

**Multiple-Reflection Time-of-Flight Mass-Spectrometry:
Not just for high precision mass measurement**

by

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Abstract

Multiple-Reflection Time-Of-Flight Mass Spectrometers (MR-TOF-MS) are a relatively new tool that has quickly become a standard instrument for high precision mass measurements in nuclear physics. The TRIUMF's Ion Trap for Atomic and Nuclear science (TITAN) MR-TOF-MS and a new technique of “post re-trapping stable calibrant beam merging” is described. The TITAN MR-TOF-MS is used for high precision mass measurements of $^{51-54}\text{K}$, improving the mass uncertainties in all cases and providing a first time direct mass measurement of ^{54}K . The improved mass uncertainties provide evidence for the theorized $N = 34$ subshell closure below Ca on the nuclear chart. The mass measurements are used to compute the empirical shell gap and empirical energy gap in potassium isotopes around $N = 34$. This is compared to theory to strengthen the argument for an $N = 34$ subshell closure.

The capabilities of the TITAN MR-TOF-MS are extended through the development of novel techniques for half-life measurement of short-lived radioactive nuclides. The half-life uncertainty of the isotope ^{54}K is improved through an approach we call “mass tagged β -decays”. This work also presents the first experimentally determined bounds on the n -neutron-delayed β -decay probabilities in ^{54}K . Half-life measurements of $^{180-182}\text{Yb}$ are made through the production and subsequent decay of a “re-trapping purified” radioactive source. This represents the first half-life measurements in ^{181}Yb and ^{182}Yb , demonstrating the potential of this new technique for measuring half-lives of lanthanide isotopes to elucidate nuclear structure and improve astrophysical isotope production simulations. Finally, a set of beam composition measurements for characterization of a new isotope separation online (ISOL) rare isotope production target are presented.

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A thesis represents a long individual academic undertaking, but I am very thankful to have had the emotional support of many people throughout that journey. I cannot thank everyone here, but know that I will always look back on this time fondly.

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I have made many good friends throughout my journey, both in Winnipeg and Vancouver. Moving to Vancouver during the height of COVID-19 lock-downs was not easy and I particularly appreciate Brian, Melissa, Philippe, Tobias and Jonas for welcoming me into Vancouver and inviting me into their bubble. I also want to thank my partner Ivana, with whom I shared the many ups and downs of my graduate school experience.

Last, I would like to acknowledge my parents, who clearly let me read too many popular science books as a young and impressionable child. Thank you for supporting me on this adventure.

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List of Acronyms

χ EFT chiral effective field theory

EBIT Electron Beam Ion Trap

FWHM full width half max

hyper-EMG hyper-exponentially modified Gaussian

IGLIS Ion-Guide Laser Ionization Source

ISAC Isotope Separation and ACceleration

ISOL isotope separation on-line

IT isochronous reflection turns

MCMC Markov-Chain Monte Carlo

MPET Measurement Penning Trap

MR-TOF-MS Multiple-Reflection Time-of-Flight Mass Spectrometer

QCD quantum chromodynamics

RFQ radio frequency quadrupole

RIB rare isotope beam

SCGF self-consistent Gorkov-Greens's function

TDC Time-to-Digital converter

TFS time focus shift

TITAN TRIUMF's Ion Trap for Atomic and Nuclear science

TOF-MS time-of-flight mass spectrometer

TRC time resolved calibration

TRILIS TRIUMF Resonant Laser Ionization Source

UHV ultra high vacuum

VS-IM-SRG valence-space in-medium similarity renormalization group

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Statement of Personal Contributions

Science more than ever before is a collaborative affair. While this PhD thesis is meant to prove a personal contribution to the body of scientific knowledge, I am indebted to the generations of graduate students, postdocs, and other scientists who have built and maintained the experimental equipment I used. For clarity, listed below are the contributions I made that are reported in this thesis, roughly in the order they are presented. Rare isotope beam measurements presented in this thesis were collected with the help of the entire local TITAN collaboration.

- Conception and implementation of direct isochronicity measurements, inspired by similar work done with the KEK-RIKEN Zero-Degree Spectrometer MR-TOF-MS.
- Conception and implementation of stable beam merging with TITAN postdoc Tobias Murböck.
- Mass analysis including calibrations and systematic uncertainty determination following previously established techniques.
- Preparation, shift leadership, and operation of the TITAN MR-TOF-MS collecting data for mass measurement of potassium isotopic chain and half-life measurements of the potassium and ytterbium isotopes.
- Organizing the nuclear structure arguments based on the presented mass measurements, guided by calculations done by our collaborators.

- Techniques for collecting and analyzing radioactive decay data collected on the MR-TOF-MS ion detector. Initial conception and exploratory data presented in section 5.6 were done with Tobias Murböck.
- Data collection, analysis, and experiment strategy with regards to the MR-TOF-MS operation for the P2N yield characterization experiments.

CHAPTER 1

An Introduction to Nuclear Theory

1.1 Introduction

A nucleus is composed of some number of protons and neutrons bound together by the nuclear force. The study of the structure of nuclei is complicated by the large number of particles involved as well as the nature of the nuclear force. It is well known that even in the low energy limit of Quantum Chromodynamics (QCD), the constituents of nuclei (nucleons) experience both two-body and three-body forces [1, 2]. A complete theoretical description of nuclei is therefore a difficult task. Instead, the behaviour of nuclei is described by nuclear theorists in various simplifying models. One very successful family of nuclear models is known (collectively) as the shell model [3]. The shell model was first proposed in 1948 by Maria Goeppert Mayer [4] and 1949 by J. Hans D. Jensen [5], work for which they would later share the Nobel prize.

One of the most basic ways of probing the nature of the nucleus is through atomic mass measurements. The mass of a nuclei with Z protons and N neutrons can be expressed as

$$M_N(Z, N) = Zm_p + Nm_n - E_B(Z, N), \quad (1.1)$$

where m_p is the mass of a proton, m_n is the mass of a neutron and $E_B(Z, N)$ is the binding energy¹ of the nucleon. Since the masses of bare protons and neutrons are well known [6], a measurement of the mass of a nucleon directly probes the binding energy of a nucleon. Historically, the field of precision mass measurement reports atomic masses rather than nuclear masses. This convention is adopted in this thesis. The atomic mass differs from the nuclear mass by the mass of the electrons, less the binding energy of the electrons, denoted in this work as

$$M(Z, N) = M_N + Zm_e - E_e(Z). \quad (1.2)$$

Here m_e is the mass of the electron and $E_e(Z)$ is the sum of the electron binding energies of all electrons in the neutral atom. Since all mass measurements reported in this thesis were performed on singly charged ions, the mass of the electron is added back to the final values reported. The binding energy of a single valence electron is on the order of 10 eV and is negligible compared to the uncertainty in the mass measurement.

Mass measurements also enable the calculation of various derived metrics, such as separation energies and empirical shell gaps. Collectively, metrics obtained from some linear combination of atomic masses are called mass filters. One group of mass filters called proton and neutron separation energies, represent the energy needed to

¹Binding energy is used somewhat loosely here. What is really meant is the mass equivalence given by $E = mc^2$. This is the origin of using eV/ c^2 as a unit of mass.

remove the proton or neutron from the nucleus. They are defined in terms of binding energy as

$$S_p = E_B(Z, N) - E_B(Z - 1, N) \quad (1.3)$$

$$S_n = E_B(Z, N) - E_B(Z, N - 1). \quad (1.4)$$

The empirical shell gap accounts for the trend of decreasing binding energy away from nuclear stability by comparing neighbouring nuclei. It serves as a sensitive indicator of abrupt changes in binding energy associated with the closure of nuclear shells. The empirical shell gap is defined for both neutrons (Δ_{2n}) and protons (Δ_{2p}) as

$$\Delta_{2n} = 2E_B(Z, N) - E_B(Z, N - 2) - E_B(Z, N + 2) \quad (1.5)$$

$$\Delta_{2p} = 2E_B(Z, N) - E_B(Z - 2, N) - E_B(Z + 2, N). \quad (1.6)$$

These types of derived metrics have become powerful tools for investigating shell structures, as demonstrated for example in [8]. In figure 1.1, the neutron empirical shell gap, computed using AME2020 mass values [7], is shown across the entire nuclear chart. A marked increase in the empirical shell gap is observed at magic numbers of neutrons. Through such mass filters, mass measurements continue to be relevant for probing nuclear structure and the nature of the nuclear force.

1.2 β -decay Radioactivity

Less than 10% of observed nuclear isotopes are stable. The vast majority of the over 3000 isotopes that have been experimentally observed [9] undergo some form of radioactive decay, which can in part be understood through nuclear binding energy.

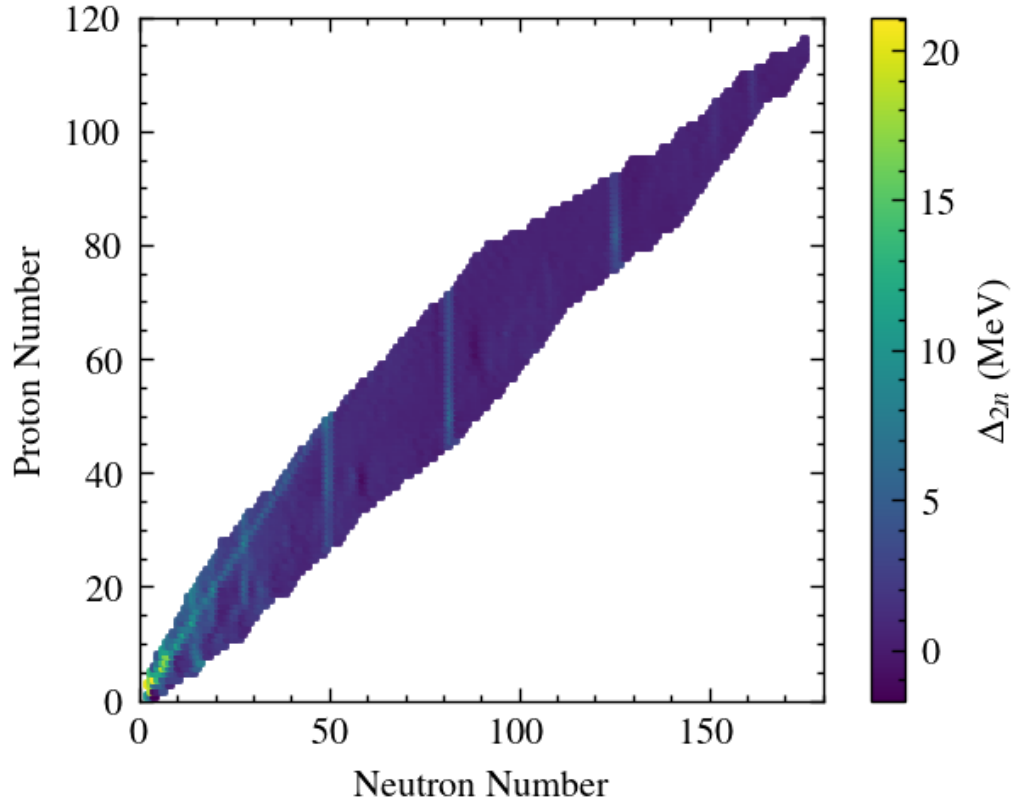


Figure 1.1: Neutron empirical shell gap calculated from the mass data in [7]. The greater binding at magic number of neutrons and along $Z = N$ is apparent as an increase in the shell gap.

The binding energy per nucleon reduces away from the valley of stability on the nuclear chart. At the most extreme ranges of the nuclear chart are the proton and neutron driplines, where the difference in binding energy is so great that a nucleus can directly emit a nucleon. Closer to stability, nuclear decay proceeds through α -decay, β -decay, or fission into much smaller nuclides. Of these decays, β -decay is special in that it can only occur through the weak force. According to the standard model, this process is mediated by the weak force through the exchange of charged $W^\pm$ boson [1]. β decay can be broken into 3 distinct forms of decay, where an electron is emitted (β^-), a positron is emitted (β^+), or where an atomic electron is captured by the nucleus (electron capture or EC). The energy available for this decay is distributed among the particles in the final state (the daughter nucleus, β particle, and neutrino). This available energy is known as the Q value and can be determined directly through the masses of the parent (M_P) and daughter (M_D) nuclides as:

$$Q_{\beta^-} = (M_P - M_D)c^2 \quad (1.7)$$

$$Q_{\beta^+} = (M_P - M_D - 2m_e)c^2 \quad (1.8)$$

$$Q_{\text{EC}} = (M_P - M_D - B_e)c^2, \quad (1.9)$$

where c is the speed of light, m_e is the mass of an electron and B_e is the atomic binding energy of the captured electron and we have neglected the mass of the neutrino².

In general, the daughter nucleus resulting from a β -decay may not be bound with respect to nucleon decay. This can occur whenever the Q -value of the decay is larger than its nucleon separation energy [12]. When a delayed emission occurs it is usually because the daughter was produced in an excited state. The daughter nucleus can

²At the time of writing the upper mass limit for a neutrino is $m_\nu < 0.12 \text{ eV}/c^2$ from cosmological considerations [10] and $m_\nu < 0.8 \text{ eV}/c^2$ from direct measurement experiments [11].

then decay through a series of prompt gamma ray emissions, or by releasing a nucleon. The latter of these is known as β -delayed neutron or proton emission, depending on the released nucleon. While the probability of β -delayed neutron emission is of interest for applications such as r-process nucleosynthesis modelling [13], as of 2020 only about 300 β -delayed neutron emitters have had their β -delayed neutron emission probabilities measured, out of about 650 identified nuclei with sufficient decay Q values [12]. There exists many theoretical approaches to estimate neutron emission probabilities, although it is questionable whether some are well suited to neutron-rich nuclei [14]. This is made more difficult by the lack of experimental measurements on neutron-rich nuclei to validate the models [14]. In chapter 5, the first experimental determination of β -delayed one-neutron and two-neutron emission probabilities for the very neutron-rich isotope ^{54}K is presented.

1.3 Nuclear Potential

There are fundamentally two ways of modelling the nuclear force for the shell model. One is nucleon interaction models, which take into account aspects of the nuclear force between nucleons. The other is mean-field models, which attempt to describe the nucleus in terms of bulk and surface properties. Both types of potentials are typically fitted to data in order to reproduce the observed nuclear properties across a large region of the nuclear chart. However, in the case of nucleon interaction models it is possible to fit the interaction strengths using simple well known systems such as ^3He and ^4He . Because the interactions are grounded in the symmetries of QCD, these potentials are universal to all nuclei. Since these interactions have the closest theoretical ties to QCD, and the fitting required to describe a large number of nuclei is minimal, these are known as *ab initio* theories.

One mean field potential of particular note is the Woods-Saxon potential. This was the earliest model of the nucleus that could capture much of the relevant physics of nuclear structure, like correct major magic numbers.

1.3.1 Woods-Saxon Potential

Phenomenologically, it was understood that to describe the nucleus, the nucleons must exist in some mean field potential that they create [1, 4, 5]. It was also quickly realized that the magic numbers observed in nuclei would naturally result from a potential somewhere between that of a harmonic oscillator and a square well [1]. This can be understood as the interactions of nucleons occurring largely on short distances, such that nucleons can be thought to only interact with their near neighbours. The potential of such an interaction is of the form $V(r) = -V_0(1 + e^{(r-R)/\alpha})^{-1}$ where $R = R_0 A^{\frac{1}{3}}$, A being the atomic mass and R_0 being a constant of about 1.2 fm. R can be interpreted as the size of the nucleus, while α would be the nuclear skin thickness and V_0 the depth of the nuclear potential (typically in the range of 50 MV)[15, 1].

Additionally, protons experience a repulsive Coulomb force due to their charge. This is modelled as a finite sphere of charge. When adding a centrifugal potential based on the nucleons' orbital angular momentum characterized by the quantum number ℓ , the nuclear potential can be written as:

$$\begin{cases} V(r) = \frac{-V_0}{1+e^{(r-R)/\alpha}} + \frac{kq}{2R}(3 - \frac{r^2}{R^2}) + \ell(1 + \ell)/2mr^2 & \text{if } r < R \\ V(r) = \frac{-V_0}{1+e^{(r-R)/\alpha}} + kq/r + \ell(1 + \ell)/2mr^2 & \text{if } r > R \end{cases} \quad (1.10)$$

where m is the mass of the nucleon and k is the Coulomb constant. This is known as the Woods-Saxon potential.

The result of calculating the energy levels in the Schrödinger equation using the

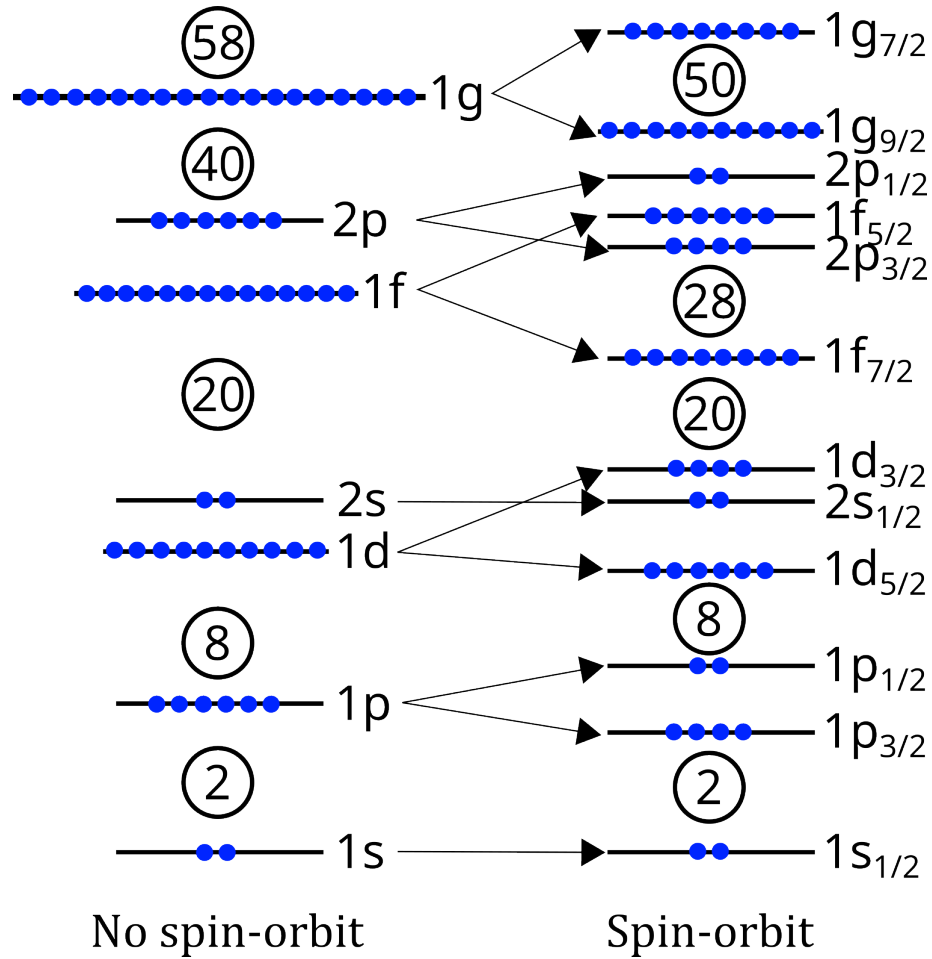


Figure 1.2: Shell model levels in a simple Woods-Saxon potential without and with a spin-orbit coupling term. The canonical magic number shell gaps are reproduced when the spin-orbit term is added. Figure made based on the calculations presented in [1].

potential of eqn. 1.10 is shown on the left in figure 1.2. The calculation fails to reproduce the observed magic numbers above nucleon number 20. Inspired by atomic physics, Mayer and Jensen [4, 5] included a spin orbit potential in their calculations. The spin orbit potential takes the form $V_{\text{so}}(r)\boldsymbol{\ell} \cdot \boldsymbol{s}$, giving rise to a new quantum number $\boldsymbol{j} = \boldsymbol{l} + \boldsymbol{s}$ known as the total angular momentum. As is seen in figure 1.2 the inclusion of this term results in level splitting and the reproduction of the canonical magic numbers.

The Woods-Saxon potential was pivotal to the early understanding of nuclear structure [3]. Many early mean field potentials were based on the potential, and it was influential in understanding nuclear magic numbers [1].

In recent years, Woods-Saxon type potentials have remained relevant as they have been extended to non-spherical nuclei [16]. This has allowed for more accurate modelling of deformed nuclei. Additionally, theory and computational power has progressed such that mass or gamma spectroscopy measurements of nuclear isomeric states can be used to determine the degree of deformation that an isotope exhibits [17].

1.3.2 Other Mean Field Potentials

In addition to the widely used Woods-Saxon potential, several other mean field approaches have been employed to describe the nuclear potential. These approaches offer alternative ways to model the average interaction between nucleons in the nucleus and provide valuable insights into nuclear structure and properties.

An early mean field potential, which had some success was the Nilsson model [18]. This model consisted of harmonic potentials of varying width in each cartesian direction. The model provided a theoretical basis for understanding the rotational

energy bands of deformed nuclei.

A more modern method for obtaining these mean field potentials is the Hartree--Fock-Bogoliubov (HFB) (or self-consistent field method) [19, 20, 21]. The HFB method allows one to calculate an effective potential on a single nucleon from the interactions of other nucleons [22]. This allows the Schrödinger equation to be solved iteratively on a single nucleon [23, 22].

1.3.3 *Ab Initio* Models

Much of the recent thrust in developing nuclear shell models has focused on bringing the description of nuclear potentials closer to their QCD foundation [24, 25]. Early *ab initio* models described the nucleon interactions using a large number of fitted phenomenological parameters [24]. As knowledge of the nuclear force developed, it became clear that the nucleus could only be properly described with a mix of two- and three-body interaction terms [26], or an effective equivalent in terms of a mean field and residual two nucleon forces [24].

Ab initio potentials can be broadly classified by the approach they use in determining the many body Hamiltonian, namely effective interactions or microscopic interactions [24]. In the case of effective interactions, the nuclear force is described in terms of a mean nucleon interaction [24, 27]. This allows the calculation of nucleon-nucleon interaction to be feasibly included as a perturbation to a mean field. However, it is known that the nucleon-nucleon interaction must be repulsive at very short range as nucleons are composed of fermionic quarks which must obey the Pauli exclusion principle. To account for this, effective interaction potentials include a short-range repulsive (scattering like) potential [24, 25]. As was pioneered with the Skyrme potential [27], this repulsive interaction is often modelled as a contact force of the

form:

$$V_1\delta(\mathbf{r}_1 - \mathbf{r}_2)$$

where V_1 is a fitted parameter and δ is the delta function. It has been noted that nuclear structure calculations are not particularly sensitive to the exact form of this repulsive interaction, allowing for significant freedom in choosing a form that allows for tractable computations [24].

In microscopic *ab initio* models, the nucleon-nucleon forces are calculated from the formalism of meson exchange or QCD [25]. Until about 2003, calculations grounded in QCD were hampered by insufficient computing power to describe the strong force at the relevant length and energy scales [28]. Therefore, microscopic *ab initio* models used interactions grounded in pion and other meson exchange to calculate the effective Hamiltonian. These models use small, well known nuclear systems such as the deuteron (^2H), ^3He , and ^4He to fit two-, three- and four-body interaction potentials [25]. These physically grounded but fitted potentials are then applied to larger systems. Still, it continues to be a challenge to apply such results to systems larger than $Z + N = A > 12$ within a no-core shell model framework [29].

1.4 Types of Shell Models: Many-body methods

1.4.1 Independent Particle Model

All shell models rest on the fermionic nature of nucleons. Due to this nature, nucleons must obey the Pauli exclusion principle. In addition, the strong force acts on all nucleons and tends to dominate the Coulomb repulsion of protons on the distance scale of a nuclei. It is therefore reasonable in a first approximation to expect protons and neutrons to each exhibit the same shell structure. This is seen clearly in the

independent particle model.

Nuclear shell models exist on a spectrum of complexity. The simplest form of shell model, as described by Mayer [4], supposes that all nucleons but one are paired. The remaining nucleon then determines the major nuclear properties of the nucleus. This is known as the extreme independent particle model [1], because it neglects any interactions between nucleons. In this way, the model ignores any coupling of nuclear levels. The model is successful at describing many of the major properties of odd mass number (A) nuclei near stability. The model is less valid away from shell closures where substantial mixing of states is expected [30].

Calculations done in this formalism begin with a mean field potential. The Schrödinger equation

$$-\frac{\hbar}{2m}\nabla^2\Psi(\mathbf{r}) + V(\mathbf{r})\Psi(\mathbf{r}) = E\Psi(\mathbf{r}) \quad (1.11)$$

then gives pure states with no coupling between nucleons in different shells. Shown in figure 1.2 is the expected shell configuration in a Woods-Saxon type potential. Large gaps in energy are marked with maximum number of protons or neutron below the gap. It can be seen that the inclusion of a spin-orbit coupling breaks the degeneracy of the levels with angular momentum greater than 0 [1]. The inclusion of this term is a defining feature in the shell model and the understanding of nuclear magic numbers [1, 3].

Other shell models can be broadly classified by how large the space of interacting valence states is in the calculations.

1.4.2 Inert-Core Models

Inert core models represent a class of nuclear shell models that treat a specific core as inert while focusing on the valence nucleons outside the core. These models assume that the core remains unchanged, providing a fixed potential within which the valence nucleons move [31]. By considering the valence nucleons as the active particles, inert core models simplify the complexity of the nuclear many-body problem, allowing for a more tractable description of nuclear structure and properties.

The concept of inert core models can be traced back to the early development of the nuclear shell model. In the original shell model formulation, the valence nucleons were assumed to move in an average potential generated by the core. This approach decoupled the valence nucleons from the core, simplifying the calculations and facilitating the analysis of nuclear states [31].

Inert core models have been particularly useful in describing the behaviour of single-particle states in nuclei. By assuming a fixed core potential, these models allow for the calculation of single-particle energies and wave functions, which are essential for understanding nuclear excitation spectra, electromagnetic transitions, and nuclear reactions. Inert core models also provide insights into magic numbers, shell closures, and the arrangement of nucleons in different energy levels [29].

While inert core models have proven valuable in capturing certain aspects of nuclear structure, they do have limitations. The assumption of an inert core neglects the dynamic interplay between the core and valence nucleons. In reality, the motion of the valence nucleons can influence the structure and properties of the core, and vice versa. Therefore, while inert core models provide a useful approximation, they are not fully self-consistent descriptions of nuclear structure [25].

Making an appropriate choice of inert core for modelling a region of the nuclear

chart is very important. A choice must be made to take a sufficiently large valence space (and thus small inert core) to capture the relevant coupling of states. However, the computational demands grow as the size of the basis increases. This is because the size of the matrices needed to represent the basis on which the Hamiltonian acts scales combinatorially with both the single particle states and the number of participating nucleons [31]. It is very common to choose an inert core to be a doubly magic nucleus, as these are strongly bound with robust shell structure [32, 29]. The assumption that the nucleons from the core play little role in the structure of the valence nucleons is thus very likely to be true [29].

Inert core models often use a middle-ground approach to the nuclear potential. In these cases the inert core is calculated using a mean field potential. Coupling of states is neglected for the states from the core. The states above this up to some maximum valence space size is calculated using some sort of nucleon-nucleon interaction potential. This can come from effective field theories, or from more microscopic *ab initio* methods [24].

1.4.3 No-core Models

While traditional shell models assume the presence of an inert core, recent advancements in computational methods have led to the development of no-core shell models [25, 33]. The no-core model is a powerful theoretical framework that provides a first-principles approach to nuclear structure calculations, without assuming the existence of a core.

In the no-core model, the nuclear wave function is expanded in a basis of harmonic oscillator (HO) states, allowing for the explicit treatment of nucleons as active particles [25]. This expansion includes both the valence nucleons and the unoccupied single

particle states, forming a complete basis for an effective Hamiltonian [29, 24, 25]. The no-core model approach considers all possible configurations of valence nucleons within the chosen model space, providing a complete description of nuclear structure.

Diagonalizing the energy basis to find the energy eigenvalues and eigenvectors is used to find the energy of the ground and excited states [30]. Exact diagonalization is very computationally demanding for the same reason as in the inert core model. This is amplified by the larger number of nucleons being considered, and therefore approximate diagonalization schemes are used [29, 25, 30]. Convergence of the results of this process is often studied by systematically increasing the model space size considered [30, 25].

No-core shell model calculations can be carried out using various many-body techniques, such as the coupled cluster method, configuration interaction method, or the Monte Carlo shell model [25]. Additionally a large number of different nuclear potentials can be used. This has allowed no-core shell model calculations to be extended to medium mass nuclei in recent years [26, 34]. These methods allow for accurate calculations of binding energies, excitation spectra, electromagnetic transition rates, and other nuclear observables.

One of the advantages of the no-core shell model is its ability to describe nuclear properties for both bound and unbound states. The model can treat resonant states and scattering processes by treating clusters of nucleons largely independently, providing a comprehensive description of nuclear structure and reactions [35].

The no-core model has been successful in describing the properties of light and medium-mass nuclei. No-core calculations have provided insights into nuclear properties, such as the evolution of shell structure and the emergence of new phenomena in nuclei far from stability. However, the no-core model also has its limitations. As the model space expands, the dimensionality of the problem increases rapidly,

resulting in computational challenges for larger nuclei. Additionally, the no-core calculations require reliable nucleon-nucleon interactions as input, obtained from effective interactions derived from nucleon-nucleon potentials or from microscopic meson exchange models.

Despite these challenges, the no-core shell model has proven to be a valuable tool for nuclear structure calculations with its ability to provide a first-principles description of nuclear systems, without relying on a specific core allows for a comprehensive understanding of nuclear phenomena across the nuclear chart.

1.5 Excited States and Isomers

The shell model qualitatively predicts the many excited states seen in nuclei. Analogously to the atomic shell model, nucleons can be excited into higher states. The nucleus can then decay from this state through direct photon emission or through emission of a massive particle like in beta decay. The energy and spin of these states can be predicted via the shell model. In some cases, the excited nuclei may not have any nuclear states close in spin to decay to. This can result in very long-lived states known as isomers [36].

Short-lived energy levels of nuclei can often be probed through beta and gamma decay spectroscopy. However, long-lived isomers often exist for much longer than the ground state of the nucleus. This makes detection of these states through decay spectroscopy difficult. In these instances, high precision mass spectrometry is an excellent tool for identifying and studying these isomeric states [37, 38]. A nucleus in an isomeric state is heavier than the ground state by an amount equal to the excitation energy of the isomer. By precisely measuring the masses of nuclei using mass spectrometry, the mass difference between an isomeric state and the corresponding

ground state can be determined. By combining these mass measurements with the predictions of the shell model, valuable insights into the structure and properties of isomeric states can be obtained. The shell model provides a theoretical framework for understanding the arrangement of nucleons in different energy levels and the associated spin and excitation energies [36, 37].

The identification and characterization of isomeric states contribute to our understanding of nuclear structure, as they represent specific configurations of nucleons within the nucleus that exhibit unique properties and behaviours. Studies of isomers can shed light on phenomena such as shape coexistence, shape isomerism, and collective excitations.

1.6 Magic Numbers

The concept of magic numbers has played a pivotal role in strengthening the evidence for the shell model and our understanding of nuclear structure. Magic numbers are specific values of proton or neutron numbers for which atomic nuclei exhibit enhanced stability and unique properties. These numbers correspond to the filling of complete shells within the nuclear potential, resulting in a particularly stable configuration of nucleons.

The discovery of magic numbers arose from observations of certain nuclei that showed increased binding energies, larger separation energies, and higher abundance compared to neighbouring nuclides. The original magic numbers identified were 2, 8, 20, 28, 50, 82, and 126 for both protons and neutrons [1, 3]. These numbers correspond to the completion of nuclear shells, analogous to the completion of electron shells in noble gases in atomic physics. The magic numbers represent a significant departure from the trend of gradual variation in nuclear properties as a function of

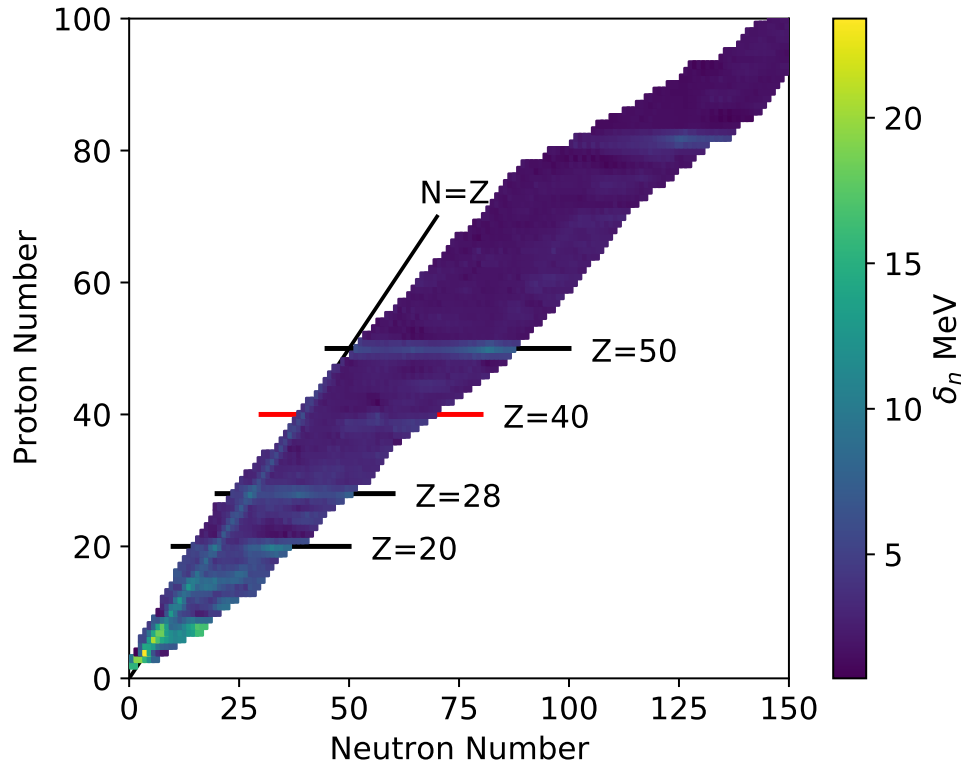


Figure 1.3: Proton empirical shell gap calculated from the mass data in [7]. Several canonical shell closures are marked as well as the $N = Z$ line. A moderate shell closure around $Z = 40$ is visible in the neutron-rich region.

nucleon numbers, providing strong evidence for the existence of nuclear shells [3].

For nuclei with magic numbers of protons or neutrons, several key properties of atomic nuclei change, demonstrating the influence of shell structure on nuclear behaviour. One of the most notable effects is the significant increase in nuclear binding energy. Nuclei with magic numbers of protons or neutrons exhibit higher binding energies per nucleon compared to neighbouring nuclei, indicating increased nuclear stability. This enhanced stability arises from the filling of complete shells,

which results in a stronger nuclear force acting on nucleons within the shell [1].

Magic numbers are the direct result of pronounced nuclear shell closures. A shell closure indicates the completion of a major shell, with a significant energy gap between occupied and unoccupied states. Nuclei with closed shells tend to be more stable, and their properties differ markedly from those with incomplete shells. For instance, closed-shell nuclei often exhibit spherical symmetry, while nuclei with fewer or excess nucleons tend to deform and exhibit different shapes [1].

The presence of magic numbers also influences the nuclear excitation spectra. Nuclei with magic numbers display distinctive energy levels and excitation patterns, resulting from the specific arrangements of nucleons in the shells. These excitation spectra can be probed experimentally through gamma spectroscopy, providing valuable information about nuclear structure and the energies of excited states [36].

1.6.1 Changing Magic Numbers

The concept of universal magic numbers is broken when examining nuclei that deviate from stability. As one moves away from stable isotopes, the magic numbers and shell closures tend to evolve, leading to the emergence of new magic numbers or the breakdown of previously established ones. This phenomenon highlights the intricate interplay between nuclear structure and the properties of exotic nuclei.

One way in which this manifests is new shell closures in regions of the nuclear chart known as islands of inversion and sub-shell closures. One such particularly strong closure is a $Z = 40$ [40]. In even-even nuclei this is apparent in gamma spectroscopy through the first excitation energy of the nucleus [41] as seen in figure 1.4. As a shell closure is approached the energy needed to excite a nucleon to a higher state increases.

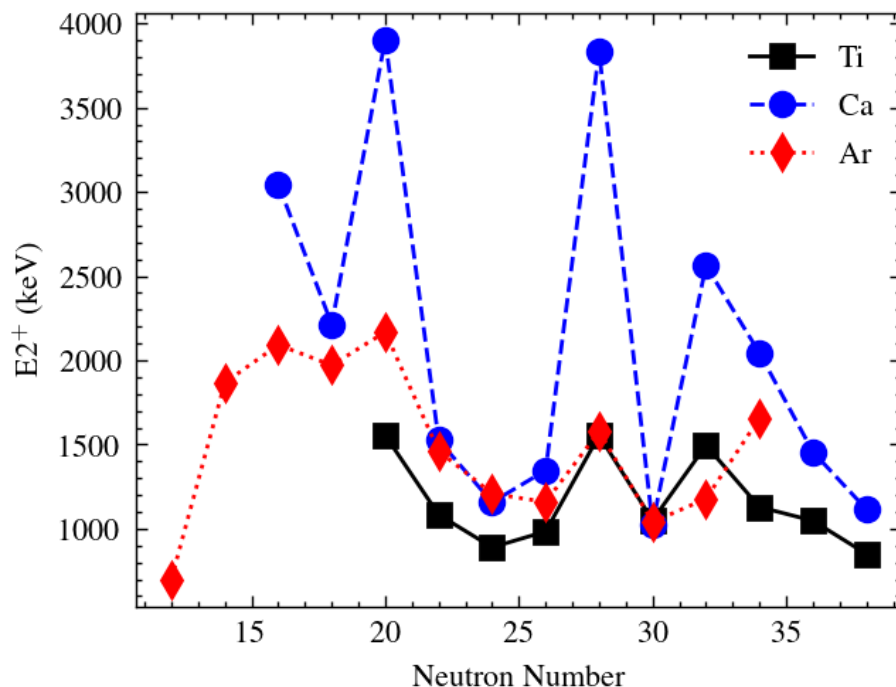


Figure 1.4: First 2^+ excitation energies in even-even nuclei around $Z = 20$. Clear signatures of magic numbers 20 and 28 are apparent. Both the Ca and Ar isotopic chains show evidence of an $N = 34$ shell closure, while this is absent in the Ti chain. The figure was generated using data from the NUDAT online database [39].

To avoid being limited to the even-even nuclei, one must accurately measure many nuclear states with gamma spectroscopy and compare to advanced shell model calculations, or measure the empirical shell gap through mass spectroscopy [8, 34]. This is seen for the same $Z = 40$ island of inversion in figure 1.3. In practice, high precision mass measurement is often the most practical technique to study these shell closures. Shown in figure 1.5 are experimental measurements of the neutron empirical gap around $Z = 20$ and $N = 32$. These represent a challenge for nuclear theory as they have a relatively large number of nucleons and include open shell and odd-odd nuclei [26, 34]. As a consequence, there is substantial coupling of states.

The calculations shown here are some of the largest mass nuclei to be calculated with *ab initio* no core shell model techniques [26].

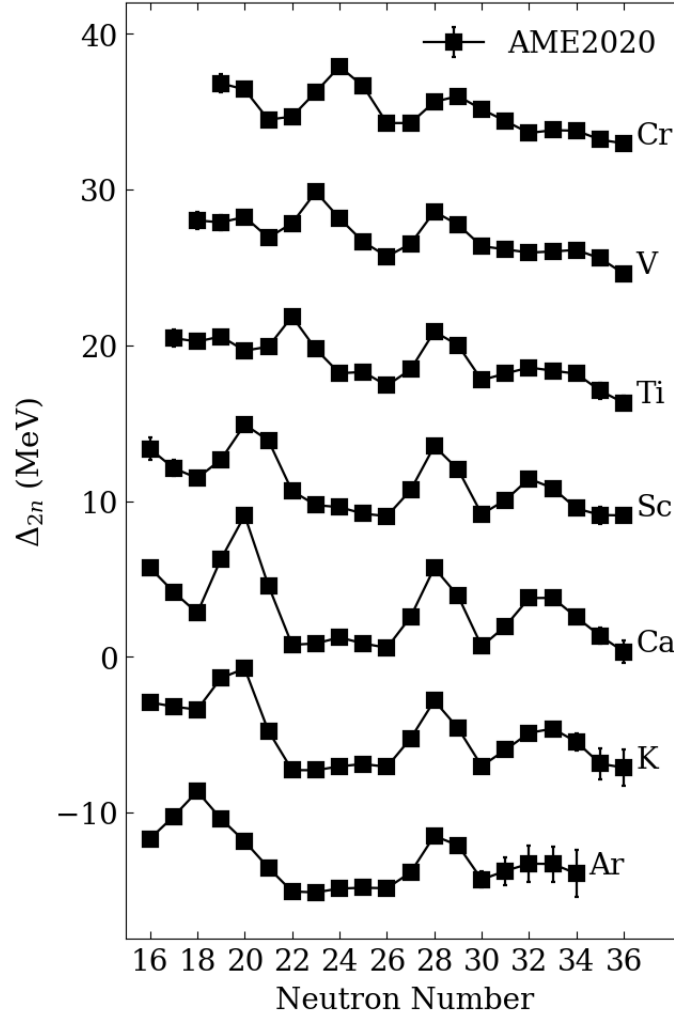


Figure 1.5: Experimental neutron empirical shell gaps near $Z = 20$ calculated from AME2020 masses [7]. It was shown that 3 body nucleon interactions can reproduce the observed shell closure at $N = 32$. Isotopic chains are offset by $(Z - 20) \times 8$ MeV for readability.

CHAPTER 2

Ion Trapping Experiments at TRIUMF's ISAC Facility

The experiments described in this this thesis were performed at the Isotope Separation and ACceleration (ISAC) facility at TRIUMF. ISAC is a rare isotope beam (RIB) facility that uses a high intensity ~ 500 MeV proton beam impinged on a range of target materials to produce radionuclides. Proton induced fission and spallation reactions in the thick targets produce a wide range of nuclides inside the target. The ISAC targets are kept at high temperature, allowing the produced nuclides to quickly effuse out of the target, where they can then be ionized and magnetically mass separated in a process called isotope separation on-line (ISOL). RIB beam produced at ISAC can be delivered to the wide range of experimental setups in the ISAC facility or be post-accelerated for higher energy experiments in the neighbouring ISAC II experimental hall. A brief description of the beam production and delivery to the TITAN experiment is given in this chapter. An overview of the

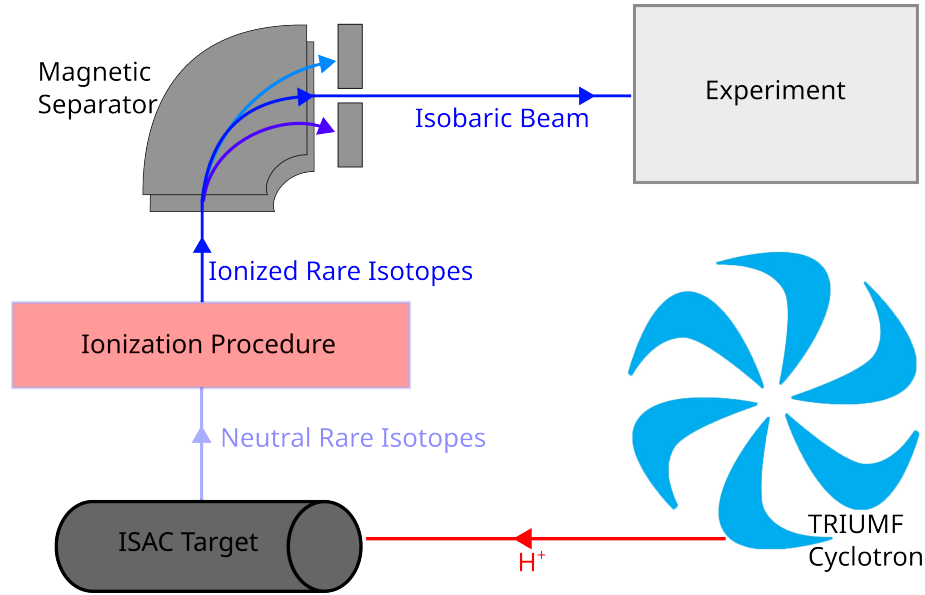


Figure 2.1: An overview of the rare isotope beam production process at ISAC.

TITAN experiment is also given, before a detailed description of the MR-TOF-MS ion trap is presented in chapter 3.

2.1 Rare Isotope Production at the ISAC Facility

Initial RIB production can conceptually be broken in to three distinct steps which all take place within the ISAC target and ion source assembly. Each element of the assembly effects the types of nuclides that can be produced for the experimenters. The range of nuclides that are generated within the target material is greatly affected by the nuclear makeup of the target material. The target material also reacts with the produced particles, determining which elements can release from the target material. The choice of ionization technique determines which of the released nuclides will be transported further downstream of the target. The experimenters choose a single

isobaric mass unit of the ionized RIB to deliver to the experiment with the use of a magnetic separator stage.

2.1.1 Production in Target

Radioisotopes are produced at ISAC within thick targets. ISAC can take the nominal 480 MeV proton beam produced in the TRIUMF cyclotron [42] onto the thick targets with primary beam current up to 100 μA . This intense primary proton beam breaks apart the nuclei of the target through fragmentation, spallation and proton-induced fission reactions forming lighter products [43]. At ISAC, a range of target materials have been used to optimize the in-target production and target release properties for the isotopes of interest to experimenters.

The experiments described in this thesis were all carried out with standard uranium-carbide (UC_x) targets with the exception of the beam composition measurements for a new type of ISAC target presented in chapter 6. In these experiments the RIB was produced by first impinging the proton beam onto a tungsten converter target. The converter target produced neutrons which then caused neutron induced fission reactions in an annular uranium target. The aim of this new target type is to produce RIB of similar intensity around the fission peaks of uranium, while reducing neutron deficient isobaric contamination.

2.1.2 Effusion from Target

The nuclei created in by the reactions in the target will generally be stopped within a very short distance, and remain within the target. In order to liberate the produced nuclides from the target material, the target is maintained at temperatures as high as 2300°C [44]. This high heat allows the particles to effuse from the target. The

efficiency of this step, along with the following ionization step, is element specific. The volatility of the element, and its chemical affinity for binding to the target material effect the speed at which the isotopes can escape. Isotopes may even form molecules before being released from the target, or even never be released within the lifetime of the target. While some elements may escape from certain target materials, some of the least volatile elements cannot be freed from any target. Isotopes of these elements are then not a good candidate for study at an ISOL facility.

2.1.3 Ionization

Once isotopes are freed from the bulk material of the target, they must be ionized in order to be manipulated into a beam through electromagnetic optics. Ionization is done by surface ionization in a hot cavity, resonant or non-resonant laser ionization or through a microwave heated plasma [45]. The target and ion source assembly is typically used for multiple experiments before being removed for refurbishment, and so the ion source can be made to produce both surface ionized and laser ionized elements in what is called a TRIUMF Resonant Laser Ionization Source (TRILIS). Alternatively, the ion source can be made as an Ion-Guide Laser Ionization Source (IGLIS) allowing laser ionization while suppressing surface ionized beam. In both cases the laser scheme can be changed to allow different elements to be produced without changing the target and ion source.

The measurements presented in the thesis were all performed with the use of TRILIS. The potassium (K) mass and half-life measurements presented in chapter 3 and 5 respectively, were done with hot cavity surface ionization only. The low ionization potential of potassium makes this an efficient ionization technique. The ytterbium (Yb) half-life measurements in chapter 5 were performed with the aid of

laser ionization.

2.1.4 Beam Delivery and Magnetic Separation

Ionized beam is accelerated and focused through electrostatic elements as it leaves the target ion source. At this point the beam is almost completely undifferentiated. The RIB is then cut to a single isobar through the use of two magnetic separator stages. A magnetic separator is the ion optical equivalent of a prism. The particles in the produced beam all have approximately the same kinetic energy (~ 20 keV), but different velocities based on the mass of the particles. The force a charged particle experiences travelling in a magnetic field is

$$\vec{F} = q\vec{v} \times \vec{B}, \quad (2.1)$$

where q is the charge, B is the magnetic field strength and m is the particle mass. A dipole magnetic separator is designed such that the magnetic field is perpendicular to the motion of the ions. Therefore we can use that the speed of the ions is given by the kinetic energy K as

$$|\vec{v}| = \sqrt{2K/m}, \quad (2.2)$$

to find that the radius of curvature r is then determined by the magnetic field, mass and kinetic energy as

$$r = \frac{\sqrt{2mK}}{qB}. \quad (2.3)$$

With this principle, it is possible to mass select the ion beam by precisely controlling the magnetic field. Adjustable aperture slits are used to control how far off axis the beam must be to be cut, allowing the mass selectivity to be tuned.

The pre-separator magnet at ISAC provides initial mass separation with mass

resolving power $m/\Delta m$ of ~ 300 , while the high resolution mass separator achieves resolving power between 2000 – 10000 [46]. This is sufficient to reduce the beam to a single isobaric mass unit, and often enough to reduce isobaric contamination in the beam. From the mass separator magnets, the RIB is delivered to the low energy beam experiments in ISAC through electrostatic benders and quadrupole lenses.

2.2 TRIUMF's Ion Trap for Atomic and Nuclear Science

TRIUMF's Ion Trap for Atomic and Nuclear science consists of four ion traps linked together to allow ions to be prepared for subsequent traps. All ion beam delivered to TITAN is first cooled and bunched in a gas-filled radio frequency quadrupole (RFQ) trap. The beam bunches can then be sent directly to any of the three science traps. The science traps consist of two mass spectrometers and a charge breeding Electron Beam Ion Trap (EBIT). The Measurement Penning Trap (MPET) mass spectrometer trap is used for the highest precision measurements, while the Multiple-Reflection Time-of-Flight Mass Spectrometer (MR-TOF-MS) is used for fast high precision measurements, high background measurements, and removal of isobaric contamination for the other science traps. The EBIT trap is used for decay spectroscopy measurements, or preparing the ions in a higher charge state to further improve the precision of measurements made in MPET. Figure 2.2 provides an overview of the trapping facility and possible ion beam paths from the RFQ cooler-buncher to the science traps. The following gives a short description of the operating principles of the traps used in this thesis (TITAN RFQ cooler-buncher and MR-TOF-MS). An in-depth discussion of the operation of the MR-TOF-MS for

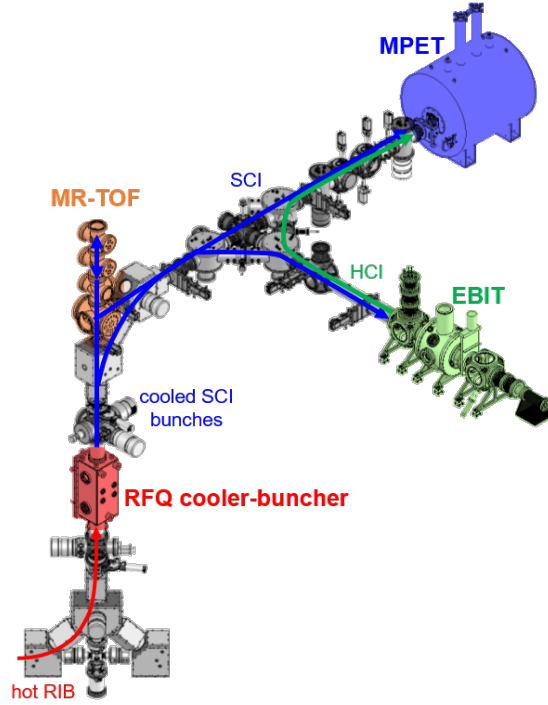


Figure 2.2: An overview of the TITAN experiment. Continuous radioactive ion beam is delivered from ISAC to the RFQ Cooler-Buncher [47, 48]. Cold ion bunches can then be sent from the RFQ to the various other traps at TITAN. For this proposal, the ion bunches will be delivered to the MR-ToF-MS where yield and mass measurements will be performed.

precision mass measurement is reserved for chapter 3, where mass measurements of $^{51-54}\text{K}$ performed with the trap are also presented.

2.2.1 TITAN RFQ Cooler-Buncher

All experiments at TITAN begin with the TITAN RFQ cooler-buncher trap. The RFQ is a gas-filled linear Paul trap which cools and bunches continuous beam delivered to TITAN from the Isotope Separator and Accelerator (ISAC) facility. An RFQ traps ions by alternating electric fields in such a way as to provide a net confining

potential to the ions in the radial direction [49]. The axial confining potential is done statically, either with the use of end-cap electrodes or, as in the case of the linear segmented design of TITAN, by keeping a region of radial oscillating electrodes at a lower potential than the neighbouring radial electrodes[50].

The entirety of the TITAN RFQ is floated at a potential of ~ 20 keV, matching the beam energy of the ions produced at the ISAC target. This careful matching keeps the ions from simply flying through the RFQ. The ions will then interact with the high purity helium gas in the TITAN RFQ. The gas flow is set to provide a pressure of approximately 1×10^{-2} Torr in the RFQ. Differential pumping maintains the ultra high vacuum (UHV) conditions outside of the RFQ ($\sim 1 \times 10^{-7}$ Torr). The ions received from ISAC interact with the gas, thermalizing from their initial energy spread on the order of tens of eV to sub eV thermal energies. The slow cooling resulting in ion accumulation at the end of the RFQ is represented pictorially in figure 2.3c. Helium is used as the buffering gas because its light weight improves the thermalization efficiency and its high ionization potential, preventing charge exchange reactions from neutralizing the ion beam.

After the ion bunch is cooled, it is ejected towards any of the three science traps at TITAN. This is done by simultaneously lowering the last electrode potential while raising the third to last electrode to form a push-pull potential. Ejecting ions in this way ensure all ions exit quickly, forming a small spread in the arrival times of ions in the pulsed drift tube after the RFQ. The pulsed drift tube after the RFQ allows the energy of ions to be matched to the trapping energy of the subsequent trap. In the case of TITAN's MR-TOF-MS the ion energy is reduced down from 20 keV to ~ 1.3 keV. TITAN's RFQ can also be set to capture ions on the input end, allowing the beam to be reverse extracted to provide bunched beam for collinear laser spectroscopy in ISAC.

2.2. TRIUMF's Ion Trap for Atomic and Nuclear Science

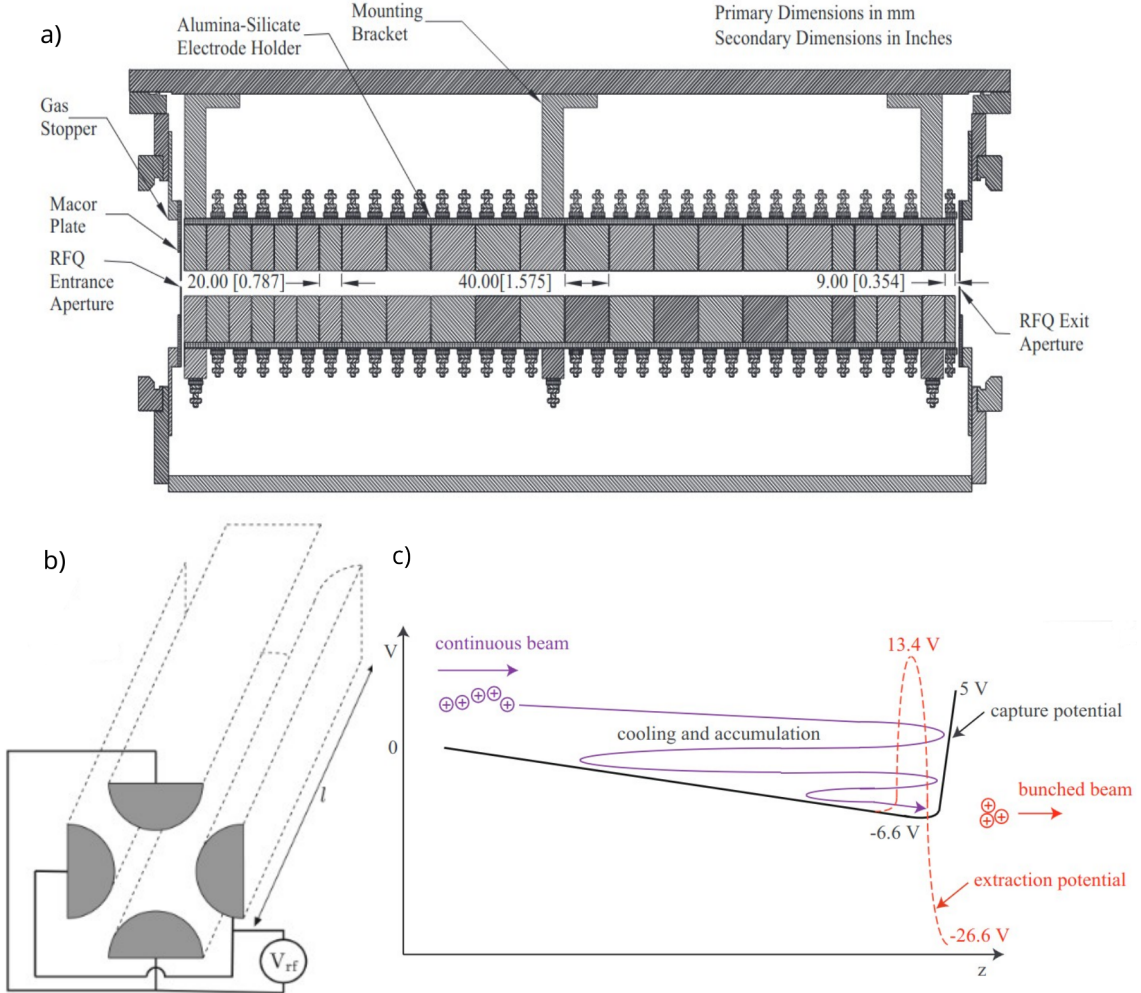


Figure 2.3: a) A schematic drawing of the TITAN RFQ cooler-buncher. b) A general drawing of the geometry of a linear RFQ trap, showing the RF voltage phases applied to the electrodes. c) A schematic of the axial dc potential applied to the TITAN RFQ during accumulation and ejection of the ions, in its normal forward ejection mode. Figure adapted from [50].

2.2.2 MR-TOF-MS

The TITAN MR-TOF-MS receives the bunched ion beam from the RFQ and re-cools the beam in its own RFQ paul trap. The MR-TOF-MS transfers the ions to just below the high vacuum analyzer section. It then lowers the potential of one mirror to inject the ions at 1.3 keV energy. The MR-TOF-MS works on the same principle as a time-of-flight mass spectrometer (TOF-MS), where ions of greater mass take longer to travel to the detector. The electrostatic mirrors of the MR-TOF-MS provides much longer flight paths for ions in a compact system by utilizing electrostatic mirrors to reflect the ions [48]. Additionally, the MR-TOF-MS benefits from the energy correcting behaviour of the electrostatic mirrors. In the same manner as a reflectron type TOF-MS, the MR-TOF-MS allows ions with greater energy to penetrate further into the reflecting field and therefore have their greater speed compensated with longer path length. This phenomenon which ensures time of flight is largely independent of energy is known as isochronicity.

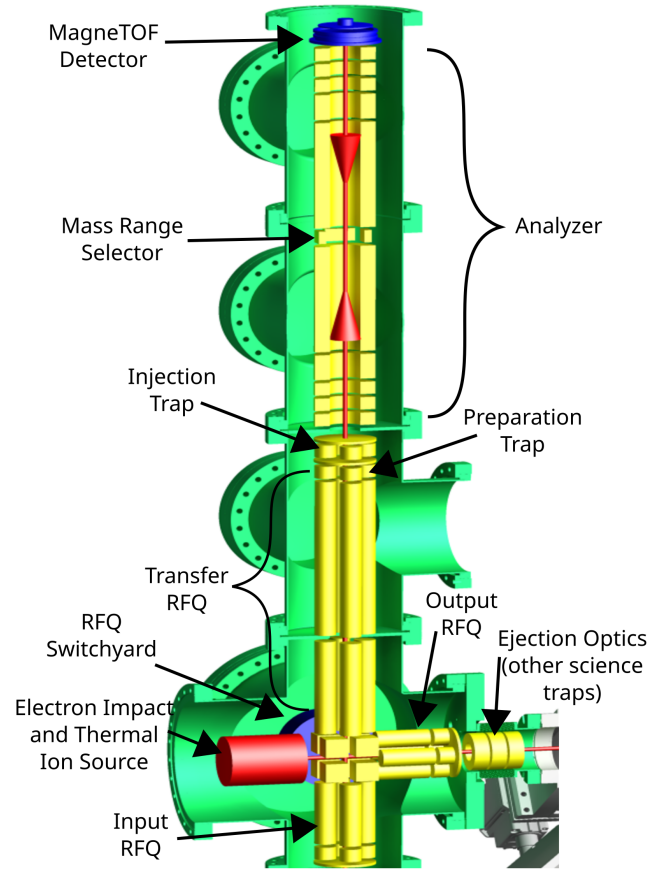


Figure 2.4: A CAD model of the TITAN MR-TOF-MS. Beam from the TITAN RFQ enters at the input RFQ at the bottom. Ions travel to the preparation trap where they are bunched and cooled before being transferred into the injection trap and subsequently injected into the electrostatic analyzer section. Figure adapted from [48].

Just High-Precision Mass Measurements with the TITAN MR-TOF-MS

3.1 Principles of Time-of-Flight Mass Spectrometry

A time-of-flight mass spectrometer (TOF-MS) uses the behaviour of a charged particle in a known electrostatic potential to determine that particle's mass-to-charge ratio. A particle with mass-to-charge ratio m/q , starting at rest will gain a kinetic energy equal to the change in potential multiplied by its charge. If the potential as a function of the position of the particle (ℓ) is defined by $U(\ell)$ this can be written as

$$K(\ell) := \frac{mv^2}{2} = q(U(\ell) - U_0), \quad (3.1)$$

3.1. Principles of Time-of-Flight Mass Spectrometry

where $K(\ell)$ is the kinetic energy at position ℓ and U_0 is the potential at the initial position. In a one dimensional model, we can then calculate the time it takes an ion to travel a length L under this potential by integrating the inverse of velocity ($1/v$) over the path as

$$t = \sqrt{\frac{m}{2q}} \int_L \frac{d\ell}{\sqrt{K(\ell)}}. \quad (3.2)$$

It is clear from eqn. 3.2 that two ions with the same charge will separate by the square root of the ratio of their masses ($t_1/t_2 \propto \sqrt{m_1/m_2}$). This allows the determination of the charge-to-mass ratio of an unknown ion from the difference in flight time from a well known calibrant ion. This eliminates the need to accurately determine the potential through which the ions travel.

A set of identical ions measured in a real TOF-MS device will not have a single well defined time-of-flight. The time-of-flight will be broadened as the ions can not all begin their flight from the same position and perfectly at rest. This broadening limits the ability to separate ion species with very similar charge-to-mass ratios. The ability to separate by charge to mass ratio is know as mass resolving power and is a common figure of merit for mass spectrometers, expressed as $R = \frac{m}{\Delta m}$ where Δm is the full width half max (FWHM) of the mass peak. For a TOF-MS device, resolving power can be expressed as

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t}, \quad (3.3)$$

where Δt is the FWHM of the time-of-flight peak.

The mass resolving power of a TOF-MS can be improved by minimizing the spatial and energy spread of the ions to be measured. Additionally, the design of a TOF-MS may be optimized to form a so-called “time focus” for the ions on the detector. This can be illustrated for an ion distribution with finite spatial extent with a simple TOF-MS device with an initial constant acceleration gradient ($U(\ell) = E\ell$), followed

3.1. Principles of Time-of-Flight Mass Spectrometry

by a constant potential drift region. In this case, the change in time-of-flight of our idealized stationary ion at some small offset from the central starting point $\ell = \Delta\ell_0$ can be calculated from eqn. 3.2 as

$$\Delta t = t_\delta - t_0 \quad (3.4)$$

$$= \int_\delta^{L_{\text{inj}}+L_{\text{drift}}} \frac{d\ell}{K(\ell)} - \int_0^{L_{\text{inj}}+L_{\text{drift}}} \frac{d\ell}{K(\ell)}, \quad (3.5)$$

where L_{inj} is the length of the constant electric field injection region and L_{drift} is the length of constant potential drift region. We then take an explicit form for the potential

$$U(\ell) = \begin{cases} E\ell & \ell \leq L_{\text{inj}} \\ EL_{\text{inj}} & \ell > L_{\text{inj}}, \end{cases} \quad (3.6)$$

where E is the constant electric field strength in the injection region. Solving the above integration with eqn. 3.6 in eqn. 3.5 and accounting for the small difference in initial potential due to the spread in initial position of the ion gives

$$\begin{aligned} \Delta t = & \sqrt{\frac{m}{2qE}} \left(\int_\delta^{L_{\text{inj}}} \frac{d\ell}{\sqrt{\ell - \delta}} - \int_0^{L_{\text{inj}}} \frac{d\ell}{\sqrt{\ell}} \right. \\ & \left. + \int_{L_{\text{inj}}}^{L_{\text{inj}}+L_{\text{drift}}} \frac{d\ell}{\sqrt{L_{\text{inj}} - \delta}} - \int_{L_{\text{inj}}}^{L_{\text{inj}}+L_{\text{drift}}} \frac{d\ell}{\sqrt{L_{\text{inj}}}} \right) \end{aligned} \quad (3.7)$$

$$= \sqrt{\frac{m}{2qE}} \left(2\sqrt{L_{\text{inj}} - \delta} - 2\sqrt{L_{\text{inj}}} + \frac{L_{\text{drift}}}{\sqrt{L_{\text{inj}} - \delta}} - \frac{L_{\text{drift}}}{\sqrt{L_{\text{inj}}}} \right). \quad (3.8)$$

If we now perform a Taylor series expansion on Δt around $\delta = 0$, and keeping up to

first order in δ we get

$$\Delta t \approx \left(\frac{L_{\text{drift}}}{2L_{\text{inj}}^{3/2}} - \frac{1}{L_{\text{inj}}} \right) \delta \quad (3.9)$$

$$= \frac{1}{2L_{\text{inj}}^{3/2}} (L_{\text{drift}} - 2L_{\text{inj}}) \delta. \quad (3.10)$$

Thus we find that we can eliminate the time dependence on the initial spread of our ions up to first order, by making the drift region twice the length of the injection region ($L_{\text{drift}} = 2L_{\text{inj}}$). A TOF-MS that has this property is said to be space-time focusing to first order. A similar concept of time-focusing applies to correcting for a set of ions' initial spread in velocity, known as velocity-time focusing. One approach to forming a velocity-time focus is through a time varying injection field, known as time-lag focusing. In general, a purely electrostatic device can not perfectly correct for both spatial and velocity spread of ions at the same time [51]. Since both the spatial spread and the thermal velocities of the ions result in an energy change from the reference ion, the combined concept of energy-time focus will be used for the remainder of the thesis. In the case of an MR-TOF-MS device, the time-of-flight spread is typically dominated by thermal velocity spread for a small number of ion laps. This thermal velocity spread results in a finite turnaround time of the ions. This turnaround time can be reduced by using a stronger injection potential at the cost of increasing the energy spread due to the spatial distribution.

3.2 Multiple-Reflection Mass Spectrometry

To increase the resolving power and precision of a time-of-flight mass spectrometer it is beneficial to extend the distance of ion flight and reduce the broadening of the TOF

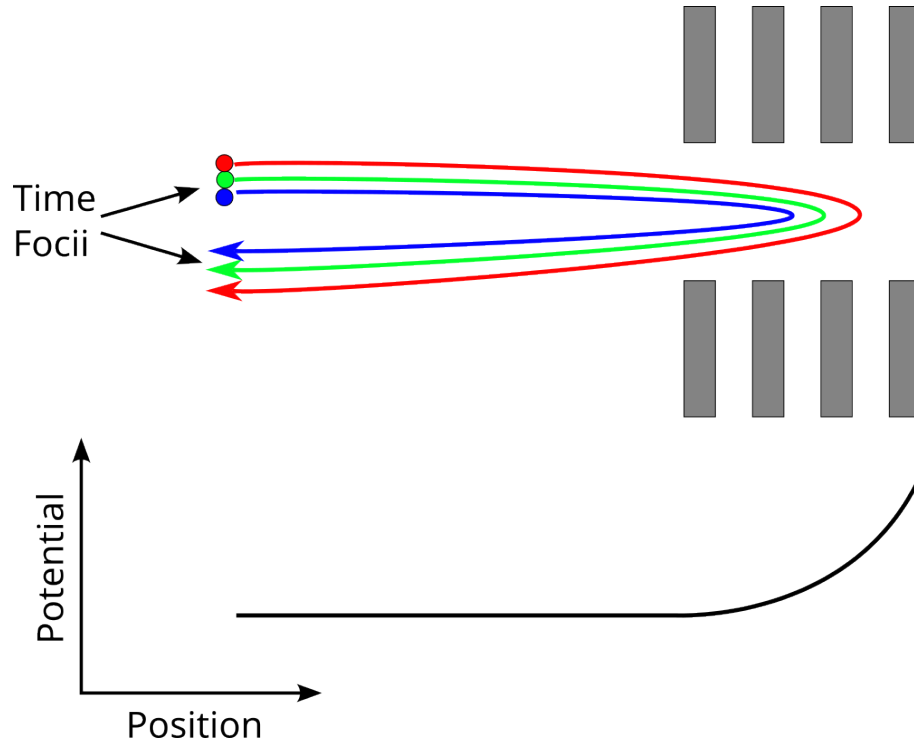


Figure 3.1: An illustration of the different paths ions take through a reflecting mirror, depending on their initial kinetic energies.

peaks due to energy spread. Adding a reflecting element can accomplish both of these tasks if appropriately designed [52]. The reflecting element minimizes the broadening to the time-of-flight peak by allowing higher energy ions to penetrate further into the reflecting region. The higher energy ions then take a longer flight path, compensating their slightly higher speed. The difference in flight path is illustrated in 3.1. The dominant source of peak broadening is then the initial turnaround time of the ions on injection, caused by the random orientation of their thermal velocity. In the case of a multi-reflection device, the ion path length can be extended to several km within a device of length ~ 1 m. If the reflecting mirrors of the MR-TOF-MS device can sufficiently prevent energy broadening, extending the path length in such a way can

improve the resolving power several more orders of magnitude compared to traditional time-of-flight mass spectrometry. The ultimate mass resolving power achievable can then be expressed in terms of the initial bunch spread due mostly to turnaround time (Δt_0) and the peak broadening per lap (Δt_{lap}) as [53]

$$R = \frac{t_0 + N_{\text{laps}} t_{\text{lap}}}{2\sqrt{\Delta t_0^2 + N_{\text{laps}}^2 \Delta t_{\text{lap}}^2}}, \quad (3.11)$$

where t_0 is the time-of-flight of the ion to the detector without any reflections (except for possibly a time focusing reflection as discussed below), t_{lap} is the time for the ion to perform a lap, and N_{laps} is the number of laps done in the analyzer.

It is necessary to separate the isochronous laps from the time-of-flight without any isochronous laps as there exist multiple schemes for putting the final time focus on the ion detector. Two schemes for adjusting the position of the time focus are by use of a post analyzer reflector [54], or by applying a different time focus shift (TFS) potential to the electrostatic mirrors during the first lap of the analyzer [55]. An isochronous lap is then defined by the mirrors having potentials to maintain the time focus of the ions in the same spatial position.

It is instructive to consider two regimes of mass measurement lap number. For small lap numbers, so long as Δt_{lap} is small compared to the turnaround time, the achieved resolving power in an MR-TOF-MS will approximately linearly increase with lap number. Since mass measurement precision is generally linearly proportional to R , increasing lap number directly improves the mass precision of the measurement.

The second regime occurs when the lap number is high ($N_{\text{lap}} \Delta t_{\text{lap}} \gg \Delta t_0$). In this regime the resolving power asymptotically approaches a maximum resolving power of $R_{\text{max}} = \frac{t_{\text{lap}}}{2\Delta t_{\text{lap}}}$. When in this regime, increasing ion laps will not improve resolving

power. In this case, mass accuracy will even be diminished for higher lap numbers as the additional time needed to perform the flight of each ion reduces the statistics that can be gathered.

3.3 TITAN’s MR-TOF-MS

The following subsections describe specific operation of TITAN’s MR-TOF-MS. This MR-TOF-MS system is currently unique among MR-TOF-MS devices at rare isotope beam facilities, in its capacity to remove isobaric contaminants from the received beam and then re-inject the cleaned beam for precision mass measurement [56]. This technique is known as “re-trapping”. To improve the ability to calibrate the subsequent mass measurement a complimentary technique of stable calibrant beam merging has been developed, and is described in section 3.3.6. As an aid to tuning the energy-time focus of the MR-TOF-MS device, an ion energy pulsing technique has been developed and is described in section 3.3.4. Standard mass calibration and data analysis techniques have been previously developed for the TITAN MR-TOF-MS and are described in subsections throughout this section.

3.3.1 Ion Transport and Preparation

During a RIB experiment at TITAN, ions are continuously produced and electrostatically accelerated from the ISAC target to the TITAN RFQ cooler-buncher. On the way to TITAN the ions from the target are mass selected according to their charge-to-mass ratio by a magnetic mass separator. The ISAC mass separator cuts the beam to a single isobaric mass unit, and may also remove some isobaric contamination with its resolving power able to be varied from $\frac{m}{\Delta m} \sim 2000$ to 12000 [57].

The TITAN cooler-buncher [50] is floated to ~ 20 kV to match the energy of the beam delivered by ISAC. Ions enter the cooler-buncher where they are confined by an RF field alternating at several hundred kHz. The ions are cooled by helium buffer gas injected into the system to maintain a pressure of $p \approx 10^{-2}$ mbar. The ions continuously collect and thermalize to the temperature of the buffer gas (~ 300 K). Ions are guided to the end of the RFQ cooler-buncher by a small DC gradient applied to the electrodes. The accumulated bunch of ions can then be extracted with push and pull electrodes to the MR-TOF-MS, with the extraction signal synced to the MR-TOF-MS cycle.

The TITAN MR-TOF-MS system has its own buffer gas filled linear input RFQ. Ions from the cooler-buncher are pulsed down in energy by a drift tube to match the nominal 1.3 kV bias of the MR-TOF-MS. Ions are guided down the input RFQ and through an RF switchyard to a transfer RFQ and then finally to a small preparation trap. While the previous ion bunch is undergoing the final mass measurement in the electrostatic mirror “analyzer” region of the MR-TOF-MS, the ions are allowed in to the injection trap by the lowering in potential of a blocking aperture. Having multiple trapping regions to separate ion bunches is necessary to enable mass selective re-trapping in the injection trap, as described in section 3.3.3. A diagram of the regions of the MR-TOF-MS and ion positions at different stages in a measurement cycle is shown in figure 3.2.

In the simplest case of a mass measurement without mass selective re-trapping, an ion bunch is injected from the injection trap into the mass analyzer to separate by mass. TITAN’s MR-TOF-MS uses a design in which the first ion mirror is brought down in potential to allow the ions in, and then brought back up while the ions are in the opposite mirror to form a trap. During the first lap of the mass analyzer, the ion mirrors are kept on a different potential to that used during the rest of the ion’s

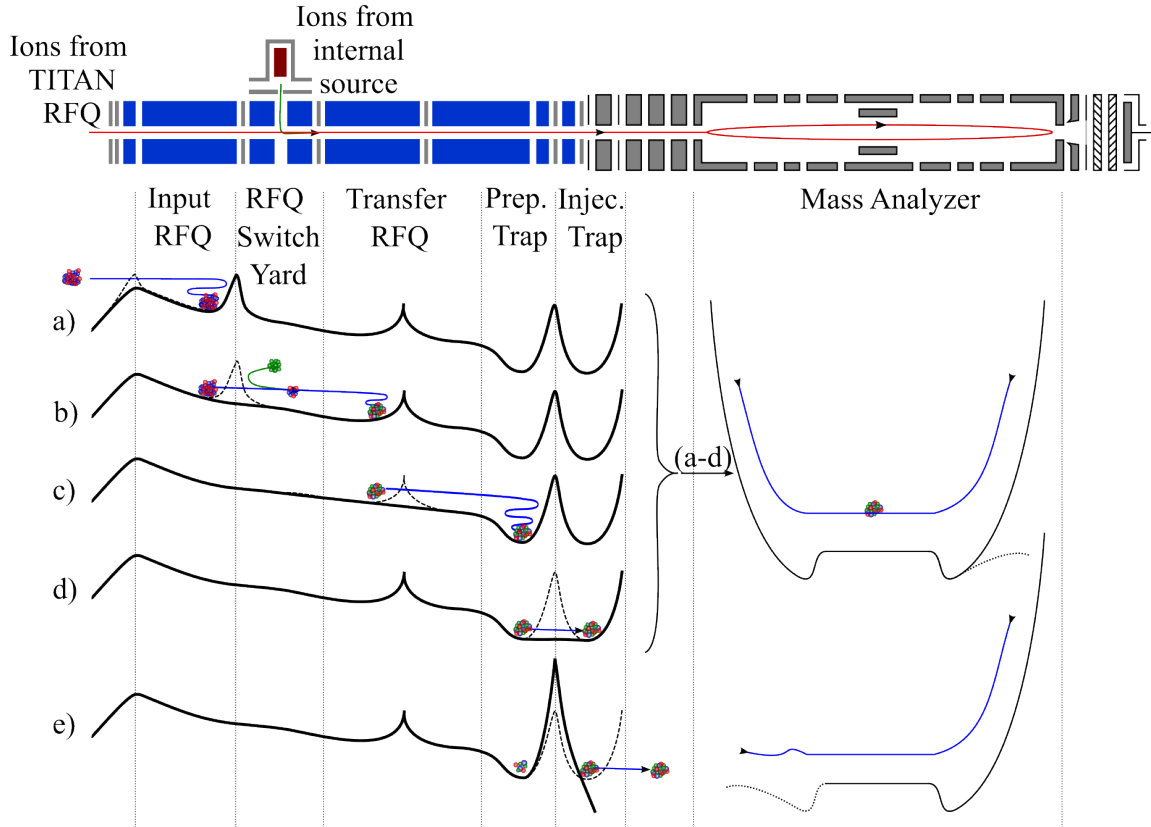


Figure 3.2: Diagram of ion positions during a simple TITAN MR-TOF-MS cycle. From a to d, ions are captured from the TITAN cooler-buncher and prepared for the mass measurement while the previous ion bunch separates by mass in the mass analyzer. At step e, the previous mass measurement has completed and the next ion bunch is injected into the mass analyzer.

flight. This potential is fine tuned to put the time focus of the ions onto the ion detector at the end of the device. This is known as a time focus shift (TFS) and the reflections which accomplish this are called the TFS turns. The mirror potentials are then switched to maintain the position of the time focus. Since these reflections maintain the time focus they are known as isochronous reflection turns (IT). The ions can be detected by lowering the end two electrodes to ground on the final revolution, allowing the ions to impact the MagneTOF ion detector [58].

The use of TFS reflections followed by IT reflections ensures that the MR-TOF-MS can be operated with ions flying a low or high number of laps without retuning the mass analyzer. This flexibility provides several benefits. It allows the MR-TOF-MS to be used at a low number of laps for measuring multiple isobars at once, which has been used to look at molecular reactions happening within the buffer gas. It also allows for maximizing the number of laps for each isotope in a high precision mass measurement. Perhaps most importantly, it allows changing the laps used for mass selective re-trapping (section 3.3.6), making the the window of captured masses tunable.

3.3.2 Mass Calibration

To make a measurement of mass-to-charge ratio of an ion using an MR-TOF-MS requires one to know how long the final flight path of the ion was. This is determined by the number of revolutions the ion completed within the device. This can be calculated from the laps done in the same period of time of a known mass reference, as the path length in a given time scales with the square root of the charge-to-mass ratios. The mass-to-charge ratio of an ion can then be determined from the time of

detection of the ion. This conversion can be parameterized as

$$\frac{m}{q} = c \left(\frac{t}{1 + N_{\text{laps}} b} \right)^2 \quad (3.12)$$

where N_{laps} is the number of isochronous revolutions the ion performs prior to ejection. The parameters c and b are determined by measuring time of flight of ions with well known mass. Cable delays and finite switching time necessitate the introduction of small time offset parameter (t_0). The conversion from measured time (t_{meas}) to charge-to-mass ratio then becomes

$$\frac{m}{q} = c \left(\frac{t_{\text{meas}} - t_0}{1 + N_{\text{laps}} b} \right)^2. \quad (3.13)$$

The calibration parameters of eqn. 3.13 are determined in a two step process. To first determine c and t_0 a measurement is made of at least two well known ions, which undergo only the initial time focus shift (TFS) reflections in the MR-TOF-MS analyzer. These ions should be well separated in mass (by many mass units), to ensure the best possible precision. For the mass measurements presented in this chapter, this calibration was performed with ^{85}Rb , ^{87}Rb , ^{133}Cs . The centroid of each peak is then determined by Gaussian fits and eqn. 3.13 is fit using the centroids and known masses with $N_{\text{laps}} = 0$. This procedure is known as a “zero turn calibration” because it is performed without the ions being subjected to the isochronous mirror potentials. The zero turn calibration for the measurements reported in this chapter is shown in figure 3.3.

The parameter b can then be set with a single calibrant ion performing a high number of isochronous reflections. Here, the advantage of parameterizing the time-to-mass conversion in this way becomes apparent. This calibration allows for freedom in

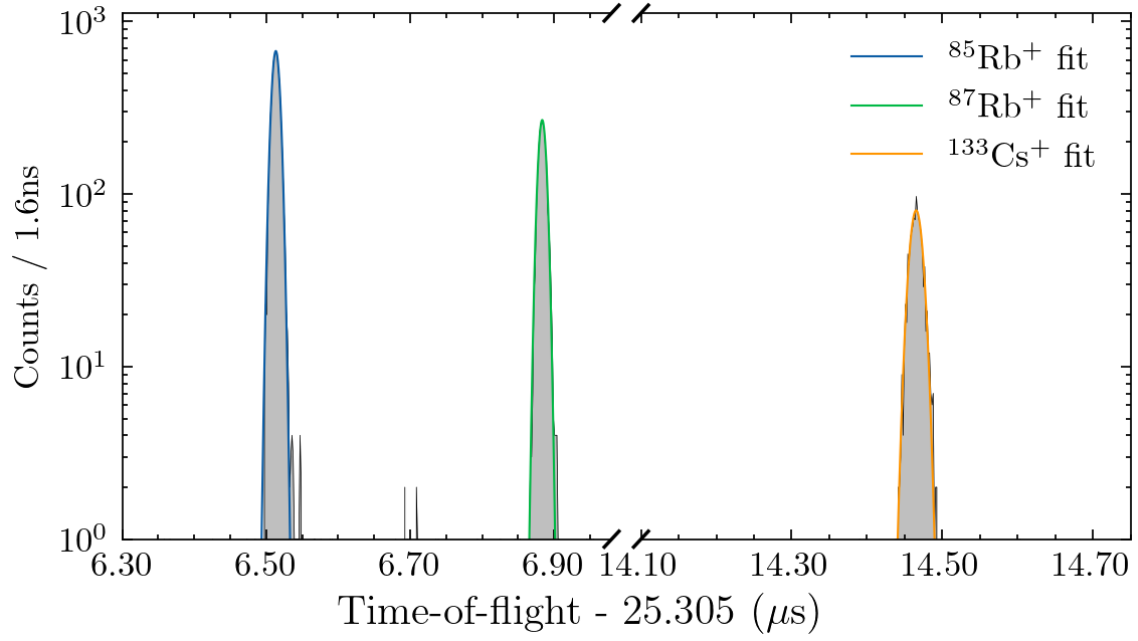


Figure 3.3: Zero turn calibration, showing the stable ion species used for calibration and the Gaussian fits done to determine the time-of-flight centroids. The TDC is triggered when the mirror potential is pulsed down for extraction. Raw TDC timings are displayed.

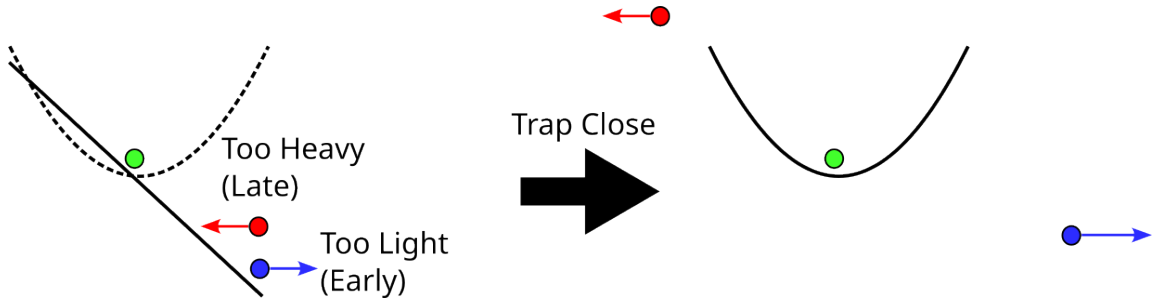


Figure 3.4: A simple diagram of mass selective re-trapping. Slightly early or late ions gain energy from the change in potential as the trap closes. Early ions are already travelling away from the trap while heavy ions gain too much energy to be trapped in the shallow trapping potential of the injection trap.

choosing an appropriate number of reflections for a given measurement. Therefore, it allows maximizing the number of laps performed by the ions within the cycle time of the MR-TOF-MS, no matter the particular number mass unit under investigation. It also allows for a non-isobaric calibrant to be used, despite the ion undergoing a different number of laps than the ion species of interest. This advantage forms the basis for the calibrant mixing technique described in section 3.3.6.

The b parameter above is a sensitive input to the final mass determination. It broadly corresponds to the ratio of ion flight time for a “zero turn” flight and an isochronous lap. Since in a mass measurement the ions will perform many isochronous reflections, small deviations to b from voltage fluctuations will have a very large effect. Therefore the b calibration must be frequently updated. This is done in a nearly continuous manner. The process for this re-calibration is described in section 3.3.5.

3.3.3 Mass Selective Re-trapping

RIB produced via the ISOL method is prone to having a large amount of isobaric contamination. This contamination is mostly stable and long-lived atomic ions and

molecular ions. This contamination can be many orders of magnitude more than the ion species of interest for a mass measurement. For the highest precision mass measurement, ion rates must be kept to ~ 1 isobaric ion in the mass analyzer per mass measurement cycle. To ensure the ion of interest can be measured in a reasonable time without violating this condition, the TITAN MR-TOF-MS uses a technique known as mass selective re-trapping. Re-trapping involves inject a large bunch of ions into the MR-TOF-MS mass analyzer to separate in time-of-flight by mass. Rather than measure the separated ions immediately, the electrostatic mirror on the injection trap side of the analyzer is opened and the trap is closed just as the ion species of interest enters the trap (see figure 3.4). The mirror is then brought back to its closed voltages preventing other ion species from also entering the injection trap.

The ions that are not re-trapped are steered into the walls of the electrostatic analyzer. The re-trapped ions are re-cooled for several ms, This technique has been shown to give the TITAN MR-TOF-MS system the ability to reduce contamination ions by a factor of 10^4 . Since the MR-TOF-MS system has demonstrated an ability to detect ions against a total detected background of 10^4 contaminant ions, the TITAN MR-TOF-MS system can be said to have a total dynamic range of 10^8 contaminant ions to ion species of interest.

The contamination suppression can be tuned by changing the number of laps the ions undergo before being re-trapped in the injection Paul trap. Increasing the number of turns will decrease the window of re-trapped masses. Contaminant suppression can also be increased by reducing the depth of the ion trap, although this will impact efficiency. For deep trap depths, separation powers in terms of FWHM of the re-trapping window of $\frac{\Delta m_{\text{fwhm}}}{m} \sim 1 \times 10^5$ can be achieved, while maintaining species of interest efficiency greater than 80%.

3.3.4 Isochronicity and Measuring Energy-Time Focusing in TITAN's MR-TOF-MS

The literature on focusing in MR-TOF-MS and MR-TOF-MS-like devices has largely focused on proving that periodic electrostatic ion optics can confine ions to stable orbits and correct for small ion energy differences [59, 60]. Some work has also been done on analytically calculating electric fields for ion mirrors with cylindrical symmetry [61], like the mirrors of the TITAN MR-TOF-MS. Since the ability to analytically describe the ion motion is limited, design of the electrostatic mirrors of an MR-TOF-MS is typically largely guided by numerical ion simulations [62]. This has proven to be sufficient for developing MR-TOF-MS devices capable of resolving powers of $\frac{m}{\Delta m} \sim 10^6$ [63, 64]. However, these simulations are heavy; taking a large amount of time to compute and requiring large amounts of memory. This makes them impractical for guiding the fine tuning process required for optimal performance. Tuning an MR-TOF-MS is therefore a time consuming task as all changes to electrode voltages must then be tested on the physical device.

The tuning procedure for the TITAN MR-TOF-MS consists of scans of electrode potentials over small ranges in steps of 100 mV or less. At each trial potential the time-of-flight of many ions is detected to measure the dispersion in time of the ions. Getting sufficient statistics to measure this dispersion at each point in a scan takes a substantial amount of time. The time necessary for each measurement can also result in the ion time-of-flight peak width being convoluted by small fluctuations of the type corrected for in section 3.3.5. In the interest of guiding and speeding up the tuning process, a method of directly measuring the isochronicity of the mirror potentials has been developed.

The isochronicity measurement is accomplished by deliberately changing the

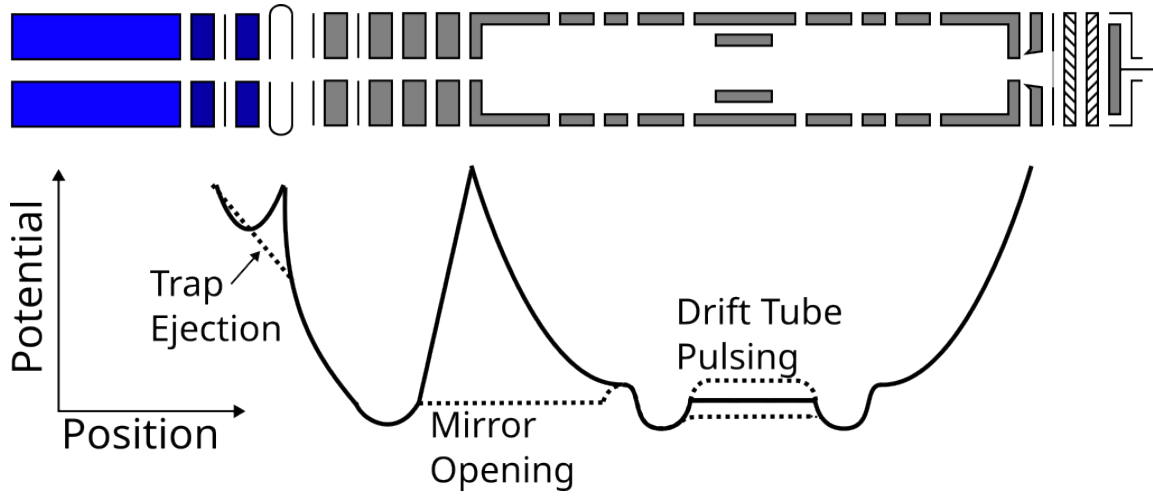
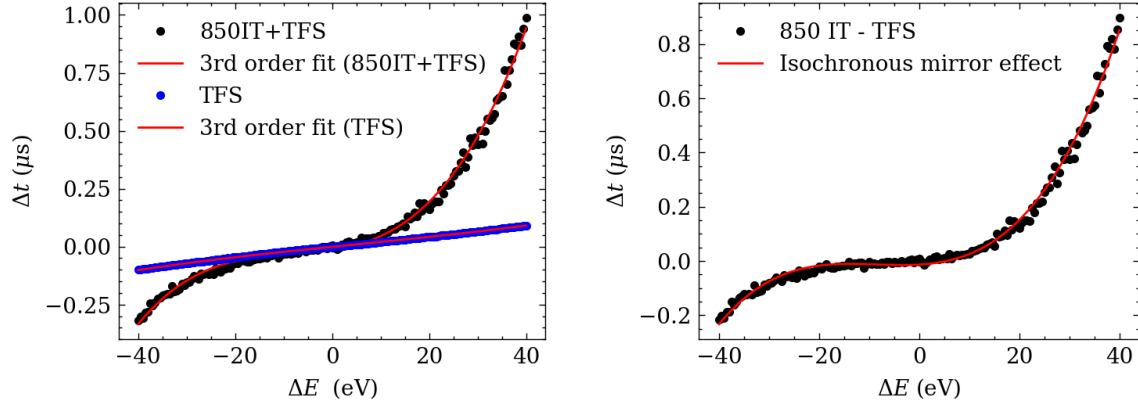


Figure 3.5: Diagram of the potentials applied during the injection phase. The drift tube of the mass analyzer is pulsed from its normal grounded potential while the ions are within in order to change the ion energy, and is only done when measuring the isochronicity. Potentials applied during injection are shown with a dashed line.

energy of the ions in the mass analyzer. The change in time-of-flight due to the energy change is measured. This is done over a range of energies typical of the ion beam emittance of the MR-TOF-MS's injection Paul trap. The beam energy is changed by pulsing the central drift tube of the mass analyzer as the ions are within. When the ions leave the drift tube they will be accelerated and continue with their kinetic energy changed by an amount equal to the voltage pulse. The drift tube is pulsed for ions on their first pass through the mass analyzer and is then kept on ground for the remainder of the ion's flight to replicate normal mass measurement conditions. A complete schematic of ion injection with drift tube pulsing is shown in figure 3.5. To minimize the influence of voltage drifts and thermal expansion, each energy pulsed measurement is followed by an unpulsed ($\Delta E = 0$) measurement. The time-of-flight change is determined by the difference between the pulsed time-of-flight and the unpulsed measurement immediately following.



(a) Change in the time of flight for 850 isochronous laps and time focus shift lap only.

(b) 850 isochronous lap effect. The effect on the time of flight from pulsing before the time focus shift has been subtracted off.

Figure 3.6: Measurement of the degree of isochronicity of the TITAN MR-TOF-MS. By subtracting the fit to the time shift with energy change with only the time focus shift lap we get the isochronicity of the isochronous mirrors.

The TITAN MR-TOF-MS has been designed to correct for ion energy spread to third order [55]. It is therefore expected that the change in ion time-of-flight (Δt) will be well approximated by a third order polynomial in energy change (ΔE). A cubic fit of time-of-flight change can be seen in figure 3.6a. The measurement was done for both a high number of isochronous laps and for zero isochronous laps. The TFS turns were done in all cases in order to place the time focus on the ion detector surface. It can be seen that there is a clear slope in Δt near the nominal beam injection energy ($\Delta E = 0$). This is explained by the initial flight from injection trap to the drift tube as the time energy focus is set for ions with different energies right from the injection trap. This can be dealt with by subtracting off the 0 IT effect from the high turn data resulting in a measurement of the isochronicity of the IT mirror potentials. The result of the subtraction of the two fits is shown in figure 3.6b.

This direct isochronicity measurement is leveraged in tuning as a partial expla-

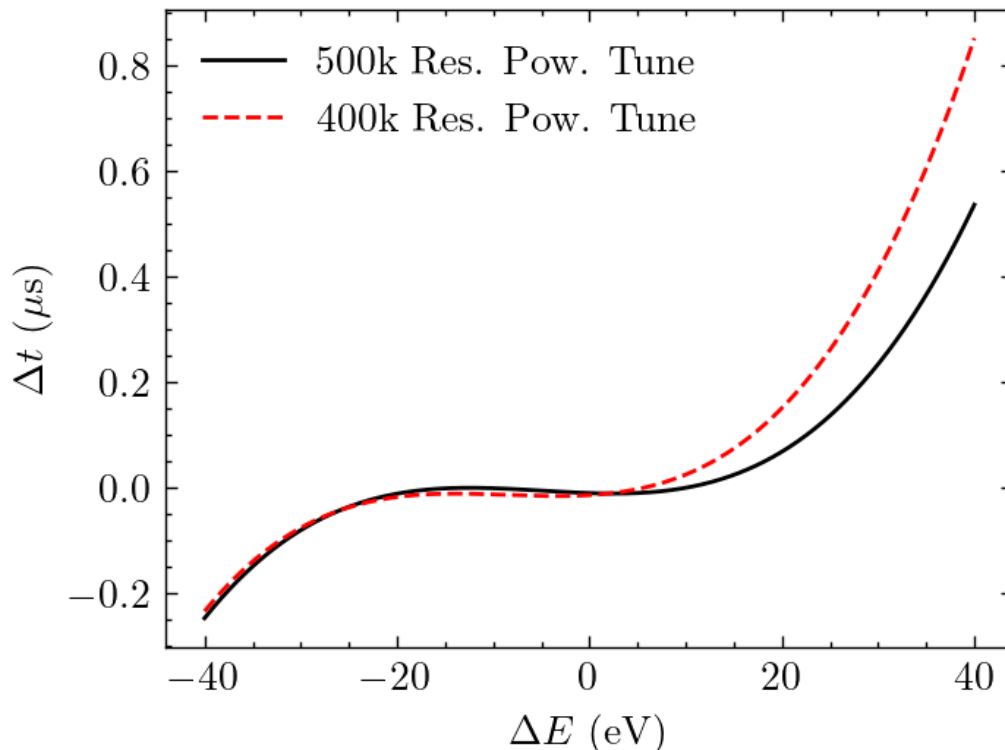


Figure 3.7: Comparing the isochronicity of two MR-TOF-MS tunes. Measurements were taken before and after the weeks long tuning procedure in preparation for a rare isotope beamtime.

nation of poor analyzer performance. While scanning and measuring time-of-flight dispersion informs which electrode potentials decrease dispersion, the isochronicity measurement can guide a procedure to improve performance. If the isochronicity measurement reveals that the analyzer is tuned to be isochronous for slightly low energy ions it can be quickly corrected by reducing the potential of the injection trap. Alternatively the mirror electrode potentials may be scaled up the ratio of the nominal energy to the isochronous energy of the current tune. If the shape of the isochronicity curve has changed, then this can reveal that adjacent electrode mirrors are not forming a good potential to be isochronous over a large energy range.

Offline experiments into measuring the isochronicity of the TITAN MR-TOF-MS have yielded encouraging results with direct isochronicity measurement. In preparation for the neutron-rich potassium measurements presented in section 3.4, a tuning campaign guided by direct isochronicity measurements was done. For each iteration of electrode adjustment, an isochronicity measurement was performed before and after analyzer changes. At the end of the tuning procedure, typical mass resolving power had been increased from 4×10^5 to 5×10^5 , a 20% improvement. A comparison of the analyzer isochronicity before and after the guided tuning is shown in figure 3.7.

3.3.5 Time Resolved Calibration

Ions in an MR-TOF-MS type device undergo many revolutions and sample the electric field of the mass analyzer many times. This makes the final ion timing fluctuate with small voltage and length changes to the analyzer from temperature drifts in the experimental hall. These timing drifts are often an order of magnitude larger than the inherent timing peak width of the ions. To get the maximum resolving power from an MR-TOF-MS system thus requires some method of correcting these slow drifts in time of flight.

To minimize the scale of the time of flight drifts due to potential changes, power supplies with low temperature coefficients may be chosen. Alternatively, active voltage or temperature stabilization may be employed. Examples of active voltage stabilization can be seen in the RIKEN SLOWRI and GARIS-II MR-TOF-MS's [65] and ISOLDE's ISOLTRAP MR-TOF-MS device [66]. In the case of TITAN's MR-TOF-MS, many of the most sensitive electrodes are biased with very low temperature coefficient high voltage supplies (E.g. ISEG EHS series). At TITAN, the mirror electrodes of the MR-TOF-MS are further stabilized by use of fourth order low pass

filters with a time constant of ~ 1 s. There is also an ongoing effort to build an actively temperature stabilized rack to hold the power supplies which bias the outer mirror electrodes. The time of flight of the ions is most sensitive to the outer mirror electrode potentials as this is where the ions are moving slowest and experience the potential for the longest period of time.

The other major source of time of flight drift is mechanical expansion of the analyzer. It is not feasible to temperature stabilize the entirety of the ISAC hall or the MR-TOF-MS system. TITAN's MR-TOF-MS is therefore made of a low thermal expansion coefficient metal known as Invar [67, 68].

Despite some effort being made to stabilize the system, time of flight drifts remain and must be corrected. This is done using an ion species coming at a rate sufficiently high that an accurate centroid time of flight for the species peak can be determined on a time scale shorter than the time of flight drifts. This high rate species is then used to perform a time resolved calibration (TRC) [69]. The known mass of the calibrant species is used to update the b parameter in eqn. 3.13. The data is then exported as a single cumulative mass spectrum with slices of the data corrected with the appropriate b parameter correction applied. This procedure is illustrated in figure 3.8 with data taken for $A/Q = 54$ u/e. The centroid was determined using the median of ion time of flight in a range of $\pm 0.029 \mu\text{s}$ (± 0.0005 u/e) of the previous centroid position. The data were chunked into 8 second accumulations, providing ~ 50 ions per chunk. The tracked peak was identified as $^{54}\text{Cr}^+$.

When performing a TRC a balance must be struck between correcting time of flight drifts on short time scales, and gathering sufficient statistics in each data chunk to accurately determine the peak position. Sufficient statistics must also be gathered to avoid artificially narrowing the TRC peak. This is particularly important if the species chosen for the TRC will also be the peak shape calibrant, or has substantial

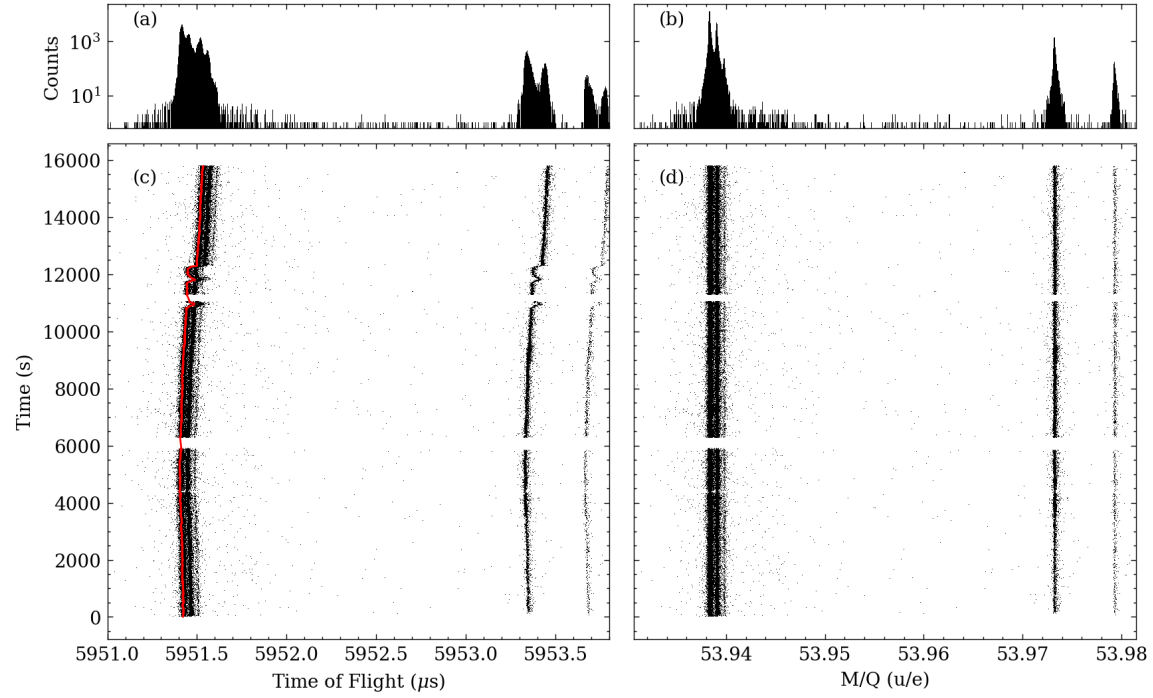


Figure 3.8: A visualization of the TRC process for $A/Q = 54\text{u/e}$. In panel (b) is the detected time of flight of ions during the experiment, sliced into single second accumulations. The centroid of the TRC calibrant is marked in red. In panel (a), the resulting time of flight spectrum is shown. In panels (b) and (d), the total mass spectrum and the sliced mass data respectively, are shown after the TRC is applied. A clear improvement in resolving power is seen after the TRC is performed, and phantom peaks due to time of flight shifts disappear.

overlap with the species of interest, or the shape or mass calibrant. In all mass measurements presented, the TRC calibrant is well separated from the ion of interest and shape calibrants, minimizing this potential source of systematic error.

3.3.6 Stable Calibrant Beam Merging

There is no guarantee that a suitable TRC calibrant will be delivered to TITAN from ISAC. TRC's are less reliable when the ion species used is not sufficiently separated in time of flight from other ion species. Additionally, the TRC ion species must come at a rate high enough to be able to be used to track fluctuations in the time of flight. However if the ion rate is too high, then there will be more than one ion flying in the MR-TOF-MS analyzer in each cycle resulting in ion-ion interaction, reducing the mass accuracy. This ion-ion interaction is particularly bad if the ions are isobaric, since they will separate in mass very slowly. It is therefore ideal to be able to mix in a controlled amount of a non-isobaric calibrant from a source internal to the MR-TOF-MS system. This can be done with TITAN's MR-TOF-MS's combined thermal and electron impact ion source [70].

To accomplish this, an ion manipulation technique dubbed "stable calibrant beam merging" has been developed. The main principle of beam merging is to collect stable ions in the MR-TOF-MS RFQ system during the short period while a radioactive ion bunch is cycling in the mass analyzer and before the next bunch of radioactive ions has been transferred from the TITAN RFQ. Stable ions first are transferred through the MR-TOF-MS switchyard and collect continuously behind the transfer RFQ aperture in the center of the transfer RFQ. Radioactive ions are re-trapped in the injection trap of the MR-TOF-MS. Then, the now purified radioactive ions are moved from the injection trap into the preparation trap. The aperture in the transfer

RFQ can now be lowered to allow stable ions to mix with the purified radioactive ions. The combined bunch is finally moved back in to the injection trap and allowed to cool before injection into the mass analyzer for mass measurement.

A schematic representation of the beam merging process is shown in figure 3.9. Not shown is the optional RF gating done prior to collecting stable ions in the transfer RFQ. This can be used to ensure that any contaminants in the radioactive ion beam that may not have been transferred to the injection trap in time for the re-trapping cycle are not reintroduced during beam merging.

A mass measurement is normally completed with ions doing the maximum number of laps possible to ensure the best possible resolving power. This is limited by the chosen cycle time of the MR-TOF-MS, chosen to balance resolving power with decay losses and the time needed to collect sufficient statistics. In the case of low mass ion species, the total laps in the MR-TOF-MS is also limited to keep isobars from lapping each other before detection. The introduction of stable calibrant beam merging adds an additional constraint to the selected lap number. As the added calibrant is usually not an isobar of the ion species to be measured, the merged calibrant will undergo a different number of laps. The lap number must be carefully chosen to ensure the merged calibrant will not overlap with the ion species of interest, or the isobaric contaminants that will be used for peak shape and final mass calibration.

3.3.7 Mass and Shape Calibration

The velocity and spatial distribution of ions in the MR-TOF-MS's Paul trap averaged over the ion micromotion can be well approximated by Gaussian functions [71]. The time of flight peaks detected by the MR-TOF-MS are therefore expected to be approximately Gaussian, as the injection and free flight spread of the ions should

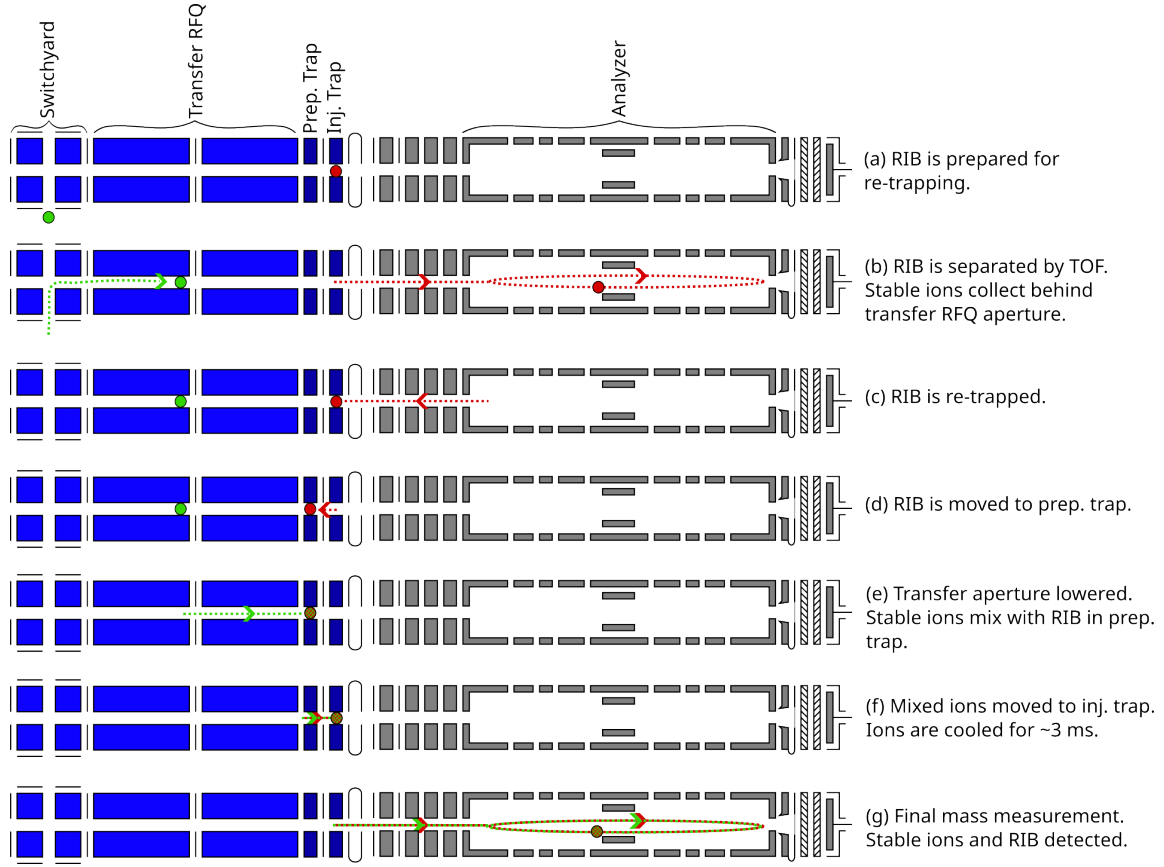


Figure 3.9: A schematic representation of beam merging. Radioactive ions in red are shown from initial transfer from the TITAN RFQ through the re-trapping cycle and subsequent mixing with stable ions. Stable ions are depicted in green.

largely preserve the characteristics of the initial ion cloud. However, the peaks detected will differ from exact Gaussian distributions as the initial distribution is not exactly Gaussian. Additionally, the ions will be effected by ion-optical aberrations as discussed in section 3.3.4. In order to make the most precise measurements, particularly when detected species are not fully resolved, the detected time of flight distribution must be modelled as accurately as possible. It has been found that a hyper-exponentially modified Gaussian (hyper-EMG) distribution can accurately model the detected peak shape of MR-TOF-MS data [72]. The hyper-EMG distribution is defined by a central Gaussian with some number of exponential tails on each side. It can be written as:

$$h_{emg}(x) = \Theta h_{emg(-)}(x) + (1 - \Theta) h_{emg(+)}(x) \quad (3.14)$$

where Θ is a weighting parameter that corresponds to the contribution of the exponential tails on each side of distribution. The $h_{emg(\pm)}$ functions are defined by a peak centroid (μ), a width parameter (σ), and an arbitrary number of exponential tail weights (η) and decay constants (τ). The two components of the distribution, with N_+ positive and N_- negative tails can then be written

$$h_{emg(\pm)}(x) = \sum_i^{N_{\pm}} \frac{\eta_{i\pm}}{2\tau_{i\pm}} \exp\left(\left(\frac{\sigma}{\sqrt{2}\tau_{i\pm}}\right)^2 \mp \left(\frac{x - \mu}{\tau_{i\pm}}\right)\right) \operatorname{erf}\left(\frac{\sigma}{\sqrt{2}\tau_{i\pm}} \mp \frac{x - \mu}{\sqrt{2}\sigma}\right). \quad (3.15)$$

Fitting the mass data is done with an in-house developed open source python package called EMGFIT [73]. The code by default will fit all peaks in the spectrum with the same peak shape. This is done because it is assumed all isobaric ions will experience similar conditions in the MR-TOF-MS analyzer. When measuring the system with light ions, it is important to keep the total laps in the analyzer low enough that isobaric species do not lap each other, and thus sample the electrode

potentials an equal number of times. The peak shape is typically fixed by first making a fit to a well separated, high-statistics peak. This is followed by a global fit to all peaks in the mass spectrum with an additional background term. At this stage, a peak with a precisely determined centroid is used to make the final mass calibration. It is important that this peak can be confidently assigned to an ion species that has had previous high-precision mass measurements made. If possible a species with no known long-lived low-energy isomers should be used to limit systematic effects that could be caused by unresolved species.

3.3.8 Systematic Uncertainty

In addition to statistical fitting uncertainty, the precision of mass measurements achieved with the TITAN MR-TOF-MS are limited by several sources of systematic uncertainty. The dominant source of this uncertainty is non-ideal injection and ejection in the MR-TOF-MS analyzer. The mirrors on each end of the analyzer must be switched from their high-voltage reflecting potential to allow the ions to pass into and out of the analyzer region. In the case of final ejection of the ions onto the ion detector, the ions will have separated substantially due to their different masses. The ejection must be done with minimal disturbance to the ions and with each species being subjected as close as possible to the same analyzer potentials.

The effect of non-ideal ejection is measured immediately prior to receiving radioactive beam. The opening time of mirror in front of the detector is scanned over almost the full lapping time of the stable ion species used. The ion species does just the TFS lap in the analyzer before measurement to ensure that drifting potentials do not cause a significant change in the time of flight of the ions during the test.

An upper bound on the systematic uncertainty caused by non-ideal injection

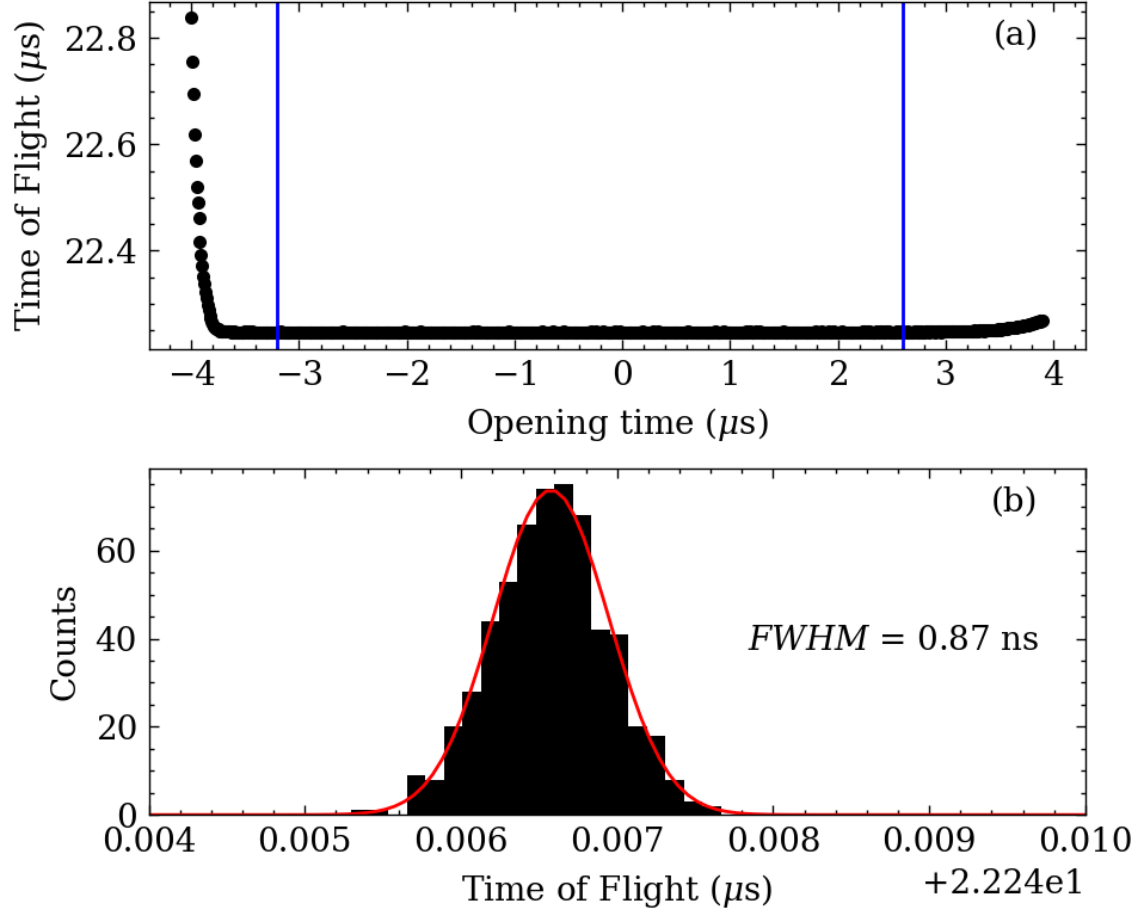


Figure 3.10: (a) A scan of the time of flight of ions as a function of the opening time of the detector side analyzer mirror. When the mirror is switched with ions in the mirror, they are greatly affected by the changing potential. The region of “good” opening times is marked in blue and used to determine the non-ideal ejection uncertainty.

(b) A histogram of peak centroids (Gaussian fit) for each opening time delay in the “good” region above.

can be calculated from the spread in ion time-of-flight peak centroids across mirror opening times. An example of this type of measurement, performed with ^{39}K is shown in figure 3.10. It can be seen that when the mirror opening time is changed to far from its nominal opening time, the ions have their final time-of-flight effected significantly. This is a result of the ions being in a quickly changing potential, and thus changing their energy. The data used for analysis for systematic uncertainty is cut to a region in which the ions do not experience a quickly changing field. The region of this cut is also used to calculate the maximum time-of-flight spread that ion's may undergo during a precision mass measurement, so that no ions will see this changing field.

To calculate the systematic uncertainty resulting from the non-ideal ejection, the time-of-flight spread of the stable ion (Δt) with mass m_{cal} is used to calculate a relative mass uncertainty as:

$$\frac{\Delta m_{ejection}}{m} = \frac{2\Delta t}{t} \sqrt{\frac{m}{m_{cal}}} \quad (3.16)$$

where t is the total time-of-flight of the ion in the mass measurement. In the case of mass measurements in this thesis, this is the dominant source of systematic error, with a magnitude of $\frac{\Delta m_{ejection}}{m} \approx 1 \times 10^{-7}$.

A potential source of systematic error is the ion-ion interaction between ions in the analyzer. As a Coulomb force, the interaction is strongest between ions close together. The effect due to ion-ion interaction can be quantified in terms of the ion rate of isobaric species. It has been found that this effect is negligible ($\frac{\Delta m_{ii}}{m} \sim 1 \times 10^{-8}$ relative uncertainty) if the isobaric ion rate is kept to ~ 1 ion per cycle or less [74]. For this reason, the isobaric ion rate was kept below 80 pps for all mass measurements presented in the thesis.

The TRC procedure described in section 3.3.5 cannot perfectly correct the position of ions at every moment in the measurement. The TRC is limited by finite statistics, and the non continuous nature of the data chunking used to compute the corrections. Since the ion species of interest is often delivered to TITAN at a low rate, the slight uncertainties in the TRC correction may not be washed away by statistics in the finite time afforded to the measurement. A TRC uncertainty can be defined in terms of the change in TRC correction between successive data chunks. The root mean square deviation of the b parameter in successive chunks is computed and the uncertainty attributed is defined by:

$$\frac{\Delta m_{TRC}}{m} \approx \frac{\Delta b_{r.m.s}}{\sqrt{N_{TRC}}} \quad (3.17)$$

where $\Delta b_{r.m.s}$ is the root mean square deviation of successive b values, and N_{TRC} is the total number of data chunks used for the TRC. In the case of all measurements in this thesis the value of $\frac{\Delta m_{TRC}}{m}$ is less than 5×10^{-8} .

The last source of systematic uncertainty comes from the uncertainty in the mass of the mass calibrant (Δm_{cal}). In all cases presented in this thesis, this uncertainty is negligibly small as the mass calibrant was a stable species having previously measured masses with precision orders of magnitude better than MR-TOF-MSs are currently capable of. In no cases during the measurement presented in 3.4 was there any concern of unresolved isomers in the mass or shape calibrant or the species of interest.

The final uncertainty in the measurement can be computed by adding in quadrature the statistical fitting uncertainty and each systematic uncertainty giving a final relative uncertainty of:

$$\frac{\Delta m}{m} = \sqrt{\left(\frac{m_{stat}}{m}\right)^2 + \left(\frac{m_{ejection}}{m}\right)^2 + \left(\frac{m_{ii}}{m}\right)^2 + \left(\frac{m_{TRC}}{m}\right)^2 + \left(\frac{m_{cal}}{m}\right)^2} \quad (3.18)$$

3.4 Mass Measurements of $^{51-54}\text{K}$

In the case of the mass measurement along the isotopic chain of potassium an MR-TOF-MS cycle rate of 80 Hz was chosen. This cycle rate was sufficient for mass resolving power to be limited by ion optical aberrations, and was short enough to avoid lapping of isobaric species. The determined half-lives in the NUBASE2020 compilation [75] of $^{51-54}\text{K}$ ranges from 365 ± 5 ms to just 10 ± 5 ms. The 80 Hz cycle time therefore keeps the cycle length to less than about ~ 1.5 half-lives of the shortest lived isotope. This has consistently been shown to be the best balance of increased resolving power, and acceptable statistics gathering for the most precise mass measurements using the TITAN MR-TOF-MS. A special alternate setup at 160 Hz and with re-trapping removed from the MR-TOF-MS scheme was also prepared. This was used to attempt a measurement of ^{55}K , with an expected half-life of 3 ms [75], although no ^{55}K was seen in the experiment.

It was found that the peak shape that best fit the mass spectrum without over-fitting was a hyper-EMG distribution with two positive and one negative exponential tail. Over-fitting of the peak shape was identified by the reduced χ^2 being less than 1 or if the amplitude of an exponential tail agreed with zero within fitting uncertainty. The global best fit of the mass spectra for A/Q 53 and 54 are shown in figures 3.11 and 3.12 respectively. Refined fitting uncertainties were calculated using a random walk Markov-Chain Monte Carlo (MCMC) resampling method [76]. This resampling of peak shape parameters and refitting allowed for fits to be performed for a representative set of realistic peak shapes, accounting for parameter correlations. Synthetic spectra were also generated from the fits and the synthetic spectra were refit. This parametric resampling give more realistic uncertainties for peak amplitudes and

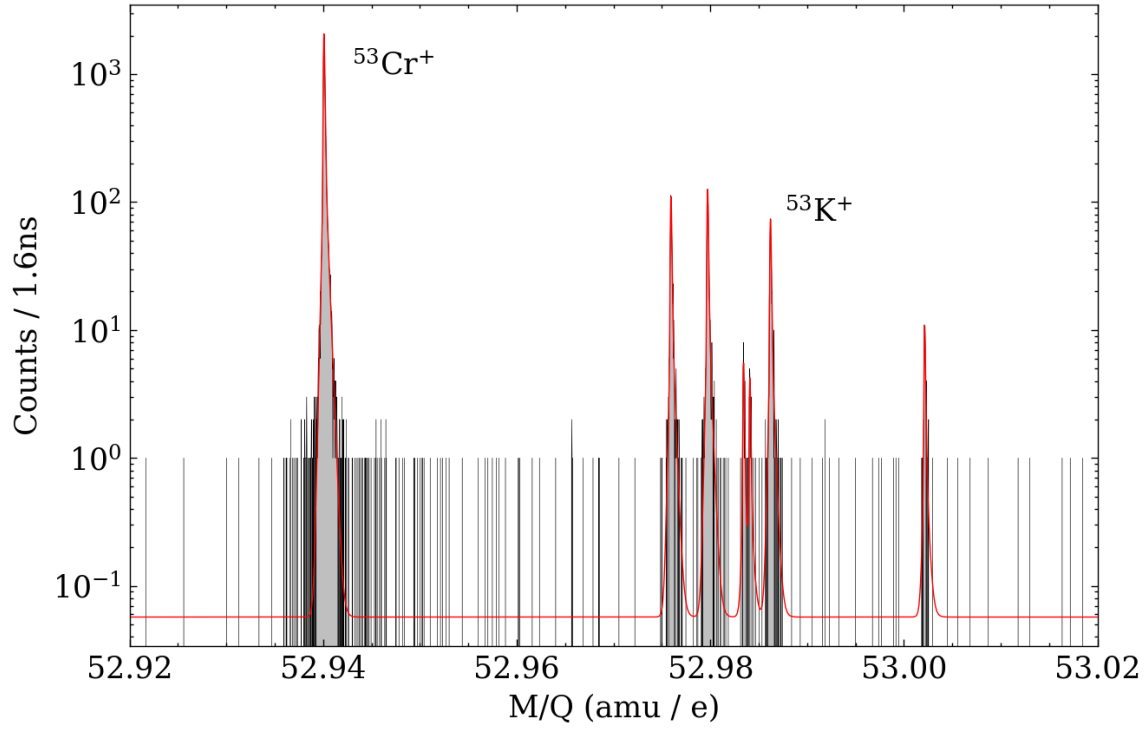


Figure 3.11: Combined mass spectrum for $M/Q = 53$. The peak shape and mass calibrant (^{53}Cr) is labelled as is the ^{53}K peak. In red is the global fit to the spectrum. Molecular contaminants are fit, but not labelled.

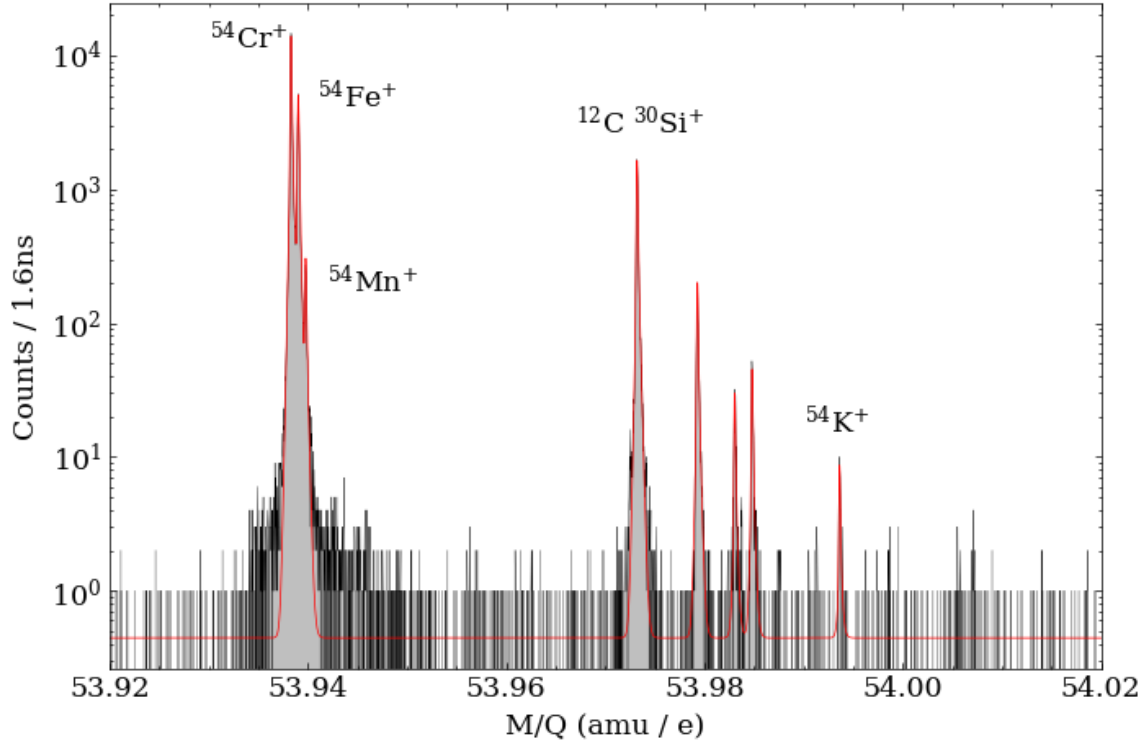


Figure 3.12: Combined mass spectrum for $A/Q = 54$. A selection of peaks are labelled according to identifications based on mass.

3.4. Mass Measurements of $^{51-54}\text{K}$

Species	Mass calibrant	Ionic mass ratio	ME (keV)	ME _{AME} (keV)	Difference (keV)
^{51}K	$^{51}\text{V}^+$	1.000625850(126)	-22504.3(6.0)	-22515(13)	11(14)
^{52}K	$^{52}\text{Cr}^+$	1.000790006(115)	-17197.6(5.6)	-17140(30)	-58(31)
^{53}K	$^{53}\text{Cr}^+$	1.000871287(123)	-12321.5(6.1)	-12300(110)	-20(110)
^{54}K	$^{54}\text{Cr}^+$	1.001025451(262)	-5413.4(13.1)	-5150(400#)	-260(400)

Table 3.1: Atomic mass excess values for the potassium isotopes measured in this work. The last two columns correspond to the literature values from AME2020 [7].

centroids for low statistics peaks. The difference in centroid values from the centroid of the mass calibrant in the MCMC refits were used to calculate the peakshape fitting uncertainty for each peak. A detailed description of the fitting uncertainty determination is give in [77]. A summary of the experimentally determined masses of potassium isotopes is given in table 3.1. In figure 3.13 the experimental mass excess values are compared to the tabulated values from the Atomic Mass Evaluation (AME2020) [7].

It is common to confirm the assignment of the ion of interest peak by blocking lasers used to resonantly ionize the species at the target [17]. In this experiment, the potassium delivered to TITAN was ionized through surface ionization in a hot cavity. This meant that no ionization lasers were in use. In the case of ^{54}K , there was no existing mass measurement to compare against. Instead, a new beta decay detection technique was used to confirm the ion species assignment. This is described in chapter 5.

The consistency of the mass uncertainties was verified by comparing confidently identified atomic and molecular contaminants to their AME2020 [7] masses. The relative deviations from the literature masses are shown in figure 3.14. The 1σ deviation of the TITAN experimentally determined masses was $\frac{m_{lit}-m_{exp}}{m_{lit}} \approx 4.5 \times 10^{-8}$. This corroborates the calculation of the systematic uncertainty of the TITAN MR-

3.4. Mass Measurements of $^{51-54}\text{K}$

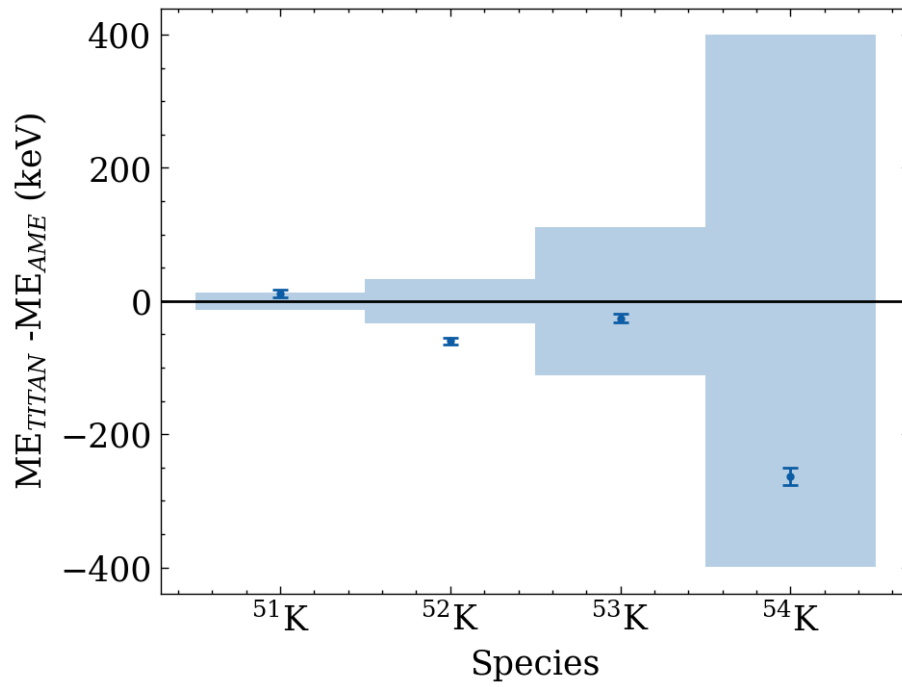


Figure 3.13: Comparison of experimentally measured masses (points) of potassium to values reported in AME2020 [7] (shaded bands). ^{54}K had no previous high-precision mass measurement and was reported in AME2020 with an extrapolated mass based on the trends of other isotopes in the region.

TOF-MS setup in this experiment.

3.4. Mass Measurements of $^{51-54}\text{K}$

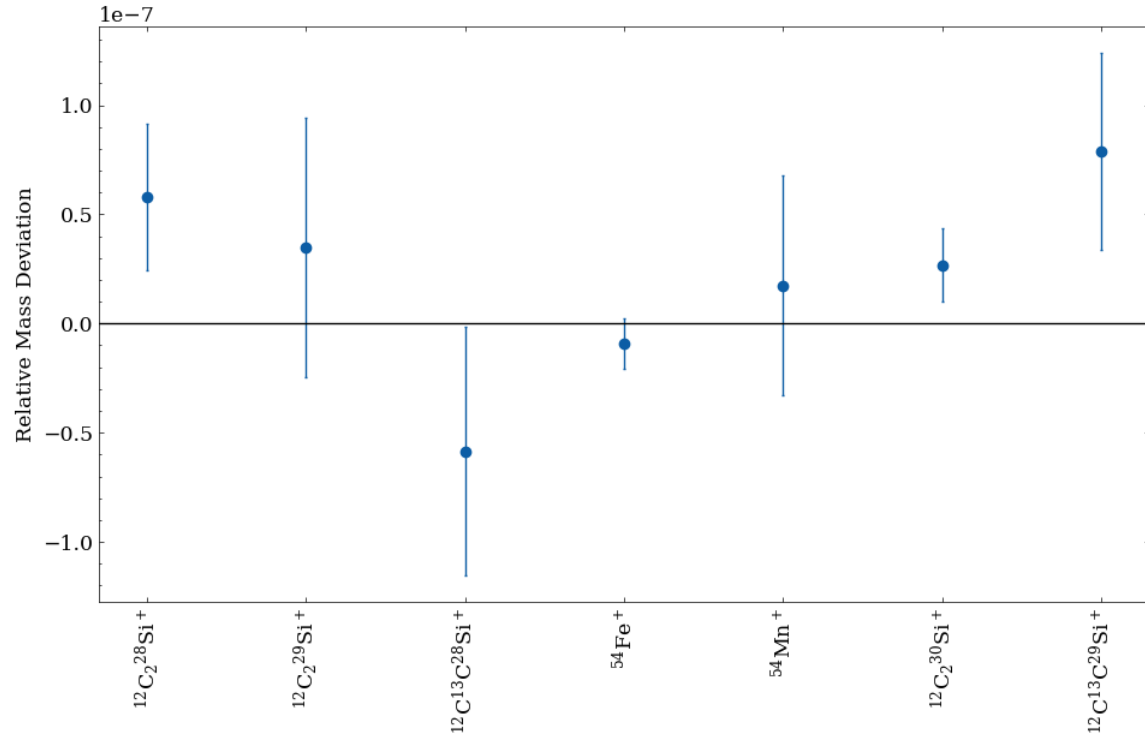


Figure 3.14: Comparison of all confidently identified contaminant masses determined during analysis of the $^{51-54}\text{K}$ data. Relative mass deviation from AME2020 [7] is shown. Uncertainties represent statistical fitting uncertainty only.

CHAPTER 4

The $N=34$ Subshell Closure in Neutron Rich Potassium

The nuclear magic numbers described in Chapter 2 are associated with nuclear shells characterized by substantial energy gaps to the subsequent shell. These pronounced energy gaps confer significant stability to these canonical magic numbers, even when examining nuclei that deviate from stability. Nevertheless, certain magic numbers are known to abruptly vanish, while novel magic numbers emerge in highly exotic nuclei. For example, the $N = 28$ magic number quickly diminishes in strength within very neutron-rich nuclei [78], and a new energy gap emerges at $Z = 40$ in moderately neutron-rich nuclei [79].

Over the last 25 years, evidence has mounted that strong magic properties appear around $N = 32$ in neutron rich nuclei. The onset of a shell gap at $N = 32$ (when approaching from stability at high Z) is at ^{56}Cr ($Z = 24$) [80] and increases in strength as Z decreases. Recently, mass spectrometry facilities have been able to

study this region with high precision mass measurements. These measurements are used to study the mass surface, where mass filters such as the empirical shell gap can also provide evidence of shell closures. These high precision mass measurements have agreed with the onset of the magicity of $N = 32$ shown in first $J^\pi = 2^+$ excited state energy ($E(2_1^+)$) measurements [81, 82] (see figure 1.4), as well as corroborating the absence of an $N = 32$ shell closure in V isotopes seen in spectroscopic data [83, 84] and mass measurements [85]. This feature of the $N = 32$ shell closure disappearing just below Cr in proton number show how fragile such shell closures can be. High precision mass measurements have also been used to show that the strength of the $N = 32$ shell closure appears to peak in Ca ($Z = 20$) [86].

As we travel more exotic and lower in proton number on the nuclear chart, a new shell closure at $N = 34$ emerges, as was first predicted in calculations by Beiner et. al. [87]. The existence of an neutron shell gap in Ca ($Z = 20$) isotopes at neutron number $N = 34$ was confirmed through with $E(2_1^+)$ measurements using proton knockout reactions [88]. However, with just 2 protons more, Ti was does not exhibit the same shell closure behaviour at $N = 34$ as Ca, resulting in some question of the robustness of the $N=34$ subshell closure [89]. Additionally, measurements in Sc using high precision mass spectrometry [8] and decay spectroscopy [90, 91] have also failed to see evidence of an $N = 34$ shell closure at $Z = 21$. In contrast $E(2_1^+)$ measurements in Ar ($Z = 18$) have shown the continued presence of an $N = 34$ closure [92], in line with with predictions [93].

The complete lack of spectroscopic for ^{53}K and precision mass data in K isotopes beyond $N = 33$ have left the robustness of the $N = 34$ shell closure in question. Using the mass measurements presented in the previous chapter, I provide the first experimental evidence in favour of a continued $N = 34$ subshell closure in K. The measurements are compared for agreement with *ab initio* calculations performed in

two different frameworks, as well as with different interaction models.

4.1 Evidence for an $N = 34$ Subshell Closure in Potassium

With mass measurement also being a direct measurement of the binding energy of nucleons, it can be a sensitive probe for shell effects in isotopic chains. However, shell gaps are not the only factor that contribute to changes in binding energy. In order to remove the mean field binding of nucleons, isolate the effect of shell closures, it is common to use nuclear masses to compute a quantity called the “empirical shell gap”. The empirical shell gap is the difference of two nucleon separation energies of isotopes or isotones 2 nucleons apart. The neutron empirical shell gap (also shown in eqn. 1.5) is given as

$$\Delta_{2n}(Z, N) = S_{2n}(Z, N) - S_{2n}(Z, N + 2) \quad (4.1)$$

$$= 2E_B(Z, N) - E_B(Z, N - 2) - E_B(Z, N + 2), \quad (4.2)$$

where the binding energy can be computed from the mass as given in eqn. 1.1. The proton empirical shell gap can similarly be computed from two proton separation energies varied over Z .

While the empirical shell gap is a commonly used mass filter for identifying shell closures, it can also be influenced by differences in pairing between valence protons and neutrons [94]. In these situations it is preferable to use the “empirical energy gap” [95]. The empirical energy gap is a 4 point formula, allowing it to account for pairing differences across a shell gap. For neutrons it is defined in terms of nuclear binding

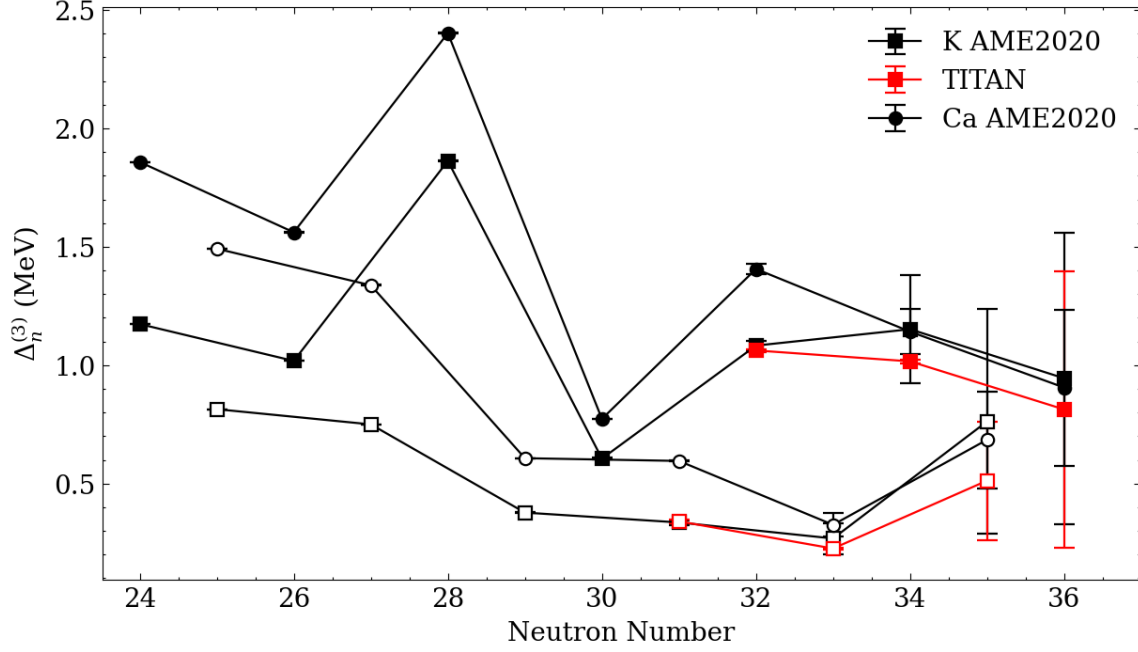


Figure 4.1: Comparison of the empirical pairing gaps in the K and Ca isotopic chains. Even neutron number pairing gaps have filled points, while odd neutron number data has empty points. Note the large difference in pairing between $N = 33$ and $N = 35$ in both isotopic chains.

energy as

$$\delta e_n = E_B(Z, N) + E_B(Z, N - 1) - E_B(Z, N - 2) - E_B(Z, N + 1). \quad (4.3)$$

Identifying areas where nuclear pairing changes quickly can also be done through mass filters. The neutron pairing gap can be estimated by a three point formula as

$$\Delta_3^n = \left(\frac{-1}{2} \right)^N (2E_B(Z, N) - E_B(Z, N - 1) - E_B(Z, N + 1)). \quad (4.4)$$

This three point mass filter is typically interpreted as the pairing gap in odd neutron number systems [95]. The pairing gap in ^{53}Ca and ^{55}Ca is known to be different due

to neutrons in the $\nu 2p_{3/2}$ and $\nu 2p_{1/2}$ orbitals having different coupling to the proton $\pi 1f_{7/2}$ orbital [96]. The $N = 34$ shell gap in Ca has therefore been studied using the empirical energy gap rather than the empirical shell gap [94]. As can be seen in figure 4.1, the pairing gap in K shows similar behaviour across $N = 34$. Therefore we also use the empirical energy gap rather than the empirical shell gap to measure the strength of the $N = 34$ shell closure in potassium. The empirical energy gap of even N potassium isotopes depicted in figure 4.2, shows the energy gap does not decrease after the $N = 32$ shell closure. In fact, we see evidence that the $N = 34$ energy gap may even be stronger than at $N = 32$. This would be in line with evidence from charge radii measurements that did not find evidence for a strong shell closure at $N = 32$ in ^{51}K [97].

The $N = 32$ and $N = 34$ shell gaps are thought to arise in neutron deficient nuclei from reduced tensor interaction between the proton $\pi 1f_{7/2}$ shell and the neutron $\nu 1f_{5/2}$ as the $\pi 1f_{7/2}$ shell becomes less populated [100]. This would manifest itself as an energy gap at neutron number $N = 32$ as the $\nu 1f_{5/2}$ reduces in binding relative to the $\nu 2p_{3/2}$ level. The reduced coupling between these f shells may even result in the normal ordering of the neutron $\nu 2p_{1/2}$ and $\nu 1f_{5/2}$ shells becoming inverted which would then be the origin of the $N = 34$ shell closure. Travelling down in proton number along the nuclear chart, the first element with no protons in the $\pi 1f_{7/2}$ shell is calcium ($Z = 20$), and heuristically, we would expect the magicity of $N = 32$ to increase up this point. High precision mass measurements have supported the $N = 32$ shell closure strength peaking in calcium [8], and the improved mass values here also support this conclusion (see figure 4.3). Similarly, we expect that the level inversion that would create an $N = 34$ shell closure would only appear in the most neutron deficient nuclei, which also appears to be supported by our results in figure 4.3. This level inversion occurring at $Z = 20$ and persisting in potassium would also explain

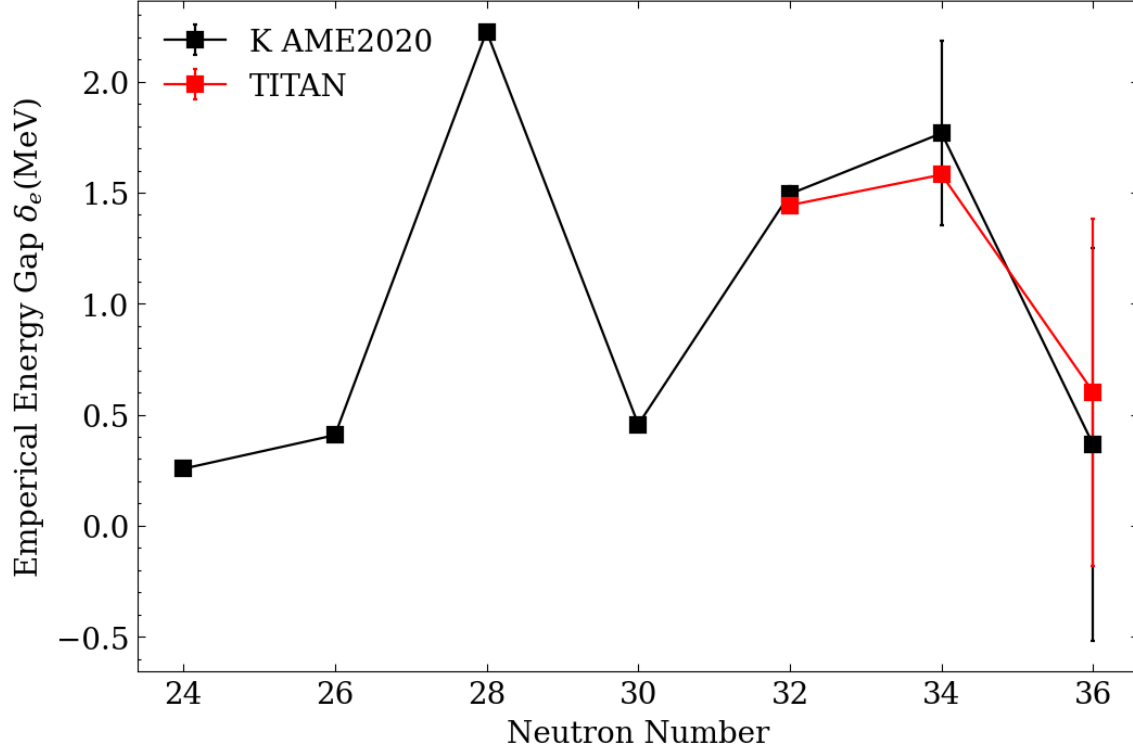


Figure 4.2: Plot of empirical energy gap in even N potassium isotopes. The $N = 34$ AME comparison is computed using an extrapolated mass for ^{54}K , resulting in large uncertainty. In the new TITAN result it is seen that the empirical energy gap at $N = 34$ is slightly larger than $N = 32$ and is approaching the strength of the canonical shell closure $N = 28$.

the consistency in behaviour of the pairing gap across $N = 34$ in these two isotopic chains.

The expected shell configuration of ^{54}K based on our mass measurements and expected structure in the region is depicted in figure 4.4. Based on this model we find it likely that the ground state spin of and parity (J^π) is likely to be $J^\pi = 4^-$ rather than the 2^- extrapolation reported in NUBASE2020 [75]. Such structure arguments show the power of nuclear mass measurements to aide in understanding nuclei in the most exotic regions of the nuclear chart, where current processes struggle to provide

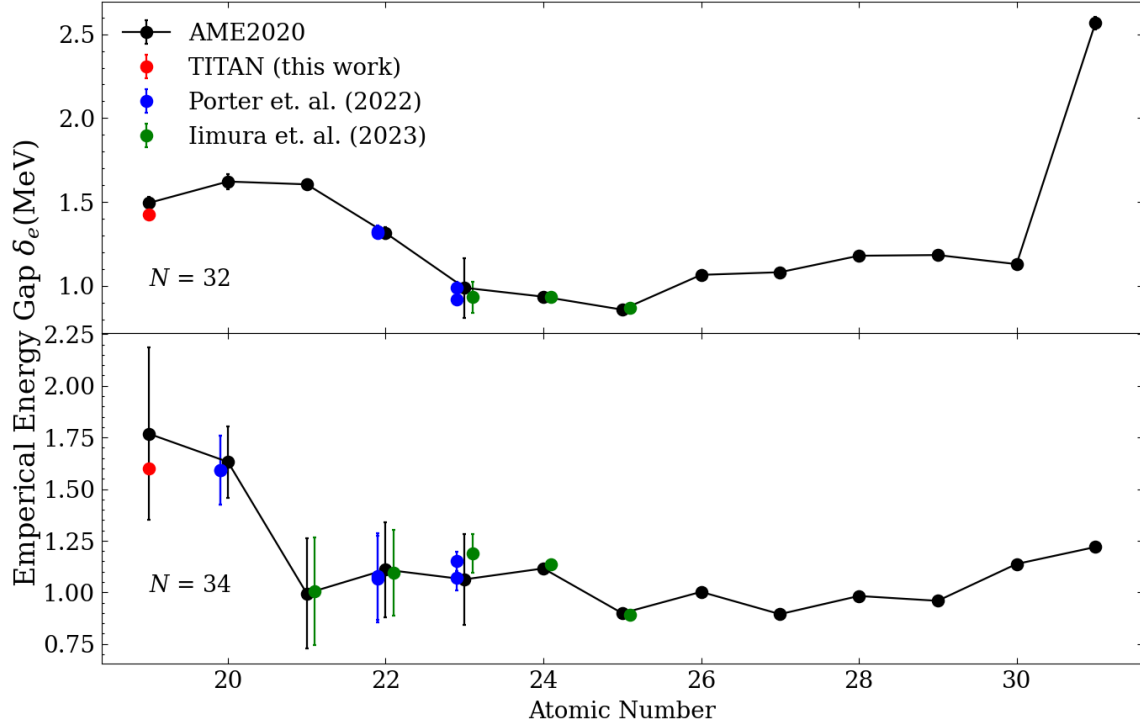


Figure 4.3: A comparison of empirical energy gap across $N = 32$ and $N = 34$ isotones. Values are computed with AME2020 masses [7], and work published after the cutoff date for the AME compilation [98, 99].

beams of sufficient intensity for nuclear spectroscopy.

4.2 Comparison of *ab initio* methods

Some *ab initio* methods are known to struggle in reproducing the total binding energy of nuclei [26]. Instead, an approach to benchmarking theory is to compare the trend of predicted masses to experiment, through first and second difference formulas like neutron separation energy and empirical shell gap.

The new and improved mass measurements in ^{54}K are compared to *ab initio* model predictions made using two frameworks. The first framework called valence-

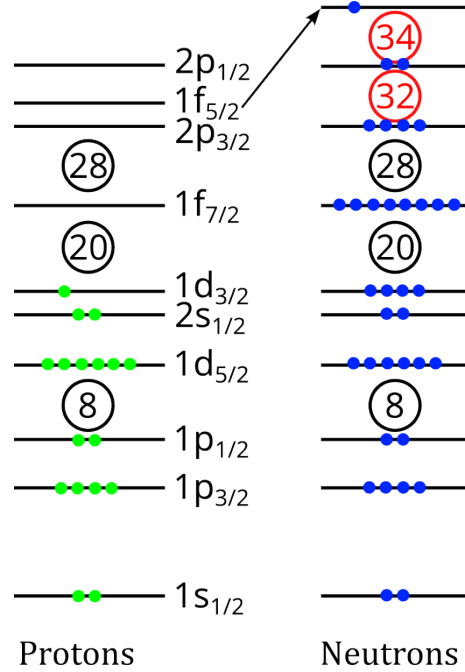


Figure 4.4: Schematic representation of nucleon shell filling for a plausible level scheme of ^{54}K . Note that the $1f_{5/2}$ level is shifted higher for the neutrons, resulting in an inversion from the normal ordering with the $2p_{1/2}$ subshell.

space in-medium similarity renormalization group (VS-IM-SRG) is computed using three different chiral effective field theory (χEFT) interactions. One of these interactions (NN+3N(lnl)) is also computed in the self-consistent Gorkov-Green's function (SCGF) framework. The calculations done in the VS-IM-SRG framework were performed by collaborators at TRIUMF and Technische Universität Darmstadt using Jülich Supercomputer resources and the SCGF calculation has been performed by collaborators at the Institute for Theoretical Physics at Université Paris-Saclay. Details of the computational frameworks and the nuclear interaction models are beyond the scope of this thesis. Only brief descriptions of each follow, with citations given to help the interested reader find more on the techniques.

4.2.1 VS-IM-SRG

The valence-space in-medium similarity renormalization group method is a technique for solving the quantum many body problem using a reference state (the core nucleus) and particle-hole excitations of that state [101]. The in-medium similarity renormalization group method is used to construct a unitary operator that drives the effective Hamiltonian towards a diagonal form under repeated operation on the Hamiltonian [102], making solving for the valence space states more computationally tractable. The VS-IM-SRG method requires that the valence space considered be built on a doubly closed reference state [34].

4.2.2 SCGF

The self-consistent Gorkov-Greens's function method does not require the symmetry allowed by a doubly closed reference state [34]. It uses approximations to the one body Green's functions to describe particle-hole excitations in the many body system, with the Green's function approximation iteratively fed into an equation which generates the perturbations of the Green's functions, allowing a refined approximation to be generated [103]. This formalism at present only allows for single odd nucleon calculations and as a result only binding energies for even A potassium isotopes.

4.2.3 χ EFT Interactions

The interactions used to generate these calculations are all derived from χ EFT. These interactions inherently obey the symmetries of quantum chromodynamics (QCD). All interactions were fit to nuclear data on 2, 3 and 4 nucleon systems, and evolved using similarity renormalization group methods [104, 105]. The interactions used are listed

below:

NN+3N(lnl) Two nucleon and chiral three nucleon interactions with local and non-local interactions [106]. The local and non-local interactions refer to interactions that explicitly depend on position and interactions that do not explicitly depend on position (such as interactions that depend on momentum).

Δ NNLO_{GO}(394) An interaction based on pion and $\Delta(1232)$ interactions, fit to reproduce the binding energy of ${}^4\text{He}$ [107, 108].

1.8/2.0 (EM) An interaction with two nucleon interactions cut-off at momentum scale 1.8 fm^{-1} and three nucleon interactions cut-off at 2.0 fm^{-1} [109].

The calculations using SCGF were performed in a harmonic oscillator basis with frequency $\hbar\omega = 18 \text{ MeV}$, and one body state basis size $e_{\text{max}} \equiv \max(2n + l) = 13$, two body basis state size $e_{2\text{max}} = 2e_{\text{max}} = 26$, three body basis state size $e_{3\text{max}} = 16$ and $e_3 < 3e_{\text{max}}$, and self-energy computed in the algebraic diagrammatic construction (ADC) truncated at second order (ADC(2)). The results of the SCGF calculations were previously reported in [34]. All of the calculations done in the VS-IM-SRG framework were performed with $\hbar\omega = 16 \text{ MeV}$, $e_{\text{max}} = 14$ and $e_{3\text{max}} = 22$ and using an $\pi sd - \nu pf$ shells valence space (i.e. an ${}^{26}\text{O}$ reference state).

4.2.4 Results

One common way of measuring relative performance of *ab initio* theory predictions is to compare average root mean square (r.m.s) deviation of some observable predicted by the theories. The SCGF method employed method is known to under-predict the degree of binding with ADC truncated to 2nd order [34], so instead the average r.m.s deviation was determined on experimentally measured S_{2n} and Δ_{2n} in the potassium

4.2. Comparison of *ab initio* methods

Interaction (model)	Even N S_{2n} r.m.s error	Even N Δ_{2n} r.m.s error	S_{2n} r.m.s error	Δ_{2n} r.m.s error
NN+3N(lnl) (VS-IM-SRG)	1.04 MeV	1.04 MeV	1.12 MeV	1.04 MeV
1.8/2.0(EM)	0.51 MeV	0.71 MeV	0.57 MeV	0.67 MeV
Δ NNLO _{GO} (394)	1.20 MeV	1.48 MeV	0.97 MeV	1.22 MeV
NN+3N(lnl) SCGF	2.02 MeV	0.87 MeV	—	—

Table 4.1: Average root mean square deviation of calculations of S_{2n} and Δ_{2n} . Deviation is calculated with new TITAN mass measurements and AME2020 masses on potassium isotopes from $N = 22$ to $N = 35$.

isotopic chain. Where the TITAN measurements improve the uncertainty on these quantities, the TITAN values are used to get the r.m.s error. To make an equal comparison across both methods we first compare only in even N potassium nuclei from $N = 22$ to $N = 34$. To compare interactions we use all potassium isotopes in the range $N = 22$ to $N = 35$ to compare the calculations from VS-IM-SRG. These results are summarized in table 4.1 and the calculated results for S_{2n} and Δ_{2n} are shown in figures 4.5 and 4.6 respectively.

It is seen that the 1.8/2.0 (EM) interaction gives the best performance over this isotopic chain. It is interesting to note that that the NN+3N(lnl) interaction performs substantially better in computing the empirical shell gap than in two neutron separation energy particularly in the SCGF framework. This may be related to the interaction missing some mean-field like contribution while still capturing the nucleon interactions that determine the exact location of shell features when using the ADC(2) truncation, as briefly mentioned in [34].

As a final note, we notice that predictions in the VS-IM-SRG framework all qualitatively predict a strong $N = 34$ shell closure signature in the empirical energy

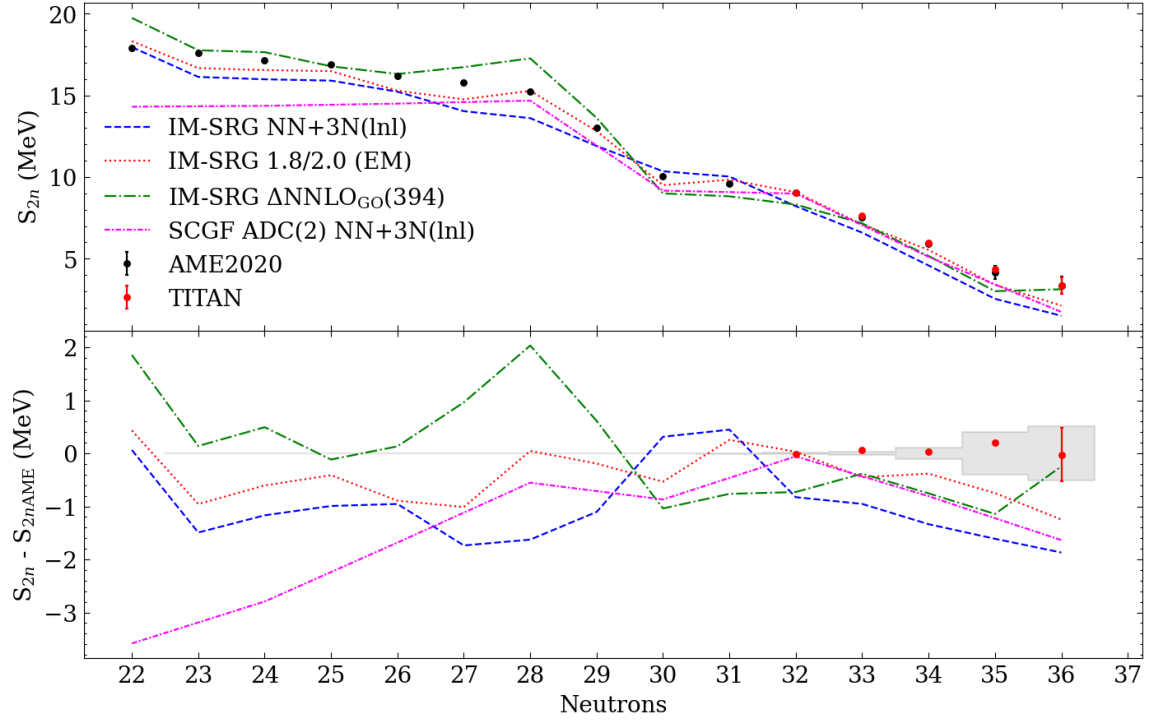


Figure 4.5: Top: Two neutron separation energy determined from masses in the AME2020 compilation [7] and this experiment and calculated in a range of *ab initio* theories. Bottom: Difference to AME2020 for the same S_{2N} estimations.

4.2. Comparison of *ab initio* methods

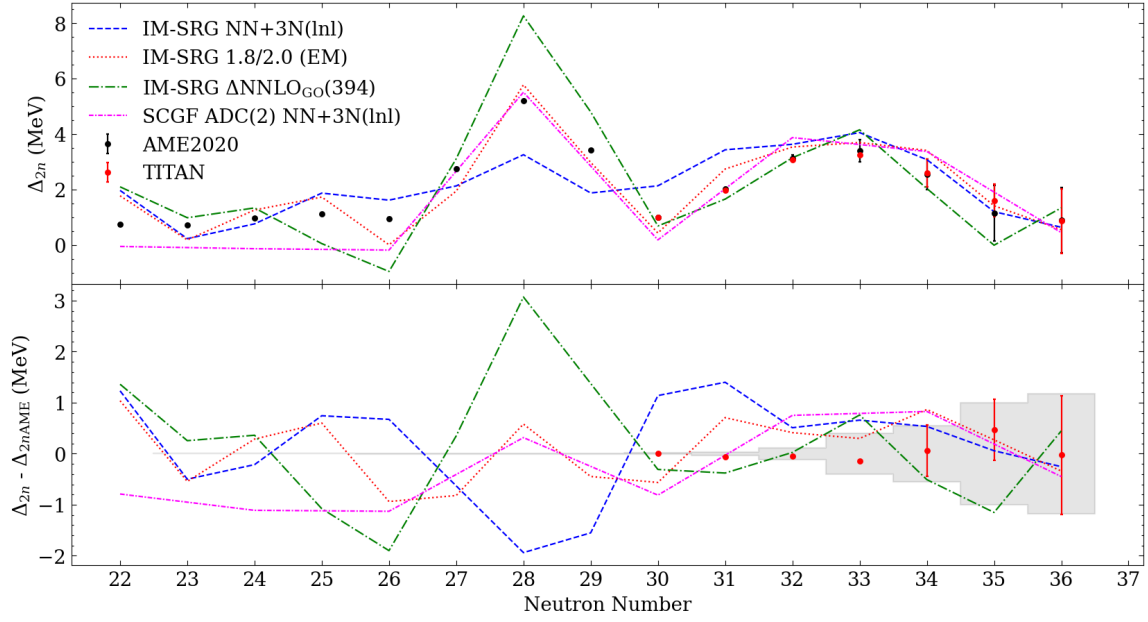


Figure 4.6: Top: Δ_{2n} as calculated from AME2020 masses and from the current work. *Ab initio* theory calculations are shown. Bottom: Difference in Δ_{2n} from AME2020 calculations.

4.2. Comparison of *ab initio* methods

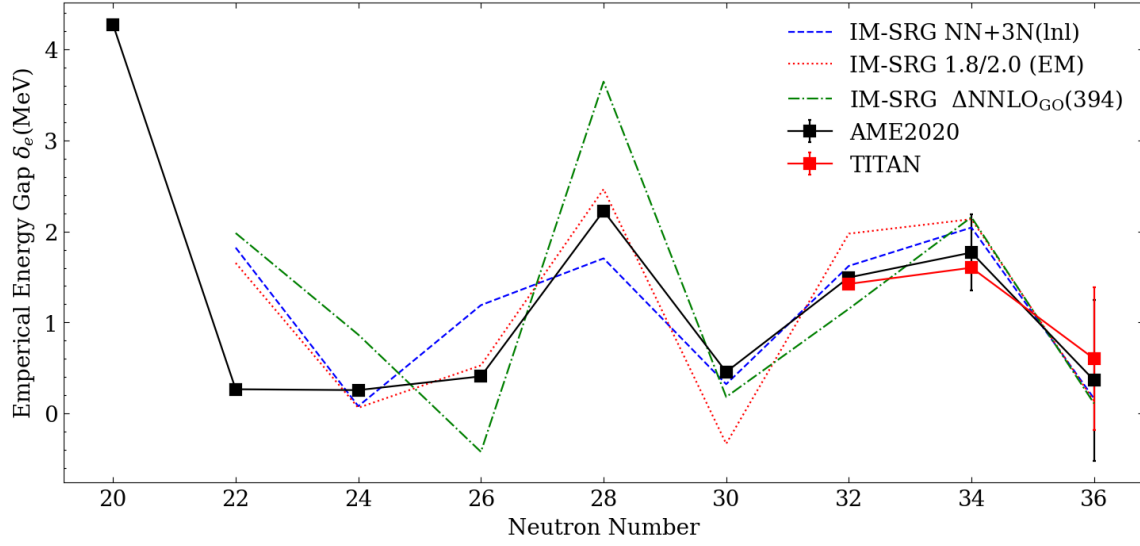


Figure 4.7: δ_{2n} with VS-IM-SRG *ab initio* calculations. Notice that all interactions tested predict an $N = 34$ empirical energy gap around the same strength as the $N = 32$ empirical energy gap.

gap. This is seen in figure 4.7. Here, we compare only the VS-IM-SRG predictions as the SCGF framework does not provide the needed binding energies in odd neutron systems to calculate empirical energy gap for the odd proton number element potassium ($Z = 19$).

CHAPTER 5

Not Just High-Precision Mass Measurement: Half-Life Measurement with TITAN's MR-TOF-MS

Half-life is a fundamental property of radioactive nuclei. The half-lives of isotopes are of interest for radiation hazard mitigation, calculating elemental abundances resulting from stellar processes [110], determining nuclear structure [111], and even precision standard model tests [112]. In the context of mass measurements, the ability to simultaneously measure the half-life of an isotope while performing a mass measurement allows for more confidence in species identification. In cases where the half-life is poorly known the combination of a mass measurement and half-life measurement can increase the science impact, without adding to the radioactive beam time requirements. Therefore, several new methods for determining half-life while performing a high-precision mass measurement with the TITAN MR-TOF-MS were

developed.

In this chapter, two new half-life measurement techniques will be discussed. The first, dubbed “mass tagged β -decay” or simply “ β -tagging”, uses the time between subsequent ion detections to search for signals from the ion detector of a later implanted ion decay. The later decay can then be uniquely associated to an ion detection and therefore a mass. By use of cuts on the β -decays based on the mass of the ions, the half-life of an ion can be determined. A measurement using this technique on ^{54}K will be shown. This measurement provided additional confirmation of our assignment of the mass peak to potassium, and resulted in an improved half-life estimate. It also represents the first experimental constraints on the β -delayed neutron branching ratios in ^{54}K .

The second technique leverages the re-trapping capabilities of the TITAN MR-TOF-MS system, similar to how Penning trap mass spectrometry has been used to purify RIB for decay experiments [113]. Re-trapping is used to remove contamination in order to build a source of a particular isotope with much higher purity than would otherwise be possible. The detection of the subsequent decays of the pure source on the detector is then done without additional ions being deposited on the detector. Examples of measurements of this nature, with small variations to get the best performance for different ion rates and half-lives, are demonstrated with $^{180-182}\text{Yb}$. Evidence supporting the decay of ^{180}Yb into a metastable state of ^{180}Lu is also presented.

5.1 Mass Tagged β -Decays

The TITAN MR-TOF-MS system is typically operated with complete cycle repetition rates of 30 to 80 Hz. The cycle includes capturing the bunched beam from the TITAN

RFQ and all ion manipulations up to the final time of flight detection. During each of these cycles, the electrostatic mirror confining the ions to the analyzer is only lowered for about 100 μ s. Furthermore, the ions in the analyzer will hit the MagneTOF detector [58] within 10-20 μ s of the mirror opening, depending on the ions mass and position in the analyzer at the moment of mirror opening. This means that there is up to ~ 33 ms during each measurement cycle in which no ions are impinging on the detector. It is with this time during each measurement cycle that we look for additional signals generated from the MagneTOF detector, which we can attribute to radioactive decays of previously implanted ions. A rough schematic showing a decay on the detector a while the following ion bunch is being prepared is shown in figure 5.1. The mass and decay data collected are also represented.

Multiple approaches to associating decay data to the mass data can be employed. One way is to uniquely associate each β -decay with the most recent ion observed in the time-of-flight measurement. In order to ensure a high likelihood of correctly assigning the β -decay, the ion rate must be low compared to the specific activity of the ion of interest. This is often easily achieved with suppression of contaminant ions with re-trapping. Additionally, ions that can be confidently identified as stable or nearly stable species can be excluded from the maximum ion rate calculation, as they can be excluded from matching with radioactive decays in the final analysis. This approach of associating each detected decay with an implanted ion, up to some maximum correlation time is described in [114, 115], where specialized detectors were used to measure α -decays and β -decays associated with an ion implantation.

The probability density of an atom with half-life $t_{1/2}$ decaying at a given time

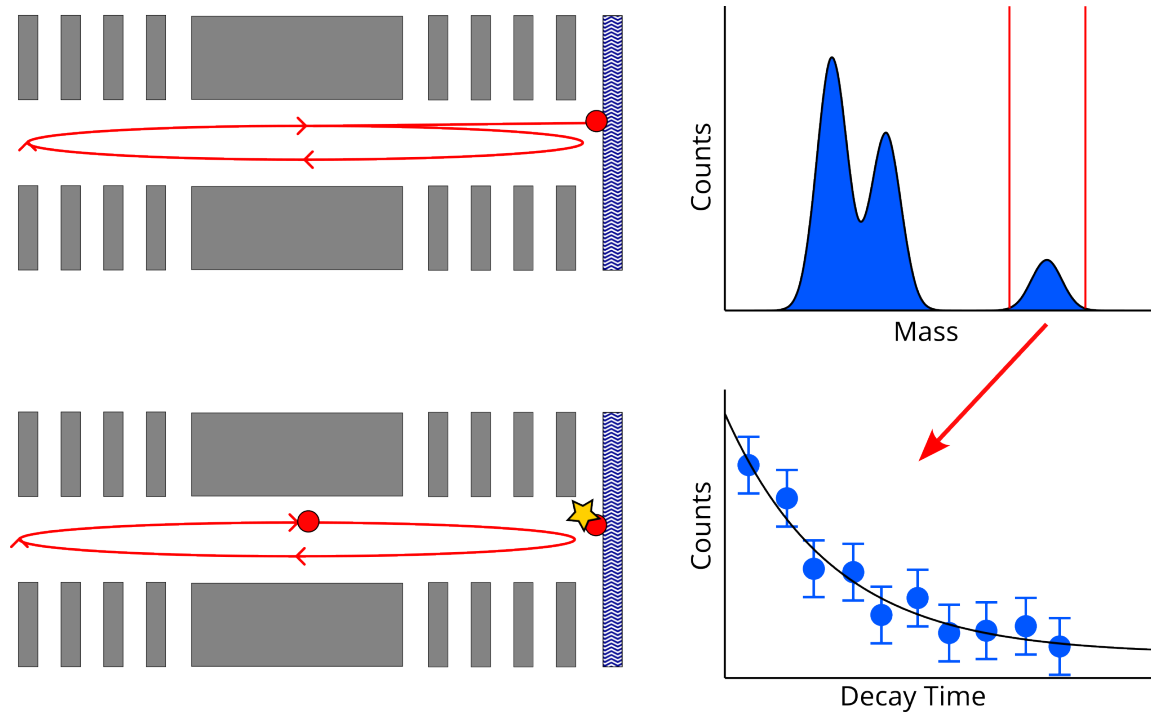


Figure 5.1: A diagram showing the decay of implanted ions (yellow star) on the detector and the flight of the ion bunches (red). A β -decay is assigned when a detector signal occurs while the mass analyzer mirror is reflecting.

after implantation can be written as:

$$p_{\text{decay}}(t) = \lambda e^{-\lambda t} = \frac{\ln 2}{t_{1/2}} e^{-\frac{\ln 2}{t_{1/2}} t}, \quad (5.1)$$

where λ is the decay rate and is related to the half-life by $\lambda = \ln(2)/t_{1/2}$.

Since the detection efficiency of the ion detector is only 50%, there is a chance that the decay of the ^{54}K isotope is missed and the subsequent daughter decay of ^{54}Ca is instead detected. If these species are the only nuclides decaying, along with a the constant background on the detector from accumulated radioactive species, the

5.2. Estimation of ^{54}K β_n Branching Ratios

following function would describe the decay curve:

$$p_{\text{decay}}(t) = \lambda_{(^{54}\text{K})} e^{-\lambda_{(^{54}\text{K})}t} + \frac{1}{2} \lambda_{(^{54}\text{Ca})} \left(1 - e^{-\lambda_{(^{54}\text{K})}t}\right) e^{-\lambda_{(^{54}\text{Ca})}t} + \lambda_{\text{back}} e^{-\lambda_{\text{back}}t}. \quad (5.2)$$

Here, the half-life of ^{54}Ca ($t_{\frac{1}{2}\text{Ca}}$) is 90 ms, giving an decay rate of $\lambda_{(^{54}\text{Ca})} = 7.7$ counts per second [75]. The background rate λ_{back} is 0.266 ± 0.005 counts per second and was determined from the total detected rate of uncorrelated β -decays. However, this curve does not fit the acquired decay data, due to the tendency of very neutron-rich nuclides to undergo β -delayed neutron emission. An estimation of the β -delayed neutron emission probabilities of ^{54}K is presented below.

5.2 Estimation of ^{54}K β_n Branching Ratios

When a nucleus undergoes β -decay the resulting daughter nucleus will in general not be in its ground state. If the energy of the excited state in the intermediate nucleus is sufficiently high, the nucleus may be unbound to direct release of one or more nucleons [116, 117]. This is known as β -delayed neutron or β -delayed proton decay. In general, isotopes further from nuclear stability are more likely to undergo β -delayed nucleon decay because the required energy to remove a nucleon is less. This tendency towards nucleon release from the daughter nuclei is described in terms of branching ratios. Predictions of β -delayed neutron (P_{1n}) and β -delayed two neutron (P_{2n}) decay probability suggest that in ^{54}K , very few of the the daughter nuclei will be ^{54}Ca [111]. However the β -decay branching ratios have never been experimentally determined for ^{54}K .

The half-lives of all three likely Ca daughters nuclei ($^{52-54}\text{Ca}$) of ^{54}K have previously been experimentally determined. These nuclei have substantially different lifetimes

5.2. Estimation of ^{54}K β_n Branching Ratios

Isotope	$t_{1/2}$ (S)	β_n Probability (%)	β_{2n} Probability (%)
^{54}Ca	0.090(6)	7#	0#
^{54}Sc	0.526(15)	*16(9)	—
^{54}Ti	2.1(1.0)	—	—
^{53}Ca	0.461(90)	40(10)	—
^{53}Sc	2.4(0.6)	*0.2#	—
^{52}Ca	4.6(0.3)	*0 (<2)	—
^{52}Sc	8.2(0.2)	*4#	—

Table 5.1: Daughter and granddaughter nuclides of ^{54}K considered in the fit to determine the ^{54}K branching ratios. Values are the recommended values from NUBASE2020 [75]. When a decay channel is to an isotope that is too long lived to be relevant, the branching ratio to that channel is marked by *. Values based on extrapolation are marked by #. Decays that are energetically forbidden are left blank.

from each other making it possible to estimate the branching ratios into each daughter solely from the mass correlated β -decay timings. The longest lived species of these is ^{52}Ca with a half-life of 4.6 ± 0.3 s. This results in a specific activity of $\lambda_{^{52}\text{Ca}} \simeq \ln(2)/4.6\text{s} = 0.15$ cps. This is close in magnitude to the background rate on the detector of 0.266 ± 0.005 cps, and so the background must not be ignored. Additionally, some Sc and Ti isotopes arising from daughter and granddaughter decay must be included in the final fit model. The decay properties of all relevant nuclides in the decay chain of ^{54}K are summarized in table 5.1.

The branching ratios of ^{54}K decay into one and two β -delayed neutron channels was estimated using an unbinned fit with the iminuit package [118, 119]. The MINOS algorithm was used to generate asymmetric fitting errors. All non-time-of-flight signals following the implantation of a ^{54}K ion up to a maximum cutoff time of 10 s were used. The fit was performed with the half-life of ^{54}K freely variable, and the β_n and β_{2n} ratios constrained such that they could individually vary between 0 and 1 and their sum remain less than or equal to 1. The parameters of all daughter decay nuclides

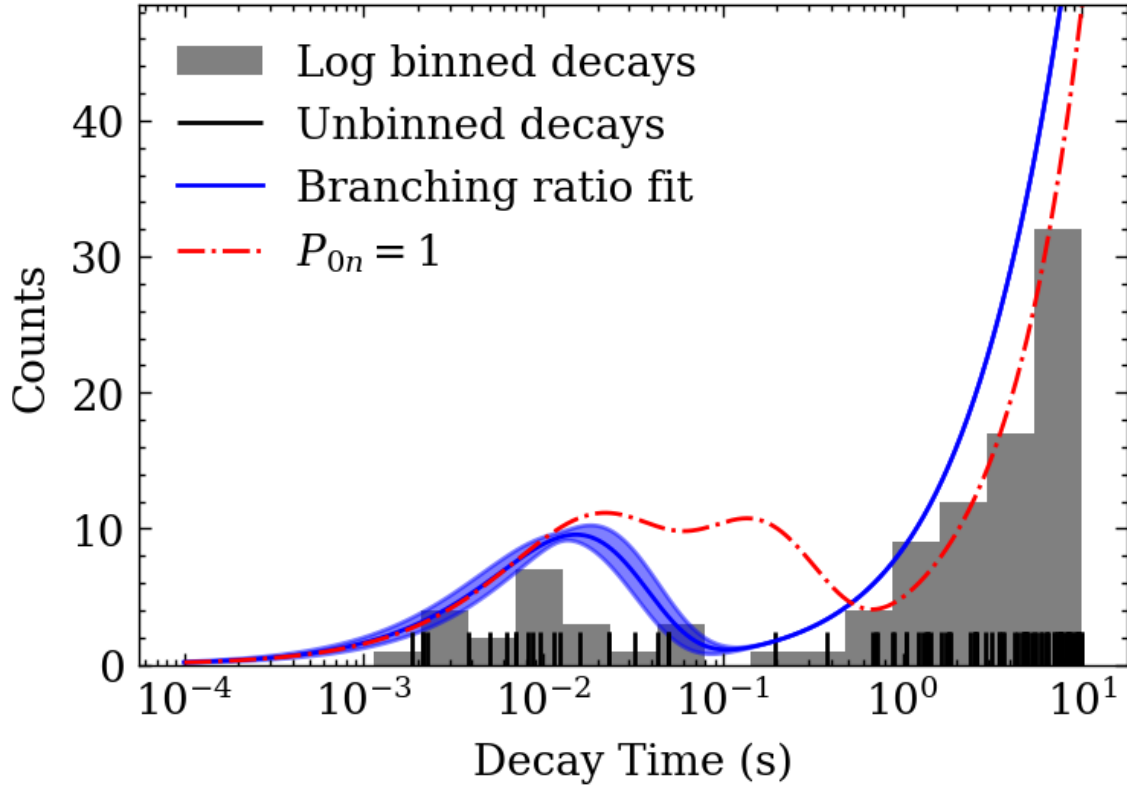


Figure 5.2: A fit with a bootstrapping-derived error band [122] of the β -decay signals associated with the implantation of ^{54}K , up to a maximum decay time of 10 s (arbitrary normalization). Binned and unbinned decay timings are shown. For reference, a decay curve assuming $P_{0n} = 1$ is shown as well.

were fixed at their NUBASE2020 [75] reported values. The model fit suggested that very little of the decaying ^{54}K was later decaying as ^{54}Ca ($P_{0n} = 0_{-0.0}^{+0.09}$). Branching into the neutron emission channels was estimated as $P_{1n} = 0_{-0}^{+0.12}$ and $P_{2n} = 1_{-0.12}^{+0}$. Consistency and numerical stability of the fit was checked by cutting off the decay data at 2 s, giving largely the same result. The data and fit are shown in figure 5.2, adjusted for display on a logarithmic time axis [120, 121].

5.3 Half-life of ^{54}K

With the branching ratio into the β_{0n} decay shown to be small, a half-life estimate of ^{54}K can be done without considering daughter and background decay events. This is done by taking only the first detection of a β -decay event after the implantation of a ^{54}K ion. Due to the 50% detection efficiency of the β -decays (section 5.6), there is a reasonably large chance that the first detection will be of a subsequent daughter decay, or the low level background. This risk of spurious decay correlations is minimized by only considering decays that occur within 100 ms of ion implantation. The maximum decay time cut-off corresponds to ~ 10 half-lives of ^{54}K and introduces minimal bias to the measurement.

The mass cuts used to identify a time-of-flight ion as ^{54}K are shown in figure 5.3. Since the β -decay timings assigned to ^{54}K are nearly background free, the maximum likelihood estimator of the mean life-time is simply the arithmetic mean of the β -decay times. Using the method described in [120] to calculate the 1σ confidence interval, the half-life of ^{54}K is determined as $10.0^{+3.2}_{-2.0}$ ms. A comparison of the probability density of the previous measurement [123] and this new value, along with the unbinned decay times is shown in figure 5.4. The trend of half-lives in the region is shown in figure 5.5.

5.4 β -tagging to Reduce Stable Contaminant Background

Many of the most interesting atomic masses to measure are on nuclides far from stability on the nuclear chart. These interesting nuclides are typically less produced

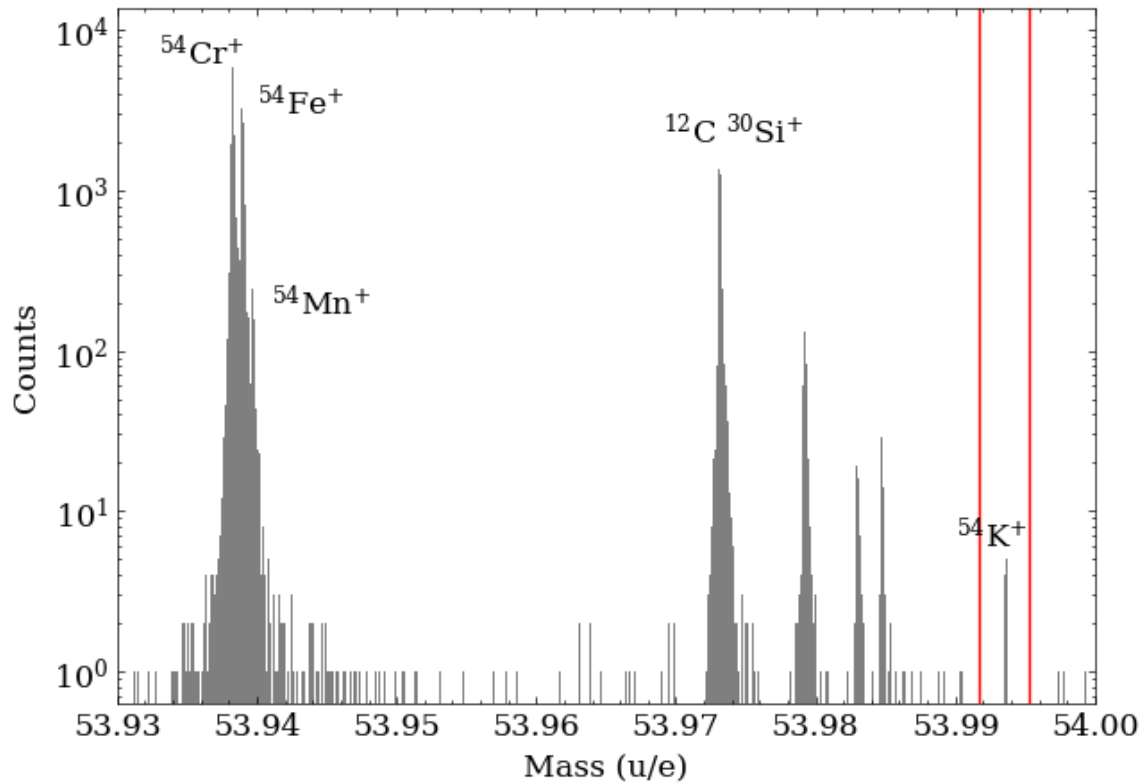


Figure 5.3: Time-of-flight spectrum with peaks labelled by ion species. The red vertical lines indicate the cut applied to attribute later β -decays to ^{54}K .

than their more stable counterparts in the reactions used in RIB facilities. ISOL facilities are especially disadvantaged when the half-lives of these nuclides is short, as the ISOL method requires time for the nuclides to be liberated from the target. The produced beam will then have more relative stable contamination.

In cases such as these it can be advantageous to reverse the idea of tagging time-of-flight ions with β -decays. If we instead tag each detected β -decay with a previous time-of-flight ion, we can discard the untagged time-of-flight ions. This will greatly reduce the background of stable and long-lived ion species in the resulting mass spectrum. The resulting mass spectra can be compared to aid the experimenter

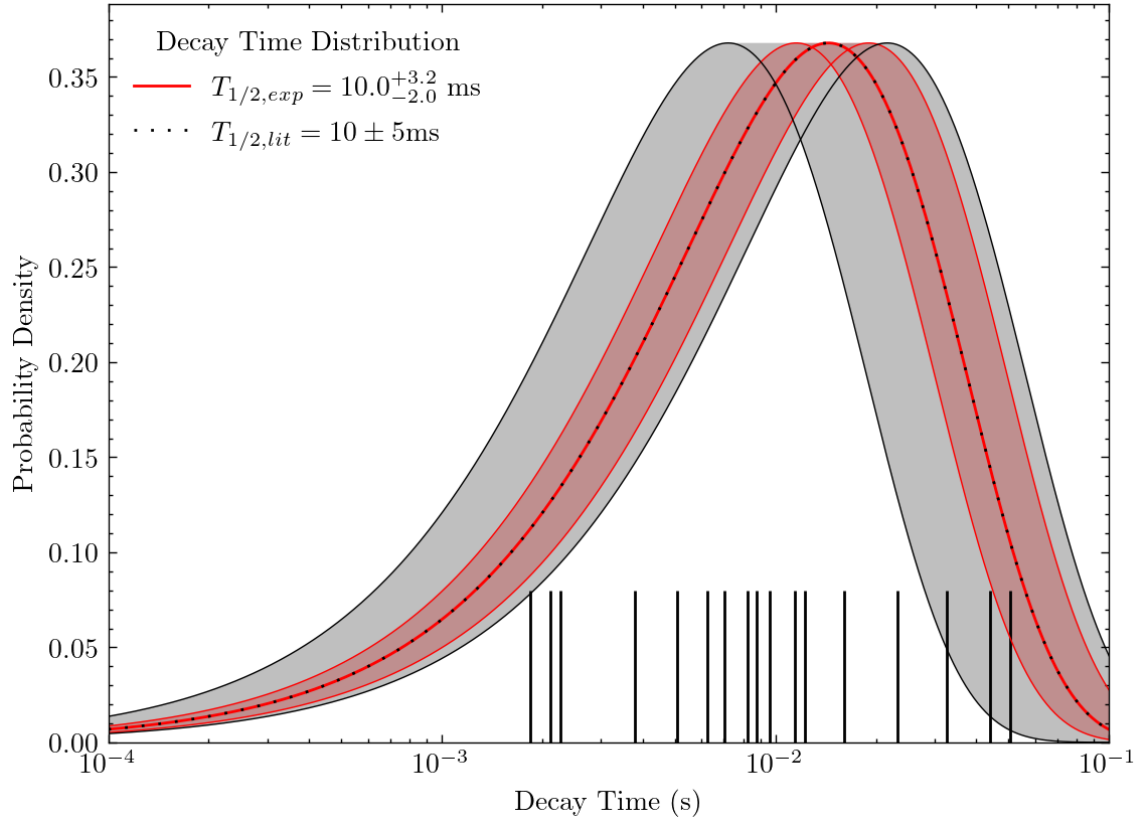


Figure 5.4: β -decays correlated with an ion detection of ^{54}K . The solid red line shows the best fit probability density function with thinner lines representing the extremes of the 1σ half-life uncertainty. A similar plot is shown in black for the previous literature value [123].

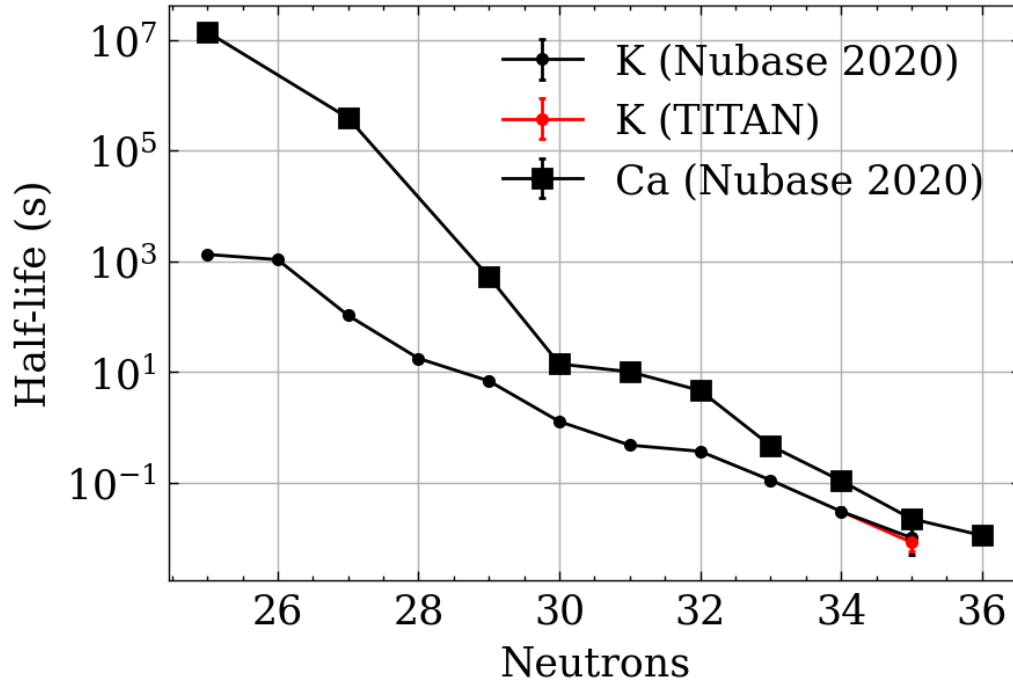


Figure 5.5: Half-lives of nuclides around $Z=20$. The new value for ^{54}K performed at TITAN is shown in red, and values for Ca and K are taken from [75]. Changes in the half-life trend can be seen around the $N=32$ subshell closure.

5.4. β -tagging to Reduce Stable Contaminant Background

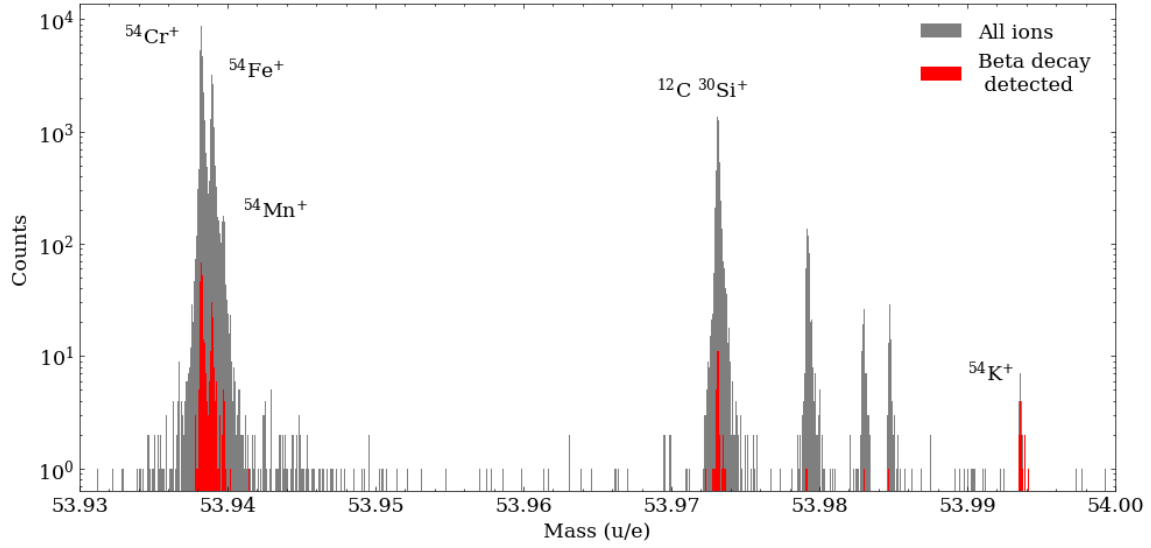


Figure 5.6: $A/Q = 54$ mass data, taken with the MR-TOF-MS set for mass selective β -tagging. In grey is all time-of-flight ions, and in red is time-of-flight ions with an associated β -decay. Stable ion peaks are suppressed by ~ 2 orders of magnitude in the β -tagged spectrum, while the $^{54}\text{K}^+$ peak is only suppressed by a factor 2.

in identifying the species in the beam. In some cases there may be substantial overlap of a radioactive isotope species with a stable contaminant close in mass. In this case, fitting the spectrum of the β -decay tagged mass data could result in much improved mass uncertainty. An example of a β -tagged mass spectrum is shown in figure 5.6. As the half-life of ^{54}K is very short, and the background activity was low, this procedure resulted in stable contamination being reduced by a factor of ~ 100 in the β -correlated mass spectrum, while the ^{54}K rate was reduced by a factor 2. In an extreme case of a background limited mass measurement, this would increase the maximum theoretically tolerable signal-to-noise ratio of the TITAN MR-TOF-MS to over 10^9 .

5.5 Decay of a Re-trapping Purified Radioactive Source

The high suppression of mass selective re-trapping in the TITAN MR-TOF-MS ($\sim 10^4$), allows for the removal of much of the isobaric contamination that is often present in ISOL beams. When dealing with high rates (approximately 100 counts per second or higher) of a radioactive species whose half-lives are significantly longer than the cycle time of the MR-TOF-MS, re-trapping can be utilized to create an extremely pure source of the radioactive species. The ion beam can then be blocked and the purified source implanted on the MagneTOF detector produces decays according to the lifetime of the species and the daughter product.

A slight complication to the fitting of the resulting decays arises from the fact that any daughter products of the isotope being implanted will also accumulate while producing the purified source. This is handled by including an initial daughter activity in the fit of the decay curve. The daughter activity can be bounded based on the detected ion rate while building the purified source and the length of time that ions were implanted for. It can be safely assumed that the daughter nuclei remain implanted on the ion detector in the case of β -decay, as the nucleus is much heavier than the electron and neutrino produced, which will necessarily carry most of the kinetic energy. This assumption is directly tested with data taken from an implantation of ^{63}Fe , in section 5.6.

Half-life measurements were performed on $^{180-182}\text{Yb}$. Due to the long half-life of ^{180}Yb it was possible to build a source of ions with re-trapping and then simply block the beam and begin measuring decays on the detector. Re-trapping was set up on Yb with a relatively low rate of ions being sent to TITAN by the TRIUMF ISAC facility.

5.5. Decay of a Re-trapping Purified Radioactive Source

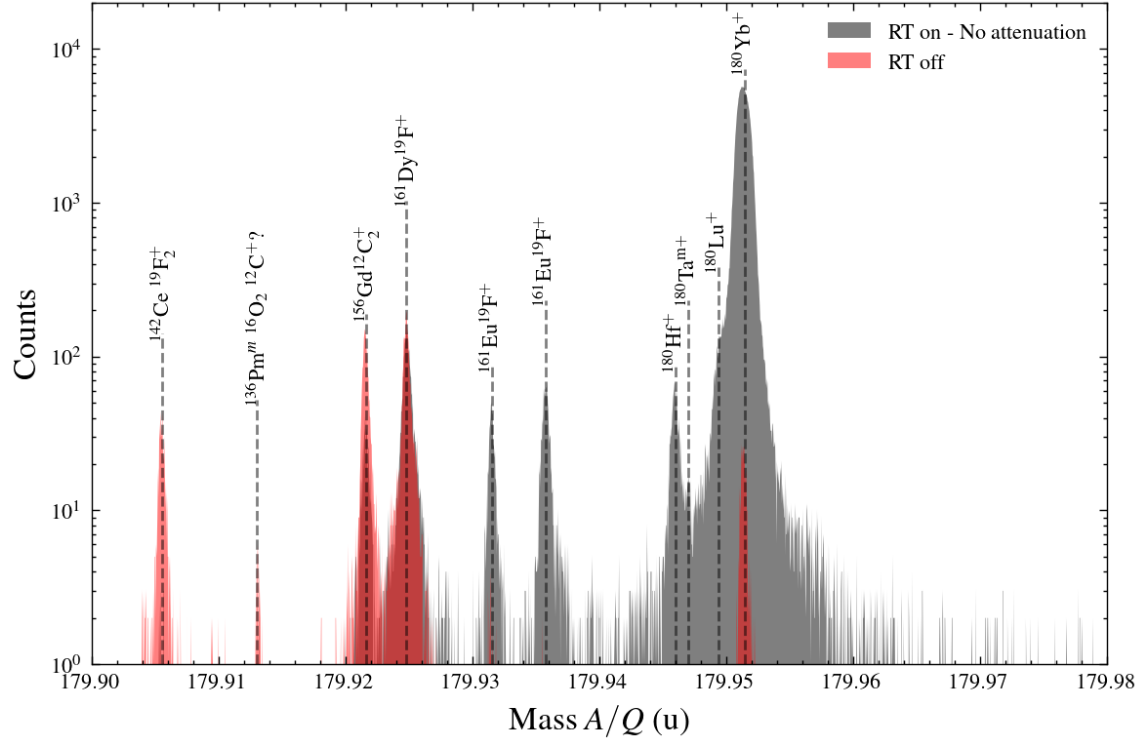


Figure 5.7: Comparison of the rate of ^{180}Yb and isobaric contaminants with re-trapping on and off. The attenuation of the ISOL beam to TITAN was removed between measurements resulting in a much higher rate of ^{180}Yb .

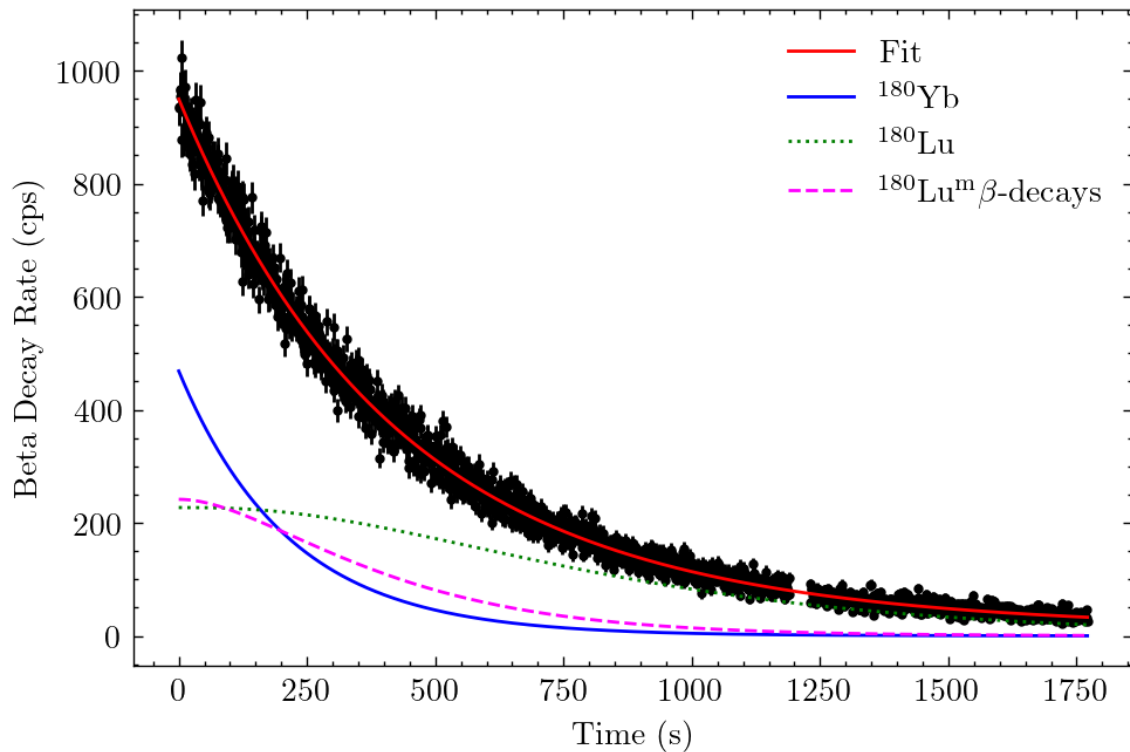


Figure 5.8: Measurement and best fit of ^{180}Yb β -decay. The contributions of β -decay of $^{180}\text{Lu}^{\text{m}}$, and $^{180}\text{Lu}^{\text{g.s.}}$ are also shown.

The ion rate was then increased by removal of an 100 times attenuator. The beam was checked during implantation with the normal operating mode to ensure that Yb was the dominant species being implanted on the detector. A comparison of the beam before and after re-trapping was enabled is seen in figure 5.7. Due to the high rate of ^{180}Yb ions, there is substantial dead time. This figure therefore provides only a lower bound on the relative rate of Yb to the isobaric contaminants. The Time-to-Digital converter (TDC) was then set to measure continuously, and the beam blocked. The resulting decay curve can be seen in figure 5.8.

It was not possible to obtain a good fit of the decay curve without the inclusion of a branching factor for the ^{180}Yb decay into an isomeric state of ^{180}Lu , even if

the initial activities of each were freely variable. This remained the case when allowing the well known ^{180}Lu half-life to vary by as much as 3 times the literature uncertainty. The presence of substantial amounts of radioactive contaminants can be excluded based on the mass spectra taken during the building of the ^{180}Yb source on the detector. We therefore conclude that the ^{180}Yb must have some decay into an isomeric state in ^{180}Lu . This isomeric state must either β -decay directly into ^{180}Hf , or have a reasonably long life-time in order to explain the observed small contribution of $^{180}\text{Lu}^{\text{g.s.}}$ in the decay curve. The final decay model included a branching ratio for Yb decay into an isomeric state ($b_{\text{Yb} \rightarrow \text{Lu}^{\text{m}}}$), a branching ratio for β -decay of the isomeric state ($b_{\text{Lu}^{\text{m}} \rightarrow \text{Hf}}$), and a half-life for the isomer ($t_{1/2\text{Lu}^{\text{m}}} = \ln(2)/\lambda_{\text{Lu}^{\text{m}}}$). The resulting differential equations governing the decay curve are then:

$$\frac{dN_{\text{Yb}}}{dt} = -N_{\text{Yb}}\lambda_{\text{Yb}} \quad (5.3)$$

$$\frac{dN_{\text{Lu}^{\text{m}}}}{dt} = N_{\text{Yb}}\lambda_{\text{Yb}}b_{\text{Yb} \rightarrow \text{Lu}^{\text{m}}} - N_{\text{Lu}^{\text{m}}}\lambda_{\text{Lu}^{\text{m}}} \quad (5.4)$$

$$\frac{dN_{\text{Lu}^{\text{g.s.}}}}{dt} = N_{\text{Yb}}\lambda_{\text{Yb}}(1 - b_{\text{Yb} \rightarrow \text{Lu}^{\text{m}}}) + N_{\text{Lu}^{\text{m}}}\lambda_{\text{Lu}^{\text{m}}}(1 - b_{\text{Lu}^{\text{m}} \rightarrow \text{Hf}}) - N_{\text{Lu}^{\text{g.s.}}}\lambda_{\text{Lu}^{\text{g.s.}}} . \quad (5.5)$$

The source was built over a period of ~ 30 minutes. The ^{180}Yb rate was continuously monitored and found to be constant. This long source building allowed all of the relevant nuclides to reach equilibrium. The initial amounts of the daughter nuclides of ^{180}Yb relative to the initial Yb ($N_{\text{Yb}}(0)$) are then:

$$N_{\text{Lu}^{\text{m}}}(0) = N_{\text{Yb}}(0) \frac{\lambda_{\text{Yb}}}{\lambda_{\text{Lu}^{\text{m}}}} b_{\text{Yb} \rightarrow \text{Lu}^{\text{m}}} \quad (5.6)$$

$$N_{\text{Lu}^{\text{g.s.}}}(0) = N_{\text{Yb}}(0) \frac{\lambda_{\text{Yb}}}{\lambda_{\text{Lu}^{\text{g.s.}}}} (1 - b_{\text{Yb} \rightarrow \text{Lu}^{\text{m}}} b_{\text{Lu}^{\text{m}} \rightarrow \text{Hf}}) , \quad (5.7)$$

and the resulting β -activities are

$$A_{\text{Yb}} = N_{\text{Yb}}\lambda_{\text{Yb}} \quad (5.8)$$

$$A_{\text{Lu}^{\text{m}}} = N_{\text{Lu}^{\text{m}}}\lambda_{\text{Lu}^{\text{m}}}b_{\text{Lu}^{\text{m}}\rightarrow\text{Hf}} \quad (5.9)$$

$$A_{\text{Lu}^{\text{g.s.}}} = N_{\text{Lu}^{\text{g.s.}}}\lambda_{\text{Lu}^{\text{g.s.}}} . \quad (5.10)$$

The model was fit with and without decay into an isomeric state of ^{180}Lu . A likelihood ratio test, as described in appendix C, with the null hypothesis as the isomer free decay scenario. The isomer model outperformed the isomer free model with a test statistic of $\lambda_{\text{LR}} = 290$, resulting in rejecting the isomer free model with significance greater than the the computers floating point precision. The full isomer model with a β -decay branching of the isomer into a ^{180}Hf state was also compared to the model constrained to 100% internal transition to the $^{180}\text{Lu}^{\text{g.s.}}$. The model with direct β -decay of the isomer outperformed the internal transition only model with $\lambda_{\text{LR}} = 9.9$ and $\alpha = 0.0016$. We therefore conclude with high confidence that ^{180}Yb decays to a β -decaying isomer of ^{180}Lu .

There have been several experiments that have previously found weak evidence of a long-lived isomer in ^{180}Lu [124, 125, 126]. Attempts to measure the half-life of the isomer and its β -branching ratio into Hf have given conflicting results. It has been argued that such an isomer should have a half-life in the range of 10-300s based on the other nuclei with similar structure [127] in the region as well as exclusion from previous searches [128]. However previous isomer searches have been limited to direct production from proton or neutron bombardment on Hf [128], Ta [126] or W [125] targets rather than the Yb β -decay presented here. It should also be noted that the only previous half-life measurement on ^{180}Yb reported a β -decay correlated gamma ray that could not be placed into the decay scheme to $^{180}\text{Lu}^{\text{g.s.}}$, speculating that it

may be a low spin isomer of ^{180}Lu β -decaying into $^{180}\text{Hf}^{\text{m}}$ [126]. The decay of $^{180}\text{Hf}^{\text{m}}$ is known to produce $^{180}\text{Ta}^{\text{m}}$ [129]. Still, explaining the large production¹ of $^{180}\text{Ta}^{\text{m}}$ has long remained a challenge [132], so the high production and clearer indication of Yb decay into an isomeric state demonstrated in this experiment may provide hope for future decay studies of ^{180}Yb and ^{180}Lu .

Due to the relatively poor knowledge of the ^{180}Yb half-life, we are unable to place tight constraints on the branching ratios of Yb into the Lu isomer or its β -decay branching ratio and half-life. This is not unexpected as many of the model parameters are highly correlated.² Despite the fitting difficulties it is possible to place bounds on the ^{180}Yb half-life. With ^{180}Lu constrained to within 3 sigma of its literature half-life ($t_{1/2} = 5.7 \pm 0.1$ min) The Yb half-life was found to be within 140 s (2.3 min) and 215 s (3.6 min) with a best-fit value of 147 s (2.45 min). The high upper range in the half-life of ^{180}Yb is primarily due to the unknown influence of the Lu isomer. This value agrees well with the 2.4 ± 0.5 min half-life reported in [126].

5.5.1 ^{181}Yb and ^{182}Yb Half-Lives

In the case of ^{181}Yb and ^{182}Yb , the rate of implantation was too low to get sufficient statistics from just a single decay curve. In these cases, RIB beam was periodically blocked and unblocked, forming a set of beam on β -decay build-ins and beam off β -decay fall-off curves. The resulting β -decay rate can be constructed as a piecewise defined function from the Batemann equations, or solved directly from the differential

¹Large is a relative term. $^{180}\text{Ta}^{\text{m}}$ is the least abundant naturally occurring isotope, but it is still unclear if known nucleosynthesis processes can fully account for the observed abundance [130, 131].

²This problem is particularly difficult. In addition to the difficulties of fitting sums of exponentials described in the amusing essay “What Not to Compute” found in [133], we have the additional problem that an infinitely long-lived isomer and an extremely short-lived 100% β -decaying isomer can give identical decay curves.

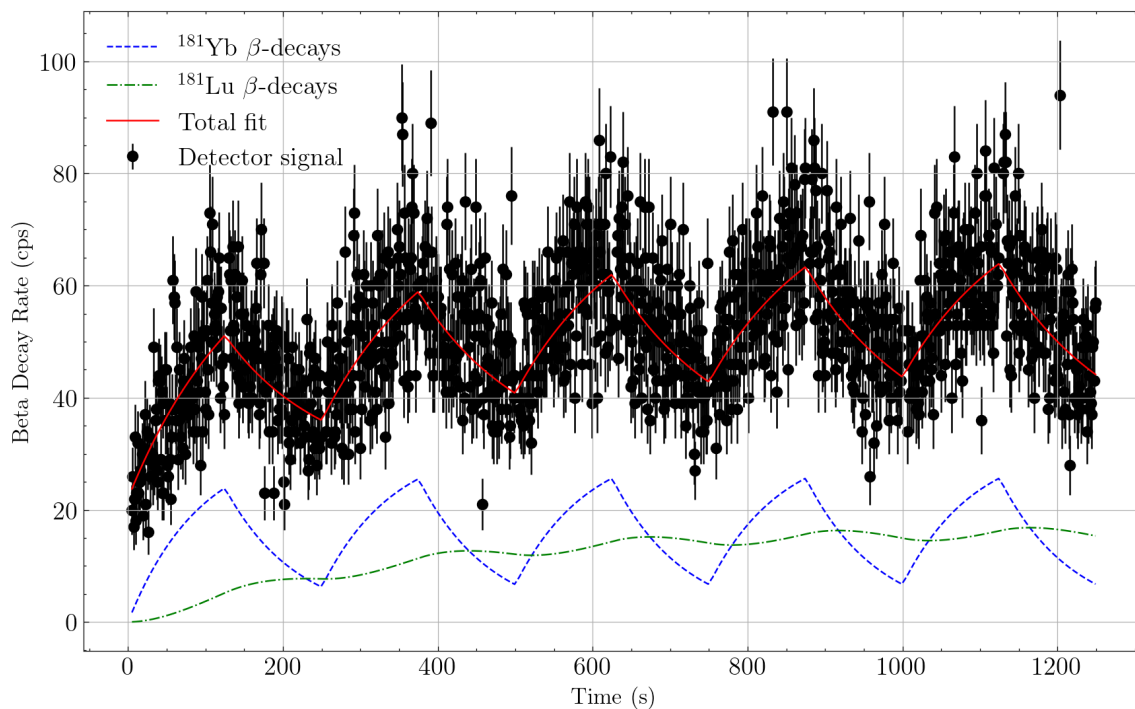


Figure 5.9: Measurement of ^{181}Yb decay. The event rate grows and decays according to the half-life of ^{181}Yb , the daughter ^{181}Lu decay and the periodically deposited ^{181}Yb ions. Ions are implanted on the detector for 125 seconds followed by the beam being blocked for an equal time.

equations. Solving the differential equations numerically has the advantage that the ion rate detected can be used directly, even if the ion rate varies over time. This advantage was not fully realized during these measurements due to software limitations requiring the TDC resolution be set too low to identify ion species. The decay curve resulting from periodically blocking and unblocking the beam will in general result in an oscillating buildup of β -decay events. The oscillations should stabilize with a mean value over a complete cycle of the primary decay equal to half the ion rate (assuming 100% detection). The ^{181}Yb decay data shown in figure 5.9 shows this clearly. To reduce the potential impact of ion rate fluctuations, the

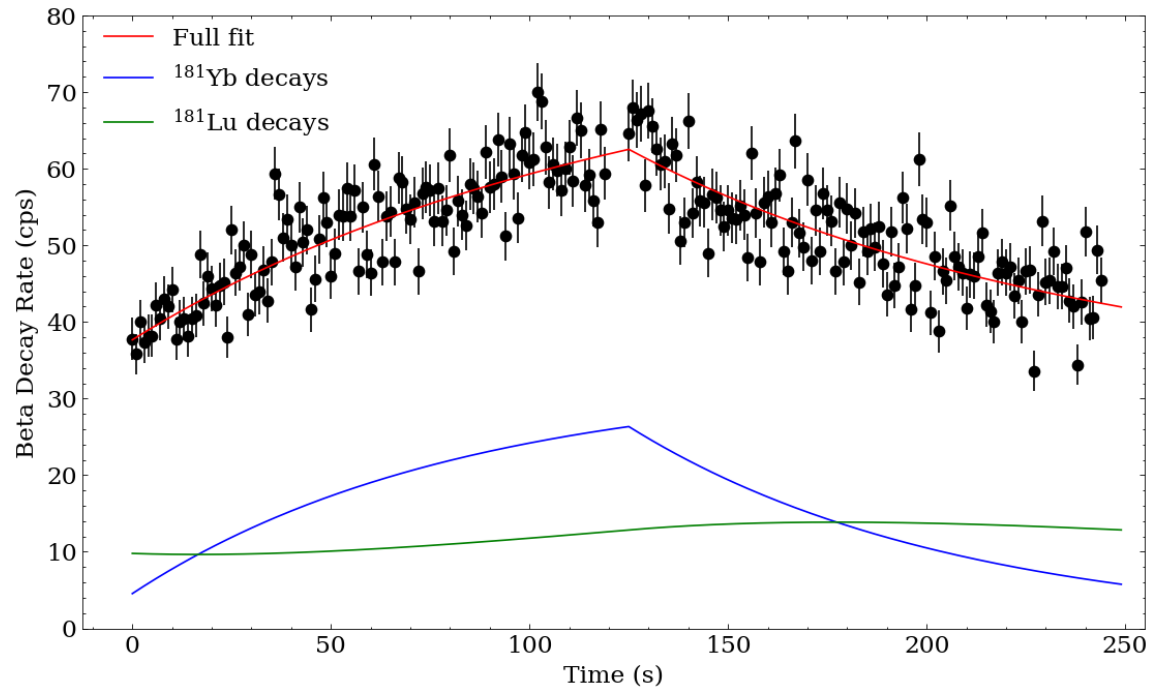


Figure 5.10: ^{181}Yb build and decay curve, with data averaged over all ion implantation cycles.

data can be summed across beam blocking cycles. One can similarly sum the fitting function across cycles to maintain the advantage of having a well defined initial state consisting solely of background signal. The cycle-summed data and fitting curve is shown in figure 5.10.

The determination of the half-life of ^{182}Yb was complicated by an order of magnitude lower production rate relative to ^{181}Yb , as well as substantial background from mass measurements performed at $A/Q = 186$. In particular, several nuclides that decay into the 2 hour half-life isotope ^{186}Pt resulted in a quasi-steady background. Measurements were done on using ion accumulation times of 90, 60, 30 and 15 seconds. The data was analyzed using the cycle summing procedure described above. Testing the statistical significance of the fit to ^{182}Yb was done with a likelihood ratio test

5.6. β -Decay on a MagneTOF Detector

Accumulation Interval (s)	Exp. Half-life (s)	Background rate (cps)	λ_{LR}	α
90	$35.5^{+15.0}_{-9.6}$	29.5 ± 0.3	24.7	1.7×10^{-5}
60	$34.2^{+25.4}_{-11.6}$	38.6 ± 0.3	10.3	0.016
30	$1000^{+\infty}_{-1000}$	32.4 ± 0.3	-0.0016	1.0
15	$9.4^{+11.8}_{-3.8}$	28.0 ± 0.3	7.8	0.050

Table 5.2: Fitting results for ^{182}Yb half-life measurements. Half-life and background uncertainties include the contributions of uncertainty on the half-life of ^{182}Lu and on the ion implantation rate.

as described for the ^{180}Yb measurement. The null hypothesis was that the data was adequately described by a constant background. Except for the case of 30 s accumulations, the likelihood ratio test excluded the null hypothesis with $p < 0.10$. The 30 s accumulation data was taken shortly after the background from $A/Q = 186$ was deposited and so had the highest background of all datasets. It was therefore excluded from further analysis. A summary of the fitting results and model statistical tests is given in table 5.2. A representative fit is shown in figure 5.11.

The final estimation of the half-life of was done with a weighted mean using the methods described in [134]. The asymmetric errors were modelled as having a distorted Gaussian shape, and the calculation was performed with the code described in [135]. The half-lives determined for all three isotopes of Yb are presented in table 5.3.

5.6 β -Decay on a MagneTOF Detector

In determining the feasibility of detecting β -decays on the MagneTOF ion detector, the ion detector signal was sent to an Ortec 142A charge-sensitive preamplifier and then to a GRIF-16 ADC card [136] for digitizing. The goal of these studies was

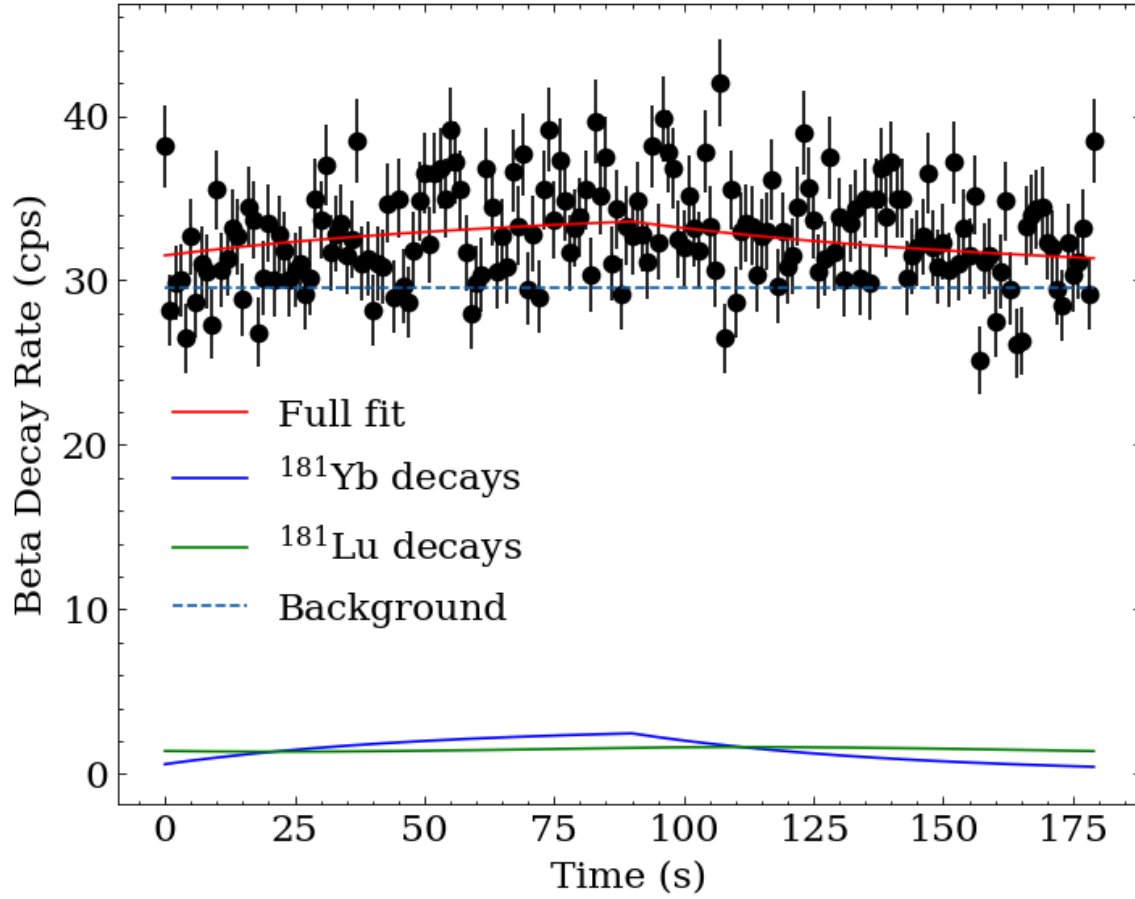


Figure 5.11: Half-life estimate for ^{182}Yb performed with 90 second accumulations and beam off decay periods. Contributions from background and the decay chain of ^{182}Yb is shown.

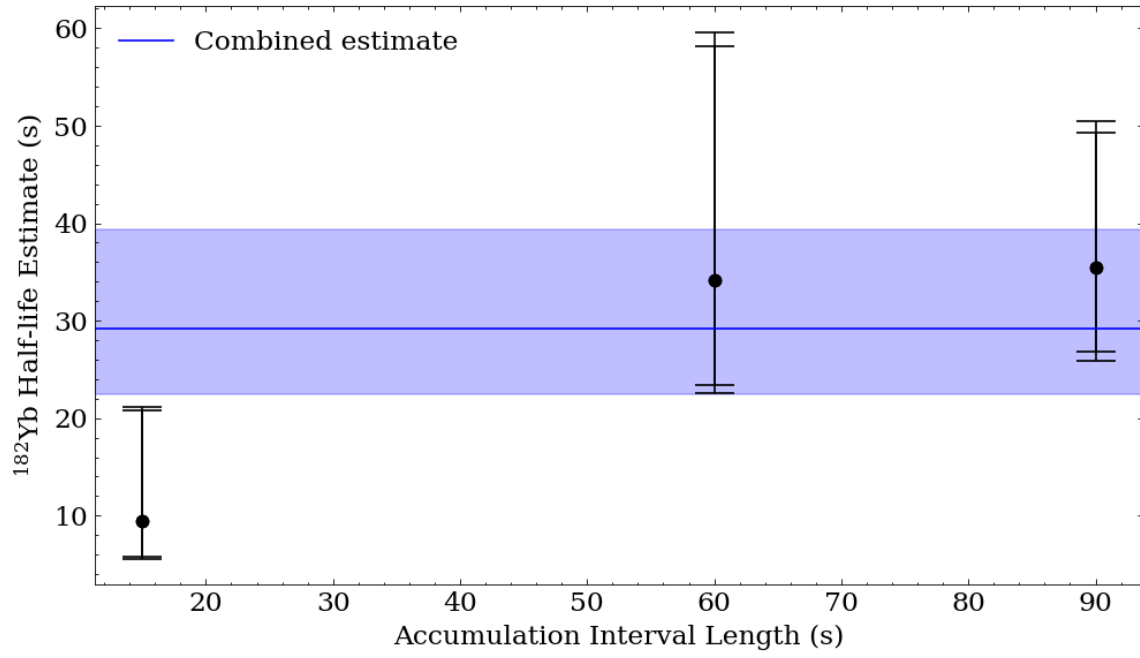


Figure 5.12: Half-life estimates for ^{182}Yb performed with different ion accumulation and decay times. Inner error bars represent pure fitting uncertainties and outer error bars include the effect of uncertainty of the ion implantation rate and half-life of ^{182}Lu . Combined half-life estimate is based on a weighted mean of the measurements.

Species	Exp. Half-life	NUBASE Half-life
^{180}Yb	$2.45^{+1.15}_{-0.15}\text{min}$	$2.4 \pm 0.5 \text{ min}$
^{181}Yb	$58^{+3.2}_{-5.8}\text{s}$	$1(\#) \text{ m } (> 300 \text{ ns})$
^{182}Yb	$29.1^{10.2}_{6.7}\text{s}$	$30(\#) \text{ s } (> 300 \text{ ns})$

Table 5.3: Yb half-lives as determined by TITAN and literature values from NUBASE 2020 [75]. Previously unmeasured half-lives are marked with (#) and the extrapolated value from NUBASE 2020 is used to compare to literature. In these cases only a lower bound on the half-life is given in the NUBASE 2020.

to determine the efficiency of β detection on the MagneTOF detector, as well as determine if the generated signal could be differentiated from an ion signal.

Data was taken by implanting ^{75}Sr which has a well known half-life of $88 \pm 3 \text{ ms}$, over 1000 s. Signals generated on the detector were continuously recorded during this time. Despite the use of re-trapping, there was also substantial contamination with the β -decaying nuclide ^{75}Rb ($t_{1/2} = 19\text{s}$). Smaller amounts of activity came from deposited $^{150}\text{Eu}^m$ ($t_{1/2} = 12.8\text{h}$), ^{150}Tb ($t_{1/2} = 4.48\text{h}$), ^{150}Dy ($t_{1/2} = 7.17\text{m}$), delivered to TITAN as doubly charged ions. Of particular concern is the decay of ^{150}Dy as this has a substantial alpha-decay branching probability of $P_\alpha = 36 \pm 5\%$. ^{150}Dy was implanted at a rate of $\sim 0.35 \text{ pps}$, resulting in a maximum rate of alpha decays of $\sim 0.1 \text{ cps}$. This represents less than 2% of the total detected decays ($\sim 6 \text{ cps}$), and so can be ignored in determining the pulse height distribution of β -decays. A comparison of the pulse heights of radioactive decays and signals generated from time-of-flight ions is given in figure 5.13, normalized by the total detections of each. While there is an excess of higher energy pulses detected for β -decays, the distribution is too similar to be able to discriminate β -decays from time-of-flight ions from pulse height alone.

The efficiency of β -decay detection was determined using ^{63}Fe . This isotope could be delivered to TITAN at relatively high rates, resulting in a total ion implantation rate of $60 \pm 2 \text{ pps}$. With the use of re-trapping the sum of all other ion species were

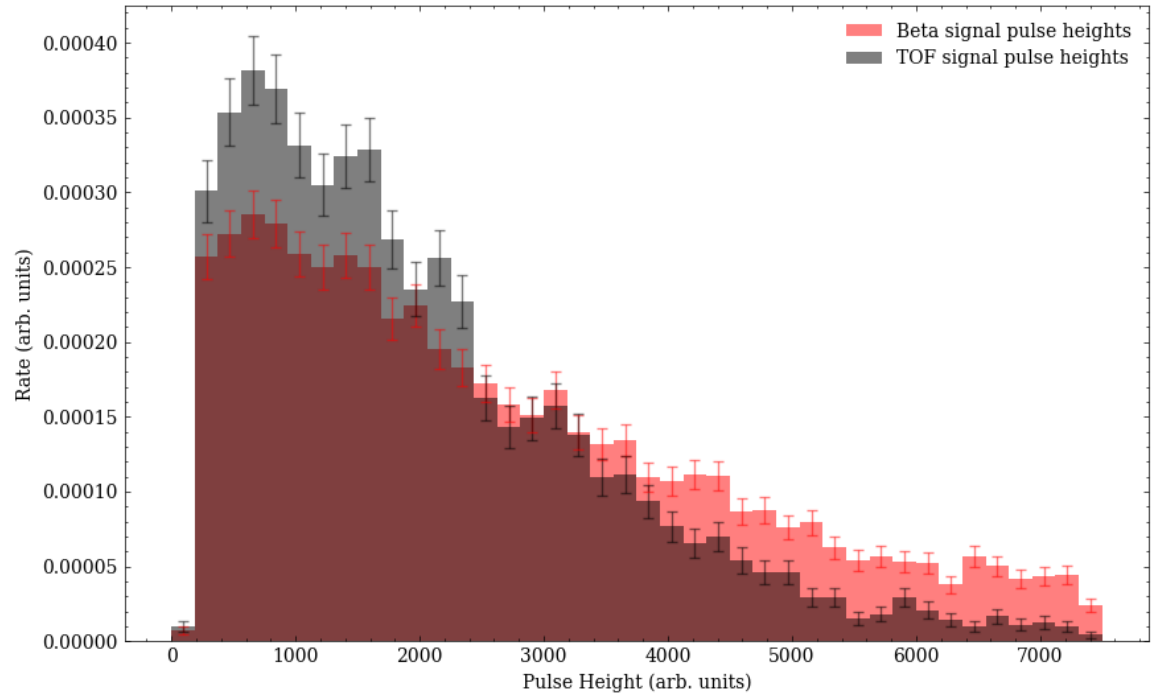


Figure 5.13: Pulse heights captured with the GRIF-16 ADC for ions and β -decays of Sr.

implanted at a rate much less than 1 pps. ^{63}Fe has the benefit of a simple and short decay chain, allowing for an accurate efficiency measurement. ^{63}Fe decays with a half-life of 6.1 ± 0.6 s into ^{63}Co which in turn decays with a half-life of 26.9 ± 0.4 s into ^{63}Ni . The half-life of ^{63}Ni is ~ 100 years and so the decay rate is negligible for this measurement.

^{63}Fe was allowed to implant on the detector for many ^{63}Co half-lives, until the rate of β -decays had stabilized, indicating that an equilibrium of ^{63}Co decay and production had been reached. The rate of time-of-flight ions and β -decays was then simultaneously measured for 100 s. The rate of time-of-flight ions was determined to be 60.8 ± 0.8 pps and the β -decay rate was 60.4 ± 0.8 pps. The detector efficiency relative to the ion detection efficiency was calculated as $49.7 \pm 0.9\%$, in line with the expected 50% from the geometry of the detector.

The RIB was then blocked and the signal rate on the detector allowed to reduce for 100 s. The resulting decay curve was fit with a model allowing some fraction of the daughter products to escape from the detector surface. Within the fit uncertainties, all recoiling nuclei remain on the detector surface after β -decay. This is expected as the energy of the recoiling nucleus will always be much less than the ~ 3 keV/e ion implantation energy. The fit indicated that the background rate during the measurement was consistent with zero within uncertainties, and so it is concluded that the efficiency estimate is not substantially inflated.

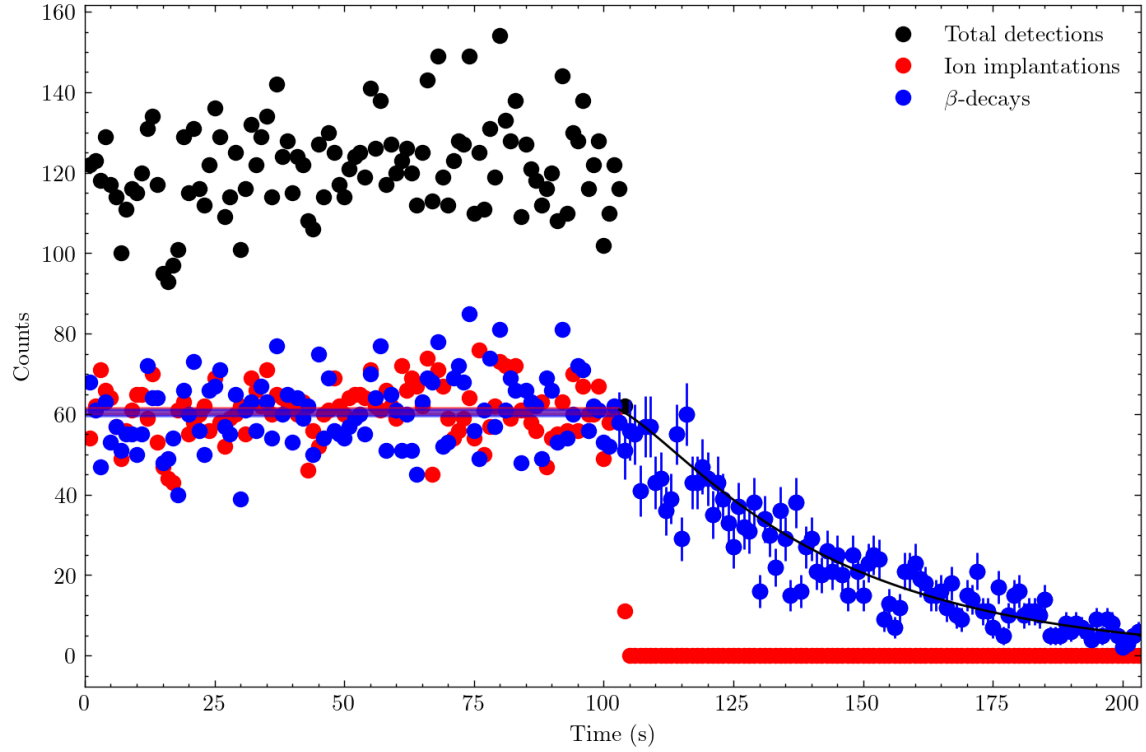


Figure 5.14: Total detected rate of ^{63}Fe implantation and β -decays in black. Time-of-flight implantation events are shown in red and β -decays in blue, with the average rates prior to blocking RIB depicted by the coloured bands. A fit of the β -decay curve when RIB was blocked is shown in black.

CHAPTER 6

Not Just High-precision Mass Measurement: Beam Composition Monitoring and Chemistry with TITAN's MR-TOF-MS

6.1 Beam Composition Measurements for a New ISAC Target at TRIUMF

6.1.1 Introduction

Successful nuclear experiments require detailed knowledge about the beam properties. The beam composition of radioactive ion beams (RIB) at Isotope Separation On-Line (ISOL) facilities depends on in-target production, element diffusion and ionization

efficiency [137, 138]. In-target production can generally be estimated well using a variety of techniques [139, 140, 141, 142]. Ionization efficiency from thermal sources can also be well predicted from each element’s ionization potential [143]. However, diffusion from a thick target is difficult to accurately model due to a myriad of chemical reactions that can take place at the high temperature of an ISOL target [138]. Choices in target material and operating parameters greatly influence the yield of the ion of interest and its contaminants, which are usually determined through gamma and particle spectroscopy.

Recently, TRIUMF has commissioned a new ISOL target using spallation neutrons to drive fission in uranium [144, 145]. This new target is referred to as a proton-to-neutron (P2N) target [145, 146, 147]. A large number of beam requests at TRIUMF’s Isotope Separator and ACcelerator (ISAC) facility [57] are for high rates of neutron-rich isotopes with neutron-deficient contamination [145]. This target aims to improve rates of neutron-rich isotopes near the $A=80$ and $A=130$ fission peaks, while reducing stable and neutron-deficient surface-ionized contamination.

Typically, ISAC uses its Yield Station [148] to measure the rates of isotopes produced from a target. The Yield Station is equipped with alpha, beta and gamma detectors allowing accurate counting radioactive isotopes produced from the target. As such the Yield Station can measure many species simultaneously and for high count rates. However, the Yield Station suffers from an inability to detect stable and long-lived isotopes as well as to distinguish species with similar half-lives when using the beta scintillator detectors. The Yield Station is also unable to detect low-rate species against high backgrounds. To address these shortcomings, TRIUMF’s Ion Trap for Atomic and Nuclear science (TITAN [149]) MR-TOF-MS [48, 56] can be enlisted to provide yields of all isobars in the beam.

6.2 TITAN MR-TOF-MS for Isotope Yields

The TITAN MR-TOF-MS [56, 48] has been used to complement the ISAC Yield Station in providing critical information about the beam and to optimize RIB delivery [150]. MR-TOF-MS operate by bouncing monoenergetic ions between a pair of electrostatic ions before detecting them. From their time of flight the mass can be deduced with high precision. This broadband technique allows for measurements of all constituents of the RIB simultaneously. The high resolution and high dynamic range demonstrated at the TITAN MR-TOF-MS have permitted detailed studies of beam composition [150], to provide yields lower than detectable at the Yield Station (e.g. [151, 17]), and to optimize the ISAC mass separator [150].

For a collaborative yield measurement, first, the ISAC Yield Station is used to measure an absolute rate of the desired element for each isotope until the rates are too low or indistinguishable from the background. Typically, these are the most active species based on rate and half-life, as this will provide the best signal-to-noise ratio. Second, the TITAN MR-TOF-MS will be used to measure the yield of a few of these isotopes to determine its own efficiency. Then measurements will continue for increasingly exotic nuclides until the rate becomes too low for the MR-TOF-MS. In addition, the broadband nature of the technique allows simultaneous estimates of the contaminant isobaric species, whether long-lived or stable. In the event that two isobars have similar half-lives, an iterative approach is adopted. The MR-TOF-MS's relative rates can inform the fitting of the Yield Station's data. This allows for more precise determinations of the absolute rates at the Yield Station. This, in turn, improves TITAN's absolute determination of all isobaric contaminants. The TITAN and Yield Station groups have collaborated on many experiments. The Yield Station

is preferred for shorter-lived species (minutes or shorter) and absolute rates due to its straightforward calibration. If the background is sufficiently low, rates as low as 50-100 pps are feasible. On the other hand, the MR-TOF-MS has a superior dynamic range due to its “self-cleaning” mode [152] and higher sensitivity (>0.03 pps [153]). In addition, by being sensitive to space charge, the MR-TOF is requisite for stable and longer-lived species (hours or longer). Thus, it can be seen that the two measurement techniques complement one another by providing critical information inaccessible to the other.

6.3 Results

6.3.1 Yield Variation as a Function of Target Heating

The target is heated to temperatures near 2000°C by heating coils, which provides more even heating than the proton beam itself. The release of radioactive nuclides from the target depends on the target temperature and the species’s chemical properties like diffusion and effusion. The variation in yields as a function of temperature is more easily executed at TITAN, due to the MR-TOF-MS’s continuous, broadband measurement capability.

For the P2N target, the target heating current was ramped from 950 A to the target’s nominal heating current of 1150 A over a 1.5 hour period in 2.4 A/min steps. Many species in the RIB were continuously monitored by the TITAN MR-TOF-MS, including Zn for which laser ionization [154] was required. Rates of the five major atomic constituents of the beam are presented in figure 6.1. Overall, the intensity of most species trends upwards, likely due to higher temperature which promotes a more efficient release behaviour. It can be noted that the constituent species

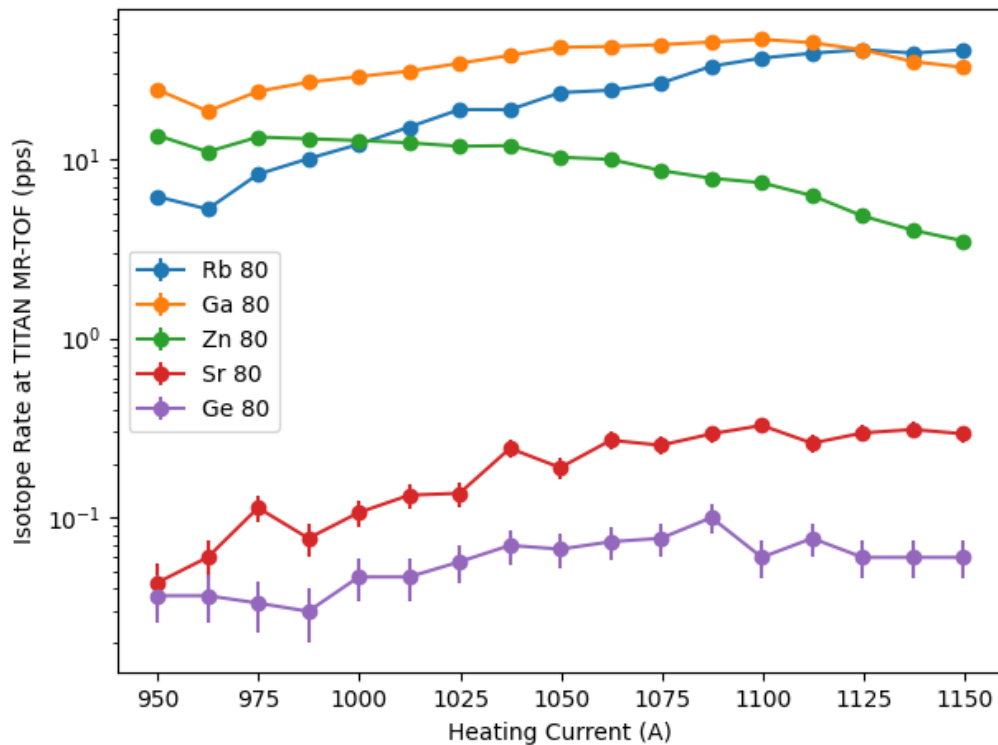


Figure 6.1: Variation in yields of five isobaric species as a function of target heating current. Data is binned into 5 min intervals. The rate of each species was measured simultaneously throughout the 1.5 hour ramping procedure. Statistical error bars are shown.

of the RIB do not increase in rate uniformly. The variance is mostly due to the different half-lives and chemical characteristics of the different species. ^{80}Zn , the only laser-ionized species, had unique behaviour. Its rate was found to fall at higher target temperatures. It is hypothesized this could be due to additional space charge in the ionizer tube resulting in slower Zn extraction and increased sticking to the ionizer tube walls. Other possibilities include chemical effects at high temperature such as alloying to the uranium-carbide matrix or contaminants. Measuring these trends is

useful to target and ion source physicists so they may choose the optimal temperature for the element or species of interest to the experimenter.

6.3.2 Quantifying Low-Rate Contamination Near the Fission Peaks

FLUKA simulations taking into account available nuclear reaction data [139, 144] have suggested that the use of a neutron converter rather than direct proton impingement on uranium would result in improved beam purity in the $A=80$ and $A=130$ fission peaks. To verify this, yields of several isotopic chains with well understood release properties were measured. This was done for several target heating currents, see Figure 6.2 for the $A=80$ peak at target currents of 800 A, 950 A, and 1150 A. Rb and Sr were chosen for being easily surface ionized. While the Yield Station could be used to confirm the fission peaks, the MR-TOF-MS was required to map their edges due to the very low rates.

Figure 6.2, when combined with Figure 6.1, demonstrates the utility of an MR-TOF-MS in experiment setup. Knowing the rate of the species of interest, the identity of the contaminant species, and their relative rates is crucial to achieve the desired precision. Near the fission peak, the long-lived Rb and Sr are a dominant source of contamination and not necessarily well separated by half-life. In these cases the MR-TOF-MS was also used to constrain the fitting of the Rb and Sr decays measured by the Yield Station.

6.3.3 Quantifying Stable and Long-lived Contamination

As TITAN's MR-TOF-MS is sensitive to all isobars in the beam, it can quantify rates of stable and long-lived contaminants, including molecular ions, unlike the Yield

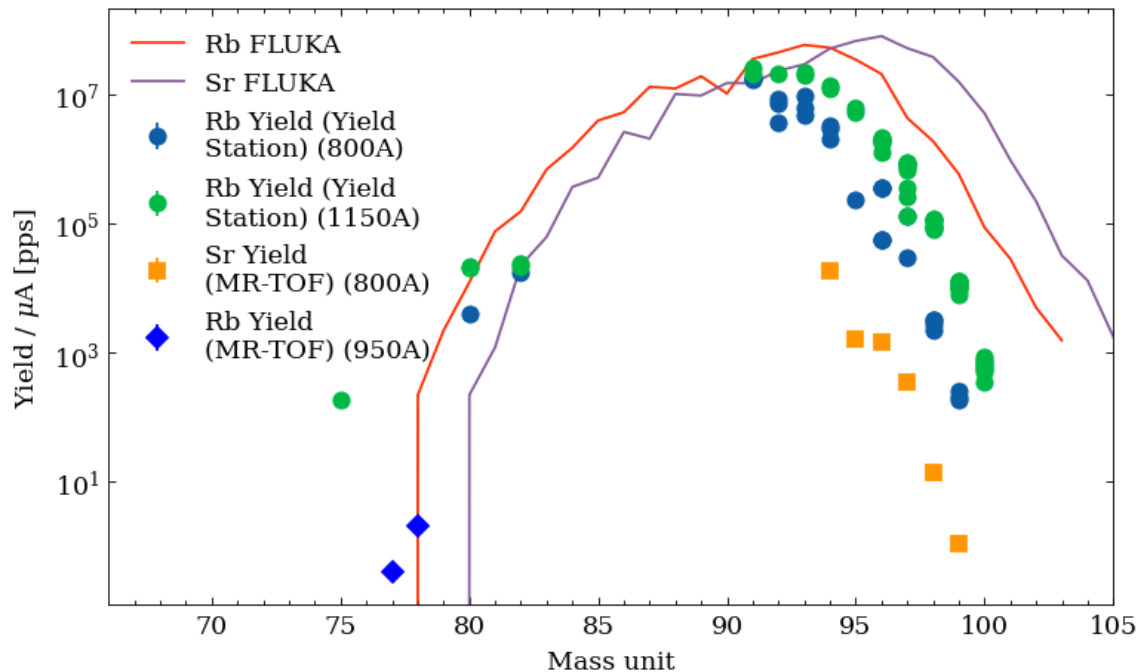


Figure 6.2: Mass spectrum of surface ionized beam from the Yield Station data [144] confirms the fission peak, while the MR-TOF-MS data is required to map the edges of the fission peak, especially at lower than nominal target heating currents, due to its superior sensitivity. In target production was estimated using the FLUKA Monte-Carlo simulation package [139, 144].

Station. For the P2N target, we chose to compare it to RIB produced with a typical ISAC uranium carbide (UC_x) target, e.g. [17] and thus compare beams produced predominantly through neutron-induced fission and proton-induced spallation, respectively. A comparison at $A/Q=65$ is shown in Figure 6.3. Stable, molecular ions were reduced at most A/Q measured and compared for the fission-produced beams. Similarly, long-lived atomic and molecular ions, like neutron-deficient $^{130}\text{Ba}^{2+}$, were suppressed or completely eliminated under fission production. See [144] for more detailed discussion on the underlying processes.

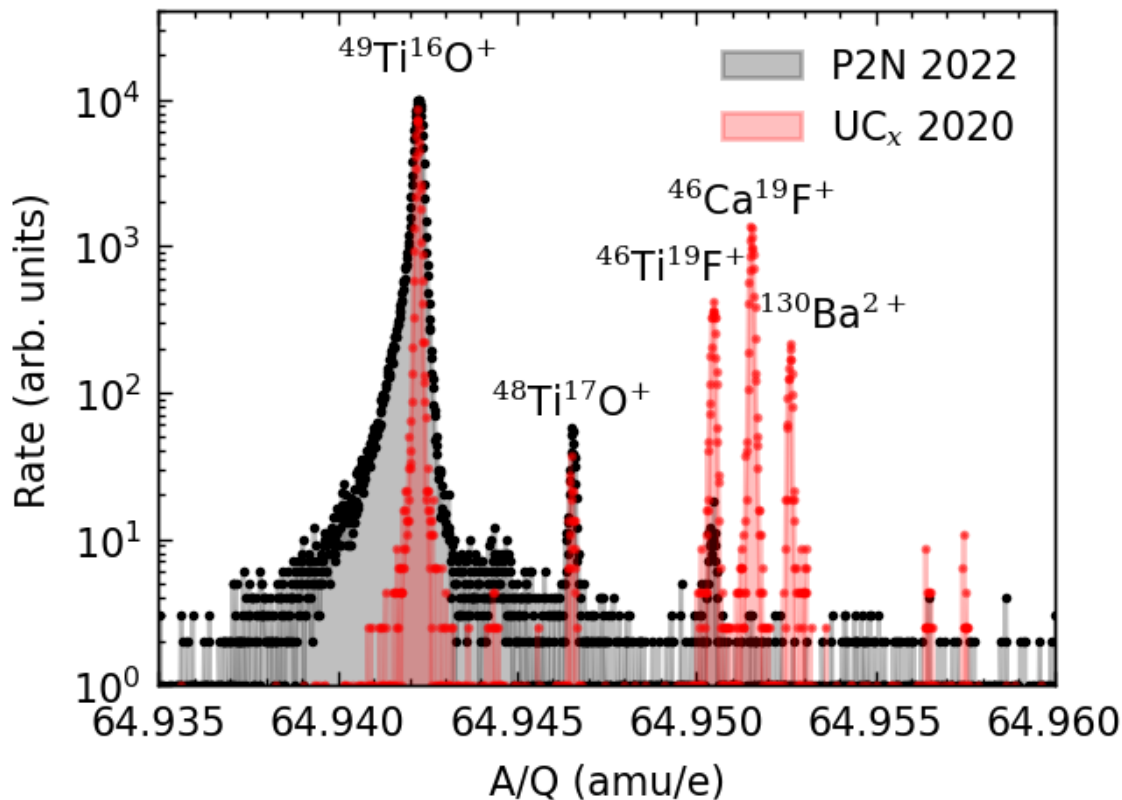


Figure 6.3: Mass spectrum of RIB at $A/Q = 65$ with target heating at 1150 A. This work is compared to a prior TITAN experiment [17] on a spallation UC_x target. Both data sets were taken with surface ionization. Rates of ^{19}F molecules and doubly charged neutron-deficient contaminants are reduced for neutron-induced fission. Data is normalized to the rate of $^{49}\text{Ti}^{16}\text{O}^+$ to evaluate the changes in beam composition.

6.3.4 Monitoring Isomer Rates

Whenever the half-lives of radioactive isobars are similar, their separation can be difficult to accomplish with the Yield Station as is the case for neutron-rich, odd- A indium isotopes. In this case, TITAN had previously distinguished the isomers from one another and the ground state [151]. As such, the MR-TOF-MS was critical not only to quantify the rates of each nuclear state. A comparison of the

fission-produced to spallation-produced rates, as shown in Figure 6.4, provides critical information about RIB production mechanisms. The $J^\pi = 1/2^-$ isomers exhibit vastly different behaviour depending on whether neutron- or proton-induced production. The variation in the ratio of ground state to isomer within this isotopic chain reflects the nuclear mechanism only since the decay losses are comparable due to the similar half-lives and the chemical processes are identical.

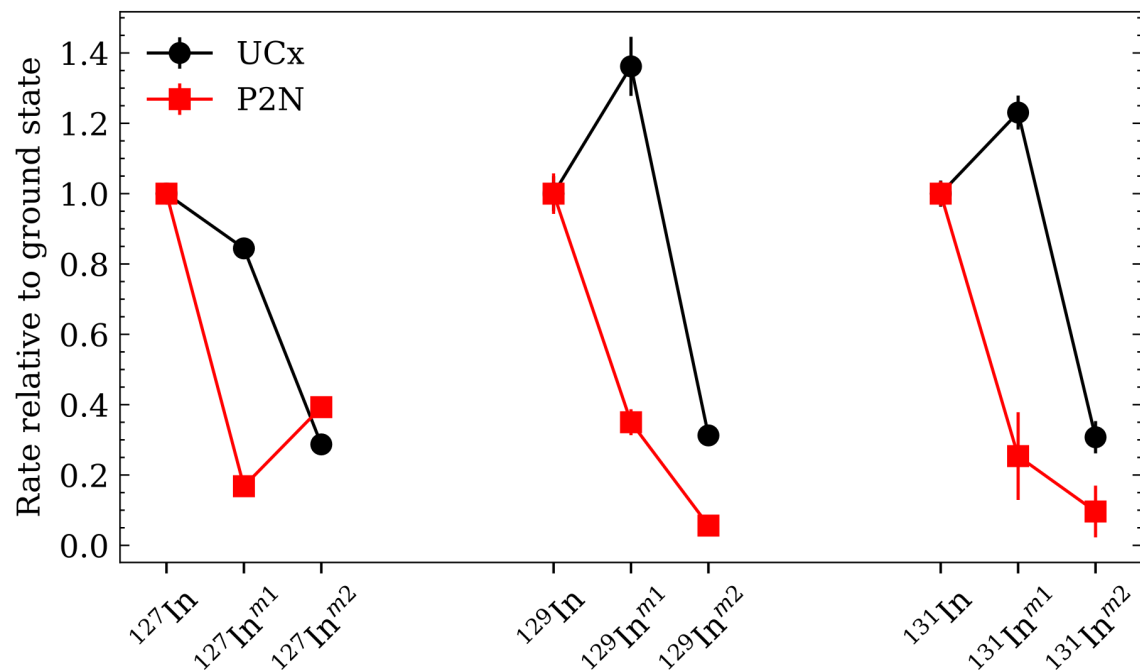


Figure 6.4: Rates of long-lived isomers of indium comparing neutron-induced fission and proton-induced spallation [151]. Error bars represent statistical fitting uncertainties only.

In addition to quantifying the isomeric rates, the MR-TOF-MS can be used to purify a particular nuclear state for further study [151]. Thus, we can understand how invaluable the MR-TOF-MS can be not only to inform experimenters' expectations but also to improve the experiment through isomeric purification.

6.4 Summary

We have demonstrated the critical and complementary data an MR-TOF-MS provides to traditional spectroscopy in characterizing targets, here a new, proton-to-neutron-converter ISOL target. We showed how the TITAN MR-TOF-MS is required to determine the rates of stable and long-lived contamination as well as those for short-lived species with similar half-lives. In addition, the constant monitoring enabled by the MR-TOF-MS, permits more detailed studies of the RIB response to variables like target heating. These data provide critical information about the production mechanisms, chemical dependencies, and other target-performance criteria. As such, target physicists can improve their models. Specific to the P2N commissioning, the dramatic reduction in contamination, especially neutron-deficient, achieved a crucial criterion for success. For experimenters, their experiments can be set up with the most accurate information about the expected rates of both the species of interest and its isobaric contamination when the Yield Station is complemented by an MR-TOF-MS.

CHAPTER 7

Conclusion and Outlook

The TITAN MR-TOF-MS has been used to perform high precision mass measurements of neutron-rich potassium isotopes at the TRIUMF ISAC facility. The mass measurements have provided evidence for continued magic character in neutron number $N = 34$ nuclides with proton number $Z \leq 20$. The mass measurements also provide new data for benchmarking the accuracy of *ab initio* methods, near the maximum mass limit of current techniques. This provided additional confirmation of the claims of Soma et. al. that three-nucleon interactions must be considered to correctly reproduce the trend of binding energies in heavy *ab initio* models [26].

The MR-TOF-MS has also been applied to problems beyond high precision mass measurement. The MR-TOF-MS has been applied as a complementary and supplementary technique for measuring yields of rare isotope beam, and contamination within the beam. In particular, MR-TOF-MS has been demonstrated as a tool for

developing the most challenging and exotic beams, and quickly characterizing new radioisotope production techniques.

The potential science impact of the TITAN MR-TOF-MS has been extended through new half-life determination methods. These were applied to four nuclides, resulting in the improvement of one previously measured half-life (^{54}K) and the first half-life determination in two isotopes ($^{181,182}\text{Yb}$). Evidence was also found for ^{180}Yb decay into an isomer of ^{180}Lu , with potential implications for the production of the rarest naturally occurring isotope $^{180}\text{Ta}^{\text{m}}$. Neutron delayed β -decay branching probabilities were measured in ^{54}K for the first time, demonstrating the range of decay data that can be gathered with these new techniques.

The demonstrated ability to measure half-lives on the order of several milliseconds to several minutes may enable half-life measurements of very neutron-rich nuclei, reducing one of the nuclear input uncertainties to r-process nucleosynthesis calculations. The ability to measure half-lives has also already been added to scientific proposals at TITAN. Combined mass and half-life measurements of neutron deficient lanthanide species near the proton drip-line promise to provide additional structure information on these isotopes [155]. Measurements of the most neutron rich nuclei will provide key nuclear inputs to improve the quality of r-process nucleosynthesis calculations.

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APPENDIX A

.LST Parsing and Data Analysis

This appendix is meant to briefly document some basic code for working with beta decay data gathered with the TITAN MR-TOF-MS. The code presented here and in the following appendix is hosted the TITANCollaboration GitHub account. The complete and up to date code is currently in a private repository and will be made public after publication of the results presented in chapter 5. The code is provided as a foundation for future analysis and for developing a comprehensive decay analysis suite, analogous to the mass analysis project led by Stefan Paul [73].

Code A.1: Convenience class for storing β -decay events.

```
class betaEvent:
    def __init__(self, tof_ev, beta_ev, cycle_time):
        self.ion_tof = tof_ev.time
        self.beta_decay_time = (beta_ev.cycle - tof_ev.cycle) * cycle_time
                               + beta_ev.time - tof_ev.time
```

Code A.2: Function for taking list type data and placing it in the spectrum class.

```
def list_to_spectrum(dataList, cycle_offset=0):
    grifList = []
    for dataFile in dataList:
        for tofData in dataFile:
            grifList.append(Event(time = tofData.time,
                                  cycle = tofData.sweep+cycle_offset,
                                  channel = tofData.channel,
                                  pulse_height=0, flags=0))
        cycle_offset = grifList[-1].cycle
    return Spectrum(events = grifList)
```

The “Spectrum” class is meant for easily working with mass and decay data gathered with either the MCS6A TDC or the GRIF16 card as described in chapter 5. It provides utilities for quickly viewing the spectrums, getting lists of events, or applying filters to the data. The basic unit of data is the “Event” defined in code A.3. The filters that can be applied to the data include simple conditions created with the “Condition” class presented in code A.4, or more complicated user defined functions.

Code A.3: Event class for storing hits detected by the GRIF16 card or MCS6A TDC.

```
@dataclass
class Event:
    """
    Contains all information for a single event given by GRIF16 card.
    """

    time: int
    cycle: int
    channel: int
    pulse_height: float
    flags: int

    def __satisfies__(self, cond):
        """Check whether an event satisfies the given simple condition."""
        value = self.__getattribute__(cond.data_field)
        return (cond.lower <= value <= cond.upper) != cond.invert
```

Code A.4: Condition class for making simple filters for the Spectrum class.

```
@dataclass
class Condition:
    """
    Simple condition class. Can be applied to any field in the event class.
```

```

"""

data_field: str
lower: float = float("-inf")
upper: float = float("inf")
invert: bool = False
__name__: None

def __post_init__(self):
    __name__ = self.data_field

```

Code A.5: Spectrum class for easily working with time-of-flight and decay data.

```

from dataclasses import dataclass

@dataclass
class Spectrum:
    """
    Contains a list of events, conditions, and a
    filtered list of events based on those conditions.
    """

    events: list = field(default_factory=list)
    conditions: list = field(default_factory=list)
    filteredEvents: list = field(default_factory=list)
    conditionsApplied: bool = False

    def addCondition(self, cond, *params, **keywordParams):
        """
        Takes either a condition, or conditionFunc,
        and applies it to the spectrum.
        """
        if type(cond).__name__ == "Condition":
            self.conditions.append(cond)
        else:
            self.conditions.append([cond, params, keywordParams])
        self.conditionsApplied = False

    def getEvents(self):
        """
        Return the filtered events, i.e. after applying conditions.
        """
        #short circuit the evaluation if we've already done the work
        if self.conditionsApplied == True:
            return self.filteredEvents

        self.filteredEvents = self.events.copy()
        for cond in self.conditions:
            self.filteredEvents = self.__satisfies__(cond)
        self.conditionsApplied = True
        return self.filteredEvents

```

```

def __satisfies__(self, cond):
    """Internally used function to apply conditions."""
    if type(cond).__name__ == "Condition":
        filteredEvents = [ev for ev in self.filteredEvents
                           if ev.__satisfies__(cond)]
        self.filteredEvents = filteredEvents
    else:
        filteredEvents = cond[0](self.filteredEvents,
                                *cond[1], **cond[2])
        self.filteredEvents = filteredEvents
    return filteredEvents

def copySpectrum(self):
    """
    Return a copy of the current spectrum, in its current state.
    """
    return copy.deepcopy(self)

def addSpectrums(self, spect):
    """
    Creates a new spectrum object containing only
    the filtered events from the original and passed
    spectra. Made without any conditions as the
    conditions shouldve already been applied.
    """

    assert self.conditionsApplied==True and spect.conditionsApplied==True
    newSpectrum = Spectrum(events=self.filteredEvents+
                            spect.filteredEvents,
                            conditionsApplied=True)

    return newSpectrum

def getField(self, data_field):
    return [ev.__getattr__(data_field)
            for ev in self.getEvents()]

def viewSpectrum(self, data_field, bins=1000):
    plt.figure()
    events = [None] * 16
    for i in range(16):
        events[i] = [ev.__getattr__(data_field)
                     for ev in self.getEvents()
                     if ev.channel == i]
    bins=np.histogram(np.hstack(events), bins=bins,
                      range=(0,max(events[10]))) [1]

    for ev in events:
        if len(ev)>0:
            h = plt.hist(ev, bins=bins,
                          range=(0,max(ev)), log=True,
                          histtype='stepfilled')
    cond = [c.__name__ for c in self.conditions

```

```
        if type(c).__name__ == "Condition"]  
plt.title("∪".join(cond))  
return h
```

APPENDIX B

Numerically Solving Decay Systems

While there exists an exact analytical expression for solving a complex chain of radioactive decays (see [156]), this can often lead to unwieldy expressions. It is often faster¹ and less error prone to instead numerically solve the initial value problem directly. This is especially the case when there exists an intermittent source of new radioactive nuclei, as the final decay curve would then have to be constructed piecewise from the Batemann equations. With the expectation that the TITAN MR-TOF-MS will be regularly used for decay measurements with the technique shown with $^{181,182}\text{Yb}$ in chapter 5, I present a simple code for quickly numerically computing decay curves. The presented code allows for optional input of an arbitrary source function of new nuclei. Such a source could be an intermittent constant source, or even the directly measured rate of ions at a given time. The code allows for as many

¹Faster in terms of scientist time, which is of course more precious than computer time.

nuclides as one cares to define, with decay chains described through “DecayBranch” class. The detection efficiency of each decay can be separately defined, allowing for example, isomer gamma decays that are missed by our ion detector.

As a warning to anyone who uses the code in the future, it is left to the user to ensure that the output does not have large numerical integration uncertainties. Convergence of the output should be checked by iteratively reducing the solution tolerances as well as comparing between different integration algorithms (e.g. LSODA, RK23, RK45). The basic code for generating decay curves is presented in code B.1. A simple example script generating a decay curve is given in code B.2. This code demonstrates a decay chain into a daughter or an isomer, and later decay of the daughter into a stable nucleus. The isomer decays into the first daughter, a secondary daughter, or directly into a stable nucleus. To demonstrate the use of a source function for new nuclei, the parent takes a user defined periodic function.

Code B.1: Classes and methods to build a decay curve of an arbitrary number of nuclides, and then fit with possible variables branching ratios, half-lives, background and source of new nuclei.

```

import numpy as np
import matplotlib.pyplot as plt
from scipy.integrate import odeint, solve_ivp
from lmfit import Model, Parameters

class Nuclide:
    def __init__(self, name, half_life=np.inf, initial_population=0,
                decay_branches=None, source_func=None,
                source_func_fit_params=None, vary_half_life=False,
                vary_initial_population=False):

        self.name = name
        self.half_life = half_life
        self.decay_constant = (np.log(2) / half_life
                               if half_life != np.inf else 0)
        self.initial_population = initial_population
        self.decay_branches = decay_branches if decay_branches else []
        self.source_func = source_func
        self.vary_half_life = vary_half_life

```

```

        self.vary_initial_population = vary_initial_population
        self.source_func_fit_params = source_func_fit_params

class DecayBranch:
    def __init__(self, daughter='Stable', branching_ratio=1,
        detection_efficiency=1, vary_branching=True):
        """
        vary_branching: if True, the branching ratio is allowed to
        vary in the fit. In the fit, free branching ratios are defined
        via weights and normalized so that the sum (plus any fixed
        contributions) equals one.
        """
        self.daughter = daughter
        self.branching_ratio = branching_ratio
        self.detection_efficiency = detection_efficiency
        self.vary_branching = vary_branching

    def get_daughter_name(self):
        return (self.daughter.name if isinstance(self.daughter, Nuclide)
            else self.daughter)

class DecayModel:
    def __init__(self, nuclides, background=0.0):
        self.nuclides = nuclides
        self.background = background
        self.lambdas = np.array([nuclide.decay_constant
            for nuclide in nuclides])
        self.N0 = np.array([nuclide.initial_population
            for nuclide in nuclides])
        for n in self.nuclides:
            total_branch = sum([b.branching_ratio for b in n.decay_branches])
            # Allow for minor rounding differences.
            if abs(total_branch - 1.0) > 1e-12:
                raise ValueError("sum_of_branching_ratios_not_1._Check_" +
                    n.name)

    def decay_system(self, t, y):
        dN_dt = np.zeros_like(y)
        for i, nuclide in enumerate(self.nuclides):
            dN_dt[i] -= self.lambdas[i] * y[i] # self-decay
            if nuclide.source_func and nuclide.source_func_fit_params:
                dN_dt[i] += nuclide.source_func(t, **nuclide.source_func_fit_params)
            elif nuclide.source_func:
                dN_dt[i] += nuclide.source_func(t)

            # Transfer to daughter(s) if not stable.
            for branch in nuclide.decay_branches:
                if branch.daughter != 'Stable':
                    j = self.nuclides.index(branch.daughter)
                    dN_dt[j] += self.lambdas[i] * y[i] * branch.branching_ratio

        return dN_dt

```

```

def solve(self, time, integration_method=None):
    if integration_method:
        return np.transpose(solve_ivp(self.decay_system, [0, time[-1]],
                                      self.N0, t_eval=time, atol=1e-12,
                                      rtol=1e-9, method=integration_method).y)

    return odeint(self.decay_system, self.N0, time, atol=1e-12,
                  rtol=1e-9, tfirst=True)

def signal(self, time, integration_method=None):
    remaining_nuclei = self.solve(time, integration_method)
    time_bin_size = (time[-1] - time[0]) / len(time)
    signals = []
    for i, lam in enumerate(self.lambdas):
        for branch in self.nuclides[i].decay_branches:
            signals.append(remaining_nuclei[:, i] * lam *
                          branch.branching_ratio * branch.detection_efficiency)
    signals.append(np.ones_like(remaining_nuclei[:, 0]) *
                  self.background)
    return np.sum(np.array(signals), axis=0)

class DecayChainFitter:
    def __init__(self, nuclides, background=0.0, vary_background=False):
        """
        nuclides: list of Nuclide objects defining the decay chain.
        background: constant background signal.
        """
        self.nuclides = nuclides
        self.background = background
        self.vary_background = vary_background

    def build_params(self):
        """
        Automatically build an lmfit Parameters object.
        For each nuclide with vary_half_life==True, add a parameter
        "half_life_<Name>".
        For each branch with vary_branching==True,
        add a free weight parameter with key
        "p_<Parent>_to_<Daughter>" and then add an expression parameter
        named "branching_<Parent>_to_<Daughter>" that normalizes these
        weights so that the free branching ratios sum to
        (1 - fixed_branch_sum).
        """
        params = Parameters()
        for nuclide in self.nuclides:
            if nuclide.vary_half_life:
                params.add(f"half_life_{nuclide.name}",
                          value=nuclide.half_life, min=0.0)

            # Gather free branches for this nuclide.
            free_keys = []

```

```

for branch in nuclide.decay_branches:
    if branch.vary_branching:
        key = f"p_{nuclide.name}_to_{branch.get_daughter_name()}"
        free_keys.append(key)
        params.add(key, value=branch.branching_ratio, min=0.0)
    if free_keys:
        # Compute fixed sum from branches that are not free.
        fixed_sum = sum(branch.branching_ratio for branch in
                        nuclide.decay_branches if not branch.vary_branching)
        available = 1.0 - fixed_sum
        # Build sum expression for free parameters.
        sum_expr = " + ".join(free_keys)
        # add an expression parameter for it branching ratio.
        for branch in nuclide.decay_branches:
            if branch.vary_branching:
                free_key = f"p_{nuclide.name}_to_{branch.get_daughter_name()}"
                expr = f"{free_key} / ({sum_expr}) * {available}"
                param_name = f"branching_{nuclide.name}_to_{branch.get_daughter_name()}"
                params.add(param_name, expr=expr, min=0, max=1)

    if nuclide.vary_initial_population:
        params.add(f"initial_population_{nuclide.name}",
                  value=nuclide.initial_population, min=0)

    if nuclide.source_func_fit_params:
        for key in nuclide.source_func_fit_params:
            params.add(key+f"_{nuclide.name}",
                      value=nuclide.source_func_fit_params[key])

if self.vary_background:
    params.add('background', value=self.background, min=0)

return params

def model_func(self, time, integration_method=None, **kwargs):
    """
    lmfit model function.
    returns the simulated signal.
    """
    # Update half-lives.
    for nuclide in self.nuclides:
        key = f"half_life_{nuclide.name}"
        if nuclide.vary_half_life and key in kwargs:
            nuclide.half_life = kwargs[key]
            nuclide.decay_constant = np.log(2) / nuclide.half_life

    # For each nuclide, update free branching ratios.
    for nuclide in self.nuclides:
        # Identify free branches and sum fixed branchings.
        free_keys = []
        fixed_sum = 0.0
        for branch in nuclide.decay_branches:

```

```

        # if branch.daughter != 'Stable':
        if branch.vary_branching:
            key = f"p_{nuclide.name}_to_{branch.get_daughter_name()}"
            free_keys.append(key)
        else:
            fixed_sum += branch.branching_ratio
    # Available branching fraction for free branches.
    available = 1.0 - fixed_sum
    # Compute total weight.
    if free_keys:
        weight_sum = sum(kwargs.get(key, 1.0) for key in free_keys)
    else:
        weight_sum = False
    # Update each free branch with normalized value.
    for branch in nuclide.decay_branches:
        # if branch.daughter != 'Stable' and branch.vary_branching:
        if branch.vary_branching:
            key = f"p_{nuclide.name}_to_{branch.get_daughter_name()}"
            if weight_sum:
                branch.branching_ratio = kwargs[key] / weight_sum * available
                # print(branch.branching_ratio)
            else:
                branch.branching_ratio = available / len(free_keys)

    for nuclide in self.nuclides:
        if nuclide.source_func_fit_params:
            for key in nuclide.source_func_fit_params:
                nuclide.source_func_fit_params[key] = kwargs[key+f"_{nuclide.name}"]

    for nuclide in self.nuclides:
        if nuclide.vary_initial_population:
            key = f"initial_population_{nuclide.name}"
            nuclide.initial_population = kwargs[key]

    if self.vary_background:
        self.background = kwargs['background']
    # Build and solve the decay model with the updated chain.
    decay_model = DecayModel(self.nuclides, background=self.background)
    return decay_model.signal(time, integration_method)

def fit(self, time, observed_signal, integration_method=None, **kwargs):
    """
    Build the lmfit model, automatically build parameters, and
    perform the fit.
    Returns the lmfit result.
    """
    model = Model(self.model_func, independent_vars=['time'],
                  nan_policy='propagate', integration_method=integration_method)
    params = self.build_params()
    result = model.fit(observed_signal, params, time=time, **kwargs)
    return result

```

Code B.2: Example decay curve using the decay curve library.

```

T_on = 60
T_off = 30
def source(t,R=50):
    cycle_time = T_on + T_off
    t_mod = (t) % cycle_time
    if t_mod < T_on:
        return R
    else:
        return 0

#defining a true model parent and a fitting parent with a deliberately
# different half-life
parent_true = Nuclide("Parent", 30, 0,
                      vary_half_life=False, source_func=source)
parent = Nuclide("Parent", 60, 0,
                vary_half_life=True, source_func=source,
                source_func_fit_params={'R':50},
                vary_initial_population=True)
daughter = Nuclide("Daughter", 3.5 * 60, 0, vary_half_life=False)
isomer = Nuclide("Isomer", 8*60, 0, vary_half_life=False)

parent_true.decay_branches = [
    DecayBranch(daughter, 1/3, vary_branching=False),
    DecayBranch(isomer, 1/3, vary_branching=False),
    DecayBranch("Stable", 1/3, vary_branching=False)
]

#suppose there is an unknown branching between an isomeric state and
#some decay to a stable isotope (maybe through alpha decay)
parent.decay_branches = [
    DecayBranch(daughter, 1/3, vary_branching=False),
    DecayBranch(isomer, 1/3, vary_branching=True),
    DecayBranch("Stable", 1/3, vary_branching=True)
]
# For Isomer, fix its branch.
isomer.decay_branches = [
    DecayBranch(daughter, 0.5, vary_branching=False, detection_efficiency=0),
    DecayBranch("Stable", 0.5, vary_branching=False, detection_efficiency=1)
]
daughter.decay_branches = [
    DecayBranch("Stable", 1, vary_branching=False, detection_efficiency=1)
]

nuclides = [parent, daughter, isomer]
nuclides_true = [parent_true, daughter, isomer]
background = 20
time = np.linspace(0, 500, 1000)

# Generate synthetic observed data with realistic error.

```

```

true_model = DecayModel(nuclides_true, background=background)
np.random.seed(0)
observed_signal = np.random.poisson(true_model.signal(time))

# Create a DecayChainFitter instance.
fitter = DecayChainFitter(nuclides, background=background,
                          vary_background=True)

# Perform the fit.
result = fitter.fit(time, observed_signal, weights=1/(observed_signal**0.5))
print(result.fit_report())

# Plot observed data and fitted curve.
plt.figure(figsize=(10, 6))
plt.errorbar(time, observed_signal, yerr=observed_signal**0.5,
             label='Simulated_Data', fmt='o', color='k', zorder=1)
plt.plot(time, result.best_fit, label='Fitted_Curve', linestyle='—',
         color='r')
plt.title('Fit_of_Decay_Chain_Parameters')
plt.xlabel('Time')
plt.ylabel('Signal_Intensity')
plt.legend(loc='upper_left')
plt.grid()
plt.show()

```

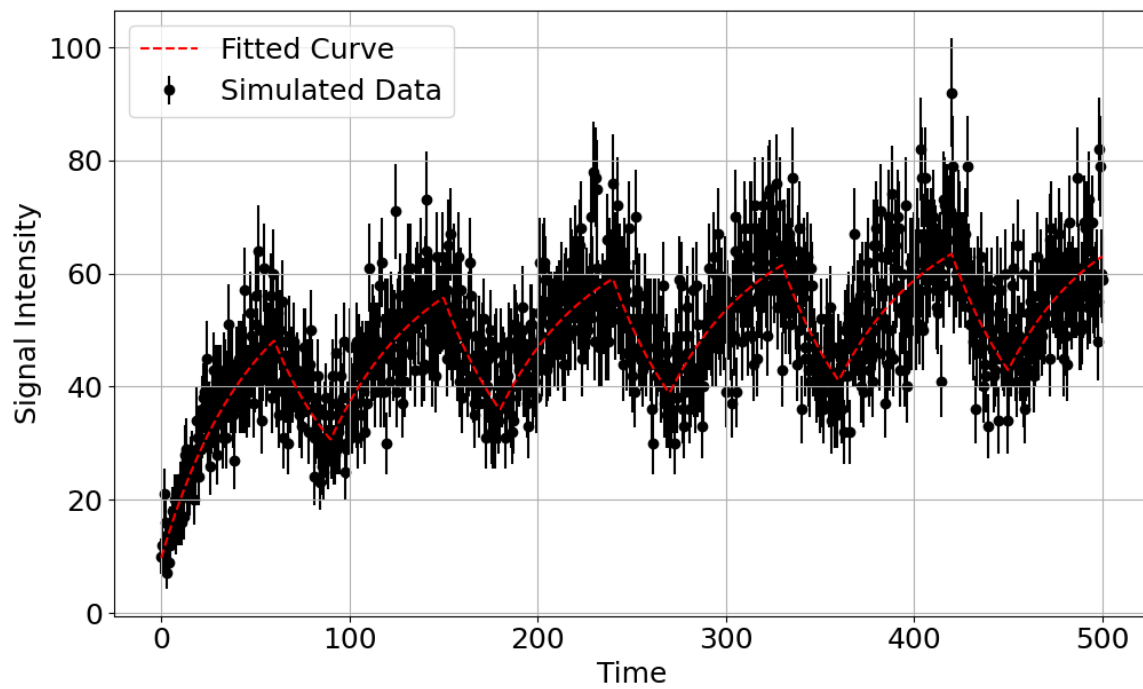


Figure B.1: Output of example decay curve python script.

APPENDIX C

Likelihood Ratio Test

The likelihood ratio test is designed to test the fit of a model to a simpler subset of that model. The subset model is labelled as the null hypothesis, and the output of the test is loosely speaking a probability of the data obtained being a result of the simpler model given the sampling uncertainty. The test compares the ratio of likelihoods of the data given the full model or the subset model. We can define the likelihood of the data given a model as a simple product of the probability of each data point, assuming that the chosen model is true. This is given as

$$\mathcal{L} = \prod_{i=1}^n p(x_i), \quad (\text{C.1})$$

where $p(x_i)$ is the probability of observation x_i . It is clear that an understanding of the uncertainty distribution for each data point is required to obtain accurate probabilities.

The likelihood ratio tests applied in this thesis were applied on count data. We can then model the distribution of expected data points as Poisson distributed around the model expectation. If we label the model expectation at each observation x_i as $f(x_i)$ and the observed value as $O(x_i)$ then we have

$$p(x_i) = \frac{f(x_i)^{O(x_i)} e^{-f(x_i)}}{O(x_i)!}. \quad (\text{C.2})$$

It is convenient at this point to work with the log of the likelihood function in order to change the product into a sum. The log-likelihood for the model f with Poisson errors¹ is

$$\ln \mathcal{L} = \sum \ln \frac{f(x_i)^{O(x_i)} e^{-f(x_i)}}{O(x_i)!} \quad (\text{C.3})$$

$$= \sum O(x_i) \ln f(x_i) - f(x_i) + \ln(O(x_i)!). \quad (\text{C.4})$$

The test statistic for the likelihood ratio test is twice the logarithm of the ratio of the likelihoods of the complete and subset models.² This can be written in terms of log-likelihoods as

$$\lambda_{LR} = 2(\ln \mathcal{L} - \ln \mathcal{L}_0), \quad (\text{C.5})$$

where $\mathcal{L}$ is the log-likelihood full model and $\mathcal{L}_0$ is the log-likelihood of the null hypothesis. The test statistic λ_{LR} approaches a χ^2 distribution with degrees of

¹If we take the data to have a normally distributed error around the model ($p(x_i) = \sqrt{2\pi\sigma^2} \exp(-(O(x_i) - f(x_i))^2/2\sigma^2)$), then the log-likelihood function obtained is $-\frac{n}{2} \ln(2\pi\sigma^2) - \frac{1}{2\sigma^2} \sum (O(x_i) - f(x_i))^2$. For count data one can approximate the correct Poisson distributed log-likelihood expression by setting $\sigma = \sqrt{O(x_i)}$. The algebraic details of the deriving the expression for log-likelihood in the normally distributed errors case is left as an exercise for the examiners.

²The physicists intuition to apply Stirling's approximation as soon as a factorial appears in a logarithm is wrong in eqn. C.4. We care most about the the low count observations where the error differs substantially from $\sqrt{O(x_i)}$. Luckily no damage is done with such an approximation, as the term exactly cancels in the test statistic.

freedom (k) equal to the difference in number of parameters between the two models, for a large number of observations. Therefore we can use the cumulative distribution function ($F(\chi_k^2(\lambda_{LR}))$) of the χ^2 distribution to reject the null hypothesis with a significance $\alpha = 1 - F(\chi_k^2(\lambda_{LR}))$.