electron signal produced when the ion impacts its surface by accelerating them through thin channels within glass plates. The TOF produces a side-band pattern due to the ion being excited for a limited duration within the trap by a monochromatic sine wave. This results in additional Fourier components appearing.

By applying a pulse of frequency ω_{rf} near the true cyclotron frequency ω_c , the resonant increase in the radial kinetic energy E_r is obtained according to the analytic expression

$$E_r \propto \frac{\sin^2(\omega_B T_{rf})}{\omega_B^2}. \quad (2.3)$$

Here, ω_B is the cyclotron frequency, and T_{rf} is the RF excitation time [43]. If T_{rf} is increased, the resonance is compressed horizontally to produce a sharper resonance peak. The characteristic side-band pattern can be seen in Figure 2.5 at various values of ω_{rf} surrounding ω_B .

The time of flight as a function of the excitation frequency ω_{rf} is given by the following expression:

$$T(\omega_{rf}) = \int_{z_0}^{z_1} \left\{ \frac{m}{2[E_0 - q \cdot E(z) - \mu(\omega_{rf}) \cdot B(z)]} \right\}^{\frac{1}{2}} dz, \quad (2.4)$$

where E_0 is the total initial energy of the ion, $E(z)$ and $B(z)$ are the electric and magnetic field along the extracted ion's path, the ion begins at z_0 and ends at the detector position, z_1 , with a maximum magnetic moment μ when ω_{rf} is the resonant cyclotron frequency [43].

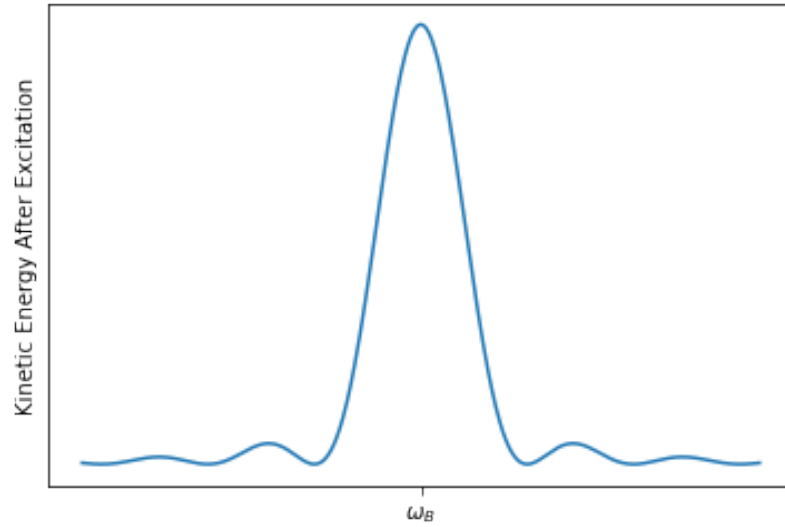


Figure 2.5: After undergoing a quadrupolar excitation, the final kinetic energy of an ion in a Penning trap depends on the deviation of the applied RF frequency from the reduced cyclotron frequency.

Only one to a few ions are trapped at a time in order to avoid ion-ion interactions. However, to perform TOF-ICR the ions under study must be collected at several different excitation frequencies (41 in TITAN) in order to produce a resonance curve for which the time-of-flight data can be fit with the analytic function. The mass is then determined from the frequency at which the peak occurs.

A variation of TOF-ICR known as *Ramsey excitation* excites the ion with two time-separated pulses instead of a single pulse. In principle, the larger separation in time allows for a more precise determination of the cyclotron frequency for the same total excitation time.

PI-ICR

A relatively new technique in Penning trap mass spectrometry is the use of the Phase Imaging Ion Cyclotron Resonance technique (PI-ICR) [44]. In principle, PI-ICR allows for fast, mass measurements with as few as a single ion, at even higher precision than TOF-ICR. While this technique is not currently in use at TITAN,

plans are in place to employ it in the future. Unpublished simulations performed by R. Steinbrugge and E.M. Lykiardopoulou have indicated that the angular width of the ion bunch $\Delta\phi$ increases with the radial energy spread as $\sqrt{\Delta E_{\text{radial}} \frac{m}{q}}$, which impacts the ability of PI-ICR to precisely resolve the location of the ion spot.

Penning Trap Precision

It is well understood that an isotope's half-life fundamentally constrains the achievable resolving power within a Penning trap [45]. This is immediately apparent when the precision is expressed as in [46] by the approximation,

$$\frac{\delta m}{m} \approx \frac{1.6\pi m}{qBT_{\text{RF}}\sqrt{N_{\text{ion}}}}. \quad (2.5)$$

The longer the duration of the radio-frequency excitation T_{RF} is, the higher the achievable precision will be. Additionally, precision is limited by the strength of the magnetic field of the trap, B , the charge of the ions, q , and the number of ions for which the time-of-flight is observed, N_{ion} .

Furthermore, Equation 2.5 assumes that the trap only contains a single species with no variation in the kinetic energy. If other species are present, the ion of interest will be observed on top of a background signal. A distribution of ion energies will introduce variations into the time-of-flight signal.

2.2.2 Brief Overview of Multiple Reflection Time-of-Flight (MRTOF) Mass Spectrometry

Time of Flight Mass Spectrometry

The most conceptually straight-forward method of determining an ion's mass is through its time-of-flight (TOF). Ions of different mass, but the same kinetic energy, will travel with different velocities, so the ion's mass can be determined by recording the time interval between two points on an ion's flight path. The ion velocity can then be used to determine the mass via the non-relativistic relation: $velocity = \frac{\sqrt{2E_k}}{m}$.

However, variability in the kinetic energy of ions can also affect their flight time. In order to achieve higher resolving power and precision, the *reflectron* type of mass

spectrometer was developed to correct for the energy spread of within the bunches of ions that are injected into a drift region for a TOF analysis. These devices accomplish this by using a single region of electrostatic reflecting potential to produce a linear electric field gradient in order to achieve a first-order correction to the time-of-flight. As illustrated in Figure 2.6, higher energy ions penetrate farther into the mirror, and when the appropriate potentials are applied, this allows their path length to be increased to compensate for their shorter time-of-flight. This increases the resolving power by narrowing the width of the TOF peak of a mass species resulting from their energy spread. Furthermore, the reflection approximately doubles the flight path for a given length of beam-line, thus increasing the separation of ions within a limited physical space. If only one reflecting potential is used, the ions are decelerated by a linear electric field gradient. Additional electrodes can be added to provide higher-order corrections to the dependence of TOF on the energy.

The MRTOF Technique

Multiple Reflection Time Of Flight Mass Spectrometry (MRTOF-MS) is a relatively new technique used to enhance the precision relative to more conventional time-of-flight mass spectrometry. This technique can obtain high-resolution mass spectra by significantly increasing the path length of the ions, solely by means of appropriately shaped electric fields. This is accomplished by allowing the ions to execute an arbitrary number of reflections between two electrostatic mirrors within a mass analyzer. The mechanism is similar to that of a reflectron type mass spectrometer [47], but MRTOF-MS allows the researcher to adjust the resolution of the measurement by specifying the number of reflections that an ion bunch executes. In addition to its usefulness for measuring the masses and relative abundances of ions, this technique can also be used as a purification technique for other traps, being able to suppress contamination by up to ~ 6 orders of magnitude [48]. Moreover, since the bandwidth sensitivity of the MRTOF is adjustable, several different mass peaks can often be measured simultaneously. This aids the identification of isobars, molecules, and even nuclear isomers within a mass unit, or can simultaneously measure dif-

ferent mass units at lower resolution. These broadband measurement capabilities make MRTOF-MS an excellent complement to PTMS, especially for masses far from nuclear stability.

While MRTOF-MS is only beginning to approach the precision of the well-established standard of Penning trap measurements, this technique is in a stage of rapid development. It is also extremely fast and possesses several other important strengths. For instance, the time-of-flight within an MRTOF-MS can be rapidly adjusted, to quickly adjust the resolving power of the spectrometer as needed. The flexibility to switch between measuring the relative masses of closely situated isobars, and to compare the relative abundances of beam constituents over a range of many mass units, make the MRTOF-MS an invaluable tool for the characterization of exotic beams.

As with a Penning trap, MRTOF-MS is performed on low-energy ion bunches. Unlike PTMS, which requires the measurement of multiple flight times in order to determine the resonant frequency, the time-of-flight of as few as a single ion can, in principle, be sufficient to measure a mass. This would require the rate of background ion counts to be low enough to identify a signal.

In contrast to the precision of PTMS given by equation 2.5, the precision of measurements using MRTOF-MS increases in direct proportion to the length of the flight path of the ions during separation. Therefore, in an MRTOF spectrometer of a given length, the precision can be expected to scale with the total number of reflections.

$$\frac{\Delta m}{m} \propto L_{total} = N \times \text{length of a single lap.} \quad (2.6)$$

However, as the number of reflections is increased, eventually collisions with residual gas in the analyzer, the initial energy spread of the ions, and fluctuations in the high-voltage power supplies will curb further improvements to the precision.

For an ion accelerated from rest, the kinetic energy in the analyzer is equal to the

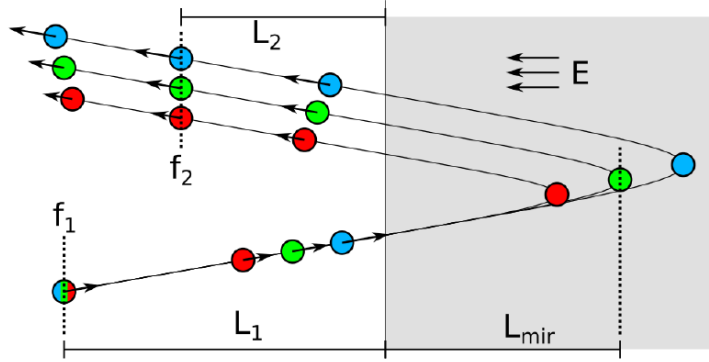


Figure 2.6: An illustration showing how reflection from an electrostatic mirror can create a new time-focus. Identical ions that begin at f_1 with different kinetic energy execute different paths through the electrostatic mirror in order to reach a new time-focus at f_2 . Diagram from [68].

product of the charge and the accelerating potential:

$$\begin{aligned} qV &= E_{kin} \\ &= \frac{1}{2}mv^2 \\ &= \frac{1}{2}m \left(\frac{d}{t} \right)^2, \end{aligned}$$

which leads to an expression that relates the mass-to-charge ratio to the time-of-flight

$$\frac{m}{q} = \frac{c(t_{exp} - t_0)^2}{(1 + N_{it}b)^2}. \quad (2.7)$$

Here, t_{exp} is the experimentally measured time-of-flight, and N_{it} is the number of isochronous turns (back and forth reflections) that take place after the two *time-focus shift* reflections. Since each reflection changes the time-focus (see Figure 2.6), measurements in this thesis use the two time-focus shift reflections to create the focus at the final detector, and subsequent turns preserve this focus. The constants t_0 , and c are determined by measuring two known species within the MRTOF prior to an experiment using so called *zero turn* measurement cycles (in which only the

PTMS	MRTOF-MS
Precision $\Delta m/m = 10^{-7} - 10^{-9}$ [51]	$< 5 \cdot 10^{-6}$ to $\sim 10^{-7}$ relative precision [52]
Extremely high precision measurements	Broadband measurements measure several species simultaneously
Reasonably “pure” beam is needed	Can handle a large “dynamic range” spanning ~ 4 orders of magnitude
Mass window is fixed by the magnet	Resolving power can be reduced to measure a broader range of masses

Table 2.1: A summary comparing some of the relative strengths and limitations of the Penning trap and MRTOF mass spectrometry techniques at TITAN.

two time-focus shift reflections are executed). t_0 is the amount of time it takes for the ions to begin their flight after the start signal is given, and accounts for electronic delay, and c results from the time of flight of the time-focus shift.

$$b = l_{it}/l_{tfs},$$

and

$$c = 2U_{eff}/l_{tfs}^2,$$

where l_{it} and l_{tfs} are the path lengths of one isochronous round-trip and the time-focus shift round-trip, respectively, and U_{eff} is the effective voltage over the complete flight path. Measurements involving many reflections are especially sensitive to the b parameter, and is therefore calculated throughout a measurement from an arbitrarily selected *time calibrant* species in the multiple-reflection TOF spectrum in order to account for any drifts in the time-of-flight.

Additional details of the theory of the MRTOF technique can be found in reference [50]. Table 2.1 summarizes the relative strengths of MRTOF and Penning trap mass spectrometry.

MRTOF Resolving Power

The resolving power within an MRTOF-MS is limited by

$$R_m = \frac{t_0 + Nt_{turn}}{2\sqrt{\Delta t_0^2 + N^2\Delta t_{turn}^2}}. \quad (2.8)$$

As the number of reflections approaches infinity, the ultimate resolving power for an MRTOF-MS is then given by

$$R = \frac{t_{turn}}{2\Delta t_{turn}}. \quad (2.9)$$

Here, t_{turn} is the time one “turn” takes, and Δt_{turn} is the increase in peak width due to chromatic aberrations (i.e. energy-dependent focusing effects) of one round-trip or turn in the analyzer [49][50]. This makes the quality of the electrostatic optics of great importance.

Since there is a random distribution in the ion TOF, the precision of a TOF peak is also statistically limited by the number of ion flight times acquired. These in turn, will depend of the rate of the incoming ion beam (see section 2.4.2). For a gaussian distribution in TOF (i.e. peak shape), the standard deviation is given by $\sigma/\sqrt{N_{ions}}$.

2.3 Mass Spectrometry Conclusion

The nuclear physics goals of this thesis are accomplished by employing cutting edge mass spectrometry techniques at TRIUMF’s TITAN facility. MRTOF-MS provides a new, flexible method of measuring masses of short-lived radioactive species at relatively high precision, and Penning traps remain a standard in the field of precision measurements. The following chapter will discuss the implementation of these, and other relevant traps at TITAN, which were used to obtain the results of this work. This will lead to a motivation for a Cooler Penning Trap, which represents an endeavour to further improve high precision Penning trap measurements of highly charged ions at extremely small abundances.

2.4 TRIUMF’s Ion Trap for Atomic and Nuclear Science (TITAN)

In order to perform precision mass measurements of exotic species, the radioactive beams produced at ISAC can be transported to the TITAN facility shown in Figure

2.7³. At TITAN, several ion traps are available to prepare and study the radioactive beams, depending on the needs of the experiment. An outline of the various traps which enable these mass measurements will be provided below.

In 2003, it was proposed to develop a Penning trap facility capable of utilizing charge-bred ions to enhance the precision of short-lived isotope measurements [53]. Since then, TITAN has used the exotic beams from the ISAC facility to measure the masses of halo nuclei [54][55], probe for the emergence and quenching of shell closures far from stability [22], study isospin symmetry breaking [56], and aid in performing r-process nucleosynthesis calculations [57], among other areas of nuclear physics.

The Penning trap at TITAN has been used to measure isotopes as short-lived as 8.8 ms (^{11}Li) [55], and TITAN's MRTOF has recently measured the mass of an isomer with a half-life of only 5 ms ($^{125}\text{In}^{m2}$), though it has yet to be published [57].

TITAN is the first online Penning trap facility to charge breed short-lived isotopes in an Electron Beam Ion Trap (EBIT) prior to performing a mass measurement, in order to enhance the measurement precision. Online commissioning of the TITAN EBIT was performed in 2010 when radioactive beam from the ISAC facility was first charge-bred and extracted at TITAN [58], and the first Penning trap mass measurements were subsequently performed on these charge-bred radioisotopes in 2011 [59].

Recently, the mass measurement Penning trap has been upgraded to operate at cryogenic temperatures in order to prevent highly charged ions from losing their charge state through collisions with residual gas. Details of the upgrade can be found in appendix B of reference [60].

When performing high precision mass measurements of short-lived species, it is necessary to find a compromise between two qualities, speed and precision. When valuable beams of exotic isotopes are being delivered to the experiment, speed is clearly of the essence, however, varying degrees of precision are required for each measurement depending upon the scientific case under study. A Cooler Penning Trap is being developed, with the goal of improving mass measurements of Highly Charged Ions (HCIs). Depending on the scientific objective being addressed, this trap can, in

³TITAN is able to study a number of scientific observables besides nuclear masses, but these techniques are outside the scope of this thesis.

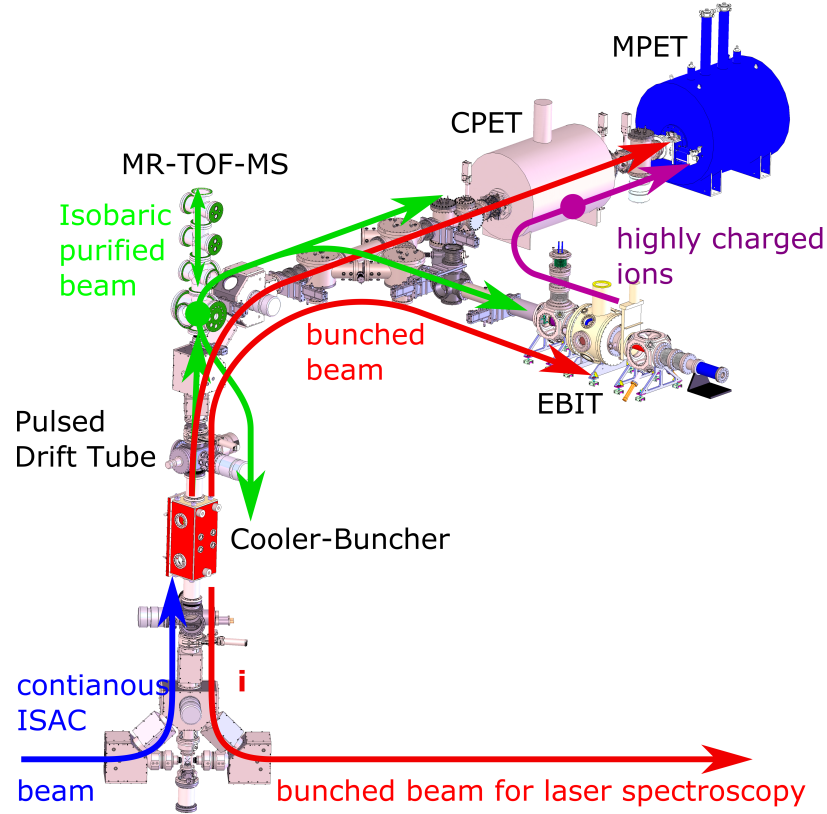


Figure 2.7: The TITAN Facility. Beam from the ISAC target is cooled and bunched in the RFQ, then transported to either the MRTOF, MPET, or EBIT. Although CPET commissioning tests took place offline, the planned location for CPET is also shown. From [71] and reproduced with permission from Springer Nature.

principle, enhance either the precision or speed in proportion to the charge state of the ion.

2.4.1 The TITAN RFQ Cooler Buncher

The ISAC facility delivers continuous beam to TITAN at an energy of ~ 20 keV. Before any precision measurement can be performed on the ions, it is therefore necessary to first prepare the beam by accumulating it into well-defined bunches with an appropriate kinetic energy before trapping in the MRTOF, EBIT or MPET.

The technique used to prepare ion bunches at TITAN is known as *buffer-gas*

cooling. Radioactive beams can be cooled and accumulated into bunches with reproducible properties by collecting them within a gas-filled Radio-Frequency Quadrupole (RFQ) trap (otherwise known as a linear Paul trap [39]) that contains a sufficient pressure of inert gas for the ions to quickly reach thermal equilibrium [61]. TITAN's RFQ is set at a floating voltage defined by the ion source in order to match it to the kinetic energy of the beam. The RFQ employs a linear geometry in which ions are radially confined using a quadrupolar AC field, and a DC potential is applied to each end of the trap to confine them axially. The continuous beam loses enough energy to the buffer gas as it traverses the trap such that ions become trapped behind the axial potential barrier over which they entered the trapping region 2.8. The ions continue to lose energy through collisions with the helium buffer gas, and as they cool they accumulate in a small region near the exit of the trap by applying an axial potential gradient. The cooling reduces the energy spread of the ions so the TITAN optics can accommodate the phase-space *emittance* of the ion bunch. The emittance is the area in position-momentum space occupied by the ion bunch, and is a conserved quantity in each spatial dimension as the ions pass through the electrostatic optics. Ions are subsequently released from the RFQ and delivered to the other traps in the system [62].

The TITAN RFQ also plays the important role of matching the kinetic energy of the ISAC beam to the TITAN system. The kinetic energy of the incoming beam is defined by the ISAC target, which floats at a potential of approximately 20 kV. Although the RFQ shares this floating voltage so the electrodes can match the beam energy, it is necessary to reduce the kinetic energy of the ion bunches produced by the RFQ so the rest of the TITAN beam-line can operate at ground potential, and the optical elements can operate at manageable voltages (within a few kV of ground). Upon exiting the RFQ, the kinetic energy of the newly-formed ion bunch is matched to the relevant trap in TITAN by quickly ($\sim 1\mu s$) pulsing the electric field inside a drift-tube that immediately follows the RFQ.

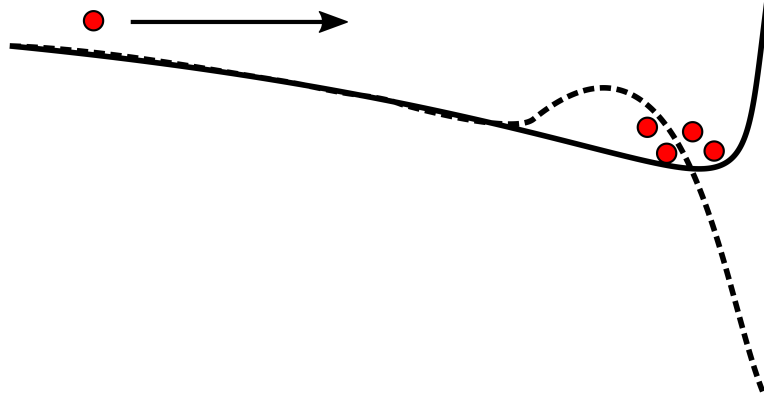


Figure 2.8: Schematic showing the basic operation of a radiofrequency quadrupole cooler-buncher (RFQ) trap, with the vertical direction representing electrostatic potential and the horizontal direction position along the beam axis. Ions enter from the left and collisions with the helium buffer gas allow ions to settle into the potential minimum (solid line). Next, the ion bunch is ejected from the trap by switching the electrode potentials to produce an extraction gradient.

2.4.2 The TITAN MRTOF

An MRTOF mass spectrometer was installed in the TITAN facility in 2017, and published its first scientific result with radioactive beam in 2018 [63]. It is one of a handful of MRTOF systems with access to short-lived radioisotopes, which includes the MRTOF system(s) at RIKEN [64][65], ISOLTRAP at CERN [66], Argonne National Lab [67], and the FRS ion catcher at GSI [50].

The MRTOF system at TITAN [68][69][70][49][52] was designed and constructed at the Justus-Liebig-University in Gießen, Germany [71][72]. Following commissioning, it has been demonstrated to achieve a mass resolving power up to ~ 400000 FWHM and a mass precision/accuracy of approximately $\frac{\delta m}{m} < 10^{-7}$ [48]. Since the number of ion reflections within the MRTOF can be varied, it allows the operator to zoom in and out in resolution, either to achieve a higher precision measurement or to observe a broader range of mass peaks as the situation demands for radioactive beam measurements.

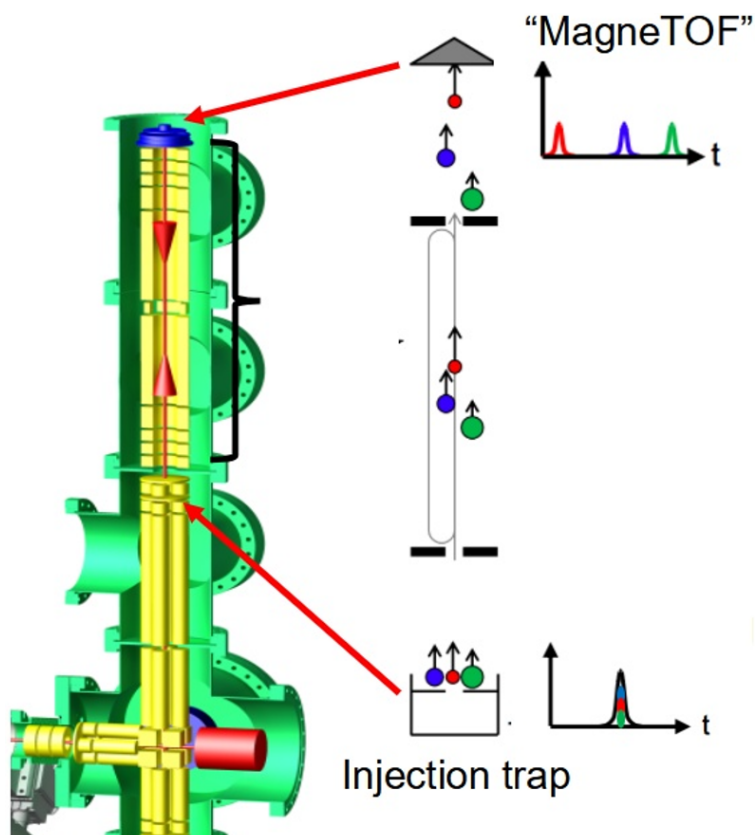


Figure 2.9: TITAN’s MRTOF mass separator. Within the MRTOF, ions are prepared inside a gas-filled injection trap before executing an arbitrary number of reflections between two electrostatic mirrors. The mirrors are tuned for isochronisity (i.e. the ion time-of-flight is not dependent on small changes in the ion energy). From [71] and reproduced with permission from Springer Nature.

Performing time-of-flight mass separation within the MRTOF at TITAN is a conceptually straightforward process that involves several distinct steps: ion bunching, time of flight separation, and detection. First, an ion bunch is prepared by collecting either the bunch of ions delivered from the TITAN RFQ, or continuous ions from the MRTOF ion source within a linear RFQ trapping section in the MRTOF. After the ions have cooled and bunched by transferring their kinetic energy to the buffer gas, the bunch is transferred to a narrow, gas-filled injection trap. From the injection trap, the bunch is released into the MRTOF analyzer section for time-of-flight separation

and the E1 electrostatic mirror is closed behind it. The bunch will then undergo an appropriate number of reflections between the two ion mirrors to achieve the desired resolving power until the E2 mirror is opened to allow the ions to be detected on the final detector. The MRTOF records the time of flight of the ions between exiting the injection trap and their detection on the final (MagneTOF®) [73][74] detector. The concept is shown with a schematic of TITAN's MRTOF in Figure 2.9.

The confining potential of the injection trap must be opened quickly as to avoid imparting more kinetic energy to heavier ions than to the lighter ions which accelerate out of the trap more quickly. The bunch of ions is “kicked” out of the injection trap by applying an electric field gradient that imparts slightly more energy to the ions that are more distant from the exit of the trap upon extraction.

A small variation in energy does not affect the measurement because the potentials of the the mirrors are carefully tuned to achieve isochronous reflections with respect to variations in energy. After leaving the injection trap, the ions rapidly come to a time-of-flight focus, and soon the ions with higher kinetic energy outpace the lower energy ions as they proceed on their flight through the analyzer. The first two reflections recover the time-energy focus such that it lies beyond the analyzer region at a position that coincides with the final detector. With each subsequent reflection, the time-of-flight focus is extended by exactly one path length across the analyzer region such that the focus remains on the detector for any number of reflections. Since each reflection within the analyzer resets the TOF focus to lie outside of the analyzer region, the possibility of ion-ion interactions impacting the flight time is minimized. The multiple electrodes in the mirrors are tuned to achieve a third order independence of the time-of-flight with respect to kinetic energy for any number of reflections (i.e. they are isochronous reflections). This energy independence in the ion time of flight allows the MRTOF to accept a wider range of ion energies without sacrificing precision or efficiency.

Isochronous reflections⁴ are achieved by carefully tuning the potentials of the mirrors to establish a kinetic energy dependent penetration of the ions into the elec-

⁴A complete cycle that includes two reflections is sometimes referred to as an *isochronous turn*

trostatic mirrors. This allows the TOF to be energy independent over a range of about 20 eV [75]. The ions can then be reflected back and forth within the analyzer any number of times, and will only again reach a time-focus after the second mirror is opened and they arrive at the detector. In this way, the energy spread allows ions of the same mass to follow slightly different trajectories. Systematic effects due to ion-ion interactions are thereby minimized by preventing the ions from reaching a time-energy focus more than once regardless of the number of reflections.

As the ions traverse the analyzer, they separate by mass. The electrostatic mirrors require two different voltages in order to reflect and transmit ions as well as a third set of voltages for the first two reflections which recover the time-focus (the time-focus shift). After ions execute a sufficient number of reflections to separate the mass peaks from one another, they are released from the analyzer and their time-of-flight is recorded when they strike the surface of a detector. Currently a MagneTOF[®] detector [73][74] is used to measure the ion flight times with high time resolution by directing each stage of the amplifying electron cascade along energy independent (isochronous) paths, by utilizing a combination of magnetic fields and accelerating voltages. The TOF can also be measured using an MCP detector.

The mass analyzer is composed of a drift tube which is held at ground potential, and two electrostatic ion “mirrors”, each composed of four cylindrical electrodes on either end of the analyzer to reflect the ions back and forth within the MRTOF. A single round-trip of an ion through the analyzer takes about 23 μ s [75].

Precision within the MRTOF has recently been enhanced by increasing the flight time of the ions. The number of reflections that ions can undergo is limited by the repetition rate of the MRTOF, which for the studies in this thesis was run at 50 Hz, and the total time over which the spectrum is acquired. It has now been established that the MRTOF can operate at 30 Hz in order to increase the resolving power while extending the measurement time.

The outer mirror electrodes are the most sensitive to fluctuations in voltage, and are therefore given the highest priority for stabilization (better than ≈ 1 ppm).

The series of isochronous reflections is terminated by lowering the reflecting potential on the second mirror so the ion time of flight can be measured on the final

detector. Alternatively, the ions can be retrapped in the injection trap to either undergo a second set of reflections in the analyzer, or to be reverse extracted for transmission to the other traps of TITAN.

The timings for switching the mirrors and other electrostatic elements during cycles of the MRTOF are controlled using an FPGA. Data acquisition of the time-of-flight of ions to the MagneTOF[®] detector is performed using a time to digital converter (TDC). The acquired time-of-flight data was recorded using the Massdata Acquisition (MAc) software developed by the MRTOF group at the University of Gießen [76][77].

The MRTOF offers several strengths at an ISOL facility. It can provide real-time feedback of adjustments to the ISAC mass separator magnet in order to optimize the yield of the desired species within a mass spectrum. The ability to adjust the number of reflections of ions within the analyzer allows for measurements with variable resolving power, and the ability to identify non-isobaric contaminants. And the simultaneous measurement of several species allows for cross-checking of mass calibrants.

Limitations on the beam rate

In order to perform precision mass measurements on exotic isotopes, both high precision and high sensitivity are required. Precision refers to the narrowness with which a mass peak can be resolved, while sensitivity refers to our ability to identify the signature of the ion being measured against some background that competes with it. The TITAN MRTOF has been shown to be sensitive to yields below one particle per second [31].

However, the sensitivity of ion traps is reduced when background species begin to dominate the spectrum. The sensitivity can be enhanced within a Penning trap by first preparing the sample by removing the contaminating mass species. This is often accomplished by applying a dipolar excitation at the resonant frequency of the contaminant species to drive the ions to a large magnetron radius [78]. Similarly, the sensitivity of an MRTOF mass spectrometer is limited by the relative spectral

abundances between the species with the highest rate and that with the lowest rate. The range of ion intensities which can be simultaneously measured is known as the *dynamic range* and is given by the total beam rate divided by the rate of the least abundant resolvable species.

The background in an MRTOF time-of-flight spectrum is dominated by the most abundant ion species within it. The lowest rate of ions that MRTOF can measure in a single mass peak is limited by the background counts on the detector from ions that scatter within the analyzer. The spectrum produced by a single round of multiple reflection mass separation within the analyzer of TITAN's MRTOF can span a range of roughly four orders of magnitude in relative peak intensities, providing the peaks do not overlap.

Furthermore, the intensity of the ion beam entering the MRTOF is adjusted so that no more than one to a few ions traverse the analyzer at a time. This is done in order to avoid systematic shifts in the time of flight due to electrostatic interactions between ions with the same mass-to-charge ratio (i.e. ion-ion interactions), especially for ions that have a very similar time of flight. The background ion intensity therefore must be controlled to maximize the sensitivity of the MRTOF.

The *time-to-digital converter* (TDC) then acquires and records all the TOF events (relative to the opening of the second mirror) over multiple cycles and they are recorded together as a single *spectrum*. For these experiments the MRTOF acquired data at 50 Hz, and 50 cycles were collected and merged to generate each spectrum. Therefore each spectrum acquired in the MAC software represents a one second block of time.

Mass-Selective Retrapping to Suppress Contaminants

The Tm and Yb measurements in 2018 could perform mass measurements down to the lowest beam rates ever measured at TITAN, largely due to a greatly enhanced suppression of contaminants. Under normal operation, the dynamic range of TITAN's MRTOF can accommodate excesses of approximately $\sim 10^4$ additional ions in the dominant peak relative to the lower abundance peak before the background

of scattered ions from the more abundant species begins to prevent the detection of the minor one. This ratio refers to the relative abundances extracted from MRTOF's injection Paul trap. During these experiments, the dynamic range was enhanced by implementing the *mass-selective retrapping* technique [79] for the first time in a radioactive beam experiment.

The retrapping technique reduces the intensity of isobaric background by trapping the ion bunch for a second time inside the gas-filled preparation trap, after it has undergone a series of passes through the analyzer (see figure 2.10). Within the trap, they once again undergo buffer gas cooling which essentially resets the properties of the ion bunch. Therefore, any heating or space-charge effects that could have caused systematic shifts in the TOF peaks during the initial pass through the analyzer will not impact the subsequent measurement cycle. Conveniently, all that matters is the population of the ion bunch that executes the final flight through the analyzer before being deposited on the MagneTOF®.

This increases the sensitivity of the MRTOF by suppressing the noise floor of the spectrum, thereby increasing the dynamic range. Furthermore, it allows for an enhancement in the speed with which statistics can be acquired when performing a precision mass measurement by increasing the maximum beam rate that the MRTOF can accept.

For several of the MRTOF mass measurements presented in this thesis, mass-selective retrapping allowed for the suppression of contaminant isobars by an additional 4 orders of magnitude. This allowed for the successful measurement of nuclear species that would otherwise be indistinguishable from the background signal.

There are two strategies that can be applied to increase the degree of suppression of unwanted ions (i.e. the mass selectivity) when employing retrapping:

1. Increase the number of reflections (i.e. lengthen the flight path) in order to enhance the time-of-flight separation
2. Reduce the depth of the potential well used to retrap ions

The second method is useful for “on-the-fly” adjustments of the selectivity. Applying a shallower retrapping potential well within the injection trap can be used

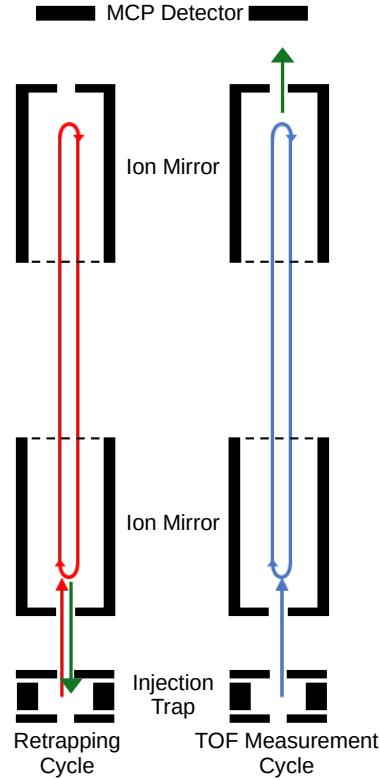


Figure 2.10: A re-trapping cycle in the analyzer is indicated by a green arrow to conclude with recapture in the injection trap (left), while a measurement cycle is shown to finish on the time-of-flight detector (right).

to retrap a narrower mass-to-charge (A/q) ratio, and a deeper well to trap a wider A/q . However, if the trap becomes too shallow the efficiency of recapturing the ions will begin to be significantly affected, thus creating a limit on the maximum possible suppression of unwanted A/q values.

The retrapping window has a width of ~ 40 ns. However, the time-selectivity of retrapping relevant to resolving two close-lying species is often limited by the time-scale of the rising (or falling) edge of the retrapping window, which has a width of ~ 15 ns. This is the result of the speed with which the electrode voltages can be

switched to close the injection trap. The resulting time-of-flight spectrum of the retrapped ions is then essentially a convolution of the initial TOF spectrum with this narrow, but smooth-edged time window.

In the future, TITAN will employ the retrapping technique to reverse extract ions from the MRTOF so it can suppress large quantities of undesirable contaminant ions before directing them to the MPET or EBIT to perform a mass measurement on singly or highly-charged ions.

Mass Range Selector

Since the ions undergo many reflections in the analyzer, non-isobaric contaminants of different A/q can execute a different number of reflections while undergoing a similar, or equal time-of-flight to the detector. After executing a different number of reflections, a faster species eventually catches up with the other (a lighter species will execute more reflections, a heavier one fewer). If the number of reflections is not explicitly known for a peak, the contaminant may imitate a species with the same A/q . As such, it will pollute the spectrum of the species of interest. These peaks will change position or vanish as the number of reflections is changed. Unfortunately, this can easily result in misidentifying or contaminating the peaks within a spectrum.

One solution to this problem is to record a mass spectrum for several different numbers of reflections. However, the TITAN MRTOF effectively addresses the challenge of non-isobaric ions using the *Mass Range Selector*, or MRS. The effect of the MRS is similar to that of a Bradbury-Nielsen gate [80] in that it provides some initial time-of-flight mass-selectivity prior to performing the measurement. Due to the slightly different velocity in the analyzer of the non-isobaric contaminants, eventually they drift out of phase with the mass unit under study. For several cycles, an asymmetrical (steering) potential is then briefly applied at the centre of the analyzer while the mass unit under study is reflecting within the distant electrostatic mirrors. At this point, the non-isobaric species will experience an asymmetrical deflecting field from two electrodes in the centre of the trap. This electric field destabilizes the trajectory of the ions, causing them to be deposited onto the an aperture near the centre

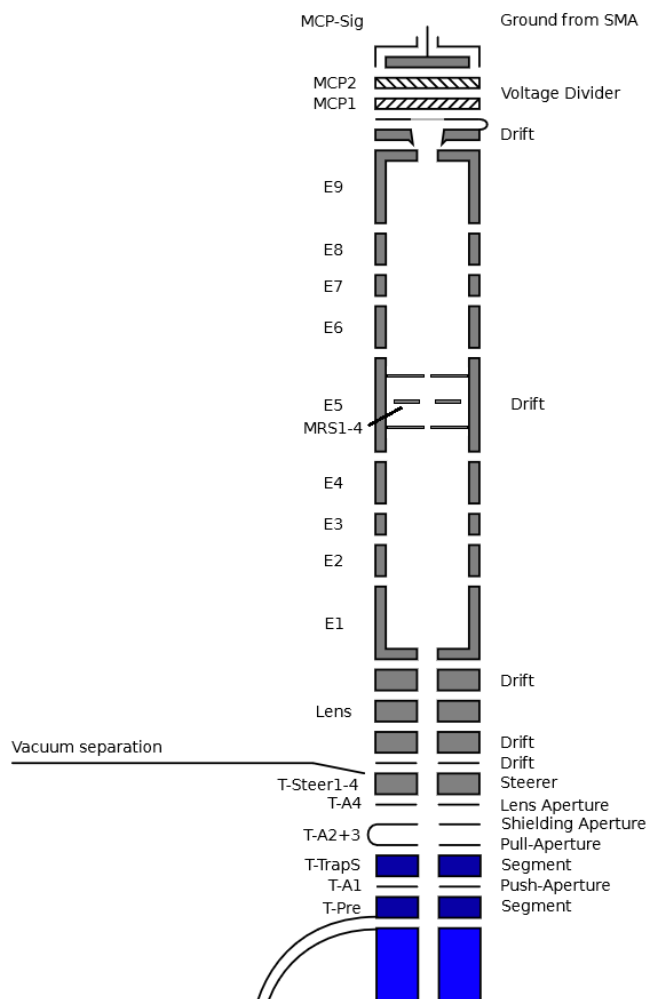


Figure 2.11: Analyzer showing the Mass Range Selector (MRS) used to remove ions from the trap which undergo a different number of reflections. Image is not to scale.

of the trap, or elsewhere. The MRS is shown in context with the other electrodes of the MRTOF analyzer in Figure 2.10.

Generally, the MRS was activated whenever ions of different mass-to-charge ratios might be present, in order to prevent ions that execute a different number of reflections from arriving at the detector after the same flight time as the ions of interest. Data with the MRS on was analyzed separately from the data with the MRS off as a precaution in case a net change in the drift potential caused a systematic shift in the time-of-flight.

Systematic uncertainties in the MRTOF

There are many potential sources of systematic uncertainties within the MRTOF. These include voltage drifts due to temperature changes, electrical noise, etc. The power supply ripple on the mirror electrodes E1 and E9 (see figure 2.11) is known to be the most critical, since this is the region where the ions are moving the most slowly.

A straightforward procedure is applied in order to account for time-dependent drifts. The multiple round-trip scaling constant b in Equation 2.7 is calculated from the peak of a known isobaric species in the mass spectrum. By continually monitoring changes to b for consecutive blocks of data collected throughout a measurement, a time-dependent correction can be applied to account for slow drifts in the electrostatic mirror power supplies that would otherwise broaden the peak. This is known as a *time-resolved calibration* (TRC). The duration of these blocks must strike a balance between correcting the drifts on a short timescale and acquiring a sufficient number of counts of the known isobaric species (which in turn, depends on its rate).

2.4.3 The TITAN MPET

Ions can be directed from the RFQ, EBIT, or MRTOF to the Measurement Penning Trap (MPET) for a high precision Penning trap measurement. In MPET ions are trapped within a harmonic axial potential with as little kinetic energy as possible while still accommodating the energy spread of the average ion “bunch”. Ions can be separated by time-of-flight using a Bradbury-Nielsen gate as an ion “shutter” [80]. They are then pulsed to a potential close to ground inside a drift tube, in order to precisely match the potential minimum within MPET as shown in Figure 2.12.

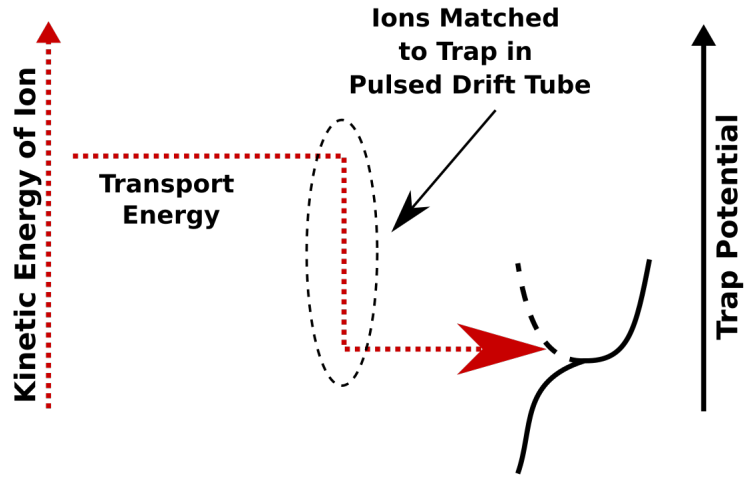


Figure 2.12: A pulsed drift tube adjusts the ion beam energy from a transport energy of about 2 keV to match the potential of MPET.

The beam rate of incoming ions is adjusted so that the typical number of ions entering the trap in a single cycle is only about one to a few. This is done to prevent the electric field of other ions from changing the effective potential inside the trap, thereby shifting the cyclotron frequency and corresponding mass value. A “count-class” analysis can be done to check for any systematic dependence of the determined mass value on the number of ions inside the trap per cycle; if the number of trapped ions has an impact on the cyclotron frequency, these cycles can either be corrected for or rejected from the analysis all together. The MCP efficiency must be taken into account when considering the true average number of ions inside the trap.

Since it is desirable to trap only one ion at a time in order to avoid ion-ion interactions in MPET, the dynamic range can be a significant limitation for mass spectroscopy measurements. For this reason MPET stands to gain significantly from having ions undergo isobaric purification in the MRTOF prior to measurement. Exotic isotopes that have been inaccessible due to significant contamination from the ISAC separator will be possible to deliver the MPET with a substantial suppression of isobars.

As an ion enters MPET, the Lorentz steerers apply an electric field transverse

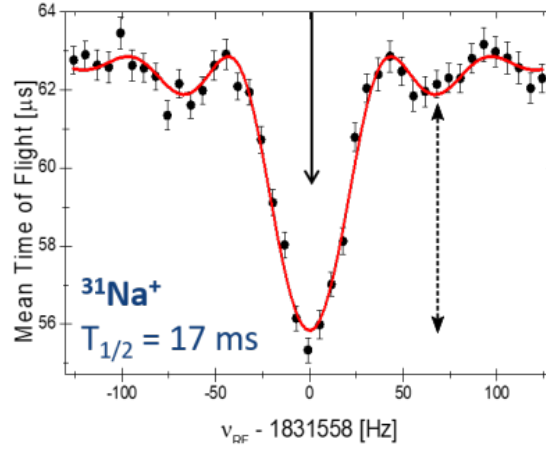


Figure 2.13: An example Time Of Flight Ion Cyclotron Resonance (TOF-ICR) spectrum showing the response in TOF to 41 different excitation frequencies. The dashed arrow indicates the *TOF effect*, which is the difference between the minimum TOF in the sidebands from the absolute minimum in TOF at the resonant frequency (indicated by the solid arrow).

the flight path of the ion in order to inject ions from the TITAN beam-line into the Measurement Penning Trap (MPET) as purely as possible in the magnetron mode. Then the TOF-ICR technique (see section 2.2.1) can be performed by applying a quadrupole excitation that converts the ions from the magnetron to the cyclotron mode, and the mass is determined by observing the resonant reduction in the time-of-flight. The quality of a resonance can be characterized by the *time-of-flight effect*, which is the difference between the peak height and the baseline noise in the sidebands (see Figure 2.13).

TITAN’s MPET can measure mass precisions on the order of $\frac{\delta m}{m} \approx 10^{-8}$ [81] and even resolve isomers separated by less than 90 keV [82], as well as nuclei with half-lives as short as less than 10 ms [55]. MPET presently uses the TOF-ICR technique discussed in section 2.2.1, although the PI-ICR method may be implemented in the future to enhance precision.

TITAN utilizes a Lorentz steerer to facilitate a fast injection cycle [42]. A steering element within the magnetic field allows for control of the radial position of the ion

upon entering the trap. By tuning these steering elements, an ion can be injected almost purely in the magnetron mode, which allows for a faster mass measurement cycle. The alternative method is to inject the ion on the central axis of the trap, increase the magnetron radius by applying a dipolar RF-field at the magnetron frequency, and subsequently apply the TOF-ICR method by scanning a quadrupolar excitation over a range of frequencies near the cyclotron frequency [78].

2.4.4 The TITAN EBIT

TITAN is unique among online (i.e. short-lived isotope beams) Penning trap facilities in that it can employ an Electron Beam Ion Trap (EBIT) to enhance the achievable precision in the Penning trap. An EBIT is a specialized trap for rapidly increasing the charge states of ions [83]. They can be used to strip additional electrons from the singly-charged radioactive ions delivered from ISAC, in order to increase the cyclotron frequency within MPET [84][85]. This is achieved by directing a high energy electron beam along the axis of the EBIT, such that the electron beam overlaps with the ions that are trapped in this region and removes even strongly bound electrons (100s or 1000s of eV) through the process of electron impact ionization. As shown in Figure 2.14, the intense electron beam within an EBIT is compressed to high density by the magnetic field of the trap and continually strips electrons from the ions present in the trap driving them to higher and higher charge states. Ultimately, their charge state reaches an equilibrium between ionization and electron recombination of the highly charged ions. Recombination occurs either by radiative recombination, in which a photon is emitted, or by rarer processes, such as the resonant process of *dielectronic recombination* [83] or by a three-body process [1]. The beam-line is then tuned to extract one of these high charge states produced in the EBIT to MPET.

Since the precision of a mass measurement scales with the mass-to-charge ratio, m/q , in principle, charge breeding allows TITAN to enhance the precision of mass spectrometry measurements in MPET by a factor of the charge state, q . To do this, the TITAN EBIT employs a 6 T, superconducting magnet in Helmholtz configuration, that compresses a 4 kV electron beam at 100-150 mA, to $\sim 200 \mu\text{m}$.

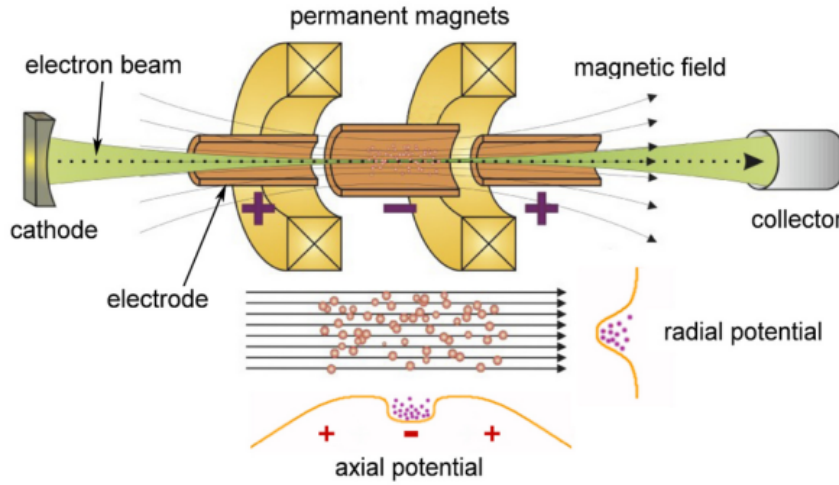


Figure 2.14: The magnetic field of an EBIT compresses the electron beam in order to create a deep potential well for ions using their space-charge. This image shows the SPARC EBIT and is from the webpage: http://web-docs.gsi.de/%7Estoe_exp/laboratory/environment/ebit/ebit.php. Image rights: GSI Helmholtzzentrum für Schwerionenforschung GmbH. While the operating principle is the same, the TITAN EBIT uses a superconducting Helmholtz coil instead of a permanent magnet configuration.

Ions are trapped within an EBIT using the same principle as a Penning trap. The combination of an axially-oriented magnetic field with an axial electrostatic potential well provides 3-dimensional confinement of positively charged ions. However, the depth of the potential well inside an EBIT is substantially enhanced by the additional space-charge potential of the electron beam, thereby increasing the space-charge capacity of the trap. This allows a much greater ionic charge to be stored inside the EBIT. The physics of EBITs is reviewed in reference [83].

Since MPET can only accept a single mass-to-charge ratio at a time, a fraction of the charge-bred ions will be lost to other charge states. However, a significant percentage of the ions can often be prepared with the same mass to charge ratio, m/q , by removing electrons until a closed electron shell is reached. This is known as “threshold charge breeding”.

Trapping ions within an EBIT heats the ions through collisions with the electron beam, and to a lesser extent, through ionization [86]. Electrons of several tens of keV impart additional kinetic energy to the ions through Coulomb collisions. Furthermore, if the ions have a non-zero energy spread, they will be able to probe regions of different potential energy within the trap. If an ion happens to have its ionization state increased while at a higher potential energy, it will receive an additional boost to its total energy in the trap. This is known as *ionization heating*. Eventually, a thermal equilibrium of the ions is approached as the energy is distributed between ions in the EBIT via the coulomb interaction.

Ion heating within the EBIT will eventually become balanced by a gradual loss of ions (along the trap axis) from the high energy tail of the Boltzmann distribution. This is an example of *evaporative cooling*, a process by which the hotter ions are released from the trap, while the cooler ones remain. Furthermore, this will preferentially release ions in lower charge states from the EBIT. Evaporative cooling represents the dominant cooling mechanism within the EBIT. Each charge state will separately reach thermodynamic equilibrium inside the EBIT due to a balance of heating and cooling processes, with the temperature increasing with the charge state [86].

Lastly, the ions can experience some additional heating when they are extracted from the EBIT. This is the result of the potential gradient which ejects them from the trap. Generally, the time-of-flight spread from EBIT to MPET is minimized by steepening the gradient that extracts the ion bunch from the EBIT. This is known as *kicking* and is represented schematically in Figure 2.15. Tuning the amount of kicking can optimize transmission to MPET by allowing them to arrive in a narrow bunch, however, increased kicking will also increase the energy spread of the ion bunch. This is fundamentally limited by the *emittance* of the ion bunch, the product of its position and momentum spreads.

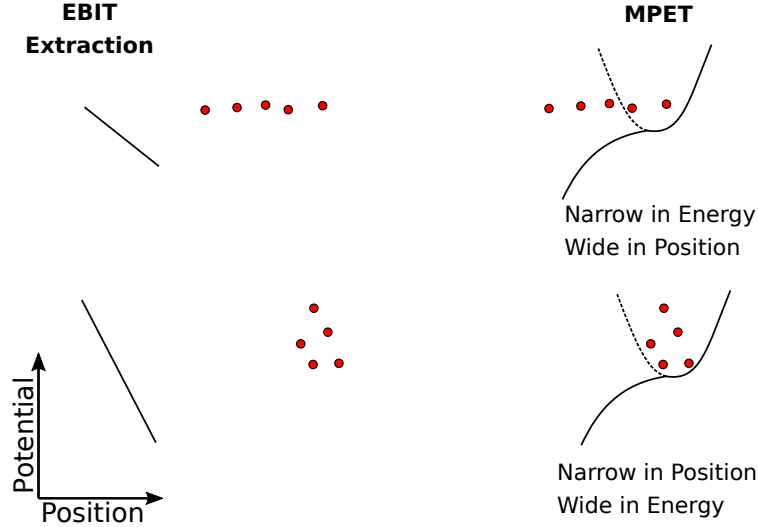


Figure 2.15: “Kicking” the ion bunches extracted from the EBIT can only achieve a compromise between a well-defined focus in position and a well-defined focus in energy. Two different potential gradients for extraction are shown on the left. The bottom gradient uses greater kicking, which imparts a larger energy spread to the extracted ions. Open and closed Penning trap potentials on the right show how ion transmission can be compromised due to a large spread in ion arrival time when kicking is too small for a given energy spread.

To date, many successful mass measurements of highly charged, short-lived ions have been performed at TITAN (e.g. [59],[87],[82],[88]).

2.4.5 Motivation for a Cooler Penning Trap

Aside from charge state distribution losses, the high energy of the electron beam used to charge breed the ions can reduce transmission by increasing the energy spread of the ion bunch. This causes an increase in the total emittance (i.e. the area in position-velocity phase space) and can reduce the transmission of ions from EBIT to MPET.

The concept of beam emittance is useful for quantifying the quality of ion bunches, because the total emittance is conserved throughout regions of beam-line in which the beam passes through constant potentials at vacuum. As the beam passes through

the various ion-optical elements, momentum is exchanged for displacement from the central focus and vice versa, in each of the three independent dimensions of travel. Figure 2.16 shown a phase space plot for many different ions passing through one point in a beam, with velocity vs position plotted in one dimension. The area of the ellipse determines the conserved quantity of the emittance in that dimension. However, interactions with other ions, gases, or time-dependent fields can alter the total beam emittance.

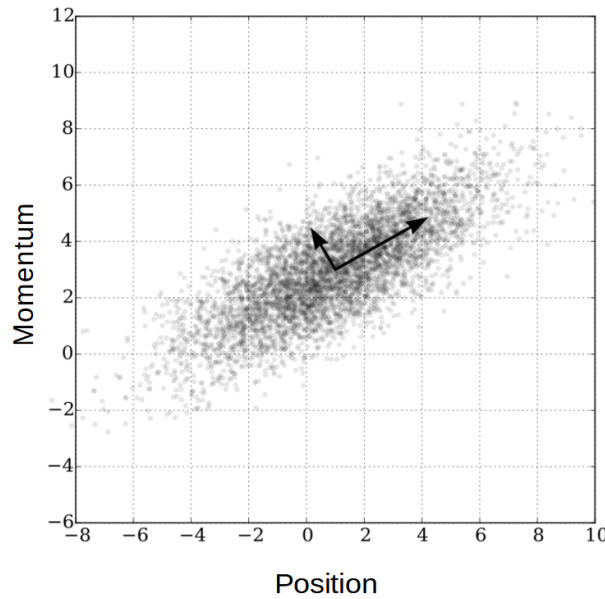


Figure 2.16: The emittance is determined by the area occupied by the beam in phase space (given here by momentum vs position in one dimension). Since the beam is statistically distributed in phase space, the area encompassing 95% of the beam is usually considered. While the shape changes, the emittance stays constant as a beam traverses electrostatic optics. Image modified from an image by Nicoguaro under CC BY 4.0.

The radial offset resulting from a large transverse emittance can impact the transmission through MPET's small trap aperture or other narrow sections in the beam-line. If the radial beam profile is too large, ions may either impact, or be deflected by these elements of the beam-line.

Furthermore, a large longitudinal emittance limits the width of the longitudinal spread in velocity and position. This results in a large time spread which limits the ability of the pulsed drift tube to uniformly match the kinetic energy of the ion bunch to the potential of the trap. A large energy spread will also require some ions to be rejected from the trap in order to prevent them from probing too far from the trap centre. If the full energy range of ions is accepted, they may probe regions with magnetic or electric field inhomogeneities, and thereby reduce the achievable precision by causing systematic shifts in the cyclotron frequency. Therefore, a large energy spread results in a reduced efficiency of ion transport to the Penning trap, since only ions from a fraction of the full phase space can be accepted.

Furthermore, an increase in energy spread can even complicate the identification of a resonance from the background noise. When TOF-ICR is used, a large energy spread in the ion bunch injected into MPET prevents ions from being injected in the pure magnetron eigenstate. Upon extraction, this reduces the signature of the so-called *time-of-flight effect*, which is the maximal reduction observed in the time-of-flight spectrum due to the increase in kinetic energy when the ion is excited. Such an increase in the variability of the time-of-flight makes it intrinsically more difficult to identify the signal of a TOF resonance. This effectively limits the species that can be measured in MPET.

These challenges motivate the development of a preparation trap to cool the highly charged ions. A Cooler Penning Trap (CPET) has therefore been planned to be incorporated into TITAN as shown in Figure 2.17, the details of which will be discussed in Chapter 5.

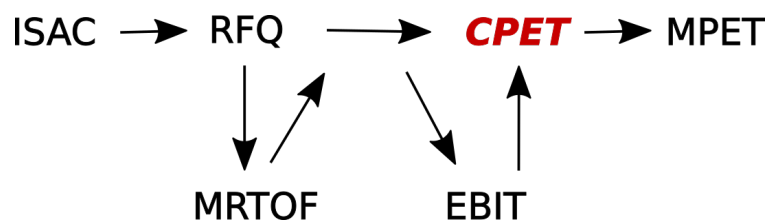


Figure 2.17: Possible flow of ions between the ion traps of TITAN. Arrows indicate direction of possible travel through connecting beam-lines. CPET is in red to indicate that this trap has not been integrated into the beam-line.

Chapter 3

MRTOF Mass Measurements of Neutron-Deficient Tm and Yb

This chapter describes the data collection and online processing of mass measurements performed using TITAN's MRTOF mass spectrometer. In order to study the $N=82$ neutron shell closure for neutron-deficient Yb ($Z=70$) and the shell closure and proton drip line for Tm ($Z=69$), exotic, proton-rich nuclei were studied at ISAC. Both elements were measured across the $N=82$ shell closure, and Tm was measured across the proton drip line. These measurements confirm the continued existence of this shell closure further into the unstable, neutron-deficient region of the nuclear chart than ever before, and establish the precise location of the proton drip line for the Tm isotopic chain. Furthermore, they provide an anchor to establish the absolute masses of alpha decay chains. These mass measurements were obtained using TITAN's recently commissioned MRTOF-MS. A description of the analysis of this data follows.

The data presented here was collected during two experimental runs with radioactive beam from the ISAC facility in 2018 and 2019. The 2018 run utilized the *Ion Guide Laser Ion Source* (IGLIS) [89] to enhance the rate of Yb ions relative to surface-ionized atoms. Resonant laser ionization enabled measurements of the most unstable Yb isotopes, with surface-ionized being suppressed using a repulsive electrostatic field.

Additionally, the 2018 experimental run represented the first use of the mass-

selective retrapping technique in a radioactive beam experiment as discussed in section 2.4.2. This allowed for measurements of species with extremely small intensities relative to the other isobars present in the beam delivered to TITAN (especially those furthest from stability). These species could differ in intensity by up to a factor of 10^8 . Retrapping was therefore utilized to suppress contaminants in both the 2018 and the 2019 experimental runs.

The 2018 data, and a corresponding analysis of the nuclear structure consequences was recently published [90]. The 2019 data provided the mass measurements necessary to identify the location of the Tm drip line and will appear in a forthcoming publication.

3.1 Rare Isotope Beam Production

The proton-rich isotopes studied in this experiment were produced at the ISAC facility before being delivered to TITAN. These exotic nuclei were initially generated when the 480 MeV proton beam from TRIUMF's driver cyclotron was impinged upon a tantalum target. The proton current on target ranged from approximately 25 to 50 μA . A cocktail of nuclear species is produced via spallation and/or fragmentation reactions within the Isotope Separator and Accelerator (ISAC) target and through the subsequent decay products of the reaction products. This creates an array of nuclei with smaller neutron number, N , and atomic (or proton) number, Z , than that of the target nucleus. Both stable and unstable species are generated, and exotic species for which observational data is sparse must be isolated for study.

In order to obtain ion beams of a single mass-to-charge ratio before delivery to experiments, ISAC employs an electromagnetic pre-separator with a resolving power of 600, followed by a high-resolution electromagnetic separator [26] which can achieve a resolving power of ~ 2500 [31].

During the 2018 experimental run, the relative purity of Yb in the beam was enhanced by utilizing the TRIUMF Resonant Laser Ion Source (TRILIS) to perform resonant laser ionization in order to enhance the rate of ionization of Yb atoms released from the target. The elemental identity of Yb could then be confirmed by

performing measurements with the lasers both blocked and unblocked, and observing the relative enhancement of the Yb peak due to laser ionization. The identity of the Tm isotopes was confirmed by using the MRS to restrict ion transmission through the MRTOF-MS to isobars, and critically, by monitoring the trends in isotopic abundance as measurements proceeded further from stability.

3.2 MRTOF Measurement and Analysis

By utilizing the MRTOF mass spectrometer, the masses of several neutron-deficient nuclei were measured to assess the nuclear shell properties and proton-bound nature of these exotic nuclei, through the neutron separation energies and proton emission Q-values. $^{157}\text{Yb} - ^{150}\text{Yb}$ and $^{157}\text{Tm} - ^{155}\text{Tm}$, ^{150}Tm mass values were obtained in 2018, and these data were supplemented by the mass measurements of $^{157}\text{Tm} - ^{149}\text{Tm}$ and ^{149}Er in 2019. Several of these masses were previously unmeasured. Calibration of the MRTOF was done with ^{133}Cs .

My analysis of these data followed several general steps:

- Data collection with the MRTOF.
- Perform time-resolved calibrations to account for slow drifts in the measured masses.
- Export the drift-corrected data as histograms.
- Simultaneously fit each peak in the spectra with a suitable function.
- Choose a suitable mass calibrant within each mass spectrum.
- Perform improved, weighted fits of the species of interest and calibrant.
- Determine the masses of the non-calibrant peaks.

The initial data acquisition and processing was done using the custom made MAC software [76]. This included the time-resolved calibration and exporting of data to histograms.

3.2.1 First Retrapping Online

The 2018 run represented the first use of the retrapping technique for online beam. The retrapping technique was particularly useful when addressing the large range in the intensities of the different species delivered to the MRTOF.

Nuclides with previously established masses must be measured alongside the ions being studied in order to allow for calibration using a known mass. However, isobaric species can interfere with the mass measurements by several different, but related, mechanisms. First, the ion of interest can be overwhelmed by a species that makes up an excessive fraction of the beam. This happens when the dominant species is so abundant that the rate at which ions are injected into the MRTOF must be limited in order to avoid ion-ion interactions that significantly impact the time-of-flight in the analyzer. Second, the scattered ions of a large peak can create a background of TOF events, against which the peak of interest becomes impossible to resolve. The small fraction of the dominant species that gets scattered within the trap effectively produces a noise floor; a background of noise which drowns out the signals of peaks of lower abundance. Therefore, there is an upper limit on the rate of ions that the MRTOF can accept. Third, the isobar peak can be so close in mass to the ion of interest that the two peaks can no longer be resolved. In this case, the uncertainty associated with the mixed composition of the peak needs to be added in quadrature to the experimental uncertainty, in accordance with the procedure in Appendix B of the Atomic Mass Evaluation [8]. Consequently, an over-abundance of any one species, or excessive overlaps in peaks of interest, must be avoided.

It is therefore important to have a degree of control over the isobaric composition of the mass spectrum. This was achieved by implementing the mass-selective retrapping technique for the first time on a beam of radioactive ions. As the fraction of contaminant ions increased, retrapping was employed to suppress the background from highly abundant isobaric species in order to accept the highest possible beam intensity (least attenuated) for the unstable isotopes with the lowest production rate. This allowed these isotopes to be resolved from the background and measured in a reasonable time frame.

3.2.2 Time-Resolved Calibration

In order to determine masses to high precision, MRTOF mass spectra require narrow time-of-flight peaks. However, while the ions are reflected back and forth, time-dependent drifts in the voltages applied to the electrostatic mirrors shift the peaks, resulting in significant peak broadening. Therefore, the first step in the data analysis requires performing a *time-resolved calibration*, or *TRC* to correct for the effect of these drifts in the TOF.

While fluctuations of the electrode voltages on a short time scale extending down to the $\sim$ Hz range can be stabilized by the system hardware using RC filters, appropriate corrections must be made for slow drifts in the voltage applied to the electrodes when acquiring TOF mass data over the course of minutes or longer. A major cause of time-dependent voltage drifts is temperature fluctuations of the power supplies. The time correction is performed by tracking the drifts of a single spectral peak of relatively large abundance, which is known as a *time-resolved calibrant*. Since different isobars will experience the same drift, the time-resolved calibrant peak is usually the one with the highest rate in order to obtain the highest time resolution in the drift-corrected spectrum.

In this experiment, the MRTOF acquired data at a rate of 50 trapping cycles per second, and 50 time-of-flight cycles were acquired per spectrum. Maintaining an ion rate below about 1 ion per cycle ensures that ion-ion interactions do not have a significant effect, but the overall rate of ions often fell below this.

The centroid of the time-resolved calibrant at a point in time can then be determined from a collection of acquired spectra which are merged into a time *block*, and all ion flight times within this block are then corrected in accordance with the position of this peak. The time-dependent correction is performed by calculating a separate value of b from this peak using equation 2.7, for each individual block. The algorithm to apply a TRC was applied in the MAC software, and the drift correction was separately determined from the mean TOF of the time-resolved calibrant peak for each block of ions. The value of b for all flight times within the spectrum are allowed to vary as a function of time so this correction can be applied. The effect of

such a correction on the TOF spectrum is shown in Figure 3.1. Part a) shows the drifts in the peaks before the TRC is performed, and part b) of the figure shows the effect of applying the drift correction. When this correction factor is used to correct the time-of-flight of all events, the peaks can be narrowed considerably.

The minimum time-scale of the drifts for which a correction is possible is limited by the rate that ions can build up statistics in the time-calibration peak. It is therefore often necessary to find a balance between correcting the drifts on a short time-scale, and having sufficient statistics per block to accurately determine the "centroid" of the TRC peak. If no appropriate time calibrant is available, stable ions from the surface ion source at the entrance to the MRTOF can be merged with the radioactive beam, however, this will also increase the background due to scattered ions accordingly.

Another consideration for the time-resolved calibration is that it may be desirable to avoid altering the true shape of the time calibrant peak. If the statistics within the individual blocks for the TRC peak are too low¹, then the shape of the calibrant peak will be changed such that the peak width is compressed and the peak shape is no longer valid in the drift-corrected spectrum. This is a consequence of the calibration also correcting for statistical variations in the peak position for each block. In order to account for this, the time calibrant should contain a sufficient number of TOF data points within a time block to accurately determine the mean time-of-flight of the peak, and thereby preserve the peak shape and width in the time-of-flight spectrum. When performing a TRC, it may be necessary to ensure that the number of data points used to determine the calibration value for b within a block is large enough to preserve the shape of the time-resolved calibration peak. For example, in the extreme limit of blocks having ≤ 1 count, the TRC peak width will resemble that of a delta function in the calibrated spectrum. However, it is often valuable to retain a meaningful shape of this peak for subsequent steps of the analysis.

The peak used for the TRC need not be the same peak that is used to calibrate the mass. The exact identity of the TRC peak can even be unknown, however if it is not confirmed to be an isobar, additional systematic effects will need to be considered.

¹The tails of a gaussian distributed variable will begin to broaden and resemble a t-distribution at low statistics

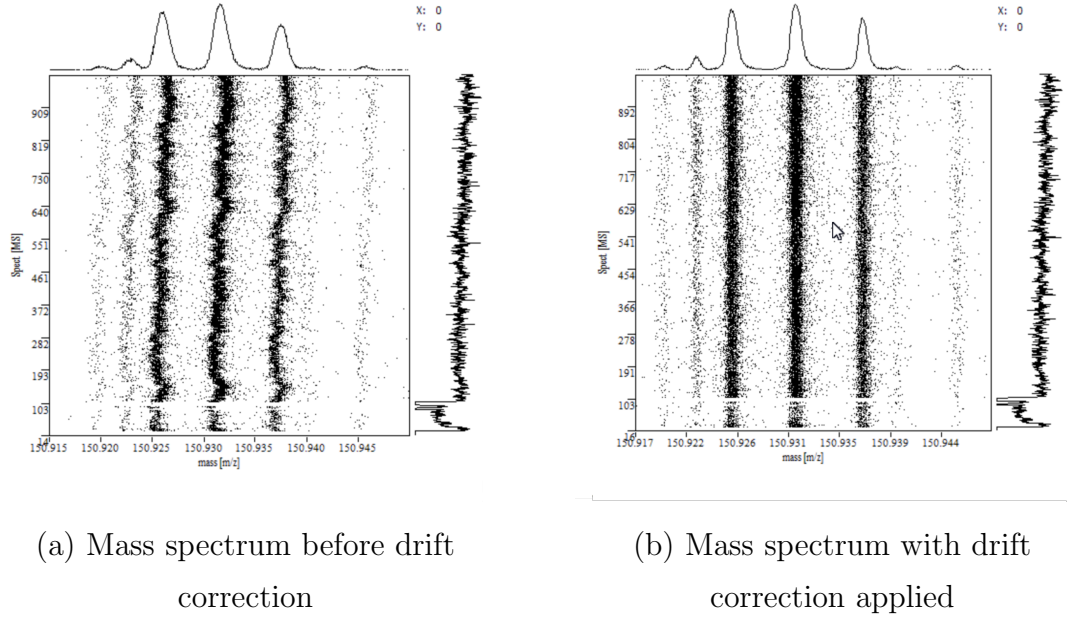


Figure 3.1: Comparison of raw vs drift-corrected MRTOF time-of-flight data. Part (a) shows a mass 151 spectrum prior to performing a time-resolved calibration. Drifts in the centroid of the calibrant peak over time are seen on the right-hand side. The effect of these time-dependent drifts on the peaks is clearly seen inside the box. The sum of all data points in this run is given at the top. Time progresses in the upward direction. Part b) shows the time-resolved calibration of the mass 151 spectrum. Drifts in the centroid of the calibrant peak over time are seen on the right-hand side. There is no visible time-dependence of the peaks inside the box when corrected using a high-rate peak. The sum of all data points in this run is given at the top and narrowing of the peaks due to this correction is evident.

Regions in which the ion count rate dropped to ~ 0 , or became too high ($> 1 - 2$ per bunch), were avoided. After time-calibration, the time-resolved data was exported as a histogram from the MAC software for fitting using the Origin Pro software package.

3.2.3 Peak Fitting Procedure

A consistent analysis procedure for fitting and calibration was applied to the data that was shown to robustly determine the masses to high accuracy.

Fit Function

I performed a weighted χ^2 minimization to fit each mass spectrum in Origin Pro using a sum of Gaussian functions, with each of the peaks being described by a single Gaussian function. Since all significant peaks are fit simultaneously, the impact from overlapping species can be reduced.

The Gaussian peak shape is an approximation, since there exists no analytical function that perfectly describes the shape of these peaks; the peak shape depends upon the specific settings to which the MRTOF is tuned. In particular, when the ion bunch is cooled to reduce its phase-space, the Gaussian character of the peaks can be seen to break down in the tails. This is due to aberrations, field imperfections, and mechanical misalignments [91]. Regardless of the fitting function used, it is essential for it to reliably and reproducibly determine the relative TOF positions of the peaks for a large range of peak intensities.

Peak Width

Ideally, the peak width used in the fit should accurately reflect the width of the true distribution. Being of practically the same A/q ratios, and subject to virtually the same fields, all peaks in a spectrum should have the same underlying shape and differ only in the number of counts they contain. For the ions under study, the peak width was generally determined from a single, high-statistics, isomerically pure, isolated peak. Other peaks in the spectrum had their width fixed at this value. An alternative would be to determine the width from a simultaneous fit of multiple acceptable peaks, but usually one peak provided the bulk of the statistics, and furthermore, it is desirable to minimize the potential influence to the peaks from overlapping isomers. The peak width could be artificially broadened if it were inadvertently determined from composite peaks containing two or more species. Therefore, in order to avoid

impacting the uncertainty, or potentially creating a systematic offset of the centroid, it was important to select an appropriate species from which to determine the width of peaks in each mass spectrum.

Weighting the Fit and Assigning a Data Cut Threshold

Another consideration was how to weight the fit. In order to account for the statistical variation in the number of counts per bin of the histogram, each bin was given a statistical uncertainty of $\sigma = \sqrt{N+1}$, which closely approximates $\sqrt{N}$ for high-statistics bins.

A standard weighting factor of $1/\sigma^2$ was applied when performing the reduced χ^2 fit, which was done in the Origin software package using an *instrumental* weighting. Since the statistics are small in the histogram bins lying within and beyond the tails of the time-of-flight peaks, they have correspondingly small absolute statistical uncertainties. This weighting can be problematic, since the weight assigned to the data points increases just as the assumed gaussian character begins to break down in the tails of the peaks, due to the reasons described above. As the fit converges, the penalty of a poorly fit centroid is often overwhelmed by the penalty of poorly fit tails and background counts, and will therefore align better to the tails than to the high-statistics centroid of the peak. This can result in a poor determination of the centroid and corresponding systematic offset of the mass value. Due to the reasonably high statistical samples in the relevant peaks, this problem can be addressed by slightly restricting the analysis to exclude the bins with the lowest number of counts. For the bins having higher statistics, the gaussian character of the peak holds, and the approximation of $\sigma \approx \sqrt{N+1}$ for the uncertainty is valid. This also reduces the possibility of undue influence from unresolved overlapping peaks that lie close to the measured or calibrant peaks, including the many isomers in this region.

Therefore, when fitting the centroid of each individual peak a cut was made at a consistent fraction of the height of that peak. The cut was made at the same fraction of the peak height for the calibrant as for all other species measured at the same mass unit. The height of the cut was guided by excluding enough of the tails to avoid any

variation in the centroid determined by the fit. In the Origin Pro (2019) software package, this cut is accomplished by applying a filter to the data. Applying this cut allowed for a consistent analysis procedure that could reliably determine the centroids of all relevant peaks.

Fit Procedure Summary

I developed a general peak fitting procedure for analyzing the current data sets. The procedure needed to determine the centroids, without the non-gaussian components in the tails systematically influencing them. In some cases, small peaks in the shoulder of another needed to be accounted for. Wherever possible, the various mass peaks were cross checked against one another for overall consistency with the known mass values listed in the Atomic Mass Evaluation (AME). Throughout the fitting procedure, the fit was monitored to ensure it converged on the appropriate peaks.

After performing a TRC and exporting the drift-corrected data, the following procedure was applied:

1. Fix the vertical offset (which accounts for the background count rate) to zero.
2. Fit all peaks in the spectrum with an unweighted, Gaussian, multiple peak function using a shared width to aid convergence.
3. Set any constraints on the fit (if applicable), making sure to tick the *enable linear constraints* box.
4. Set a threshold at 20% of the height of the calibrant peak (usually one of the largest peaks).
5. Using the parameters of the initial fit, fix the position, area, and width of all peaks except for the calibrant.
6. Perform a weighted fit of all parameters of the calibrant peak (making certain the *scale Error with sqrt(Reduced Chi-Sqr)* box is not ticked because this is done manually).

7. Record the centroid of the calibrant.
8. Set a 20% threshold on the next ion of interest.
9. Fix all calibrant parameters and assign the calibrant width to the ion of interest.
10. For ^{149}Er , fix the area and width of the ground state at the initial fit values, but allow the position to be a free parameter. The ground state only serves as a small correction to the tail of the ^{149m}Er isomer.
11. Ensure that the *enable linear constraints* box is ticked.
12. Fit for the centre and area of the ion of interest.
13. Repeat for any potential calibrants and ions of interest, generally proceeding from most abundant to least abundant species.

3.2.4 Note on Fitting with Exponentially Modified Gaussian

Mass peaks generated by the TITAN MRTOF have previously been fit using Lorentzian and Gaussian functions [60] [92]. A recently developed method for fitting MRTOF spectra utilizing the *hyper-exponentially modified gaussian* function (hyper-EMG) has also been applied to MRTOF mass spectra [91]. The hyper-EMG allows for accurate modelling of the asymmetric and non-gaussian components in the tails of peaks, and thus can provide an accurate description of regions where peaks overlap [91]. Here, *hyper* refers to the flexibility to arbitrarily add additional degrees of freedom to better describe the shape of the peak. In order to determine the peak positions, this method models the peak as a gaussian function convoluted with an arbitrary number of exponential functions which are used to better describe the tails of the peaks. The contributions of all functional parameters is generally obtained by fitting the function to a single, well-separated peak of high statistics.

The EMG was applied to the 2018 Yb data to extract the excitation energy of the ^{151}Yb isomer [90]. The masses which were determined using a Gaussian have been shown to be quantitatively consistent with independent analyses using the EMG

function to fit much of the present data. No improvement to the precision of mass measurements was observed for these data, since uncertainties are dominated by the statistics of the smallest peaks at each mass, and the various systematic effects.

While, the present analysis using Gaussian fit functions could determine the centroid of the dominant ^{151}Yb isomer, the ground state is not presented here due to a large uncertainty that arises when using this method on a low abundance peak residing in the tail of another. However, the ground state peak was included in the overall fit procedure, in order to account for the influence of these counts on the mass of the isomer.

Furthermore, it is not always possible to apply the full EMG fitting procedure to the rich mass spectra that were observed in this experiment. A well-resolved, high-statistics, peak, unpolluted by the presence of possible isomers or other unresolved species, is required to determine the parameters of the peak shape. Such a peak was not available within some of the mass spectra under consideration in this data set. For future experiments, it would be very useful to employ a laser-ablation source to ionize stable atoms within the MRTOF, thus ensuring a well-separated isobaric calibrant (e.g. carbon clusters) is always present [93]. Such a source could be turned off to accurately assess any contribution from the background. For these reasons, it was sensible to fit the peaks using gaussian functions which were applied consistently across all mass units under analysis.

The peak shape was more symmetrical during the 2019 runs due to an improved tune of the MRTOF, but even the asymmetry of the 2018 runs was not observed to cause any systematic shift to the mass values when fitting with a gaussian versus the hyper-EMG.

3.2.5 Mass Calibration

Once the centroids of the peaks are determined by fitting the data at a single mass unit, it is necessary to select appropriate mass calibrants for each of the isotopes under study.

There are several criteria which must be considered when selecting a peak as a

mass calibrant:

- The calibrant peak should be an atomic or molecular isobar (systematic effects of non-isobars was unknown).
- The calibrant peak must have a low statistical uncertainty.
- The calibrant mass must have been previously measured both accurately, and with sufficient precision.
- Calibrant peaks that could be influenced by underlying isobars (including isomers, molecules, and doubly-charged species), must be avoided.

Although non-isobaric species that execute a different number of reflections can, in principle, be used as mass calibrants, the systematic uncertainty associated with doing so was not established during these measurements. Therefore, only isobaric species were used for mass calibration.

An appropriate mass calibrant also must have been delivered at a sufficient rate to have acquired adequate statistics for a robust fit of the peak that can determine the centroid both accurately and precisely, while minimizing any systematic uncertainty. No matter how well the isotope under study is measured, the mass determination can only be as good as the uncertainty on both the current measurement and the previously established mass of the calibrant species. The mass of the calibrant isotope therefore must be well-established, having been previously determined to high precision, and ideally measured via a direct measurement technique. The established masses of the calibrants were extracted from the 2020 Atomic Mass Evaluation [94].

A calibrant must not be unduly influenced by the presence of any other ion species that overlap with it. Since the spectra under study were rich in isobars, many possible calibrants were available, but care was taken to select a calibrant for each mass that was not influenced by unresolved isobars, as this would cause the centroid of the peak to be shifted from the true mass value. I carefully considered potential influences from elemental species, molecules, and isomers, but multiply-charged species were not of concern at these atomic masses. Some isobars with very similar masses, prevented

several potential mass calibrants from being fully resolved due to their overlapping peaks, and likewise, some of the species under study could only be measured as a combined mass of the ground state and one or more isomers; in many cases, the beam delivered from the target contains a greater abundance of the isomer than of the ground state. To avoid any systematic shifts to the mass, the mass calibrant should not significantly overlap with an adjacent TOF species, or if it does, the influence of that species to the centroid of the calibrant must be well-understood.

This required carefully choosing appropriate mass calibrants, and adjusting uncertainties in measured masses accordingly. When selecting an isotope to use as a mass calibrant, isotopes that were believed to contain long-lived isomers that could shift the centroid of the peak were avoided. For example, Figure 3.2 shows the ^{151}Ho peak, for which the centroid is believed to be influenced by an unresolved isomer at 41 keV.

The mass calibrant need not be the same peak as the time-resolved calibrant, but there should be no ambiguity as to the identity of the calibrant used; both existing nuclear data and trends in the chemical elements extracted from ISAC that carry over from one mass unit to the next were therefore used to identify peaks and exclude potential calibrants that were likely to be polluted by an unresolved species. Additionally, no calibrant or measured species should lie so close to the edge of the time-of-flight window that the fields of switching electrodes may have shifted or broadened it.

An example of a mass calibrant that required particular consideration was the ^{152}Dy peak. Systematic trends in the relative abundances of Dy to Tb and its isomers were considered to determine that Dy is expected to exceed any Tb species by an order of magnitude in abundance. This prevents any of the possible contaminants from having a significant influence on the centroid of ^{152}Dy . A similar trend in the abundance indicates that ^{152}Nd is also expected to make a negligible contribution to this peak. Likewise, a check was done that the tail of the $^{150}\text{Ho}/^{150m}\text{Ho}$ peak is not expected to impact the centroid of the ^{150}Er calibrant peak, as shown in Fig. 3.3.

Calibrants were thus selected to balance the statistical uncertainty of potential calibrants with the various factors that can impact their reliability. Difference in

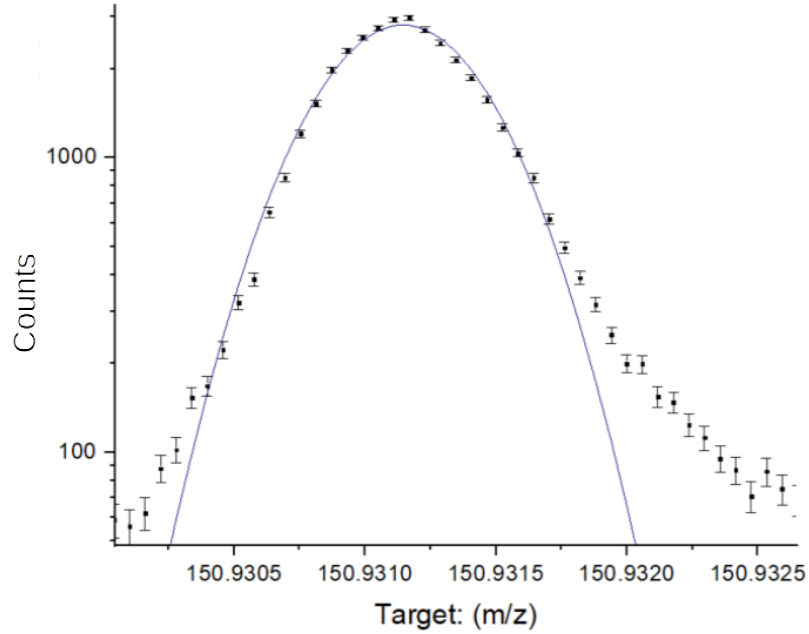


Figure 3.2: This fit of the ^{151}Ho peak is an example of a peak that is believed to contain an unresolved overlap of the isomer and ground state. Since the relative abundance of each species is not precisely known, this highlights the importance of judiciously selecting which species to use as mass calibrants. Only peaks believed to contain a single species were used as calibrants.

beam composition between the 2018 and 2019 data sets due to variations in the target temperature, the presence of lasers, retrapping windows, etc., necessitated the use of different species as mass calibrants. The 2018 runs had a large variation in the best available elements for calibration, using ^{157}Tm , ^{156}Tm , ^{155}Eu , $^{138}\text{Ce}^{16}\text{O}$, ^{153}Dy , $^{136}\text{Ce}^{16}\text{O}$, ^{151}Er , and ^{150}Dy . The 2019 calibrants were more consistent, using ^{157}Yb , ^{156}Yb , ^{155}Er , ^{154}Dy , ^{153}Dy , ^{152}Dy , ^{151}Dy , ^{150}Dy , and ^{149}Dy . At mass 155 the Er mass had excellent agreement with ^{155}Dy and ^{155}Ho , but Er was used to minimize the risk of contamination from species with similar mass. Masses 156 and 157 could only calibrate the Tm mass using Yb, since changing isotopic distributions resulted in low Dy statistics.

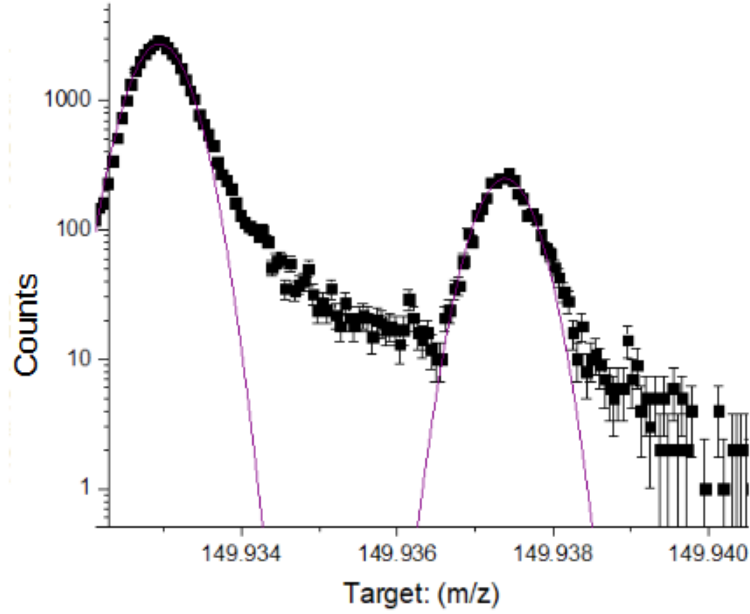


Figure 3.3: The ^{150}Er calibrant required caution due to its proximity to the tail of the $^{150}\text{Ho}/^{150m}\text{Ho}$ peak. The possible influence of tails from adjacent peaks was one motivation for fitting the peaks only to the bins above a certain fraction of the total peak height for the Gaussian fits. The points near the peak have an uncertainty of $\sim \pm 16$ counts per bin while the background is constant within a couple counts from the left side of the peak to the right.

3.2.6 Statistical Uncertainty Characterization

The uncertainty on the determined mass must include that of the measured peak, the calibrant peak, and any systematic error. The statistical uncertainty in the centroids of the measured and calibrant isotopes were added in quadrature to the fit error, and the systematic error. Needless to say, this also must incorporate the inherent uncertainty in the previously established mass of the calibrant peak, and this was extracted from the 2020 AME [94].

Unfortunately, a fully analytic model of the mass peaks obtained by the MRTOF does not exist, the peak width and shape being optimized before an experiment by careful tuning of the voltages on the mirror electrodes in order to obtain a high re-

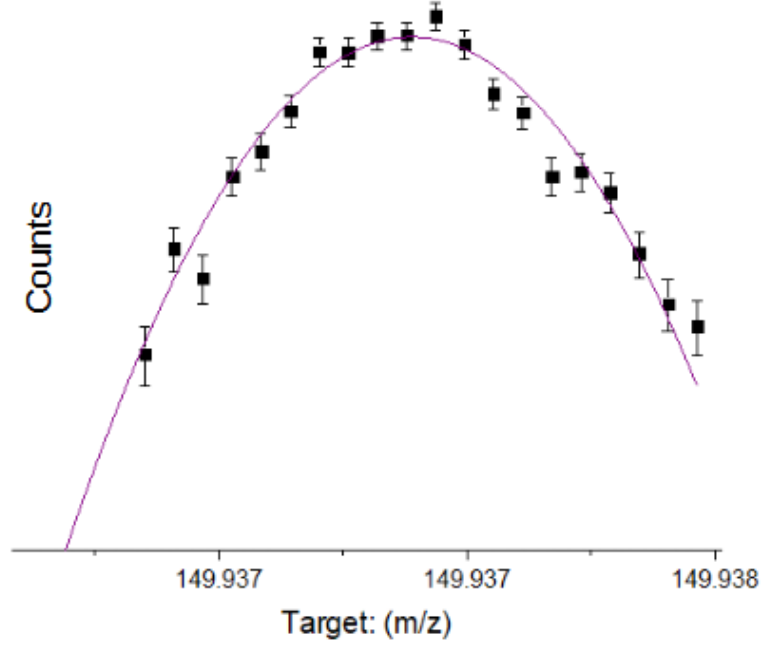


Figure 3.4: A weighted fit of the ^{150}Er mass calibrant.

solving power. Although the MRTOF peaks from these experiments are not perfectly gaussian, the centres of the peaks, which contain the highest statistics, can be well described by a gaussian peak shape.

Therefore, all mass peaks believed to be composed of a single species were simultaneously fit using multiple gaussian functions sharing a common peak width. The width was generally determined from a pure peak of high statistics. The uncertainty in the centroid of a gaussian function is known as the *standard error of the mean* or *standard uncertainty of the mean* and is given by $\sigma_x = \sigma/\sqrt{(n)}$, where σ is the population standard deviation (i.e. the expected deviation of a single measurement from the mean), and n is the total number of detected TOF events contained within the peak.

The statistical uncertainty must also depend on how precisely the mass calibrant peak is measured in the mass spectrum. The total statistical error for a measured mass is determined by adding the measurement uncertainty and calibrant uncertainty

in quadrature.

If a measured peak contains a single unresolved isomer or other isobar for which the relative composition is unknown, a standardized procedure is given in the Atomic Mass Evaluation for determining the mass and uncertainty [8]. The relevant expressions for a mass determined from two unresolved mass peaks is given by

$$M_{exp} = \frac{1}{2}(M_0 + M_1), \quad (3.1)$$

with an uncertainty of

$$\sigma = \frac{\sqrt{3}}{6}(M_1 - M_0) = 0.290(M_1 - M_0), \quad (3.2)$$

but efforts were made to avoid using overlapping peaks as calibrants when possible.

In addition to the statistical error, the error from the fit to the peak performed in Origin Pro, and the systematic uncertainty were also added in quadrature to obtain the total uncertainty.

3.2.7 Systematic Characterization

Additionally, several systematic effects associated with the measurement within the MRTOF need to be considered [50].

At this point, the systematic uncertainty was dominated by ringing in the voltages when the electrostatic mirrors of the MRTOF were closed and opened. A systematic error of 3×10^{-7} was included for the initial online mass measurements performed with the MRTOF in 2018 of Yb and some Tm isotopes. In March of 2019, additional filtering was introduced to slow the speed at which the potentials of the electrostatic mirrors is switched in order to minimize this ringing while maintaining sufficient switching speeds [77]. This reduced the estimated systematic uncertainty to 1.5×10^{-7} for the 2019 Tm measurements, and it is now dominated primarily by ripple generated within the power supplies themselves.

To avoid systematic shifts to the mass from ion-ion interactions, the rate of ions was kept small during the measurements, so that only approximately one ion at a time traversed the analyzer. The 2018 measurement mass 153 had a slightly higher

rate than usual, but ion-ion effects should still not be significant enough to add to the overall systematic uncertainty.

Chapter 4

Results of the Lanthanide Mass Measurements

In order to address questions related to the shell closure, the proton drip line, and trends in the energies of isomers, mass measurements were taken of neutron-deficient isotopes of Yb and Tm, near the N=82 shell closure.

The proton drip line nucleus is the last nucleus for which the proton separation energy S_p remains positive. It is defined by the difference in mass between the nucleus and its prospective decay products, which consist of a proton and the recoiling nucleus with $Z - 1$ protons. Beyond the drip line, the proton emission Q-value (where $Q_p = -S_p = M(A, Z) - M(A - 1, Z - 1) + M(^1\text{H})$) also serves as a necessary input to calculate the proton emission half-life of the unbound nucleus, along with the angular momentum.

Although ^{147}Tm and ^{148}Tm (at N=78 and N=79 respectively) had previously been established to lie beyond the drip line [95], the precise location of the drip line in the Tm (Z=69) isotopic chain has been undetermined up until now. The region around the proton drip line and nuclear shell closure is of particular interest, because the single-proton nuclear potential is known to be more strongly impacted by the neutron density (and vice-versa) [12]. The present mass measurements finally allow for the identification of the limit of proton-bound Tm isotopes.

The extent of the N=82 shell closure at the extreme neutron-deficient limit is

also unknown. Both two-neutron separation energies, as well as the neutron pairing gap are able to provide reliable evidence of neutron shell closures, but rely on the availability of ground state masses. Prior to this work, measurements of two-neutron separation energies (S_{2N}) of neutron-deficient isotones along the $N=82$ shell closure extended as far from stability as ^{150}Er ($Z=68$). The S_{2N} of both ^{151}Tm and ^{152}Yb , located at $N=82$, were previously estimated by extrapolations from the Atomic Mass Evaluation, but had never been measured experimentally.

4.1 Results and Discussion

As described in Chapter 3, the masses of neutron-deficient isotopes near the $N=82$ neutron shell closure were measured over the course of two experimental runs in 2018 and 2019, to precisions in the range of 3×10^{-7} using the MRTOF-MS at TITAN. High beam rates of exotic neutron-deficient isotopes of the lanthanide elements were achieved by impinging the proton beam from TRIUMF's cyclotron on Ta targets. Both laser-ionized and surface-ionized species were filtered through the ISAC mass separator and delivered to the TITAN facility.

The masses of ytterbium ($Z=70$) isotopes ranging from ^{150}Yb ($N=80$) to ^{157}Yb ($N=87$) were measured, as well as ^{149}Tm ($N=80$) to ^{157}Tm ($N=88$). These measurements included $^{149,150}\text{Tm}$, and $^{150,153}\text{Yb}$, which had never been measured before, as well as $^{151,152}\text{Yb}$ which had previously only been measured indirectly [?] [?]. The masses of these isotopes allow for neutron and proton separation energies in this region to be determined, thereby allowing the neutron shell closure to be probed and establishing which nuclei are proton-bound. Furthermore, the previously measured mass of ^{149m}Er was also measured to deviate from the value listed in the Atomic Mass Evaluation. This shifts the binding energy enough to directly impact the determined location of the drip line.

Since the MRTOF often also measures other isobars delivered in a beam, the masses of ^{149}Er to ^{154}Er were measured as well. ^{155}Er was also delivered in the beam, but is used as a calibrant and confirmed to be consistent with the AME 2020 values by both ^{155}Dy and ^{155}Ho . TITAN's MRTOF-MS was used to measure atomic masses

for species with beam rates down to $\lesssim 0.08$ pps at the final MagneTOF[®] detector.

The Yb data was mainly gathered during the 2018 beamtime, and the Tm data mainly during the 2019 beamtime. Yb yields were significantly enhanced by employing element-specific resonant laser ionization during the 2018 measurement. On the other hand, the yields of Tm only began to improve in the later part of the 2018 experiment, primarily due to an increase in the target temperature. This allowed for the first measurement of ^{150}Tm and opened the door to the more precise and comprehensive set of Tm measurements that was taken in 2019. The reach of the Tm measurements was thereby extended down to ^{149}Tm . The present measurements of the Yb and Tm mass values are compared to the 2020 Atomic Mass Evaluation [94] in Figures 4.1 and 4.2 respectively. It can be seen that the mass uncertainty is substantially reduced, especially for species with no prior measurement.

The 2019 experimental run offered improved statistical and systematic uncertainties over the 2018 run. This was made possible with a better tune of the electrodes in TITAN's MRTOF and by improvements made to the reproducibility of the potentials applied to the electrostatic ion mirrors. Furthermore, several Lu masses can now be determined via their known decay energies. The unmeasured mass of ^{151}Lu is measured through the known S_p of -1241.0 ± 1.8 keV, and ^{154}Lu its known S_p of -204 ± 14 keV. The mass of ^{153}Lu at $N=82$ can also be improved via this species' known proton decay energy of -606 ± 10 keV[96] from its $h_{11/2}$ ground state into ^{152}Yb . Additionally, the masses of several heavier nuclei which are connected through alpha decay chains can now be anchored via a direct mass measurement, which has been discussed in a recent publication [97].

4.2 Scientific Impact of Tm and Yb Mass Measurements

The measurements of Tm and Yb isotopes discussed in this work extend the boundary of experimentally-determined, ground-state binding energies across the $N = 82$ neutron-deficient shell closure. Additionally, the mass measurements identify

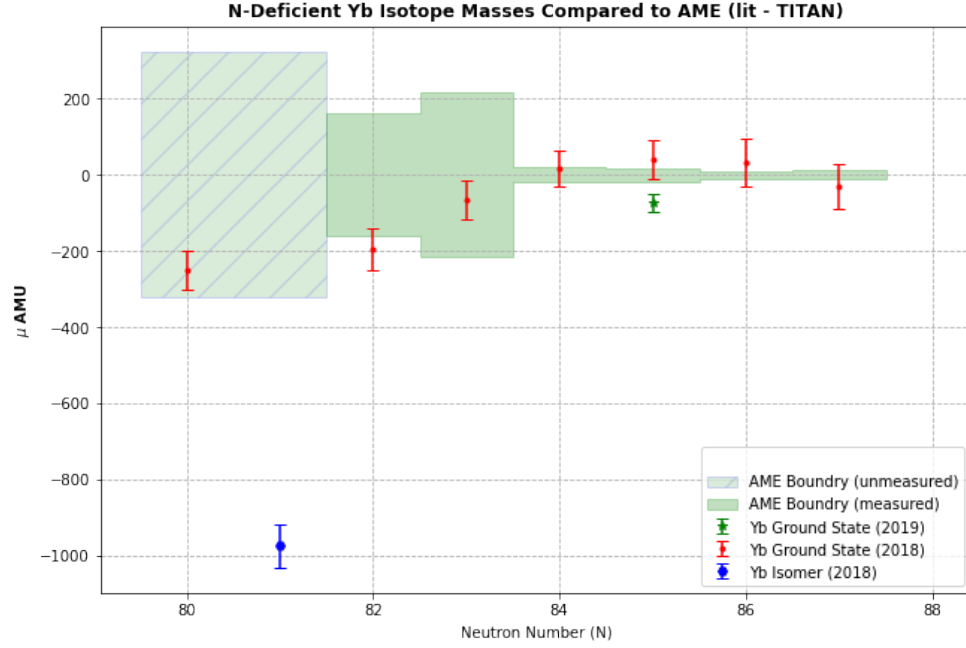


Figure 4.1: Comparison of the Yb ($Z=70$) isotope masses measured at TITAN with the 2020 Atomic Mass Evaluation [94]. The blue point shows the excitation of the measured isomer from the ground state, which was determined separately in [90].

the location of the proton drip line in Tm ($Z=69$), and in the process seem to uncover a likely error in the accepted mass value of ^{149}Tm .

The mass surface of neutron-deficient lanthanide isotopes was determined by measuring the masses of several hitherto unmeasured nuclear ground states. Ultimately, these masses test mass models near the $N=82$ nuclear shell closure, and established the location of the proton drip line in Tm.

4.2.1 Comparison to Theoretical Models

One interesting line of inquiry in the field of exotic isotopes is to look for deviations from the classical *magic numbers* of closed neutron shells, which may change for nuclei in regions far from stability [12]. As discussed in Chapter 1, key indicators of a shell closure include a sudden change in the two-neutron separation energy (S_{2n}), as well as sharp changes in the two-neutron separation energy, and a spike in the corresponding

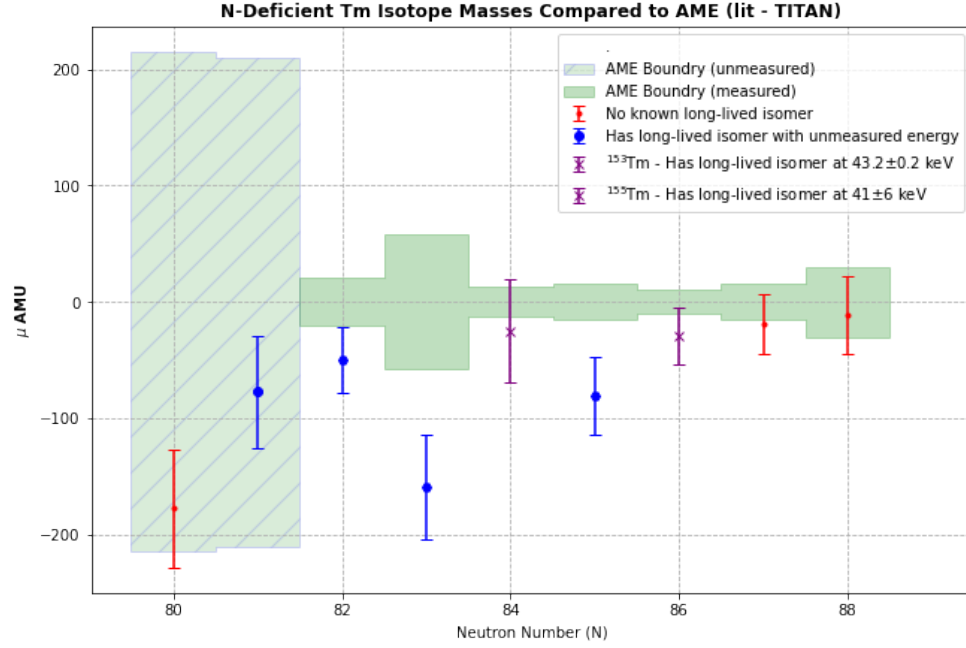


Figure 4.2: Comparison of the Tm isotope masses measured at TITAN to the 2020 Atomic Mass Evaluation [94]. Differences from the AME values can be explained by the presence of known isomers. Although ^{150}Tm has a long-lived isomer at 671 keV, the energy is high enough for the ground state to be resolved from it.

neutron pairing gap.

The strong shell model characteristics seen in the ratio of the first two single-particle excitation energies ($4^+/2^+ \approx 1.3$) of the even-even $N=82$ isotones is observed to soften slightly further from stability (Figure 4.3). However, as can be seen in Figure 4.3, the ratio of these excitation energies for ^{152}Yb has not yet been measured. Therefore, an alternate means of testing the extent of the shell closure is thus needed.

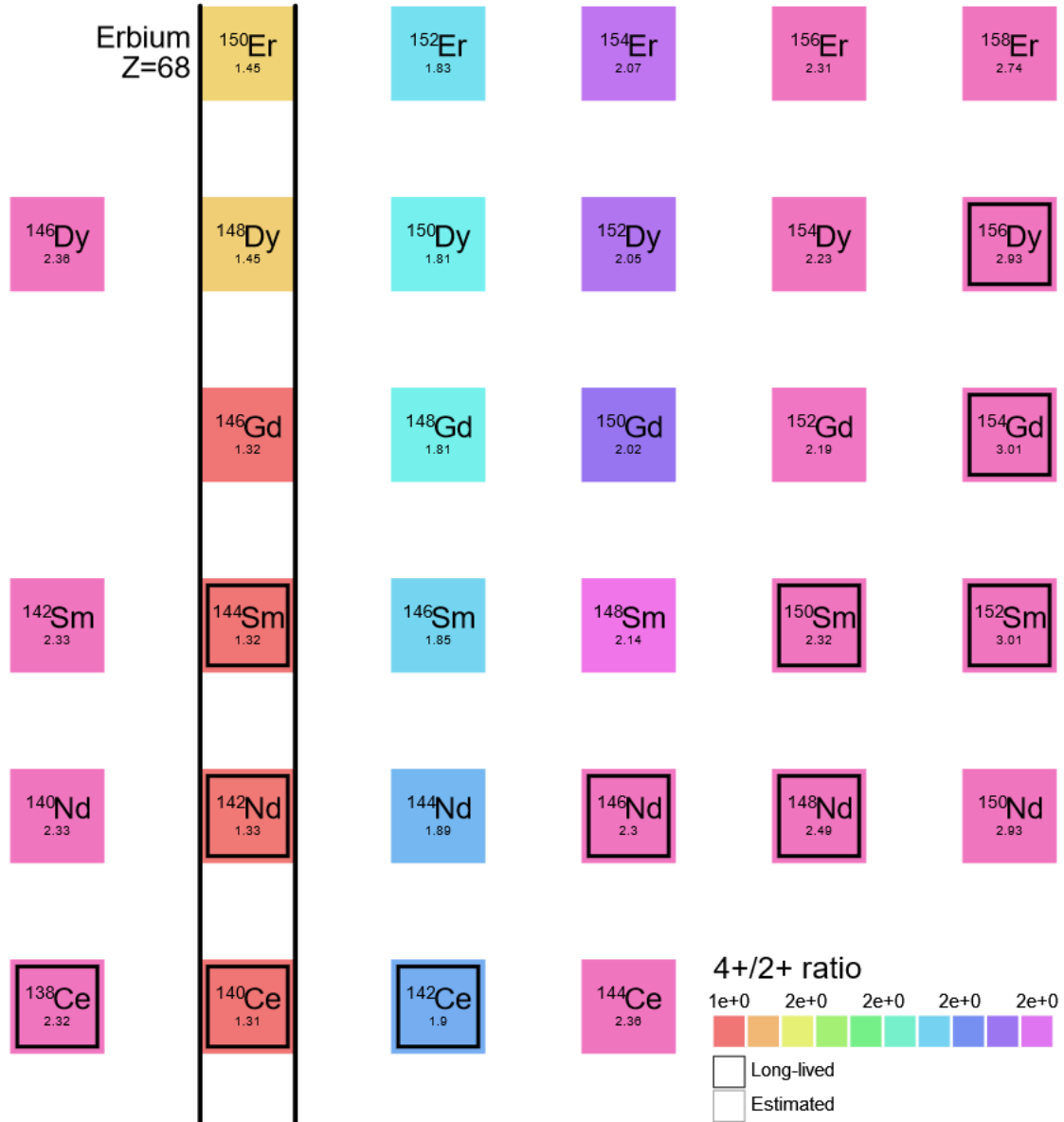


Figure 4.3: The ratio of the $4^+/2^+$ excitation energies for N=82 nuclei begins to increase slightly from a value of 1.33, which is associated with a shell closure, as Yb (Z=70) is approached. Measurements are therefore needed further from stability to test whether the shell closure becomes quenched. Here the colours of even-even species on the table of nuclei indicate the magnitude of this ratio on a scale from 1.3 to 2.3, with the $4^+/2^+$ excitation ratio written under each nuclide. Image generated by The Colourful Nuclide Chart (<https://people.physics.anu.edu.au/~ecs103/chart/>) and used with permission [98].

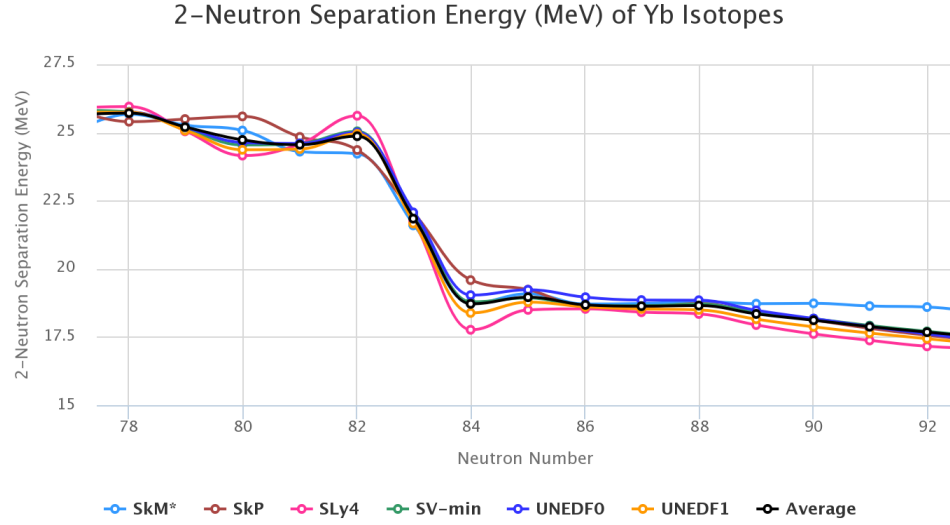


Figure 4.4: DFT predictions of the two-neutron separation energy of Yb around the N=82 shell closure. From massexplorer.frib.msu.edu using data from [99].

Since the *two-neutron separation energy*, or S_{2n} , is a common gauge for observing the trends in the binding energies along isotopic chains, and assessing whether they exhibit shell-like behaviour, the mass results were fed into equation 1.3 of section 1.3. Figure 4.4 shows Density Functional Theory (DFT) predictions of the S_{2n} using various energy density functionals.

The trend observed in the binding energies obtained from the direct mass measurements obtained by TITAN's MRTOF shows that a strong neutron shell closure at N=82 continues to stabilize isotopic chains as far from stability as Z=70 (Yb). This can be clearly seen in the step increase in the two-neutron separation energy between N=82 and N=84 visible in Figure 4.5. This persistence of the shell closure supports the DFT theoretical predictions for this region.

Similarly, as discussed in [90], the ^{152}Yb nucleus (Z=70) exhibits a pairing gap that does not deviate significantly from other isotones at the N=82 shell closure. This is consistent with predictions that ^{153}Lu is the first proton unbound nucleus at the N=82 shell closure [94][14].

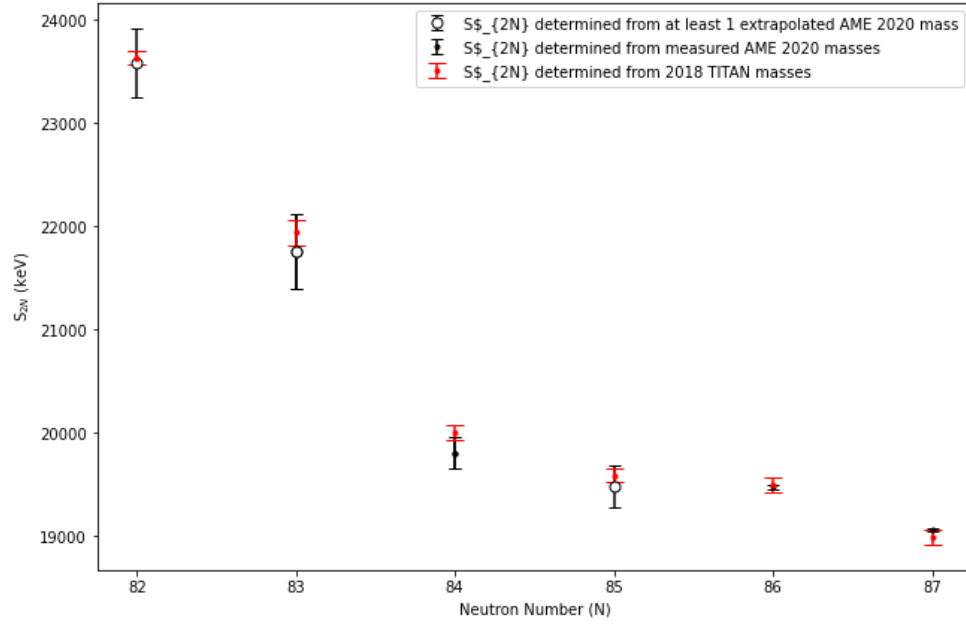


Figure 4.5: The shell closure is apparent in the two-neutron separation energies (S_{2N}) given by the newly measured masses of the Yb ($Z=70$) isotopic chain.

4.2.2 Establishing the Limit of Proton-Bound Thulium

The single-proton separation energy is equivalent in magnitude to a Q-value for radioactive decay by proton emission, but opposite in sign. Proton emission becomes a possible decay mode when the separation energy, S_p , drops to ≤ 0 , i.e. the nucleus is no longer proton-bound. The separation energy obtained through mass measurements of Tm isotopes along with an anomalous Er mass that deviates from the current value in the Atomic Mass Evaluation, suggest that ^{149}Tm is the first proton-unbound isotope in the Tm chain.

Mean Field Theories, which consider the interaction of an individual nucleon with the overall potential of the nucleus, are a well-established tool for predicting the ground state properties and low-lying energy levels of nuclei, especially near nuclear shell closures. Figure 4.6 shows that although different theoretical predictions of the location of the Tm drip line exhibit some variation, ^{150}Tm ($Z=81$) is predicted to be proton-bound. The plot was extracted from massexplorerer.frib.msu.edu which

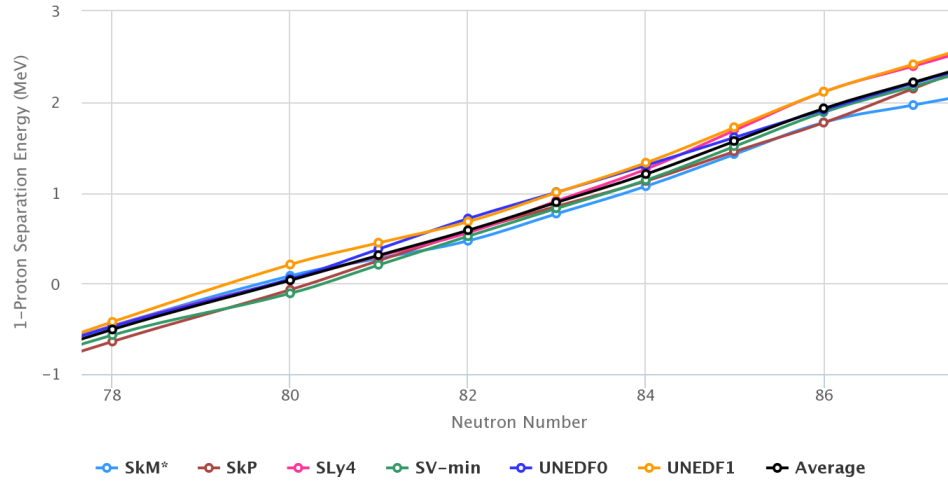


Figure 4.6: When the proton separation energy drops below zero, the drip line is crossed. Theoretical (DFT) predictions of proton separation energy of Tm disagree upon whether the drip line occurs at $N=80$. From massexplorer.frib.msu.edu using data from [99].

uses the data calculated in [99]. The models cluster around a separation energy of 0 MeV for ^{149}Tm ($N=80$), so little can be said regarding whether this species lies beyond the drip line.

A careful analysis of the second set of Tm data has provided the first experimental data indicating the location of the proton drip line at $Z=69$. This was accomplished through mass measurements of Tm isotopes across the drip line, as well as a correction to the proton emission decay product ^{149}Er .

Previously, a bayesian analysis with nuclear density functional theory employing several different energy density functionals had predicted the lightest nucleus at which the thulium isotopic chain ($Z = 69$) remains bound with respect to proton emission to be ^{150}Tm which has a neutron number of $N = 81$ [14].

The Impact of ^{149}Er and ^{149m}Er

When only the newly measured masses of Tm are used to supplement the existing Atomic Mass Evaluation (AME 2020 [94]) data, the calculated proton separation

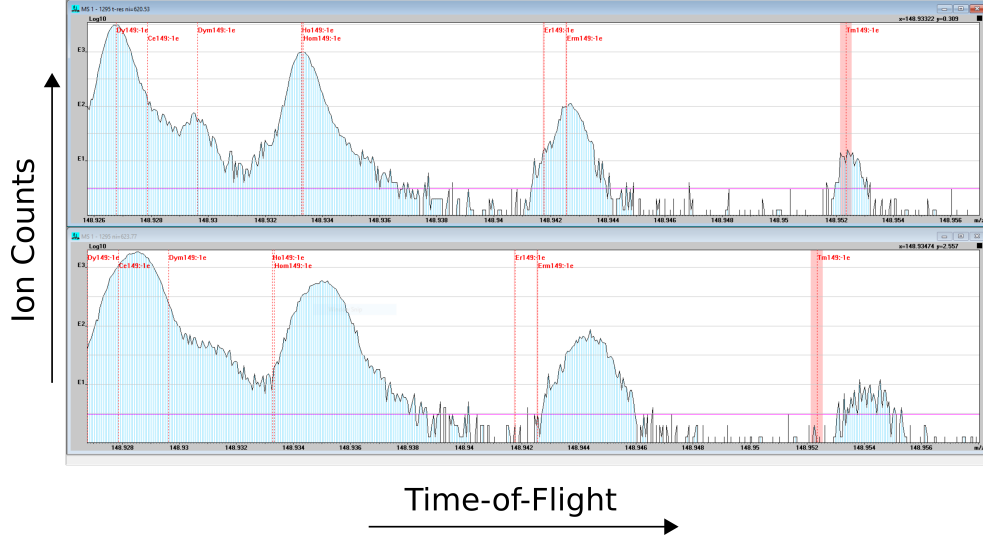


Figure 4.7: Raw TOF data at $A=149$. The unexpectedly low proton separation energy of ^{150}Tm could be accounted for by a deviation in the mass of the ^{149}Er isomer and ground state. This amounts to an increase of more than 100 keV from the present AME value. When the drift correction is applied to the raw data of the lower spectrum, and a careful analysis is applied to the time-resolved spectrum in the top of the figure, the centroid of ^{149}Er is revealed to lie further to the right than predicted by the 2020 AME [94]. The ground state of ^{149}Er may also be present in the time-calibrated spectrum pictured here.

energy of ^{150}Tm suggests that it could be either proton-bound or unbound, with a weak preference for it being unbound. This initial evaluation of the Tm data was therefore insufficient to make a definitive conclusion about the drip line in Tm. Furthermore, unbound ^{150}Tm would be in tension with the theoretical prediction in reference [14] that it is a proton-bound isotope. However, as outlined below, a more complete assessment of the data taken with TITAN's MRTOF strongly suggests that ^{150}Tm is, in fact, proton-bound.

An anomalous mass of an Er isotope appears to significantly impact the location of the Tm drip line. The proton separation energy of a nucleus depends not only on the mass of the candidate proton emitter, but also on the mass of the daughter nucleus of the decay. Although a definite location for the drip line cannot be extracted from

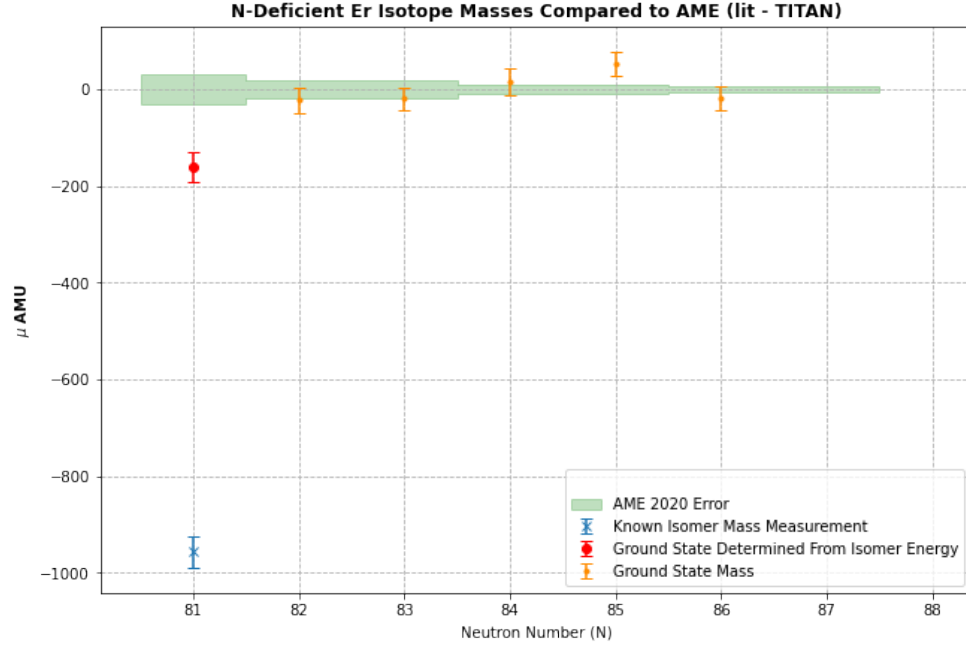


Figure 4.8: Comparison of the Er isotope masses measured at TITAN in 2019 to the literature values of the 2020 Atomic Mass Evaluation [94].

the ^{149}Tm and ^{150}Tm masses alone, revised values for the masses of $^{149g,m}\text{Er}$ allow for an unambiguous identification of the Tm drip line.

Since Er is one element closer to stability than Tm for neutron-deficient nuclei, a peak corresponding to ^{149m}Er was measured concurrently with Tm at mass 149 during the 2019 experiment. The ^{149m}Er peak is clearly seen in figure 4.11. The shoulder of the peak likely contains some of the ground state as well, but not enough to significantly influence the centroid. Although a prior measurement of the ^{149}Er isomer using Schottky mass spectrometry [100] had determined these masses with an uncertainty of ± 28 keV, the present data set obtained with the MRTOF indicates that the masses of ^{149}Er and ^{149m}Er lie $\sim 150 \pm 29$ keV/ c^2 higher than this literature value.

Using the previously established excitation energy separating the $^{149g,m}\text{Er}$ ground state and isomer, the mass of the ground state is determined through the present mass measurement of the isomeric state. This gives a ground state mass of ^{150}Tm that firmly establishes its proton-bound nature.

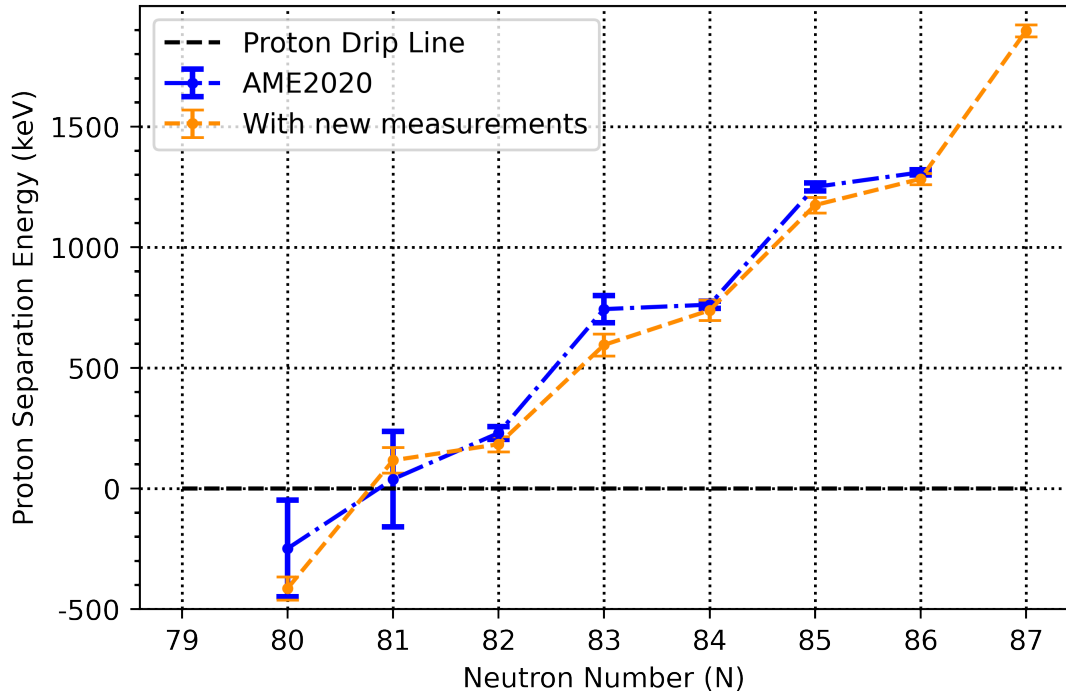


Figure 4.9: The proton separation energy (S_p) is shown in orange using the new measurements of masses in the Tm isotopic chain, and the newly ground state mass of ^{149}Er , as determined from the mass of the isomer. The blue points show the separation energy determined from the literature values contained within the 2020 AME [94].

The excitation energy of the ^{149m}Er isomer is known to be 741.8 ± 0.2 keV according to the NUBASE 2016 evaluation [101]. This was determined through gamma ray spectroscopy experiments, the isomer being known to transition to the ground state through a $630.5(\text{M}4) \rightarrow 111.0$ (111.3)(M1) gamma ray cascade [102][103]. Furthermore, as discussed in the recent paper detailing results from the 2018 data-set, the $N=81$ isomers have been shown to have a remarkably consistent excitation energy of approximately 750 keV due to a clustering of single-proton energy levels for these mildly deformed nuclei at $N=81$ [90]. This is due to the single hole in the $h_{11/2}$ neutron orbital common to ^{149}Er and many of its isotones [102].

The mass of the ^{149m}Er isomer had been previously determined by a direct mass

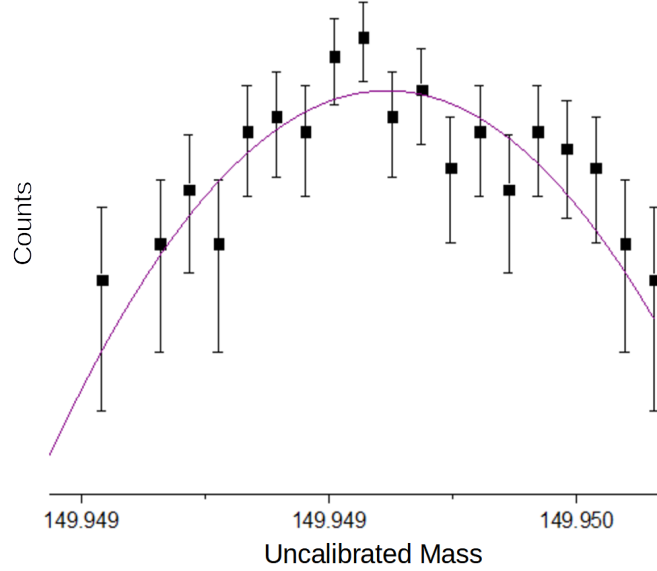


Figure 4.10: A weighted fit of the ^{150}Tm peak after a cut is made at 20% of the peak height (prior to mass calibration).

measurement using Schottkey mass spectrometry (SMS) at the FRS-ESR storage ring at GSI [100], which fixes the ground state mass through the known excitation energy. This mass was published as part of a vast data-set in 2005 alongside the masses of 70 other nuclides which had not previously been measured. SMS is known to be a reliable technique, so the origin of the discrepancy is unclear. However, the new mass of ^{149}Er obtained by the MRTOF strongly suggests that ^{149}Tm , with 20 fewer neutrons than the only stable isotope of Tm (^{169}Tm , $N=100$), is indeed the first ground state in this isotopic chain capable of radiating a proton. This demonstrates how a single discrepancy in a critical database can directly impact the experimental benchmarks that nuclear physicists rely on to test theoretical models.

Although the ground state of ^{149}Er is likely also present, being hidden 741.8 ± 0.2 keV to the left of the centroid of the isomeric peak in the spectrum, its influence is minimal. However, as a precaution, an additional gaussian peak was added to the spectrum with the centroid fixed 741.8 keV lower in mass. In a worst case scenario, this additional peak could only shift the centroid of the isomer in the heavier direction

by accounting for some of the tail of the isomeric peak, so ^{150}Tm would remain bound. Since the tails of peaks are underestimated by the gaussian fits, a fit of the ground state will always slightly overestimate the relative contribution from of the ground state to the overlapping peaks. As a result, the centroid of the isomer would be shifted to a higher mass, and the separation energy of ^{150}Tm would be increased. Regardless, the relative contribution of the ground state will not change the conclusion that ^{150}Tm is proton-bound. Excluding the ground state shifted the mass by a maximum of 28 keV.

The mass of the Er isomer was determined under the assumption that the isomeric peak and its low-statistics ground state are relatively gaussian, and that the excitation energy is fixed. An independent fit was also performed to confirm this result with an analysis that uses a hyper (i.e. many degrees of freedom) exponentially-modified gaussian function (EMG) to fit both peaks. This analysis agreed with the mass value determined using a fixed excitation energy. The EMG allows for the excitation energy to be a free parameter, but the peak shape is determined from a calibrant, so it must assume that the peak shape calibrant is not significantly impacted by isomers.

A revised ground state mass of ^{149}Er was therefore determined through the known excitation energy. Figures 4.11 and 4.12 show a fit of the mass peak of the previously measured $^{149}\text{Er}^m$ isomer. This yields a value for the mass which is heavier than the SMS value in [100] by 150 keV. This in turn, reduces the Q-value of the ^{150}Tm decay enough to establish it as being proton-bound.

Influence from the ^{150}Tm isomer should also be minimal due to the 5.2ms half-life, but any effect would only imply a reduction in the true mass of the ground state, further solidifying its status as a proton-bound isotope. The only caveat would be that if an unknown, long-lived isomer of ^{149}Tm was measured instead of the ground state, it could then be possible for the true ground state to be proton-bound, however, no such state is presently known to exist.

In summary, the newly measured ^{150}Tm mass, and the corrected ^{149}Er measurements establish ^{150}Tm to be proton-bound. Additionally, the measurement of ^{149}Tm indicates that it is the first isotope lying beyond the proton drip line.

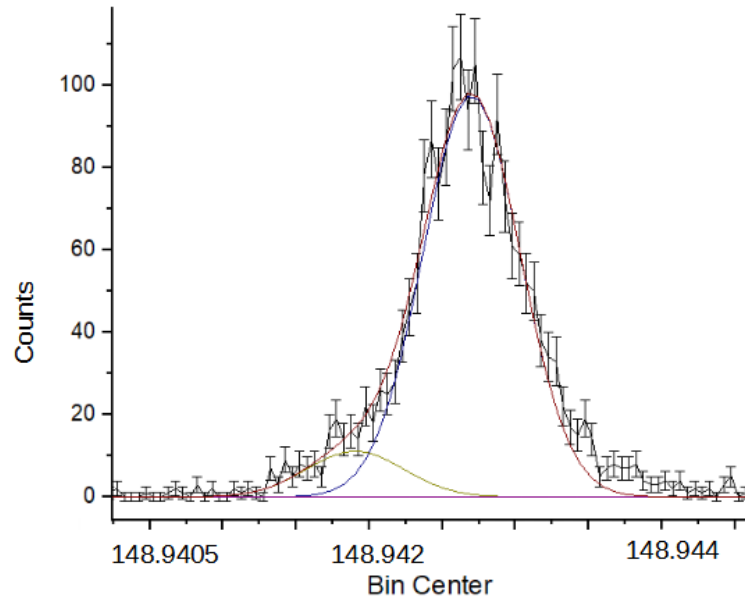


Figure 4.11: Gaussian fits of ^{149}Er and ^{149m}Er with the distance between them constrained by the known excitation energy which was measured in [102] via a 2 gamma ray internal transition and later confirmed by [103].

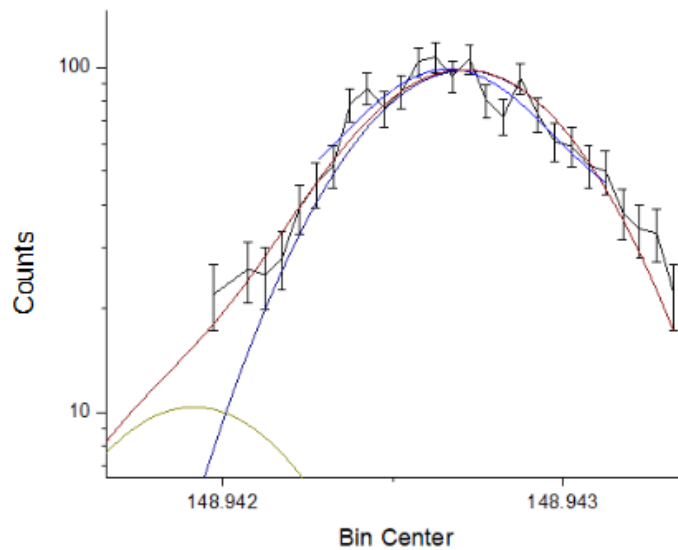


Figure 4.12: This log plot of the ^{149m}Er peak is fit after a cut is made at 20% of the peak height. This further reduces the influence of the adjacent ground state.

4.3 Newly Determined Mass Values

The mass data for the neutron-deficient lanthanide elements was obtained during two experimental runs, which took place in 2018 and 2019. The 2018 data primarily contains the masses of the Yb isotopes, and the 2019 data the masses of the Tm isotopes and the correction to the anomalous Er mass. The uncertainty was reduced in the later due to improvements made to the MRTOF settings. The measured masses are summarized below.

Species Measured	Measured Mass (AMU)	Measurement Uncertainty (AMU)	Calibrant	Calibrant Mass (AMU)	Calibrant Uncertainty (AMU)	Mass Ratio	Mass Ratio Uncertainty
^{157}Yb	156.9426823	5.8×10^{-5}	^{157}Tm	156.936973	3.00×10^{-5}	1.0000363793	3.70×10^{-7}
^{156}Yb	155.9427850	6.4×10^{-5}	^{156}Tm	155.938985746	1.53×10^{-5}	1.0000243638	4.10×10^{-7}
^{155}Yb	154.9457432	5.1×10^{-5}	^{155}Eu	154.922899847	1.34×10^{-6}	1.0001474500	3.29×10^{-7}
^{155}Dy	154.9257377	4.9×10^{-5}	^{155}Eu	154.922899847	1.34×10^{-6}	1.0000183180	3.16×10^{-7}
^{154}Yb	153.9463792	4.8×10^{-5}	$^{138}\text{Ce}^{16}\text{O}$	153.900908799	5.36×10^{-7}	1.00029545245	3.12×10^{-7}
$^{153}\textbf{Yb}$	152.9494386	5.0×10^{-5}	^{153}Dy	152.925771729	4.30×10^{-6}	1.0001547607	3.27×10^{-7}
^{152}Yb	151.9505210	5.5×10^{-5}	$^{136}\text{Ce}^{16}\text{O}$	151.902043875	3.48×10^{-7}	1.0003191342	3.62×10^{-7}
$^{151m}\textbf{Yb}$	150.9563755	5.7×10^{-5}	^{151}Er	150.937448567	1.77×10^{-5}	1.0001253958	3.78×10^{-7}
$^{150}\textbf{Yb}$	149.9585633	5.0×10^{-5}	^{150}Dy	149.925593068	4.64×10^{-6}	1.0002199108	3.33×10^{-7}
$^{150}\textbf{Tm}$	149.9501910	5.1×10^{-5}	^{150}Dy	149.925593068	4.64×10^{-6}	1.0001640674	3.40×10^{-7}

Table 4.1: A summary of the 2018 measurements of Yb atomic masses. **Boldface** font represents a previously unmeasured mass. The mass of the ^{151}Yb ground state was determined separately using an EMG analysis in [90].

Species measured	Measured Mass (AMU)	Measurement Uncertainty	Calibrant	Calibrant Mass (AMU)	Calibrant Uncertainty	Mass ratio	Mass Ratio Uncertainty
^{157}Lu and ^{157m}Lu	156.9502270	2.7×10^{-5}	^{157}Yb	156.942651368	1.17×10^{-5}	1.0000482699	1.72×10^{-7}
^{157}Tm	156.9369844	3.4×10^{-5}	^{157}Yb	156.942651368	1.17×10^{-5}	0.9999638915	2.17×10^{-7}
^{156}Tm	155.9390041	2.6×10^{-5}	^{156}Yb	155.942817096	9.99×10^{-6}	0.9999755490	1.67×10^{-7}
^{155}Tm	154.9392385	2.4×10^{-5}	^{155}Er	154.93321571	6.52×10^{-6}	1.0000388736	1.549×10^{-7}
^{155}Yb	154.9458561	2.5×10^{-5}	^{155}Er	154.93321571	6.52×10^{-6}	1.0000815858	1.61×10^{-7}
^{154}Tm	153.9416510	3.4×10^{-5}	^{154}Dy	153.92442892	7.98×10^{-6}	1.0001118864	2.21×10^{-7}
^{154}Er	153.9328096	2.6×10^{-5}	^{154}Dy	153.92442892	7.98×10^{-6}	1.0000544468	1.69×10^{-7}
^{153}Tm	152.9420831	4.4×10^{-5}	^{153}Dy	152.925771729	4.30×10^{-6}	1.0001066619	2.88×10^{-7}
^{153}Er	152.9350348	2.5×10^{-5}	^{153}Dy	152.925771729	4.30×10^{-6}	1.0000605725	1.63×10^{-7}
^{152}Tm	151.9446353	4.5×10^{-5}	^{152}Dy	151.924725274	4.93×10^{-6}	1.0001310520	2.96×10^{-7}
^{152}Er	151.9350348	2.9×10^{-5}	^{152}Dy	151.924725274	4.93×10^{-6}	1.0000678597	1.91×10^{-7}
^{151}Tm and ^{151m}Tm	150.9455444	2.9×10^{-5}	^{151}Dy	150.926191279	3.49×10^{-6}	1.0001282291	1.92×10^{-7}
^{151}Er	150.9374680	2.3×10^{-5}	^{151}Dy	150.926191279	3.49×10^{-6}	1.0000747166	1.52×10^{-7}
^{150}Tm	149.9501904	4.8×10^{-5}	^{150}Dy	149.925593068	4.64×10^{-6}	1.0001640636	3.20×10^{-7}
^{150}Er	149.9379390	2.5×10^{-5}	^{150}Dy	149.925593068	4.64×10^{-6}	1.0000823470	1.67×10^{-7}
^{149}Tm	148.9530058	5.0×10^{-5}	^{149}Dy	148.927327516	9.86×10^{-6}	1.0001724214	3.36×10^{-7}
$^{149m}\text{Er}^*$	148.9432630	3.1×10^{-5}	^{149}Dy	148.927327516	9.86×10^{-6}	1.0001070020	2.08×10^{-7}

Table 4.2: A summary of the 2019 measurements primarily of Tm atomic masses. **Boldface** font represents a previously unmeasured mass. The * indicates a re-measurement with a significant departure from the literature value.

A wide array of nuclear masses were measured over two distinct experimental runs. These included $^{150-157}\text{Yb}$, $^{149-157}\text{Tm}$, and $^{149-154}\text{Er}$. The masses of $^{149,150}\text{Tm}$, and $^{150,153}\text{Yb}$ were previously only as extrapolations in the Atomic Mass Evaluation. Additionally, the mass uncertainty of ^{151m}Yb was reduced from 300 keV and a separate analysis directly determined its excitation energy from the ground state by measuring both mass peaks [90]. This work confirmed the strong trend in the excitation energy of the single-hole isotones at $N=81$ that justified fixing the excitation energy when performing the fit of ^{149m}Er .

The uncertainty in the mass of ^{152}Yb was reduced from 150 keV, and a correction was made to the mass of ^{149m}Er and thus its corresponding ground state. This allowed for a definitive determination of the drip line location.

The correction to ^{149m}Er highlights the significant advantage offered by simultaneously measuring many isobars in the MRTOF, even if they have been previously measured.

Chapter 5

The Cooler Penning Trap (CPET)

As discussed in Chapter 2, there are several potential benefits to developing a Cooler Penning Trap and incorporating it into the TITAN facility. A dedicated trap to reduce the energy spread of the Highly Charged Ions (HCIs) extracted from the EBIT could improve the precision, speed, efficiency and isotopic range of Penning trap mass measurements conducted at TITAN. More generally, this technique is of broader interest in experimental physics, as it would provide a new technique with which to cool trapped HCIs. Evaporative ion cooling is a well-established cooling technique [104], but relies on evaporating the majority of a sample of trapped ions. Sympathetic cooling using a laser-cooled atomic species can cool HCIs sufficiently to produce Coulomb crystals [105][106], but the cooling rate is fastest when the species to cool has already fallen to thermal energy levels [107]. Sympathetic cooling with trapped electrons holds the potential to achieve rapid cooling of HCIs at 10s of eV, with the prospect of high ion throughput and ion transport efficiency that are not presently possible using other methods.

The purpose of TITAN's CPET is to dissipate the additional spread in kinetic energy that ions acquire as a result of the various heating mechanisms within the EBIT, before using them to perform a precision mass measurement within MPET. The quality of the TOF-ICR mass spectrometry within MPET can be negatively impacted by a large energy spread in several ways. Although higher charge states horizontally compress a resonance such as the one seen in Figure 2.13, larger variations

in the kinetic energy will make the TOF resonance less prominent. Charge breeding can increase this energy spread, and the corresponding reduction in a discernible change in time-of-flight of ions excited at their resonant frequency can even prevent the identification of a resonance. Additionally, a reduced energy spread can also improve the ion transport efficiency into MPET. A larger fraction of ions can be injected with an appropriate energy for trapping, since the energy must be matched to the floor of the potential well. Lastly, if ions have a smaller kinetic energy inside the measurement trap, this helps to minimize the possibility of systematic shifts in the cyclotron frequency due to trapped ions probing regions further from the centre of the trap where the magnetic field becomes less homogeneous.

The beam emittance is defined by the starting temperature of the ion bunch upon ejection from the EBIT, and is a conserved quantity for particles travelling through the conservative field of the beam optics when under vacuum. It is important to minimize the longitudinal beam emittance because it creates a trade-off between the energy spread of the ion bunch delivered to MPET, and the spread in its time-of-flight. *Kicking* the bunch (i.e. applying a steep potential gradient to rapidly extract the ions from the EBIT), can therefore be used to reduce the TOF spread, but will, in turn, increase its energy spread ¹. Since MPET can only trap ions that are centred in the trap when it closes, a large TOF spread will critically reduce the transport efficiency to the trap. On the other hand, a large energy spread allows the ions to probe regions far from the centre of the measurement trap, which can cause systematic shifts in the determined masses.

Although the energy spread of ions that reach MPET can be reduced by trapping ions with a shallower potential, thereby restricting acceptance to ions with the ideal energy, many of the most interesting ‘exotic’ nuclear species that TITAN studies are primarily limited by the rate at which ions can arrive at MPET for mass measurements. When planning an experiment these rates need to be carefully considered for each species in order to assess the feasibility of the measurement. Unlike offline facilities which study stable species, and thus have quantities of atoms on the scale of

¹A *bunch* can include as little as a single ion, so the energy spread must be considered in terms of many identical transport cycles from the EBIT to the Penning trap

Avagadro's number available, TITAN cannot afford to be extremely selective about the kinetic energy of ions that are transported to MPET. It is not unlikely to expect as few as 10 ions per second of a nuclide under study to arrive at MPET under favourable conditions! Therefore, by enhancing the ion acceptance of MPET, CPET could enhance both the rate, and the isotopic range of possible mass measurements in MPET.

Therefore, in order to efficiently transport HCIs through TITAN with a small energy spread, it is necessary to somehow minimize the temperature of the trapped ions through either a dynamic or a dissipative process.

5.1 Sympathetic Cooling for Highly Charged Ions

The TITAN facility is unique for being the only Penning trap facility in the world currently employing an Electron Beam Ion Trap (EBIT) to quickly remove additional electrons from the singly-charged short-lived isotopes produced by an Isotope Separator On-Line (ISOL) facility. Penning trap mass spectrometry of short-lived isotopes stands to benefit significantly from utilizing highly charged ions, because the precision of mass measurements is limited by the mass-to-charge ratio m/q . TITAN therefore utilizes an EBIT to charge breed the low energy ion bunches produced in the RFQ from the continuous ISAC beam.

However, the process of *electron impact ionization* which generates these high charge states, as well as a handful of related processes that take place within an EBIT [83], can result in significant heating of the ions² (e.g. Electron Beam Ion Trap, In-Flight production, etc.). It has been observed that it is possible for ion bunches exiting the EBIT to have been heated to 20eV by interactions with the electron beam. Data to support this fact was collected with the TITAN EBIT by Ish Mukul, and is shown in Figure 5.1.

The Cooler Penning Trap (CPET) has therefore been under development to ad-

²Since measurements often involve as few as one ion per cycle, *temperature* in this context is better thought of as a measure of the reproducibility of ion kinetic energies from one cycle to the next, than as an equilibrium thermal distribution

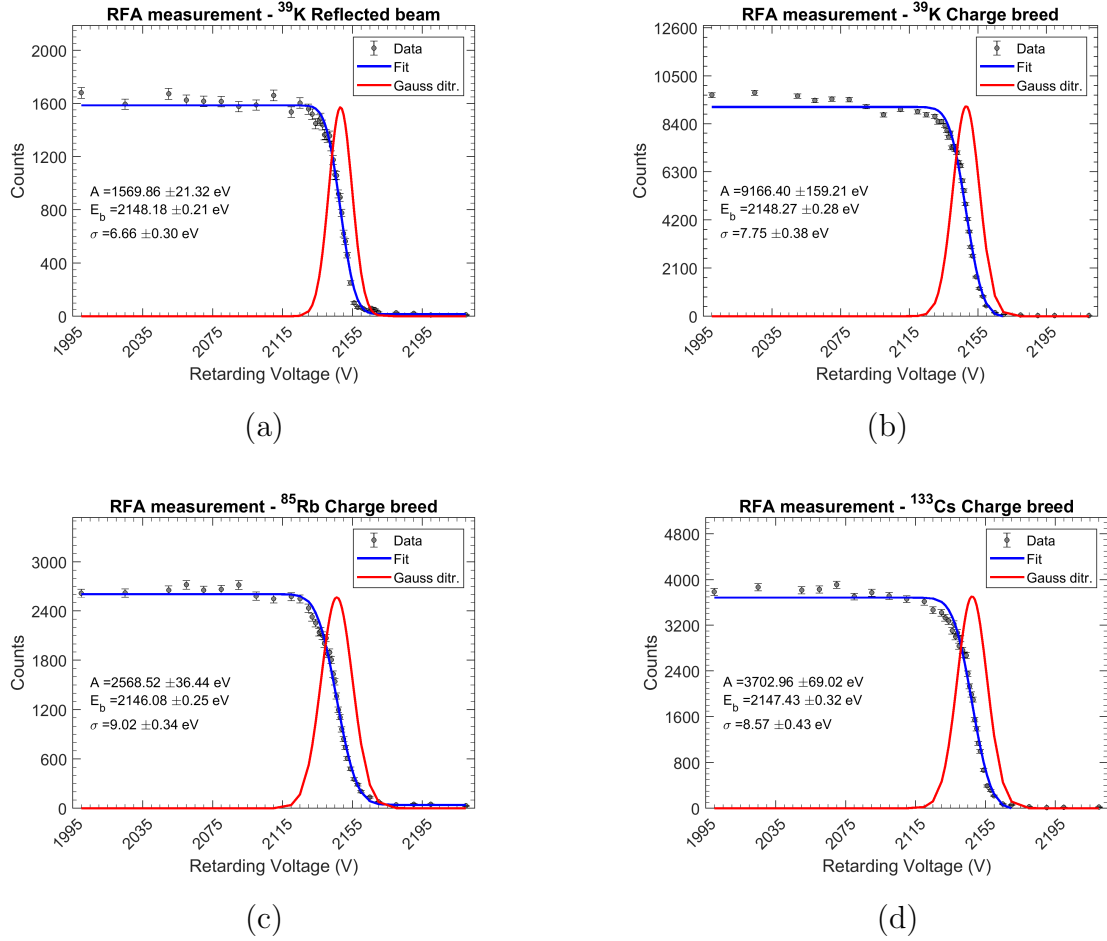


Figure 5.1: The energy spread of ions exiting the EBIT was previously probed using a *retarding field analyzer* to scan over a range of blocking potentials for the ions. Images courtesy of Ish Mukul.

dress the issue of the energy spread of charge-bred ions exiting the EBIT. Significant experimental efforts have been directed towards the realization of sympathetic cooling of highly charged, radioactive ions using electrons³. The benefits, developments, and challenges of this method will be discussed below.

An appropriate cooling mechanism is therefore required in order to meet the various demands of rapidly cooling the unstable, highly charged ions studied at TITAN.

³This technique may be referred to as simply *electron cooling*, which should not be confused with the process of the electrons themselves cooling by emitting synchrotron radiation.

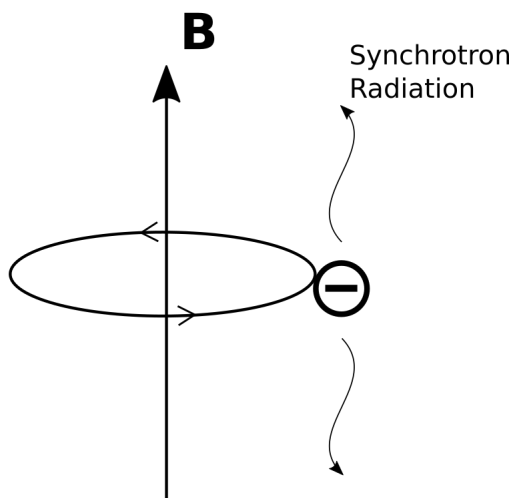


Figure 5.2: Electrons orbit the magnetic field in accordance with the left-hand rule, and emit synchrotron radiation in the plane perpendicular to the acceleration vector.

A successful cooling technique should offer the following qualities to mass measurements:

- Fast cooling - needed if used for exotic species ($t_{1/2}$ could be as low as ~ 10 ms) or high throughput is needed.
- Efficiency - to handle species with low abundance.
- Reliable, continuous operation - to accommodate the limited time window of experiments.
- Preservation of the highly ionized states.

This last item is of particular importance, and creates a unique constraint. Unlike the MRTOF, which utilizes an internal Radio Frequency Quadrupole (RFQ) trap to prepare ions in a bunched, low kinetic energy state, standard RFQ cooling procedure cannot be used to cool HCIs. This is because RFQ traps rely on dissipating the kinetic energy of the ions via *buffer gas cooling* in which the ions lose energy through collisions with molecules of an inert gas. However, if this technique was applied to HCIs the electrons in such a gas would promptly recombine with the ions and destroy

their high charge states, due to the large ionization energies/electron affinities of these highly-stripped ions.

Alternatively, if TITAN's experimental focus was on stable atomic beams, utilizing macroscopic samples of elements, evaporative cooling could be employed to quickly cool the ion sample. Evaporative cooling is an effective cooling mechanism that allows the high energy tail of a population of ions to escape the trap. However, this would be far too inefficient for beams containing only a small number of exotic isotopes as it necessarily discards the majority of a sample.

Having eliminated these techniques, there remains the use of *sympathetic cooling* with charged particles, by which a colder charged species cools a hotter one by the Coulomb interaction, and laser cooling. Conventional laser cooling is not an option since appropriate optical transitions are a special case which exist for only a small fraction of HCIs. Sympathetic cooling allows a cooler charged species to provide a reservoir that can sink the excess kinetic energy of the hotter species as the particles collide with one another. There exist several different possible configurations for sympathetic cooling.

One means of sympathetic cooling that has been considered for CPET is a reservoir of cold, positive ions which could dissipate kinetic energy from the HCIs [1]. H^+ could serve as an effective coolant that could be trapped in the same potential as HCIs. Although cooling with protons would prevent any recombination with electrons, its implementation poses several challenges. Being much heavier, protons will not radiate synchrotron radiation quickly enough to cool, so evaporative cooling would need to be used to prepare the ions in a cool state. Alternatively, a hybrid laser/sympathetic cooling approach could be applied using a positive ion with an optical transition, such as Be^+ , that can be cooled via laser [106]. A significant issue with utilizing a positive ion coolant lies in the fact that it has the same charge as the ions that need to be cooled; extraction of the HCIs would also likely purge the previously prepared coolant ions from the trap. In this case, such a coolant would need to be trapped and cooled for each cooling cycle of the radioactive ions. This would require very intense beams of H^+ ions or any other ion that is selected for cooling, in order to trap and cool them very rapidly in between cooling the bunches of HCIs.

5.2 The Electron Advantage: Benefits of Sympathetic Cooling Using Electrons

Due to the aforementioned constraints, sympathetic cooling with a plasma of free electrons been proposed as the most promising method for cooling HCIs at TITAN. Electrons offer several notable advantages as a sympathetic cooling particle:

- Electrons are relatively straightforward to produce in large quantities.
- Free electrons recombine with ions much more slowly than atomic electrons.
- Electrons self-cool in high magnetic fields.
- The negative charge of electrons makes them potentially reusable.
- Electrons are more efficient and cost-effective than positrons to produce.

In particular, a major advantage of using electrons for cooling in CPET is their ability to radiate away their thermal energy when held in the 7 T field of the trap's superconducting magnet. This in principle, makes it possible to continually cool the system without employing laser cooling, buffer gas, or a tank circuit.

The electrons must also conduct thermal energy away from the ions by interacting with them. As shown in Figure 5.3, ions having a non-zero kinetic energy can be trapped in the same region as the electrons by applying the appropriate potentials. The negative charge of electrons requires that the trap use *nested potentials* in order to allow for their spatial overlap with positively charged ions. They can then interact by Coulomb scattering to lose their energy to the colder electrons.

It is important that the charge state of most of the ions remains in-tact. Perhaps surprisingly, a free electron charge-exchanges with an HCI more slowly than does a buffer gas, which leads to a survival time that could allow cooling to take place [1]. For all these reasons, and since electrons are straightforward to produce using a simple thermal source composed of a tungsten filament, this makes them a very attractive cooling medium for HCIs.

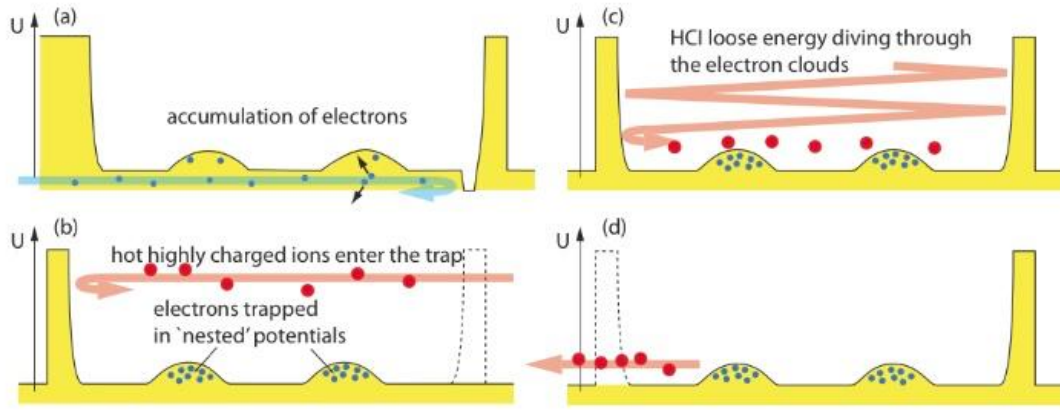


Figure 5.3: A conceptual illustration of how cooling can be performed with oppositely-charged species by implementing a nested potential configuration. In a) the negatively-charged electrons cool into regions of more positive potential and in b) the positively-charged ions are trapped in an overlapping region. The ions are shown losing energy to the electrons in c) via Coulomb scattering and in d) they are shown to be extracted from the far side of the ion trap when they have given up a sufficient amount of energy. Image courtesy of Gerald Gwinner.

5.2.1 Timescale of Electron Self-Cooling

There are two timescales which are intrinsic to using electrons as a coolant particle. The first is the time it takes for the electrons to cool, and the second is the time it takes for the ions to lose their kinetic energy to the electrons by means of the Coulomb interaction. Electrons cool within CPET primarily by emitting synchrotron radiation (also known as cyclotron radiation for the non-relativistic case), and the time constant of this process can be determined as follows:

The Larmor formula gives the power radiated by an accelerating, non-relativistic point charge (in SI units)[108]:

$$P = \frac{2}{3} \frac{q^2 a^2}{4\pi\epsilon_0 c^3}.$$

Here, q is the charge, a the acceleration, c the speed of light, ϵ_0 the permittivity of free space, and the relationship $c^2 = \frac{1}{\epsilon_0\mu_0}$ is employed. The Lorentz force acting on

an electron for which the motion is dominated by the magnetic field is given by:

$$\vec{F} = e(\vec{v} \times \vec{B}),$$

where e is the elementary charge. By applying $\vec{F} = m\vec{a}$ we obtain

$$\frac{2}{3} \frac{e^2}{4\pi\epsilon_0 c^3} \left(\frac{evB}{m_e} \right)^2$$

for the component of the velocity that is orthogonal to the magnetic field. The electron velocity v can then be expressed in terms of its kinetic energy as $2E_k/m_e$ giving

$$P = \frac{2}{6} \frac{e^4 E_k B^2}{\pi \epsilon_0 c^3 m_e^3},$$

and since $P = \frac{dE}{dt}$ the cooling timescale can be determined by integration to yield:

$$t_{cooling} = \frac{3\pi\epsilon_0 c^3 m_e^3}{e^4 B^2} \ln |E_k| \Big|_{E_{init}}^{E_{final}} = 0.0527 \times \ln\left(\frac{E_{init}}{E_{final}}\right) \text{seconds}. \quad (5.1)$$

Taking the definite integral from the initial kinetic energy of the electron to the final energy allows for an estimation of the expected timescale of cooling. For example, when cooling electrons from 50 eV to 1 eV, a timescale of about 206 ms is expected. This should be adequate, especially when the possible reuse of electrons from cycle to cycle is considered.

This is consistent with the timescale necessary for cooling radioactive ions, but it would be preferable to reuse a single electron bunch for several cooling cycles rather than loading and cooling a new electron plasma for each ion bunch. This would help to achieve a high throughput of ions to MPET, and thereby reduce the overall measurement time.

The time it takes for the ions to cool in a bath of ions depends not only on the Coulomb scattering, but also on the capacity of the electrons to sink this heat. Therefore, a large number of electrons is needed to provide an adequate thermal reservoir. Figure 5.4 from [1] shows the predicted sympathetic cooling curves for two extreme cases of electron-cooled HClIs (C^{6+} and U^{92+}).

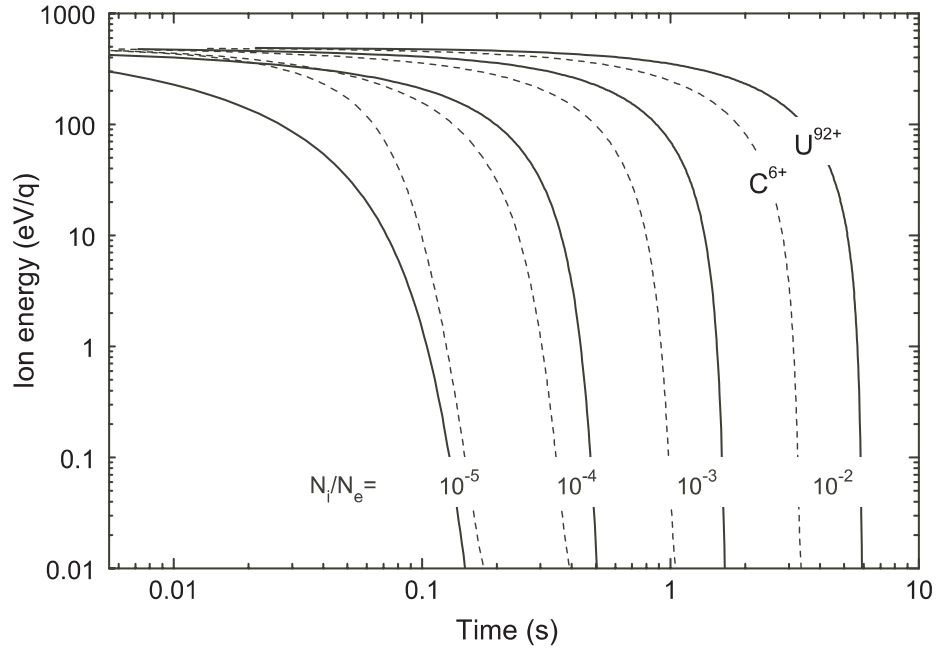


Figure 5.4: Predictions for the sympathetic cooling of ions with different ratios of ions to electrons. Image from [1] and reproduced with permission from Springer Nature.

5.3 Design of CPET

The aforementioned requirements to achieve electron cooling of HCIs informed the design of the Cooler Penning Trap (CPET). An operational cooling trap will need to collect electrons in a nested potential well, cool the electrons, accept a bunch of ions, cool the ions through interaction with the electrons, and ultimately extract the cold ions on the opposite side of the trap from which they entered.

Electrons and ions are radially confined in CPET within a uniform, 7 Tesla magnetic field produced by a cryogenic superconducting magnet. The magnetic field also serves the critical purpose of removing kinetic energy from the trapped electrons by cooling them through the emission of synchrotron radiation. The 7 Tesla field allows for a cooling time constant of $\tau \approx 0.07$ seconds for the electrons [1].

The trap electrodes are mounted within a vacuum pipe, which itself is mounted and aligned within the bore of the magnet. The vacuum pipe is lined with a titanium getter material that can be activated with heat in order to improve the vacuum

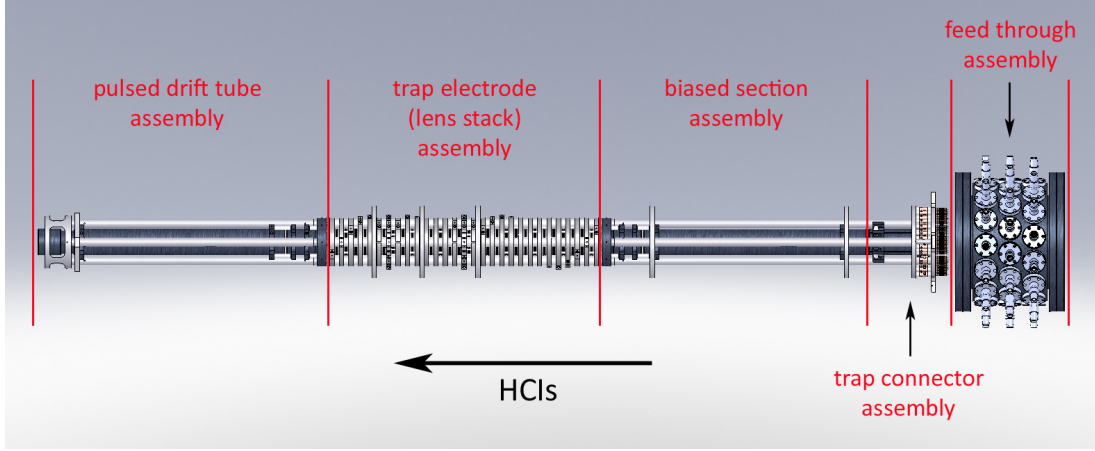


Figure 5.5: An image of the CPET trap electrodes and drift tubes, which are housed within the superconducting magnet, and the feedthrough section which is located outside the magnet. From [131].

conditions.

CPET is designed to have enough independently biasable electrodes to capture and simultaneously hold two different species of charged particles with opposite charge. In order to do so, *nested* potential wells must be shaped in which one species is trapped within a local extremum inside the larger trapping region of the other. Figure 5.5 shows the 29 gold-plated trap electrodes, and the two drift tubes for changing the ion energy. The thicker gate electrodes are located at the two ends of the trap electrodes, positioned between them and the drift tubes. Together these comprise the electrodes used for control of electric potentials within the CPET magnet. A number of the cylindrical trap electrodes are also divided into two or eight segments in order to allow the application of either dipole or octopole fields (see Figure 5.6), which allows the use of rotating-wall compression or other RF manipulations of the ions. However, a limited number of vacuum feed-throughs on the feed-through assembly restricted the number of electrodes that could be independently biased, and the number of feed-throughs and biasing wires being utilized in the vacuum was further reduced to help prevent discharges as discussed in section 5.5.3.

For the purposes of ion tests in this thesis, a stable thermal ion source was installed on the upstream (EBIT) side of CPET, which is shown in Figure 5.7, and in context



Figure 5.6: An 8-segmented electrode (octopole) was included with the intention of using it for rotating-wall compression of ions. However, many segments have now been shorted to reduce the complexity of the electrical connections and feed-throughs, making quadrupoles the highest order trap electrode configuration currently in use.

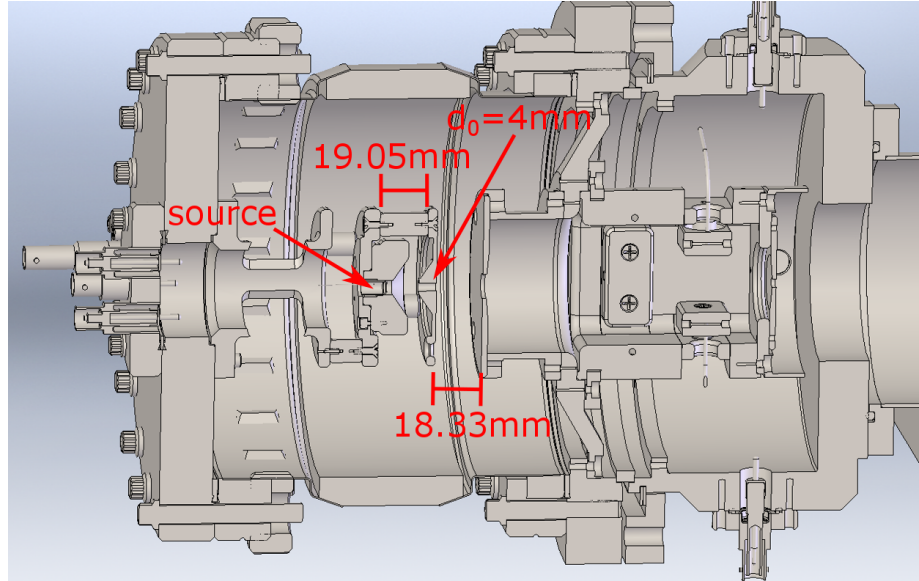


Figure 5.7: The stable ion source used to deliver thermally ionized positive ions (including Na) for CPET tests.

of the optics in Figure 5.8. The ion source is designed to produce a beam of surface-ionized singly-charged Na ions, which are then steered through the corresponding ion source assembly and delivered to CPET. The assembly uses two sets of x-y steering

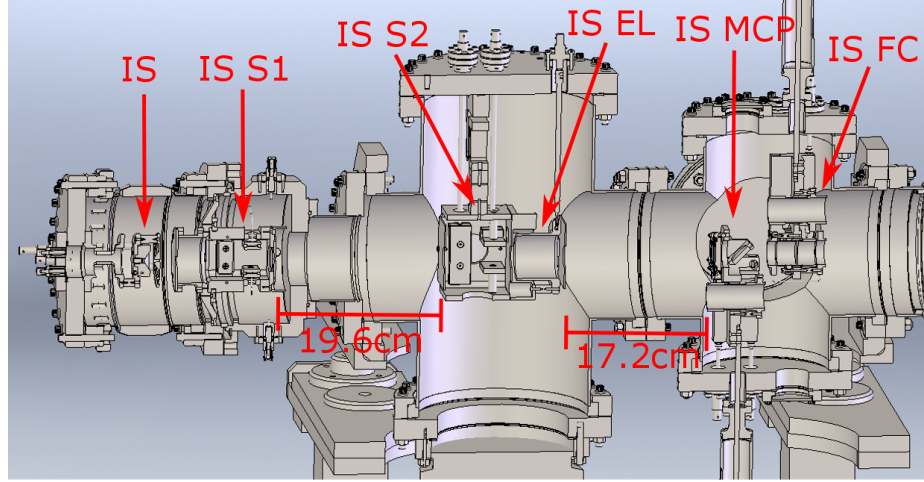


Figure 5.8: An image of the stable ion source used for CPET tests in context of the current CPET optics. Surface ions generated at the source (IS) are steered with two sets of steerers (IS S1/S2) and focused using an Einsell lens (IS EL). The microchannel plate detector (IS MCP) can be inserted to characterize the beam before it enters the trap, and the Faraday cup (IS FC) can be inserted to measure continuous beams of electrons transmitted from the far side of the trap.

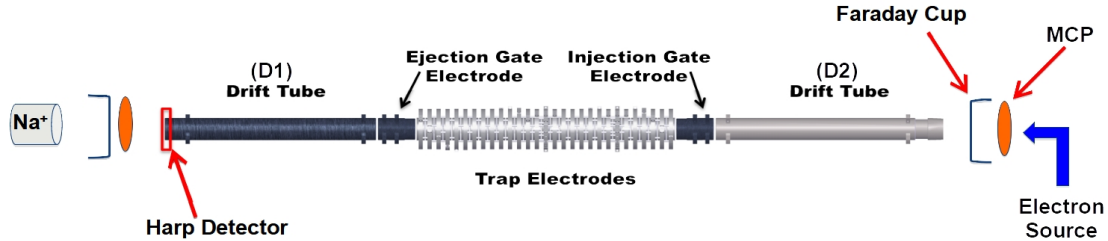


Figure 5.9: Electrodes and detectors surrounding the Cooler Penning Trap (CPET). Ions are injected from the left side of the image and extracted on the right.

elements and an electrostatic lens to prepare the ion beam for injection into CPET.

Efforts were made to optimize the ion injection into CPET both through experimental tests as well as simulations. Simulations indicated that it is best to avoid large net potentials on each of the axially-separated X and Y steering elements, in order to prevent radial asymmetries in the beam from arising later within the magnetic field of the trap.

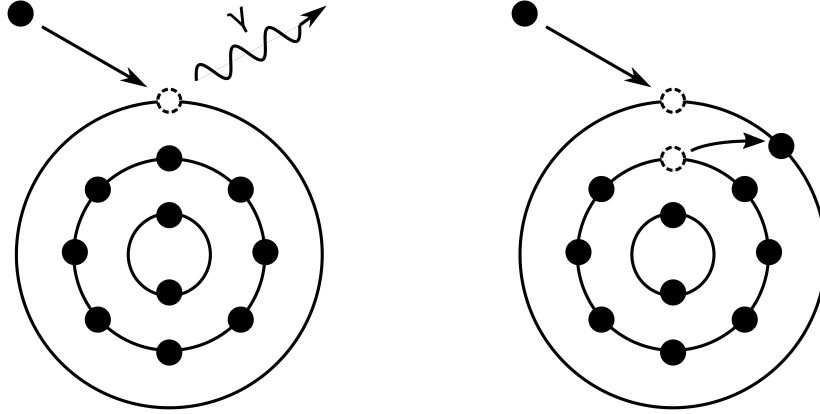


Figure 5.10: Radiative recombination of electrons (left) vs dielectronic recombination (right). Being a resonant process, the dielectronic channel can occur much more quickly, but requires the energy gained by capturing an electron to match an available transition energy of an orbital electron.

5.3.1 Possibility of Electron Recombination in CPET

One challenge, unique to cooling HCIs is that electron-ion recombination could destroy the charge state of the ions. It is essential that the cooling process in CPET preserves the charge state of the ions being cooled. However, highly-charged ions could potentially recapture electrons from the background gas or the free electron plasma used as coolant. Free electrons are being used as a coolant because recombination is much slower than for a buffer gas, but this mechanism must be considered.

The processes which allow for recombination with the free electrons and thereby destroy the high charge states of the ions include *dielectronic recombination*, *three-body recombination*, and *radiative recombination*. The recombination processes that could affect the operation of a cooler Penning trap are discussed in Ke et al. [1]. Radiative recombination involves the capture of a single electron by the ion, while

a photon is radiated away to maintain the energy balance. In contrast, dielectronic recombination is a resonant process that requires the energy of an excited electronic state of the highly charged ion (after recombination) to be matched to the energy change of the electron recombination. These two processes are compared in Figure 5.10. Three-body recombination relies upon a second electron to be involved in the process and is unlikely except in very dense trapped electron plasmas.

The electron recombination rate, $\frac{dP(t)}{dt}$, is related to the electron density, n_e , by

$$\frac{dP(t)}{dt} = (\alpha_{RR} + \alpha_{DR} + \alpha_{TBR})n_e, \quad (5.2)$$

where $P(t)$ is the fraction of ions that remain. The radiative recombination rate coefficient is given by

$$\alpha_{RR} = 5.2 \times 10^{-14} Z_{\text{eff}} \sqrt{\frac{E_{\text{inf}}}{T_{\text{eff}}}} \left(0.43 + \frac{1}{2} \ln(E_{\text{inf}}/T_{\text{eff}}) + 0.469(E_{\text{inf}}/T_{\text{eff}})^{-1/3} \right) \text{cm}^3 \text{s}^{-1}, \quad (5.3)$$

where E_{inf} is the ionization energy of the ion ($E_{\text{inf}} \approx 13.6q^2$ eV), T_{eff} is the effective temperature of the electron-ion collision ($T_{\text{eff}} = T_e + (m_e/m_i)T_i$), and the three-body coefficient is given by

$$\alpha_{TBR} = [2.0 \times 10^{-27} \text{cm}^6 \text{s}^{-1}] q^3 T_{\text{eff}}^{-4.5} n_e. \quad (5.4)$$

However, the dielectronic recombination rate, α_{DR} , is more complicated to predict.

In the case of ions that are not fully stripped of electrons, resonant dielectronic recombination processes could potentially enhance the rate of recombination. Unfortunately, there is not a general formula for the dielectronic recombination rate of ions, since it depends on the electronic structure of the particular ion. Both the atomic number and the charge state will affect whether such a transition is available. Although the precise conditions for dielectronic recombination are unlikely to be met by a given species, this mechanism for the destruction of the charge state must be considered when cooling highly-charged ions.

The time scale of radiative and three-body recombination is expected to exceed the required timescale for cooling radioactive ions. When these recombination processes have been considered for electron densities $n_e > 10^7 \text{cm}^{-3}$, more than 95% of fully

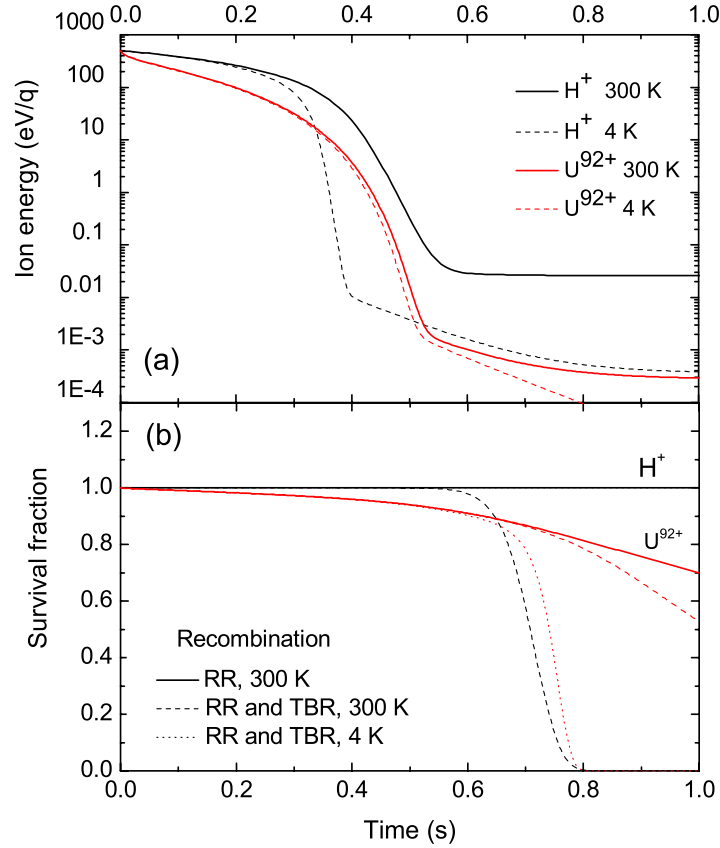


Figure 5.11: Predictions for the sympathetic cooling of ions over time, showing the ion energy and the fraction of the ions that have not been lost to radiative or 3-body recombination (RR and TBR). Image from [1] and reproduced with permission from Springer Nature.

ionized ions will retain their original charge state after 500 ms for any elements that can be studied at ISAC (see Figure 5.11). This represents a worst case scenario for the recombination processes that are likely to occur.

Therefore the time limits imposed by the need to quickly cool species with short radioactive half-lives, as well as the need to maintain reasonably short measurement

cycles, will likely pose the more demanding constraints on the cooling time. Recombination is not expected to destroy the charge state of a significant fraction of the HCl⁺s being cooled by CPET.

5.3.2 Vacuum Requirements in CPET

Although the recombination rate of HCl⁺s with free electrons is expected to be low within CPET, it is also possible for HCl⁺s to lose their high charge state by capturing electrons from residual background gas. This makes management of the pressure within a Penning trap critical for storing HCl⁺s. As such, MPET and CPET have the most stringent vacuum requirements at TITAN. Vacuum pressures down to $< 10^{-9} - 10^{-10}$ Torr are achieved by implementing a rigorous ultra high vacuum cleaning procedure. CPET presently employs three turbomolecular pumps backed by an additional turbomolecular pump which is, in turn, backed by an oil-free scroll pump.

To further improve the vacuum conditions when highly-charged ions are trapped in CPET, the vacuum pipe which houses the trap electrodes is lined with a titanium getter material. This getter can be activated using an electric heating blanket that surrounds the tube while installed within the magnet.

5.3.3 Status of Sympathetic Electron Cooling

Sympathetic cooling with electrons (or positrons) has been employed in a few specialized settings including antimatter research [109], and storage rings (as an electron beam) [110][111], but to date, it has not been used to cool radioactive ions. Aspirations to carry out this technique on highly-charged ions are currently under development at the HITRAP facility located at GSI Darmstadt [112][113].

The purpose of CPET is to achieve cooling of HCl⁺s with a focus on the fast cooling times needed for radioactive isotopes. This chapter outlines the current status of TITAN's CPET, including its recent upgrades. The characterization of ion and electron trapping, the merging of the two different species within the trap, and early evidence of an interaction are discussed.

5.4 Technical Challenges of Electron Cooling

Several technical challenges are inherent to commissioning a cooler trap as described above.

Functional operation of the trap requires the switching of voltages (sometimes in the kV range) in regions of high magnetic field. In the presence of dense plasmas, this creates conditions that enhance the risk of runaway Penning discharges occurring.

The physical space constraints within the bore of the magnet significantly limit the instrumentation that can be incorporated near the trapping region. Also, the trap must operate under strict ultra-high vacuum (UHV) conditions in order to preserve the high charge states of the ions. Any modifications that need to be made to the system are therefore non-trivial. Furthermore, manipulating two different charges of particle increases the complexity of the electrodes, voltages, and control systems.

A large charge density of electrons must be trapped to satisfy several constraints on the efficiency and timing for the ultimate operating conditions of the trap. Since the electrons are produced outside of the trap, they must be injected with high kinetic energy in order to overcome reflections of the light electrons from the magnetic field gradient. In order to inject electrons efficiently several modifications were made to optimize the trajectories of the incoming electron beam, including extending the length of the biased trap drift tube toward the electron source.

5.4.1 Penning Discharge

A significant challenge to trapping oppositely-charged ions in a nested trap is its susceptibility to Penning discharges. Electrical discharges can create electromagnetic noise within a Penning trap [114], significantly increase the experimental background, and can even cause damage to equipment [115]. Traditionally, electrical arcing due to static electric fields in vacuum (glow discharge) is assumed to be reasonably well described by Paschen's law. The DC breakdown voltage is given by

$$V_b = \frac{B p d}{\ln(A p d) - \ln \left[\ln \left(1 + \frac{1}{\gamma_{se}} \right) \right]}, \quad (5.5)$$

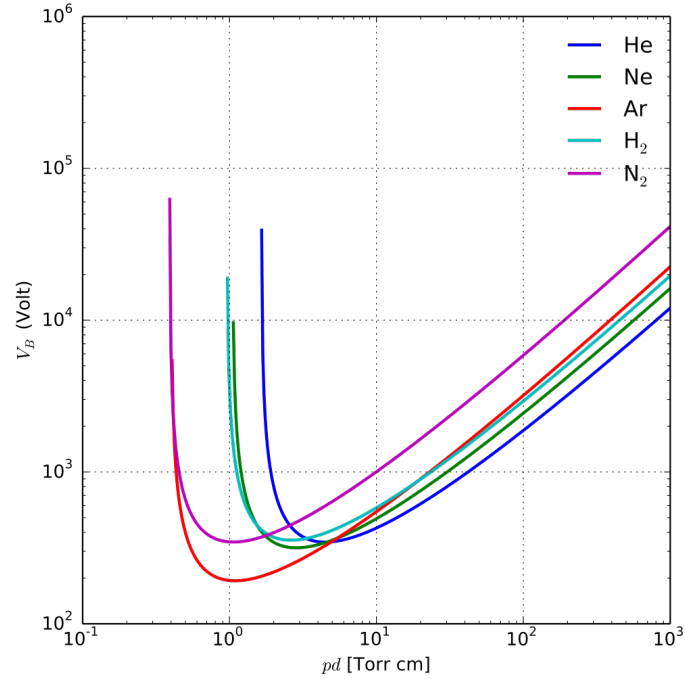


Figure 5.12: The Paschen curve shows the expected electrical breakdown voltage (V_B) vs. the product of pressure and separation distance (pd) for various gasses. Paschen curves are shown here for several gasses, when no magnetic field is present. See https://commons.wikimedia.org/wiki/File:Paschen_curves.svg for original image and source code. CC BY-SA 4.0 Krishnavedala.

where B and A are gas-dependent constants, p is the pressure (Pascal), d is the distance between surfaces (cm), and γ_{se} is a coefficient of secondary electron production[116]. The relationship between the breakdown voltage and the gas pressure is known as the *Paschen Curve*, and can be seen in Figure 5.12 for various gasses. A consequence of this relationship is that electrical discharges are greatly suppressed when UHV conditions can be maintained.

Electrical discharges can be initiated by the presence of free charge carriers in the vacuum. The presence of the electron plasma in CPET, as well as fast switching of electrode potentials, make electrical breakdown a cause for concern. For this reason, CPET was carefully designed with ceramic spacers to maintain sufficiently large gaps between conductors to prevent the electric field from inducing discharges within the

vacuum. However, it is also well-known that for certain conditions in the vacuum, an increase in the magnetic field can exacerbate the problem of discharges, substantially increasing the discharge current over a range of vacuum pressures [117].

Discharges can occur when the difference in electrostatic potential between the negative cathode and the positive electrostatic maximum allows the accelerated electrons to ionize the background gas. If the kinetic energy of the free electron is greater than or equal to the ionization potential of a gas particle (~ 10 eV), then an orbital electron can acquire sufficient energy to be liberated from the atom or molecule with a single collision.

In the absence of a magnetic field, an electron liberated from a negatively charged electrode, or from a gas atom within the vacuum, will simply follow the electric field gradient until it strikes a more positive surface. This is because the mean free path length of the electrons under UHV conditions ($\sim 10^{-9}$ Torr in CPET) is long. When an electron ionizes a residual gas molecule, at least one additional electron as well as one positively charged ion is produced. The free ion will then be accelerated by the electric field towards an electrode held at a relatively negative potential, and the electron(s) toward an electrode having a positive bias relative to the position at which the ionization occurred. As the charged particles accelerate towards the electrodes, they can ionize the background gas. When the ion strikes the surface of the negative electrode, yet more electrons can be released and accelerated into the vacuum. As long as the average electron generated in the vacuum produces more than one additional electron, this creates a positive feedback loop and the conditions are satisfied to initiate a self-sustaining discharge known as a *Townsend Discharge*. This process is illustrated in Figure 5.13.

Only a single charged particle is needed to trigger (or ‘seed’) the avalanche discharge. However, even when no free charge carriers are initially present in the trap to seed the discharge, discharges can still be initiated under Ultra High Vacuum (UHV) conditions by cosmic rays striking the electrode surfaces, as well as any other high-energy processes such as radioactive decay. Furthermore, rough surfaces or sharp edges can create localized points which have above-average electric field strength. These fields can then overcome the electronic work function at lower voltages than

expected. If any of these conditions create free charged particles at any instant, a continuous Townsend discharge can begin to take place.

It has long been known that high magnetic fields can lower the ignition voltage and increase the current associated with a discharge in vacuum [118][119][120]. Under certain conditions, an avalanche effect known as a *Penning discharge*[121] can be triggered, even at UHV pressures and relatively low voltages [122]. A Penning discharge can occur if the vacuum contains a minimum in the electrostatic potential which overlaps with the field lines of a sufficiently strong magnetic field. It then becomes possible for electrons to become trapped along these magnetic field lines. When electrons are trapped in a potential minimum (such as when they are accumulated for cooling in CPET), the distance traversed by the trapped electrons is greatly extended. A trapped electron can oscillate along the magnetic field lines within the potential minimum long enough to eventually collide with a particle of the low-pressure background gas. This, in turn, lowers the voltage and/or the pressure at which an electrical discharge can take place. The mechanism is illustrated in Figure 5.14.

5.4.2 Plasma Behaviour of Trapped Electrons

Both electrons and ions can be trapped within a Penning trap, but the cyclotron frequency of the lighter electron is higher than that of the ion in proportion to the fractional difference in their masses. However, at the electron densities needed for effective cooling, collective behaviour of the self-interacting, non-neutral, electron plasma cloud is also a factor. When the electrons no longer behave like isolated trapped ions, collective motions can emerge, and may give rise to various forms of instabilities.

Specifically, the $m = 1$ diocotron mode has been observed to often dominate the motion of the trapped electrons [123]. The cylindrical column of trapped electrons rotates about the central axis of the trap (at a radius much larger than the Larmor radius), due to the electrons interacting with their image charge on the trap wall.

Various techniques have previously been developed to control the properties of

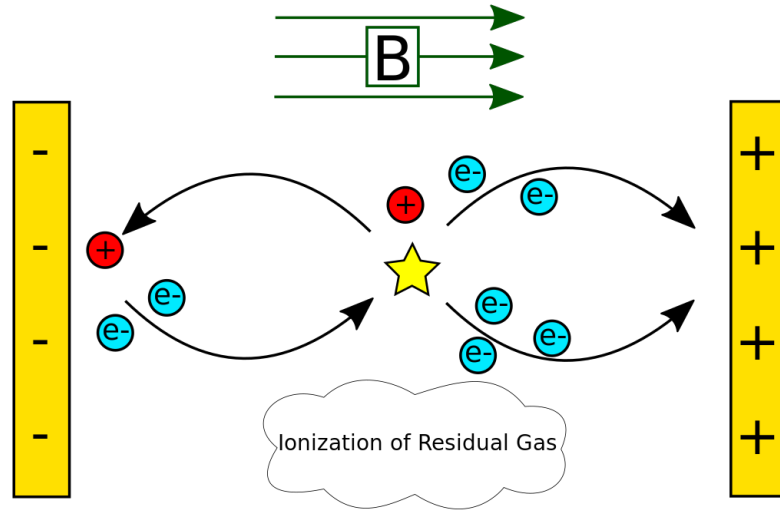


Figure 5.13: Mechanism of a Townsend discharge. The stronger the magnetic field is, the more electrons are restricted to traveling along the magnetic field lines. This increases the probability that on average, each liberated electron will generate at least one additional electron, thereby initiating a runaway discharge.

plasmas within Penning traps. It has been shown that the density of an electron plasma can be precisely controlled (for the purpose of controlling the overlap of positrons with anti-protons) by applying a combination of rotating-wall compression and evaporative cooling [124]. The diameter of an electron plasma can be controlled by the voltage that accelerates the electrons into the trap [125], and radio frequencies can also be applied to reduce the diocotron radius [126]. There is also interest specifically in studying two-component (electron and ion) plasma in a Penning trap by combining the two species using nested potential wells [127][128]. The study of trapped plasmas within Penning traps remains an area of active research.

The complicated behaviour inherent to trapped electron plasma makes these Penning traps a rich area of study with many subtle intricacies that need to be considered.

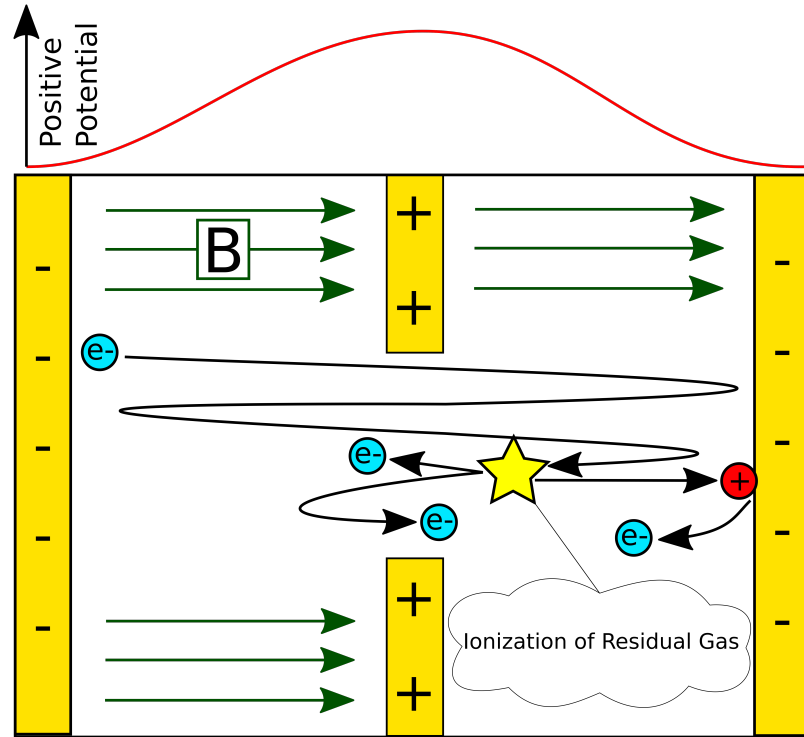


Figure 5.14: Mechanism of a Penning discharge. Stronger magnetic fields restrict electrons to travelling along the magnetic field lines. This increases the probability that each liberated electron will generate, on average, at least one additional electron, thereby initiating a runaway discharge.

5.5 Hardware Improvements to CPET

Initially, the nature of many of the processes inhibiting successful operation of the trap were poorly understood. Several important lessons were learned regarding the trapping of electron plasmas inside Penning traps. In addition to improvements to the controls and diagnostics needed to accommodate the complexities of trapping and merging two oppositely-charged species, greater control over the voltages in the trap were needed. A series of critical improvements made to the system are therefore outlined below.

5.5.1 An Improved Electron Source

One challenge of implementing effective electron cooling in CPET is the need to quickly collect large numbers of trapped electrons in order to achieve cooling on a reasonable timescale. The greater the density of trapped electrons, the more quickly the ions can be cooled. Furthermore, if too few electrons are used, the flow of energy from the ions to the electrons can begin to appreciably increase the temperature of the electron plasma, thereby delaying the cooling of ions, or even evaporating the electrons. Appreciable heating of the electrons would limit ion cooling to the timescale of electron synchrotron emission ($\tau_{1/2} \approx 0.07$ s). Therefore, a large number of electrons must be trapped quickly enough to not significantly extend the total measurement time of the ions under study.

This requires electron beam currents of up to ~ 1 mA, having a minimal transverse component of the velocity, to prevent reflections upon entering the magnetic field. These currents have been observed to be sufficient to overload power supplies, overwhelm sensitive detectors, and even heat the walls of the vacuum chamber within CPET. Additionally, the presence of electrons increases the probability of initiating Penning discharges. Therefore, the electron beam needs to be well-controlled, allow for reasonable rate of electron injection, and ideally, provide the option of being turned off whenever not needed for injection into the trap.

Additionally, cooled ions must also be able to continue downstream from CPET to be detected on an MCP. This requires a mechanism to produce the electron beam off of the ion beam axis, and then bend it back onto the axis. Control of the bent electron beam was first attempted by constructing an off-axis electron source that utilized a curved bending electrode and a steering element, both segmented into 4 quadrants, to bend the beam emitted from an electron source 90 degrees and aligned onto the ion beam axis (see Figure 5.15).

This initial off-axis steering setup was plagued by poor injection efficiency, and required additional features to be added. The 4-segmented bending and steering electrodes proved insufficient to overcome the Lorentz force imparted by the fringe field of the magnet (on the order of ~ 100 Gauss) when being bent by 90 degrees onto

the beam axis. Even the side of the vacuum chamber was observed to be warmed by the mis-steered electrons as seen in the thermal camera image pictured in Figure 5.16. The electron source has been subsequently upgraded to a more sophisticated electron gun for which the optics was thoroughly motivated by simulations. The electron beam is now intense enough to accumulate a sufficiently large number of electrons within CPET on an appropriately short time scale for cooling radioactive ions [129].

The new off-axis electron source has several advantages over the initial design. The improved electron source positions the filament or cathode within a Wehnelt cylinder with a tapered Pierce geometry within which the electrons are accelerated towards the anode [129]. This configuration produces a focused electron beam for steering onto the ion beam axis. Additionally, the anode has been divided into four independently biasable segments with the spacing maintained by a Macor[®] ceramic insulator. By dividing the anode into four separate segments, the anode can direct the electron beam into the magnetic field at an angle that further compensates for the Lorentz force, thereby preventing it from striking electrodes when being steered on axis. These improvements lead to high electron beam intensities, improved transmission onto the ion beam axis, and ultimately faster loading into the trap.

Furthermore, the Wehnelt cylinder also provides a mechanism whereby the electron beam can be pulsed on and off in order to reduce stray electrons and their associated background in the vacuum. When the voltage on the Wehnelt cylinder is reduced by 70 V, emission from the tungsten filament was sufficiently suppressed to eliminate any detectable electron emission.

The electron source can use either a hot tungsten filament, or a BaO disc cathode to produce the electrons, the former being observed to operate more stably and also being far more tolerant to venting of the vacuum chamber. The majority of tests were done using the hot filament, but the use of a BaO cathode can improve vacuum conditions.

Some representative values for electron transmission after the updated electron gun was installed on the off-axis source is here given for a DC beam injected into CPET at a typical electron gun emission current of 440 μA . After bending this beam onto the ion beam axis, the improved transmission resulted in 270 μA as detected on

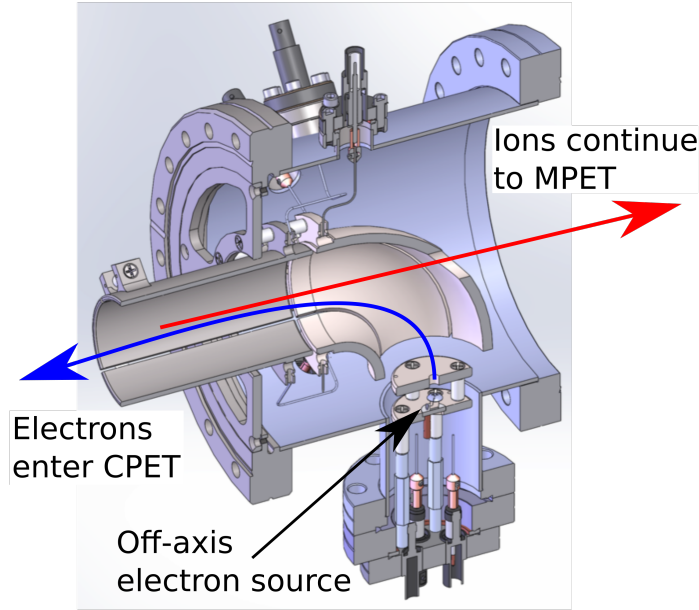


Figure 5.15: Electrons from the off-axis electron source are steered onto the ion beam axis using the potentials applied to the electron source anode, a curved bending element, and a steering element, all three of which consist of four independently-biased segments in order to compensate for the fringe field of the magnet. The off-axis positioning of the source allows ions from the cooler Penning trap (CPET) to continue through a hole in the bending electrode, to the measurement trap (MPET).

the electron-side Faraday cup located in the fringe field of the magnet, and about $3 \mu\text{A}$ inside the magnet on the far side of the trap, detected using the harp detector as a current sink, which will be discussed later in this chapter.

Upon injection into a magnetic trap, magnetic compression will result in the radius of the electrons being compressed in proportion to the ratio of the field inside the trap, to the field where the electron beam begins, outside the trap.

5.5.2 A Modification for Detecting Trapped Electrons

One obstacle that hampered electron detection efforts was that when a large number of electrons are trapped, they can be observed to execute diocotron motion inside the trap. This takes the plasma column slightly off the central axis of the trap,

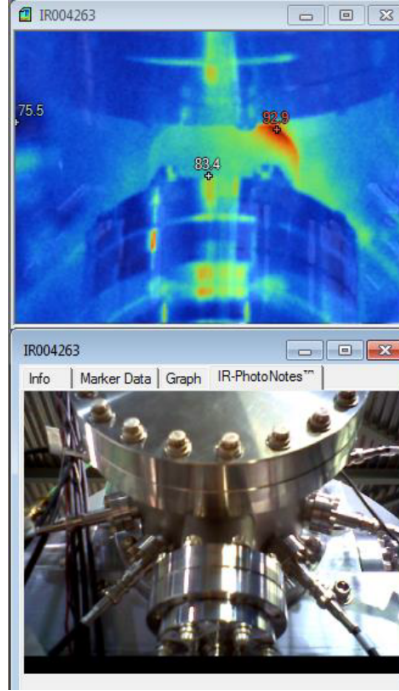


Figure 5.16: A thermal camera image shows heating of the vacuum chamber due to poorly steered electrons. Shown here is the housing the off-axis electron source photographed from just downstream and below the beam axis. Before installing an improved electron gun, electrons bent by the magnetic field preferentially heated one side of the vacuum chamber, despite electrostatic steering.

and any radial offset of the electron plasma from the axis is amplified by a factor of $\sqrt{\frac{B_{trap}}{B_{detector}}}$ as it exits the CPET magnet to the detector along the diverging magnetic field lines [123].

Two different methods were used to detect electrons that had been trapped. The first type of detector for trapped electrons successfully implemented in CPET was a phosphor screen placed inside the DT-1 drift tube. This allowed for a direct observation of the diocotron orbit of the trapped electrons [123][130], and the damping of the plasma radius could be clearly observed. These tests revealed that electrons could be centred within CPET.

However, a long-term solution for electron detection needs to be located well-within the magnetic field of the trap, while still allowing for the transmission of ions.

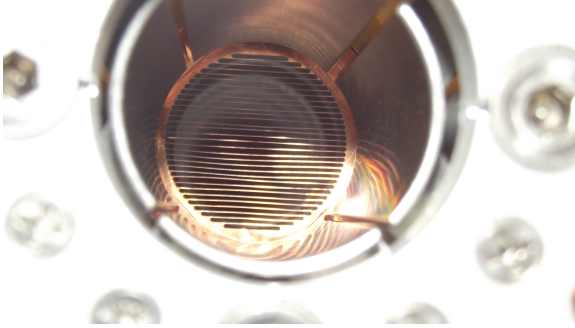
To this end, a custom-made, photo-chemically etched, copper detector with a harp-like geometry was developed and is shown in Figure 5.17. It consists of 0.1 mm detector wires separated by a 1 mm pitch. This was essential to measure the quantity of trapped electrons while still allowing ions to pass through CPET. It functions like a simple Faraday cup, detecting the deposited charge of a bunch of electrons on a copper electrode. However, the small cross-sectional area of the harp-like geometry allows the majority of ions to be transmitted when the harp is biased to the drift tube potential in that region. Details can be found in reference [131].

After steering the beam emitted by the electron gun onto the beam axis, transmission through the trap needed to be confirmed on the harp. Both the DC electron beam and the accumulated electron bunches could be detected on the harp by simply grounding it through an oscilloscope without any charge pre-amplification. However, detection efficiency could be improved by applying a more positive potential to the harp.

Detection of the electron beam and resulting trapped electrons on the harp was facilitated by an amplifier circuit which allows the harp to measure an electron signal while a positive high voltage bias is applied to it (700 V). This provides increased flexibility in the potential configuration, since detection of cooled electrons requires the harp to be located at the bottom of a potential well. The active isolation amplifier circuit makes it possible to measure the potential across a floating 100 k Ω through a readout at ground potential. The signal of the electron bunch on the harp can then be detected on an oscilloscope or other data acquisition system. Zener diode protection was added to prevent damage to the amplifier from voltage spikes due to deposited electrons or the pulsing electrodes.

5.5.3 Minimizing Penning Discharges

Although the Penning discharge is a known effect [114], it was not known how it might impact the co-trapping of ions and electrons within CPET. At just over 4 T, TITAN's MPET had experienced electrical discharges, but these did not prevent it from operating. However in CPET, the effect of the discharges on the electrode



(a) Harp detector installed between the injection steering electrodes (S3) and the drift tube (DT1).



(b) Harp detector shown within the Macor® ceramic mount

Figure 5.17: Figure a) shows the harp detector (also referred to as the *mesh* detector) mounted within the injection electrode structure, just downstream from where ions pass through the steering electrodes just downstream from the SHV feed-through section. Figure b) shows the harp detector within its ceramic (Macor®) mount as presented in [131].

power supplies, the EM radiation in the vacuum, and on the ion signal detected on the final MCP detector necessitated that these discharges be suppressed.

In order to minimize the persistent challenges with Penning discharges observed in CPET, several steps were taken. Discharges become more likely wherever 2 different voltages with a large potential difference, or a rapidly switched voltage are in close proximity to one another. This required reconfiguring the positions of the copper wires which bias the electrodes within the trap, and the electrodes themselves, in order to keep the highest voltages as well as the switched voltages well-separated from each other over the entire path from the trap electrodes to the high-voltage vacuum feed-throughs. Also, electrically connecting the segments of several electrodes to one another allowed for the complexity of the wiring to be reduced. Figure 5.18 shows the re-stacking of electrode segments during reassembly of the trap in a configuration designed to minimize high-voltage discharges, and Figures 5.19 and 5.20 show



Figure 5.18: Electrodes within CPET were reconfigured and/or shorted to minimize the possibility of Penning discharges.

the former and revised layout of the trap electrodes. The old and new configuration of the wires which bias the CPET electrodes is shown in figure 5.21. Furthermore, the ceramic insulators and conducting wires were carefully positioned within the vacuum to prevent electrical arcing between any two surfaces. The electron detection harp/mesh spacing was increased due to it occupying a region that was especially subject to discharges. The physical conditions of the trap would be more than adequate in the absence of a magnetic field, but discharges were nevertheless observed due to the increased probability of electrical discharges within the 7 T magnetic field.

Every time components were added to the trap or modified in any way, rigorous UHV handling and cleaning procedures were followed. This regimen of cleaning and pumping protocols prevented the introduction of any volatile substances to the UHV system, and allowed us to maintain a vacuum pressure in the range of 10^{-10} Torr as measured at the vacuum gauges. After each round of modifications, high-voltage resistance testing was done between all possible pairs of the biased elements in CPET, including ground.

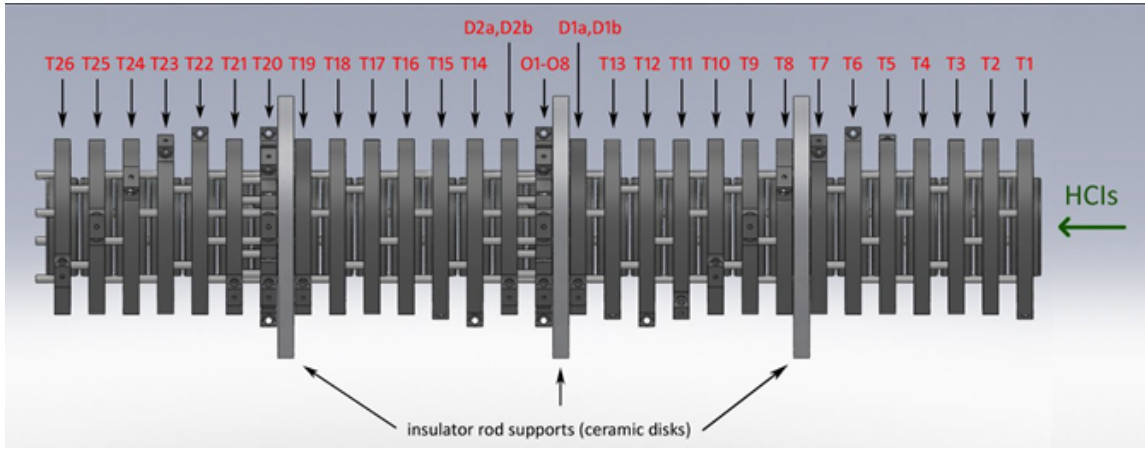


Figure 5.19: The previous CPET electrode configuration before improvements were made. Image from [131].

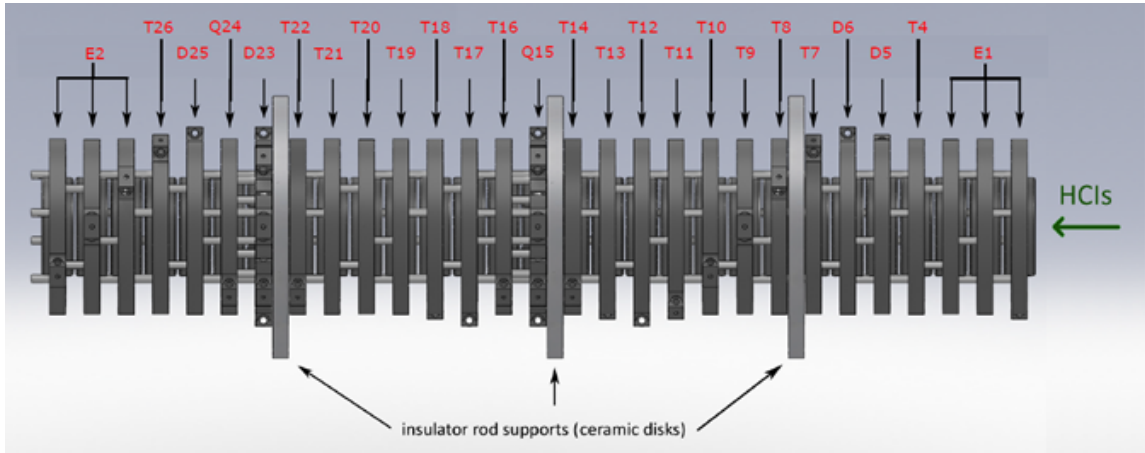
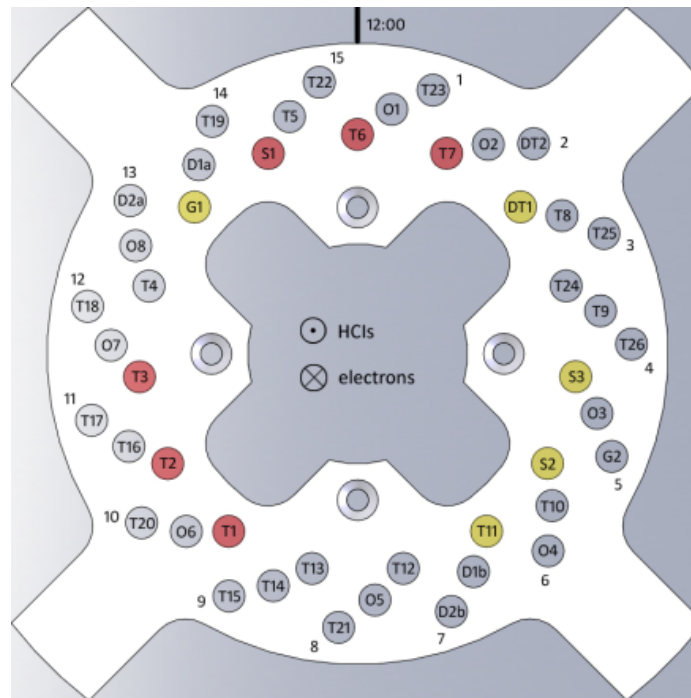


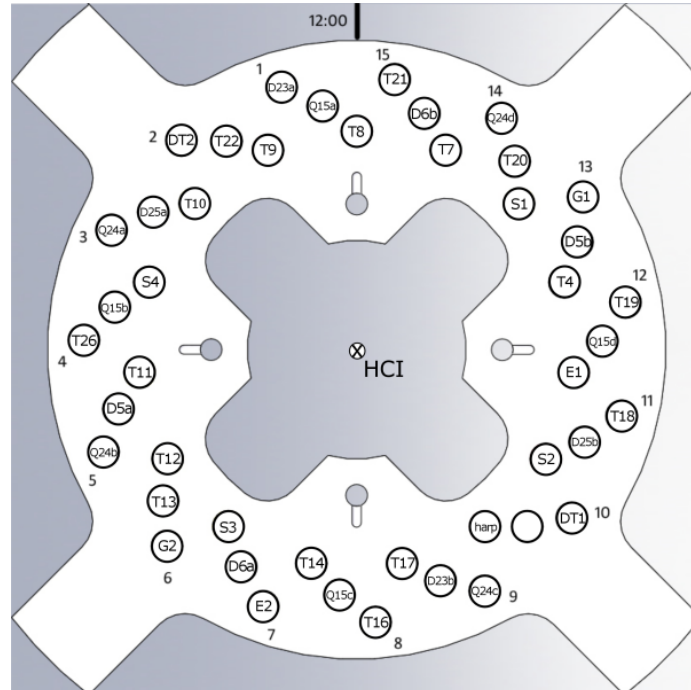
Figure 5.20: The new configuration of the CPET electrodes maximized the distance between higher-voltage, and switching electrodes. Shorting the end-cap electrodes to one another helped to simplify the setup, and also prevented electric field penetration from seriously reducing the trapping potential. Image modified from [131].

5.5.4 Slowing Electrode Switching

The fast switching of the electrodes was problematic, since it created noise on the harp detector and the MCP, and also increased the likelihood of discharges. The switching speed was therefore reduced by adding additional resistance and filtering



(a)



(b)

Figure 5.21: Old wiring (a) vs the new wiring (b) for biasing the CPET electrodes. Image courtesy of Stefan Paul.

to the switching electrodes to slow the changing voltage. This suppression of the high frequency components of the switched voltages produces a cleaner, more reliable signal on the detectors. For instance, noise was minimized on the harp by adding filtering between the upstream, outermost trap electrode (i.e. the *Injection Gate Electrode* shown in Figure 5.9) and the switching box, and by including an additional $3.15\text{ M}\Omega$ resistance in series with this low-pass filter. This made it possible to reliably detect the negative electron signal above the switching-induced background signal.

5.5.5 High Voltage DAC box

Critical to applying and manipulating nested potentials, a high-voltage Digital to Analogue Converter (DAC) box was developed to shape the axial potential in the trap using the cylindrical electrodes. The DAC box was designed to independently bias 16 channels up to about 180 V (with hardware capacity for more channels), with the option of operating all channels at a common floating voltage up to 800 V above ground.

Voltage settings are triggered using TTL pulses delivered to four different input channels to a Raspberry Pi computer, each of which triggers a distinct set of voltage settings that are configured in software. The software allows for gradual switching of the voltages by performing incremental voltage changes, for which the number of steps are defined by the user in the software. Any change to the voltage settings is immediately sent to a floating 32-channel evaluation board via a one-way fibre-optic link that communicates using the Serial Peripheral Interface (SPI) protocol. The evaluation board is made by Analog Devices, and is capable of outputting 0-200 V⁴. A single channel responds to an input in about $180\text{ }\mu\text{s}$ and all 16 channels can respond to an input signal in about $500\text{ }\mu\text{s}$. A $12\text{ k}\Omega$ resistor limits the current on each output, and a 1000:1 voltage divider is also included for each output to monitor the voltages supplied to the electrodes and bleed off any excess charge. The output is accurate to better than 0.1 V, improving for the lower voltages. The software also allows voltages

⁴The maximum operating voltage of the channels is about 10% less than 200V due to the range lost when calibrating the channels.

to be changed more gradually, over several steps in order to make it less likely to initiate a discharge or transfer energy into the trapped particles.

The DAC box substantially improved the configurability of the trap by augmenting the handful of 2-state high voltage switches that were previously available. It proved to be an essential tool to control the axial electrostatic potential in the trap in order to successfully manipulate the two species of charged particles in CPET and to test their properties.

5.6 Developments Toward Achieving an Electron-Ion Interaction

The most basic requirement to achieve an exchange of kinetic energy between the ions and electrons is for them to occupy the same spatial region. This will rely on using the trap electrodes to produce a *nested* potential configuration in order to allow the oppositely-charged species to overlap. The position of the electrons along the axis depends on the axial potential as defined by the voltages applied to the trap electrodes. This was made possible by using the DAC biasing box and other power supplies to apply voltages to the appropriate ranges of electrodes.

In addition to the axial position, the two species must overlap radially. Ideally, the electrons and ions will be centred inside the trap. Although the electrons have a small orbital radius due to their mass, they may not be centred within the electrodes. The alignment of the trap with the magnetic field, the injection steering and focusing potentials, and the plasma effects of the electrons can all result in an uncentred electron plasma. In particular, the diocotron mode was found to result in a collective motion of the electron plasma. This results from the interactions between all electrons and the induced charge on the electrode walls, and causes the entire the electron cloud to orbit around the central axis of the trap wall.

Switching cycles on the electrodes must allow electrons to be accumulated within the trap efficiently, while still allowing for ions to be trapped as well. Additionally, some means of measuring the effect of the interaction between the ions and electrons

is necessary. Cooling could either be characterized by measuring a compression in the TOF from CPET to a detector, or by using a Retarding Field Analyzer (RFA) that scans over a range of blocking potentials to test what range of energies are contained in the extracted ion bunch. To this end, a *microchannel plate detector* (MCP) was installed on the downstream side of CPET for sensitive ion detection.

5.6.1 Electron Injection Into CPET

In order to establish effective sympathetic electron cooling, electrons must be accumulated in CPET as quickly as possible. The first step to inject electrons into CPET is to transport a continuous (i.e. DC) electron beam through the trap. To tune the electron beam, a continuous electron beam current of $\sim 1\text{mA}$ is large enough to detect on Faraday cups on both the near and far sides of the trap when the transmission has been well-tuned. This allows for the transmission to be optimized through the anode, onto the ion beam axis, and through the trap to the Faraday cup on the far side. After optimizing transmission, a continuous electron beam of $\sim 100\text{ }\mu\text{A}$ can be transmitted into the trap with $\sim 1\%$ efficiency (due to reflections off the magnetic field) to deliver $\sim 1\text{-}2\text{ }\mu\text{A}$ to the harp detector within the magnet after the losses due to reflections from the magnetic field.

Unless they are generated within the trap using RF [132][133] or by laser excitation of residual gas, the electrons must next be trapped within CPET. While it is straightforward to dynamically trap ions by rapidly raising an electrode voltage in order to close the trap when an ion enters the desired region (before the ion can exit), this is not the case for electrons. Due to the relatively low mass and corresponding greater velocity of the electrons, trapping large numbers of electrons requires an alternative loading mechanism.

Experimental tests have revealed that the fastest loading mechanism for injecting a sufficient density of electrons from outside the trap, appears to require the electrons to scatter off of one another in order for them to become trapped behind the axial electrostatic potential barrier. Although electrons are expected to traverse the trap in less than a microsecond, the number of electrons in the trap could often increase

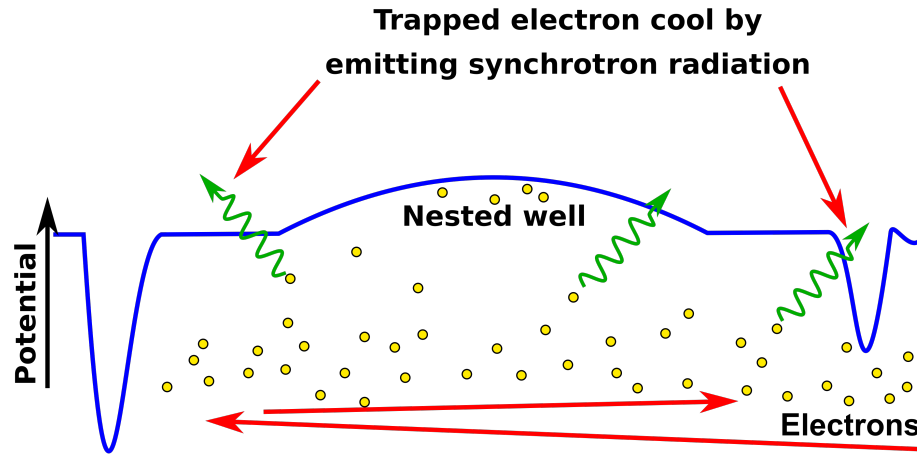


Figure 5.22: Electrons were trapped by scattering them behind the gate electrodes. They will subsequently cool into the potential minimum of this region by emitting synchrotron radiation.

over the course of a few seconds, and was found to depend strongly on the intensity and tune of the incoming beam. This is explained by collisions between the electrons which allow some of the scattered particles to lose enough energy from the axial degree of freedom to become trapped behind the potential barrier [134]. Rapid accumulation of electrons therefore depends sensitively upon injecting a high current of electrons into the trap to increase the rate of electron-electron scattering.

Effective accumulation of dense electron plasmas was therefore accomplished in CPET by creating a potential configuration in which the electrons first lose energy to electron-electron scattering to become trapped behind the potential barrier of the second gate electrode, and then cool into the nested potential well, as shown in Figure 5.22. They can then cool into a local potential minimum by emitting synchrotron radiation in accordance with equation 5.1.

The plasma could be quantified by detecting the trapped charge on the harp detector. Using the injected electron current of $\sim 100 \mu\text{A}$, an integrated charge signal of $1.8 \mu\text{Vs}$ could be detected on an oscilloscope after approximately 1 second of electron loading. This amounts to 1.8 pC or about 1.1×10^7 electrons that can be routinely accumulated in approximately 1 second.

5.6.2 Electron Trapping Lifetime

One important result of tests in CPET is the trapping of a sample of electrons within the trap for several minutes.

Online Penning trap measurements of exotic isotopes must conduct experiments within the limited time-frames dictated by beam availability. This demands an efficiently conducted experiment, with an acceptably short total cycle time. This is both due to the fact that the precision depends on the number of TOF data points collected during the experiment, as well as the fact that short-lived species can have half-lives down to $< 10\text{ms}$. This is sufficiently short-lived to potentially reduce the availability of exotic species, since they can decay before being measured if the cooling cycle is too long.

In order to effectively cool ions, electrons must be trapped for a sufficient amount of time for an ion to cool. However, in addition to this, the time spent loading electrons should be minimized in order to maximize the duty cycle of measurement to total cycle time and gather the highest number of statistics possible. Section 5.6.1 addresses how the speed at which electrons are loaded was enhanced, but ideally electrons would be used and reused for many cooling cycles. To this end, the persistence of electrons in the trap has been established in CPET through studies of the evolution of the quantity of electrons over time [135]. A scan of the number of detected electrons as a function of trapping time is shown in Figure 5.23. The data shows that after an initial jump, the number of trapped electrons can be seen to decrease over time as electrons escape the trap. The initial increase in the number of detected electrons can be explained as an initial reduction of the diocotron radius that allows a greater number of electrons to reach the harp detector (vs. getting steered into trap electrodes). The initial data indicated that while electrons slowly escape from the trap as expected, they have a half-life of just under a minute inside the trap. Since then, the lifetime has been extended to well over 3 minutes for certain tunes of the axial potential. This suggests that it may be possible to use the same electrons for many cooling cycles in the trap.

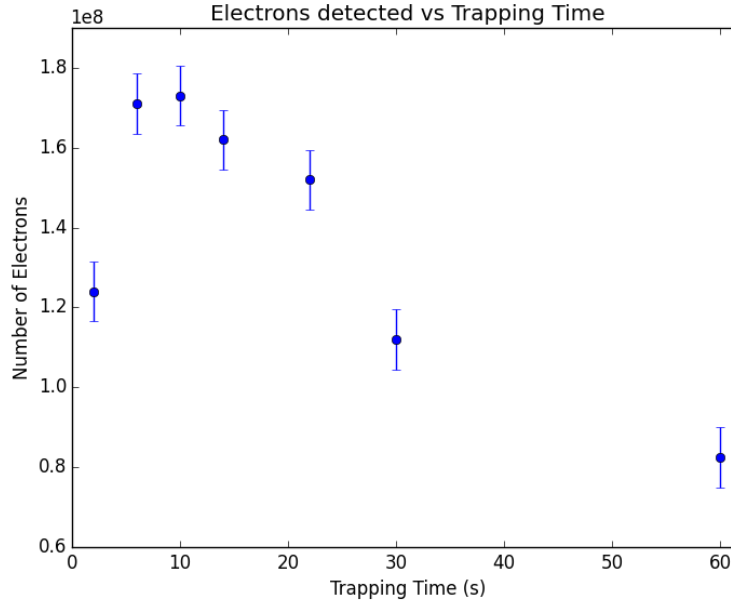


Figure 5.23: This plot shows the number of electrons for various electron trapping times as determined from the average charge deposited on the detector during an early electron trapping test. Electrons were detected on the copper harp detector by biasing it to the highest positive potential inside the trap (i.e. ground in this test). The initial rise in signal is believed to be the result of the diocotron motion damping to smaller radius. This plot shows that more than 10^8 electrons were detected, and that their half-life in CPET is slightly under one minute. This data that was previously published in reference [135].

5.6.3 Evidence for Electrons Cooling

An important initial result was the preparation and characterization of a cold electron plasma with which to cool ions. It is necessary to establish the presence of cold electrons within the trap which will provide a sink for the thermal energy of the ions.

Cooling Electrons Within a Shaped Potential Well

Since the axial magnetic field of the trap constantly accelerates the trapped electrons in circular orbits, they will cool by emitting synchrotron radiation if trapped for a sufficient duration.

Although it was established in section 5.2.1 that the electrons should cool quickly enough, it is not immediately obvious how much coupling to expect between the various degrees of freedom. The overall timescale of electron cooling was therefore established by observing the settling of electrons into a potential minimum,

A non-trivial potential configuration in the trap is required to trap store electrons and ions together/ Preparation of the electrons within a nested potential well was therefore achieved using the multiple-channel DAC box as a high-voltage source to shape the potential landscape produced by the trap electrodes shown in Figure 5.9.

In the simplest electron accumulation procedure, incoming electrons are first trapped by scattering behind the outermost electrostatic potential barrier on the downstream side of the trap (the Injection Gate Electrode). Figure 5.22 shows the relative voltages for loading a beam of hot electrons. Injection from the electron source was then blocked in order to allow the electrons to cool by emitting synchrotron radiation. Finally, the electrons were released from the upstream side of the trap and detected on the harp detector.

It was however, also necessary to explicitly demonstrate that the plasma effectively cools to the needed kinetic energy. In order to show this, the main blocking electrode on the upstream (harp) side of the trap was opened after the electrons had time to cool. The 4th, 5th, and 6th electrodes from the left were raised to a higher potential than the others, and the electrons would then settle into this region as they lost their kinetic energy. Only electrons which remained trapped behind the remaining negative potential on the first three electrodes from the left of the trap would then remain. When this final potential was then lowered to below the potential of the nested well, the remaining electrons could exit the trap, and a detectable electron signal could be measured on the harp detector. The magnitude of the signal was determined by subtracting the integrated signal from cycles without electrons from the same number

of cycles with electrons.

A relative voltage of 100 V was applied to the three trap electrodes producing the nested well, and the electrons were first stored between the two negatively-biased gate electrodes on the outer edges of the trap for a duration of seconds. Since the gate electrode on the extraction side was open for several hundred milliseconds before unloading the nested trap, it could be confirmed that the electrons are losing their kinetic energy to settle into this potential minimum [135].

5.6.4 Co-Trapping of Ions with Electrons

A fundamental step in accomplishing sympathetic cooling of ions with electrons is to arrange for them to occupy the same physical space. Co-trapping of oppositely-charged particles poses some intrinsic complexities. Although both positive and negative species are confined radially by the magnetic field (but rotating in opposite directions), the electrostatic potentials that are attractive to a positive ion are repulsive to an electron, and vice-versa.

After extensive technical improvements to CPET enabled the successful trapping of both electrons and ions individually, modifications needed to be made to the system and its operation to create a nested potential configuration that is able to simultaneously trap both species in the same region. This required developing a several step trapping cycle:

- Accumulate electrons
- Allow electrons to cool
- Dumping any hot electrons from the trap that did not cool into the local positive potential well
- Inject ions.

Furthermore, to confirm that co-trapping took place, both species needed to be detected after executing this cycle. Having opposite charge, and very different mass, special considerations must be made for detecting both ions and electrons within the

CPET system. In order to enable this, the trap electrode potentials on each side of the trap needed to accommodate both the injection of one charged species, and the detection of the other.

Ions were detected on the MCP downstream from CPET at the end of the cycle, thereby confirming that ions survived the entire co-trapping process. Additionally, the potentials around the harp/mesh detector could be configured to deposit the trapped electrons on it, thereby confirming their presence after having applied the nested well settings for co-trapping inside the trap. At the end of the cycle all potentials were then reset to the starting conditions. Electrons were often purged at the end of each cycle to prevent a discharge from being initiated. The necessary changes to the potentials on the trap electrodes was accomplished using the DAC box, and the surrounding electrodes and drift tubes were switched using high voltage switch boxes employing commercial Behlke switches.

5.6.5 Ion (Injection and) Extraction

Injection of an ion bunch was achieved through pulsing the voltage on the anode of the ion source. The duration of the pulse could be further shortened by also pulsing one of the steering elements in the ion source assembly in order to quickly deflect the ion beam.

Appropriate injection settings for the ions were determined by performing a series of simulations using the SIMION software. The goal was to achieve efficient injection of ions into CPET, while minimizing the radius of the ions inside the trap. The ion radius was sensitive to the focusing potential applied to the Einsel lens that ions pass through before entering the trap.

Although the steering electrodes can also be used as focusing elements for the incoming ion beam, it was found to be beneficial to minimize the DC offset of the field applied to the first ion beam steering elements immediately downstream of the CPET ion source (i.e. “S1” in Figure 5.8). If these elements are used to apply significant focusing to the beam, the ions will ultimately adopt a larger beam radius after they enter the trap. The difference between the optimal injection settings and

when the first set of steering electrodes is used for focusing can be seen in Figure 5.24. This is believed to be the result of the axial separation between the horizontal and vertical steering plates in this component of the optics. Since the beam is not surrounded by electrodes in every transverse direction, the large region between the plates can be influenced by the surrounding vacuum chamber, which is at ground potential. This likely introduces non-dipole components of the electric field that cannot be compensated for as ions enter the magnetic field of the trap. Ideally, the average potential near the centre of each steering element should be ~ 0 V to minimize this effect, so that the ions can be centred on the beam axis and aligned by an approximately uniform field in each of the two steering elements (i.e. S1 and S2). Figure 5.25 shows a sagittal view of ions injected into the trap when the injection settings are optimized in the simulation as in Figure 5.24 a).

Ion extraction is another critical step if the cooled species is to be transported to MPET for measurement. Simulations indicate that it is important to consider the field penetration into the large diameter electrodes in order to effectively extract the cold ions. The extraction gradient must be balanced so that it is steep enough for the ions exit the trap as a well-defined bunch, but shallow enough to prevent reheating of the ions.

Outside of the trap region on the downstream side of ions leaving CPET, voltages applied to the electrostatic optics are tuned for two different configurations. The first allows for the loading of electrons into the trap from the electron source, and the second allows for the transmission of ions to the final MCP on the downstream side of CPET. Ultimately, an Ortec VT120 preamp and a multi-channel scaler needed to be used to detect the ion pulses on this MCP.

5.7 Studies Performed on the Two Plasma Species

This section discusses the studies performed on the co-trapped ions and electrons in order to facilitate and verify sympathetic cooling of the ions.

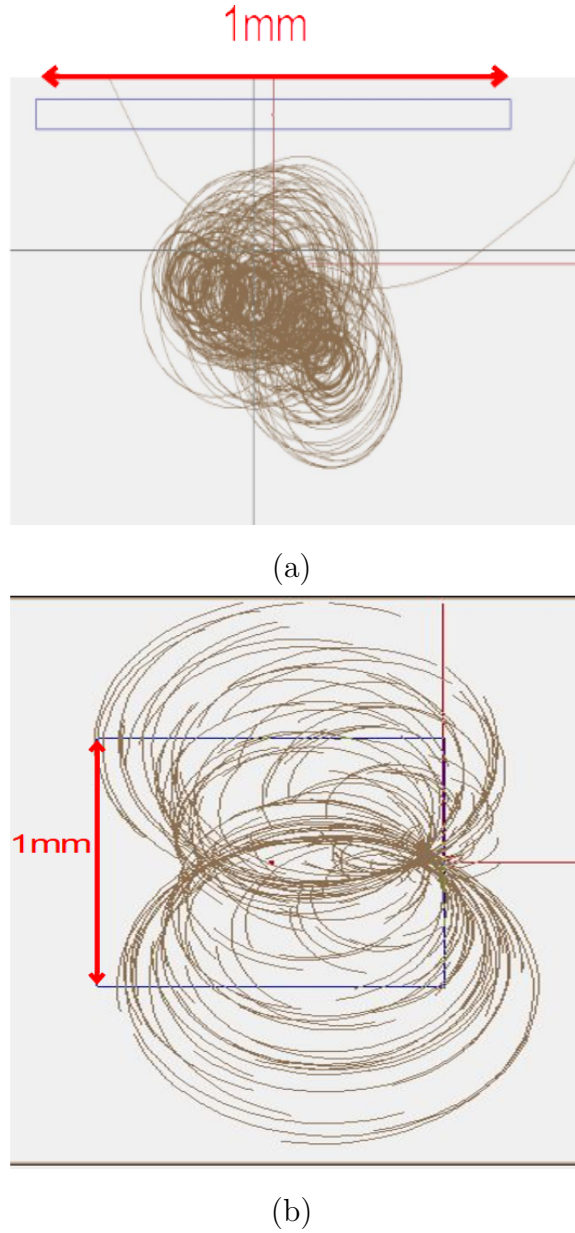


Figure 5.24: SIMION simulation of ions injected from the stable surface ion source. The image in (a) shows a cross-sectional view of ion trajectories in the central trapping region of CPET immediately after injection. The injection settings are optimized to minimize the radius of ions inside CPET while maintaining transmission into the trap (cross-sectional view).

The image in (b) shows a cross-sectional view of ion trajectories in the central trapping region of CPET immediately after injection. The ion radius in CPET increases when the second steering element after the ion source (IS S2) is used for focusing (cross-sectional view).



Figure 5.25: SIMION simulations of the ion radius inside CPET upon injection without using the second ion source steering element (IS S2) to apply focal corrections to the beam. The radius could be kept as small as ~ 0.2 mm with these injection settings (sagittal view).

5.7.1 Ion Detection

Ion detection is critical both to optimize ion capture within CPET, as well as to study the effects of endeavours to cool the trapped ions. High sensitivity approaching single ion detection is especially important for measurements of the few trapped ions ejected from the trap. The ion beam travelling through CPET can be detected on MCPs in the beam line, allowing as few as one ion at a time to be discriminated at the typically low-rate beam of $< 100/\text{s}$. Ions which have been trapped are also detected in this way, but they must be transported from the trapping region to the detectors. The size restrictions within the magnet bore, as well as the high magnetic field in the trap prevent the use of MCP detectors inside the trapping region itself.

Further Discharge-Like Behaviour in CPET

Initial attempts to observe the ion signal were complicated by a large background signal that was not initially understood. In particular, a large background signal complicated detection when ions were extracted from the trap. Varying degrees of Penning discharge and ionization of background gas were initially proposed as a mechanism to explain a variety of observations in CPET suggestive of high-voltage discharges, and which can prevent the observation of ions.

In order to facilitate the rapid cooling of electrons, CPET utilizes a 7 T magnetic field in the trap. This is a relatively high magnetic field as compared to many other

experiments that trap large quantities of electrons. For instance, other plasma experiments that study plasmas of simultaneously trapped electrons with ions operate with a field of only 0.13 T [127], [128], and [125].

Over the course of experimental investigations in CPET, substantial evidence was uncovered to indicate the presence of Penning-like discharges. This included prompt MCP and phosphor screen signals, as well as increased current draw and overloads on electrode power supplies. A continuous signal observed on the electron-side MCP at different times throughout the experimental tests are indicative of large runaway discharges. Furthermore, the amplified signal on the MCP was strong enough to be visible on the phosphor screen behind it.

Frequently, discharges were also indicated by the presence of large currents or voltage overloads on the electrode power supplies which coincided with significant activity on the MCP. These currents would continue until the bias was removed from the electrodes. The presence of discharges at a given point in time increased the likelihood that discharges would be observed again in the near future. In order to prevent the effect from immediately resuming, the voltages generally needed to be removed from the trap, end-cap, gate, and drift-tube electrodes for a cool-down time of at least 30 minutes in order to restore functionality to the trap.

It is known that under high vacuum conditions, the ignition of a Penning discharge can be delayed by a significant period of time at very low gas pressures [114]. Stable operation of electron trapping within CPET was often achieved on the $\sim$ hour timescale, before evidence of discharge conditions was discovered. The presence of these delayed discharges further supported the Penning discharge hypothesis.

In fact, Penning discharges have been exploited since 1937 to produce beams of ions at relatively low voltages from low pressure background gas [136][137]. Although a time-dependent electric field can be used for the source, only the static electric field is needed to impart energy to the charged particles inside a Penning ion source. The free electrons can be liberated from the surface of an electrode with a relatively negative bias, or else generated from ionization of residual gas. Ions can be produced between the electrodes with a potential energy that leaves them trapped. Electrons produced with total energy $\Phi < \Delta V_{trap}$ become trapped, as do electrons that lose

enough of their initial kinetic energy to collisions with the residual gas inside the vacuum chamber. Since the electrons can become trapped in this way, they can collide with residual gas atoms even in regions where the electrons have a path length much longer than the vacuum chamber itself. If the region where the electrons become trapped is nested within a positive potential minimum, any ion that is produced will be accumulated in this region, rather than being accelerated to an electrode. This mechanism is believed to be the source of the ions that were observed to be spontaneously generated in CPET when electrons are loaded into the trap.

Sensitive dependence of ion generation on the electrode voltages suggests that ionization likely occurred either during electron injection, or extraction of the hot electrons. Also, it was discovered that by reducing the heating of the electron source and ion source within CPET, it was possible to suppress many of the electron-generated ions from the signal. This may be the result of limiting residual gas in the vacuum. Reducing the voltage on the E2 end-cap also helped to limit the discharge-like ion background.

However, although some of the observed behaviours were most likely true discharges, this is not the full story. Further consideration suggests that the most persistent challenges were likely due to an additional ionization mechanism within the trap that was not initially considered.

Penning Ion Accumulation

One behaviour that had been attributed as arising from the Penning discharge mechanism, is now thought to have a different origin. Studies of the ions exiting the electron plasma revealed an apparent increase in the number of ions due to the introduction of a dense electron plasma, presumably due to the ionization of residual gas. A clear signature was observed of ions generated from a surface ion source located on the upstream side of CPET after they had been trapped within a nested potential well in CPET, and then ejected to a downstream MCP. In order to produce a cooling effect on the ions, it is necessary that electrons be trapped alongside them. However, whenever an electron plasma was introduced into the trap, a much larger

signal resembling many trapped ions would generally be observed on the downstream MCP upon ejecting positive ions from the trap.

This unanticipated signal posed a significant problem, since the injection and trapping of an electron plasma produced a signal on the downstream MCP that was indistinguishable from the trapped ions, even when no ions had been injected. Since the signal of the injected ions were mixed with, or hidden behind this signal, it was at first not possible to identify any collective or relative change in the ion time-of-flight associated with cooling. Understanding this behaviour proved to be a significant challenge in order to effectively address the problem before cooling could be demonstrated.

The number of plasma-induced time-of-flight(TOF) signals on the downstream MCP was observed to increase for longer durations of electron loading and other specific conditions. Settings needed to be optimized in order to study the signal of the injected ions after the CPET. The generated ion signal was observed to drop over time like regular trapped ions. Although the presence of MCP signals resembling ions when no ions had been injected into the trap was at first thought to only indicate high-voltage sparking, this was later interpreted to be the signal of ions generated through electron-induced ionization of the background gas. This effect is discussed below.

Under the appropriate conditions, the Penning trap can actually function as a source that bunches either ions or electrons. Within the nested potential configuration of CPET, ions produced from electron collisions with residual gas can accumulate within the negative potential well that exists for the purpose of confining the ions being cooled. Electron-induced ionization can therefore contaminate the time-of-flight signal of the ion under study. This is now believed to be the source of these ions.

The evidence strongly suggests that both the trapped electron plasma and the greater number of electrons that are reflected back out of the trapping region act as an ion source; the neutral atoms of the background gas within the trap can be ionized either by the confined electrons or by the incoming ion beam. This is problematic for CPET, since it obscures the signal of the injected ions, thereby preventing them from being detected on the MCP. In a 2017 publication by Himura et al. and a sub-

sequent 2018 proceeding from Himura [127] [128], it was observed that in a Penning trap intended for nesting two-component plasmas, the number of ions seemed to be increased due to the introduction of a dense electron plasma. Our measurements using the MCP detector also shows a similar significant enhancement of the ion signal in CPET. These results indicate that trapped electrons can ionize low-pressure background gas similarly to an Electron Beam Ion Source, albeit less efficiently and with a much lower energy beam.

Since it seems that the injection of electrons into the trap create ions within the nested potential, they need to be eliminated from the signal of the ions injected from upstream that CPET is designed to cool. It was therefore found to be essential to release the hot electrons after the electron beam is blocked. The accumulation of these ions can also be mitigated by implementing a more complex cycle that allows any positive ions that are generated to escape from the trapping region during the initial accumulation and cooling of the electrons. While the generation of these ions could not be eliminated entirely, these measures were sufficient to meaningfully observe the signal of the injected ions. This was absolutely critical to establishing a system in which ion cooling could be observed.

Application of RF to Electron Trapping

The electron-generated ions are likely the result of the same effect that was recently found to be able to generate electrons within a Penning trap at ultra high vacuum [132][133]. Existing charge-carriers within the vacuum can be driven by an RF field to ionize atoms of residual background gas to produce electrons. The difference in the case here, being that when the RF is applied to a nested potential, ions are collected in the trap as well as electrons.

Interestingly, it was seen that the electron-dependent ion signals on the MCP were enhanced when radio frequency signals were applied to electrodes. While monitoring the electron signal on the harp, RF can be applied to the plasma to see if this has an impact on its radius or other properties. It was found that the onset of this ion-like signal could be hastened by applying a radio-frequency *rotating wall potential* to the

four-segmented trap electrodes at the centre of the trap. Applying an amplitude of ~ 3 V caused the electron signal observed on the harp to be increased by an order of magnitude. Additionally, when the pulsed electron beam was turned off entirely, an electron signal could still be observed upon extraction when a sufficiently high frequency was applied. Even the application of the RF potential to only one segment of the four-segmented electrode (which produces an approximately dipole field at the central axis) was able to make this ion signal appear. It would thus seem that the presence of RF electric fields in the trap in general is sufficient to generate ions.

Onset of the ion and electron signals happened at a higher, but more reproducible frequency when the trap was first purged of any stray electrons from previously stored electrons. This was accomplished by executing several cycles of electron trapping and ejection with the pulse of emitted electrons from the electron gun disabled, before administering the RF signal. While no electrons were being injected into the trap, it was observed that when the rotating wall frequency was raised to ~ 1.5 kHz, a signal could be reliably produced on the harp that was indistinguishable from a large pulse of trapped electrons. Above 1.5 MHz, the plasma could be generated in only 0.1 s, but if the frequency was too high, or applied for too many cycles, signatures characteristic of discharges would appear.

We understand this effect to be a signature of electron plasma generation by the same mechanism first addressed in reference [126]. Electrons are produced when the RF voltage serves to heat any free electrons that are present inside the trap until they have enough kinetic energy to ionize the residual gas within the vacuum, even under UHV conditions. This can be a useful technique for generating electrons within the vacuum of a Penning trap.

In summary, all of these effects can find a common explanation in electrical discharges that are particularly exacerbated by ionization within the trapping potentials and strong magnetic field of CPET.

5.7.2 Searches for an Electron-Ion Interaction

In order to demonstrate an interaction between ions and electrons, it is necessary to establish some property of the ions which depends on the presence of electrons (or vice-versa, but CPET traps many orders of magnitude more electrons than ions, so the effect of the ions on the plasma is expected to be minimal). Electron dependent heating or cooling (total energy or energy spread), space charge effect, or even destruction the charge state of the ions can provide a signal to indicate the interaction between the two species.

Using Space-Charge to Test and Optimize Electron-Ion Interaction

Of course, for a reduction in the energy spread of ions to occur, there must be a spatial overlap between the ions and the electron plasma during trapping. Therefore, efforts were made to first optimize the physical overlap of the two trapped species when ions undergo a single pass through the trap. Cooling on a reasonable time scale will require a tune that injects the ion beam into the trap at a radial position that allows them to significantly overlap with the trapped electron plasma. One approach I explored to allow us to search for such a tune involves taking advantage of the *space-charge* of the electrons.

Space charge is the repulsive (or attractive) potential due to the net charge of the particles which occupy a region of an ion beam or trap, resulting directly from the Coulomb interaction. The practical consequence of the space-charge effect in ion traps is that it sets a fundamental limit on the maximum number of particles of a given charge that can occupy a region of space within the trap. However, in the case of CPET, the high density of negatively charged electrons in the trap might instead be used to lower the on-axis potential enough such that ions could occupy a region which would be impossible for them to inhabit if electrons were absent from the trap. In Figure 5.28, this would result in a reduced effective voltage in the electron well.

The space-charge effect can be used to engineer a situation in which ion transmission through the trap depends on the presence of the electron plasma. The ions will only experience a reduced space-charge potential due to electrons when the two

species have a radial overlap. CPET was configured to probe this overlap by using the trap electrodes to provide a positive blocking potential in the centre of the trap that falls off harmonically. The same positive potential applied to electrodes can be used to both block the passage of positive ions, as well as to provide a potential minimum for negatively charged electrons to cool into. By blocking the passage of ions with the trap electrodes, this potential acts on the ions similarly to the repulsive effect of a retarding field analyzer. However, rather than sensitively probing the ion energy, by tuning to the threshold voltage for blocking ions, the overlap of ions passing through the trap with trapped electrons due to changes in the electric field is tested.

If the (axial) harmonic potential well within the trap becomes filled with a sufficiently large number of electrons, their space charge will alter the electric potential on the central axis of the trap. This essentially provides a target in the transverse plane at which to aim the ion beam. If the presence of electrons allows ions to pass through this region, then we have demonstrated that the two different species pass close enough to one another to interact via the Coulomb interaction. In this way the trap electrodes can provide a spatially-dependent retarding field in the xy-plane, that could allow for an optimization of the electron-ion interaction.

Due to the heavier mass of the ions, and correspondingly lower cyclotron frequency, it is significantly easier to manipulate the radial position of ions within the trap using a set of Lorentz steerers (symmetrical steering element) than it is for electrons. The cyclotron radius of the ions is also correspondingly larger. The voltages on the Lorentz steerer in the fringe field of the magnet can be used to tune the injection of the ion beam to maximize the overlap of the two species.

The technique of manipulating the on-axis potential of partially blocked bunches of ions using the electron plasma was attempted on several occasions. Initial efforts to observe an electron-dependent transmission of ions through the trap electrodes were unsuccessful. These measurements were hampered by discharges that interrupted the setup of the complicated switching cycles, and limitations of the pulse generator. Measurement of the ion transmission rate was also complicated by the presence of ions that seem to be generated by the electron plasma. There are several possible explanations for this; for example, a misalignment of the trap electrodes or steering

elements with respect to the magnetic field lines inside the trap could cause the ions to be injected on an angle. It is essential that the ions radially overlap with the ions over a sufficiently long portion of the trap length in order for space charge to allow ion transmission.

After a number of upgrades, it appears that the presence of electrons does indeed have an effect on the ion transmission. However, rather than increasing the number of ions that are transmitted through the highest potential region of the trap, the negatively charged electron plasma significantly reduces the rate of transmission when a sufficient quantity of electrons has been collected in the trap. No tune could be found to increase the ion transmission. In spite of this somewhat surprising result, the observation of this effect nevertheless provides the *first direct evidence of an electron-ion interaction*.

The Lorentz steerer near the entrance to the CPET magnet can be scanned to determine the impact of steering on the electron-ion interaction when passing bunches of ions through the trap. Various settings on the Lorentz steerers were tried to see if the ion transmission could be increased when electrons were present in the trap, but optimal ion transmission occurred at the same voltages with or without electrons, with the presence of electrons inhibiting ion transmission. Any ions generated from background gas by the electron plasma would be expected to also increase the ion rate at the MCP detector. This leaves a limited number of possible explanations for this decrease in rate. The conditions for dielectronic recombination are thought unlikely to be met, since these are singly-charged ions which only pass through the trap once. Furthermore, the ion transmission was seen to be systematically reduced for a range of incoming ion energies.

The decrease in ion transmission may be due to a defocusing effect of the electrons, or the fact that their presence couples the axial momentum of the ions into the radial plane. Since the mechanism could not be easily determined, optimization of the overlap of the two species shifted to trapping both species simultaneously instead of only passing the ions through the trap region. With the switching cycle changed to a configuration that traps the ions, the expectation is that the ions will begin to lose their kinetic energy to the electrons if they are indeed interacting.

A significant effort in seeking an interaction was directed toward co-trapping the two species, since this is the ultimate goal of CPET, and because it allows for long interaction times of the two species. Figure 5.26 shows example electrostatic trapping potentials for accumulating electrons, and for combining ions with electrons, superimposed on the magnetic field of the trap. First, electrons need to cool into the trap centre. After this has happened, the three combined electrodes of E1 and the 3 combined electrodes of E2 can have their voltages increased to create blocking potentials that confine the positively-charged ions. For the potentials pictured here, ions will overlap with the previously accumulated electrons at the centre of the trap.

For these tests, new electrons were loaded into the trap each cycle. A potential gradient was applied to the trap electrodes to dump electrons from the trap after every ion cooling cycle and maximize reproducibility. The hot electrons were released after each loading cycle. Electrons were loaded for 300 ms and subsequently allowed to cool for 1 second, with a 30 V deep rectangular nest. Background ion rates as low as ~ 1 count per cycle were present in the MCP signal.

Time Of Flight Approach

Since the ion velocity $v \propto \sqrt{E_k}$, the simplest method to search for cooling of the trapped ions is to look for changes to their time-of-flight from the trapping region to the final MCP. This results from ions losing kinetic energy due to the presence of electrons. If the electrons significantly reduce the total kinetic energy of the ions, their arrival at the MCP after extraction (by opening the E2 end-cap electrode) is delayed. If the spread in the distribution of the ion energies is reduced, the width of the time-of-flight pulse on the detector becomes more narrow. One possible caveat of this method is that the space charge of the electrons could impact the ion TOF as they exit the trap.

Although, the detection of this signal was initially hampered by the injection of electrons into the trap causing a large number of overlapping ion counts on the detector when ions were extracted, after this was resolved an apparent cooling effect of the electrons was observed in the ion signal. Ultimately, the incremental improvements

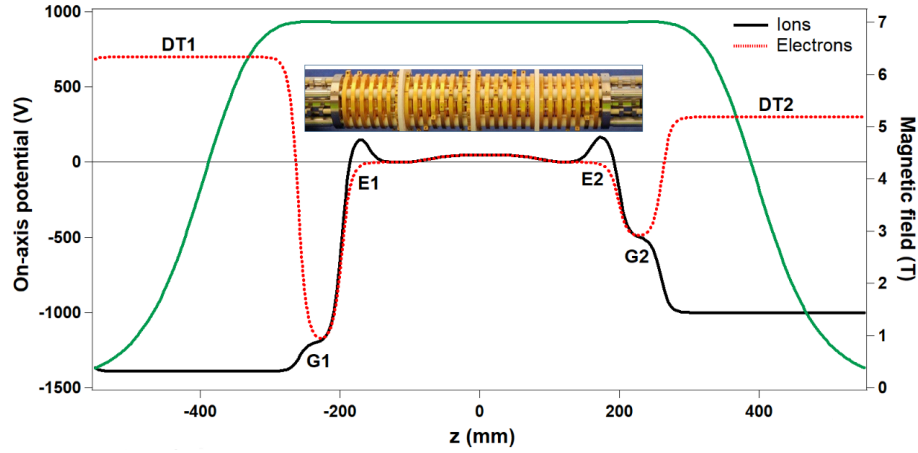


Figure 5.26: An example potential configuration for combining ions with electrons. Magnitudes of the potentials varied over the course of these tests, but this general form of the potentials was used for most studies producing the two species plasma. The dotted red line is the potential used when electrons were accumulated in the centre of the trap using the blocking potentials applied to the gate electrodes (G1/G2). Next, the potential configuration represented by the black line was used so ions could be trapped as well using the endcap electrodes (E1/E2), such that they overlapped with the trapped electrons. The drift tubes (DT1/DT2) require different potentials for ion and electron transmission, as well as a dynamic change to the potential when matching ions to the trap potential. The green line represents the magnitude of the magnetic field on the central axis. Image from [138] CC BY 3.0 (<https://jacow.org/ecris2020/papers/nacb04.pdf>).

made to the system allowed both an overall delay and a compression in the ion time-of-flight of the extracted ions to be observed above the background when electrons were present.

Figure 5.27 shows that for sufficiently long trapping times, the hot (fast) ions become both significantly delayed and more narrow. Part a) shows the endcap voltage being smoothly decreased in order to avoid discharges, and to see if the ions cool to the bottom of the potential well. The delayed arrival time when electrons are present in part c) relative to when they are absent in part b) is suggestive of reduced

kinetic energy. Even more significant is the narrowing of the time-of-flight peaks when ions are trapped for longer. The fact that a longer interaction time with the electrons causes the peak in the ion TOF to become more narrow strongly suggests that sympathetic cooling is occurring.

Retarding Field Approach

Additionally, it was possible to observe the energy spread of the ion bunch using an electrode on the extraction side of the trap as an improvised *retarding field analyzer* (RFA). With the correct blocking potential applied, we would expect to see some ions that would have just enough energy to reach the final detector when hot, but not if they are cooled. Using this method also provided evidence of sympathetic cooling of the ions.

The kinetic energy profile of an extracted ion bunch was probed by progressively raising the potential of the trap electrode until transmission of the ion bunch to the MCP was completely blocked. A clear picture of the energy spread is obtained by measuring the ion transmission as a function of the blocking voltage.

However, since the potential at the centre of the trapping electrodes can deviate significantly from the potential at the surface of the electrode, one must be cautious when relating the electrode voltage to the ion energy. As can be seen in Figure 5.28, this is a consequence of the field penetration that is possible for the large inner diameter of the electrodes, and is most significant in regions where the potential applied to the electrodes is changing rapidly along the axis. It is important to ensure that ions are subject to the appropriate electric potential on the beam axis in the blocking region, as this may differ substantially from the voltage applied to the electrodes at the corresponding axial position.

The blocking potential voltages were therefore applied to the 3 electrode wide end-cap electrode in order to limit field penetration when performing a retarding field scan. By scanning over a range of blocking voltages, it was revealed that the number of ion counts arriving at the final MCP detector could be suppressed when the ions were held for a sufficient length of time within an electron plasma. Figure

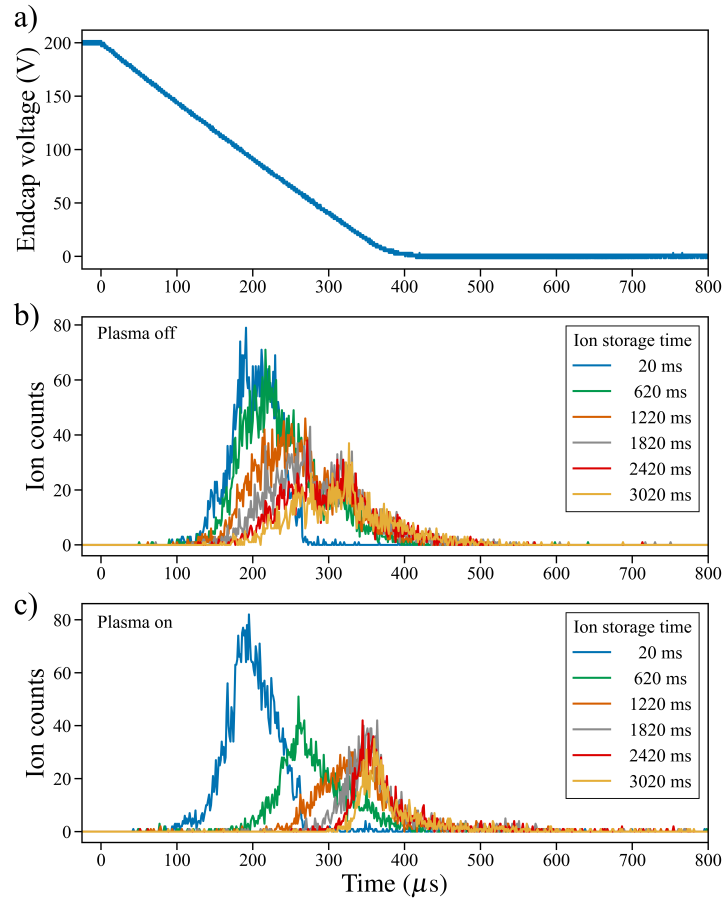


Figure 5.27: Sympathetic cooling of ions is demonstrated by observing the effect of trapping duration with the electrons on their time-of-flight to the final MCP detector when extracted from the trap. When the electrons are introduced in part c), TOF peaks are observed to narrow as compared to trapping without electrons in part b). This suggests a reduced ion energy spread. Image courtesy of Stefan Paul.

5.29 shows how the retarding field curves are effected by the plasma.

Figures 5.30, 5.31, and 5.32 shows how the ion cooling timescales of ^{23}Na , ^{39}K , and ^{85}Rb respectively are impacted by the presence of an electron plasma. A modest cooling effect over time is observed even when ions are trapped without electrons. This is

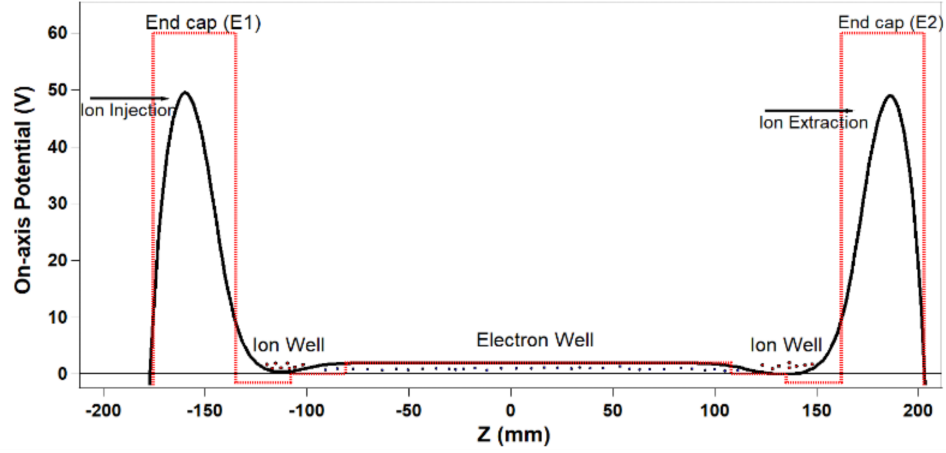


Figure 5.28: On-axis potential with a triple electrode end-cap. The large inner radius of the electrodes allows the field from other electrodes to influence the on-axis potential of adjacent electrodes. This substantially smooths the transitions between regions of different potentials and reduces the maximum on-axis potential that the end cap electrodes can apply. Image from [138] CC BY 3.0 (<https://jacow.org/ecris2020/papers/nacb04.pdf>).

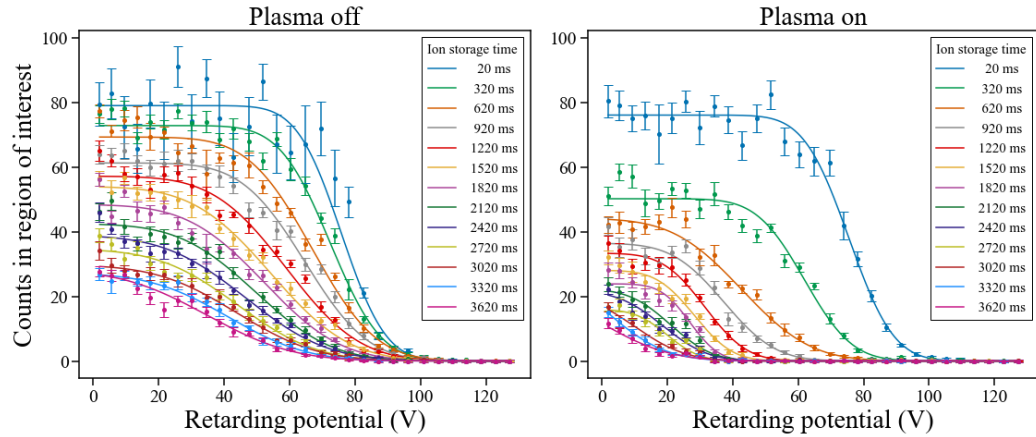


Figure 5.29: The effect of electron cooling on scans of the number of detected ions versus the blocking voltage applied to the end-cap electrode. Image provided by Stefan Paul.

most likely due to some combination of cooling through collisions with residual gas, and/or evaporative cooling due to gradual losses of the hottest ions. The essential question is whether an additional cooling effect can be observed through the introduction of electrons. When electrons are introduced to the trap they are shown to significantly reduce the number of ions that are able to overcome the potential barrier after a given trapping time. This provides compelling evidence of electron-induced cooling.

While the reduced transmission of ions could also be explained by the electrons increasing ions losses, recombination is very unlikely for singly-charged ions, and when combined with the increased time-of-flight discussed above, provides strong evidence of the cooling effect. Singly-charged Na, K, and Rb show evidence of interacting with the electron plasma to give up kinetic energy. This is a promising result for the technique when one considers that the interaction of HCIs with electrons is expected to be greater due to the enhanced scattering of ions in higher charge states. By comparing the curve in Figure 5.30 to the other cooling curves it can be seen that the Na ions have the lowest *mean ion energy* at all stages during the trapping cycle. This is consistent with the expectation that ions with smaller mass to charge ratios will exhibit enhanced scattering, and bodes well for the cooling of highly-charged species.

The energy decay curves shown here are fit to an exponential decay of the form $E(t) = (E_0 - E_{inf})e^{-\frac{t}{\tau}} + E_{inf}$, and the results are summarized in table 5.1. E_0 is the initial energy and E_{inf} is the energy after long times. The cooling time constant due to electrons is given by $\tau_{ecool} = 1 / \left(\frac{1}{\tau_{combined}} - \frac{1}{\tau_{bkg}} \right)$, which uses the time constant of ion cooling when electrons are introduced $\tau_{combined}$, and the time constant of ion cooling when trapped without an electron plasma τ_{bkg} . Although the species with lower mass-to-charge ratios cool more quickly, all results are consistent with electrons enhancing the rate of ion cooling.

Ion losses due to the electron plasma are reasonable. As can be seen in Figures 5.33 and 5.34, there is better ion retention for lower mass-to-charge ratios with Na and K suffering a modest plasma-induced ion loss of $\sim 50\%$. Clearly the errorbar for the fit shown in Figure 5.35 is too large to conclude anything. This provides yet another indication of the utility of electron cooling for HCIs.

	^{23}Na	^{39}K	^{85}Rb
Background Cooling Time Constant (s)	2.5 ± 0.4	3.5 ± 0.8	5.1 ± 2.8
Electron Cooling Time Constant (s)	1.4 ± 0.3	1.2 ± 0.2	9 ± 14

Table 5.1: A comparison of ion cooling time constants when trapped with electrons to when they are trapped without introducing electrons into the trap.

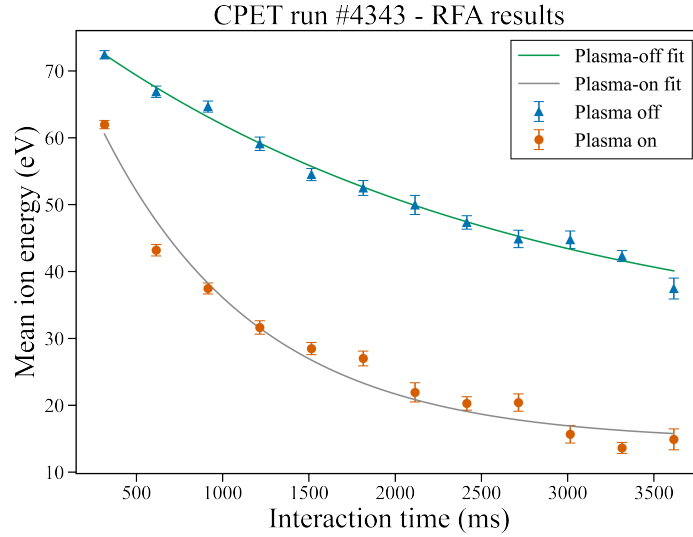


Figure 5.30: The energy of trapped $^{23}\text{Na}^+$ ions in the trap was measured for various trapping duration using the RFA approach. A comparison of the time evolution of the required blocking potential downstream from the trap reveals how the cooling rate changes when an electron plasma is present to speed the cooling of ions. In this case, the electrons are held in a 40V square potential well. Some loss of ion energy is observed for long trapping times, presumably due to losing the hottest ions. Plot provided by Stefan Paul.

5.8 Conclusion of Sympathetic Cooling Studies

In summary, sympathetic cooling of ions was achieved in CPET using electrons as a cooling medium. However, the strong magnetic field required for electrons to cool by synchrotron radiation on a short timescale, coupled with the necessary nested potential configuration, creates a more challenging environment in which to consistently

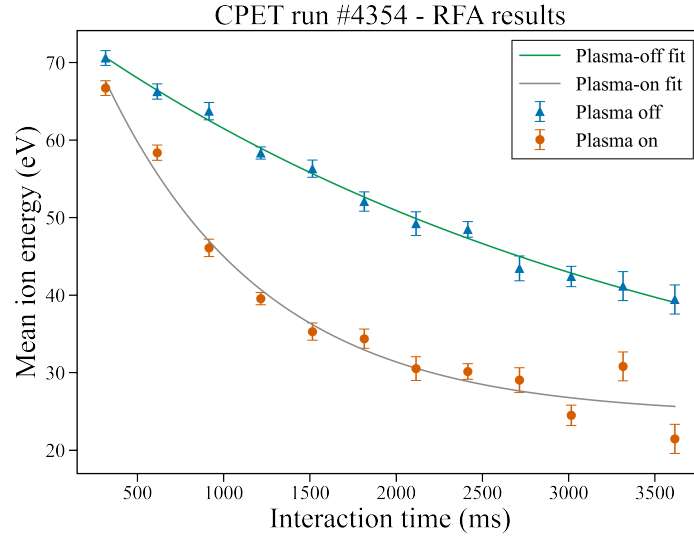


Figure 5.31: Same as Figure 5.30, for the energy of trapped $^{39}\text{K}^+$ ions as a function of trapping time. Provided by Stefan Paul.

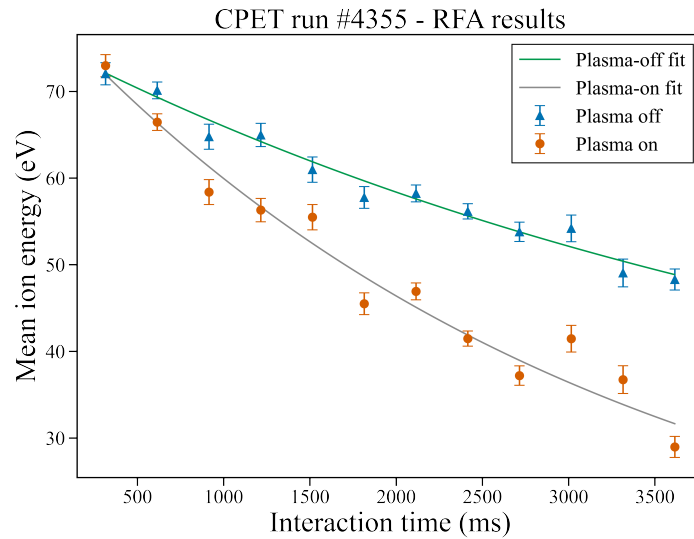


Figure 5.32: Same as Figure 5.30, for the energy of trapped $^{85}\text{Rb}^+$ ions as a function of trapping time. Provided by Stefan Paul.

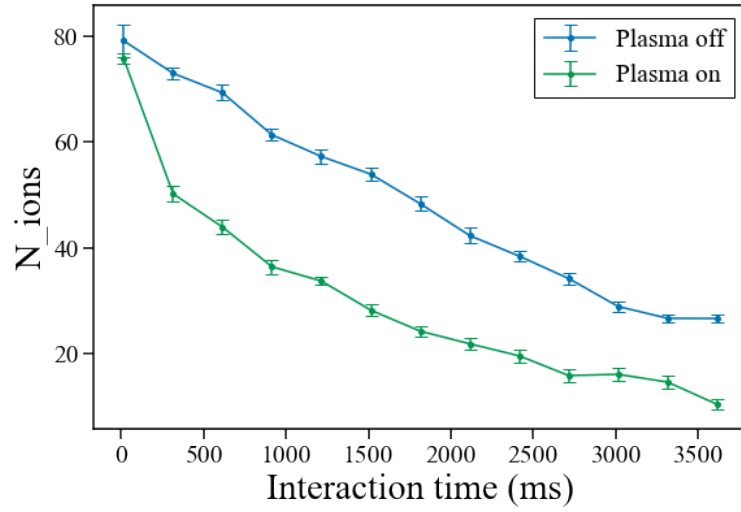


Figure 5.33: Ion losses in the trapped $^{23}\text{Na}^+$. Plot provided by Stefan Paul.

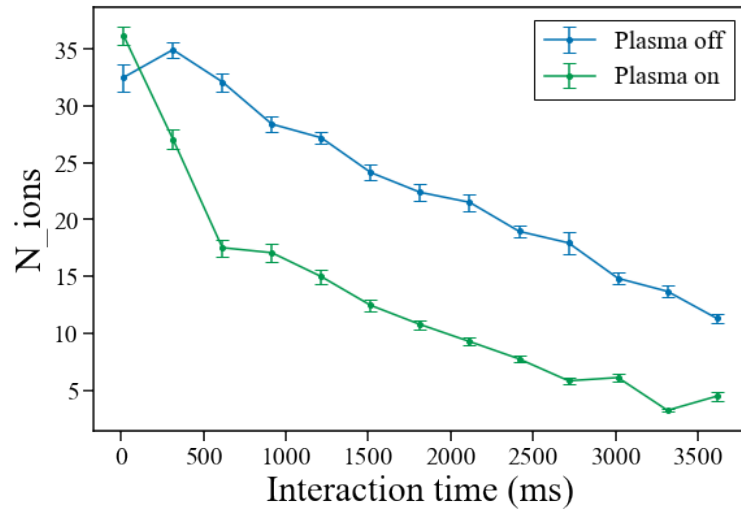


Figure 5.34: Ion losses in the trapped $^{39}\text{K}^+$. Plot provided by Stefan Paul.

trap and cool ions. Hardware and trapping procedures must be carefully configured in order to avoid Penning discharges and the accumulation of ionized background gas. Using both a retarding field approach and a time-of-flight approach, evidence was uncovered to indicate that sympathetic cooling using electrons in a Penning trap was achieved for the heaviest ions ever cooled in this way. Although the extreme reliability requirements of studies on radioactive beams of highly-charged ions (HCIs) would

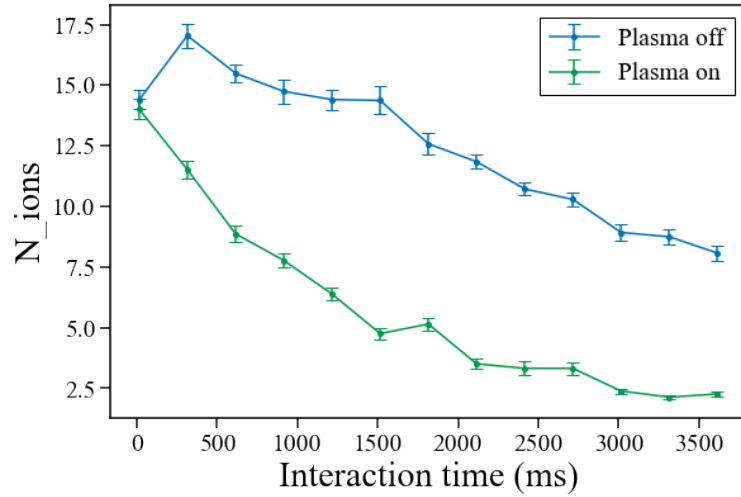


Figure 5.35: Ion losses in the trapped $^{85}\text{Rb}^+$. Plot provided by Stefan Paul.

be challenging to address using the current CPET system, this method opens the door for the rapid cooling of ions within a Penning trap. Additionally, the obstacles encountered in this endeavour highlight the fact that flooding a trap with electrons makes it possible to accumulate ions from residual gas within a Penning trap, even under ultra high vacuum (UHV) conditions, and with relatively low voltages.

Moving forward, sympathetic cooling using electrons could find utility in cooling HClIs, since the interaction with the electron plasma increases with the charge of the ions.

Chapter 6

Outlook

6.1 Sympathetic Cooling with Electrons

After making significant technical improvements to the Cooler Penning Trap system, strong evidence indicating the sympathetic cooling of ions using electrons has been obtained using CPET. The cooled species included stable $^{23}\text{Na}^+$ and $^{39}\text{K}^+$ ions (as well as $^{85}\text{Rb}^+$ but with larger uncertainty). To our knowledge, these represent the highest mass-to-charge ratios for species cooled using this method. This demonstrates the general applicability of the technique for fast cooling of ions. With ion cooling times on the order of a second, this could prove to be a valuable technique for cooling time-sensitive species that require cooling at room temperature in order to reduce the energy spread of trapped ions ranging from 10s to 100s of eV.

Ultimately, the results of these investigations into commissioning the first (electron-mediated) sympathetic cooling of radioactive ions inside a Penning trap have led to the conclusion that the technical and infrastructure requirements associated with integrating CPET within TITAN's radioactive beamline is not the optimal use of experimental resources at this time. The unique requirements of very rapid cooling, high efficiency, and maximal up-time during online radioactive beam experiments pose a tight set of constraints for the particular case of cooling highly-charged radioactive ions at TITAN. The efforts necessary to achieve a more favourable duty cycle using this technique have therefore been decided to be best directed toward other improve-

ments at TITAN. However, the evidence obtained for sympathetic cooling of ions using this technique, as well as the observations that enabled this, remain of broad interest to the scientific community.

This work is relevant to studies on two-species plasmas [128] [125]. In particular, the ionization of stray atoms observed in the CPET could possibly impact such experiments in unexpected ways. As CPET has shown, the energetic trapped electrons tend to ionize other low-pressure atoms (or molecules) of residual gas, even under UHV conditions. The observations of this electron-induced ionization in CPET suggests of inherent difficulties when attempting to trap electrons with only a single species of positively-charged ions. When seeking agreement between experiment and the theory of two-species plasmas, the possible influence from unanticipated species of ions contaminating the trap should therefore be considered when modeling such systems.

The cooling demonstrated in CPET can also be used to guide the endeavour to cool bunches of HCIs at GSI's HITRAP facility [112] [113]. Sympathetic cooling using electrons remains a promising method for precision experiments requiring the cooling of Highly-Charged Ions (HCIs).

When used as a cooling trap for HCIs, steps may be necessary to prevent the sample from being contaminated by singly-charged ions produced by the electrons interacting with the background gas, including the possibility of charge exchange. Possible measures to prevent contamination from in-trap ionization could include dumping any positive species from the trap after the electrons have been injected and cooled, or the minimizing the kinetic energy of electrons inside the trap.

Furthermore, the consistent production of ions by trapping electrons inside a nested trap, even suggests the possible use of this configuration as a cold, RF-free source of ions from residual gas under UHV conditions. For example, this could be useful in situations in which using an electron source is more practical than a gaseous discharge ion source to generate small quantities of ions for a Penning trap.

6.2 Scientific Outlook for Mass Measurements of Neutron-Deficient Lanthanides

Mass data of neutron-deficient lanthanide elements in the vicinity of the $N=82$ shell closure was collected during two experimental runs with radioactive beam from TRIUMF's ISAC facility in 2018 and 2019. This was accomplished using the MRTOF at TITAN and the brand new (as of the 2018 experiment) re-trapping technique to purify the beam, solidifying its utility for mass measurements of rare isotopes at an ISOL facility. The $N=82$ shell closure was thereby confirmed to persist up to Yb ($Z=70$) from the perspective of ground state energies. Future work measuring the energy ratios of the 4^+ to 2^+ excited states in neutron-deficient Yb would provide a useful additional confirmation of the magicity of the $N=82$ shell closure in this region.

Additionally, the location of the proton drip line in Tm ($Z=69$) was unambiguously identified through newly measured values of the proton separation energy, establishing ^{150}Tm ($N=81$) as the last proton-bound isotope. This result was facilitated both by a redetermination of the mass of ^{149}Er through a new measurement of its isomeric state, and by the direct measurement of the previously unmeasured ^{149}Tm and ^{150}Tm . The limits of the proton-bound nuclei are of fundamental interest, and ^{149}Tm is now confirmed to be the first proton-unbound Tm isotope, with a proton separation energy of $S_p = -415 \pm 48$ keV. This also provides a new constraint on mean-field theory calculations of the proton drip line, which were consistent with ^{149}Tm being either bound or unbound.

Furthermore, the angular momentum represents a fundamental quantity that is necessary to understanding the structure of nuclei. The separation energy of any proton-unbound nucleus can be used in conjunction with the angular momentum of the valence proton (spin), and the nuclear radius, to calculate the proton emission half-life. Alternatively, if the half-life is known instead, the separation energy can be used to discriminate between different spin states. Future spin and proton emission measurements could be used to explore this relationship within the scope of various theoretical models.

Appendix A

Overview of Personal Contributions

Below I have included a selection of my personal scientific and technical contributions which were necessary to accomplish the scientific outcomes presented within this thesis.

A.1 Contributions to Lanthanide Mass Measurements

- Acquired data from radioactive beam measurements and acted as “MRTOF expert”
- Planning of analysis and atomic/molecular calibrant selection
- Prepared data by performing time-resolved calibrations and generating histograms
- Fit data using Gaussian functions; identified species present; applied appropriate limits (cuts)
- Calibrated all mass peaks to known masses; cross-checked results of various known isomers, molecules, and ground states

- Visualizing of data
- Literature review to interpret results

A.2 Contributions to Cooler Penning Trap

- Planning technical improvements to enable cooling
- Simulated ion source injection optics to enhance ion rate
- Documentation of technical improvements and experimental procedures
- Setup, cleaning, and installation of ion source
- Installation of electron harp (but not design)
- Collected electron trapping data and optimized electron injection
- Installation of off-axis and on-axis electron sources
- Testing and troubleshooting of off-axis electron gun and identification of mis-steered beam
- Cleaning and installation of components under ultra high vacuum (UHV) conditions
- Operation of vacuum system; sealing, pumping, and venting of vacuum
- Monitoring of vacuum pressure, voltages, currents, etc.
- High voltage testing for electrical shorts (both in and out of vacuum)
- Measurements of residual gas composition
- MCP and other diagnostics installations and modifications as required
- Design of custom made floating HV bias box (PCB design and mechanical fabrication done by TRIUMF staff) and subsequent hardware improvements

-
- Installation and testing of custom HV bias box
 - Installation and cabling of HV power supplies
 - Construction and use of filters/coupling boxes etc.
 - Performed tests trapping electrons and ions
 - Optimization of trap voltages
 - Quantification of trapped charged particles
 - Preparation of dense electron plasmas
 - Confirmed the cooling of electrons into a potential minimum
 - Performed experiment combining electrons with ions
 - Performed extensive tests to identify and understand the sources of electrical discharges and other spurious signals
 - Identified ions produced by electrons
 - Performed time-of-flight and retarding field measurements to search for reductions in ion energy

Bibliography

- [1] Z. Ke, W. Shi, G. Gwinner, K. Sharma, S. Toews, J. Dilling, and V. L. Ryjkov, “A Cooler Ion Trap for the TITAN On-Line Trapping Facility at TRIUMF,” *Hyperfine Interactions*, vol. 173, no. 1-3, pp. 103–111, 2006.
- [2] E. Rutherford, “The British Association at Winnipeg,” *Nature*, vol. 81, no. 2078, p. 263, 1909.
- [3] E. Rutherford and H. Brooks, “The New Gas from Radium,” *Trans. R. Soc. Canada*, vol. 7, pp. 21–25, 1901.
- [4] F. W. Aston, “Bakerian Lecture. A New Mass-Spectrograph and the Whole Number Rule,” *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character*, vol. 115, no. 772, pp. 487–514, 1927.
- [5] F. Aston, “LXXIV. A Positive Ray Spectrograph,” *The London, Edinburgh and Dublin philosophical magazine and journal of science*, vol. 38, no. 228, pp. 707–714, 1919.
- [6] G. Gamow, “Mass defect curve and nuclear constitution,” *Proc. R. Soc. Lond. A*, vol. 126, pp. 632–644, 1930.
- [7] C. F. v. Weizsäcker, “Zur Theorie der Kernmassen,” *Zeitschrift für Physik*, vol. 96, no. 7, pp. 431–458, 1935.
- [8] W. Huang, M. Wang, F. G. Kondev, G. Audi, and S. Naimi, “The AME 2020 Atomic Mass Evaluation (I). Evaluation of Input Data; and Adjustment Proce-

- dures,” *Chinese Physics C: High Energy Physics and Nuclear Physics*, vol. 45, p. 030002, Mar 2021.
- [9] M. Goeppert Mayer, *On Closed Shells in Nuclei*. AECD ; 1818, Oak Ridge, Tenn: U.S. Atomic Energy Commission, Technical Information Division, 2 1948.
- [10] M. Goeppert Mayer, “On Closed Shells in Nuclei. II,” *Physical Review*, vol. 75, pp. 1969–1970, Jun 1949.
- [11] O. Haxel, J. H. D. Jensen, and H. E. Suess, “On the “Magic Numbers” in Nuclear Structure,” *Physical Review*, vol. 75, pp. 1766–1766, Jun 1949.
- [12] J. Dobaczewski, I. Hamamoto, W. Nazarewicz, and J. Sheikh, “Nuclear Shell Structure at Particle Drip Lines,” *Physical Review Letters*, vol. 72, no. 7, pp. 981–984, 1994.
- [13] P. J. Woods and C. N. Davids, “Nuclei beyond the proton drip-line,” *Annual Review of Nuclear and Particle Science*, vol. 47, no. 1, pp. 541–590, 1997.
- [14] L. Neufcourt, Y. Cao, S. Giuliani, W. Nazarewicz, E. Olsen, and O. B. Tarasov, “Beyond the Proton Drip Line: Bayesian Analysis of Proton-Emitting Nuclei,” *Physical Review C*, vol. 101, p. 014319, Jan 2020.
- [15] H. Geiger and J. Nuttall, “The Ranges of the α Particles from Various Radioactive Substances and a Relation Between Range and Period of Transformation,” *Philosophical Magazine*, vol. 22, no. 130, pp. 613–621, 1911.
- [16] H. Geiger and J. Nuttall, “The Ranges of α Particles from Uranium,” *Philosophical Magazine*, vol. 23, no. 135, pp. 439–445, 1912.
- [17] C. Qi, R. J. Liotta, and R. Wyss, “Generalization of the geiger-nuttall law and alpha clustering in heavy nuclei,” *Journal of Physics: Conference Series*, vol. 381, p. 012131, sep 2012.
- [18] G. Gamow, “Zur Quantentheorie des Atomkernes,” *Zeitschrift für Physik*, vol. 51, pp. 204–212, 1928.

- [19] C. Qi, F. R. Xu, R. J. Liotta, and R. Wyss, “Universal decay law in charged-particle emission and exotic cluster radioactivity,” *Physical Review Letters*, vol. 103, p. 072501, Aug 2009.
- [20] W. Nazarewicz, J. Dobaczewski, and T. R. Werner, “Physics of Exotic Nuclear States,” *Physica Scripta*, vol. T56, pp. 9–14, Jan 1995.
- [21] Z.-X. Zhang and J.-M. Dong, “A Formula for Half-life of Proton Radioactivity,” *Chinese Physics C*, vol. 42, p. 014104, Jan 2018.
- [22] E. Leistenschneider, M. P. Reiter, S. Ayet San Andrés, B. Kootte, J. D. Holt, P. Navrátil, C. Babcock, C. Barbieri, B. R. Barquest, J. Bergmann, J. Bollig, T. Brunner, E. Dunling, A. Finlay, H. Geissel, L. Graham, F. Greiner, H. Hergert, C. Hornung, C. Jesch, R. Klawitter, Y. Lan, D. Lascar, K. G. Leach, W. Lippert, J. E. McKay, S. F. Paul, A. Schwenk, D. Short, J. Simonis, V. Somà, R. Steinbrügge, S. R. Stroberg, R. Thompson, M. E. Wieser, C. Will, M. Yavor, C. Andreoiu, T. Dickel, I. Dillmann, G. Gwinner, W. R. Plaß, C. Scheidenberger, A. A. Kwiatkowski, and J. Dilling, “Dawning of the $N = 32$ Shell Closure Seen through Precision Mass Measurements of Neutron-Rich Titanium Isotopes,” *Physical Review Letters*, vol. 120, p. 062503, Feb 2018.
- [23] M. P. Reiter, S. Ayet San Andrés, E. Dunling, B. Kootte, E. Leistenschneider, C. Andreoiu, C. Babcock, B. R. Barquest, J. Bollig, T. Brunner, I. Dillmann, A. Finlay, G. Gwinner, L. Graham, J. D. Holt, C. Hornung, C. Jesch, R. Klawitter, Y. Lan, D. Lascar, J. E. McKay, S. F. Paul, R. Steinbrügge, R. Thompson, J. L. Tracy, M. E. Wieser, C. Will, T. Dickel, W. R. Plaß, C. Scheidenberger, A. A. Kwiatkowski, and J. Dilling, “Quenching of the $N = 32$ Neutron Shell Closure Studied via Precision Mass Measurements of Neutron-Rich Vanadium Isotopes,” *Physical Review C*, vol. 98, p. 024310, Aug 2018.
- [24] J. C. Hardy and I. S. Towner, “Superaligned $0^+ \rightarrow 0^+$ nuclear β decays: 2014

- critical survey, with precise results for V_{ud} and CKM unitarity,” *Physical Review C - Nuclear Physics*, vol. 91, no. 2, pp. 1–27, 2015.
- [25] A. G. Brenton and A. R. Godfrey, “Accurate Mass Measurement: Terminology and Treatment of Data,” *Journal of the American Society for Mass Spectrometry*, vol. 21, no. 11, pp. 1821 – 1835, 2010.
- [26] P. Bricault, R. Baartman, M. Domsbky, A. Hurst, C. Mark, G. Stanford, and P. Schmor, “TRIUMF-ISAC Target Station and Mass Separator Commissioning,” *Nuclear Physics A*, vol. 701, no. 1, pp. 49–53, 2002. 5th International Conference on Radioactive Nuclear Beams.
- [27] J. Bagger, R. Laxdal, Y. Bylinsky, O. Kester, A. Gottberg, P. Schaffer, K. Hayashi, S. Koscielniak, M. Marchetto, and F. Ames, “TRIUMF in the ARIEL era,” *Proc. IPAC’18*, p. 6, 2018.
- [28] A. Laxdal, R. Baartman, I. Bylinskii, S. Ganesh, A. Gottberg, F. Jones, P. Kunz, L. Lopera, T. Planche, and A. Sen, “Recirculated Electron Beam Photo-Converter for Rare Isotope Production,” in *8th Int. Particle Accelerator Conf. (IPAC’17), Copenhagen, Denmark, 14â 19 May, 2017*, pp. 2526–2529, JACOW, Geneva, Switzerland, 2017.
- [29] J. Dilling, R. Krücken, and G. Ball, “Isac overview,” *Hyperfine Interactions*, vol. 225, pp. 1–8, 2014.
- [30] F. Maldonado Millan, T. Day Goodacre, and A. Gottberg, “Multiphysics Simulation of a FEBIAD Ion Source,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 463, pp. 302–304, 2020.
- [31] M. Reiter, F. Ames, C. Andreoiu, S. A. San Andrés, C. Babcock, B. Barquest, J. Bergmann, J. Bollig, T. Brunner, T. Dickel, *et al.*, “Improved Beam Diagnostics and Optimization at ISAC via TITAN’s MR-TOF-MS,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 463, pp. 431–436, 2020.

- [32] A. Kwiatkowski, “Ion Traps for Precision Experiments at Rare-Isotope-Beam Facilities,” 2016.
- [33] G. Bollen, P. Dabkiewicz, P. Egelhof, T. Hilberath, H. Kalinowsky, F. Kern, H. Schnatz, L. Schweikhard, H. Stolzenberg, R. B. Moore, H. J. Kluge, G. M. Temmer, and G. Ulm, “First Absolute Mass Measurements of Short-Lived Isotopes,” *Hyperfine interactions*, vol. 38, no. 1-4, pp. 793–802, 1987.
- [34] K. S. Sharma, R. C. Barber, H. Fukutani, F. Buchinger, J. E. Crawford, X. Feng, S. Gulick, J. K. P. Lee, P. Martinez, R. B. Moore, G. Hackman, D. Hofman, G. Savard, D. Seweryniak, J. Uusitalo, and J. C. Hardy, “Status of the Canadian Penning Trap Mass Spectrometer at the Argonne National Laboratories,” *AIP Conference Proceedings*, vol. 455, 12 1998.
- [35] J. Wang, G. Savard, K. Sharma, J. Clark, Z. Zhou, A. Levand, C. Boudreau, F. Buchinger, J. Crawford, J. Greene, S. Gulick, J. Lee, G. Sprouse, W. Trimble, J. Vaz, and B. Zabransky, “The Canadian Penning Trap Mass Spectrometer,” *Nuclear Physics A*, vol. 746, pp. 651–654, 2004. Proceedings of the Sixth International Conference on Radioactive Nuclear Beams (RNB6).
- [36] T. Eronen, V. S. Kolhinen, V. V. Elomaa, D. Gorelov, U. Hager, J. Hakala, A. Jokinen, A. Kankainen, P. Karvonen, S. Kopecky, I. D. Moore, H. Penttilä, S. Rahaman, S. Rinta-Antila, J. Rissanen, A. Saastamoinen, J. Szerypo, C. Weber, and J. Äystö, “JYFLTRAP: A Penning Trap for Precision Mass Spectroscopy and Isobaric Purification,” *The European physical journal. A, Hadrons and nuclei*, vol. 48, no. 4, pp. 1–21, 2012.
- [37] R. Ringle, S. Schwarz, and G. Bollen, “Penning trap mass spectrometry of rare isotopes produced via projectile fragmentation at the lebit facility,” *International Journal of Mass Spectrometry*, vol. 349-350, pp. 87–93, 2013. 100 years of Mass Spectrometry.
- [38] J. Dilling, K. Blaum, M. Brodeur, and S. Eliseev, “Penning-Trap Mass Measure-

- ments in Atomic and Nuclear Physics,” *Annual Review of Nuclear and Particle Science*, vol. 68, no. 1, pp. 45–74, 2018.
- [39] F. G. Major, V. N. Gheorghe, and G. Werth, *Charged Particle Traps: Physics and Techniques of Charged Particle Field Confinement*, vol. 37. Springer-Verlag Berlin Heidelberg, 2005.
- [40] L. S. Brown and G. Gabrielse, “Geonium Theory: Physics of a Single Electron or Ion in a Penning Trap,” *Reviews of Modern Physics*, vol. 58, no. 1, pp. 233–311, 1986.
- [41] G. W. Fouad G. Major, Viorica N. Gheorghe, *Charged Particle Traps*. Springer-Verlag Berlin Heidelberg, 2005.
- [42] R. Ringle, G. Bollen, A. Prinke, J. Savory, P. Schury, S. Schwarz, and T. Sun, “A “Lorentz” Steerer for Ion Injection into a Penning Trap,” *International Journal of Mass Spectrometry*, vol. 263, no. 1, pp. 38 – 44, 2007.
- [43] M. König, G. Bollen, H.-J. Kluge, T. Otto, and J. Szerypo, “Quadrupole Excitation of Stored Ion Motion at the True Cyclotron Frequency,” *International Journal of Mass Spectrometry and Ion Processes*, vol. 142, no. 1, pp. 95 – 116, 1995.
- [44] S. Eliseev, K. Blaum, M. Block, C. Droese, M. Goncharov, E. Minaya Ramirez, D. A. Nesterenko, Y. N. Novikov, and L. Schweikhard, “Phase-Imaging Ion-Cyclotron-Resonance Measurements for Short-Lived Nuclides,” *Physical Review Letters*, vol. 110, p. 082501, Feb 2013.
- [45] G. Bollen, “Mass Measurements of Short-Lived Nuclides with Ion Traps,” *Nuclear Physics A*, vol. 693, no. 1, pp. 3 – 18, 2001. Radioactive Nuclear Beams.
- [46] M. Brodeur, *First Direct Mass Measurement of the Two and Four Neutron Halos ${}^6\text{He}$ and ${}^8\text{He}$ Using the TITAN Penning Trap Mass Spectrometer*. PhD thesis, University of British Columbia, 2010.

- [47] B. Mamyrin, V. Karataev, D. Shmikk, and V. Zagulin, “The Mass-Reflectron, a New Nonmagnetic Time-of-Flight Mass Spectrometer with High Resolution,” *Zh. Eksp. Teor. Fiz*, vol. 64, no. 1, pp. 82–89, 1973.
- [48] M. Reiter, S. A. S. Andrés, J. Bergmann, T. Dickel, J. Dilling, A. Jacobs, A. Kwiatkowski, W. Plaß, C. Scheidenberger, D. Short, C. Will, C. Babcock, E. Dunling, A. Finlay, C. Hornung, C. Jesch, R. Klawitter, B. Kootte, D. Lascar, E. Leistenschneider, T. Murböck, S. Paul, and M. Yavor, “Commissioning and Performance of TITAN’s Multiple-Reflection Time-of-Flight Mass-Spectrometer and Isobar Separator,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 1018, p. 165823, 2021.
- [49] A. Jacobs, “Collision Induced Dissociation and Mass Spectrometry with the TITAN Multiple-Reflection Time-of-Flight Mass-Spectrometer,” Master’s thesis, University of British Columbia, 2019.
- [50] S. Ayet San Andrés *et al.*, “High-resolution, accurate MR-TOF-MS for short-lived, exotic nuclei of few events in their ground and low-lying isomeric states,” *Physical Review C*, vol. 99, no. 6, p. 064313, 2019.
- [51] A. A. Kwiatkowski, C. Andreoiu, J. C. Bale, T. Brunner, A. Chaudhuri, U. Chowdhury, P. Delheij, S. Ettenauer, D. Frekers, A. T. Gallant, A. Grossheim, G. Gwinner, F. Jang, A. Lennarz, T. Ma, E. Mané, M. R. Pearson, B. E. Schultz, M. C. Simon, V. V. Simon, and J. Dilling, *TITAN: An Ion Trap Facility for On-line Mass Measurement Experiments*, pp. 143–155. Dordrecht: Springer Netherlands, 2014.
- [52] T. Dickel, S. A. San Andres, S. Beck, J. Bergmann, J. Dilling, F. Greiner, and C. Hornung, “Recent Upgrades of the Multiple-Reflection Time-of-Flight Mass Spectrometer at TITAN, TRIUMF.(Report),” *Hyperfine Interactions*, vol. 240, no. 1, pp. 1–9, 2019.

- [53] J. Dilling, P. Bricault, M. Smith, and H.-J. Kluge, “The Proposed TITAN Facility at ISAC for Very Precise Mass Measurements on Highly Charged Short-Lived Isotopes,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 204, pp. 492 – 496, 2003. 14th International Conference on Electromagnetic Isotope Separators and Techniques Related to their Applications.
- [54] V. L. Ryjkov, M. Brodeur, T. Brunner, M. Smith, R. Ringle, A. Lapierre, F. Ames, P. Bricault, M. Domsby, P. Delheij, D. Lunney, M. R. Pearson, and J. Dilling, “Direct Mass Measurement of the Four-Neutron Halo Nuclide ^8He ,” *Physical Review Letters*, vol. 101, p. 012501, Jul 2008.
- [55] M. Smith, M. Brodeur, T. Brunner, S. Ettenauer, A. Lapierre, R. Ringle, V. L. Ryjkov, F. Ames, P. Bricault, G. W. F. Drake, P. Delheij, D. Lunney, F. Sarazin, and J. Dilling, “First Penning-Trap Mass Measurement of the Exotic Halo Nucleus ^{11}Li ,” *Physical Review Letters*, vol. 101, p. 202501, Nov 2008.
- [56] M. P. Reiter, K. G. Leach, O. M. Drozdowski, S. R. Stroberg, J. D. Holt, C. Andreoiu, C. Babcock, B. Barquest, M. Brodeur, A. Finlay, M. Foster, A. T. Gallant, G. Gwinner, R. Klawitter, B. Kootte, A. A. Kwiatkowski, Y. Lan, D. Lascar, E. Leistenschneider, A. Lennarz, S. Paul, R. Steinbrügge, R. I. Thompson, M. Wieser, and J. Dilling, “High-Precision Q_{EC} -Value Measurement of the Superallowed β^+ Emitter ^{22}Mg and an Ab Initio Evaluation of the $A = 22$ Isobaric Triplet,” *Physical Review C*, vol. 96, p. 052501, Nov 2017.
- [57] C. Izzo, J. Bergmann, K. A. Dietrich, E. Dunling, D. Fusco, A. Jacobs, B. Kootte, G. Kripkó-Koncz, Y. Lan, E. Leistenschneider, E. M. Lykiardopoulou, I. Mukul, S. F. Paul, M. P. Reiter, J. L. Tracy, C. Andreoiu, T. Brunner, T. Dickel, J. Dilling, I. Dillmann, G. Gwinner, D. Lascar, K. G. Leach, W. R. Plaß, C. Scheidenberger, M. E. Wieser, and A. A. Kwiatkowski, “Mass Measurements of Neutron-Rich Indium Isotopes for r-Process Studies,” *Physical Review. C*, vol. 103, no. 2, 2021.

- [58] A. Lapierre, M. Brodeur, T. Brunner, S. Ettenauer, A. Gallant, V. Simon, M. Good, M. Froese, J. Crespo López-Urrutia, P. Delheij, S. Epp, R. Ringle, S. Schwarz, J. Ullrich, and J. Dilling, “The TITAN EBIT Charge Breeder for Mass Measurements on Highly Charged Short-Lived Isotopes—First Online Operation,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 624, no. 1, pp. 54–64, 2010.
- [59] S. Ettenauer, M. C. Simon, A. T. Gallant, T. Brunner, U. Chowdhury, V. V. Simon, M. Brodeur, A. Chaudhuri, E. Mané, C. Andreoiu, G. Audi, J. R. C. López-Urrutia, P. Delheij, G. Gwinner, A. Lapierre, D. Lunney, M. R. Pearson, R. Ringle, J. Ullrich, and J. Dilling, “First Use of High Charge States for Mass Measurements of Short-Lived Nuclides in a Penning Trap,” *Physical Review Letters*, vol. 107, p. 272501, Dec 2011.
- [60] E. Leistenschneider, *Dawning of nuclear magicity in $N = 32$ seen through precision mass spectrometry*. PhD thesis, University of British Columbia, 2019.
- [61] F. Herfurth, J. Dilling, A. Kellerbauer, G. Bollen, S. Henry, H.-J. Kluge, E. Lamour, D. Lunney, R. Moore, C. Scheidenberger, S. Schwarz, G. Sikler, and J. Szerypo, “A Linear Radiofrequency Ion Trap for Accumulation, Bunching, and Emittance Improvement of Radioactive Ion Beams,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 469, no. 2, pp. 254 – 275, 2001.
- [62] J. Dilling, R. Baartman, P. Bricault, M. Brodeur, L. Blomeley, F. Buchinger, J. Crawford, J. C. López-Urrutia, P. Delheij, M. Froese, G. Gwinner, Z. Ke, J. Lee, R. Moore, V. Ryjgov, G. Sikler, M. Smith, J. Ullrich, and J. Vaz, “Mass Measurements on Highly Charged Radioactive Ions, a New Approach to High Precision with TITAN,” *International Journal of Mass Spectrometry*, vol. 251, no. 2, pp. 198 – 203, 2006. ULTRA-ACCURATE MASS SPECTROMETRY AND RELATED TOPICS Dedicated to H.-J. Kluge on the occasion of his 65th birthday anniversary.

- [63] E. Leistenschneider, M. P. Reiter, S. Ayet San Andrés, B. Kootte, J. D. Holt, P. Navrátil, C. Babcock, C. Barbieri, B. R. Barquest, J. Bergmann, J. Bollig, T. Brunner, E. Dunling, A. Finlay, H. Geissel, L. Graham, F. Greiner, H. Hergert, C. Hornung, C. Jesch, R. Klawitter, Y. Lan, D. Lascar, K. G. Leach, W. Lippert, J. E. McKay, S. F. Paul, A. Schwenk, D. Short, J. Simonis, V. Somà, R. Steinbrügge, S. R. Stroberg, R. Thompson, M. E. Wieser, C. Will, M. Yavor, C. Andreoiu, T. Dickel, I. Dillmann, G. Gwinner, W. R. Plaß, C. Scheidenberger, A. A. Kwiatkowski, and J. Dilling, “Dawning of the $N = 32$ Shell Closure Seen through Precision Mass Measurements of Neutron-Rich Titanium Isotopes,” *Physical Review Letters*, vol. 120, p. 062503, Feb 2018.
- [64] Y. Ito, P. Schury, M. Wada, S. Naimi, T. Sonoda, H. Mita, F. Arai, A. Takamine, K. Okada, A. Ozawa, and H. Wollnik, “Single-Reference High-Precision Mass Measurement with a Multi-Reflection Time-of-Flight Mass Spectrograph,” *Physical Review C*, vol. 88, 07 2013.
- [65] P. Schury, M. Wada, Y. Ito, D. Kaji, F. Arai, M. MacCormick, I. Murray, H. Haba, S. Jeong, S. Kimura, H. Koura, H. Miyatake, K. Morimoto, K. Morita, A. Ozawa, M. Rosenbusch, M. Reponen, P.-A. Söderström, A. Takamine, T. Tanaka, and H. Wollnik, “First Online Multireflection Time-of-Flight Mass Measurements of Isobar Chains Produced by Fusion-Evaporation Reactions: Toward Identification of Superheavy Elements via Mass Spectroscopy,” *Physical Review C*, vol. 95, no. 1, 2017.
- [66] R. N. Wolf, F. Wienholtz, D. Atanasov, D. Beck, K. Blaum, C. Borgmann, F. Herfurth, M. Kowalska, S. Kreim, Y. A. Litvinov, D. Lunney, V. Manea, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, and K. Zuber, “ISOLTRAP’s Multi-Reflection Time-of-Flight Mass Separator/Spectrometer,” *International Journal of Mass Spectrometry*, vol. 349-350, pp. 123–133, 2013.
- [67] T. Y. Hirsh, N. Paul, M. Burkey, A. Aprahamian, F. Buchinger, S. Caldwell, J. A. Clark, A. F. Levand, L. L. Ying, S. T. Marley, G. E. Morgan, A. Nystrom,

- R. Orford, A. P. Galván, J. Rohrer, G. Savard, K. S. Sharma, and K. Siegl, “First Operation and Mass Separation with the CARIBU MR-TOF,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 376, pp. 229 – 232, 2016. Proceedings of the XVIIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11-15 May 2015.
- [68] C. Jesch, *The Multiple-Reflection Time-of-Flight Isobar Separator for TITAN and Direct Mass Measurements at the FRS Ion Catcher*. PhD thesis, Justus-Liebig-Universität Gießen, 06 2016.
- [69] A. Finlay, “Integration of a multi reflection time of flight isobar separator into the titan experiment at triumph,” Master’s thesis, University of British Columbia, 2017.
- [70] D. Short, “Nuclear Isobar Separation for Penning Trap Mass Measurements at TRIUMF,” Master’s thesis, Simon Fraser University, 04 2018.
- [71] C. Jesch, T. Dickel, W. R. Plaß, D. Short, S. A. San Andres, J. Dilling, H. Geissel, F. Greiner, J. Lang, K. G. Leach, *et al.*, “The MR-TOF-MS Isobar Separator for the TITAN Facility at TRIUMF,” in *TCP 2014*, pp. 175–184, Springer, 2017.
- [72] T. Dickel, W. Plaß, J. Lang, J. Ebert, H. Geissel, E. Haettner, C. Jesch, W. Lipert, M. Petrick, C. Scheidenberger, and M. Yavor, “Multiple-Reflection Time-of-Flight Mass Spectrometry for In Situ Applications,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 317, pp. 779–784, 2013. XVIth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 2–7, 2012 at Matsue, Japan.
- [73] D. Stresau, K. Hunter, W. Sheils, P. Raffin, and Y. Benari, “A New Class of Robust Sub-Nanosecond TOF Detectors with High Dynamic Range,” in *54th ASMS Conference Proceedings*, 2006.

- [74] A. Takkinen, *Characterization of MagneTOF Ion Detector and Bradbury-Nielsen Ion Gate*. PhD thesis, University of Jyväskylä, 2018.
- [75] R. Bass, *Investigating Systematic Position Effects in TITAN's Multiple-Reflection Time-of-Flight Mass Spectrometer*. PhD thesis, The University of Edinburgh, 2020.
- [76] J. Bergmann, "Masters thesis," Master's thesis, Justus-Liebig-Universität Gießen, 2015.
- [77] T. Dickel, S. A. San Andrés, S. Beck, J. Bergmann, J. Dilling, F. Greiner, C. Hornung, A. Jacobs, G. Kripko-Koncz, A. Kwiatkowski, *et al.*, "Recent Upgrades of the Multiple-Reflection Time-of-Flight Mass Spectrometer at TITAN, TRIUMF," *Hyperfine Interactions*, vol. 240, no. 1, pp. 1–9, 2019.
- [78] M. Mukherjee, D. Beck, K. Blaum, G. Bollen, J. Dilling, S. George, F. Herfurth, A. Herlert, A. Kellerbauer, H. J. Kluge, S. Schwarz, L. Schweikhard, and C. Yazidjian, "ISOLTRAP: An On-Line Penning Trap for Mass Spectrometry on Short-Lived Nuclides," *The European Physical Journal A*, vol. 35, no. 1, pp. 1–29, 2008.
- [79] T. Dickel, W. R. Plaß, W. Lippert, J. Lang, M. I. Yavor, H. Geissel, and C. Scheidenberger, "Isobar Separation in a Multiple-Reflection Time-of-Flight Mass Spectrometer by Mass-Selective Re-Trapping," *Journal of The American Society for Mass Spectrometry*, vol. 28, no. 6, pp. 1079–1090, 2017.
- [80] N. E. Bradbury and R. A. Nielsen, "Absolute Values of the Electron Mobility in Hydrogen," *Physical Review*, vol. 49, no. 5, pp. 388–393, 1936.
- [81] B. E. Schultz, M. Brodeur, C. Andreoiu, A. Bader, A. Chaudhuri, U. Chowdhury, A. T. Gallant, A. Grossheim, R. Klawitter, A. A. Kwiatkowski, K. G. Leach, A. Lennarz, T. D. Macdonald, J. Lassen, H. Heggen, S. Raeder, A. Teigelhöfer, and J. Dilling, "Precision Q_{EC} -Value Measurement of ^{23}Mg for Testing the Cabibbo-Kobayashi-Maskawa Matrix Unitarity," *Physical Review C*, vol. 90, p. 012501, Jul 2014.

- [82] C. Babcock, R. Klawitter, E. Leistenschneider, D. Lascar, B. R. Barquest, A. Finlay, M. Foster, A. T. Gallant, P. Hunt, B. Kootte, Y. Lan, S. F. Paul, M. L. Phan, M. P. Reiter, B. Schultz, D. Short, C. Andreoiu, M. Brodeur, I. Dillmann, G. Gwinner, A. A. Kwiatkowski, K. G. Leach, and J. Dilling, “Mass Measurements of Neutron-Rich Indium Isotopes Toward the $N = 82$ Shell Closure,” *Physical Review C*, vol. 97, p. 024312, Feb 2018.
- [83] F. Currell and G. Fussmann, “Physics of Electron Beam Ion Traps and Sources,” *IEEE Transactions on Plasma Science*, vol. 33, no. 6, pp. 1763–1777, 2005.
- [84] G. Sikler, J. R. Crespo López-Urrutia, J. Dilling, S. Epp, C. J. Osborne, and J. Ullrich, “A High-Current EBIT for Charge-Breeding of Radionuclides for the TITAN Spectrometer,” *The European Physical Journal. A, Hadrons and Nuclei*, vol. 25, no. S1, pp. 63–64, 2005.
- [85] A. T. Gallant, M. Brodeur, T. Brunner, S. Ettenauer, M. Good, A. Lapierre, R. Ringle, V. V. Simon, P. Delheij, and J. Dilling, “TITAN-EBIT — Charge Breeding of Radioactive Isotopes for High Precision Mass Measurements,” *Journal of Instrumentation*, vol. 5, pp. C08009–C08009, Aug 2010.
- [86] F. J. Currell, F. J. Currell, H. Kuramoto, S. Ohtani, C. Scullion, E. J. Sokell, and H. Watanabe, “Measurements of Temperature Dynamics of Ions Trapped Inside an Electron Beam Ion Trap and Evidence for Ionisation Heating,” *Physica Scripta*, vol. T92, no. 1, pp. 147–149, 2001.
- [87] S. Malbrunot-Ettenauer, T. Brunner, U. Chowdhury, A. Gallant, V. V. Simon, M. Brodeur, A. Chaudhuri, E. Mané, M. Simon, C. Andreoiu, *et al.*, “Penning Trap Mass Measurements Utilizing Highly Charged Ions as a Path to Benchmark Isospin-Symmetry Breaking Corrections in Rb 74,” *Physical Review C*, vol. 91, no. 4, p. 045504, 2015.
- [88] D. Lascar, R. Klawitter, C. Babcock, E. Leistenschneider, S. R. Stroberg, B. R. Barquest, A. Finlay, M. Foster, A. T. Gallant, P. Hunt, J. Kelly, B. Kootte,

- Y. Lan, S. F. Paul, M. L. Phan, M. P. Reiter, B. Schultz, D. Short, J. Simonis, C. Andreoiu, M. Brodeur, I. Dillmann, G. Gwinner, J. D. Holt, A. A. Kwiatkowski, K. G. Leach, and J. Dilling, “Precision Mass Measurements of $^{125-127}\text{Cd}$ Isotopes and Isomers Approaching the $N = 82$ Closed Shell,” *Physical Review C*, vol. 96, p. 044323, Oct 2017.
- [89] M. Mostamand, R. Li, J. Romans, F. Ames, P. Kunz, A. Mjøs, and J. Lassen, “Production of clean rare isotope beams at triump ion guide laser ion source,” *Hyperfine Interactions*, vol. 241, no. 1, pp. 1–9, 2020.
- [90] S. Beck, B. Kootte, I. Dedes, T. Dickel, A. A. Kwiatkowski, E. M. Lykiardopoulou, W. R. Plaß, M. P. Reiter, C. Andreoiu, J. Bergmann, T. Brunner, D. Curien, J. Dilling, J. Dudek, E. Dunling, J. Flowerdew, A. Gaamouci, L. Graham, G. Gwinner, A. Jacobs, R. Klawitter, Y. Lan, E. Leistenschneider, N. Minkov, V. Monier, I. Mukul, S. F. Paul, C. Scheidenberger, R. I. Thompson, J. L. Tracy, M. Vansteenkiste, H.-L. Wang, M. E. Wieser, C. Will, and J. Yang, “Mass Measurements of Neutron-Deficient Yb Isotopes and Nuclear Structure at the Extreme Proton-Rich Side of the $N=82$ Shell,” *Physical Review Letters*, vol. 127, no. 11, pp. 112501–112501, 2021.
- [91] S. Purushothaman, S. Ayet San Andrés, J. Bergmann, T. Dickel, J. Ebert, H. Geissel, C. Hornung, W. Plaß, C. Rappold, C. Scheidenberger, Y. Tanaka, and M. Yavor, “Hyper-EMG: A New Probability Distribution Function Composed of Exponentially Modified Gaussian Distributions to Analyze Asymmetric Peak Shapes in High-Resolution Time-of-Flight Mass Spectrometry,” *International Journal of Mass Spectrometry*, vol. 421, pp. 245 – 254, 2017.
- [92] E. Dunling, *Exploring Magicity Around $N = 32$ & 34 in $Z \geq 20$ Isotopes via Precision Mass Measurements and Developments with the TITAN MR-TOF Mass Spectrometer*. PhD thesis, University of York, February 2021.
- [93] J. Kartheim, *Next-Generation Mass Spectrometry of Exotic Isotopes and Isomers*. PhD thesis, Ruprecht-Karls-Universität, Heidelberg, 2020.

- [94] M. Wang, W. Huang, F. Kondev, G. Audi, and S. Naimi, “The AME 2020 Atomic Mass Evaluation (II). Tables, Graphs and References,” *Chinese Physics C*, vol. 45, p. 030003, Mar 2021.
- [95] C. Rauth, D. Ackermann, K. Blaum, M. Block, A. Chaudhuri, Z. Di, S. Eliseev, R. Ferrer, D. Habs, F. Herfurth, F. P. Heßberger, S. Hofmann, H.-J. Kluge, G. Maero, A. Martín, G. Marx, M. Mukherjee, J. B. Neumayr, W. R. Plaß, S. Rahaman, D. Rodríguez, C. Scheidenberger, L. Schweikhard, P. G. Thirolf, G. Vorobjev, and C. Weber, “First Penning Trap Mass Measurements beyond the Proton Drip Line,” *Physical Review Letters*, vol. 100, p. 012501, Jan 2008.
- [96] R. Irvine, C. Davids, P. Woods, D. Blumenthal, L. Brown, L. Conticchio, T. Davinson, D. Henderson, J. Mackenzie, H. Penttilä, *et al.*, “Proton Emission from Drip-Line Nuclei ^{157}Ta and ^{161}Re ,” *Physical Review C*, vol. 55, no. 4, p. R1621, 1997.
- [97] E. M. Lykiardopoulou, G. Audi, T. Dickel, W. J. Huang, D. Lunney, W. R. Plaß, M. P. Reiter, J. Dilling, and A. A. Kwiatkowski, “Exploring the limits of existence of proton-rich nuclei in the $z = 70 - -82$ region,” *Phys. Rev. C*, vol. 107, p. 024311, Feb 2023.
- [98] E. C. Simpson, “The Colourful Nuclide Chart.” <https://people.physics.anu.edu.au/~ecs103/chart/>.
- [99] J. Erler, N. Birge, M. Kortelainen, W. Nazarewicz, E. Olsen, A. M. Perhac, and M. Stoitsov, “The Limits of the Nuclear Landscape,” *Nature (London)*, vol. 486, no. 7404, pp. 509–512, 2012.
- [100] Y. A. Litvinov, H. Geissel, T. Radon, F. Attallah, G. Audi, K. Beckert, F. Bosch, M. Falch, B. Franzke, M. Hausmann, *et al.*, “Mass Measurement of Cooled Neutron-Deficient Bismuth Projectile Fragments with Time-Resolved Schottky Mass Spectrometry at the FRS-ESR Facility,” *Nuclear Physics A*, vol. 756, no. 1-2, pp. 3–38, 2005.

- [101] G. Audi, F. Kondev, M. Wang, W. Huang, and S. Naimi, “The NUBASE2016 Evaluation of Nuclear Properties,” *Chinese Physics C*, vol. 41, no. 3, p. 030001, 2017.
- [102] K. S. Toth, Y. A. Ellis-Akovali, F. T. Avignone, R. S. Moore, D. M. Moltz, J. M. Nitschke, P. A. Wilmarth, P. K. Lemmertz, D. C. Sousa, and A. L. Goodman, “Single-Particle States in ^{149}Er and ^{149}Ho , and the Effect of the $Z=64$ Closure,” *Physical Review C*, vol. 32, pp. 342–345, Jul 1985.
- [103] R. B. Firestone, J. M. Nitschke, P. A. Wilmarth, K. Vierinen, J. Gilat, K. S. Toth, and Y. A. Akovali, “Decay of $^{149}\text{Er}^{g+m}$ by Positron and Delayed Proton Emission and by Electron Capture,” *Physical Review C*, vol. 39, pp. 219–232, Jan 1989.
- [104] K. Yao and R. Schuch, “Evaporative Cooling of Hydrogen Molecular Ions in a Penning Trap,” *Hyperfine interactions*, vol. 227, no. 1-3, pp. 191–196, 2014.
- [105] L. Schmöger, O. O. Versolato, M. Schwarz, M. Kohnen, A. Windberger, B. Piest, S. Feuchtenbeiner, J. Pedregosa-Gutierrez, T. Leopold, P. Micke, A. K. Hansen, T. M. Baumann, M. Drewsen, J. Ullrich, P. O. Schmidt, and J. R. C. López-Urrutia, “Coulomb Crystallization of Highly Charged Ions,” *Science (American Association for the Advancement of Science)*, vol. 347, no. 6227, pp. 1233–1236, 2015.
- [106] L. Schmöger, M. Schwarz, T. M. Baumann, O. O. Versolato, B. Piest, T. Pfeifer, J. Ullrich, P. O. Schmidt, and J. R. Crespo López-Urrutia, “Deceleration, Pre-cooling, and Multi-Pass Stopping of Highly Charged Ions in Be^+ Coulomb Crystals,” *Review of Scientific Instruments*, vol. 86, no. 10, 2015.
- [107] M. Bussmann, U. Schramm, D. Habs, V. Kolhinen, and J. Szerypo, “Stopping Highly Charged Ions in a Laser-Cooled One Component Plasma of Ions,” *International Journal of Mass Spectrometry*, vol. 251, no. 2-3, pp. 179–189, 2006.
- [108] D. Griffiths, *Introduction to Electrodynamics*. Cambridge University Press, 4 ed., 2017.

- [109] C. Amole, G. Andresen, M. Ashkezari, M. Baquero-Ruiz, W. Bertsche, P. Bowe, E. Butler, A. Capra, P. Carpenter, C. Cesar, *et al.*, “The ALPHA Antihydrogen Trapping Apparatus,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 735, pp. 319–340, 2014.
- [110] M. Steck, K. Beckert, H. Eickhoff, B. Franzke, F. Nolden, and P. Spadtke, “Electron Cooling of Heavy Ions at GSI,” in *Proceedings of International Conference on Particle Accelerators*, pp. 1738–1740 vol.3, 1993.
- [111] T. Radon, T. Kerscher, B. Schlitt, K. Beckert, T. Beha, F. Bosch, H. Eickhoff, B. Franzke, Y. Fujita, H. Geissel, M. Hausmann, H. Irnich, H. C. Jung, O. Klepper, H.-J. Kluge, C. Kozhuharov, G. Kraus, K. E. G. Löbner, G. Münzenberg, Y. Novikov, F. Nickel, F. Nolden, Z. Patyk, H. Reich, C. Scheidenberger, W. Schwab, M. Steck, K. Sümmerer, and H. Wollnik, “Schottky Mass Measurements of Cooled Proton-Rich Nuclei at the GSI Experimental Storage Ring,” *Physical Review Letters*, vol. 78, pp. 4701–4704, Jun 1997.
- [112] W. Quint, J. Dilling, S. Djekic, H. Häffner, N. Hermanspahn, H.-J. Kluge, G. Marx, R. Moore, D. Rodriguez, J. Schönfelder, *et al.*, “HITRAP: A Facility for Experiments with Trapped Highly Charged Ions,” *Hyperfine Interactions*, vol. 132, no. 1, pp. 453–457, 2001.
- [113] Z. Andelkovic, G. Birkel, S. Fedotova, V. Hannen, F. Herfurth, K. König, N. Kotovskiy, B. Maaß, J. Vollbrecht, T. Murböck, *et al.*, “Status of Deceleration and Laser Spectroscopy of Highly Charged Ions at HITRAP,” *Hyperfine Interactions*, vol. 235, no. 1-3, pp. 37–44, 2015.
- [114] F. M. Fränkle, F. Glück, K. Valerius, K. Bokeloh, A. Beglarian, J. Bonn, L. Bornschein, G. Drexlin, F. Habermehl, M. L. Leber, A. Osipowicz, E. W. Otten, M. Steidl, T. Thümmel, C. Weinheimer, J. F. Wilkerson, J. Wolf, and S. V. Zadorozhny, “Penning Discharge in the KATRIN Pre-Spectrometer,” *Journal of Instrumentation*, vol. 9, pp. P07028–P07028, Jul 2014.

- [115] M. Aker, K. Altenmüller, A. Beglarian, J. Behrens, A. Berlev, U. Besserer, K. Blaum, F. Block, S. Bobien, B. Bornschein, *et al.*, “Suppression of Penning Discharges Between the KATRIN Spectrometers,” *The European Physical Journal C*, vol. 80, no. 9, pp. 1–12, 2020.
- [116] M. A. M. A. Lieberman, *Principles of Plasma Discharges and Materials Processing*. Hoboken, N.J: Wiley-Interscience, 2nd ed. ed., 2005.
- [117] N. V. Mamedov, A. S. Rohmanenkov, and A. A. Solodovnikov, “Magnetic Field Influence on the Penning Discharge Characteristics,” *Journal of Physics: Conference Series*, vol. 2064, p. 012039, Nov 2021.
- [118] E. Kay, “Magnetic Field Effects on an Abnormal Truncated Glow Discharge and Their Relation to Sputtered Thin-Film Growth,” *Journal of Applied Physics*, vol. 34, no. 4, pp. 760–768, 1963.
- [119] W. D. Gill and E. Kay, “Efficient Low Pressure Sputtering in a Large Inverted Magnetron Suitable for Film Synthesis,” *Review of Scientific Instruments*, vol. 36, no. 3, pp. 277–282, 1965.
- [120] K. Wasa and S. Hayakawa, “Low Pressure Sputtering System of the Magnetron Type,” *Review of Scientific Instruments*, vol. 40, no. 5, pp. 693–697, 1969.
- [121] S. I. Ivanov, “Formative Time of Penning Discharge,” *International Journal of Electronics*, vol. 34, no. 6, pp. 769–775, 1973.
- [122] F. M. Fränkle, F. Glück, K. Valerius, K. Bokeloh, A. Beglarian, J. Bonn, L. Bornschein, G. Drexlin, F. Habermehl, M. L. Leber, A. Osipowicz, E. W. Otten, M. Steidl, T. Thümmel, C. Weinheimer, J. F. Wilkerson, J. Wolf, and S. V. Zadorozhny, “Penning Discharge in the KATRIN Pre-Spectrometer,” *Journal of Instrumentation*, vol. 9, no. 7, pp. P07028–P07028, 2014.
- [123] U. Chowdhury, *A Cooler Penning Trap to Cool Highly Charged Radioactive Ions and Mass Measurement of ^{24}Al* . PhD thesis, University of Manitoba, Winnipeg, 7 2016.

- [124] M. Ahmadi, B. X. Alves, C. J. Baker, W. Bertsche, A. Capra, C. Carruth, C. L. Cesar, M. Charlton, S. Cohen, R. Collister, S. Eriksson, A. Evans, N. Evetts, J. Fajans, T. Friesen, M. C. Fujiwara, D. R. Gill, J. S. Hangst, W. N. Hardy, M. E. Hayden, C. A. Isaac, M. A. Johnson, S. A. Jones, S. Jonsell, L. Kurchaninov, N. Madsen, M. Mathers, D. Maxwell, J. T. McKenna, S. Menary, T. Momose, J. J. Munich, K. Olchanski, A. Olin, P. Pusa, C. Rasmussen, F. Robicheaux, R. L. Sacramento, M. Sameed, E. Sarid, D. M. Silveira, C. So, G. Stutter, T. D. Tharp, J. E. Thompson, R. I. Thompson, D. P. Van Der Werf, and J. S. Wurtele, “Enhanced Control and Reproducibility of Non-Neutral Plasmas,” *Physical Review Letters*, vol. 120, no. 2, p. 25001, 2018.
- [125] T. Kato, H. Himura, S. Sowa, and A. Sanpei, “Controlling the Diameter of a Pure Electron Plasma to Produce an Exact Two-Fluid Plasma State in a Nested Trap,” *Plasma and Fusion Research*, vol. 14, pp. 1201039–1201039, 2019.
- [126] G. Maero, B. Paroli, R. Pozzoli, and M. Romé, “Stabilizing Effect of a Non-resonant Radio Frequency Drive on the $m=1$ Diocotron Instability,” *Physics of Plasmas*, vol. 18, p. 032101, Mar. 2011.
- [127] H. Himura, “Recent Experiments with Lithium Ion and Electron Plasmas in the BX-U Linear Trap,” vol. 1928, p. 020005, 01 2018.
- [128] H. Himura, S. Yamada, T. Kato, and S. Masamune, “First Observation of Independent Ion and Electron Plasmas after Elapse of Two-Fluid Plasma State,” *Plasma and Fusion Research*, vol. 12, pp. 1–3, 2017.
- [129] S. F. Paul, B. Kootte, D. Lascar, A. A. Kwiatkowski, G. Gwinner, J. Dilling, and the TITAN collaboration, “Off-Axis Electron Injection Into a Cooler Penning Trap,” *Hyperfine Interactions*, vol. 240, no. 1, p. 50, 2019.
- [130] U. Chowdhury, M. Good, B. Kootte, D. Lascar, B. E. Schultz, J. Dilling, and G. Gwinner, “A Cooler Penning Trap for the TITAN Mass Measurement Facility,” *AIP Conference Proceedings*, vol. 1640, pp. 120–123, JAN 2015.

- [131] D. Lascar, B. Kootte, B. Barquest, U. Chowdhury, A. Gallant, M. Good, R. Klawitter, E. Leistenschneider, C. Andreoiu, J. Dilling, J. Even, G. Gwinner, A. Kwiatkowski, and K. Leach, “A Novel Transparent Charged Particle Detector for the CPET Upgrade at TITAN,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 868, pp. 133 – 138, 2017.
- [132] B. Paroli, F. De Luca, G. Maero, R. Pozzoli, and M. Romé, “Radio Frequency Generation of an Electron Plasma in a Malmberg-Penning Trap,” in *AIP Conference Proceedings*, vol. 1242, pp. 343–348, American Institute of Physics, 2010.
- [133] B. Paroli, F. D. Luca, G. Maero, R. Pozzoli, and M. Romé, “Broadband Radio Frequency Plasma Generation in a Penning–Malmberg trap,” *Plasma Sources Science and Technology*, vol. 19, p. 045013, Jun 2010.
- [134] T. Mohamed, “Successful production of non-neutral electron plasma of high density in the multi-ring trap,” *Plasma Devices and Operations*, vol. 16, no. 3, pp. 181–188, 2008.
- [135] B. Kootte, M. Alanssari, C. Andreoiu, J. Bale, B. Barquest, U. Chowdhury, J. Dilling, J. Even, A. Finlay, D. Frekers, A. Gallant, G. Gwinner, R. Klawitter, R. Schupp, A. A. Kwiatkowski, D. Lascar, K. G. Leach, E. Leistenschneider, A. Lennarz, T. Li, A. J. Mayer, and D. Short, “Quantification of the Electron Plasma in TITAN’s Cooler Penning Trap,” *Int. Workshop on Beam Cooling and Related Topics (COOL’15)*, pp. 39–42, 2016.
- [136] F. Penning and J. Moubis, “Eine Neutronenröhre Ohne Pumpvorrichtung,” *Physica*, vol. 4, no. 11, pp. 1190–1199, 1937.
- [137] N. V. Mamedov, S. P. Maslennikov, A. A. Solodovnikov, and D. I. Yurkov, “Effect of the Magnetic Field on the Characteristics of a Pulsed Penning Ion Source,” *Plasma Physics Reports*, vol. 46, no. 2, pp. 217–229, 2020.
- [138] R. Silwal, S. F. Paul, B. Kootte, T. Joseph, R. Simpson, A. A. Kwiatkowski, G. Gwinner, J. Dilling, and the TITAN collaboration, “Ion Simulations, Re-

cent Upgrades and Tests with TITAN's Cooler Penning Trap," in *ECRIS 2020 Conference Proceedings*, pp. 181–187, 2020.