Eur. Phys. J. A (2024) 60:87
https://doi.org/10.1140/epja/s10050-024-01241-6

THE EUROPEAN PHYSICAL JOURNAL A

Regular Article - Experimental Physics

15 years of precision mass measurements at TITAN

Anna A. Kwiatkowski[2,1,a], **Jens Dilling**[3,4,1,b], **Stephan Malbrunot-Ettenauer**[1,5,c], **Moritz Pascal Reiter**[6,d]

[1] Science Division, TRIUMF, 4004 Wesbrook Mall, Vancouver, BC V6A 2K3, Canada
[2] Department of Physics and Astronomy, University of Victoria, 3800 Finnerty Road, Victoria, BC V8O 5C2, Canada
[3] Oak Ridge National Lab., ORNL, 1 Bethel Valley Road, Oak Ridge, TN 37830, USA
[4] Department of Physics, Duke University, 120 Science Street, Durham, NC 27708, USA
[5] Department of Physics, University of Toronto, 60 St. George St., Toronto, ON M5S 1A7, Canada
[6] School of Physics and Astronomy, The University of Edinburgh, Peter Guthrie Tait Road, Edinburgh, Scotland EH9 3FD, UK

Received: 23 July 2023 / Accepted: 9 January 2024 / Published online: 17 April 2024

Communicated by Nicolas Alamanos

Abstract Atomic masses represent key ingredients to understand the structure of atomic nuclei. In particular, they provide insights into the nuclear binding energy and, thus, into the combined forces that govern the stability of atomic nuclei. Given their high experimental precision and accuracy, these observables serve as stringent benchmarks for modern nuclear theory and are critical input for nuclear astrophysics and tests of fundamental symmetries. Here, we review the current status of precision atomic mass measurements, with a focus on short-lived radioactive species and relevant techniques employed at TRIUMF's Ion Trap for Atomic and Nuclear science (TITAN). Coupled to the ISAC facility, TITAN has been in operation since 2007. Over the years, it has evolved in its capabilities, taking advantage of its unique combination of ion traps, including Paul traps, an electron beam ion trap (EBIT), a Penning trap, and most recently, an electrostatic multiple-reflection time-of-flight (MR-TOF) system.

1 Introduction

Mass measurements have since long been recognised as key contributions to our understanding of nuclear structure. The obtained atomic masses and resulting nuclear binding energies provide unique information about the combined forces that govern atomic nuclei. Historically, they played a decisive role in the discovery of magic numbers which ultimately led to the formulation of the nuclear shell model [1].

A major shift in the field of mass spectrometry occurred with the advent of radioactive ion beams (RIB) which became available in sufficient quality and quantity to extend direct mass measurements also to short-lived radionuclides enabling the use of atomic-physics based precision measurements [2]. Today, a wide variety of instrumentation is available to produce 'exotic' radioactive isotopes, and correspondingly, a broad range of specialized techniques for direct mass measurement methods has been developed. These methods differ in their experimental performance characteristics such as achievable precision, accuracy, speed, and ability to suppress undesired contamination. Common to all these methods is their capability to determine atomic masses of radioactive isotopes by coupling the mass measurement system to a RIB facility, typically driven by a particle accelerator. The most common types of isotope production facilities, are (1) isotope-separator on-line (ISOL), using spallation, fragmentation, or fission reaction of a heavy target nucleus, (2) in-flight separation, using fragmentation processes at or near relativistic projectile energies, or (3) fusion-evaporation and in-flight separators at low energies [3].

The field of precision mass measurements of short-lived radionuclides was pioneered by ISOLTRAP at ISOLDE/CERN [4], a Penning-trap mass spectrometer which started operation in the late 1980s [5]. Its ground-breaking concept of coupling an ion trap, which was originally designed for off-line atomic-physics experiments, with an accelerator facility for carrying out nuclear-physics motivated mass spectrometry laid the foundation for a flourishing research landscape today [6]. As a result, unique ion-trap systems have been developed for mass measurements at accelerators or nuclear

[a] e-mail: AniaK@triumf.ca
[b] e-mail: DillingJ@ornl.gov (corresponding author)
[c] e-mail: sette@triumf.ca
[d] e-mail: mreiter@ed.ac.uk

reactors, each optimized to the respective production site of short-lived radioactive isotopes. At ISOL facilities, there are currently the following systems operational (o) or planned (p): ISOLTRAP (o) [7] at ISOLDE-CERN, TITAN (o) [8] at ISAC-TRIUMF, PIPERADE (p) for DESIR-GANIL [9], [10] as well as Penning traps at quasi ISOL-systems, i.e. JYFL-TRAP(o) at the IGISOL-facility at Jyväskylä-University [11], TAMUTRAP (o) at the cyclotron facility at Texas A&M [12], TRIGA-TRAP (o) at the research reactor in Mainz [13] and the CPT system (o) at the CARIBU facility at Argonne national Lab [14]. At in-flight facilities: the LEBIT-system (o) at the NSCL/FRIB [15], HITRAP (p) at FRS-GSI [16], MATS (p) at FAIR S-FRS [17]. At fusion-evaporation facilities: SHIPTRAP (o) at SHIP-GSI [18] and the Lanzhou Penning Trap (LPT) at the heavy ion accelerator facility in Lanzhou-China (p) [19,20].

In recent years, the field of low-energy, ion-trap based precision mass measurements has been extended by new technical developments, specifically by the advent of electrostatic ion traps operated as multi-reflection time-of-flight (MR-ToF) mass spectrometers. These devices are nowadays integrated in many of the aforementioned ion-trap systems. Some (newer) programs on mass spectrometry at RIB facilities rely entirely on MR-ToF devices, for example at the RIBF facility at RIKEN (o) [21,22], at the FRS Ion Catcher at GSI (o) [23], the JINR-facility in Dubna (p) [24] or the NEXT facility at the Groningen AGOR facility (p) [25].

As a common feature, ion-trap systems for mass measurement at RIB facility consist of multiple components connected in series, such as various instruments for ion manipulation and preparation prior to the actual mass spectrometer. The purpose of these preparation devices is to match the ion beams delivered by the production facility to the requirements of the mass spectrometer. This may involve, the adjustment and control of the ions' kinetic energy, the reduction of the energy spread, the conversion of a continuous beam into a well-defined ion bunch, the purification from contaminants in the RIB, or other ion manipulation to enhance the performance of the mass measurement.

Preparation and manipulation devices include linear Paul traps, which capture the ion beam as delivered by the RIB facility. These Paul traps usually employ buffer gas cooling – using inert gases such as hydrogen or helium – as an effective and efficient scheme to obtain cold ion samples universally applicable to all chemical elements.

The next step may exploit further separation techniques to provide a cleaner, ideally mono-species sample to the measurement apparatus. Methods for this ion beam purification include the use of gas-filled Penning traps or MR-ToF instruments.

An additional preparation technique to enhance mass measurements in Penning traps is found in charge-breeding, i.e. the removal of additional electrons from the initially singly-charged isotopes. This is currently implemented at two on-line systems; HITRAP which provides access to long-lived species (beyond seconds in half-life) and TITAN, capable to also work with very short-lived species.

The experimenters' choice for specific instruments for preparation and mass measurement depends on the production facility, its beam characteristics in terms of energy and purity, as well as on the yields and half-lives of the ion species to be investigated by the mass spectrometer. In the following we discuss TRIUMF's Ion Trap for Atomic and Nuclear science (TITAN), an ion trap system coupled to the ISAC (isotope seperator and accelerator) facility [26] at TRIUMF.

1.1 TITAN at TRIUMF

The ISAC facility at TRIUMF (Vancouver, Canada) is currently the world's highest-power RIB facility of the ISOL type with production capabilities up to 100 kW [27]. It excels in the production of a wide range of radionuclides facilitated by extensive developments in target materials, ion sources, and driver beam. The produced elements range from helium to uranium [28]. New developments include the rotating driver proton beam on the ISOL target [29], proton beam currents exceeding 10 μA on UCx targets [30], and a proton-to-neutron-converter target for neutron-induced fission of UCx targets. The availability of rare isotopes and associated capabilities in mass separation techniques will be further enhanced by TRIUMF's new Advanced Rare IsotopE Laboratory (ARIEL) [31], which is currently under construction. ARIEL will include two independent production beam lines, one based on electrons and one based on protons. With this ARIEL will provide unprecedented intensities of clean, neutron-rich nuclides, produced by electron-driven photon-fission of actinide targets. The 30 MeV, 200 kW superconducting electron linear accelerator will be supplemented with an additional proton driver such that ARIEL will provide two more RIB production facilities to ISAC experimenters, ultimately having three independent isotope production lines. The RIBs from ARIEL will be separated using the isotope separator system CANREB (CANadian Rare-isotope separator facility with Electron Beam ion source) [32], capable of high mass resolving powers reaching $R = m/\Delta m = 20,000$. The combined operation of ARIEL, ISAC, and CANREB will enable delivery of three simultaneous RIBs to experiments. CANREB was commissioned in December 2020 [33], while ARIEL is expected to come on-line 2025 for electron-driven photo-fission beams and soon after for proton-induced beams.

Within the suite of state-of-the-art experimental stations hosted by ISAC, TRIUMF's Ion Trap for Atomic and Nuclear science (TITAN) is dedicated to high-precision mass spectrometry of rare and short-lived isotopes as well as in-trap decay spectroscopy. Initiated by the commissioning of its

Penning trap mass spectrometer MPET, TITAN's ion trap system has been in operation since 2007. It distinguishes itself from other online Penning-trap facilities by its ability to employ fast measurement cycles and by its capability to increase the charge state of the analyzed ions by charge breeding in an electron ion beam trap (EBIT). Mass measurements can thus be achieved at high precision and accuracy also for isotopes with very short half-lives. For example, ^{11}Li with $T_{1/2} = 9$ ms remains a world record as the shortest-lived radionuclide ever characterized in Penning trap mass spectrometer [34]. Its diverse combination of state-of-the-art ion traps, including the recent addition of a multiple-reflection time-of-flight (MR-TOF) mass spectrometer [35], coupled to the intense isotope production sources available at ISAC and ARIEL makes TITAN a unique facility in the world.

1.2 TITAN setup

A schematic and a photo of the TITAN facility are shown in Fig. 1. The system consists of the following main components:

RF cooler and buncher: the ion beam delivered from ISAC is accumulated in a radio-frequency quadrupole cooler and buncher (RFCB) [36]. The RFCB is a helium buffer-gas-filled preparation trap which allows cooling of the 'hot' incoming ion beams. The RFCB prepares and delivers well defined ion bunches with small emittance to the other ion traps at TITAN. In addition to ISAC beams, the RFCB can receive stable beams from a surface ion source installed directly underneath the RFCB injection beamline.

EBIT: an electron beam ion trap (EBIT) is used as an ion charge breeder, removing additional electrons from the injected ions through electron impact ionization and thus increasing the charge state of the ion population. The TITAN EBIT [37] is designed to operate with an intense electron beam with electron energies of up to $E_{kin} = 60$ keV which is sufficient to reach a helium-like configuration for species up to $Z = 70$. Employing ions at high charge states boosts precision and resolving power of mass measurements in Penning traps. Since the additional charge-breeding step can typically be completed in a period shorter than a nuclide's half-life, this approach is particularly powerful for shorter-lived radionuclides in which the measurement time in the Penning trap and thus attainable precision with single charge ions is limited by the lifetime until a nuclide's radioactive decay.

MR-TOF-MS: a multiple-reflection time-of-flight mass separator (or spectrometer) uses two opposing electrostatic ion-optical mirrors to guide charged particles back and forth, often for an accumulative flight path of multiple kilometers, while maintaining a compact instrument design. Facilitated by this elongated flight path one achieves spatial separation of various ion species, which in return allows for high resolution isobar separation or precision mass measurements. See Sect. 1.4 for more details.

Penning trap mass spectrometer: a Penning trap combines a strong homogeneous magnetic field ($B = 3.7$ T in the case of TITAN [38]) and a weak quadrupolar electrostatic field, which enables ion confinement in three dimensions. Various techniques can be employed in order to determine the ion's cyclotron frequency which links the movement of the ion species in the electromagnetic field to the mass over charge ratio of the trapped particle.

1.3 The TITAN Penning trap system

Ion traps provide unique confinement capabilities for charged particles leading to ample opportunities for measurements in a well-controlled and stable environment. As a consequence, systematic errors in the determination of atomic masses are kept well under control, which translates into a superb accuracy of mass spectrometers based on ion traps.

In particular, Penning traps have been exploited for mass measurements at accelerator facilities since many decades [39]. Although the basic trapping mechanism remained the same, the techniques to determine an ion's cyclotron frequency and thus its mass have evolved over the years to complementary approaches. These involve the methods of (1) time-of-flight ion cyclotron resonance (TOF-ICR) [40], (2) Fourier-transform ion cyclotron resonance (FT-ICR) [41], and – most recently added – (3) phase-imaging ion cyclotron resonance (PI-ICR) [42].

The TITAN measurement penning trap (MPET) [38] is a hyperbolic shaped Penning trap mass spectrometer designed and optimized to provide fast measurement cycles. Moreover, it is the only online Penning trap system in the world able to perform mass spectrometry of short-lived, highly-charged ions.

In order to facilitate fast measurement cycles, the ions are directly injected from the RFCB into MPET, without a separate gas-filled preparation Penning trap, whose absence further allows direct measurements of highly charged ions. Moreover, TITAN takes advantage of so-called Lorentz steerers [43] installed directly in front of MPET. This enables fast ion preparation during the ion-injection process compared to a more time-consuming excitation of the ion motion in the trap itself. MPET currently employs the TOF-ICR method and can be operated for mass measurements at a precision and accuracy as good as one part-per-billion.

A key element at TITAN is its ability to measure isotopes in ionic states higher than $q = 1+$ by changing the ions' charge state in the TITAN EBIT charge breeder prior to the mass measurement. In order to fully capitalize on the resolution and precision gains provided when operating with highly charged ions (HCIs) it is important to understand the various factors involved.

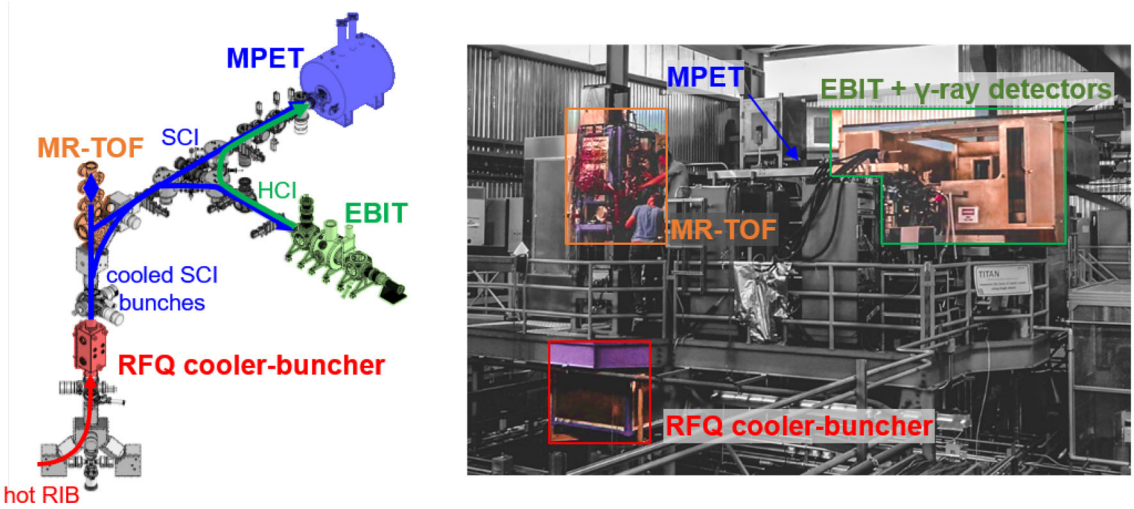

Fig. 1 TITAN's ion-trap system shown as a schematics (left) and picture (right). A continuous ion beam from ISAC or the TITAN offline ion source is injected into the RFQ cooler buncher for ion beam preparation. Ion bunches are subsequently transferred to the MR-TOF device, EBIT, and/or measurement Penning trap (MPET). See text for details

The major gain by using HCIs arises from the fact that the cyclotron frequency

$$\nu_c = \frac{q}{m} \cdot \frac{B}{2\pi} \tag{1}$$

is directly proportional to the charge state q, with B representing the magnetic field strength, and m the mass of the ion. The resolution $\Delta\nu_c$ expressed in absolute frequency remains however unaffected by the charge state. Instead, it is governed by the Fourier limit $\Delta\nu_c \propto 1/T_{RF}$ where T_{RF} is the RF-excitation time in the Penning trap [44]. In a mass measurement the magnetic field is typically determined via a measurement of the cyclotron frequency $\nu_{c,ref}$ of a reference ion with well known mass m_{ref}. The unknown mass m of the ion of interest can than be determined via the mass ratio

$$\frac{m}{m_{ref}} = \frac{\nu_{c,ref}}{\nu_c}. \tag{2}$$

The attainable mass resolving power R is proportional to $\nu_c \cdot T_{RF}$ and can thus be expressed as

$$R = \frac{m}{\Delta m} = \frac{\nu_c}{\Delta\nu_c} \propto \frac{qBT_{RF}}{m}, \tag{3}$$

which illustrates that large gains in resolution and consequently measurement precision can be obtained with HCI in cases where a radionuclide's half-life limits the excitation time T_{RF} [45].

The increase in mass resolving power enabled by HCI for Penning-trap mass spectrometry of short-lived radionuclides can be either exploited to improve the measurement precision, to reduce required measurement time per ion species, or to resolve close lying isobaric ion species present in the RIB from ISAC/ARIEL [46], including nuclear isomers. As an example, this is shown for the case of ^{126}In and its 90 keV isomeric state in Fig. 2 (top) which shows ^{126}In in charge state $q = 13+$ configuration.

The achievable statistical measurement precision $\delta m/m$ scales with the detected number of ions N_{ions} and the mass resolving power according to

$$\frac{\delta m}{m} \propto \frac{1}{R\sqrt{(N)}} \propto \frac{m}{qBT_{RF}\sqrt{(N)}}. \tag{4}$$

As such, the optimal excitation time for best precision is $T_{RF} = 2T_{1/2}/\ln 2 \approx 2.9T_{1/2}$. If T_{RF} is extended beyond this point, radioactive decay losses become dominant, statistics is reduced too much, and the measurement precision is compromised.

Equation 4 represents the idealised boost in precision by a factor identical to the charge state q but assumes that the charge breeding proceeds without losses and is fast compared to the radionuclide's half-life, $T_{1/2} >> T_{cb}$, with T_{cb} being the charge breeding time. The actual improvement in precision is described by an effective gain factor G_{HCI} according to

$$G_{HCI} = q \times \sqrt{2^{-T_{cb}/T_{1/2}} \cdot \eta_{pop}(q, T_{cb})} \leq q \tag{5}$$

which also reflects losses due to radioactive decay during charge breeding and the achievable population of the desired charge state η_{pop} [47]. For given electron beam energies, currents and densities in the EBIT, the population factor η_{pop} depends on the charge state and on the charge breeding time T_{cb}. It is typically evaluated in simulation and then determined experimentally through optimisation. Note however that Penning-trap mass measurements are carried out with

only one or very few ions in the trap at any one time to eliminate space-charge effects. Thus, the effective gain G_{HCI} remains equal to q, as long as the radionuclide's production yield and the efficiency of the ion trap system allows for one ion to be studied in the MPET per measurement cycle.

Additionally, other mechanisms of ion losses or perturbations may have to be taking into account. These include the vacuum conditions in the Penning trap, in particular vacuum pressure and residual gas composition. Interactions with residual gas may lead to charge exchange and charge state losses due to electron transfer. HCI ions are more likely to be effected given the increased cross section [48] as well as their higher cyclotron frequency and thus velocity inside the trap, probing a larger 'volume' per unit of time. Studies have shown, that the vacuum inside the trapping region of the MPET is $p_{trap} = 3 \times 10^{-9}$ to 1×10^{-11} torr [49]. The ideal vacuum to maintain high charge states is below 1×10^{-12} torr, when assuming a natural distribution of H, N, and O species in the trap. Based on these requirements, TITAN's room-temperature MPET, successfully operated for about one decade, is currently being replaced by a new cryogenic version held at an operational temperature of $T_{op} \approx 20\,\mathrm{K}$, see Sect. 3 which will provide a significant improvement in vacuum conditions and hence boost MPETs performance beyond current limitations.

1.4 The TITAN MR-TOF mass measurement system

In 2017, the TITAN facility has been upgraded with a multiple-reflection time-of-flight mass spectrometer (MR-TOF-MS) [50,51]. The device consists of a beam transport system based on a helium gas-filled RFQ and a vacuum time-of-flight analyzer.

Mass measurements rely on the time-of-flight technique relating mass m and charge q of an ion to its time-of-flight t_{tof} required to travel a certain flight path l in the electric potential $U(l)$:

$$qU(l) = \frac{1}{2}m(v(l))^2 \rightarrow t_{\mathrm{tof}} = \int_l \frac{dl}{\sqrt{\frac{2q}{m}U(l)}} \tag{6}$$

$$\rightarrow \frac{m}{q} = \frac{2U(l)}{l^2}t_{\mathrm{tof}}^2. \tag{7}$$

Due to signal propagation times and electronic delays the real time of flight t_{tof} is shifted from the experimentally measured time of flight t_{exp}, resulting in a calibration function of

$$\frac{m}{q} = a(t_{\mathrm{exp}} - t_0)^2 \tag{8}$$

with parameter a being a device-specific calibration parameter, which depends on the kinetic energy of the ions as well as the effective path length given by the geometrical path and the voltages applied to the electrode system. The time offset t_0 can be measured offline from a measurement of the time-of-flight and known mass-to-charge of stable calibration species [52].

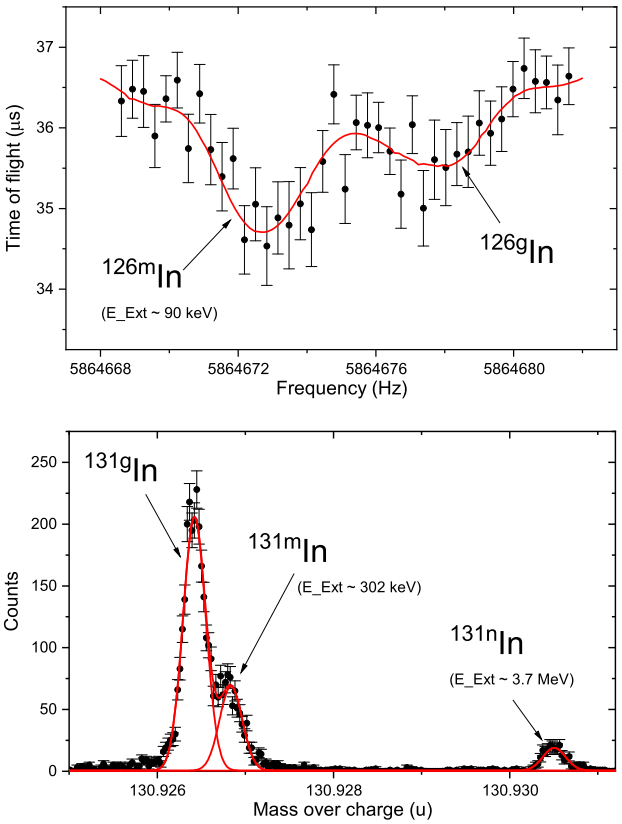

Fig. 2 (Top) MPET Time of flight resonance of $^{126g+m}In^{13+}$ using a 400 ms excitation time. The red lines shows a fit with the theoretical line shape taking a convolution of two species into account. The isomer at 90 keV can be well resolved with a mass resolving power of about 2,600,000. (bottom) MR-TOF-MS time of flight spectrum showing beam at mass number 131. The red line shows a fit with a HyperEMG function. The isomeric and ground states in ^{131}In can be resolved after a time of flight of 15.4 ms (685 turns) reaching a mass resolving power of about 450,000

The mass resolving power can be linked to the total time of flight t_{tof} and the full width half maximum (FWHM) of the time-of-flight distribution Δt_{tof} as

$$R = \frac{m}{\Delta m} = \frac{t_{\mathrm{tof}}}{2\Delta t_{\mathrm{tof}}}. \tag{9}$$

An elegant way to increase the resolving and separation power of the time-of-flight analyzer is to extend the flight path by trapping ions for multiple reflections between two electrostatic isochronous ion mirrors [53]. The total time of flight t_{tof} can be written as the sum of the time of flight from the initial ion trap to the detector t_{initial} and I times the time of flight of one turn t_{turn} resulting in $t_{\mathrm{tof}} = t_{\mathrm{initial}} + I \cdot t_{\mathrm{turn}}$. In an ideal system, the ion optical mirrors completely preserve the isochronicity and thus the time spread of the initial ion bunch $\Delta t_{\mathrm{inital}}$ does not increase with each reflection. However, in

a real time-of-flight mirror system ion optical aberrations cause some degree of additional broadening per turn Δt_{turn}. The mass resolving power of such a multiple turn system can be rewritten as

$$R = \frac{t_{\text{initial}} + I \cdot t_{\text{turn}}}{2\sqrt{\Delta t_{\text{initial}}^2 + (I \cdot \Delta t_{\text{turn}})^2}}, \tag{10}$$

and it can be shown that at high number of turns the mass resolving power becomes limited by aberrations (including instabilities in the mirror potentials). Resolving powers on the order of $\frac{m}{\Delta m}$ of $\sim 10^5$ can be achieved after about 2 ms of time of flight e.g. for ^{85}Rb [51]. The statistical precision of such a mass measurement can be derived similar as discussed above and is proportional to $1/(R\sqrt{(N)})$.

The TITAN MR-TOF-MS operates in three distinct operation modes [51], being:

- Mass measurement mode: After a certain number of turns ions are guided to a fast time-of-flight detector. There, their time-of-flight and thus their mass-over-charge ratio is measured. Being a non-scanning mass measurement technique, each individual measurement cycle allows for a determination and characterization of the composition of the RIB. Therefore unwanted isobaric as well as isomeric contaminations can be detected, identified and quantified prior to a possible isobar separation. This feature has established the TITAN MR-TOF as useful device for yield measurements and supporting beam development [54]. An example time-of-flight spectrum in mass measurement mode is shown in Fig. 2 (bottom) for ^{131}In and its two isomeric states at 302 keV and 3.7 MeV.
- Isobar separation mode: Compared to other MR-TOF-MS, which rely on either Bradbury–Nielsen gates [55–58], selective switching of an ion mirror [59] or drift tube [60], the TITAN MR-TOF-MS is the only online system employing mass-selective re-trapping [61]. In this mode, ions are guided back towards the initial RF injection trap where the ions-of-interest are selectively re-trapped by switching the injection trap potentials from a retarding potential to a trapping potential [61]. Choosing the correct moment in time captures only the ions of interest, while ions with a different mass-to-charge ratios arrive at different times and are discarded. Using this operation mode the MR-TOF-MS can provide isobarically purified beams for all experiments at TITAN.
- Combined isobar separation and mass measurement mode: Combining both operation modes the TITAN MR-TOF-MS can uniquely act as its own high-resolution, high-throughput isobar separator. This allows additional background suppression and increases the dynamic range of the measurement within the same device by several orders of magnitude.

1.5 Performance comparison

Both mass spectrometers at TITAN are optimized to ensure fast measurement cycles. The combination of a high-throughput MR-TOF-MS with the fast MPET spectrometer gives access to the shortest-lived and rarest isotopes produced at TRIUMF. A key challenge working with beams produced via the ISOL method is the need to resolve ions of interest from contaminant species, which are co-produced in proton-induced reactions due to the non-differential production channels of spallation, fragmentation, fission acting at the same time. As discussed above, for both spectrometers the achievable resolving power depends on the mass-over-charge ratio of the species of interest and the measurement time; time-of-flight within the electrostatic analyzer in case of MR-TOF-MS, see Eq. 10, and excitation time in case of MPET, see Eq. 3.

An overview of the achievable performance for different ions of interest is given in Fig. 3 along with representative values of resolving powers achieved during online experiments.

The TITAN MR-TOF-MS excels at measuring short-lived isotopes and has been shown capable of high mass resolving powers [51,62]. The device benefits from its internal dedicated injection trap producing narrow ion bunches matched to the acceptance of its time of flight analyzer. As such, e.g. a resolving power of 470,000 FWHM is reached already after 10 ms time of flight for ^{85}Rb$^+$ ions, as shown in Fig. 3. At long flight times, beyond 15 ms, the resolving power is dominated by ion optical aberrations and instabilities in the analyzer power supplies, which currently limit the mass resolving power to about half a million. Masses can than be determined from the time of flight spectra with a typical precision $\delta m/m$ on the order of a few 10^{-7}.

The measurement penning trap MPET has shown its capabilities to measure short-lived isotopes [34], but particularly excels at performing mass measurements with highest precision and accuracy. Up to a few millions the resolving power is not limited by field imperfections and only depends on the measurement time. As such, e.g. FWHM resolving powers of 400,000 and 1.6 million have been reached after 125 ms and 500 ms excitation times for ^{22}Mg$^+$ ions, respectively [63]. Uniquely, MPET can measure masses of highly charged ions, boosting its performance in terms of mass resolving power and precision. Using HCI, e.g. via a 13+ charge state, FWHM resolving powers of 1.2 and 2.8 million have been archived e.g. for ^{126}In^{13+} ions after 175 ms and 400 ms, respectively [64]. Based on the obtained resonance curves masses can then be obtained with relative precision $\delta m/m$ on the order of a few 10^{-9} to 10^{-8}.

As isotope production yields drop at RIB facilities when approaching the limits of nuclear binding, the resulting relative precision is often limited by the available statistics. The

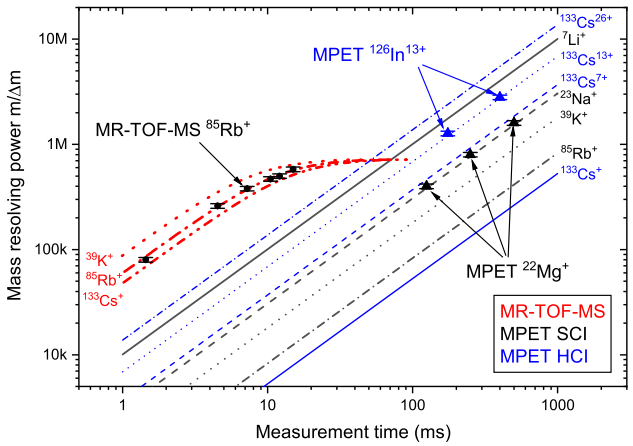

Fig. 3 Mass resolving power FWHM routinely reached with the spectrometers at TITAN for different mass-over-charge ratios. The lines show predictions for MPET and MR-TOF based on Eqs. 3 and 10, respectively. The symbols indicate representative resolving powers reached during online experiments

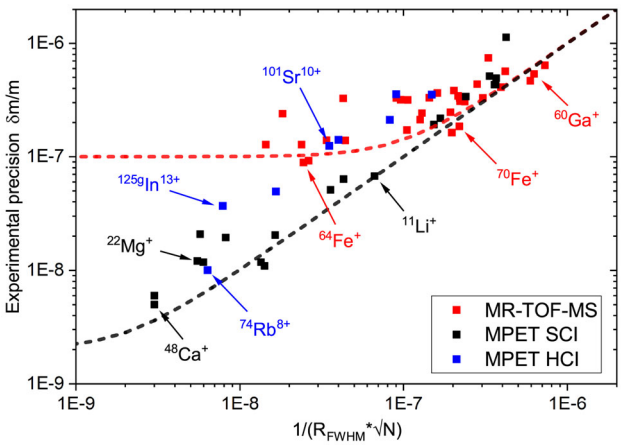

Fig. 4 Precision reached during MPET (SCI vs HCI) and MR-TOF-MS mass measurements in compassion to the respective expected precision based on mass resolving power and number of detected ions. The dashed lines show performance predictions assuming a perfect spectrometer with systematic uncertainties of 2×10^{-9} for MPET and 1×10^{-7} for MR-TOF-MS. A selection of different measurement results are shown

TITAN spectrometers have been fine tuned to cope with low production yields and successful high precision measurements have been performed with intensities as low as a few ions per hour (MR-TOF) and a few ions per second (MPET) delivered to the TITAN facility. On the other hand, even for high-statistics cases systematic uncertainties limit both spectrometers at some level. In Fig. 4 the experimental precision of a subset of published TITAN mass values is shown in comparison to the pure statistical precision given via $1/(R \cdot \sqrt{N})$. Measurements limited by the statistical precision lie close to the diagonal, whereas results limited by systematic effects diffuse off the diagonal. For the TITAN MR-TOF-MS systematic effects limit the precision to around $1 \cdot 10^{-7}$, where as for MPET systematic effects dominate on the few 10^{-9} level. Using HCI high precision mass measurements of the short-lived isotope ^{74}Rb have been performed, but more commonly HCI have been used to resolve close-lying nuclear isomeric states. In particular HCI mass measurements using high charge states are limited by vacuum quality and charge exchange which has limited their reach.

This comparison of the two mass spectrometers at TITAN illustrates their past, present and future areas of application. TITAN's MPET has been designed for fast and accurate mass measurements enabling the first Penning-trap measurements of halo nuclei as well as of other short-lived radionuclides in the light and mid-mass region, see Sects. 2.1 and 2.2. TITAN's focus on isotopes with short lifetimes has been further strengthened by the later advent of its MR-TOF-MS which facilitate even shorter measurement cycles at high mass resolving power. Thus, the MR-TOF device has become TITAN's primary instrument to address the 'shortest-lived', low-yield isotopes. However, given MR-TOF-MS limitations in ultimate accuracy, MPET remains the tool of choice – even for very short-lived radionuclides – in case the physics demands for higher measurement precision and accuracy.

TITAN's unique program of online-charge breeding for Penning-trap mass spectrometry represents another significant development along this rationale. In order to further increase resolution and to utilise rare ions at MPET as effectively as in the MR-TOF-MS instrument, the frequency scanning of MPET's present TOF-ICR scheme will in the near future be complemented by the PI-ICR technique [42] in which every detected ion represents a measurement, analogously to MR-TOF mass spectrometry. TITAN's PI-ICR application will represent its first implementation with HCI [65].

2 TITAN mass measurements

Over the last 15 year, TITAN has pursued a rich program of mass measurements which has been addressing intriguing questions in nuclear structure and fundamental sciences. Results range from the light halo nuclei all the way up to the persistence of the $N = 82$ neutron shell closure towards the proton drip-line in heavy nuclei.

An overview of all published mass values from TITAN is given in Fig. 5. Since its commissioning in 2007 MPET has measured more than 86 isotopes, whereas the newly installed MR-TOF-MS was able to determine masses of 78 nuclei since 2017.

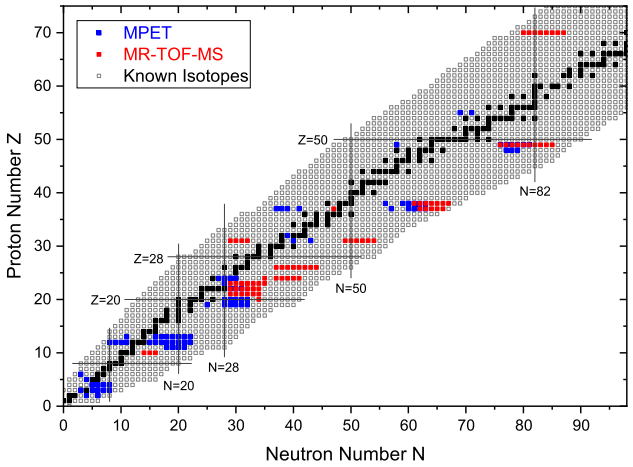

Fig. 5 Overview of all mass values published by TITAN since the initial commissioning until end of year 2022. Using the TITAN MPET and MR-TOF-MS spectrometers masses of 86 and 78 nuclei have been published, respectively

2.1 Mass measurements of neutron halo nuclei

Halo nuclei [66–68] are weakly bound nuclear systems which exhibit remarkably large matter radii. Qualitatively, this is understood by one or more loosely-bound valence nucleon(s) tunneling deeply into the classically forbidden region. Experimentally well-studied neutron halos are, for example, ^{6,8}He, ^{11}Li, or ^{11}Be.

Precision mass measurements of halo nuclei are important for two reasons. First, they provide the nucleon separation energies which evaluate the weak binding of a halo nucleus and, thus, characterize the large tail of the nuclear wave function. Second, because of neutron(s) orbiting far away from the nuclear core, neutron halos show a sizable difference between their matter and charge radii. The determination of the latter requires precise values of nuclear masses when they are extracted from optical isotope-shift measurements [2].

Representing a (small) difference in an atomic transition frequency ν when studying different isotopes of the same chemical element, the isotope shift $\delta\nu^{A',A} = \nu^{A} - \nu^{A'}$ between isotopes A and A' relate to the difference in their nuclear root-mean-square charge radii $\delta\langle r_c^2\rangle$ according to

$$\delta\nu^{A',A} = \nu_{MS}^{A',A} + K_{FS}\delta\left\langle r_c^2\right\rangle.$$

with $\nu_{MS}^{A',A}$ representing the mass shift and K_{FS} the field shift factor. For the lightest halo nuclei, both $\nu_{MS}^{A',A}$ and K_{FS} can be calculated with sufficient accuracy in high precision atomic theory [69]. However, experimental atomic masses are crucial input parameters in these calculations. Since the mass shift largely dominates over the field shift in these light systems, the accuracy and precision of the experimental input masses is imperative for the evaluation of the charge radii of a halo nuclei. Ideally, it should be $\delta m \lesssim 1$ keV corresponding to $\delta m/m \lesssim 1.3 \cdot 10^{-7}$ in ^{8}He for instance.

Building on its unique design for fast measurement cycles, TITAN's very first online campaigns at ISAC have hence been dedicated to direct Penning trap mass measurements of the halo nuclei ^{6,8}He [70,71], ^{11}Li [34], and ^{11}Be [72]. As far as experimental precision is concerned, the case of ^{8}He impressively demonstrates the strength of Penning-trap mass spectrometry at TITAN. Despite its short halflife of only $T_{1/2} = 119$ ms, a mass uncertainty of 14 parts-per-billion (ppb) could be achieved, i.e. a factor of about 70 better than the previously reported value [73]. For the even shorter-lived halo nucleus ^{11}Li ($T_{1/2} \approx 8.8$ ms) TITAN's measurement uncertainty remains as low as 62 ppb.

Note that a measurement's precision is only valuable if matched in its accuracy, too. For instance, the previously reported mass of ^{6}He with an uncertainty of $\delta m \approx 0.76$ keV was found to be off by 4 standard deviations, or 3.0 keV, compared to TITAN's direct Penning trap mass measurement. MPET's accuracy itself was carefully evaluated, including a series of benchmark measurements with stable nuclides of well known masses [38,74].

The improved experimental precision and accuracy on the halo masses as determined by TITAN have notable consequences also for the determination of their nuclear charge radii. In all cases, the atomic masses are essentially eliminated as a source of uncertainty when the charge radii R_c of ^{6,8}He, ^{11}Li, and ^{11}Be are extracted from isotope shifts obtained in high-resolution laser spectroscopy [75–77]. For instance, R_c of ^{8}He is corrected from 1.934(26) fm based on the mass value in Ref. [73] to 1.959(16) fm using the more accurate and precise TITAN mass dictum [71]. This represents a shift in R_c by one standard deviation and a reduction in uncertainty by $\sim 40\%$.

Masses and charge radii of halo nuclei studied at TITAN have played an important role in testing modern nuclear theory, especially ab-initio approaches which exploit nuclear forces based on chiral effective field theory. Both observables are extracted from experiment in a nuclear-model independent way and, thus, represent stringent benchmarks at high accuracy and high precision. The extended nuclear wavefunction of a halo nucleus often constitutes a theoretical challenge which can for instance be addressed by an appropriate choice of basis functions such as in the approach of effective interaction hyper-spherical harmonics [78].

Due to their low mass numbers, however, these halo nuclei can be theoretically modeled by few-body methods, e.g. by treating all individual nucleons inside the nucleus on equal footing. Thus, the comparison of experimental results and 'exact' theoretical calculations in light system also provides insights into the quality of the employed nuclear potentials. These also involve 3-body forces which have been shown to be essential ingredients to reproduce experimental observ-

ables of halo nuclei. In particular, a simultaneous calculation of 2-neutron separation energy and charge radius in ^{6}He only matches the experimental values based on TITAN's mass measurement when 3-body forces are considered [71].

2.2 Shell model tests and islands of inversion

At first glance, the nuclear shell model appears universally successful in describing all nuclides. Yet even its inventors, Goeppert–Mayer [1] and Jensen [79], suggested excursions from immutable shells in their Nobel Laureate lectures. The shell model was derived, among others, from observations of exceptional binding energy for filled shells and is a cornerstone of nuclear structure. Today a preponderance of experimental evidence from RIB facilities points to the evolution of these shells. Loss of magicity and Islands of Inversions are hallmarks of the evolution of nuclear shell structure far away from stability. Areas of interest have been identified where classical shells vanish and new shells appear. Modern theoretical frameworks have been adapted to understand and to predict these phenomena. TITAN led the high-precision cartography of the first Island of Inversion at $N = 20$ through measurements of Ne, Na, Mg, and Al isotopes [80–83]. The TITAN MPET campaign, in addition to tremendously improving the precision despite the short half-lives ($\leq$ 13 ms) [82] also found large deviations to reported literature values.

Another intriguing example of exotic shell formation has been the emergence of a neutron shell at $N = 32$ around ^{52}Ca [84], which was first predicted in self-consistent energy density functional calculations [85] and experimentally identified by measurements of first excited 2^+ states [86]. Within the picture of tensor-force-driven shell evolution it forms by a weakening of the attractive nucleon-nucleon force between the proton $\pi f_{7/2}$ and the neutron $\nu f_{5/2}$ orbitals due to reduced proton occupancy numbers in the $\pi f_{7/2}$ orbital [87]. TITAN has tackled this region by performing mass measurements of neutron-rich potassium to vanadium isotopes using both, Penning trap and MR-TOF systems, in combination with state-of-the-art ab initio shell model calculations [88]. Initial results helped to confirm the existence of the $N = 32$ shell closure in calcium [89].

Furthermore, subsequent findings precisely identified the emergence of the shell closure from vanadium [90], where no closed-shell signatures could be observed, to titanium [35,91], where weak signatures could be observed. Measurements of scandium isotopes [92] demonstrated a progressive enhancement of shell signatures across the mass surface, peaking at the double magic ^{52}Ca.

In recent work TITAN pushed toward the $N = 40$ Island of Inversion by performing mass measurements of Fe, Cr and Mn isotopes [62,93]. The measurements span across the island, as shown in Fig. 6, and in combination with VS-IMSRG(2) calculations could pin down ^{64}Cr as the summit of the $N = 40$ Island of Inversion.

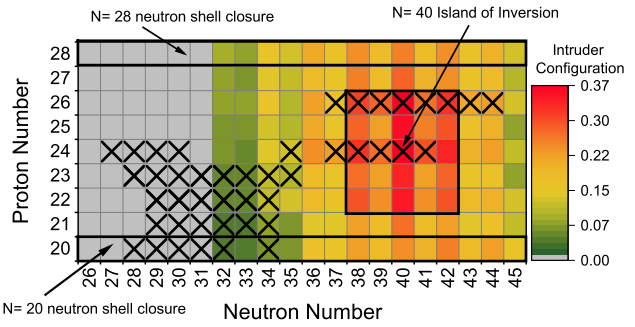

Fig. 6 Section of the nuclear chart around the $N = 40$ Island of Inversion between the $Z = 20$ and 28 proton shell closures. Theoretical calculations indicating the contribution to the ground-state wave function via VS-IMSRG(2) are shown in colour, for grey isotopes no calculations were performed. TITAN mass measurements are marked and played a key role in bench marking and testing the predictive power of the VS-IMSRG(2) calculations in the region. Data taken from [93]

The $N = 82$ neutron shell closure, forming due to the large energy gap between the $\nu h_{11/2}$ (or close by $\nu s_{1/2}$ and $\nu d_{3/2}$) and the $\nu f_{7/2}$ neutron orbital, has been under intense investigation. Its evolution has been an open question and particularly below $Z < 50$ has profound consequences for the modeling of explosive neutron-capture nucleosynthesis. TITAN has investigated the evolution of shell effects along the $N = 82$ neutron shell closure in neutron-rich nuclei around $Z = 50$ [64,94,95] as well as on the neutron deficient side close to the proton drip line [96]. Here first high precision mass measurements of Yb and Tm isotopes were able to prove the existence of the shell closure up to the proton drip-line, solving this side of the $N = 82$ prescience puzzle [96]. In fact, TITAN's mass measurement of ^{150}Yb marked the discovery of this isotope, the first isotope to be discovered with the MR-TOF technique and the first at TITAN. Ongoing work now addresses more neutron-rich isotopes further below $Z < 50$.

2.3 Nuclear astrophysics

Atomic mass measurements are key tools in modern nuclear structure studies and play an important role in identifying ground-state and isomeric-state structure phenomena and couple them to their theoretical description. Additionally, nuclear ground state properties are important inputs for large scale network calculations describing astrophysical processes. Particularly explosive processes, such as neutron star merger or X-ray burst require precise masses of participating, exotic, nuclei [97]. Mass measurements were performed using the MPET and MR-TOF-MS spectrometers addressing neutron-rich Rb and Sr isotopes [98–100] as well as investigating ground and isomeric states of In and Cd iso-

topes [64,94,95]. Following the recent detection of Sr in the aftermath of a binary neutron star merger, more recent work looks at the formation of the first r-process abundance peak and explored the impact of new Ga masses on the nucleosynthesis in neutron star merger [101].

2.4 Superallowed β decays and CKM matrix

Fundamental symmetries investigated via nuclear probes typically demand for the highest level of experimental precision. A prime example of this research is the study of superallowed nuclear β decays which provide a unique precision window to the weak interaction [102]. Experimentally, the ft value of a superallowed β emitter is determined which, among others, depends on the transition energy and thus on the masses of the involved parent and daughter nuclides. Once theoretical corrections at the $\approx 1\%$ level are applied to the ft values, the precisely studied superallowed β emitters provide stringent limits on physics beyond the Standard Model of particle physics. For example, they have confirmed the conserved vector current (CVC) hypothesis at a level of $9 \cdot 10^{-5}$ and constrain the potential existence of a scalar interaction in the weak force to the 0.1% level of the vector strength. Moreover, superallowed β decays allow for the most precise determination of the up-down quark mixing element V_{ud} in the Cabbibo–Kobayahsi–Maskawa (CKM) matrix, leading to a value of $V_{ud} = 0.97373 \pm 0.00031$ [102] when considering recent advances in the calculation of involved radiative corrections. This corresponds to a shift of 1 σ from the previous 2015 survey [103] as well as a 50% increase in its uncertainty. The updated value of V_{ud} impacts the unitarity test of the top row in the CKM matrix, $|V_{ud}|^2 + |V_{us}|^2 + |V_{ub}|^2 = 1$, which now falls short of unity by $\approx 2\sigma$ [104]. Such a tension to CKM unitarity motivates a careful examination of theoretical corrections applied to superallowed β decays, especially those which depend on nuclear structure. These comprise nuclear-structure dependent contributions to the radiative corrections as well as isospin symmetry breaking (ISB) due to the Coulomb and other charge-dependent nuclear forces. As new theoretical approaches emerge for both of them, e.g. in Refs. [102,105–107], experiments are critical to guide these models. For example, ISB corrections in superallowed β decays are linked to the isobaric multiplet mass equation (IMME). TITAN investigated the breakdown of the IMME in light nuclei, a common tool from nuclear theory to estimate nuclear properties and limits of the isospin description of nucleons [108–110] as well as addressed the superallowed β-decay Q-values of ^{22,23}Mg [63,111].

One of the flag-ship measurements in this regard was the first demonstration of a mass measurement of on-line charge-bred, short-lived radionuclides, ^{74}Rb, with a half-live as short as $T_{1/2} = 65$ ms where we were able to obtain a charge state of $q = 8+$ [45]. As the heaviest of the precisely studied superallowed β emitters, ^{74}Rb exhibits the largest ISB corrections and, thus, could serve to benchmark present and upcoming theoretical models [112]. Although TITAN's mass measurement led to a reduction in the uncertainty of ^{74}Rb's superallowed transition energy, the latter remains the leading uncertainty in its ft value. This motivates another ^{74}Rb measurement campaign at TITAN, at an even higher charge state. A limiting factor in TITAN's first measurement was the total time the HCI could be stored and excited in the Penning trap. The maximally attainable storage duration depends on the vacuum quality in the Penning trap and, hence, developments are under way to upgrade the system to a cryogenic system with significantly reduced background pressures.

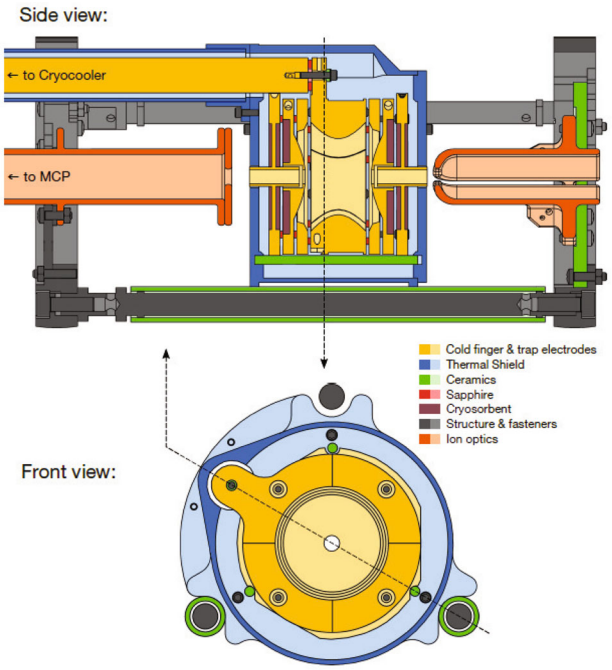

Fig. 7 Rendered concept of the TITAN CryoMPET front and side view. The various materials and functions are indicated in color. Taken from [113]

3 A next-generation Penning trap for TITAN

The design of the cryogenic measurement Penning trap (CryoMPET) follows the design of the original TITAN Penning trap [114], adapted to the needs of the new setup, that is, operational at a temperature of a few Kelvin. Further optimization based on operational experiences unrelated to the cryogenic upgrade were incorporated to improve its overall performance. A schematic representation of the CryoMPET construction is shown in Fig. 7.

The trap geometry is formed using hyperbolic shaped electrodes, essentially following the original TITAN Penning trap

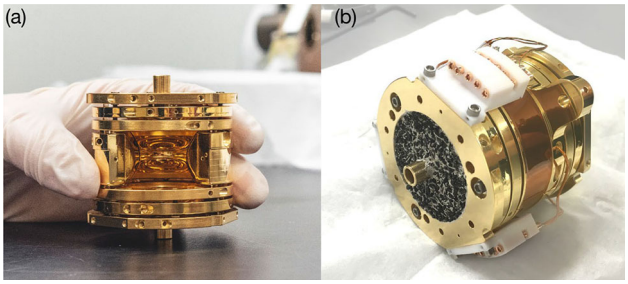

Fig. 8 Pictures of the assembled electrode structure of the cryogenic Penning trap: **a** without one segment of the central ring electrode showing its interior, courtesy of Stuart Shepherd (TRIUMF); **b** fully assembled with cryosorbent, wires, connectors and instrumentation. Taken from [113]

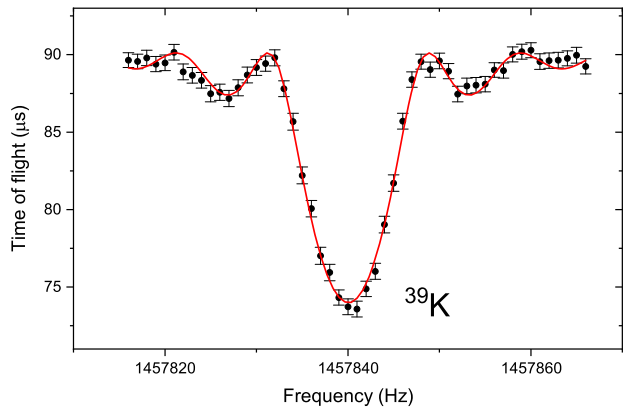

Fig. 9 First resonance of $^{39}K^{+1}$ ions obtained with the newly commissioned CryoMPET operated at cryogenic temperature ($T = 20$ K) using a 100 ms excitation time

version. The structure of the electrodes was designed to be massive and more solid to facilitate thermal conduction, necessary to reach the low temperatures.

The electrodes were all constructed using ultra-pure copper and finished with a gold plating. In order to achieve the required vacuum conditions, CryoMPET utilizes cryo-absorption and cryo-condensation by applying a material of high porosity to strategic surfaces (see Fig. 8). Moreover, the trap surface is continuously cooled using a closed-cycle cryocooler and thermally connecting it to a cold-head via a gold-plated high purity copper feed.

The new Penning trap system has been installed in the previous MPET magnet and the trap electrodes have been cooled down to temperatures of around 20 K. First system tests have been performed and an initial cyclotron resonances of stable off-line ions ($^{39}K^{+}$) have been recorded, see Fig. 9, indicating that the complete system meets specifications and on-line measurements will be possible in the upcoming experimental campaigns.

4 Conclusion

Over the last 15 years TITAN has pursued a rich program of high-precision mass measurements to address contemporary questions in nuclear physics research. The ambitious program has led to the development of ion traps and associated techniques to measure extraordinarily short-lived, low-production-yield, severely contaminated beams of radionuclides. Consequently, the topics investigated ranged from the evolution of the nuclear structure towards the proton and neutron driplines and exotic halo nuclei to explosive end-of-life stars synthesizing heavy elements to rigorous tests of fundamental symmetries.

The system was originally conceived as an ideally matched mass measurement system for on-line produced isotopes with the ISOL method and followed principles developed over the years at the ISOLTRAP facility [115] and adding capabilities for rapid charge breeding via an EBIT, a concept originally developed for the HITRAP facility [116] at GSI Darmstadt.

The TITAN system has since then evolved and multiple additions, improvements, and unforeseen developments and upgrades have enriched the science opportunities, leading to over 120 original publications and more than 25 graduate student thesis. Over the years, the TITAN collaboration together with collaborators on individual projects has constituted a group of more than 80 individual scientists from 15 countries and 4 continents. Major contributors include the Max-Planck Institute for Nuclear Physics which played a major role in designing and building the EBIT charge breeder, and GSI-Darmstadt and University of Giessen, who were responsible in conceiving, developing, and building the MR-TOF system. The TITAN system is located at the ISAC facility at TRIUMF and hence builds upon a strong Canadian community in atomic and nuclear physics; specifically members of the following institutions were instrumental in making the experiments and the TITAN community a success: University of British Columbia, Simon Fraser University, University of Victoria, University of Calgary, University of Manitoba, McGill University, University of Toronto, University of Windsor, and TRIUMF.

All of this was only possible with support from the National Science and Engineering Research Council of Canada (NSERC), the Canada Foundation of Innovation (CFI) and contributions from the National Research Council of Canada (NRC).

Acknowledgements We thank the TITAN collaboration for their hard work, dedication, and collegiality over many years. The many undergraduate students from around the world who contributed, the graduate students from the TITAN partner institutions, and the over 30 post-doctoral fellows who carried a major part of the success at TITAN. Since TITAN is a highly technical system, operating at the edge of technology, it is only possible with dedicated experts, many at TRIUMF and the partner institutions. In particular, we want to highlight Mr. M. Good, for

almost 20 years of technical support of the highest level. Moreover, the success of TITAN is based on many people contributing including those who help on the user facility side of TRIUMF, and as such we thank the ISAC team, including the beam development and beam delivery groups to support and enable our collaborations to explore new target designs and new beams to ISAC as well as for scientific opportunities and discussion.

Data availability This manuscript has no associated data or the data will not be deposited. [Authors' comment: This review article discusses 15 years of TITAN operation. It does not contain any new, original physics data.]

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