

First on-line applications of a
multi-reflection time-of-flight mass separator at ISOLTRAP
and the mass measurement of ^{82}Zn

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Nomenclature

A	mass number
BE	binding energy
D	characteristic ion-trap size parameter
E	energy
G	Gibbs free energy
G_0	Gravitational constant, $G_0 = 6.673\,84(80) \times 10^{-11} \text{ m}^3/\text{kg}/\text{s}^2$ [NIS13]
M	mass of an astronomical object
ME	mass excess
N	neutron number
N_A	Avogadro constant, $N_A = 6.022\,141\,29(27) \times 10^{23} \text{ mol}^{-1}$ [NIS13]
N_b	baryon number
R	mass resolving power or radius
S_i	separation energy, $i = n, 2n, p, 2p, \dots$
T	temperature
T_0	revolution time in MR-ToF-MS
T_{RF}	RF excitation time
T_{obs}	observation time
$T_{1/2}$	half life
V_0	trapping potential difference
V_Q	quadrupolar excitation amplitude
Z	charge/proton/electron number
ΔT_0	revolution ToF difference in MR-ToF-MS
Δt	ToF difference
Δt_{th}	thermal ToF spread
Δ	difference
Δ_i	shell gap, $i = n, p$
δ	uncertainty or cyclotron frequency detuning
γ	gamma radiation or damping coefficient
$\mathbf{B}$	magnetic field vector
$\mathbf{F}$	force vector
$\mathbf{a}$	acceleration vector
$\mathbf{v}$	velocity vector
$M_\odot$	solar mass, $M_\odot = 1.988\,55(25) \times 10^{30} \text{ kg}$ [Wil13]
u	atomic mass unit, $u = 1.660\,538\,921(73) \times 10^{-27} \text{ kg}$ [NIS13]
μ_i	chemical potential, $i = e, b = \text{electrons, baryons}$
∇	Nabla operator
ν	frequency
ν_i	eigenfrequencies in a Penning trap, $i = c, -, +, z = \text{cyclotron, magnetron, reduced cyclotron, axial}$

Nomenclature

ω	angular frequency, $\omega = 2\pi\nu$
ω_{RF}	RF excitation frequency
ω_i	angular eigenfrequency in a Penning trap, $\omega_i = 2\pi\nu_i$
$\bar{\omega}_R$	complex extension of the Rabi oscillation frequency
ϕ	electric potential
ρ	mass density or radius
ρ, z	cylindrical coordinates
σ_{rms}	root-mean-square deviation
ε_i	energy density, $i = L, e, tot, \dots = \text{lattice, electron, total}$
$^A_Z X_N$	isotope X
a	distance from the center of the Penning trap or lattice spacing
c_0	speed of light in vacuum, $c_0 = 299\,792\,458\,\text{m s}^{-1}$ [NIS13]
g	coupling parameter of RF excitations
k	Fermi momentum
m	mass
m_e	electron rest mass, $m_e = 9.109\,382\,91(40) \times 10^{-31}\,\text{kg}$ [NIS13]
m_n	neutron rest mass, $m_n = 1.674\,927\,351(74) \times 10^{-27}\,\text{kg}$ [NIS13]
m_p	proton rest mass, $m_p = 1.672\,621\,777(74) \times 10^{-27}\,\text{kg}$ [NIS13]
n	number density or number of revolutions in MR-ToF-MS
p	pressure
q	electric charge
r	radius
t	time, ToF
t_s	time of flight without MR-ToF-MS trapping
x, y, z	Cartesian coordinates
AME	Atomic Mass Evaluation [Aud95; Wap03; Aud03; Aud12b; Wan12]
BPS	neutron-star crust model of [Bay71]
CERN	European Organization for Nuclear Research
DC	direct current
EoS	equation of state
GPS	general purpose separator
HRS	high-resolution separator
ICR	ion-cyclotron resonance
ISOLDE	isotope separator online
MR-ToF-MS	multi-reflection time-of-flight mass separator/spectrometer
MS	mass spectrometry
PT	Penning trap
RF	radio frequency
RFQ	radio-frequency quadrupole ion trap
RILIS	resonance-ionization laser ion source
ToF	time of flight
ToF-ICR	time of flight ion-cyclotron resonance

1 Introduction

100 years ago, in 1913, J. J. Thomson¹ reported about his recent experiments and results on “Rays of positive electricity” [Tho13]:

The composition of these positive rays is much more complex than that of the cathode rays, for whereas the particles in the cathode rays are all of the same kind, there are in the positive rays many different kinds of particles. We can, however, by the following method sort these particles out, determine what kind of particles are present, and the velocities with which they are moving.

The method Thomson mentions uses a combination of electric and magnetic fields to deflect the components of a beam of charged particles according to their mass-over-charge ratios m/q . This discovery and the invention of the mass spectrograph are regarded as the cornerstone of mass spectrometry (MS), a research tool which contributed essentially to our understanding of nature in the last 100 years. In an address to the Royal Society in the same year, he discussed the observation of a so far unknown mass [Mla92]:

...in addition [to the neon-20 line] there is a line corresponding to an atomic weight 22, which cannot be identified with the line due to any known gas ...

Under all conditions he could examine, this “atomic weight 22” was tightly connected to the simultaneous appearance of the well-known mass 20 of neon. This was the discovery of the isotopic nature of the chemical stable elements and the first groundbreaking result of mass spectrometry. However, Thomson was not convinced of this explanation. Even when his colleague F. W. Aston² wanted to show him the first mass spectrum photographs of chlorine, indicating two lines of the isotopes chlorine-35 and 37, he refused to look at the photographs [Mla92]. At this time, it was known that radioactive atoms can have different masses and the same chemical properties, but the known radioactive nuclides were much heavier than neon or chlorine. F. Soddy³, who introduced the term “isotope”, was enthusiastic about Thomson’s discovery and wrote [Mla92]:

...the discovery is a most dramatic extension of what has been found for the elements at one extreme of the periodic table, to the case of an element at the other extreme, and strengthens the view that the complexity of matter in general is greater than the periodic law alone reveals. Although

¹1856-1940, Nobel Prize in physics 1906 for the discovery of the electron

²1877-1945, Nobel Prize in chemistry 1922 for, amongst others, the discovery of the isotopy of numerous stable elements with mass spectrographic methods

³1877-1956, Nobel Prize in chemistry 1921 for, amongst others, his research in radioactive decay

1 Introduction

the complexity is greater, the problem of atomic structure has been much simplified, because the generalisation gives a probable explanation of the absence of exact simple numerical relations among the atomic weights. [\[Sod13\]](#)

What Soddy means is that isotopy can be found on both ends of the periodic table, not only among the heavy elements. Furthermore, the reason why the atomic mass is not an integer and does not follow a pattern many researchers have tried to define, is based on the fact that the atomic mass is the mean of the isotope masses weighted by their abundances.

The isotope mass m of an atom is the sum of the mass of its constituents, electrons $Z \cdot m_e$, protons $Z \cdot m_p$ and neutrons $N \cdot m_n$, minus their mass-defect m_{BE} , released as binding energy $BE = m_{BE}c_0^2$,

$$m(Z, N) = Z \cdot m_e + Z \cdot m_p + N \cdot m_n - BE/c_0^2. \quad (1.1)$$

The nuclear binding energy results from the interaction among nucleons (protons and neutrons) due to the so called nuclear force, a residual of the strong interaction, and the interaction between protons due to the electromagnetic force. Therefore, the binding energy is the “fingerprint” of the nucleus, reflecting the sum of all interactions present within, giving each combination of protons and neutrons bound in a nucleus a unique binding energy.

While the long-standing goal of theoretical nuclear physics, to give a unified microscopic description of all properties of all nuclei based on a nuclear Hamiltonian, has not yet been achieved, precision mass measurements are still essential in order to quantify the binding energy. Because the nuclear force is very short-ranged, e.g., in the range of 2.5 fm, it acts only between neighboring nucleons, in contrast to the infinite range of the Coulomb interaction. The binding energy per nucleon peaks around mass number $A = N + Z = 60$, whereas smaller and larger nuclei are less bound. For smaller nuclei, more nucleons are found on the surface, therefore have fewer neighbors and are less bound by the nuclear force, while very heavy nuclides have many protons and therefore the Coulomb interaction reduces the binding energy. ^{62}Ni has the highest binding energy per nucleon of all nuclides with 8794.546(8) keV/ A , followed by ^{58}Fe and ^{56}Fe [\[Aud12b\]](#). These nuclei are about 0.9 % lighter than the sum of the mass of its constituents. In contrast, the mass defect due to the electromagnetic force between the proton and the electron in a hydrogen atom is on the order of 10^{-6} %, therefore orders of magnitude smaller.

An early attempt to calculate nuclear binding energies was the mass formula by Bethe and Weizsäcker [\[Wei35; Bet36\]](#), utilizing the liquid drop model of the nucleus and phenomenological corrections. By splitting up the binding energy in terms related to volume, surface, and Coulomb, symmetry and pairing energy, the binding energies of medium mass nuclei could be reproduced with a deviation to the experiment of only about 1 % by just fitting five coefficients to the experimental data. More recent mass models will be described at the end of this chapter.

By the end of 1919, Aston had already recorded over 200 of the 287 different isotopes known at this time⁴ [Ast21; Ast20]. In 1918, A. J. Dempster enhanced the mass spectrometric techniques with a device 100 times more accurate than spectrographs before [Dem18] and discovered the ²³⁵U isotope in 1935, which should be separated from the much more abundant ²³⁸U isotope by the “Calutrons”, giant mass spectrometers built in the 1940’s in the course of the Manhattan Project. In 1934, J. Mattauch and R. Herzog developed the first achromatic mass spectrograph by combining the advantages of Aston’s and Dempster’s developments [Mat34]. Mattauch-Herzog type MS are still in use today for precision measurements and abundance analysis.

The next milestone in MS was enabled by the advancements in fast electronics: In 1946, W. E. Stephens presented the first “pulsed mass spectrometer with time dispersion”, which paved the success of time-of-flight mass spectrometry (ToF-MS), i.e., relatively small, rapid, non-magnetic and non mass-range limited mass analyzers [Ste46]. This concept was crucially enhanced by W. C. Wiley and I. H. McLaren in 1955 [Wil55] and B. Mamyrin in 1973 [Mam73], improving the performance of ToF-MS by orders of magnitude with respect to distinguishable signals in mass, defined as mass resolving power $R = m/\Delta m$. In the following decades, the invention of the ion trap, either based on radio-frequency (RF) electric fields, the Paul trap [Pau53], or on static electric and magnetic fields, the Penning trap [Bro86], paved the way for today’s most versatile and accurate mass spectrometers, see Figure 1.1. The most accurate mass measurements on stable isotopes are performed by measuring the oscillation frequencies of single ions in a Penning trap via their induced image currents [Mye13]. A comparable analytical technique for samples of many constituents is FT-ICR (Fourier-transformation ion-cyclotron resonance) MS, developed by A. G. Marshall in 1973, see [Mar00] for a review.

While in the 1970s and 1980s not much progress has been made in the field of TOF-MS, despite the fact that it became a widely used, commercially available, technique. Compared to FT-ICR devices, the mass resolving power was poor (a few 10^3 compared to 10^6), however, spectra could be obtained within microseconds, compared to seconds. An important step came in the 1990, when H. Wollnik discussed the possibility to improve the moderate performance of ToF-MS by using the same ion flight path multiple times [Wol90]. A comparable design had already been realized in 1960 by W. Tretnner [Tre60], but at this early time, the achievable performance was limited due to technical reasons. The possibility to confine ions by reflections between multiple ion-optical elements, comparable to a high-energy ion storage ring, and at the same time preserve a time-of-flight focus plane for the temporal dispersive ion species, was successfully demonstrated [Wol90]. The development, implementation and application of a so called multi-reflection time-of-flight (MR-ToF) mass analyzer for on-line mass separation are the subject of this thesis (cumulative thesis articles [Wol12a; Wol13b]). The concept was technically enhanced and simplified to facilitate the challenges of on-line mass analysis, separation and spectrometry (cumulative thesis article [Wol12b]). The world-wide first successful applications of this type of device under on-line conditions is as well presented

⁴Today, about 3350 isotopes are known, 254 of them are considered as stable plus 35 considered as primordially stable (half-life higher than the age of the earth), 3061 are radionuclides, 1256 isotopes have at least one excited state considered as isomer ($T_{1/2} > 100$ ns) [Aud12a]

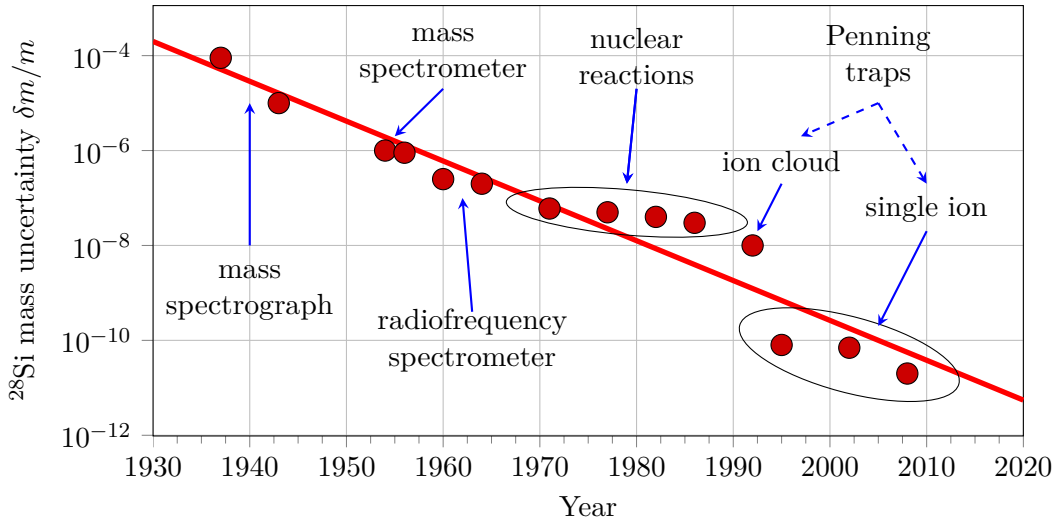


Figure 1.1: The evolution of the ^{28}Si mass determination in terms of achievable mass uncertainty from the 1930s until today (red lines shows linear fit to the data). The most recent improvements of over two orders of magnitude are contributed by Penning-trap mass spectrometry on a single highly charged ion (Figure adapted from [Aud04], recent data from [Ber02; Red08]).

in the articles [Wol12a; Wol13a; Wol13b] and [Wie13] (not part of the cumulative thesis).

Similar ion traps, often referred to as “electrostatic ion beam traps” (EIBT, introduced simultaneously in 1997 by Zajfman and coworkers [Zaj97] and Benner and Henry [Ben97]) or “electrostatic linear ion trap” (ELIT) [Hil13], are used for a wide range of atomic, molecular, and cluster studies [Rei10; Lan10; Gre11]. In many cases the focus of these devices not on their mass spectrometric aspects. Instead, they provide ion storage for an extended time by use of space-charge effects [Ped01; Str03; Fro12; Ros13] of dense ion clouds and the additional advantage of the detection of neutral decay products.

1.1 Precision mass measurements of short-lived isotopes

The development of the Penning trap for direct mass measurement with high precision and high accuracy (in the following these two terms are abstracted as low uncertainty) and high sensitivity allows detailed studies of the “mass surface”, i.e. the propagation of the mass along isotopic, isotonic and isobaric chains of the nuclear chart. Based on the binding energy, other quantities can be derived which reveal the influence of the underlying forces. These are, for example, the separation energies S , i.e., the energy needed to separate some nucleons from the nucleus, and the shell gap Δ , providing insights to nuclear shell structure and shape transitions, see Figure 1.2. Either single or double differences of binding energies (for protons and neutrons) can be evaluated, e.g.,

the one and two-neutron separation energies and the neutron shell gap

$$S_n(Z, N) = BE(Z, N) - BE(Z, N - 1) \quad (1.2)$$

$$= ME(Z, N - 1) - ME(Z, N) + ME(0, 1) \quad (1.3)$$

$$S_{2n}(Z, N) = BE(Z, N) - BE(Z, N - 2) \quad (1.4)$$

$$= ME(Z, N - 2) - ME(Z, N) + 2ME(0, 1) \quad (1.5)$$

$$\Delta_n(Z, N) = S_{2n}(Z, N) - S_{2n}(Z, N + 2) \quad (1.6)$$

$$= 2BE(Z, N) - BE(Z, N - 2) - BE(Z, N + 2) \quad (1.7)$$

$$= ME(Z, N - 2) - 2ME(Z, N) + ME(Z, N + 2). \quad (1.8)$$

A commonly used measure for the mass of an atom is the mass excess $ME(Z, N) = m - A \cdot u$, with u the atomic mass unit.

By measuring the mass of an atom, or in practice an ion, with a sufficiently low mass uncertainty $\delta m/m$, insights into various fields of nature can be gained. Today, the lowest uncertainties are achieved by measuring revolution frequencies of ions in a Penning trap or a storage ring. Facilities pursuing Penning-trap mass spectrometry on radioactive ions are ISOLTRAP [Muk08] at ISOLDE/CERN in Geneva/Switzerland [Kug00], JYFLTRAP [Äys08] at IGISOL in Jyväskylä/Finland, SHIPTRAP [Blo13] at GSI in Darmstadt/Germany, TITAN [Ett13] at ISAC/Triumf in Vancouver/Canada, CPT [Sav06] at ANL near Chicago/USA, LEBIT [Rin13] at MSU in East Lansing/USA and TRIGA-TRAP [Smo12] at the TRIGA research reactor in Mainz/Germany. Furthermore, under construction are MLL-Trap [Web13] at the Maier-Leibnitz laboratory of the LMU Munich which will be installed in the future DESIR/SPIRAL-2 facility/France [Bla13] and LPT [Wen12] at Lanzhou/China. The HITRAP [Klu08] Penning-trap facility at the future FAIR [Rod13] complex at GSI is being commissioned and will deliver highly charged stable and radioactive ions up to bare uranium for precision experiments including mass spectrometry [Klu13]. Furthermore, frequency-measurement based MS on short-lived isotopes is performed also with ion storage rings such as the ESR at GSI [Bos13; Fra08] and the CSRe in Lanzhou [Xu13]. High-energy ToF-MS uses projectile fragmentation reactions coupled to a magnetic rigidity time-of-flight spectrometer (ToF-B ρ), performed e.g. with SPEG at GANIL/France, TOFI at LANL/USA and at the S800 at NSCL/USA, see [Mei13] and references therein. The following list gives examples for mass uncertainties and their application, focused mainly on radioactive isotopes.

- $\delta m/m < 10^{-5}$: Analysis of chemical compounds regarding its constituents in general purpose analytical chemistry, environmental science and physics. Most commercial mass spectrometers offer this level of uncertainty. Necessary observation times are often very short, e.g., in the millisecond range per process, which, e.g., offers the possibility for time-resolved analysis of the evolution of chemical processes. For radioactive isotopes, this level of uncertainty can, in cases of very exotic nuclides with very low binding energies, be used for identification [Cra10], claim of discovery [Fra08] and basic nuclear shell evolution studies [Mei13]. However, for detailed nuclear structure analysis, this uncertainty is often insufficient.
- $\delta m/m < 10^{-6}$: Nuclear shell effects in the vicinity of the magic numbers can be

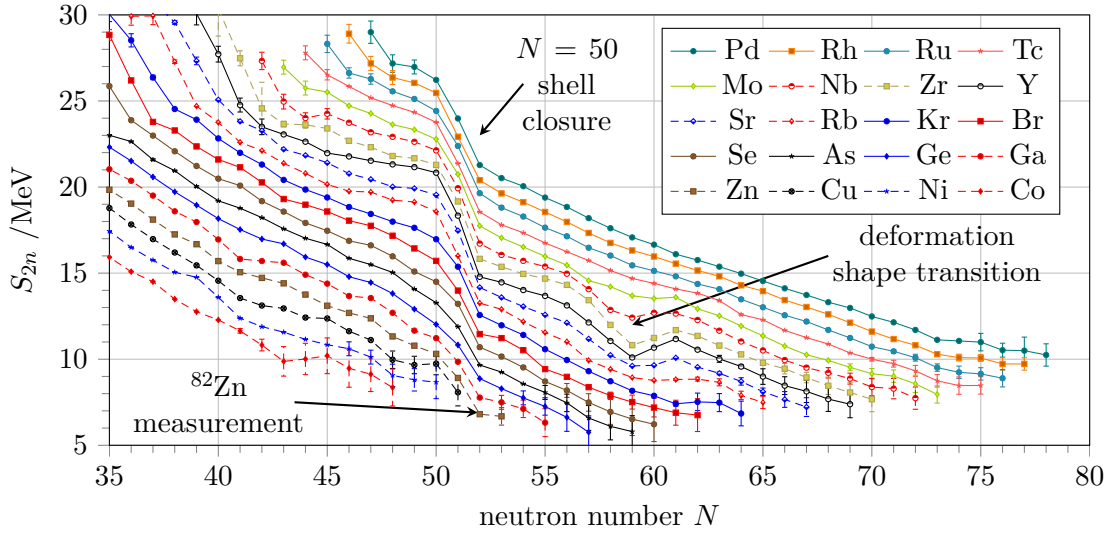


Figure 1.2: Two-neutron separation energies S_{2n} in the $Z = 27$ to $Z = 46$ region around the $N = 50$ shell closure. The shell closure is clearly seen by the drop in two-neutron separation energy by about 4 MeV. Furthermore, a strong nuclear deformation is found around $Z = 40$ and $N = 60$. The new S_{2n} value obtained with the first mass measurement of ^{82}Zn is shown as well.

evaluated from BE -uncertainties of 100 keV or less, see Figure 1.2. Furthermore, astrophysical models require separation energies as input parameters to calculate, e.g., r and rp -process pathways that finally aim to reproduce and clarify the elemental abundances found in our solar system [Lun03; Bre12; Sch13; Pea13; Cla13]. The first mass measurement of the neutron-rich nuclide ^{82}Zn (with an uncertainty of $\delta m/m < 4 \times 10^{-8}$) has been applied to the scenario of the isotope abundance expected in the outer crust of a neutron star as will be discussed in chapter 3 (cumulative thesis article [Wol13a]).

- $\delta m/m < 10^{-7}$: Variations in the binding energy due to interactions between neutrons and protons occupying the highest orbits can be evaluated using differences in binding energy to multiple neighboring isotopes [Cak05; Nai12]. The magnitudes of these variations are on the order of 150 keV to 250 keV, therefore, to draw a clear conclusion, the mass uncertainties of all involved nuclides should be not more than a few 10 keV. To unambiguously identify low-lying isomers with excitation energies of 100 keV and below, Penning traps are so far the primary mass spectrometers that are able to achieve this [Van04]. Isomers with some hundreds of kiloelectronvolts excitation energy can also be identified with other techniques, e.g., storage-ring mass spectrometry [Fra08].
- $\delta m/m < 10^{-8}$: High-precision Q -value measurements of the β -emitters ^3H , ^{187}Re and the electron-capture Q -value of ^{163}Ho are needed to enable a data analysis of experiments to determine the (anti)neutrino mass. The antineutrino mass, today upper-limit estimation with about $2 \text{ eV}/c_0^2$, can be obtained from the end-point

of the β^- -decay spectrum by a comparison of the fit to the β -decay spectrum of ^3H and the measured Q -value of the decay to ^3He [Eli13; Ott08]. The end-point spectrum will be determined with, e.g., the KATRIN experiment [Dre13], while the precise Q -value will be determined by, e.g., the THe-Penning trap experiment with an aimed uncertainty of $\delta m/m < 10^{-11}$ [Die11]. Furthermore, it is not clear if neutrinos are Majorana or Dirac particles. A possible way to clarify this is through neutrinoless double β^- decay or neutrinoless double electron-capture processes. The observation of one of these processes would unambiguously identify the neutrino to be its own antiparticle and a violation of lepton-number conservation (Physics beyond the standard model). If a neutrinoless double- β -decay occurs, the energy of the emitted electrons sum up to the Q -value, which is therefore needed with high precision. Further experiments are the test of the unitarity of the Cabibbo-Kobayashi-Maskawa (CKM) matrix and tests of the conserved-vector-current (CVC) hypothesis [Ero13]. Stable isotopes have been measured with uncertainties of $\delta m/m = 10^{-10}$ and below [Mye13] as well as antimatter, in particular the antiproton [Gab06] to test the CPT-theorem [DiS13]. Furthermore, the electron mass and the fine structure constant can be determined from high precision mass measurements.

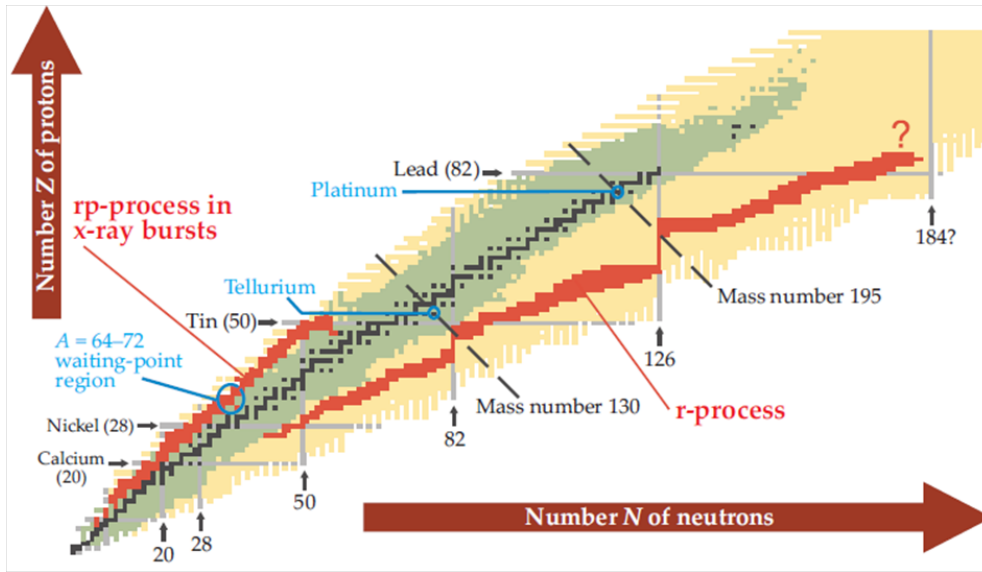


Figure 1.3: Nuclide chart showing the stable nuclides in black, nuclides with known mass in green, unobserved nuclides between a proposed proton and neutron dripline in yellow and possible paths of the r and rp process. The grey lines mark the magic neutron and proton numbers. The dashed lines mark the major waiting points of the r process at mass numbers $A = 130$ and $A = 195$ [Sch08].

While for all stable and most radioactive isotopes with half-lives in the order of several tens of seconds experimental mass data is available, mass models have to be used to extrapolate masses of unknown nuclides. Based on experimental mass data,

these models can derive binding energies from the proton to the neutron dripline, e.g., nuclides where protons or neutrons are still bound, therefore have a positive neutron or proton separation energy [Lun03]. This is especially important for astrophysical processes on the outskirts of the nuclear chart, like the rapid proton capture (rp) process and rapid neutron capture (r) process [Sch13; Pea13], see Figure 1.3. These processes, besides the slow neutron capture s process [Kae11], are believed to be the origin of the heavy elements above iron ($Z = 26$), which cannot be produced in stellar nuclear fusion reactions due to the high binding energy per nucleon around ^{56}Fe [Arn99]. While the exact astrophysical site, temperature and pressure conditions of these processes are not known today, the following is assumed:

- rp process: The rp process occurs in the thermonuclear explosion at the surface of a neutron star accreting hydrogen-rich matter from a companion star and is the source of so-called X-ray bursts [Sav09; Sch13]. Sequences of fast proton captures ($(p, \gamma) - (\gamma, p)$ equilibrium within an isotonic chain) and slow β^+ decays create nuclides along and close to the $N = Z$ line of the nuclear chart, from He up to Sn, Sb, Te, the end points of the rp process. Proton captures occur up to and in some cases even beyond the proton dripline (even-to-even Z). The observed X-ray bursts last between ten and hundred seconds and reoccur with times from hours to days. The nuclear reactions determine the composition of the “ash” which is continuously buried by newly accreted material, the heat transferred to the neutron star crust, and the shape of the observed X-ray burst light curve [Est11].
- r process: The exact site of the r process is still unknown and one of the biggest open questions in nuclear astrophysics [Arn07; Has02]. Possible sites are core collapse supernovae explosions and neutron star mergers [Sch13]. It consists of a seed nuclide (in the Fe to Se region), followed by a process of fast neutron captures ($(n, \gamma) - (\gamma, n)$ equilibrium within isotopic chains) and slow β^- decays up to the uranium region. Due to a variety of possible r process pathways, depending on the temperature, pressure and neutron density of the particular astrophysical site, masses of many, very neutron-rich nuclides are necessary to validate the possible scenarios.
- Neutron stars: The composition of the outer crust of cold, non-accreting neutron stars depends on the binding energies of several neutron-rich isotopes, the electron energy and the energy of a lattice composed of nuclei [Bay71; Rüs06; Pea11]. This will be described in more detail in chapter 3.

In general, nuclear masses affect astrophysical models in the following ways [Sch13]:

1. Changes in the Q -values lead to different energy consumption or release of a nuclear reaction and therefore change the overall energy generation. Mass uncertainties thus contribute to the energy uncertainty which is of special importance for neutron-star crust heating.
2. The abundance distribution within an isotopic (isotonic) chain in the r (rp) process ($(n, \gamma) - (\gamma, n)$ or $(p, \gamma) - (\gamma, p)$ equilibrium condition, respectively) is determined by the Saha equation, where the neutron (proton) separation energy

enters exponentially. Therefore, within a chain the binding-energy differences influence the abundance of the isotopes.

3. Nuclear resonances, especially in the rp process, influence (p, γ) reaction rates and therefore the speed and reaction path. These depend on the excitation energies and the Q -values.

1.2 Mass models

Since the development of the semi-empirical Bethe-Weizsäcker mass formula, many different approaches have been developed in order to predict the structure and properties of unknown nuclei, e.g., their masses. The main objective of theoretical nuclear physics is to calculate all properties of the nuclei from first principles. This has not yet been reached due to the missing knowledge of the character of the underlying force between the nucleons and due to the complexity of the many-body problem. Nuclear scattering data of, e.g., deuterons is used to fit the nucleon-nucleon (NN) interaction potential, e.g. Argonne V18 [Pie08], however, calculations of light nuclei ($A < 12$) with only NN interaction already show a relatively high discrepancy in binding energy to the experiment. Therefore, also many-nucleon forces such as three-nucleon (3N) forces have to be included, but their parameters cannot be obtained from experiment and have to be calculated, e.g. the Illinois-7 potential [Pie08]. Recently, microscopic models with realistic interactions based on chiral effective field theory been developed to also calculate masses [Hol12; Wie13]. However, due to the complexity the later can so far only calculate masses of light nuclei and of medium-mass nuclei with closed shells.

For heavy nuclei other approaches have to be used. Models ranging from local mass formulas such as the Garvey-Kelson (GK) relations [Gar66] over global mass models using a combination of microscopic and macroscopic parameters, e.g., the finite range droplet model (FRDM) [Mö195]. Non-relativistic models covering the whole mass table for even-even nuclides – from the proton to the neutron dripline – are, for example, based on the self-consistent effective mean-field Hartree-Fock-Bogoliubov (HFB) method [Ben03; Sto06; Sto07].

Self-consistent non-relativistic effective mean-field models employ an effective force, i.e., an averaged interaction of all nucleons within a nucleus [Ben03; Sto07], to calculate the ground-state properties of nuclides with a certain parametrization of an effective force [War10], e.g. Skyrme-type [Sky56; Sky59] or Gogny-type [Dec75] interaction, see Figure 1.4. The method has much in common with the density functional theory of electronic systems. However, the difference is that the Coulomb interaction is well known and therefore the electronic energy-density functional can be derived ab initio. In the nuclear case this is different because in contrast to electrons, nucleons are compound particles; therefore a nucleonic interaction is already an approximate concept. Nuclear many-body theory cannot yet provide all the necessary input for effective mean-field models but it helps to provide a guidance to develop energy-density functionals [Sto07].

The effective interactions, such as the ones of Skyrme type, use a set of parameters which have to be fitted to experimental data and is therefore phenomenologically adjusted. Skyrme's interaction [Sky59; Vau72] for example uses only contact interaction for its two- and three-nucleon parts. In the series of Hartree-Fock-Bogoliubov (HFB) mass

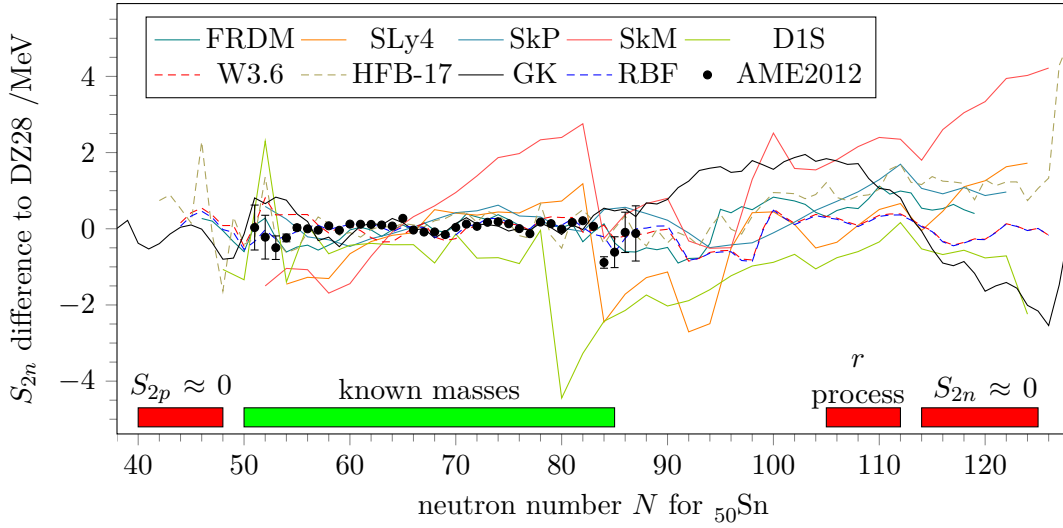


Figure 1.4: Comparison of the two-neutron separation energy of the ${}_{50}\text{Sn}$ isotopes of the most frequently used or recently developed mass models. Indicated are the proton and neutron drip “zones” and a possible crossing of the r process. The values are presented as differences to the DZ28 (Duflo-Zuker) mass model [Duf95]. The other models are: FRDM (Finite Range Droplet Model) [Mö195], SLy4 (Skyrme-Lyon 4 parametrization, Hartree-Fock-Bogoliubov method with Transformed Harmonic Oscillator basis) [Cha95; Cha98; Sto03; Dob04], SkP (Skyrme-Pairing parametrization, Hartree-Fock-Bogoliubov method) [Dob84; Dob04], SkM (Skyrme parametrization, Hartree-Fock-Bogoliubov method) [Kri80; Bar82; Ben03], D1S (Gogny [Dec75] parametrization with constrained-HFB method) [Del10], WS3.6 (Weizsäcker-Skyrme 3, semi-empirical mass formula with Skyrme-force correction) [Wan10b; Wan10a; Liu11], HFB-17 (Skyrme parametrization with more realistic pairing, Hartree-Fock-Bogoliubov method) [Gor09b; Gor09a], GK (Garvey-Kelson relations) [Com88; Gar66; Gar69] and WS3.6-RBF (WS3.6 with radial basis functions and Garvey-Kelson relations) [Wan11]. Experimental binding energies are available from $N = 50$ to $N = 85$.

models of the BRUSLIB collaboration [Uni13], the most recent Skyrme interactions HFB-19, 20 and 21 use a set of 16 parameters for the Skyrme interaction and are fitted to all the experimental mass data available [Gor10]. Furthermore, the parameters of the interaction are adjusted to reproduce different equations of state of infinite symmetric and neutron matter obtained from realistic microscopic calculations including NN and 3N forces. The nucleon pairing is included with 5 additional parameters which are fitted to the 1S_0 pairing gaps of homogeneous nuclear matter of the appropriate charge asymmetry [Gor10]. The HFB formalism is a further advancement of the Hartree-Fock formalism combined with BCS (Bardeen, Cooper, Schieffer) theory of superconductivity to account for the pairing of nucleons in nuclei and to describe phases of e.g. neutron superfluidity in neutron stars. The root-mean-square deviation of all the calculated masses from the experimental masses for nuclides with $N, Z > 8$ is in the order of 0.6 MeV, which puts these models among the best mass models. However, they do not reproduce the so-called Wigner energy of $N = Z$ nuclei. Therefore, an additional

phenomenological correction depending on the proton-neutron asymmetry is included.

A common measure for the quality of a model is the root-mean-square deviation σ_{rms} with respect to the experimental data. However, this quantity tells how well the adjustment of the parameters of this model can reproduce the known data, but it cannot estimate how the model evolves in the unknown territory. Systematic variations like over- or under-binding can only be evaluated by further measurements and judging the quality of an extrapolation on the validity of the model data compared to new, complementing experimental data. In [Figure 1.4](#), the two-neutron separation energies of $_{50}\text{Sn}$ isotopes are plotted for various mass models and experimental data [\[Aud12a\]](#) as differences to the Duflo-Zuker (DZ28) mass model [\[Duf95\]](#). As can be seen, the models agree best in the region of the known masses to which their parameters have been adjusted. In the interesting region towards the proton and neutron driplines, where the rp and r process are located, extrapolations differ by more than 1 MeV, while the astrophysical reaction flows and sequences of the processes need mass uncertainties below 50 keV to be defined with respect to binding energy. Therefore, new mass data is needed on the one hand to confirm or falsify the models in particular regions of the nuclear chart (and find the most suitable to date to extrapolate), and on the other hand to improve them. A comprehensive compilation can be found in [\[Lun03\]](#).

2 On-line mass measurements at ISOLTRAP/ISOLDE

The evolution of the ion-trap mass spectrometer ISOLTRAP [Bol96; Muk08] – named after the facility where it is localized, ISOLDE (Isotope Separator On-Line) [Kug00] at CERN – started more than 25 years ago, when the first masses of radionuclides ever measured with a Penning trap (PT) were reported [Bol87; Klu13]. By then, the masses of short-lived nuclides were mostly determined by measuring the Q -value of their nuclear decay or that of nuclear reactions [Roe13], or by sector field mass spectrometers [Thi75].

ISOLDE is a facility dedicated to the production of a very large variety of radioactive ion beams. Connected to CERN’s proton-synchrotron booster (PSB), nearly half of CERN’s accelerated protons with energies of 1.4 GeV are guided towards one of the two target stations, connected to magnetic isotope separators: the general-purpose separator (GPS) and the high-resolution separator (HRS). By proton irradiation of either the target (e.g., uranium carbide, refractory metal powder, molten metal) itself or a close-by neutron-converter (tungsten or tantalum) [Cat03], fission reactions are induced, as well as spallation and fragmentation in the case of direct proton irradiation, which produce the exotic nuclides.

Besides the production cross-sections, the suppression of unwanted nuclides (created as well in the target), and the release time from the target are of main importance for experiments on very short lived species. By controlling the temperature of the target and transfer line towards the ion source, the effusion and diffusion times can be influenced, facilitating or suppressing the transfer of the created elements. Different ion sources can be used to ionize the wanted species with highest efficiency while ideally ionizing as little as possible of the contaminating species. This is perfected by using the (multi-step) resonance-ionization laser ion source (RILIS) [Fed12]. However, surface ionized atoms are not suppressed. If no laser ionization scheme is available, hot/cold plasma or simply surface ionization is used.

Successively, the ion beam is mass separated in the magnetic separators (GPS or HRS), providing mass resolving powers of up to $R = m/\Delta m \approx 5000$, sufficient to suppress neighboring isotopes by many orders of magnitude, but inadequate in purifying the beam from isobaric and isomeric contaminations. Therefore, the suppression of unwanted nuclides delivered in the beam – even with RILIS – remains a major challenge for on-line experiments. Figure 2.1 shows an excerpt of isotope yields from the ISOLDE yield data base [The13]. As can be seen, exotic isotopes are in general produced in minute amounts, e.g., a few hundred ions per second, while stable or long lived isobars can drown the beam by orders of magnitude in yield.

At ISOLDE, the beam of singly charged ions is transferred with kinetic energies typically between 30 keV to 60 keV in an electrostatic beam-transfer system to the user experiments. ISOLTRAP catches the quasi-continuous beam electrostatically in a radio-

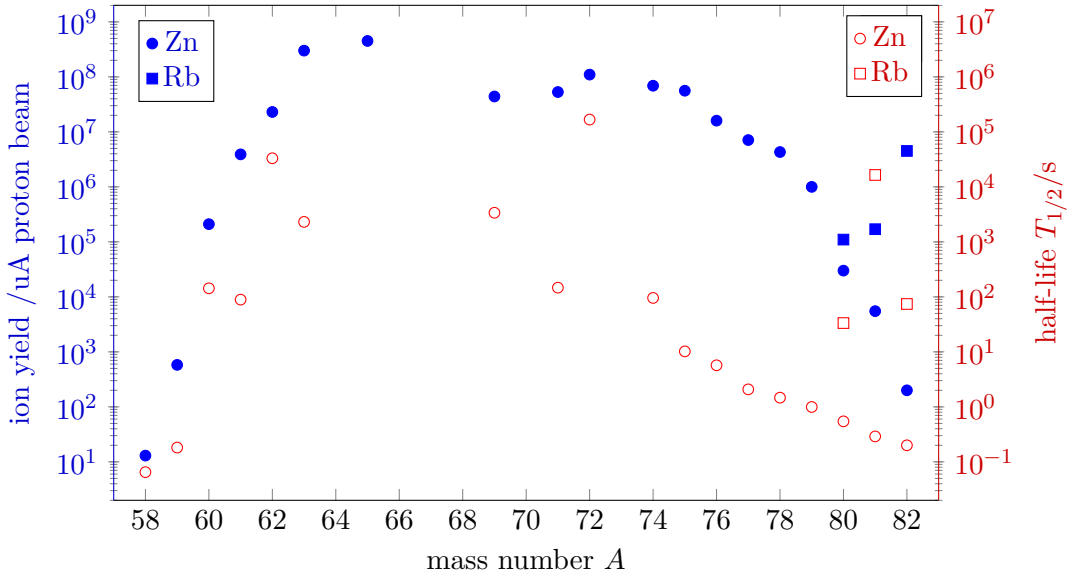


Figure 2.1: Isotope production of Zn and Rb isotopes at ISOLDE (left ordinate) and the corresponding half-life (right ordinate). The production of Zn decreases by over one order of magnitude per mass number for very neutron rich or very neutron deficient isotopes. The ratio in yield of ^{82}Rb to ^{82}Zn is about 20000 [The13] (Ground-state half-lives and their PSB production yields; if two or more yield values were given, the highest has been taken. For Rb, only these three PSB values are published online).

frequency (RF) quadrupole ion-beam cooler and buncher (segmented linear Paul trap, in short RFQ) on a floatable high-voltage platform [Her01]. The ions are decelerated to a few electronvolts and finally thermalize in a helium buffer-gas environment, confined by RF and DC electric fields. If a sufficient number of ions are accumulated, they are ejected in bunches into a pulsed drift tube, which is switched from a high-voltage potential (close to the RFQ potential) to ground. Up to 2010, the ion bunches were then directly transferred to the preparation Penning trap (PT1), see Figure 2 of [Wol13b]. In the framework of this PhD project, a multi-reflection time-of-flight mass separator (MR-ToF-MS) [Wol90] and a Bradbury-Nielsen gate (BNG) [Bra36] have been developed, installed and brought to operation between the RFQ and the PT1 and will be discussed below. In the PT1 the ions are prepared for the mass measurement in the following precision Penning trap (PT2). The main functions are the simultaneous minimization of the ions phase-space volume by helium buffer-gas cooling and contamination separation by RF-excitations of the ion motion. The technique developed at ISOLTRAP, mass-selective resonant buffer-gas centering [Sav91], has been the method of choice for nearly two decades. The limits of this technique and the need for a different method will be described in section 2.2 and section 2.3. After the preparation, the cooled and (in the majority of cases) isobarically cleaned ion bunches are transferred into the PT2. Here, the ion motion is excited by RF-fields to determine the cyclotron frequency via the time-of-flight ion-cyclotron resonance (ToF-ICR) method [Grä80], from which the mass

can be extracted as will be described in [section 2.1](#). To purify the ion ensemble from very close-by isobars or even isomers necessitating mass resolving powers well above $R = m/\Delta m = 10^5$, cleaning via RF-excitations can also be applied in PT2 preceding the mass measurement [Bol92]. Furthermore, a tape station setup can be used to perform nuclear decay spectroscopy [Kow12].

To link the cyclotron frequency to the mass of the ion, the magnetic field at the time of the measurement has to be known. This is accomplished by reference measurements of ions with well-known masses, provided by two off-line ion sources. An alkali ion source is located in front of the RFQ to optimize the complete setup and to perform reference and cross-check measurements. A carbon-cluster laser-ablation ion source is located between the MR-ToF-MS and PT1 to perform systematic studies on the uncertainty of the apparatus over a wide mass range (12 u to 240 u) [Kel03] and to use these clusters as references for absolute mass measurements, thus linking the unknown mass directly to the atomic mass-standard of $u = 1/12 \cdot m(^{12}\text{C}) = 1 \text{ g}(^{12}\text{C})/N_A$ [Bec13]. Since its first experiments in 1987 [Bol87], ISOLTRAP has measured the masses of close to 600 nuclides.

Section 2.1 will discuss the performance of modern Penning-trap mass spectrometers for short-lived species and explain the most commonly used techniques as the ToF-ICR technique and the mass-selective resonant buffer-gas centering. In [section 2.2](#), the limits of these techniques are discussed. Section 2.3 and 2.4 will summarize the MR-ToF mass separator related improvements at ISOLTRAP which have been published in the respective papers and will give additional information on the operation principal of the device.

2.1 Penning traps as state-of-the-art mass spectrometers

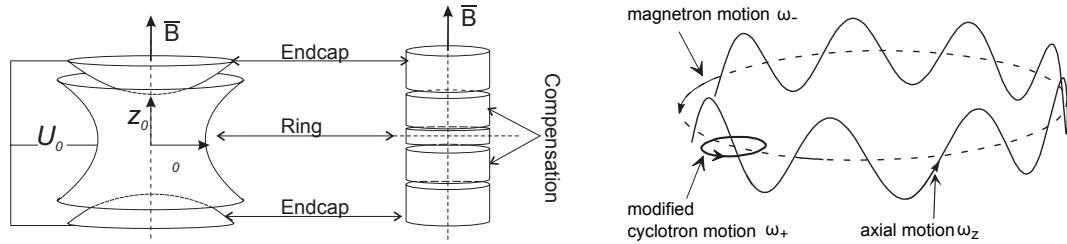


Figure 2.2: Sketch of the electrodes of a hyperbolic and a cylindrical Penning trap (left) and the ion motion in a Penning trap composed of the three eigenmotions, magnetron motion (metastable), cyclotron motion (stable) and axial motion (stable) (right) [Muk08].

The success of Penning trap mass measurements originates from the fact that they allow a direct mass measurement with only a few ions, stored for an extended period in time, in principle until they decay, in a very well defined, limited space in the homogeneous field of a superconducting magnet, see [Figure 2.2](#). Furthermore, the mass determination relies on a frequency, which can be provided and measured with lowest

uncertainty. The cyclotron frequency,

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

of an ion with charge q and mass m , from which the mass can be determined, is ideally only related to the magnetic field B . Due to the superior performance of modern superconducting magnets, providing highly homogeneous magnetic fields with relative deviations of $\delta B/B \leq 1 \times 10^{-8}$ in volumes of about a cubic centimeter, and a temporal stability of $\delta B/B \leq 1 \times 10^{-8}$ per hour (limited by flux creep), the magnetic-field related systematic uncertainty on the determination of the cyclotron frequency is usually $\delta\nu/\nu_c \leq 1 \times 10^{-8}$. However, the temporal stability of the magnetic field is influenced by the environmental conditions acting on the dewar vessel of the magnet and therefore also on the superconducting coils surrounded by liquid helium. Deviations of the laboratory temperature and air pressure as well as floor vibrations can be observed as shifts in the cyclotron frequency due to deformations of the superconducting coils.

To preserve the homogeneity, care has to be taken of the material in the trapping region as well as of the surroundings of the superconducting magnet. The trap electrodes and their insulating material are ideally machined as thin as possible from material with vanishing magnetic susceptibility, such as oxygen free copper and sapphire. Besides magnetic field imperfections and instabilities, the deviations from the ideal quadrupolar electric field and the orientation of the magnetic field axis with respect to the electric (ion trap) axis have to be considered. Therefore, ion traps are equipped with dedicated correction electrodes to compensate the deviations of the quadrupolar field due to the finite electrode structure and re-correct for machining tolerances. Furthermore, the surfaces of the electrodes are gold plated to prevent electric field distortions originating from patch oxidations. To align the axis of the magnetic and electric field, the vacuum tube housing and supporting of the ion trap is mounted in a gimbal support attached to the magnet's vessel, allowing one to adjust the trap axis with respect to the magnetic field axis.

Finally, the measurement of the cyclotron frequency is perturbed by the presence of other ions loaded in the trap as a result of their mutual Coulomb interactions. This is the case for many ions of the same species and, more critical, for ions of different species. The existence of a space charge modifies the electric field inside the trap and leads to significant shifts in the frequencies even for low ion numbers (more than a few tens) on the one hand, and on the other hand leads to ion-bunch coalescences for higher ion numbers. This phenomenon, which is also known from FT-ICR applications, ion-beam accelerators, storage rings and electrostatic ion beam traps, couples the revolution frequencies of different species and prevents their individual investigation [Str02; Bol11; Ros13].

As mentioned before, the Penning trap, as shown in Figure 2.2, confines ions by a superposition of a homogeneous magnetic field

$$\mathbf{B} = B\mathbf{z} \quad (2.2)$$

along the axis of a cylindrical symmetric electrostatic quadrupole potential

$$\phi(\rho, z) = \frac{V_0}{4D^2} (2z^2 - \rho^2), \quad \rho^2 = x^2 + y^2, \quad 4D^2 = 2z_0^2 + \rho_0^2. \quad (2.3)$$

2.1 Penning traps as state-of-the-art mass spectrometers

The equation of motion for an ion in the Penning trap is therefore

$$\mathbf{a} = \frac{q}{m} (-\nabla\phi + \mathbf{v} \times \mathbf{B}) \quad (2.4)$$

$$\begin{pmatrix} \ddot{x} \\ \ddot{y} \\ \ddot{z} \end{pmatrix} = \frac{qV_0}{m2D^2} \begin{pmatrix} x \\ y \\ -2z \end{pmatrix} + \frac{qB}{m} \begin{pmatrix} \dot{y} \\ -\dot{x} \\ 0 \end{pmatrix}. \quad (2.5)$$

In axial direction, the motion is a harmonic oscillator, $\ddot{z} + \omega_z^2 z = 0$, with the axial angular eigenfrequency

$$\omega_z^2 = \frac{qV_0}{mD^2} \quad (2.6)$$

which is uncoupled from the radial motion. In the radial plane, the two angular eigenfrequencies are

$$\omega_{\pm} = \frac{\omega_c}{2} \pm \sqrt{\frac{\omega_c^2}{4} - \frac{\omega_z^2}{2}}, \quad (2.7)$$

with the cyclotron frequency

$$\omega_c = \omega_+ + \omega_- = qB/m, \quad (2.8)$$

the modified cyclotron frequency ω_+ (reduced from ω_c by ω_- due to the presence of the quadrupolar electric field) and the magnetron frequency ω_- (radial $E \times B$ drift motion around the axis of the trap). While there are other invariant relations between the frequencies of the eigenmotions, this is the most important in the context of mass measurements at ISOLTRAP. Since $\omega_z \ll \omega_c$, it can be seen in a Taylor expansion of ω_- in ω_z that

$$\omega_- \approx \frac{\omega_z^2}{2\omega_c} = \frac{V_0}{2D^2B} \quad (2.9)$$

is mass-over-charge independent in first order. Typical values for a $^{82}\text{Zn}^+$ ion at $B_{\text{PT2}} = 5.9 \text{ T}$, $D_{\text{PT2}} = 10.23 \text{ mm}$ and $V_{0,\text{PT2}} = 8.5 \text{ V}$ are ($\nu = \omega/2\pi$):

$$\nu_- \approx 1.1 \text{ kHz} \ll \nu_z \approx 49 \text{ kHz} \ll \nu_+ \approx \nu_c \approx 1.1 \text{ MHz}.$$

The ring electrodes of both Penning traps are segmented to allow azimuthal RF excitations of the ion motions (the ring of PT1 is eight-fold and the ring of PT2 is four-fold segmented). Two schemes are necessary for mass measurement and mass-selective resonant buffer-gas centering, explained for the case of a four-fold segmented ring electrode (the same applies for an eight-fold segmented ring if two neighboring segments are interconnected):

- dipolar excitation: Using two opposing segments with an RF-voltage phase shifted by 180° , either the radius of the cyclotron or magnetron motion can be modified by resonant RF excitations at ω_- or ω_+ (first order mass independent).
- quadrupolar excitation: Using all four segments with an RF-phase shift of 180° between neighboring segments, the magnetron motion can be converted to cyclotron motion and vice versa by resonant excitation at the cyclotron (sum) frequency $\omega_c = \omega_+ + \omega_-$ (sideband excitation) [Bol90].

The excitation scheme is basically the same for PT1 (purification and cooling with mass-selective resonant buffer-gas centering) and PT2 (mass measurement with the ToF-ICR method). At first, the magnetron radius of all ions is increased by a dipolar excitation at ω_- . Then, the quadrupolar excitation frequency ω_{RF} is applied for a duration T_{RF} which results in a resonant conversion between magnetron and cyclotron radius if the cyclotron frequency ω_c is hit. The initial magnetron radius is decreased while the energy of the fast cyclotron motion is increased, which is maximized in resonance. Including a damping force $\mathbf{F} = -2m\gamma\mathbf{v}$ with the damping coefficient γ , the radial energy of the ion after the quadrupolar excitation can be derived [Kre08; Geo11] as proportional to

$$E_{\text{rad}} \propto \frac{\rho_+^2(T_{\text{RF}})}{\rho_-^2(0)} = \frac{4g^2}{|\overline{\omega_R}|^2} \left| \sin\left(\frac{\overline{\omega_R} T_{\text{RF}}}{2}\right) \right|^2 \exp(-2\gamma T_{\text{RF}}) \quad (2.10)$$

with

$$\overline{\omega_R} = \sqrt{4g^2 + \delta^2 - \tilde{\gamma}_1^2 + i2\delta\tilde{\gamma}_1} = \sqrt{4g^2 + (\delta - i\tilde{\gamma}_1)^2} \quad (2.11)$$

the complex extension of the Rabi oscillation frequency,

$$g = \frac{q}{m} \frac{2V_Q}{a^2(\omega_+ - \omega_-)} \quad (2.12)$$

the coupling to the quadrupole excitation amplitude V_Q at a radius a from the center of the trap, the detuning from the cyclotron frequency $\delta = \omega_{\text{RF}} - \omega_c$ and

$$\tilde{\gamma}_1 = 2\gamma \frac{\omega_c}{\sqrt{\omega_c^2 - \omega_z^2}} = 2\gamma \sqrt{1 - \frac{\omega_z^2}{\omega_c^2}} \approx 2\gamma \left(1 + \frac{1}{2} \frac{\omega_z^2}{\omega_c^2} - \dots\right) \quad (2.13)$$

Depending on the damping coefficient and therefore the gas pressure, the two previously mentioned applications result, which are realized in ISOLTRAP's two Penning traps.

1. In the precision Penning trap at background pressures of $p_{\text{PT2}} < 1 \times 10^{-8}$ mbar, damping can be neglected for excitation times $T_{\text{RF}} < 2$ s. By varying the detuning around the cyclotron frequency, $\delta = \omega_{\text{RF}} - \omega_c$, the radial energy of the ion is varied. To probe this, the ion is ejected from the high magnetic-field region of the Penning trap towards an ion detector outside the magnetic field. The radial motion creates a magnetic moment $\mu = E_{\text{rad}}/B$ and therefore the ion experiences an axial force $\mathbf{F} = -\mu \frac{dB}{dz} \mathbf{e}_z$ when moving through the gradient. The mean time of flight is recorded and plotted against the excitation frequency value. To precisely extract the cyclotron frequency, multiple different excitation frequency values around the cyclotron frequency have to be probed to produce a characteristic resonance pattern. Due to the well known line shape, the precise cyclotron frequency can be determined via a fit to the data points. This procedure is called time-of-flight ion-cyclotron resonance (ToF-ICR) technique [Grä80].
2. In the preparation Penning trap, high-purity helium is used at buffer-gas pressures around $p_{\text{PT1}} \approx 1 \times 10^{-4}$ mbar. All ions are initially brought on a magnetron radius larger than the radius of the exit aperture of the trap. To mass-selectively re-center only the species of interest for further transfer to the precision Penning trap and

removal of all other species in the ejection process, a quadrupolar excitation at ω_c is used as in the previous case to convert the magnetron motion of the species of interest into the fast cyclotron motion. The high cyclotron energy of the species of interest is more rapidly cooled by collision than their increase in magnetron radius due to collisions. Therefore, the wanted species is recentered to the trap axis while all other species are further drifting out of the trap [Sav91]. In the ejection process from the trap, the ions have to pass a small aperture of a 3 mm diameter. Therefore, only the re-centered ions are transmitted.

The full-width-at-half-maximum (FWHM) of the respective mass (frequency) signal of the resonance curves obtained with the ToF-ICR method is inversely proportional to the observation time,

$$\Delta\nu \propto \Delta m \propto T_{\text{obs}}^{-1} \quad (2.14)$$

which makes these devices the most precise mass spectrometers if the observation time is not limited. However, for short observation times, i.e., a few tens of milliseconds as demanded for very short-lived species, other techniques like the multi-reflection time-of-flight method can exceed their precision, see [chapter 4](#) and [Wie13].

2.2 Limits of on-line Penning-trap mass spectrometry

As described above, the preparatory steps for an on-line mass measurement have to be as fast as possible while maintaining a mass resolving power and contaminant suppression high enough to purify the ion ensembles from unwanted isobaric species. Due to the low mass resolving power of ISOLDE's separator magnets, the purification has to be performed at the ISOLTRAP setup.

The need for a pure ion ensemble arises from the requirement of a perturbation-free measurement environment. To reach a mass uncertainty of $\delta m/m = 1 \times 10^{-8}$ necessary to address, e.g., nuclear structure studies or weak interaction investigations, Coulomb interactions between different species, as well as between numerous ions of the same species have to be avoided. As shown in [Bol92; Bec01; Wol13a] these effects shift the resonance frequency on a relative level of $\Delta m/m = 10^{-7}$ or above, even for a few tens of ions trapped simultaneously.

Comparable effects related to the same cause have also been observed in gas-filled PTs used for mass selection [Sch05; Stu09; Her11; Gus11]. The purification from as many unwanted ions as possible is limited by the so-called bunch coalescence, which hampers the re-centering of the few wanted ions. The conditions for coalescence are not yet completely understood, but the investigations have shown that the maximum ion number is limited to some hundreds or a few thousand [Bol11; Ros13].

At ISOLTRAP, PT1 can be operated with mass-selective resonant buffer-gas centering and PT2 with dipolar excitation on the modified cyclotron frequency. As described in [Wol12a; Wol13a; Ros12b], the first method can provide high mass resolving powers of up to $R = 3 \times 10^5$ but only at low buffer-gas pressure, i.e., 10^{-6} mbar. Since buffer-gas collisions are essential to reduce the phase-space volume of the ion cloud, on the one hand to match the diameter of the ejection aperture, and on the other hand to assure a well-defined starting region in the precision Penning trap, the cooling time has to be

extended if the gas pressure is low. To reach mass resolving powers of 1×10^5 , the total operation period is around 400 ms. The second separation technique can be applied in the PT2 and can provide highest mass resolving powers of above 1×10^6 . To this end, the envelope of a dipolar RF excitation is modulated with a Gaussian shape which results in a Gaussian-shaped frequency spectrum and therefore reduces the possibility to unintentionally excite a close-neighboring species by one of the side lobes which are produced by the standard rectangular modulation. The mass-resolving power is Fourier-limited by the excitation time and can be approximated with [Bol01]

$$R = m/\Delta m = \omega_c/\Delta\omega \approx 1.25\nu_c T_{\text{RF}} \quad (2.15)$$

Mass resolving powers of up to 1×10^7 have been reached for excitation times of 10 s [Eli10]. Furthermore, also buffer-gas free excitation schemes have been developed to simultaneously purify the ion species from multiple different contaminations [Ros12a]. Because this cleaning is performed in the measurement trap itself, care has to be taken that the remaining contamination ions, possibly gyrating on high radii in the trap, are not perturbing the successive mass measurement.

2.3 The MR-TOF mass separator upgrade at ISOLTRAP

The development, implementation and first on-line operation of the isobar purification system are the main technical subjects of this thesis. The technical setup, operation parameters and performance are described in detail in [Wol12a; Wol13b] and will only be summarized briefly. The in-trap lift method [Wol12b], which is unique among the MR-ToF mass analyzers already operational world-wide, will be summarized in section 2.4. The system was the first of its kind operational at an on-line facility. The isobar purification system consists of two main parts: the MR-ToF mass analyzer to separate the ions and the Bradbury-Nielsen gate to select them. The mass analyzer is designed and optimized to achieve highest mass resolving power by extending the flight time through a folded trajectory and maintain a temporal well defined bunch in the detector plane. This is accomplished by use of two electrostatic mirrors in a coaxial arrangement to reflect the ions axially and focus them transversally to achieve a stable trajectory. The mass resolving power of this type of time of flight mass spectrometer can be approximated with

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t} \approx \frac{t_s + nT_0}{2\sqrt{\Delta t_{\text{th}}^2 + (\Delta t_s - n\Delta T_0)^2}}, \quad (2.16)$$

with $t_s = t_1 + t_2$ being the flight time in “shooting through”, thus the flight time from the ion source to the center of the analyzer t_1 , and the flight time from the center of the analyzer to the detector t_2 . T_0 is the revolution time and n the number of revolutions. $\Delta t_s = \Delta t_1 + \Delta t_2$ is the time difference between the slowest and the fastest ion in “shooting through”, while ΔT_0 is the time difference of these ions per revolution, see Figure 4 in [Wol12b]. Highest mass resolving power for a given flight time t is achieved at the time focus plane, which is maximal when $\Delta t = \Delta t_{\text{th}}$. Δt_{th} is the thermal time spread originating from the finite temperature of the ions in the ion source. To achieve a

compensation, $\Delta t_s - n\Delta T_0 = 0$, and maintain a stable trajectory, the electric potentials of the mirror electrodes have been optimized numerically [Wol11].

The Bradbury-Nielsen gate [Bra36; Pla08] is used to deflect the separated unwanted species from the optical axis of the beamline. Its advantage compared to commonly used parallel-plate deflectors [Tok09] lies in the fact that its mechanical layout provides the highest spatial resolution. The resolution is defined as the minimum spacing between ions needed in order that one ion species experiences the full deflection voltage while the other is not affected. This is achieved by two sets of wires, installed in a plane and biased with switchable potentials of alternating polarity but same strength, see Figures 2 and 6 in [Wol12a]. Thus, the electric fields cancel out in the vicinity of the gate, typically at a distance of one wire spacing before and behind the gate. The MR-ToF mass analyzer has been operated with mass resolving powers up to $R = 2 \times 10^5$ which is one to two orders of magnitude higher than for commercial linear and reflectron-type time-of-flight mass spectrometers, see for example [Bru13; Jor13]. This resolving power is reached at flight times of $t \approx 30$ ms, therefore also an order of magnitude faster than the time needed to achieve a comparable mass resolving power with a Penning trap. The transmission is mainly limited by losses due to the bunch emittance increase during injection.

2.4 The in-trap lift method to facilitate the MR-TOF MS application

To inject and eject ions “in-line” into and from the MR-ToF mass analyzer, i.e., in parallel to the optical axis of the system, their kinetic energy has to be high enough to overcome the mirror potentials. This is usually achieved by keeping the ion’s energy constant and sufficiently lowering the mirror electrode voltages. However, as introduced in [Wol12b] one can also keep the mirror voltages constant and modify the ions kinetic energy.

The first method is used in most MR-ToF systems and electrostatic ion beam traps, possibly because it is the most common technique known from the dynamical capture used in other ion traps. However, the quality of the mass spectrum relies strongly on the quality of the applied mirror potentials, on the one hand with respect to the correct voltages to cancel time-of-flight deviations for a particular number of revolutions, on the other hand with respect to the voltage fluctuations (ripple and noise) which reduce the mass resolving power.

Switching electrical potentials, in this case charging and uncharging a capacitive load, induces a load change in the electrical circuitry of the power supplies, which therefore have to regulate for readjust the voltage. This leads to a voltage drift in the time range of the regulation speed of the powersupplies, typically a few tens of milliseconds. The regulation time corresponds to the total flight time of the ions and thus changes in the potential distribution in the mirrors. Therefore, a constant potential distribution, which is necessary for a high mass resolving power, cannot be achieved without modifying the regulation circuits to low frequencies and/or using external low-pass filters with a cutoff frequency of less than 1 Hz which makes the reaction speed for wanted changes slow as well. This drawback is overcome by the new method, keeping the mirror voltages on a

constant potential and modifying the ions kinetic energy.

By use of a switched drift tube in the spacing between the mirrors, the ions' kinetic energy can be changed. To capture ions, the so called in-trap lift electrode is on a "high" potential when ions enter the MR-ToF system. Once they passed the mirror and entered the in-trap lift electrode, its potential is switched to "ground". The ions are now captured. The process is repeated to eject the ions.

The main advantages are that the mirror potentials are not disturbed by a switching process, the energy of the ions in the MR-ToF system can be varied without changing the kinetic energy of the ions in the adjacent parts of the beamline (and vice versa). Furthermore, the position of time-of-flight focus can be adjusted to be on the detector for any revolution number. By changing the kinetic energy of the ions also their revolution time changes and also their revolution-time differences ΔT_0 . This effect is used to achieve the condition $\Delta t_s - n\Delta T_0 = 0$ for any revolution number and for any Δt_s (isochronicity), see Figure 5 and 6 in [Wol12b] and Figure 3 in [Wol13b]. In contrast, when switching the mirror potentials the kinetic energy cannot be adjusted and ΔT_0 is fixed (despite its unwanted drift due to the load-change regulation). Therefore one would have to adjust the mirror potentials itself to achieve isochronicity for every number of revolutions, which is hard to achieve when the reaction of the system is very slow due to e.g. low-pass filtering with cut-off frequencies of less than 1 Hz. The in-trap lift method facilitates the use of the MR-ToF mass analyzer and its optimization largely over the method of switching the mirror and contributes significantly to the success of the measurements at ISOLTRAP.

3 The mass measurement of ^{82}Zn

The first successful on-line application of the MR-ToF mass separator that resulted in a mass measurement of a nuclide with so far unknown mass was the measurement of ^{82}Zn at ISOLTRAP. The masses of neutron-rich zinc isotopes are of interest for astrophysical calculations such as for the r process [Bar08] and the crustal composition of neutron stars [Wol13a; Kre13b]. Furthermore, the $N = 50$ neutron shell closure and the nuclear structure of the first two neutrons occupying the $2d_{5/2}$ neutron orbit (according to the HFB models [Uni13]) can now be investigated.

The measurement was attempted several times before at ISOLTRAP and at other mass-measurement setups, but always failed due to the high yield of contaminations and the relatively low half-life $T_{1/2}(^{82}\text{Zn}) = 228(10)$ ms [Mad12]. The main contaminant is the alkaline metal rubidium. By use of a temperature-controllable quartz tube embedded in the transfer line between the ISOLDE target and the ion source, the effusion and diffusion times of the ions can be varied. At a temperature of about 680°C , this results in a suppression of four orders of magnitude for rubidium without reducing the zinc yield [Bou07]. Furthermore, resonant multi-step laser ionization (RILIS) [Fed12] was used to selectively ionize only the zinc atoms. However, still about 6000 ^{82}Rb ions per second were present in the beam, coming from surface ionization in the target and ion-source region. In contrast, the ^{82}Zn yield was about 200 ions per second. The MR-ToF mass separator and the Bradbury-Nielsen gate were used to purify the ^{82}Zn ions from the ^{82}Rb contamination [Wol13a].

Figure 3.1 shows the two-neutron separation energies of zinc isotopes around the $N = 50$ shell closure and the $N = 50$ neutron shell gap of the measured data as well as calculated masses from the most widely used and accurate mass models. These models are the Duflo-Zuker mass model (DZ28), resulting in standard deviation to all experimental masses of $\sigma_{\text{rms}} = 375$ keV [Duf95], the finite range droplet model (FRDM) with $\sigma_{\text{rms}} = 669$ keV [Möl95], the Weizsäcker-Skyrme (WS3.6) mass model with $\sigma_{\text{rms}} = 336$ keV [Liu11] and the mean-field Hartree-Fock-Bogoliubov (HFB-19 and HFB-21) mass models with Skyrme energy-density functionals, reproducing the experimental masses with $\sigma_{\text{rms}} = 580$ keV [Gor10]. The later were used in the study of the crustal composition of a neutron star. With the new ^{82}Zn mass measurement, the $N = 50$ shell closure can be evaluated for the first time. With a mass excess of $ME_{\text{ISOLTRAP}}(^{82}\text{Zn}) = -42.314(3)$ MeV, the nuclide is less bound by $\Delta ME = 296$ keV than predicted by the Atomic Mass Evaluation (AME) 2012 ($ME_{\text{AME2012}}(^{82}\text{Zn}) = -42.610(300)$ MeV) [Aud12b], based on systematic trends in that mass region. Therefore, the two-neutron separation energy decreased to about $S_{2n}(^{82}\text{Zn}) = 6.8$ MeV. The neutron shell gap energy is about $\Delta_n(^{80}\text{Zn}) = 3.5$ MeV, which is an increase compared to ^{80}Ga and ^{80}Ge , therefore strengthening the shell closure again towards the $Z = 28$ proton shell closure of the doubly magic $^{28}\text{Ni}_{50}$ nucleus.

As can be seen from Figure 3.1, all models reproduce the steeper decrease in S_{2n} at

the shell closure. It is interesting to note that the FRDM and DZ28 models are fitted to the AME1995 [Aud95] and therefore to a much smaller data set of experimental masses (1654 nuclides for FRDM and 1751 nuclides for DZ28). In comparison to WS3.6, HFB-19 and HFB-21, whose parameters are adjusted with experimental data of 2149 nuclides from the AME2003 [Aud03], the accuracy of the prediction is remarkable, especially for FRDM, which reproduces the strength of the $N = 50$ shell gap with smaller deviations than HFB-19.

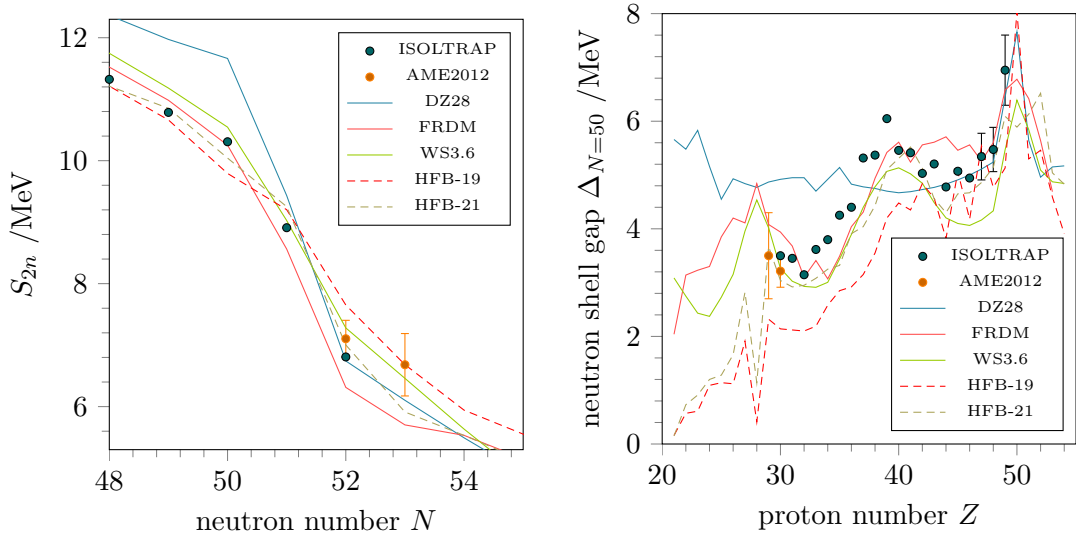


Figure 3.1: Two-neutron separation energies of the $_{30}\text{Zn}$ isotopes (left) and $N = 50$ neutron shell gap (right) of different mass models (see text for explanation) and experimental data (filled black circles) and extrapolated values from the AME2012 (filled orange circles).

3.1 Introduction to neutron stars

Neutron stars are the most compact objects known in the observable universe. They are created in a type I or type II core collapse supernovae. When a massive star of more than about $8 M_{\odot}$ (solar mass) has converted enough material via nuclear fusion to iron, the endpoint of solar nucleosynthesis, the iron core collapses under its own gravitational pressure and a neutron star is born [Heg03]. The birth mass of a neutron star can be very different from the idealized picture of the Chandrasekhar mass limit due to competing effects, which yields to neutron-star birth masses between $\approx 1 M_{\odot}$ and $\approx 1.6 M_{\odot}$ [Kiz10]. For even lighter stars, when the nuclear fusion processes ends, the electron degeneracy pressure stabilizes and balances the star against gravitational collapse. These are then called white dwarfs [Cha35]. However, above the Chandrasekhar limit, the degeneracy pressure cannot balance the gravitation, thus electrons and protons are converted via inverse β -decay into neutrons and neutrinos. The equation of state of the resulting ultra-dense matter with densities of 10^6 g/cm^3 to 10^{15} g/cm^3 , which is even above the saturation density of nuclear matter $\rho_0 = 2.8 \times 10^{14} \text{ g/cm}^3$ ($n_0 = 0.16$ nucleons per fm^3),

is an object of intense research. The main problem is the understanding of the nuclear force at such high densities, which cannot be reproduced in a laboratory.

Due to their small diameter of about 20 km, neutron stars are very hard to detect. They are mostly discovered and observed by their strong gravitational force in binary systems, e.g., with a white dwarf, a black hole or a star, or by their emission of radiation, if they periodically radiate in direction of the earth. Therefore, neutron stars are often referred to as “pulsars”. This name was given by J. Bell Burnell and A. Hewish in 1967, who discovered the first neutron star (Nobel Prize in Physics 1974 for Hewish). Due to their small size of a few kilometers in radius, the rotational period can be as small as 1.39 ms (PSR J1748-2446ad) [Hes06]. Today there are only two heavy neutron stars known with their masses precisely determined to about $2 M_{\odot}$ [Ant13]. These are PSR J1614-2230 [Dem10] and PSR J0348+0432 [Ant13], which put a lower limit on the maximum mass of a neutron star. The precise value, which is the analogue to the Chandrasekhar limit for the electron degeneracy pressure, is not yet known because of the lack of the theoretical treatment of highly neutronized matter above the nuclear saturation density. However, most of the discovered neutron stars have masses around $1.4 M_{\odot}$, see [Lat04; Lat12] for a review. For very heavy progenitor stars of above $25 M_{\odot}$, the supernova remnant is not a neutron star but a black hole [Heg03; Hae03]. Other scenarios to create a black hole are from merging neutron stars and maybe also matter accretion by a neutron star from a companion, e.g. a star or white dwarf.

3.2 The model of a cold, non-accreting neutron star

A commonly used assumption, especially when focusing on the composition profile, is to use the model of a cold, non-rotating and non-accreting neutron star. The matter composing this object is assumed to be in the ground state, therefore in complete thermodynamic equilibrium at zero temperature with respect to all interactions [Hae01]. This ground-state structure is illustrated in Figure 3.2. The profile of a neutron star can be divided into three distinct regions: the outer crust (including the envelope), the inner crust and the core. At pressures of 10^4 g/cm^3 to 10^8 g/cm^3 the outer crust consists of a crystal lattice of ^{56}Fe atoms which represents the energetically most favorite nuclide at this density. With increasing density atoms become fully ionized and at densities above 10^7 g/cm^3 the electrons can be treated as almost uniformly distributed and fully relativistic. Due to the high matter density, “neutronization” (inverse beta decay) starts to create more and more neutron rich nuclei. At pressures around $4 \times 10^{11} \text{ g/cm}^3$ the neutron Fermi energy reaches zero and no more neutrons can be bound to a nucleus. This marks the neutron dripline and the transition to the inner crust. In the inner crust, nuclei are therefore immersed in a sea of unbound neutrons and electrons. At densities of about 10^{14} g/cm^3 various non-spherical phases, called “pasta”, are predicted until the liquid core is reached with densities about and beyond the nuclear saturation density.

3.2.1 The outer crust

In their classical paper Baym, Pethick and Sutherland [Bay71] described the outer crust of cold, non-accreting neutron stars (BPS model) and calculated the ground-state equilibrium composition of nuclides between densities of 10^4 g/cm^3 and $4.3 \times 10^{11} \text{ g/cm}^3$

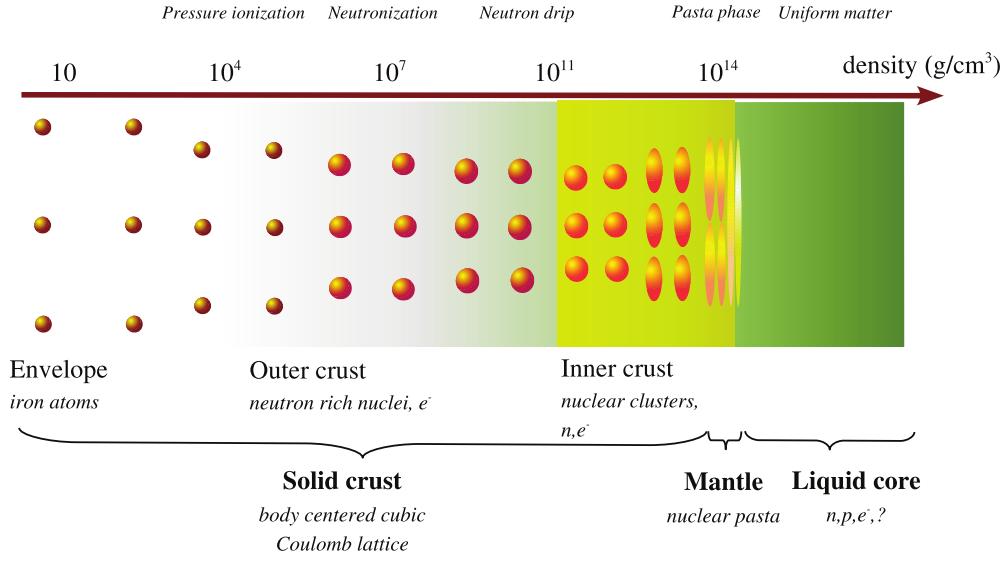


Figure 3.2: Schematic illustration of the ground state profile of a neutron star along the density axis [Cha08].

based on the mass table of Myers and Siwatecki [Mye66]. In the BPS model, the matter forms in a perfect crystal with a single nuclear species at lattice sites, mixed lattices of different species occur only in very thin layers in between two shells, whereby the transition can be approximated to be sharp¹. The energy density of nuclei with mass number A and proton number Z is

$$\varepsilon_{\text{tot}}(A, Z, n_N) = n_N E_N + \varepsilon_L + \varepsilon_e \quad (3.1)$$

where n_N is the nuclei density, $E_N = m \cdot c_0^2$ is the energy (mass) of an isolated nucleus, ε_L is the lattice energy density and ε_e is the energy density of the electrons, see also e.g. [Hem06; Rüs06; Pea11].

To find the ground state for a given density and pressure inside the outer crust, the total energy ε_{tot} has to be minimized. Besides the well-known electron and lattice energy, the masses of mostly short-lived nuclides are the relevant input parameters. Therefore, the specie which minimizes the total energy for a given density and pressure represents the ground state configuration. For nuclides with experimentally known mass (up to so far ^{80}Zn), the composition sequence and the equation of state (EoS, relation between pressure and density, $p(\varepsilon)$ for $T = 0$) is fixed. Where no experimental data is available, models are used to extrapolate the masses. Different mass extrapolations thus result in different composition profiles and sequences as well as higher-density equation of states [Rüs06; Pea11]. To finally determine the depth of the individual species in the neutron star outer crust, the general relativistic Tolman-Oppenheimer-Volkoff (TOV) equations [Tol39; Opp39] have to be solved for a neutron star of given mass and radius by including the equation of state. In the following, the basic procedure is explained according to

¹A treatment of the transition can be found for example in [Jog82]

[Bay71]. However, a more complete description can be found in, e.g., [Rüs06; Pea11]. Furthermore, no correction terms as are applied [Pea11].

For each shell within the outer crust charge neutrality has to be fulfilled. Thus, the number of electrons and protons has to be equal, $n_e = n_p = Zn_N$. Since at a temperature $T = 0$ the nuclei exert no pressure, the total pressure p of the system is given only by the electron pressure and the lattice pressure [Cha08],

$$p = p_e + p_L = p_e + \varepsilon_L/3 \quad (3.2)$$

with the electron pressure $p_e = \mu_e n_e - \varepsilon_e$, the chemical potential of the electrons $\mu_e = (k_e^2 c_0^2 + m_e^2 c_0^4)^{1/2}$ and the Fermi momentum of the electrons $k_e = \hbar (3\pi^2 n_e)^{1/3}$. The lattice energy represents the Coulomb interaction of the electrons and the nuclei. A Wigner-Seitz sphere with radius $R \approx a/2$ (approximately half the lattice spacing a) is constructed around every nucleus, arranged in a body-centered cubic (bcc) lattice, which is the energetic most favored composition. The lattice energy density can be calculated from the Coulomb energy of the electric field [Cha08]

$$\varepsilon_L = n_N E_L = -\epsilon_M e^2 \left(\frac{4\pi}{3} Z^2 n_e^4 \right)^{1/3} \quad (3.3)$$

with $\epsilon_M = 0.89593$ the Madelung energy per particle in a Coulomb bcc lattice [Bai01]. To calculate the ground state composition of the crust, the total energy density ε_{tot} has to be minimized for a given baryon density $n_b = An_N$ under the condition of charge neutrality, $n_e = Zn_N$. In the one-component plasma approximation used here (no mixed phases of multiple nuclear species), the baryon density suffers from jumps when the ground state shifts from one nuclear species to another, $(A, Z) \rightarrow (A', Z')$. In contrast, the pressure increases continuously towards the inner of the neutron star. Therefore, a minimization using Gibbs free energy $G(T, p, N_b)$ can be performed. By using the Gibbs-Duhem relation, $G = \mu_b N_b$, with the baryon number N_b , the baryon chemical potential μ_b can be minimized. The later can be written as [Rüs06]

$$\mu_b = (\varepsilon_{\text{tot}} + p) / n_b = (\varepsilon_{\text{tot}} + p_e + \varepsilon_L) / n_b = (E_N + 4E_L/3 + Z\mu_e) / A. \quad (3.4)$$

The equation has to be evaluated for every pressure p and for every $E_N(A, Z)$ from experimental data or mass models. The species (A, Z) which minimize μ_b is the ground-state nucleus for the particular pressure. Furthermore, the EoS $p = p(\varepsilon)$ is determined by the two expressions $\varepsilon(n) = c_0^2 \rho(n)$ and $p(n) = n^2 \frac{\partial(\varepsilon/n)}{\partial n}$.

To relate the pressure p and energy density ε of neutron star matter to a region within the a neutron star, the TOV equations [Tol39; Opp39],

$$\frac{dp(r)}{dr} = -\frac{G_0 \varepsilon(r) M(r)}{c_0^2 r^2} \left(1 + \frac{p(r)}{\varepsilon(r)} \right) \left(1 + \frac{4\pi r^3 p(r)}{c_0^2 M(r)} \right) \left(1 - \frac{2G_0 M(r)}{c_0^2 r} \right)^{-1}, \quad (3.5)$$

$$M(r) = 4\pi \int_0^r \rho(r') r'^2 dr' = \frac{4\pi}{c_0^2} \int_0^r \varepsilon(r') r'^2 dr' \quad (3.6)$$

have to be solved, see e.g. [Sto07; Pea11; Hem08]. They describe the hydrostatic equilibrium for a spherically symmetric body of isotropic material in general relativity.

G_0 is the gravitational constant, $M(r)$ is the mass inside a sphere of radius r and $p(r)$ is the pressure at this position. The coupled equations can be integrated numerically starting from $r = 0$ with $M(0) = 0$ and a given assumption for pressure in the central region $p(0) = p_0$, towards the surface of the neutron star with $M(R) = M_0$ and $p(R) = 0$. On the other hand, the equations can be integrated from the surface towards the core with proper assumptions for the total mass and radius, without the need of an assumption for the pressure in the center of the star. Since the high-density region of the EoS and thus the inner of the neutron star is still very uncertain, the latter method can be advantageous. This was used for the calculation of the depth profile and composition of the outer crust [Wol13a], assuming a neutron star of $M_0 = 1.4 M_\odot$ and $R = 10$ km radius.

3.2.2 The inner crust and the core

The deepest layers of the outer crust are composed of nuclides close to or directly at the neutron drip line. At this point the density reaches about $4 \times 10^{11} \text{ g/cm}^3$ and the neutron Fermi energy reaches zero. No more neutrons can be bound to a nucleus and the inner crust is reached. Below this layer, the nuclei become surrounded by a sea of free neutrons. Therefore, the border of nuclei becomes unclear and one often refers to “clusters of nucleons” instead of nuclei. The pressure is now dominated by the free neutrons, not the electrons anymore. As shown in Figure 3.2, the high densities at the deepest layers of the inner crust gives rise to predictions about strangely deformed nuclear matter, referred to as pasta phases. The clusters come so close that interaction with neighboring clusters must be taken into account. The core is reached at densities above 10^{14} g/cm^3 . The speculation of species of matter therein reach from hyperons, pions over kaons to strange quark matter.

4 Summary and Outlook

This thesis describes the implementation and first on-line application of a multi-reflection time-of-flight (MR-ToF) mass analyzer for high-resolution mass separation at the ISOLTRAP mass spectrometer at ISOLDE/CERN. On the one hand, the major objective was to improve ISOLTRAP's mass-measurement capabilities with respect to the ratio of delivered contaminating ions to ions of interest. On the other hand, the time necessary to purify wanted from unwanted species should be reduced as much as possible to enable access to even more exotic nuclei.

The device has been set up, optimized and tested at the University of Greifswald before its move to ISOLTRAP. The achieved performance comprises mass resolving powers of up to 2×10^5 reached at observation times of 30 ms and a contamination suppression of about four orders of magnitude by use of a Bradbury-Nielsen gate. With the characteristics, it outperforms clearly the so far state-of-the-art purification method of a gas-filled Penning trap. To improve the utilization of the MR-ToF mass analyzer, the in-trap lift method has been developed. It simplifies the application and optimization of the device, which is a crucial time factor in an on-line experiment. The device was the first of its kind successfully applied to radioactive ion beams for a mass analysis, for a mass separation (in combination with the Bradbury-Nielsen gate) as a preparatory step for a subsequent Penning-trap mass measurement and as a high-precision mass spectrometer of its own. The later was recently used for the first mass measurement of the neutron-rich calcium isotopes ^{53}Ca and ^{54}Ca , revealing a neutron shell closure at the neutron number $N = 32$ [Wie13]. The so-far achieved mass-resolving power of 2×10^5 belongs to the highest reported for time-of-flight mass analyzers at all.

The first successful application of the MR-ToF system as the only mass separator at ISOLTRAP resulted in the mass measurement of ^{82}Zn . The new mass value has been compared to mass extrapolations of the most recent Hartree-Fock-Bogoliubov (HFB) mass models, HFB-19 to HFB-21, of the BRUSLIB collaboration. The mass of the nuclide is of high interest for the compositions and depth profile of the outer crust of neutron stars. In the classical model of the outer crust of a cold, non-accreting and non-rotating neutron star, the sequence of nuclides found within this parts is determined mainly by the binding energy of exotic nuclides. The crustal compositions determined with the three HFB mass models differed with respect to the appearance of a layer of ^{82}Zn , originating from different mass extrapolations of this mass. With the new experimental data, the extrapolations could be evaluated. It was found that the HFB-21 mass value differs less from the experimental data than the ones from HFB-19 and 20. Therefore, in the classical model, ^{82}Zn does not appear anymore in the outer crust.

Due to its high resolution and very fast measurement time, the MR-ToF mass analyzer will be an important instruments for future activities at ISOLTRAP [Kre13a], at the ISOLDE facility in general [Mar13], and at other radioactive ion-beam facilities. Devices of this type, mainly for precision mass measurements, are already operational at the FRS

ioncatcher at GSI Darmstadt [Dic13] and at SlowRI/RIKEN [Ito13]. Devices are under construction at CARIBU/Argonne [Sav13], IGISOL/University of Jyväskylä [Jok13], GANIL/Caen [Del13] and Triumf/Vancouver [Dil13]).

Besides the mass purification and the precision mass measurement, other applications have been realized, for example regarding the target and ion-source development at ISOLDE. The optimization of different target and ion source parameters as well as the ion beam composition analysis and suppression of unwanted species is a preparatory step for every beamtime. The standards techniques to qualitatively and quantitatively evaluate radioactive ion beams, nuclear decay stations and Faraday cups, fail for example for long-lived species, competing decay modes and very low-intensity ion beams. The MR-ToF mass analyzer allows an analysis of ion beams independent of these parameters, for half-lives down to the 100-ms range and yields of about 10 ions per second. Furthermore, the hyperfine structure and isotope shift of atoms and therefore their spin (and g-factors) can be investigated background free with the MR-ToF mass analyzer in combination with RILIS [Kre13b; Mar13]. Therefore, the device enhances not only the mass measurement capabilities at ISOLTRAP but also the beam production and investigation at ISOLDE in general.

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6 Cumulative thesis articles

6.1 Author Contribution

Wol12b: Static-mirror ion capture and time focusing for electrostatic ion-beam traps and multi-reflection time-of-flight mass analyzers by use of an in-trap potential lift,

R. N. Wolf, G. Marx, M. Rosenbusch, L. Schweikhard, [Int. J. Mass Spectrom. 313, 8-14 \(2012\)](#)

R. W. invented the method with respect to the application at an MR-ToF-MS device. R. W. performed the calculations presented in the paper. R. W. prepared the manuscript, which was edited by all coauthors. R. W. designed and realized the hardware modifications necessary for the application presented in [\[Wol13b\]](#).

Wol12a: On-line separation of short-lived nuclei by a multi-reflection time-of-flight device,

R. N. Wolf, D. Beck, K. Blaum, Ch. Böhm, Ch. Borgmann, M. Breitenfeldt, F. Herfurth, A. Herlert, M. Kowalska, S. Kreim, D. Lunney, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, K. Zuber, [Nucl. Instrum. Methods Phys. Res., Sect. A 686, 82-90 \(2012\)](#)

R. W. performed the simulations and calculations. R. W., Ch. B., M. K., S. K., M. R. and F. W. performed the measurements. R. W. performed the data analysis and prepared the manuscript, all coauthors edited the manuscript.

Wol13b: ISOLTRAP's Multi-Reflection Time-of-Flight Mass Separator/ Spectrometer,

R. N. Wolf, F. Wienholtz, D. Atanasov, D. Beck, K. Blaum, Ch. Borgmann, F. Herfurth, M. Kowalska, S. Kreim, Y. Litvinov, D. Lunney, V. Manea, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, K. Zuber, [Int. J. Mass Spectrom. 349-350, 100 years of Mass Spectrometry, 123-133 \(2013\)](#)

R. W. performed the calculations. R. W., D. A., Ch. B., M. K., S. K., V. M., M. R. and F. W. performed the measurements. R. W. and F. W. performed the ^{137}Eu data analysis. R. W., M. R. and F. W. prepared the manuscript, all coauthors edited the manuscript.

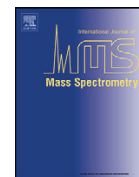
Wol13a: Plumbing Neutron Stars to New Depths with the Binding Energy of the Exotic Nuclide ^{82}Zn ,

R. N. Wolf, D. Beck, K. Blaum, Ch. Böhm, Ch. Borgmann, M. Breitenfeldt, N. Chamel, S. Goriely, F. Herfurth, M. Kowalska, S. Kreim, D. Lunney, V. Manea, E. Minaya Ramirez, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, and K. Zuber, [Phys. Rev. Lett. 110, 041101 \(2013\)](#)

R. W., Ch. Bö, Ch. B., M. B., M. K., S. K., V. M., E. M. R., D. N., M. R., J. S. and F. W. performed the measurements. R. W. and S. K. performed the ^{82}Zn data analysis. S. G. and N. C. performed the neutron star calculations. R. W., D. L., S. G. and L. S. prepared the manuscript, all coauthors edited the manuscript.

Obige Angaben werden bestätigt.

6.2 Static-mirror ion capture and time focusing for electrostatic ion-beam traps and multi-reflection time-of-flight mass analyzers by use of an in-trap potential lift, International Journal of Mass Spectrometry 313, 8-14 (2012)



Static-mirror ion capture and time focusing for electrostatic ion-beam traps and multi-reflection time-of-flight mass analyzers by use of an in-trap potential lift

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ABSTRACT

Capture and ejection of ions in electrostatic ion-beam traps and multi-reflection time-of-flight (MR-ToF) devices can be accomplished by pulsing the potential of only a single drift tube inside of the device in contrast to the conventional switching of the ion-mirror voltages. In addition to its simplicity, the new method allows to set the position of the time-focus plane at a given ion detector or ion-beam separator without the need of any further hardware. The position can be adjusted easily by the choice of the pulse height of the potential switch.

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1. Introduction

Electrostatic ion-beam traps are a recent development in the area of ion storage devices [1,2]. Similar to electrostatic ion storage rings [3–10] they provide confinement for ions of defined kinetic energies, independent of their mass. Although they are used for various applications in atomic and molecular science [11–17], electrostatic ion-beam traps are based on similar designs and operation principles. Typically, they consist of two ion-optical mirrors creating a potential barrier $qU(z)$ (with the ion charge q and the electric potential $U(z)$ as a function of position along the axis, z) at the ends of a drift section. To achieve storage the potential maxima have to exceed the ions' total energy, i.e., their kinetic energy E in the drift section,

$$qU(z) > E \quad (1)$$

with the potential energy in the drift section defined as zero. Thus, the ions are confined axially and bounce back and forth between the two ion mirrors, see Fig. 1. Transversal confinement is achieved by focusing elements in front of or integrated in the mirrors.

Such an arrangement was already reported five decades ago as "Farvitron" [18,19]. The two electrostatic mirrors were adjusted for identical revolution time independent of the ions initial param-

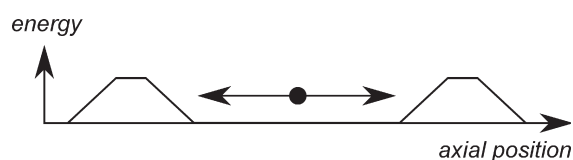


Fig. 1. Schematic illustration of the trapping criterion Eq. (1). Ions are confined axially between the two ion mirrors as long as their kinetic energy is lower than the potential maximum in the mirrors.

eters (for ions of the same mass-to-charge ratio). The ions were produced between the mirrors, i.e., already inside of the device and thus no voltage switching was necessary for the capture. This device was developed as a compact residual-gas analyzer in the UHV range. A related design was later proposed by Wollnik and Przewlaka [20] to enhance the performance of ToF mass spectrometers. In this application the ions fly along the same track multiple times while repeatedly being focused by two reflectron ion mirrors in an antiparallel, coaxial configuration. These multi-reflection ToF mass analyzers/spectrometers (MR-ToF MS) were later set up and successfully tested [21–24]. For appropriate electrode arrangements and voltages, high mass-spectral resolving powers $R = m/\Delta m$ have been reached [25–30] which make these devices attractive for precision mass spectrometry and the purification of ion ensembles with respect to their masses [28–30]. In the following we will not distinguish between the various applications and refer to all devices as MR-ToF MS.

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While in the case of the Farvitron the ions were created inside the trap they are nowadays in general produced in dedicated sources outside of the MR-ToF MS. Thus, they have to be injected into the device by sending them through one of the ion mirrors (the entrance mirror). Similarly, in most cases, the ions are finally ejected through a mirror (exit mirror) for detection or additional investigation. Entrance and exit mirrors may be identical, but in any case, during both injection and ejection, the total ion energy has to be higher than the maximum potential energy in the entrance/exit mirror in order to allow the ion passage. However, this is in contradiction to the trapping criterion, Eq. (1), i.e., different voltage settings are required for the storage and injection/extraction periods of an experimental sequence.

Injection and ejection of ions from MR-ToF MS has been achieved by switching the electric potentials of the entrance and exit mirrors, respectively, to appropriate lower values while the ions are passing. In the following, an alternative method is presented that simplifies the ion transfer to and from the MR-ToF MS considerably: instead of lowering a sufficient number of mirror electrodes, a single capture pulse is applied to just one drift tube between the ion mirrors. In addition, by varying the height of this voltage pulse applied to the in-trap drift tube, the ions' time focus can be adjusted with respect to the distance from the trapping region. Thus, by use of appropriate settings the resolving power for mass spectrometry or for the separation of particular ions of interest from contaminant species can be maximized. Furthermore, by use of the new in-trap potential-lift technique the trapping energy inside the MR-ToF device becomes independent of the transfer energy in the up-/downstream beamline. This decoupling of the MR-ToF MS from the beamline has several advantages, in particular with respect to their individual optimizations. Moreover, it is not necessary that injection and ejection pulses are of equal height which gives the possibility to change the kinetic energy after the MR-ToF device. In the following these features will be discussed in detail.

2. Comparison of ion capturing and ejection with switched mirrors and in-trap lift

Fig. 2 illustrates the differences between the conventional mirror-switching technique (Fig. 2 left) and the in-trap potential-lift technique (Fig. 2 right).

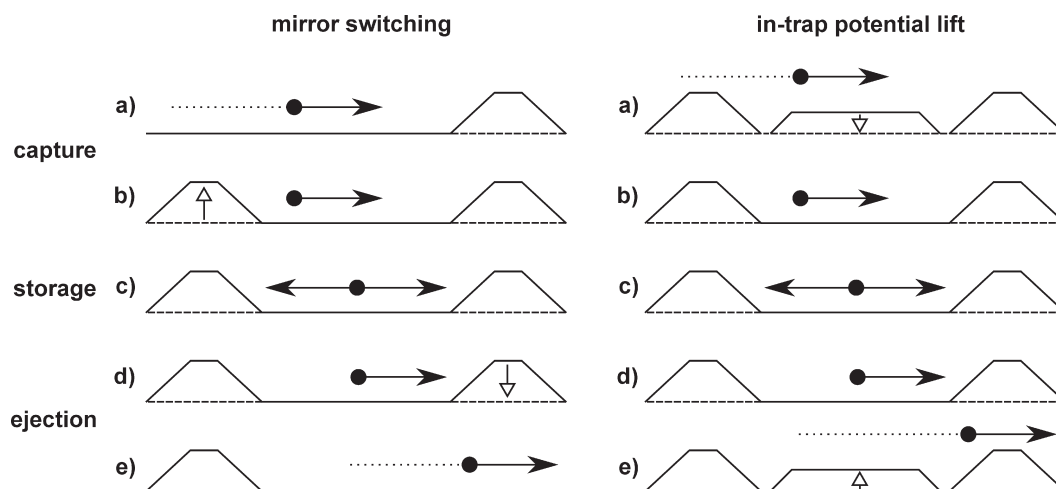


Fig. 2. Schematic illustration of ion capture, storage and ejection with a mirror-switching design (left) and with a static mirror design using an in-trap potential lift (right). For details see text.

2.1. Mirror switching

When the ion bunch arrives from the left, the potential of the entrance mirror is lowered such that the ions can pass the mirror and enter the trap region (a). After the ions passed the entrance mirror region, the mirror is raised back to the trapping value (b). Thus, externally created ions are captured (c). Similarly, the potential of the exit mirror is lowered below the total ion energy for ejection (d) and ions can escape via the exit mirror (e).

2.2. In-trap lift switching

In the framework of the new in-trap potential-lift method, the incoming ions have a kinetic energy $qU_{\text{transfer}} > qU(z)$ that exceeds the maxima of the mirror potentials. Thus, they pass the first mirror and enter the drift tube from the left (a). While the in-trap lift is activated, i.e., a potential U_{lift} is applied, their kinetic energy is reduced by qU_{lift} when entering the lift electrode. Once the ions have entered the lift it is deactivated, i.e., it is switched to ground potential, (b) and the ions are trapped as their energy is no longer high enough to pass the mirrors (c). After a certain number of reflections, the ions can be ejected by activating the in-trap lift voltage again while they are inside the drift tube (d). Thus, they are regaining enough energy to leave the MR-ToF MS (e).

Due to the strong focusing effect of the inhomogeneous electric field inside the mirror, special care has to be taken with respect to the injection ion optics in front of the device and the ejection ion optics behind the device. As an example a set of cylindrical lenses can be used to focus the beam near the turn-around point to prevent radial ion losses.

2.3. Comparison

The use of the in-trap potential lift instead of switched mirrors has several advantages. The nominal trapping energy inside the MR-ToF device becomes decoupled from the transfer energy of the beamlines in front and behind the MR-ToF MS. As long as the transfer energy of the ions in the beamline is high enough to let the ions pass the mirrors, they can be captured with the in-trap lift to any nominal trapping energy below the transfer energy. This simplifies the optimization of antecedent parts of the MR-ToF MS, because changing the transfer energy qU_{transfer} does not affect the trapping conditions as long as the difference $U_{\text{transfer}} - U_{\text{lift}}$ is kept

constant. On the other hand, the nominal trapping energy can be varied without the need of adjusting the ion-source potential or other ion-optical elements outside the MR-ToF MS. In addition, the tuning of the trapping energy is a very effective and efficient way to maximize the resolving power, as described in the next section.

On the other hand, the in-trap lift reduces the accepted time-of-flight distance of different species or, in case of extended bunches or continuous ion beams, the trapped number of ions. Depending on the particular layout of the device, the in-trap lift has a length of l_{lift} and incoming ions have a kinetic energy of E . The time of flight they travel through the lift electrode is

$$t_{\text{lift}} = \frac{l_{\text{lift}}}{v} = l_{\text{lift}} \cdot \sqrt{\frac{m}{2E}}. \quad (2)$$

For example, the velocity of light ions with a mass of $m = 28 \text{ u}$ and a kinetic energy of $E = 2 \text{ keV}$ is $v \approx 120 \text{ mm } \mu\text{s}^{-1}$. Thus they need $t_{\text{lift}} \approx 2.6 \text{ } \mu\text{s}$ to pass a $l_{\text{lift}} = 300 \text{ mm}$ drift tube. Edge effects have not been included in the present study. The edge field region of about one to two times the drift-tube diameter should be avoided, i.e., no ions should be within this region when the voltage is switched since they would acquire a different energy, between $q(U_{\text{transfer}} - U_{\text{lift}})$ and qU_{transfer} . Using fast solid-state switches with transition times well below $0.1 \text{ } \mu\text{s}$, the accepted time-of-flight distance is close to the drift time in the lift electrode minus the edge regions. Furthermore, ions that are far enough away from the accelerating or decelerating edge regions used to increase or decrease the ions' kinetic energy for ejection or injection (either in the mirror regions or somewhere else in the drift tube) are not affected by potential changes of the in-trap lift electrode. Therefore, once the distance between two species is large enough, only one of them can be addressed selectively for ejection or capturing.

The main difficulty in using an MR-ToF device for high mass-resolving power is to keep the mirror voltages as constant as possible over the whole measurement period. In general, the power supplies produce voltage fluctuations ranging from several hundreds of kilohertz down to a few microhertz. High-frequency fluctuation, ripple or noise is produced by the transformers used to create high voltages. On top of that, there may be a 50/60 Hz component originating from line voltage. This contribution is usually well known and specified for the device. In contrast, the low-frequency fluctuations or drifts below 1 Hz are often unknown. They are, in general, much higher in amplitude than the high-frequency components and very hard to suppress.

One has to distinguish, which of these components degrade the performance of the device in which state of operation. The high-frequency components change the potential distribution inside the mirrors in the time range of the ions' revolution time. Thus, they decrease the mass-resolving power achieved for a single-shot spectrum. The mid-frequency fluctuations change the potential distribution between several spectra, assuming experimental repetition rates between about 1 Hz and 1 kHz . This leads to a blurred accumulated spectrum that is often several times worse than the single-shot spectra. The low-frequency contributions change focusing and arrival times in the range of minutes to hours, which needs to be compensated by continuous recalibration and reoptimization of the apparatus.

There is an additional difficulty connected with the switching of the mirrors: Their potential distribution changes as a function of time after the mirror voltages are switched on. This is due to the drop of the high-voltage line caused by the load change, which is then compensated by the power-supply biasing the mirror-electrode voltage. Thus, there is a different optimal trapping potential distribution that compensates ToF aberrations best for every number of revolutions n . For an optimization of the resolving power the mirror-potential distribution has to be re-adjusted for each specific number of revolutions.

As an example, assume a mirror electrode switched from 0 for injection to V_{nom} nominal trapping voltage and a capacitance of the load circuitry of C_{load} . To recharge the electrode within a few hundred nanoseconds, an energy storage in form of a buffer capacitor may be used, for example $C_{\text{buffer}} = 100 \cdot C_{\text{load}}$. After switching, the electrode is only recharged to $V = V_{\text{nom}} \cdot C_{\text{buffer}} / (C_{\text{buffer}} + C_{\text{load}}) = 0.99 \cdot V_{\text{nom}}$. This voltage drop of 1% is then recharged by the powersupply, leading to a voltage change typically in the millisecond range, dependent on the current output and regulation speed of the power supply. Therefore, the potential distribution inside the mirror changes continuously for milliseconds, which decreases the performance considerably. In contrast, the in-trap lift is free from this drawback. The ions are only influenced by the pulsed drift electrode when they are entering or leaving the MR-ToF MS. This period is very short, of the order of a microsecond, and thus power-supply drifts due to load changes are negligible.

3. Time-of-flight focusing

To maximize the resolving power of a ToF MS, it is most important to control the position of the time-focus plane. Ideally, for mass analysis or ion separation it is placed on the detector or on the device for ion selection, respectively. The time-focus plane of an arbitrary ion source can be shifted in space by use of a reflectron [31] as well as by a multi-reflection ToF device as discussed in the following.

Ions (of the same mass-over-charge ratio) oscillate in an MR-ToF MS with a revolution time $T(E)$ depending on their kinetic energy E . More quantitatively, the relative revolution-time difference

$$\delta_T = \frac{T}{T_0} - 1 \quad (3)$$

of any ion with respect to a reference ion with a period T_0 can be expressed as a function of its relative kinetic-energy difference

$$\delta_E = \frac{E}{E_0} - 1 \quad (4)$$

with respect to the reference ion's kinetic energy E_0 . In the following, small deviations ($\delta_T \ll 1$, $\delta_E \ll 1$) are assumed which allows to limit the discussion about the energy dependent ToF differences to the linear coefficient $\partial\delta_T/\partial\delta_E$. Moreover, ToF aberrations with respect to geometric deviations are not considered, i.e., the ions are assumed to fly close to the optical axis of the system (for a more detailed treatment, see references [24,25,31]). The settings can either be adjusted to $\partial\delta_T/\partial\delta_E > 0$, i.e., ions with higher energy have a longer revolution time or to $\partial\delta_T/\partial\delta_E < 0$, i.e., ions with lower energy having a longer revolution time. Both of these cases can be achieved in an MR-ToF device either by adjusting the mirror-potentials for a fixed kinetic energy or by adjusting the kinetic energy for fixed mirror potentials. The latter is illustrated in Fig. 3. It is considerably easier than the first method as there is no need to adjust the mirror voltages once these values have been optimized. Depending on the value of $\partial\delta_T/\partial\delta_E$, the position of the time-focus plane of the MR-ToF device will change: For $\partial\delta_T/\partial\delta_E < 0$, it moves closer to and for $\partial\delta_T/\partial\delta_E > 0$ further away from the ion source. With the in-trap lift, the average trapping energy of an ion bunch is chosen when the ions enter the devices, $E = q(U_{\text{transfer}} - U_{\text{lift}} \pm \Delta U)$. In other words, the in-trap lift aids to navigate the ion bunch on the $\delta_T(\delta_E)$ curve, see Fig. 3.

Most MR-ToF mirror-potential optimizations aim at the energy-isochronous state $\partial\delta_T/\partial\delta_E = 0$ [22–30]. However, this condition is usually not easily achieved and conserved. In contrast, it will be shown in the following that states with $\partial\delta_T/\partial\delta_E \neq 0$ can be used to gain a comparably high mass-resolving power. In addition, this method relies on the variation of only one voltage which favors

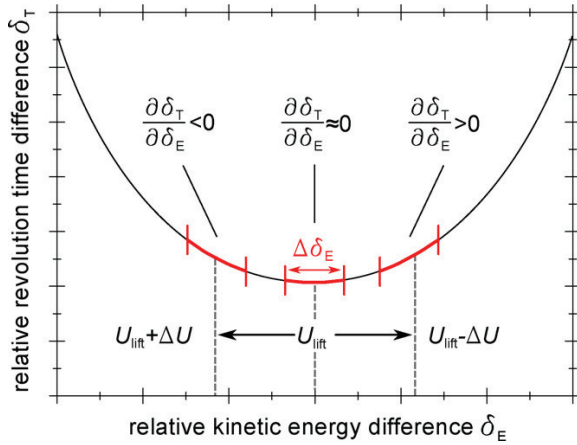


Fig. 3. Schematic example of the relative revolution-time difference δ_T as a function of the relative kinetic-energy difference δ_E . Sections of different slopes are indicated in red as well as the energy distribution $\Delta\delta_E$ of a bunch. To change the average kinetic energy of a bunch and therefore the corresponding ToF energy dispersion relation $\partial\delta_T/\partial\delta_E$, the in-trap potential lift voltage U_{lift} can be changed.

a quick optimization of an MR-ToF device. Moreover, for each number of revolutions n and each position of the ion source's time-focus plane, an optimal time focusing of the MR-ToF device can be achieved as long as there is a change in sign of $\partial\delta_T/\partial\delta_E$. The two cases of a positive and a negative ToF energy-dispersion coefficient are discussed in the following.

3.1. Positive ToF energy-dispersion coefficient, $\partial\delta_T/\partial\delta_E > 0$

Fig. 4 illustrates the case where the time-focus position of the ion source is located in front of the MR-ToF device. At passing this plane, the faster ions outpace the slower ions and the signal width Δt of the ion bunch starts to increase again due to the different kinetic energies originating from different axial starting positions in the ion source. Within the flight time t_1 from the time-focus position

of the source to the center of the MR-ToF device, the time spread increases by Δt_1 and in the flight time t_2 from the center of the MR-ToF device to the detector by Δt_2 . By capturing the ion bunch and storing at a kinetic trapping energy with positive energy-dispersion coefficient $\partial\delta_T/\partial\delta_E > 0$ (Fig. 3), the time difference between ions of different kinetic energies becomes smaller with each revolution. After a certain number of revolutions, i , a second time-focus plane can be found in the center of the device (Fig. 4 middle), i.e., the ToF deviation Δt_1 is compensated. However, after the ions are released towards the detector, they will again start to spread in time (Fig. 4 middle, right). Therefore, to compensate the ToF difference Δt_2 due to the flight path from the center of the analyzer to the detector, an additional number of revolutions j has to be performed before the ions are ejected (Fig. 4, bottom).

Let $\Delta t_s = \Delta t_1 + \Delta t_2$ be the ToF difference between the slowest and the fastest ion at the detector plane while “shooting through” the MR-ToF device with an activated in-trap lift (Fig. 4 top). This time difference has to be compensated in $n = i + j$ revolutions, thus $\Delta t_s/n$ per revolution. This leads to

$$\frac{\Delta t_s}{n} \stackrel{!}{=} T_0 \frac{\partial\delta_T}{\partial\delta_E} \Delta\delta_E \quad (5)$$

to compensate the ToF deviation with respect to energy.

To achieve a high mass-resolving power

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t}, \quad (6)$$

the absolute flight time $t = t_1 + nT_0 + t_2 = t_s + nT_0$ has to be high compared to the ToF distribution Δt at the detector plane. When the ToF deviations Δt_s originating from energy differences $\Delta\delta_E$ and initial ToF differences Δt_{th} (thermal ion distribution in source region, turn-around time in pulsed ion converters) are add up, the overall ToF distribution width is

$$\Delta t = \sqrt{\Delta t_{th}^2 + \left(\Delta t_s - nT_0 \frac{\partial\delta_T}{\partial\delta_E} \Delta\delta_E \right)^2} \quad (7)$$

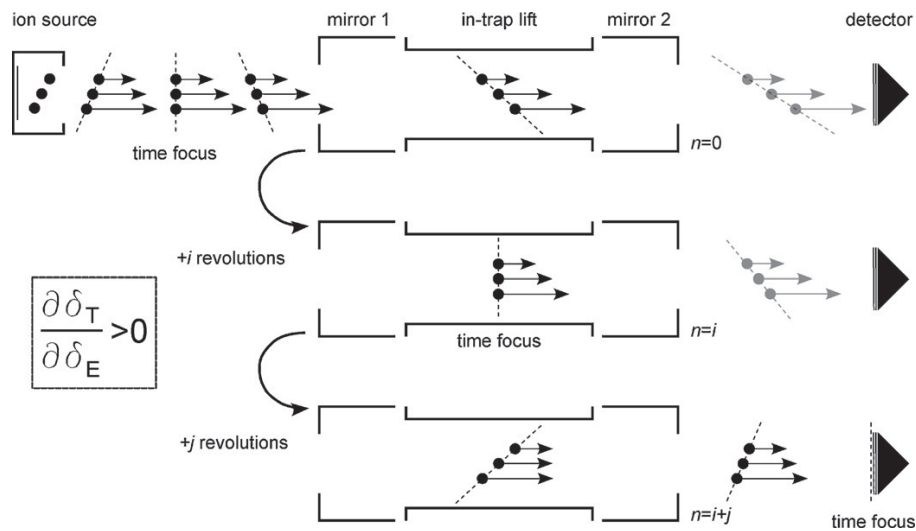


Fig. 4. Illustration of the time focusing by use of the in-trap lift for a positive ToF energy dispersion coefficient. The ion source's time-focus plane is located between the ion source and the MR-ToF device. After i revolutions, a time focus plane is found in the middle plane of the MR-ToF and after $i + j$ revolutions, the time focus has moved onto the detector. For further explanations see text.

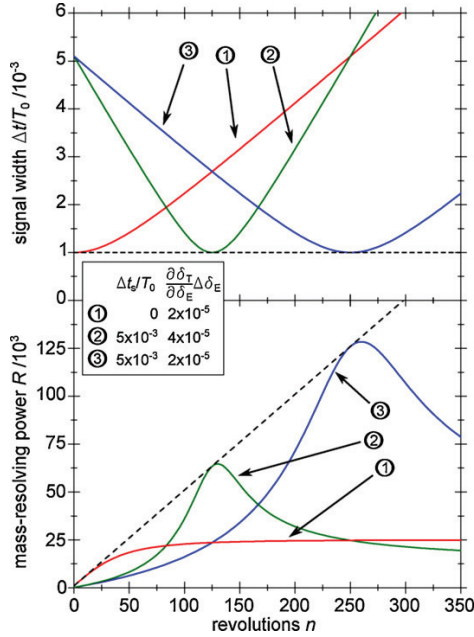


Fig. 5. Relative time width (top) and mass-resolving power (bottom) as a function of the number of revolutions according to Eqs. (8) and (9), respectively (for $t_s/T_0 = 2$ and $\Delta t_{th}/T_0 = 1 \times 10^{-3}$). The curves represent three sets of ToF energy dispersions $\partial \delta_T / \partial \delta_E \cdot \Delta \delta_E$ and time spreads $\Delta t_s/T_0$, the dashed curve represents the case of a ToF energy-isochronous state.

which results in a mass-resolving power

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t} = \frac{t_s + nT_0}{2\sqrt{\Delta t_{th}^2 + (\Delta t_s - nT_0(\partial \delta_T / \partial \delta_E) \Delta \delta_E)^2}} \quad (8)$$

Fig. 5 shows the relative ToF distribution width

$$\frac{\Delta t}{T_0} = \sqrt{\frac{\Delta t_{th}^2}{T_0^2} + \left(\frac{\Delta t_s}{T_0} - n \frac{\partial \delta_T}{\partial \delta_E} \Delta \delta_E \right)^2} \quad (9)$$

and the mass-resolving power R as a function of the number of revolutions inside the MR-ToF device.

The first curve (red) exemplifies the cases when the time-focus plane of the ion source was initially set on the detector, $\Delta t_s = 0$, and the ions with an energy difference $\Delta \delta_E$ are confined in the MR-ToF with a weak positive ToF energy dispersion, $\partial \delta_T / \partial \delta_E \cdot \Delta \delta_E = 2 \times 10^{-5}$. In this case, the signal width is increasing continuously, starting from its minimal value, i.e., the thermal time spread $\Delta t_{th} = 10^{-3} T_0$ (Fig. 5 top). This induces a fast, quasi-linear increase in mass-resolving power for the first few tens of revolutions until it converges slowly to the maximum for higher revolution numbers (Fig. 5 bottom). This maximum, for $n \rightarrow \infty$, is limited by the ToF energy dispersion $\partial \delta_T / \partial \delta_E \cdot \Delta \delta_E$. For the second (green) and third (blue) curves, different ToF energy coefficients and relative energy differences $\partial \delta_T / \partial \delta_E \cdot \Delta \delta_E$ were chosen for a fixed initial time-focus plane in front of the detector, such as the primary time focus position of the ion source. The ToF from this plane to the detector was set to $t_s = 2T_0$ and the ToF difference was chosen as $\Delta t_s = 5 \times 10^{-3} T_0$. The initially high time spread Δt is reduced until a minimum, namely Δt_{th} , is obtained according to

$$\frac{\partial \Delta t(n)}{\partial n} \Big|_0 = 0 \quad (10)$$

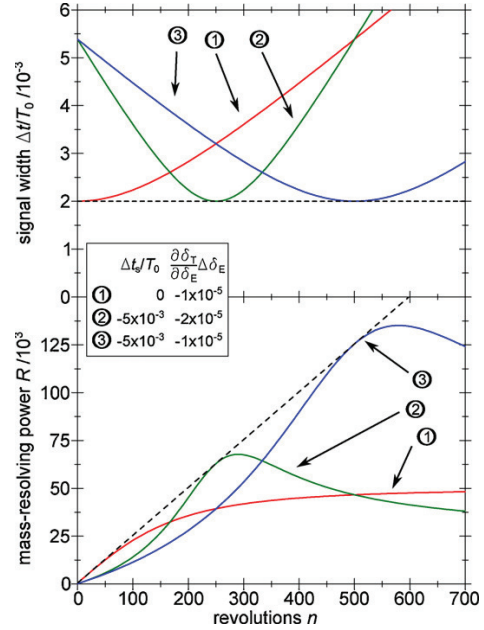


Fig. 6. Relative time width (top) and mass-resolving power (bottom) as a function of the number of revolutions according to Eqs. (8) and (9), respectively (for $t_s/T_0 = 2$, $\Delta t_{th}/T_0 = 2 \times 10^{-3}$). The curves represent three sets of different ToF energy dispersions $\partial \delta_T / \partial \delta_E \cdot \Delta \delta_E$ and time spreads $\Delta t_s/T_0$, the dashed curve represents the case of a ToF energy-isochronous state. Note the changed abscissa compared to Fig. 5.

after

$$n_{\Delta t} = \frac{\Delta t_s}{T_0} \frac{1}{(\partial \delta_T / \partial \delta_E) \Delta \delta_E} \quad (11)$$

revolutions where a mass-resolving power of

$$R(n_{\Delta t}) = \frac{t_s + n_{\Delta t} T_0}{2\Delta t_{th}} = \frac{1}{2\Delta t_{th}} \left(t_s + \frac{\Delta t_s}{(\partial \delta_T / \partial \delta_E) \Delta \delta_E} \right) \quad (12)$$

is reached. The maximum of the mass-resolving power is derived from

$$\frac{\partial R(n)}{\partial n} \Big|_0 = 0 \quad (13)$$

and is reached at

$$n_R = n_{\Delta t} \left(1 + \frac{\Delta t_{th}^2}{\Delta t_s (\Delta t_s + t_s (\partial \delta_T / \partial \delta_E) \Delta \delta_E)} \right) \quad (14)$$

revolutions with a value of

$$\begin{aligned} R(n_R) &= \frac{1}{2(\partial \delta_T / \partial \delta_E) \Delta \delta_E} \sqrt{\left(\frac{\Delta t_s + t_s (\partial \delta_T / \partial \delta_E) \Delta \delta_E}{\Delta t_{th}} \right)^2 + 1} \\ &= R(n_{\Delta t}) \sqrt{\left(\frac{\Delta t_{th}}{\Delta t_s + t_s (\partial \delta_T / \partial \delta_E) \Delta \delta_E} \right)^2 + 1} \\ &= R(n_{\Delta t}) + \frac{1}{2} R(n_{\Delta t}) \left(\frac{\Delta t_{th}}{\Delta t_s + t_s (\partial \delta_T / \partial \delta_E) \Delta \delta_E} \right)^2 - \dots \quad (15) \end{aligned}$$

In comparison to case 1 of Fig. 5, i.e., an ion-source time-focus position initially set on the detector and an MR-ToF with a weak positive ToF energy-dispersion coefficient where a maximum mass-resolving power of $R \approx 25,000$ is approached asymptotically, in case 2 the maximum $R \approx 65,000$ is reached at only $n_R \approx 130$ revolutions. This holds even for a ToF energy-dispersion coefficient further away from the energy-isochronous condition $\partial \delta_T / \partial \delta_E = 0$,

because the minimum time spread $\Delta t = \Delta t_{th}$ is found at a higher number of revolutions, $n_{\Delta t}$, and therefore at a longer flight time $t = t_s + n_{\Delta t} T_0$. In case 3, a mass-resolving power of $R \approx 128,000$ is reached at $n_R \approx 260$ revolutions with the same ToF energy dispersion as in case 1 (particularly the same potential distribution of the MR-ToF mirrors), but with the initial focus point in front of the detector. The number of revolutions where the minimum of Δt is found is proportional to the time spread gained between the ion source's time-focus point and detector, Δt_s , and inverse proportional to the ToF energy dispersion $\partial \delta_T / \partial \delta_E \cdot \Delta \delta_E$, Eq. (11).

3.2. Negative ToF energy-dispersion coefficient, $\partial \delta_T / \partial \delta_E < 0$

In the case that the time-focus plane of the ion source is behind the detector, the MR-ToF device can be operated in a negative-dispersion mode to move this plane onto the detector and achieve maximum mass-resolving power.

All considerations can be performed in analogy to the case discussed above, Section 3.1. Again, Δt_s is the time difference between the slowest and the fastest ion at the detector plane and is now of negative sign because the slowest ion still leads the bunch and therefore has the shortest flight time, $\Delta t_s < 0$. This has to be compensated within n revolutions, analog to Eq. (5). The slowest ions leading the ensemble will now have a higher revolution time inside the MR-ToF and therefore with each revolution the time-focus plane will move closer to the detector. With the given definition of Δt_s , which in this case turns negative, the mass-resolving power and relative ToF distribution width are still expressed by Eqs. (8) and (9). For consistency, the value of t_s is kept the same. The corresponding curves for graphical illustration are shown in Fig. 6. In order to create a focus point behind the detector, the time spread $\Delta t_{th}/T_0 = 2 \times 10^{-3}$ was doubled while the ToF energy dispersions were halved. All other considerations are the same as in the case of positive ToF energy dispersion, discussed above. As a consequence of the reduction of the ToF energy dispersions by a factor of two, the number of revolutions to reach the minimum time spread or maximum mass resolving power is doubled in cases 2 and 3 of Fig. 6.

4. Summary and outlook

By use of a pulsed drift tube between the ion mirrors of an MR-ToF device the operation can be significantly facilitated. Several implications of this approach have been outlined. In particular, there is no lack of mass resolving power with respect to the conventional method of switching of the mirror voltages. Moreover, the mass resolving power can be maximized for any number of revolutions, ion-source configuration and position as well as detector position by adjustment of a single, minor-critical voltage and without the need of further external ion optics. The new method is currently been tested at the on-line mass spectrometer ISOLTRAP [30].

In addition, the single ion description is presently extended by computer simulations including ion–ion interaction. In particular, a possible advantage of the negative dispersion mode may be the avoidance of ion bunch coalescence, previously referred to as self bunching [32–34], where a bunch of ions stays focused in time, regardless of their (close but different) mass-over-charge ratios. This effect of the Coulomb interaction and corresponding energy transfer between the ions is thought to appear in positive dispersion mode only. By avoiding this regime, higher ion numbers could be injected and mass analyzed or separated. Further investigations and comparisons of the modes discussed

based on Coulomb-interaction simulations are currently under way.

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On-line separation of short-lived nuclei by a multi-reflection time-of-flight device

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ABSTRACT

A multi-reflection time-of-flight (MR-ToF) mass analyzer has been integrated into ISOLTRAP, the precision mass spectrometer for on-line mass determinations of short-lived nuclides at ISOLDE/CERN. The new instrument improves ISOLTRAP by providing a fast separation of isobaric contaminant species as well as subsequent ion selection using the fast Bradbury–Nielsen gate. Suppression ratios of up to 10^4 and mass-resolving powers of over 10^3 have been reached in off-line experiments. Preliminary data from on-line applications illustrate the benefit and performance of the device and its potential in the context of the ISOLTRAP setup.

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1. Introduction

Penning traps have proven to be the devices of choice with regard to high-precision mass measurements of stable and exotic nuclei [1,2]. One of the primary conditions to minimize uncertainties of such a measurement is the availability of pure ion ensembles. In the presence of contaminations, the motion of the ions of interest is altered via the Coulomb interaction [3–6]. In general, the resulting deviations of the trapped-ion frequencies prevent precision experiments. Therefore, it is of utmost importance to ensure contamination-free measurements.

The yield of nuclei produced at on-line facilities decreases far away from the valley of stability whereas usually the ratio of the number of ions of interest and contaminant ions becomes more and more unfavorable. Penning-trap mass spectrometers suffer from

systematic effects in presence of contaminants either if the ratio is too large or if the required resolving power is high (usually the case for isobaric or isomeric purification). Thus, one of the most important challenges for ISOLTRAP [7] at ISOLDE/CERN [8], as for other comparable setups installed at on-line facilities [9–14], is the efficient transfer of the ions of interest to the location of the precision measurement and the elimination of all contaminating species. Such contaminations may be delivered from the on-line separator directly or can be created inside the apparatus as products from nuclear decay [15] or charge exchange [16], in particular in the case of stopping in a gas cell [17].

In addition, for nuclides with half-lives on the order of the measurement duration or below (≤ 1 s), the ion preparation and purification should be as fast as possible to avoid decay losses. Currently, this contamination removal is performed by mass-selective resonant buffer-gas cooling in a Penning trap [18] which requires several 100 ms. Therefore, a faster mass selection would be a significant improvement for experiments on nuclides with short half-life. For this purpose, a second isobar-purification

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system was developed and installed in 2010 at ISOLTRAP. It is based on a multi-reflection time-of-flight (MR-ToF) technique [19–25] in combination with a fast Bradbury–Nielsen gate (BNG) [26,27] and provides a significantly faster purification without degradation in effectiveness. The system can be used either as

- an auxiliary device for contamination removal in combination with the existing preparation Penning trap,
- an exclusive mass separator for very short-lived nuclides, or
- a fast mass spectrometer in its own right for very short-lived nuclides.

In the following we report on the implementation of the first MR-ToF mass analyzer at a Penning-trap facility for exotic nuclei. In addition to the design of the MR-ToF section and the BNG, the results of off-line as well as of the first on-line experiments on mass separation of short-lived nuclei are presented.

2. Principle of isobar purification with the MR-ToF/BNG device

The MR-ToF/BNG isobar-purification principle is illustrated in Fig. 1. The setup consists of an MR-ToF section for the separation of the different ion species with respect to their different mass-over-charge ratios and thus different arrival times at the BNG where the actual selection of the species of interest takes place.

2.1. MR-ToF mass separator

The MR-ToF device uses two electrostatic coaxial ion mirrors to reflect charged-particle bunches. Thus, the mirrors act as an anti-parallel arrangement of two ToF reflectrons and create a multiply-folded flight path [19]. The resulting trajectories can be several hundreds or thousands of meters long all within a compact device of 1 m length. All ion species i of mass m_i and charge state z_i start on a well-defined potential U and gain the same average kinetic energy per charge state $E/z_i = eU$, where e is the elementary charge. Due to their different velocities $v_i = \sqrt{2Uez_i/m_i}$ the ions have m_i/z_i -specific revolution times $T_i \propto v_i^{-1} \propto \sqrt{m_i/z_i}$. Thus, the difference in ToF between two ion species $\Delta t_{ij} \propto n(T_i - T_j)$ increases linearly with the number of revolutions n . When the ToF difference becomes larger than the time width Δt of the individual ion bunches, $|\Delta t_{ij}| > \Delta t$, the ions can be ejected from the MR-ToF device towards the BNG.

To achieve the high resolving power $R = m/\Delta m = t/(2\Delta t)$ necessary to separate, e.g., isobaric species, the total time of flight (ToF) t has to be long compared to the temporal width of an ion bunch Δt . The total ToF can be extended by increasing the number of revolutions n of the ions in the MR-ToF, since $t \propto nT$. For an

increase in resolving power, the signal width has to stay constant. Ideally, the complete ToF system consisting of the flight path from the ion source to the MR-ToF device, the trajectory inside the MR-ToF device itself, and the flight path from the MR-ToF device to the detector, or to a device for selection, is isochronous with respect to the ion energy. In other words, the flight time should be independent of the different kinetic energies of the ions, originating in particular from different starting positions in the ion source. This can be achieved by applying optimized mirror potentials [28] in combination with the use of a pulsed drift electrode between the ion mirrors that also serves as an injection and ejection device [29]. The upper limit for the time-of-flight separation is given by the maximum time difference between species before they start to approach each other again, $\Delta t_{ij} < T_{ij}/2$.

2.2. Bradbury–Nielsen gate

The principle of operation of the BNG [26,31] is illustrated in Fig. 2. It consists of two sets of parallel wires in a planar arrangement such that neighboring wires belong to different sets. During the passage period of unwanted species, the gate is activated (closed), i.e. voltages of alternating sign but equal value are applied to neighboring wires, which results in the deflection of the charged particles. When the ions of interest arrive the gate is deactivated (open) until they have passed. A BNG achieves higher time resolutions than parallel-plate deflectors or similar arrangements. The electric field of the activated wires decays exponentially with increasing distance from its plane [31], and thus is negligible at the distance of one wire spacing d in the longitudinal direction, as shown in Fig. 2 (right). The achievable time resolution τ_{res} , i.e. the ion flight time through the region of the deflecting electric field can be approximated by the ion flight time through twice the wire spacing [31]

$$\tau_{res} = \frac{2d}{v_i} = 2d \sqrt{\frac{m_i}{2z_i e U}}. \quad (1)$$

Thus, for singly charged ions with $m=100$ u starting from a potential of $U \approx 3$ kV and passing a BNG with $d=0.5$ mm, the time resolution is $\tau_{res} \approx 13$ ns. For the selection of ToF-separated ion species, the transition times of the pulsed voltages τ_{trans} needed to change the state of the gate from closed to open and vice versa has to be added. Ions that penetrate the field region while the gate potentials are being switched experience a change in kinetic energy which thus broadens the energy distribution of the ensemble. To deflect one species without disturbing the other, the minimum time-of-flight difference between the end of the first ion bunch and the beginning of the second ion bunch at a given relative signal height should therefore not be smaller than $\tau_{res} + \tau_{trans}$. Thus, this condition does not include the bunch width.

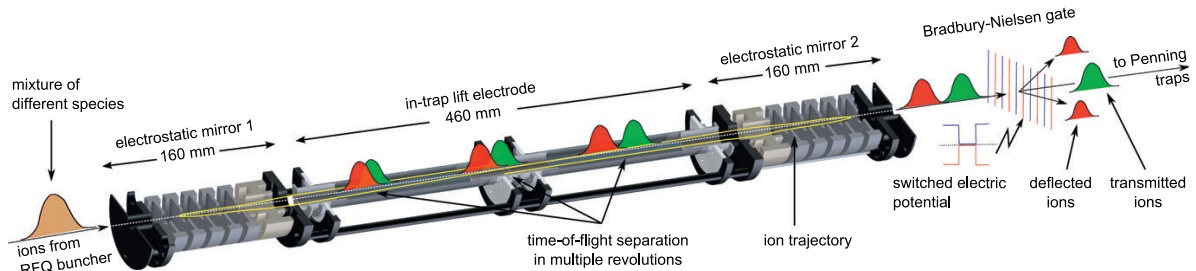


Fig. 1. Illustration of the isobar-separation principle. A mixture of two species (colored Gaussian-shaped ToF distributions) enters the MR-ToF mass separator (sectional view). The nuclides are separated due to their different mass-over-charge ratios in multiple revolutions inside the MR-ToF mass separator. After ejection, they pass the Bradbury–Nielsen gate where the contaminant species are deflected.

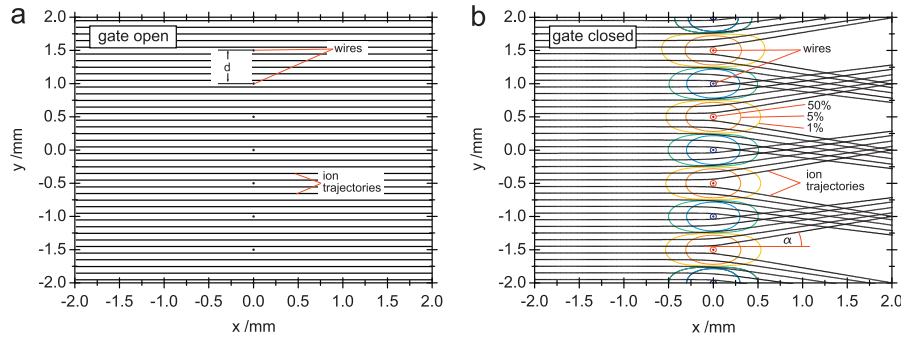


Fig. 2. Simulation (SIMION 8 [30]) of the Bradbury–Nielsen gate operation with a wire distance of $d=0.5$ mm. For ion transmission (left), both wire sets are at the same voltage, ideally the potential of the beam line. To deflect the ions (right), the wires are activated. Note the equipotential lines of 50%, 5% and 1% of the wire voltage.

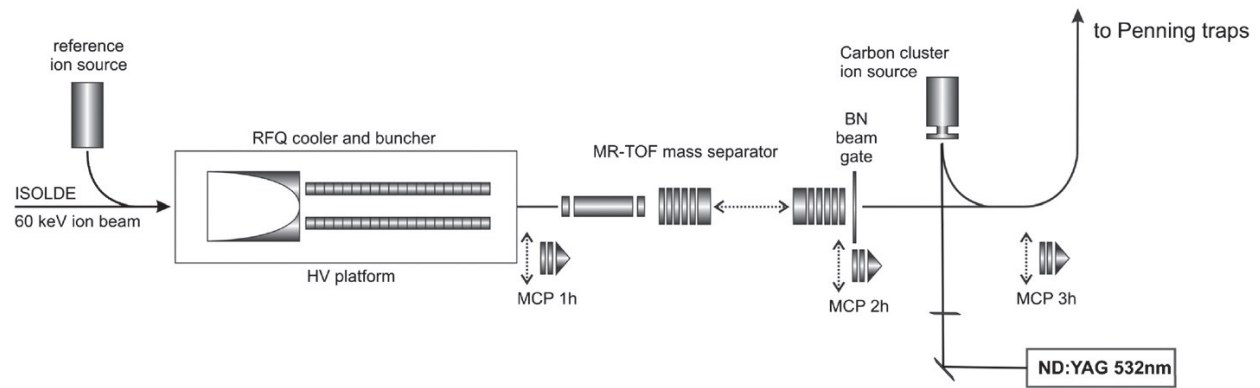


Fig. 3. Layout of the horizontal ISOLTRAP beam line after implementation of the MR-ToF mass separator between the RFQ buncher and the carbon-cluster ion source in front of the bender that directs the ion bunches towards the vertical Penning traps (not shown). For details see text.

3. Overview of the ISOLTRAP setup and beam purification in a Penning trap

The layout of the horizontal ISOLTRAP beam line including the MR-ToF mass separator and the BNG is illustrated in Fig. 3. ISOLTRAP consists of a radiofrequency-quadrupole (RFQ) ion-beam cooler and buncher [32], the new MR-ToF/BNG section, and in the vertical part – not shown in the figure – two Penning traps for the preparation [33] and precision [34] measurement steps mentioned above as well as a tape-station [35] for trap-assisted nuclear-decay studies. Two off-line ion sources (alkali ion source and carbon-cluster ion source [36]), one in front of the RFQ buncher and one in front of the preparation Penning trap deliver the stable reference nuclides for the mass measurements. The status of the setup before the implementation of the MR-ToF mass separator has been described in detail in [7]. Only a short review of the parts needed for this work will be given in the following.

The low-energy part of the ISOLDE facility delivers a quasi-continuous ion beam with an ion energy of up to 60 keV, in most cases consisting of a mixture of isobaric short-lived and stable nuclides as the separation resolving power of the sector-field magnets is limited to about 7000 without higher-order corrections [8]. For mass measurements, the ions are decelerated electrostatically in front of the ISOLTRAP RFQ buncher located on a high-voltage platform floatable up to 60 kV. To accumulate ions and to reduce the beam emittance, the RFQ buncher is filled with high-purity helium buffer gas of 10^{-2} mbar. After an accumulation and cooling period, thermalized ion pulses are reaccelerated to 3.1 keV by injection into a switchable drift electrode which transfers the ions to ground potential of the

beam line. In the adjacent MR-ToF mass separator, a mass-over-charge separation of the different species can be performed, which are then ejected onto the BNG for selection of the species of interest. After the ions have passed the BNG they are deflected by 90° to the vertical direction and are injected into the first Penning trap for further purification and cooling as a preparation step for the actual precision mass measurement in the second Penning trap.

The standard method to purify the ion ensemble from contaminating species is the mass-selective resonant buffer-gas centering (so-called “quadrupolar cooling”) in a preparation Penning trap: By use of an azimuthal dipolar radiofrequency (RF) excitation the magnetron radius of all ions is simultaneously increased to a value higher than the radius of an aperture used for ejection (in the case of ISOLTRAP, $r=1.5$ mm) in the endcap electrode of the trap. To re-center only the species of interest mass-selectively, the slow magnetron motion is converted into the fast cyclotron motion by applying an azimuthal quadrupolar RF excitation at the cyclotron frequency $\nu_c = qB/(2\pi m)$, with mass m , charge q , and magnetic field strength B . Due to the damping by buffer-gas collisions, the fast cyclotron motion is effectively cooled, leading to a decrease of the cyclotron radius. When the ions are ejected axially, e.g., towards the measurement Penning trap, only the centered ones pass through the aperture, whereas the others collide with this electrode and are lost. Mass-resolving powers of up to 300,000 have been reached with ISOLTRAP’s preparation Penning trap [37]. However, such high resolving powers are achieved at very low helium-gas pressures only, where this essential preparation step takes several hundreds of milliseconds. In contrast, if the ensemble is already

contamination-free and mass separation is not needed, the cooling period can be reduced to only a few milliseconds (at increased helium pressure).

After the preparation, the ions are transferred to the precision Penning trap for the determination of their cyclotron frequency ν_c in a homogenous magnetic field. To this end, a quadrupolar excitation is performed at several frequencies around the expected cyclotron frequency. In resonance, i.e. at the cyclotron frequency, the magnetron motion is converted to cyclotron motion [38]. This ion-motional response is probed by axial ejection of the ions and a subsequent measurement of their drift time to a micro-channel plate (MCP) detector outside of the strong magnetic field. In the axial magnetic-field gradient the radial (cyclotron) motion is converted to axial kinetic energy, i.e. the conversion becomes visible as a reduction in the time of flight. For short-lived nuclides, a statistical uncertainty in the order of $\delta\nu_c/\nu_c \approx 10^{-9} \dots 10^{-8}$ can be reached by use of this time-of-flight ion-cyclotron resonance (ToF-ICR) technique [38–40] and the Ramsey excitation scheme [41]. The magnetic-field strength is deduced from the cyclotron frequency measured for a reference ion with a well-known mass.

If necessary, the measurement trap itself can also be used for contaminant reduction, in particular for ions with very close-lying masses: Dipolar RF excitation at the reduced cyclotron frequency can be applied to resolve and remove ions of different isomeric nuclear states [42,43] with a mass-resolving power that is roughly proportional to $\nu_c T_{rf}$ where T_{rf} is the excitation time. As an example, mass-resolving powers of 10^7 have been reached at ISOLTRAP for excitation durations of $T_{rf} = 12$ s for $A \approx 85$ at $\nu_c \approx 1$ MHz. This technique is an important feature also for trap-assisted nuclear spectroscopy with the recently installed tape-station setup as it allows beta and gamma spectroscopy of pure isomeric ion ensembles.

4. Experimental setup

Fig. 4 shows an overview of the ISOLTRAP MR-ToF purification system which consists of the MR-ToF mass analyzer, injection and ejection ion optics, the BNG and an ion detector. In the following, the technical details of the different parts will be described.

4.1. Vacuum system

The MR-ToF device is installed in a CF100/200 cross of 800 mm length while the adjacent parts are installed in CF100 double-cross chambers. The background pressure determines the mean free path of the ions traveling in the MR-ToF device and thus the transmission on long flight paths. Therefore, an ultrahigh vacuum has to be achieved to avoid ion losses. The injection side is evacuated with a 350-l/s turbo-molecular pump (TMP 1) while at the ejection side a 150-l/s pump (TMP 4) is used. In the MR-ToF central section, a 1500-l/s pump (TMP 2) is used in series with a 50-l/s pump (TMP 3) to achieve a low pressure at high gas loads.

Two 250-l/min oil-free scroll pumps maintain a prevacuum of about 1×10^{-3} mbar. With closed valves to the neighboring sections, the background pressure of the system is 2×10^{-10} mbar measured at the center of the MR-ToF device, 6×10^{-9} mbar at the injection side and 1×10^{-8} mbar at the ejection side. Under normal operation conditions with helium-gas flow from the RFQ buncher ($\approx 1 \times 10^{-2}$ mbar(He)) and the preparation Penning trap ($\approx 5 \times 10^{-4}$ mbar(He)), the pressure at the central MR-ToF section is 5×10^{-9} mbar, and 3×10^{-7} mbar as well as 1×10^{-7} mbar at the injection and ejection crosses, respectively. In order to block the helium gas entering into the MR-ToF section from both sides, two adjustable iris apertures are installed at the injection side and another on the ejection side. A reduction of a factor of six in pressure at the MR-ToF section could be observed by closing the irises to their minimal permissible diameter (≈ 3 mm) without a decrease in the ion transmission.

4.2. Injection and ejection ion optics, MCP detector

To control the radial position and diameter of the injected ion bunches, two sets of x-y steerers and an einzel lens are installed in front of the MR-ToF device (see Fig. 4). Small deflection potentials in the range of a few volts are sufficient to maximize the transmission efficiency of the MR-ToF. These voltages depend on the mass-over-charge ratio of the injected ions. They correct the trajectory variations due to different ejection conditions of the RFQ buncher as well as the mass-dependent transfer due to the residual magnetic stray field of the 4.7 T-magnet of the preparation Penning trap. The einzel lens is used to focus the beam into the activated ion mirror near the reflection point.

In analogy, an einzel lens at the ejection side collimates the ion bunches for further transfer to the MCP detector or to the BNG. The latter devices are both installed on a movable feedthrough (denoted by the arrow in Fig. 4) to allow choosing between either ion monitoring or selection.

4.3. MR-ToF setup

The mechanical construction of the MR-ToF device has already been described in detail in [28] and will only be summarized here. It consists of two ion mirrors of six electrodes each, which are separated by a drift section (with length of 460 mm with an inner diameter of 26 mm), which is used as an in-trap lift [29] for ion injection, ejection and time-of-flight focusing. A sketch of the MR-ToF and a typical potential configuration is shown in Fig. 5. The voltages applied to the mirror electrodes are supplied by high-precision modules (ISEG DPS-series) controlled via a 16-bit digital-to-analog converter (DAC, National Instruments NI-6703). All other voltages, for the injection/ejection ion optics and the in-trap lift electrode, are generated by further modules (Applied Kilovolts HP-series) and controlled via the same DAC.

The performance of the MR-ToF separator depends critically on the potentials applied. In particular, a given optimized mirror-potential distribution has to stay as constant as possible over the whole measurement period. For a discussion on how the different

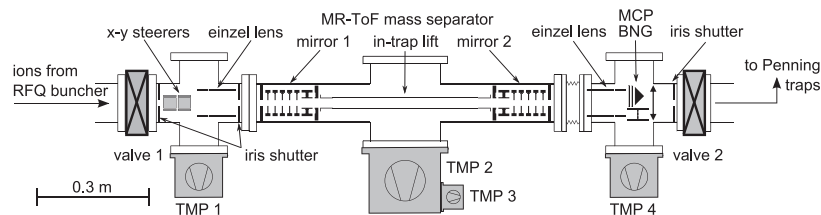


Fig. 4. Time-of-flight isobar-separation section of ISOLTRAP including injection and ejection ion optics and vacuum system.

spectral components of voltage fluctuations lower the performance, see [29]. The controls of the setup are fully integrated in the experimental control system used at ISOLTRAP (ISOLTRAP CS [44,45]). The timing signals for switching the electrode voltages are generated by an FPGA card (National Instruments NI-7811R) programmed as a pulse-pattern generator [46]. This controls among others the activation and deactivation of the in-trap lift electrode via a solid-state MOSFET switch (Behlke HTS 61-03-GSM).

4.4. Bradbury–Nielsen gate

The BNG construction and weaving mechanism is similar to the one presented in [31]. The gate is constructed by weaving 10 μm diameter gold-plated tungsten wire on a frame of poly-ether ether ketone (PEEK) as shown in Fig. 6. Small grooves are milled on the top side of the frame for parallel guiding of the wires at a defined spacing of $d=0.5$ mm. On the front side, a comb structure is machined to enhance the distance for the electrically separate contacting. It guides the odd-numbered wires onto the bottom while the even-numbered wires stay on top for some more distance. To fix the wires, strips of PEEK and copper are glued onto the substrate with non-conductive and conductive vacuum-compatible adhesives, respectively. Finally, the wires on the bottom are removed. The gate is placed between two drift

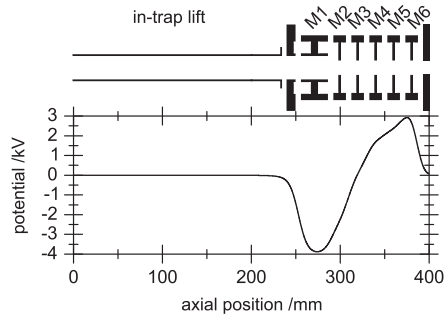


Fig. 5. Axial potential distribution in the MR-ToF device. The first two mirror electrodes (M1 and M2) form a negative electrostatic lens to create stable trajectories. The last four (M3–M6) are on positive voltage to reflect the ions and compensate their energy-dependent ToF. The in-trap lift is on ground potential while the ions are stored and will only be switched for in- or ejection.

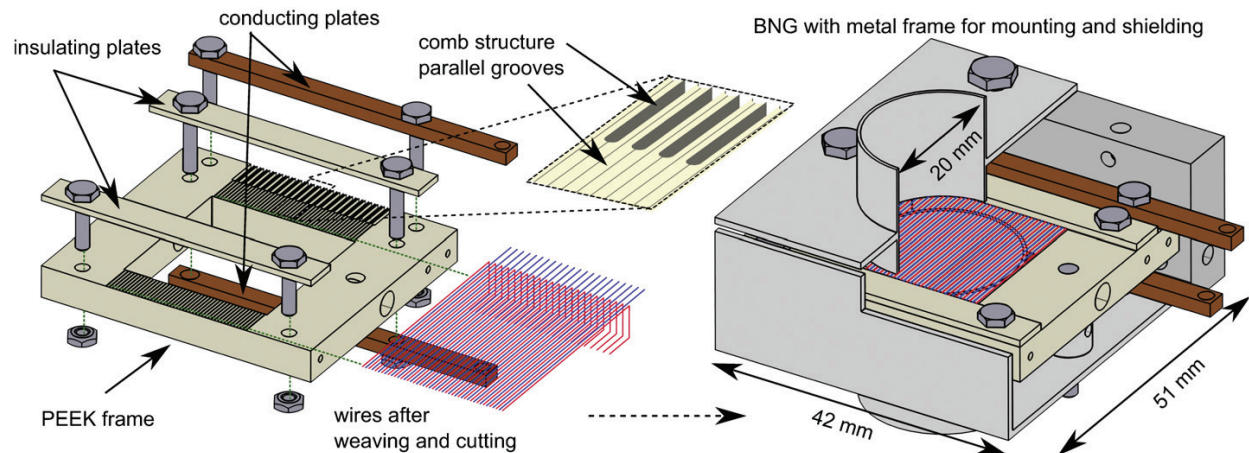


Fig. 6. Construction and mounting mechanism of the BNG. Left: Exploded view of the BNG showing the PEEK frame, the woven and cut wires and the four plates for fixing and contacting. Middle: guiding grooves (magnified) and comb structure. Right: final BNG assembly and partly sectional view of the BNG metal frame for mounting and shielding of the insulating parts.

tube electrodes to prevent ions from hitting non-conductive surfaces, thus charging them. After curing the adhesives and baking the gate at temperatures around 100 $^{\circ}\text{C}$, no vacuum degradation could be observed. The present gates have an active diameter of 20 mm. Their transmission is about 95% in the “open” state, which is close to the calculated open area of 98%. The advantage of this method of production is that the PEEK frames for wire spacings down to 0.5 mm can be produced with standard milling techniques and are therefore easily replaceable.

As described above, the time resolution of a BNG depends on the wire spacing and the voltage transition time on the wires between the states “closed” and “open” and vice versa. Fast push-pull MOSFET switches (model AMX-500-3, CGC Instruments) with voltage transition times of $\tau_{\text{trans}} \approx 20$ ns are used to switch voltages in the range of $U_{\text{BNG}} = \pm 250$ V.

5. Off-line operation and performance

To reach the full operation capability of the MR-ToF/BNG combination, the RFQ buncher had to be modified to provide bunches with a shorter time structure. By decreasing the length of the axial RFQ trapping region from 20 mm to 10 mm and by increasing the extraction field strength from ≈ 3 V/mm to ≈ 20 V/mm, the time structure of the bunches measured in front of the MR-ToF (MCP1h, Fig. 3) was reduced from about 1 μs to 60 ns (FWHM) at a cooling and bunching efficiency of 10–20%, i.e. the same as reported for the earlier buncher parameters. An initial short time width is necessary to achieve a fast separation in the MR-ToF section. The energy distribution of the ion bunches was determined by use of a repelling-grid arrangement in front of MCP2h (see Fig. 3, with the MR-ToF separator deactivated). The measured energy distribution of about $\Delta E_{90\%}^{\text{buncher}} \approx 60$ eV is within the energy acceptance of about 100 eV of the preparation Penning trap.

5.1. MR-ToF performance

The mean kinetic energy of the ions in the transfer beam line and in the MR-ToF separator is $E_{\text{transfer}} = 3.1$ keV and $E = 2.1$ keV, respectively, where the reduction is due to the in-trap lift method [29]. Mass-resolving powers of up to $R_{\text{FWHM}} = m/\Delta m \approx 2 \times 10^5$ at storage times of $t \approx 30$ ms (about 1000 revolutions for ^{133}Cs or 2000 revolutions for ^{39}K ions, respectively) have been achieved. As an example, Fig. 7 shows the corresponding time-of-flight

spectrum for ^{39}K . It was acquired with a multi-channel analyzer adding 200 single-shot spectra of MCP2h. The repetition rate was approximately 1.5 Hz. Thus, the recording time of ≈ 120 s in this example is short compared to the typical duration of mass measurements with the precision Penning trap which can take several hours if very low statistical uncertainties are to be obtained. Therefore, long-term voltage drifts on the MR-ToF electrodes have to be considered as well (see next paragraph). Fig. 8 (top) shows the mass-resolving power as a function of the time of flight for ^{133}Cs ions. Eq. (8) from [29]

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t} = \frac{t_s + nT_0}{2\sqrt{\Delta t_{th}^2 + (\Delta t_s - nT_0)(\partial\delta_T/\partial\delta_E)\Delta\delta_E)^2}} \quad (2)$$

was used to fit the mass-resolving power for the fixed experimental parameters, i.e. the revolution time $T_0 \approx 27.7 \mu\text{s}$ and the ToF difference between the ion source's time-focus plane and the detector, $t_s \approx 30 \mu\text{s}$. The resulting free fit parameters are: the turn-around time $\Delta t_{th} \approx 58$ ns in the ejection section of the RFQ buncher when ejecting (which is the lower limit of the time width), the additional time-spread due to MR-ToF-external flight time $\Delta t_s \approx 105$ ns, and the ToF-dispersion relation with respect to energy $(\partial\delta_T/\partial\delta_E)\Delta\delta_E \approx 5 \times 10^{-6}$ (see [29] for further details).

The dependence of the average time of flight on the mirror voltages is shown in Fig. 9. Variations of mirror electrode 5 (M5) have the largest influence, namely $(\delta t/t)/(\Delta V/V|_{M5}) = 0.5$ because it is closest to the reflection point and therefore defines the penetration depth. For example, a fluctuation of $\Delta V/V|_{M5} = 5 \times 10^{-6}$ in the spectrum shown in Fig. 7 would result in a ToF center shift of $\delta t/t = 2.5 \times 10^{-6}$, i.e. $\delta t \approx 75$ ns for a flight time of $t \approx 30$ ms. Thus, the signal width would be doubled and hence the mass-resolving power reduced by a factor of 2. As mentioned above the Penning-trap mass measurements can last several hours, and thus the possible shift of the time of flight of the ions of interest in the MR-ToF has to be taken into account. The major contribution to the long-term instability is given by the temperature variation in the laboratory of ≈ 1 K between day and night and a temperature coefficient of the voltage sources of $\Delta V/V = 5 \times 10^{-5}/\text{K}$.

The transmission of the MR-ToF is mostly affected by the injection focusing and steering of the pulses delivered by the RFQ buncher. Small changes in the focus position and angle with respect to the optical axis can increase the losses in the first few

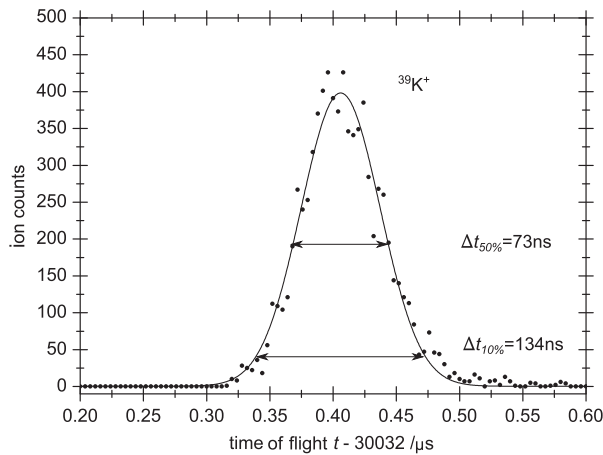


Fig. 7. Ion counts (dots) and Gaussian fit (line) of $^{39}\text{K}^+$ ions from the reference ion source at a total flight time of about $t \approx 30$ ms (corresponding to 2000 revolutions at mass $A=39$ in the MR-ToF separator). The mass-resolving power is $R_{FWHM} \approx 2 \times 10^5$ and $R_{10\%} \approx 1 \times 10^5$.

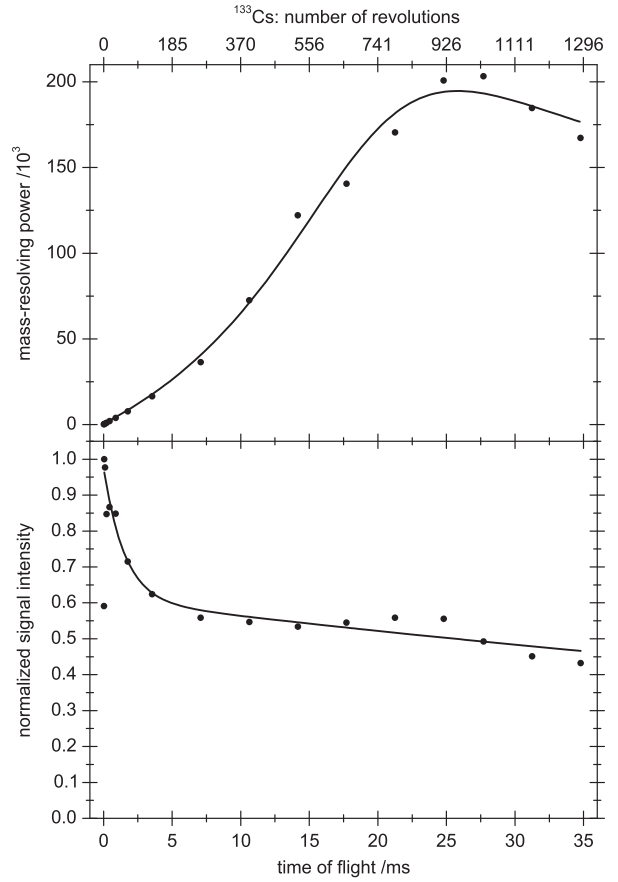


Fig. 8. Mass-resolving power (top) and signal intensity (bottom) of ^{133}Cs with respect to the time of flight (number of revolutions) in the MR-ToF mass separator. The solid lines are fits to the data points. See text for details.

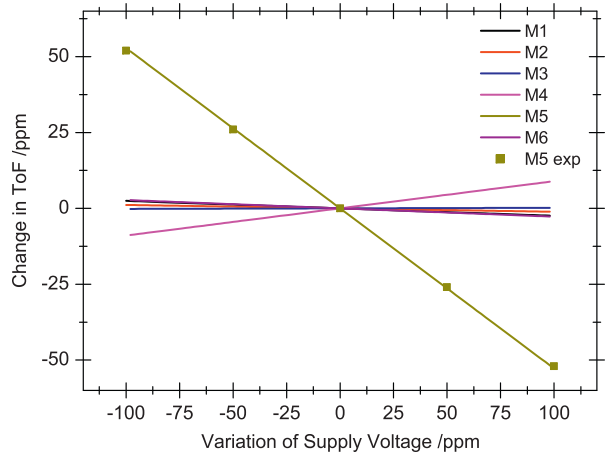


Fig. 9. Time-of-flight deviation from the nominal value as a function of the mirror-electrode voltages (mirror electrodes 1–6 = M1–M6) for a given kinetic ion energy. For the electrode numbering, see Fig. 5. The simulations agree with measured data points (of ^{133}Cs) for the electrode M5.

milliseconds of trapping as shown in Fig. 8 (bottom). The solid line shows an exponential fit to the data with two constants, $\tau_1 \approx 1.5$ ms and $\tau_2 \approx 130$ ms. The fast losses are most likely a

result of a mismatch of the pulse emittance and the capture acceptance of the MR-ToF separator. Furthermore, residual stray magnetic fields in the order of 1 mT due to the superconducting magnet of the preparation Penning trap and due to the steel girders of the surrounding support structures of ISOLTRAP influence the injection and trapping efficiency mass dependently. A transmission of about 50% can be maintained even for storage times up to a few tens of milliseconds. Losses due to collisions with residual gas have a minor influence on the transmission due to the pressure of $< 10^{-8}$ mbar, resulting in the high time constant τ_2 .

The overall efficiency of the ISOLTRAP setup for low ion yields is on the order of 0.5–2%, deduced from the ratio of the beam current in the ISOLDE beam line in front of the RFQ buncher and the ion events counted on the MCP detector used for the mass measurement. The transmission loss in the MR-ToF mass separator of 50% is thus small compared to other section of ISOLTRAP.

5.2. BNG performance

The suppression factor of ions as a function of the wire-voltage difference of the BNG is shown in Fig. 10. ^{85}Rb ions from the alkali reference ion source were ejected towards the BNG while it was either “open” or “closed”. The transmitted ions were captured in the preparation Penning trap and ejected towards an MCP detector after the cooling procedure as explained above. Below $\Delta U_{\text{BNG}} = 440$ V, corresponding to $U_{\text{BNG}} = \pm 220$ V on the wire sets, the suppression is low since the deflection angle is too small to

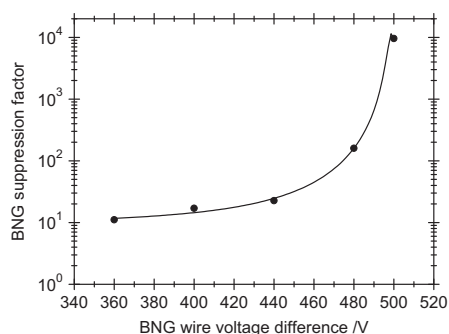


Fig. 10. BNG suppression (ratio of ^{85}Rb ions transmitted for BNG open vs. BNG closed) as a function of the wire voltage difference, solid line to guide the eye. For the determination of the deflection efficiency the dark-count rate has been subtracted.

prevent the ions from being transported by the subsequent ion optics into the preparation Penning trap. Above deflection voltages of $\Delta U_{\text{BNG}} = 480$ V the ratio between ions that passed the “closed” and the “open” gate decreases by orders of magnitudes, i.e. the deflection angle is sufficient. A reduction of four orders of magnitude has been achieved at $\Delta U_{\text{BNG}} = 500$ V, which was the maximum voltage used to avoid damaging the BNG. However, this is no general limit for the suppression ratio.

6. On-line application

In the following, the potential of the new sections at ISOLTRAP is illustrated by first on-line data. Fig. 11 (left) shows a time-of-flight spectrum of short-lived $A=80$ zinc and rubidium ions (with half-lives of 540 ms and 33.4 s, respectively).

After the ions had been transferred through the ISOLDE HRS sector-field mass separator and captured at the ISOLTRAP RFQ buncher they performed 400 revolutions in the MR-ToF, corresponding to a flight time of $t \approx 8.6$ ms. Thus, the ion species were separated by $\Delta t_{80\text{Zn},80\text{Rb}} \approx 1.2 \mu\text{s}$ while their bunch width was $\Delta t \approx 70$ ns. This corresponds to a mass-resolving power of over 60,000, fully sufficient to separate both species by multiple signal widths. After the separation the ^{80}Zn ions were selectively transmitted through the BNG to the preparation Penning trap. There, they were cooled for only 15 ms at a high helium buffer-gas pressure, i.e. a second mass-separation step was not necessary, before they were forwarded to the precision Penning trap. Thus, the distribution of isobar separation and ion-sample preparation into two separate steps and on two devices reduced the overall processing time by a factor of ten to < 25 ms. Fig. 11 (right) shows a ToF-ICR spectrum of ^{80}Zn ions including the detailed ToF ion distributions within the individual time-of-flight spectra [47]. Obviously, there is no ^{80}Rb contamination, as these ions would show up at flight times of about $400 \mu\text{s}$ at the center of the ^{80}Zn resonance. This proves that the MR-ToF and BNG system were used effectively to purify the ensemble.

The fast ion separation in the MR-ToF device will allow high-precision mass measurements of nuclides with half-lives as low as a few milliseconds. The feasibility of this approach is demonstrated in Fig. 12 which shows an MR-ToF time-of-flight spectrum of the $A=137$ nuclides barium, praseodymium, neodymium, promethium and samarium for a MR-ToF storage time of ≈ 22.5 ms. Such spectra allow the determination of the mass of one ion species from the known masses of the others, which act as internal mass references. This implies the advantage that they are exposed to the same fluctuations and drifts of the system, which

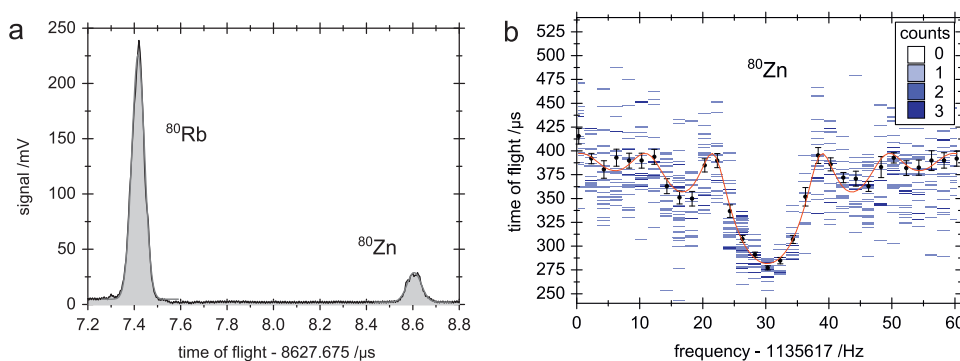


Fig. 11. Left: MR-ToF spectra of ^{80}Rb and ^{80}Zn after 400 revolutions recorded with the MCP2h (see Fig. 3), with Gaussian fit curves. The ISOLDE yield of ^{80}Zn was about 10^3 /s. Right: Time-of-flight matrix of the ^{80}Zn ToF-ICR spectrum showing that the species of interest was cleaned from ^{80}Rb contaminations. The shading represents the number of counts recorded in the respective time-of-flight bin.

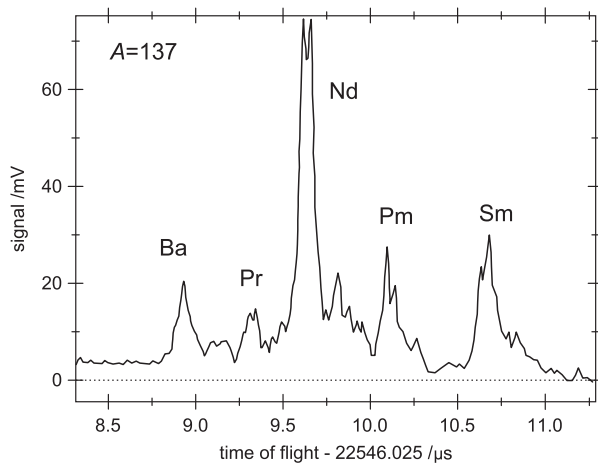


Fig. 12. MR-ToF time-of-flight spectrum of exotic nuclides at mass number $A=137$ after 800 revolutions in the MR-ToF.

reduces the systematic uncertainty significantly. This proof-of-principle data has been taken in a region of the nuclides which has already been explored by Penning-trap measurements [48]. In these previous experiments, the necessary mass resolving power of 50,000 was reached by excitation durations of several 100 ms while the MR-ToF mass separator storage time for comparable mass-resolving power was just ≈ 22.5 ms. The relative deviation to the literature value of e.g. ^{137}Nd , by using ^{137}Ba and ^{137}Sm as calibrants, derived from this data is $\Delta m/m \approx 1 \times 10^{-6}$ which is in good agreement taking the 1-sigma width of the ToF signals ($> 1 \times 10^{-6}$). A more detailed investigation of the uncertainties will be performed in the upcoming measurement periods.

7. Summary and outlook

The implementation of an MR-ToF mass separator in the on-line Penning-trap mass spectrometer ISOLTRAP has been described in detail and its performance has been characterized with respect to systematic measurements during both off-line and on-line experiments. The results confirm that the MR-ToF mass separator is a valuable addition to the ISOLTRAP setup. While it adds further complexity as it is the fourth ion-storage device besides the RFQ buncher and two Penning traps, it has significantly improved the purification speed and facilitates measurements of nuclides with shorter half-lives, of capital importance for advancing nuclear physics. These data are under evaluation and will be published soon.

To evaluate the effect of ion-ion interaction [49–51] on the performance of the MR-ToF device, numerical and experimental investigations are under way.

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6.4 ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer, International Journal of Mass Spectrometry 349-350, 100 years of Mass Spectrometry, 123–133 (2013)



ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer

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Mass of ¹³⁷Eu nuclide

ABSTRACT

The online precision mass spectrometer ISOLTRAP at ISOLDE/CERN was recently upgraded by adding a multi-reflection time-of-flight mass separator/spectrometer (MR-ToF MS) between the linear radio-frequency ion trap and the two Penning traps already in place. As a mass separator, the MR-ToF device has improved significantly ISOLTRAP's capability of purification of contaminated ion beams. In addition, the MR-ToF MS can be operated as a mass spectrometer, either to analyze the ISOLDE ion beam or for precision mass measurements of nuclides that are shorter-lived or that have lower yields than those accessible for Penning-trap mass spectrometry. The MR-ToF MS and corresponding components, its integration into ISOLTRAP, and its various operation modes are reviewed. Furthermore, a precision measurement of the ¹³⁷Eu mass is presented, determined with the help of the MR-ToF device as a mass separator.

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1. Introduction

The investigation of short-lived nuclides continues to contribute significantly to our understanding of nuclear structure, fundamental interactions and astrophysical processes [1–9]. The mass difference between an atom and the sum of its constituents, the total binding energy, reflects the sum of all their interactions. The differences in binding energy within isotopic and isotonic chains reveal the general structural evolution of atomic nuclei. In particular, they point to configurations of enhanced binding at so-called magic numbers. Other filters, like the empirical pairing gap, reveal more subtle changes in nuclear structure such as deformation, collectivity or residual proton-neutron interaction [10–18].

To understand the abundance distribution of the elements in our universe, a robust modeling of astrophysical processes which occur far from any earth laboratory is of major importance [19–21]. The mass, as the most fundamental ground state property, is among the most important input parameters for the reaction paths of nucleosynthesis, e.g. the *r* process [22–25], *rp* process [26–30], *s* process [31,32] and *vp* process [33,34].

A major step for precision mass spectrometry of short-lived nuclides was the introduction of the time-of-flight ion-cyclotron resonance (ToF-ICR) technique [35], pioneered with the ISOLTRAP experiment at ISOLDE–CERN over 20 years ago [36–39]. Here, ions of the radioactive species of interest are stored in a Penning trap to determine their cyclotron frequency $\nu = qB/(2\pi m)$, which is inversely proportional to their mass-over-charge ratio m/q (and proportional to the magnetic field B). To achieve smallest mass uncertainties δm , the measurement has to be perturbation-free, in particular with respect to the Coulomb interaction with ions of other species, i.e. contaminations of the sample. Facilities like ISOLDE [40] produce a large variety of stable and unstable isotopes. These are usually mass-over-charge separated by magnetic separators before being sent to the experiments. The separators' mass resolving power on the order of $R = m/\Delta m = 10^3$ is sufficient to suppress neighboring isotopes, e.g. atomic ions with a different mass number $A = Z + N$, where Z and N are the number of protons and neutrons of the nucleus, respectively. However, usually one of the major challenges for subsequent experiments is the selection of the one ion of interest from the remaining isobaric ensemble, i.e. ions with the same number of nucleons. These contaminations may originate from surface ionization in the hot environment of the target/ion source, like the atomic isobaric species in the case of alkali ions, or molecules formed in the ion source, e.g. oxides, fluorides,

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hydroxides, the so-called molecular sidebands. The required mass resolving power to suppress these isobars can vary from $R \approx 10^3$ up to 10^6 . Furthermore, investigations of isotopes far from stability are hindered by the steep decrease in production yield, alongside with a drop of half-life. This often reduces the abundance ratio between the ions of interest and the contamination ions. A fast and efficient removal of all contaminating species and the transfer of the ions of interest to the actual mass-measurement device is therefore of utmost importance for a successful experiment.

The MR-ToF mass separator/spectrometer (MR-ToF MS) is a recent development in time-of-flight mass analysis. Actually, a predecessor, the “Farvitron” [41], was introduced already in the early 1960s as a small high-vacuum gauge where electrostatic mirrors were used to periodically increase the density of a mass-over-charge specific ion bunch and to detect its mirror charge signal. However, it was not until 30 years later when Wollnik and Przewłoka [42] described electrostatic time-of-flight mass spectrometers with multiple folded trajectories, see [43] for an overview. Due to their extended flight path which was no longer limited by the dimension of the device, long flight times could be achieved, leading to mass resolving powers several times higher than obtained up to then with table-top mass spectrometers. This property, combined with a fast operation cycle, single-shot measurement of several ion species (although of a limited mass range) and single-ion detection made the MR-ToF device appear as a promising mass spectrometer for short-lived species [44–46]. Furthermore, in combination with a fast ion selector, it was suggested to supply highly-resolved mass-over-charge selected bunches for subsequent experiments [47]. In the meantime, several of these systems are in operation or under construction [48–55].

Besides their applications related to mass-spectrometry, similar devices, often referred to as “electrostatic ion beam traps” (EIBT), are used for a wide range of atomic, molecular, and cluster studies [56–61]. In many cases the focus of EIBTs is not on their mass spectrometric aspects. Instead, they provide ion storage with the additional advantage of detection of neutral decay products.

In the following, the ISOLTRAP MR-ToF device will be reviewed. After its first successful on-line operation in 2010, the device has been applied in several different innovative experiments. The techniques and applications are presented, as well as preliminary results from on-line experiments.

2. Experimental setup and operation principle

Precision mass measurements with a sub parts-per-million (ppm) mass uncertainty utilizing a Penning trap demand a pure sample of one or at most a few ions of interest per measurement. The presence of a different species in the measurement trap leads to a perturbation of the individual mass-to-charge specific cyclotron frequencies due to Coulomb interaction, commonly referred to as space charge [62–66]. This is exemplified in Fig. 1 with ToF-ICR spectra of $^{229}\text{Fr}^+$ and $^{229}\text{Ra}^+$. In one case, about 40 ions of both species were simultaneously injected into the ISOLTRAP precision Penning trap with nearly equal abundances. In a second experiment, the $^{229}\text{Ra}^+$ ions were removed by dipolar excitation prior to the ToF-ICR measurement of $^{229}\text{Fr}^+$. In the presence of the $^{229}\text{Ra}^+$ ions, there is a shift in the cyclotron frequency of $^{229}\text{Fr}^+$ of $\Delta\nu \approx 0.5$ Hz to lower values, i.e., higher mass, attributed to the Coulomb interaction between the two ion species. Furthermore, the cyclotron frequency of $^{229}\text{Ra}^+$ is shifted as well by about 0.4 Hz in the same direction. The relative difference of $\Delta\nu/\nu_c = 10^{-6}$ is about two orders of magnitude higher than the typical total uncertainty achieved for measurements in the absence of space-charge effects. This example emphasizes the importance of contamination-free samples in Penning traps for precision measurements in the sub-ppm range.

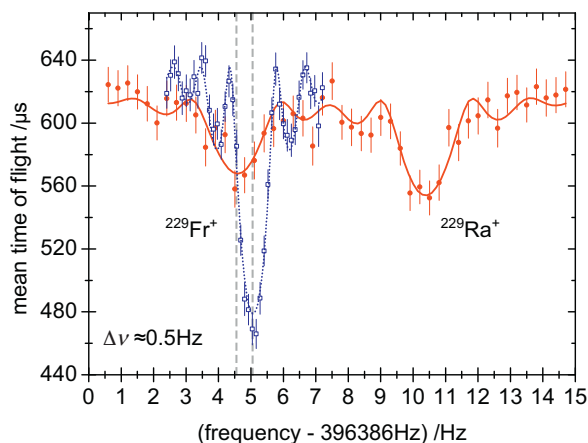


Fig. 1. Simultaneous ToF-ICR resonances, i.e., time of flight of ions ejected from the precision Penning trap as a function of excitation frequency, of $^{229}\text{Fr}^+$ and $^{229}\text{Ra}^+$ (red) and single-species resonance of $^{229}\text{Fr}^+$ (blue). In the first case, the cyclotron frequency of $^{229}\text{Fr}^+$ is shifted by almost $\Delta\nu \approx 0.5$ Hz to lower values, i.e., higher mass, due to Coulomb interaction in the Penning trap. The cyclotron frequency of $^{229}\text{Ra}^+$ is shifted by a comparable value in the same direction. About 40 ions were injected into the trap for each measurement step. In case of the double resonance, the excitation time was $T_{\text{eff}} = 0.6$ s, while for the single resonance, the excitation time was $T_{\text{eff}} = 1.2$ s. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

For on-line experiments, besides the purity of the sample, the number of the ions delivered is an additional limiting factor. Short-lived nuclides with half-lives below a second, the usual measurement time of a Penning trap, are challenging due to the decay losses during the preparation and actual measurement. Therefore, the preparation time should be kept as short as possible. The MR-ToF mass separator combines high-resolution separation with a fast operation. It serves as a universal beam-composition analyzer and mass separator for species with half-lives down to a millisecond. Furthermore, it offers the possibility to use well-known species as simultaneous mass-markers, i.e., reference masses for MR-ToF mass measurements, which would be considered unwanted contaminants in the case of the Penning trap experiments.

2.1. Overview of ISOLTRAP

Before the implementation of the MR-ToF MS and corresponding components, the ISOLTRAP setup already included three ion-trapping devices: a segmented linear Paul trap (radio-frequency quadrupole (RFQ) cooler and buncher) in a horizontal beamline and two vertically installed Penning traps, each with its own superconducting magnet. Behind the RFQ, space was available for the installation of an additional MR-ToF MS device. Thus, this component had to be built to meet the restrictions of the already existing ISOLTRAP setup.

After the implementation of the MR-ToF MS, the experimental setup of ISOLTRAP, as shown in Fig. 2, currently includes four ion traps:

1. A helium buffer-gas filled RFQ cooler and buncher is used to accumulate the ions from ISOLDE and to convert the quasi-continuous beam to a pulsed beam [68]. The typical accumulation durations range from a few microseconds up to several hundreds of milliseconds, depending on the yield and half-life of the species. To thermalize the ions, a cooling period of around 10 ms is sufficient.
2. The MR-ToF MS can be used in combination with a Bradbury-Nielsen gate (BNG) as a fast ion selector [47,69] to separate the ions of interest from contaminations. Alternatively, it can be

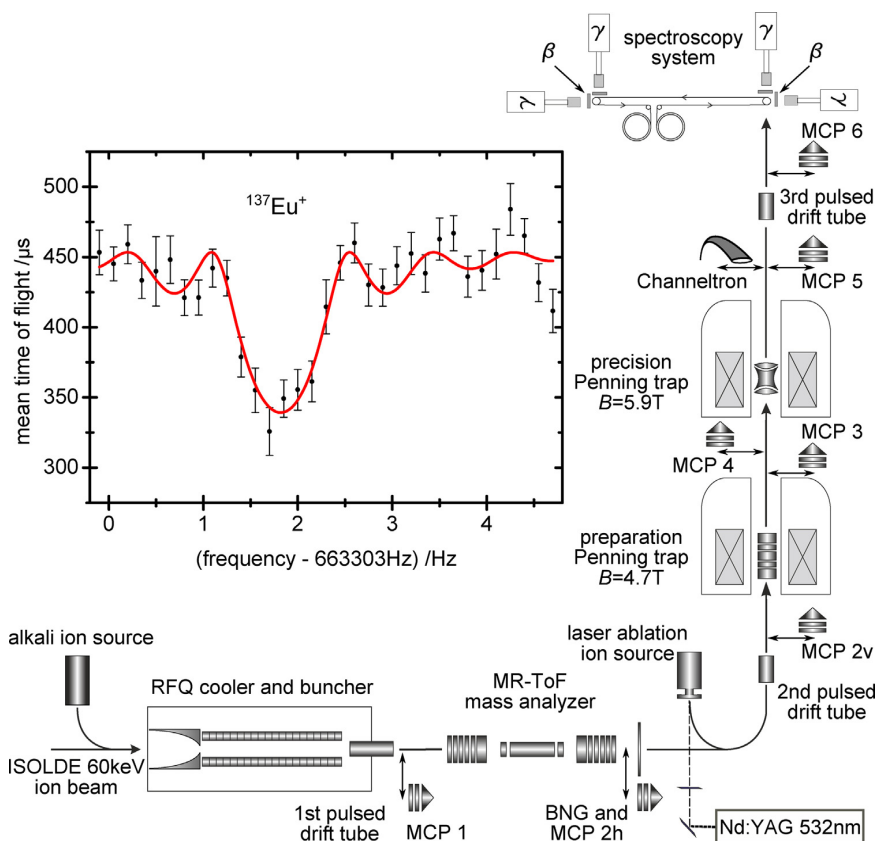


Fig. 2. Schematic overview of the ISOLTRAP setup (adapted from [67]). The inset shows a ToF-ICR spectrum of $^{137}\text{Eu}^+$. See text for further explanation.

utilized in combination with an ion detector to directly observe the composition of the ion ensemble for mass spectrometry and other applications, which will be discussed below. The operation period for maximum mass resolving power of $R_{\text{FWHM}} \approx 200,000$ is about 30 ms.

3. A helium buffer-gas filled preparation Penning trap is used to further cool and mass-select the ions of interest [70] for the subsequent mass measurement in the precision Penning trap. The trap can be operated in cooling and bunching mode, only, at high gas pressures of about 10^{-3} mbar if the ion ensemble is already delivered essentially pure. The required centering can be accomplished in a few tens of milliseconds. For contaminated ensembles, mass-selective resonant buffer-gas centering is applied at typical pressures of about 10^{-4} mbar. The lower pressure is essential to achieve mass resolving powers in the order of $R = 10^5$. However, it also results in cooling periods of several hundreds of milliseconds, affecting the ion throughput and leading to losses in particular for ions with short half-lives.
4. The precision Penning trap, combined with a drift section in a magnetic-field gradient for time-of-flight measurements of the ejected ions, is used to determine their cyclotron frequency and thus their mass. To this end, the ion motion is probed by quadrupolar excitation at different frequencies (one at a time) to find the resonant conversion between magnetron and cyclotron motion. The increase in motional energy at the cyclotron frequency is detected by monitoring the ions' time of flight to a detector outside of the superconducting magnet. Dipolar excitation at the reduced cyclotron frequency can be applied prior to the actual mass measurement to clean the ensemble with mass resolving powers of up to $R = 10^6$.

In addition to the four ion traps, there are several supplementary devices. In particular, two off-line ion sources deliver the reference ions required for the calibration of the magnetic field: An alkali ion source in front of the RFQ produces potassium, rubidium and cesium ions. These ions are available to optimize the whole setup. A laser-ablation ion source located in front of the preparation Penning trap can be used to generate atomic or cluster ions by irradiating various bulk materials with 532-nm laser light [71]. Furthermore, a tape-station setup subsequent to the precision Penning trap has been installed for trap-assisted decay spectroscopy of isobarically and isomerically pure ion ensembles [72].

2.2. Details of the MR-ToF device

The MR-ToF mass analyzer, the BNG, a multichannel-plate (MCP) ion detector and adjacent ion optics were installed at ISOLTRAP in the CERN shutdown period 2009/2010. An ion-optical transfer section of about 1.8 m length in the horizontal ISOLTRAP beam line between the RFQ cooler and buncher and the laser-ablation ion source was used to accommodate the devices. A detailed description of the setup can be found in references [54,73]. It will only be summarized in the following.

The MR-ToF mass analyzer consists of two 160 mm-long electrostatic ion-optical mirrors, each incorporating six electrodes, surrounded by inner and outer shielding electrodes. The mirrors are separated by an 460 mm-long in-trap lift, a drift electrode whose electric potential can be pulsed to facilitate ion injection, ejection and control of the time-of-flight focus plane [74]. Ion species with different masses m_i , ejected from the RFQ at a potential U , gain the kinetic energy $E_{\text{kin}} = z_i e U = m_i v_i^2 / 2$, where e is the elementary

charge and z_i , v_i are their charge state and velocity, respectively. Therefore, their time of flight t_i for the same flight path is mass and charge dependent, $t_i \propto v_i^{-1} \propto \sqrt{m_i/z_i}$. Thus, they separate in time and can be resolved on an ion detector if their difference in time of flight Δt_{ij} is larger than the individual signal widths Δt_{ij} , $\Delta t_{ij} > \Delta t_{ij}$.

High mass resolving power $R = m/\Delta m = t/(2\Delta t)$ can be achieved by either increasing the time of flight or decreasing the signal width at the detector plane. The latter is limited by the turn-around time of the ions in the ion source, i.e., depending on the ion velocity and the extraction field strength, and the Coulomb repulsion. The former can be increased by either reducing the average kinetic energy or extending the flight path. Decreasing the average kinetic energy E_{kin} leads to an increase in the relative kinetic energy spread $\delta_E = \Delta E_{kin}/E_{kin}$, mostly unfavorable for the mass analyzer because the time-of-flight dispersion with respect to energy increases. Extending the flight path considerably is only possible by folding the flight path, i.e. the ions have to travel multiple times through the device. This technique is well known from, e.g. storage rings [5]. However, for high-resolution time-of-flight mass spectrometry, the successful application was shown just a few years ago [44–54]. Here, the individual species i travel with m_i/z_i -specific revolution times $T_i \propto \sqrt{m_i/z_i}$. After n revolutions, the time-of-flight difference of species i and j is grown by $\Delta t_{ij} = n |T_i - T_j|$, i.e. their separation increases linearly with the number of revolutions. By compensating time-of-flight dispersions with respect to the energy, angular and spatial distributions of the ions, the minimum signal width can be adjusted on the detector plane.

To store ions in an MR-ToF device, the potential maximum $U(x)$ inside a mirror has to exceed the ions' energy, i.e., their kinetic energy $E_{kin} < zeU(x)$ in the drift section. Injection and ejection from the MR-ToF device can be achieved by lowering the potential distribution of an ion mirror to a value that ions can pass the mirror without modifying the ions energy. An alternative approach is to use static mirrors and inject and eject the ions with a kinetic energy that exceeds the mirror potential maximum, $E_{kin} > zeU(x)$. The confinement can then be achieved by changing the ions' energy once they are between the mirrors. For this purpose, the in-trap lift has been introduced [74]. Furthermore, electrically switching the sensitive mirror potentials is avoided which reduces the electrical noise and fluctuations on the corresponding electrodes. The incoming ions have a kinetic energy $E_{kin}^{transfer} = zeU_{transfer} > zeU(x)$ that exceeds the maxima of the mirror potentials. Thus, they pass the first mirror and enter the activated in-trap lift electrode on a potential U_{lift} . While the ions travel through the electrode, it is switched to ground. Thus, the ions' energy is reduced to $E_{kin}^{trapping} = ze(U_{transfer} - U_{lift}) < zeU(x)$ and the ions are trapped. For ejection, the in-trap lift electrode is activated again while the ions are inside. Thus, they can pass the exit mirror for further transfer to the downstream experimental parts. The transfer energy in the horizontal beam line of ISOLTRAP is $E_{kin}^{transfer} \approx 3.1$ keV, while a trapping energy of $E_{kin}^{trapping} \approx 2.1$ keV is used for the MR-ToF mass analyzer. Furthermore, the in-trap lift can be used to maximize the mass resolving power. Depending on the ejection settings from the RFQ and the number of revolutions performed in the MR-ToF, the position of the time-focus plane changes. By modifying the kinetic energy of the ions inside the MR-ToF, i.e., changing U_{lift} , the position of the time-focus plane can be readjusted on the ion detector, as demonstrated in Fig. 3. This provides a simple and independent method to tune the mass resolving power, without changing the transfer energy outside the MR-ToF device or switching of the sensitive mirror voltages.

The minimum separation in time needed to apply a selection with the BNG can be derived from its wire spacing [75] and the rise/fall times of the switches used to drive the two sets of wires. Suppression of an ion signal can be achieved if the transversal spread in beam radius (for a BNG in the "closed" state) is increased

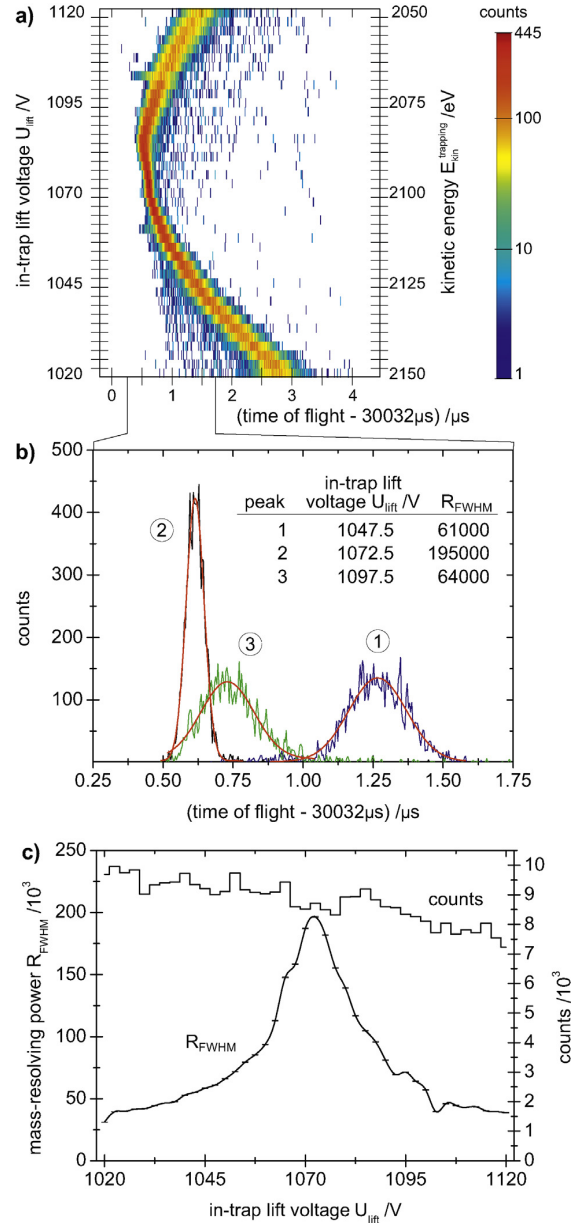


Fig. 3. In-trap lift time-of-flight focusing for $^{39}\text{K}^+$ ions at 2000 revolutions. (a) Color-coded number of counts as a function of the time of flight for different in-trap lift voltages and the corresponding kinetic energies. (b) Corresponding time-of-flight distributions for three in-trap lift voltages: the ToF focal plane is behind (peak 1), precisely at (peak 2) and in front of (peak 3) the detector. (c) Mass resolving power and number of integrated ion counts as a function of the in-trap lift voltage. A maximum mass resolving power of 200,000 is reached for about 1070 V.

sufficiently to not meet the acceptance of a subsequent aperture. This increase is determined by the wire distance, the voltage difference applied to neighboring wires, and the ions' energy. The voltage dependence sets an upper limit on the selection capabilities as shown in Fig. 10 of reference [54] due to the finite transmission even for closed BNG. Presently, the transmission of an unwanted species can be reduced by up to 4 orders of magnitude. Higher voltage differences would presumably increase the suppression factor, but have not been tested yet to avoid damaging the BNG.

2.3. Modifications of the RFQ

The RFQ buncher acts as an ion source for the MR-ToF device and the minimum bunch width provided by this source determines the time necessary to reach the maximum mass resolving power. Thus, the RFQ's trapping and ejection section has been modified to meet the MR-ToF requirements. The bunch width Δt after MR-ToF MS trapping is a quadratic sum of the initial bunch width Δt_0 provided by the RFQ and contributions from the dispersion per turn, ΔT , of the mass analyzer, $\Delta t = \sqrt{\Delta t_0^2 + (n\Delta T)^2}$. The mass resolving power

$$R = \frac{t}{2\Delta t} = \frac{t_0 + nT}{2\sqrt{\Delta t_0^2 + (n\Delta T)^2}} = \frac{t_0/n + T}{2\sqrt{\Delta t_0^2/n^2 + \Delta T^2}} \quad (1)$$

is, therefore, limited by the dispersion of the MR-ToF device, $R = T/2\Delta T$ for $n \rightarrow \infty$. In contrast, the initial bunch width determines the number of revolutions necessary to approach the maximum. It is limited by the turn-around time $\Delta t_0 = 2v_0m/(qE_{ex})$ of the ions moving in the pulsed ion source (the RFQ), i.e. their velocity v_0 and the extraction-field strength E_{ex} applied when the source is emptied [76]. This, in combination with the spatial distribution Δx along the axis, determines the absolute energy spread, $\Delta E = E_{ex}\Delta x$. To decrease the initial time spread it is necessary to apply high extraction-field strengths. However, this increases the energy spread of the bunch. Therefore, to achieve low initial time spreads and low absolute energy distribution as well, an efficient cooling of the ion motion is required. This is accomplished in the RFQ by the application of a helium buffer gas. To decrease the spatial distribution, the RFQ trapping region was shortened. Originally, it had consisted of two segments with a total length of 20 mm (segments 24 and 25, see Fig. 4). This was modified by separating the two segments to form a trapping region of only 10 mm length. A dipolar extraction field is generated by switching the adjacent segments. By increasing the field strength from $E_{ex} \approx 3$ V/mm to $E_{ex} \approx 20$ V/mm, the bunch width on a detector in front of the MR-ToF device (MCP 1 in Fig. 2) was reduced from ≈ 900 ns to ≈ 60 ns for an ion-transfer potential of $E_{kin}^{transfer}/e z_i = 3.17$ kV, see Fig. 4. Thereby, the energy spread increased from about $\Delta E_{kin}^{90\%} \approx 6$ eV to about $\Delta E_{kin}^{90\%} \approx 60$ eV. This results in a relative energy spread of $\delta_E^{trapping} \approx 3\%$ at a trapping potential of $E_{kin}^{trapping}/e z_i = 2.1$ kV.

3. Operation modes and applications

The MR-ToF mass analyzer has become a versatile tool for ISOLTRAP as well as for the ISOLDE facility. Originally intended as an auxiliary device to enhance the mass-purification capabilities for Penning-trap mass spectrometry (Fig. 5 bottom), it has additionally become a mass spectrometer in its own right, see (Fig. 5 top). Its advantage with respect to both mass separation and mass spectrometry lies in the combination of the following characteristics:

- fast measurement cycle, i.e., typically 30 ms, compared to Penning traps and therefore lower half-life limitations,
- high mass resolving power sufficient for most isobars, i.e., in excess of 10^5 ,
- easy set up and fast operation readiness, e.g., less elements of the setup have to be prepared and optimized in preparation of a measurement campaign,
- non-scanning mode (in comparison to ToF-ICR): The whole (narrow-band, e.g. isobar) spectrum is obtained from a few injections. Contaminations are observed and identified simultaneously with the MR-ToF measurement, i.e. in the same mass spectra, and can even be utilized as calibrants,

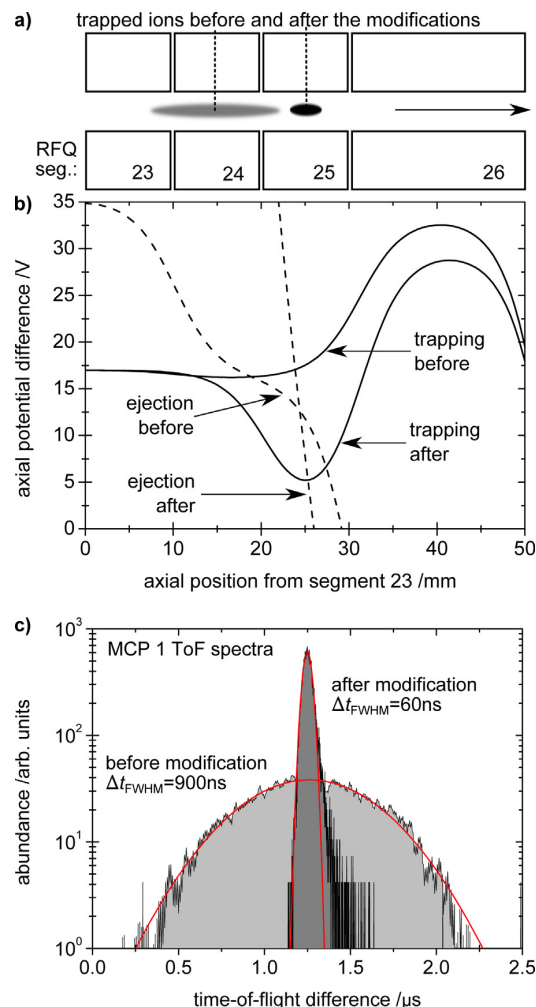


Fig. 4. Modification of the RFQ extraction section, properties before and after the intervention are shown. (a) Schematic illustration of the electrode configurations; (b) axial potential distributions for trapping and ejection; (c) time-of-flight distributions at MCP 1 (see Fig. 2).

- single-ion sensitive detection,
- contaminations do not perturb the measurement, as long as space-charge effects are avoided.

In the following some specific applications of the MR-ToF MS will be discussed in more detail.

3.1. Ion-beam analysis

The development and advancement of ionized rare-isotope beams continues to be a major issue at on-line facilities like ISOLDE, see e.g. [77,78]. A prerequisite is the qualitative and quantitative analysis of the ion-beam composition, influenced by e.g., rare-isotope production cross sections, ionization efficiencies, and time structure, e.g. effusion and diffusion flux, and half-lives of the isotopes.

The time-of-flight spectra obtained with the MR-ToF mass analyzer and an ion detector can be used to analyze the composition of unknown ion beams. For this purpose, the ToF spectrum has to be converted into a mass spectrum. This is achieved by the use of well-known calibrant ions, which connect mass and time of flight

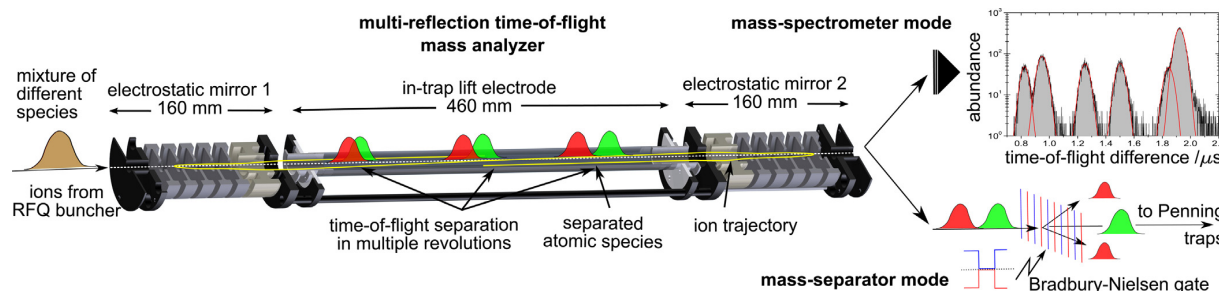


Fig. 5. Sectional view (adapted from [54]) of the MR-ToF device. The mass-separated ions are either detected by a microchannel-plate detector to record a time-of-flight spectrum (right top) or selected by the BNG (right bottom).

with respect to a set of operation parameter of the mass analyzer. In general, for a rough identification of nuclear species, an “external”, i.e. non-simultaneous, calibration with two or more different ion species, e.g., with stable isotopes from the alkali ion source, is sufficient. This allows assigning a mass-over-charge value, and therefore a likely species, to every flight time. As a start, if one species in the spectrum is clearly identified, the relative distances to the neighboring time-of-flight signals can be used to further analyze the ensemble. This procedure is advantageous in case of voltage drifts, e.g., due to a change in the environmental conditions, which might cause time-of-flight deviations. While these may shift the absolute positions of the peaks, the changes in the distances between close-neighboring signals remain negligible.

In cases where ISOLDE’s resonance-ionization laser ion source (RILIS) [79] is employed, the unambiguous identification of one of the unknown ions in the time-of-flight spectrum can easily be accomplished: The RILIS system can be tuned to selectively ionize only the species under investigation. Blocking the laser beam leads to an immediate drop of the signal of the corresponding ion in the time-of-flight spectrum, i.e. a straightforward identification, as shown for $^{185}\text{Au}^+$ in Fig. 6: The surface-ionized isobar $^{185}\text{Tl}^+$ is well separated after a flight time of about 33 ms. By monitoring the count rate the ionization efficiency can be optimized even for very low yields, independent of branching ratio and half-life, as described below. Furthermore, the hyperfine structure of the isotope under investigation can be studied by scanning of the resonances, as show in Fig. 7 as a proof of principle with a relatively large laser line width [80]. A high count-rate range of four orders of magnitude could be recorded background free. A detailed discussion about the application of the MR-ToF MS in the field of in-source laser spectroscopy is out of the scope of the present treatment and will be given elsewhere [81].

The abundance ratios of the different species composing the MR-ToF time-of-flight spectrum are in many cases a direct representation of the ion-beam composition delivered from ISOLDE. Depending on the isotope, corrections may have to be applied with respect to, e.g., the half-lives and charge-exchange rates from collisions with buffer-gas atoms in the RFQ. By varying the delay between the ISOLTRAP RFQ collection-time window and the proton irradiation pulse on the ISOLDE target, the time structure of the release of the nuclides from the target can be sampled. To derive absolute yield numbers, the efficiency of the ISOLTRAP setup up to the MR-ToF MS can be calibrated by the signal intensity of a reference beam.

Currently, the intensity of an ion beam from ISOLDE is monitored with Faraday-cups, which give only an integral number and no information on the composition. In addition, the sensitivity on the order of picoamperes is not sufficient to measure the yield of very exotic species far away from the valley of stability. The beam composition is determined by nuclear decay spectroscopy. This method, however, comes to its limits if the half-life exceeds

a few tens of seconds, if the branching ratio is unfavorable (as, e.g., in Fig. 7, with an α -decay branching fraction of ^{185}Au of only 0.26%), or if the production in the target is low. In contrast, the MR-ToF MS offers an efficient alternative to both Faraday-cup beam-current measurement as well as decay spectroscopy as it has no intrinsic upper half-life limit, provides single-ion counting and can distinguish the species delivered by its ultra-high mass resolving power. Thus, the MR-ToF MS is a universal analytical tool that provides yield information for stable to short-lived species down to the 10-ms half-life range.

The ion-identification capability can be used to optimize the ISOLDE performance for particular experimental requirements, e.g., to find the best parameters to enhance or suppress the yield of specific isotopes. For example, the temperature of an ISOLDE target determines the diffusion time of the ions out of the target. In Fig. 8, the target temperature was varied from about 1740 °C to 1810 °C and the effect on the ion ensemble was observed with the MR-ToF MS: At mass $A=72$ the spectrum shows, amongst others, the short-lived ions $^{72}\text{Ga}^+$ and $^{72}\text{Cu}^+$, with half-lives of 14.10(2) h and 6.63(3) s, respectively [82]. The proposed species are the ones

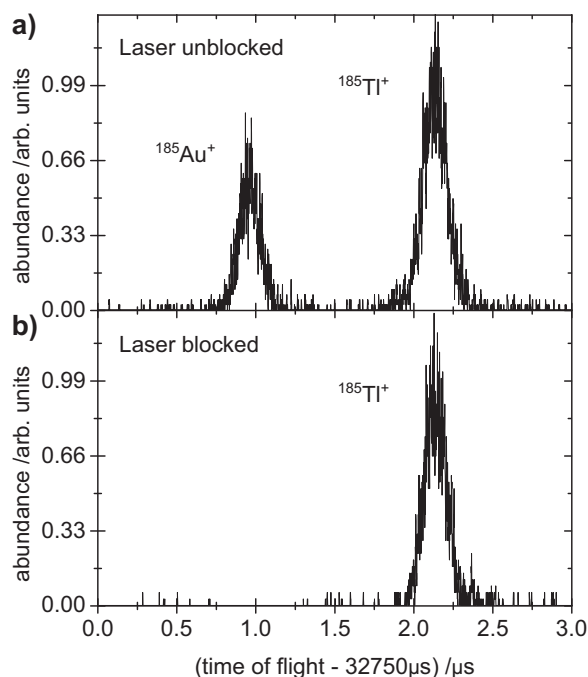


Fig. 6. $A=185$ time-of-flight spectra after 1000 revolutions in the MR-ToF MS. (a) The RILIS lasers were tuned to ionize $^{185}\text{Au}^+$ atoms. The clear separation from $^{185}\text{Tl}^+$ isotopes enables a background-free gold-yield analysis. (b) Furthermore, blocking the laser beam unambiguously identifies the gold isotope.

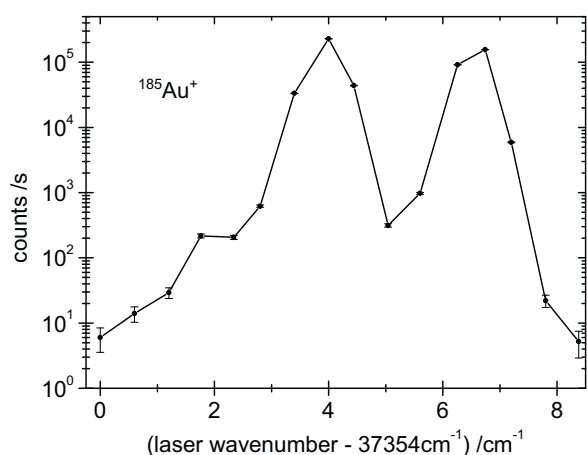


Fig. 7. Signal intensity of $^{185}\text{Au}^+$ (scaled to RFQ accumulation time of 1 s), representing the ionization efficiency, as a function of the wavenumber of the RILIS first excitation step ($6^2S_{1/2} \rightarrow 6^2P_{1/2}$; the two resolved resonances of the hyperfine-structure correspond to the transitions $F=3 \rightarrow 2,3$ (left peak) and $F=2 \rightarrow 2,3$ (right peak)). The laser line width was 0.33 cm^{-1} .

with the best-matching flight times. In the case of close matches, additional arguments are taken into account, such as target material, production mechanism and associated cross sections. One can clearly see in Fig. 8 that the copper yield increases by a factor of two due to faster diffusion whereas the number of counts of the contaminating isotopes stays the same. It is noteworthy, that even the $^{40}\text{Ca}^{32}\text{S}^+$ signal can be evaluated although it is only a shoulder of the dominant $^{72}\text{Cu}^+$ peak.

3.2. Ion-beam purification

The first successful on-line application of ISOLTRAP's MR-ToF mass separator was during mass measurements of neutron-rich argon isotopes in July 2010. The MR-ToF MS was used in addition to the preparation Penning trap to purify $^{45}\text{Ar}^+$ ions from contaminations, mainly consisting of molecules delivered from ISOLDE. An on-line operation resulting in a new mass value was just two months later: In the rare-earth region at $A=137$, the mass of ^{137}Eu could be measured for the first time. Here, the MR-ToF was used again as an auxiliary purifier in combination with the preparation Penning trap. Contaminations of isobaric barium, praseodymium, neodymium, promethium and samarium were present in the ISOLDE beam (see Fig. 12 in [54]). The MR-ToF MS resolved these species with a flight time of about 22 ms. The signal of $^{137}\text{Eu}^+$ was not visible in the spectrum due to the low dynamic range of the transient recorder used for data acquisition during the first MR-ToF MS experiments. At this early time, the BNG could suppress the contaminants by only a factor of 10, since only small deflection voltages were applicable. However, the suppression was still essential for the success of this measurement. Three resonances could be recorded, one with $T_{\text{rf}} = 0.1\text{ s}$ and two with $T_{\text{rf}} = 1.2\text{ s}$ excitation time. In total 1678 ion events were recorded. $^{133}\text{Cs}^+$ from the off-line alkali ion source served as a reference. From the mean frequency ratio, derived from a standard evaluation procedure [83], $r = \nu_{133\text{Cs}}^{\text{ref}}/\nu_{137\text{Eu}} = 0.970570108(35)$, the mass excess is determined to be $\text{ME}(^{137}\text{Eu})_{\text{ISOLTRAP}} = m(^{137}\text{Eu}) - A u = -60145.87(4.28)\text{ keV}/c^2$, where A is the mass number and u the unified atomic mass unit. The relative mass uncertainty of $\delta m/m = 3.5 \times 10^{-8}$ is dominated by the statistical uncertainty of the cyclotron frequencies. With a difference of $\Delta\text{ME}(\text{AME2012} - \text{ISOLTRAP}) = 25.87\text{ keV}/c^2$ this agrees well with the extrapolated AME2012 [84] value of $\text{ME}(^{137}\text{Eu})_{\text{AME2012}} = -60120\text{ (200\#) keV}/c^2$.

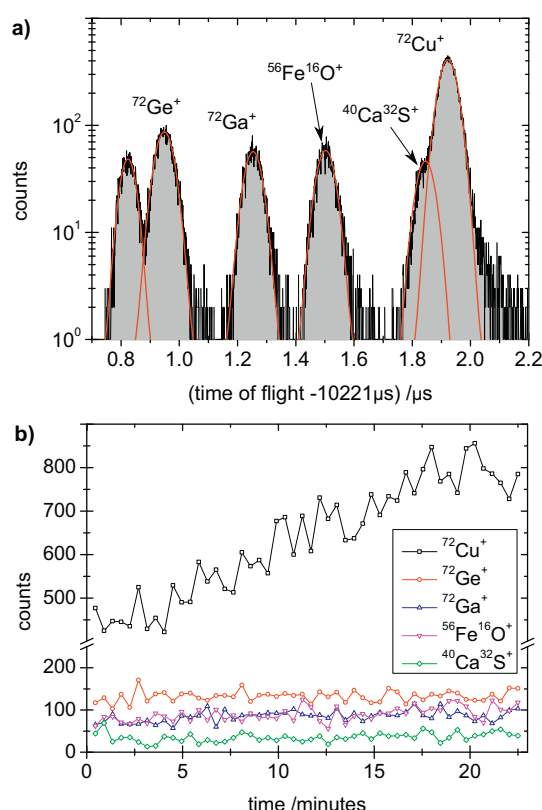


Fig. 8. (a) Integral time-of-flight spectrum for $A=72$ as delivered from ISOLDE during heating of the target material; (b) number of ion counts of the isotopes from the spectrum above as a function of the time during increase of the target heating by approximately 70°C .

As described above, the MR-ToF mass separator and the BNG can be used as an auxiliary mass purifier in combination with the preparation Penning trap. Furthermore, the system has been used as the only mass separator while the preparation Penning trap served as an ion accumulator and cooler. The conditions favoring one of these two operation modes depend on the half-lives of the species to be investigated, yields of the contamination ions, suppression ratios of the contamination, and mass resolving power necessary.

If the half-life of the ion of interest is comparable to, or less than, the usual separation period of the preparation Penning trap, i.e. 300 ms, ion losses due to decays reduce the efficiency of ISOLTRAP as a whole. In this case, using the MR-ToF mass separator as the only purifier is favorable because of its fast separation cycle, in the range of a few 10 ms. The preparation Penning trap is then used just for re-bunching of the selected species in a high buffer-gas environment, which is typically achieved in a few 10 ms as well. This scheme has been applied, e.g., for the mass measurement of the short-lived isotope ^{82}Zn [85] with a half-life of 228(10) ms [86], that could be separated from the contaminating ^{82}Rb and prepared for the mass measurement in less than 25 ms.

The suppression achievable by the MR-ToF mass separator and the BNG is limited by the distributions of the time-of-flight signals and the deflection angle of the BNG. The time-of-flight distribution of different species, i.e. their separation in time at a given height of the signal, is limited by the maximum mass resolving power. Furthermore, asymmetric peak shapes, large tails, and unfavorable abundance ratios between the species will limit the contamination suppression. For Gaussian-shaped time-of-flight

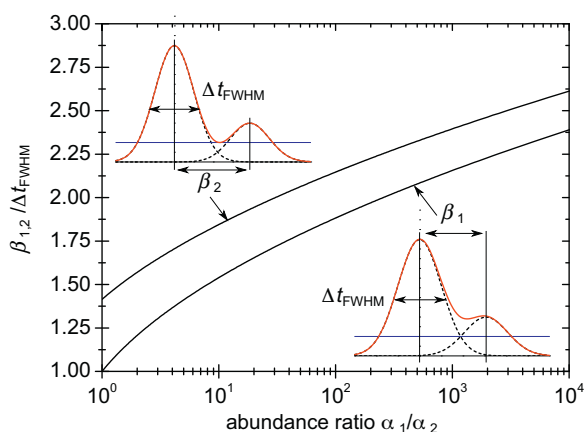


Fig. 9. Necessary increase β in peak separation, i.e. mass resolving power, as a function of the abundance ratio α_1/α_2 of two species. The curves are given exemplarily for two cases where the individual signals are separated to a distance $\beta_{1,2}$ as shown in the insets. In the case of β_1 , both species are separated to a distance where the more abundant signal has decreased to the half-height of the less abundant signal (blue line). In the case of β_2 , the summed signal (red curve) has decreased to the half-height of the less abundant signal. Both signals are assumed to be Gaussian shaped, with the same Δt_{FWHM} . (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

signals, the required increase $\beta(\alpha_1, \alpha_2)$ in mass resolving power, i.e., $R_\beta = \beta \cdot R_{FWHM}$, as a function of abundance ratio α_1/α_2 is shown in Fig. 9.

For asymmetric ToF distributions, the suppression depends also on whether the ion of interest is on the high-mass side or on the low-mass side of the contaminant, see for example Fig. 10. Here, the contaminant $^{50}\text{Cr}^+$ has a tailing edge below the 5%-intensity level of the high-mass side. A clean mass selection of an ion of interest on this side would not be possible. Therefore, in practice, the separation of multiple species and consequently their purification depends strongly on the peak shapes, their abundance ratio and the difference in mass.

For ions of interest with half-lives reasonably longer than the usual separation period of the preparation Penning trap, both devices can be used in combination to increase the accessible contamination to ion-of-interest ratio. This is preferable if the separation by the MR-ToF/BNG combination is hampered by large abundance differences.

Another operation mode in the case of heavily contaminated ion beams is the repeated MR-ToF MS application and accumulation of the ion bunches delivered from the MR-ToF separator in the preparation Penning trap. Due to the limited measurement time available in an on-line experiment, it is desirable to have on average at least one ion of interest processed by the measurement trap per cycle. If the ratio between contaminants and ions of interest is too high, the total number of simultaneously mass-separated ions required to provide at least one purified ion of interest can be so large that space-charge effects due to Coulomb interactions inhibit the MR-ToF MS from separating the different species [87,88]. This effect is known as peak coalescence, but a detailed evaluation with respect to the corresponding conditions is not yet available. Therefore, in practice the number of ions in the MR-ToF has to be increased with care. The maximum number of ions that can be handled by the MR-ToF MS depends on the mass difference and abundance ratio of the species of interest and the contaminations. Typically, it is in the order of only some hundreds up to several thousands. However, when the half-life of the ion of interest is sufficiently long (about a second or more), the number of available ions per mass-measurement cycle in the precision trap can

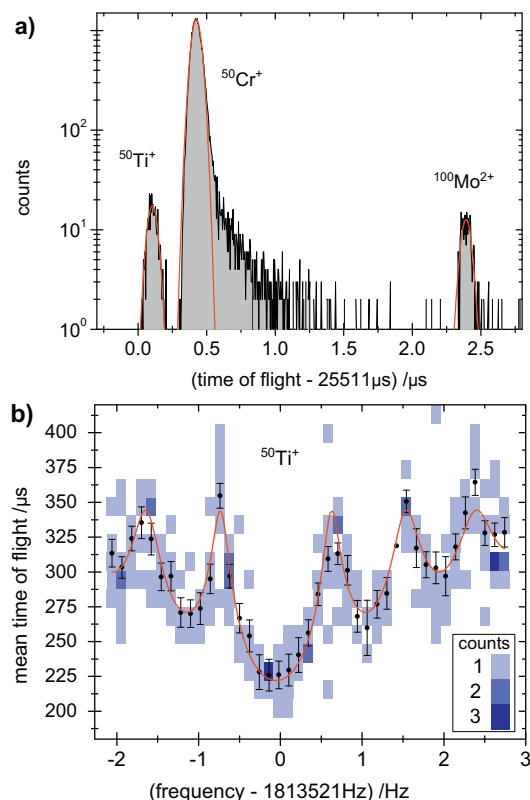


Fig. 10. (a) MR-ToF mass spectrum of $A=50$ isobars at 1500 revolutions, showing $^{50}\text{Ti}^+$, $^{50}\text{Cr}^+$ and $^{100}\text{Mo}^{2+}$. (b) ToF-ICR measurement of $^{50}\text{Ti}^+$ with $T_{\text{eff}} = 1.2$ s excitation time, only 300 ions recorded. The much more abundant $^{50}\text{Cr}^+$ contamination has been removed and simultaneously the number of detected $^{50}\text{Ti}^+$ per second has been increased by a factor of 10, see text for explanation.

be increased significantly by repetitive injections of mass-selected bunches from MR-ToF MS into the preparation Penning trap. In this case, the preparation Penning trap acts as an accumulator to capture the mass-selected species of interest, ejected from the MR-ToF MS in intervals of some ten milliseconds. Therefore, the available number of ions for a subsequent mass measurement increases in principle linearly with the number of injections, while the overall increase in cycle time is comparably low. If the contamination is too strong and exceeds the suppression limits of the MR-ToF MS, or the accumulated ions get significantly contaminated by their own decay products, a final cleaning process in the preparation Penning trap can be applied before the transfer to the precision Penning trap is performed. With a contamination suppression of 10^4 by the BNG (see above) and an additional cleaning process in the preparation Penning trap removing another two orders of magnitude of contamination, ratios of up to 10^6 contaminating ions to ions of interest can be handled.

Alternatively, if the purification by the MR-ToF MS is sufficient and the decay of accumulated ions is not critical (decay products not trapped or sufficiently long half-life of ion-of-interest), additional cleaning in the preparation Penning trap is not required. To save measurement time, only cooling and bunching with a high buffer-gas pressure and a permanent quadrupolar excitation is applied. This accumulation scheme has been used with up to 50 injections, where the resulting number of separated ions for the mass measurement scaled with the number of injections.

Fig. 10 shows a ToF-ICR test measurement of stable $^{50}\text{Ti}^+$ ions, which were contaminated with a 74 times higher yield of $^{50}\text{Cr}^+$

ions. A separation time of about 25 ms has been used to achieve a resolving power of $R_{\text{FWHM}} \approx 150000$, whereas the time-of-flight difference is 278 ns since the mass difference is $1.17 \text{ MeV}/c^2$. The ISOLDE beam was accumulated for 80 ms in the RFQ, which resulted in an average count rate of about $1.6 \text{ }^{50}\text{Cr}^+$ per mass-measurement step in the precision Penning trap. The count rate for $^{50}\text{Ti}^+$ was too low to perform a measurement within an adequate time. Thus, the number of accumulations in the preparation Penning trap was increased from 1 to 30, the average count rate of $^{50}\text{Ti}^+$ in the precision Penning trap could be increased to 0.65 per measurement step. The measurement period for a $T_{\text{rf}} = 1.2 \text{ s}$ excitation time ToF-ICR increased by a factor of 3 from 1.7 s (including all other experimental steps) to about 5 s (this includes additional 29 accumulation steps consisting of 80 ms RFQ accumulation, 10 ms RFQ cooling and 25 ms MR-ToF mass separation, thus 3.335 s). In summary, the increase in count rate was a factor of 30, while the measurement time increased by just a factor of 3, which amounts to a 10-fold reduction of on-line time to obtain a ToF-ICR measurement of $^{50}\text{Ti}^+$. This has been achieved without increasing the space charge in the MR-ToF MS and is therefore a suitable method to tackle high contamination-to-ion-of-interest ratios. As can be seen in Fig. 10, the individual time-of-flight distribution of the ions for each frequency step of the radio-frequency excitation scan show that there are no contaminants “leaking” into the resonance (these would assemble at flight times around $350 \mu\text{s}$, independent of the frequency).

The total on-line measurement time (typically limited to few hours per nuclide) plays an important role for experiments with rare ion beams with only a few ions of interest per second. A more detailed description of this important method is out of the scope of the present article. It will be presented in a forthcoming publication [89].

3.3. MR-ToF precision mass spectrometry

The possibilities for precision mass spectrometry of exotic isotopes are limited with respect to half-life and yield of the ion of interest. As a consequence, this limits observation time t and the number of ions N detectable within an on-line experiment. For conventional Penning-trap mass spectrometry, the mass resolving power [90] $R = \frac{m}{\Delta m} = \frac{v_c}{\Delta v_c} \approx 1.25 v_c t$ increases linearly with the observation time and can reach millions. For MR-ToF devices, the mass resolving power given in Eq. (1) reaches a maximum value when the growth in time-of-flight dispersion per revolution is nearly linear. Fig. 11 compares the conventional Penning-trap mass resolving power of ISOLTRAP ($B = 5.9 \text{ T}$) with that of its MR-ToF MS

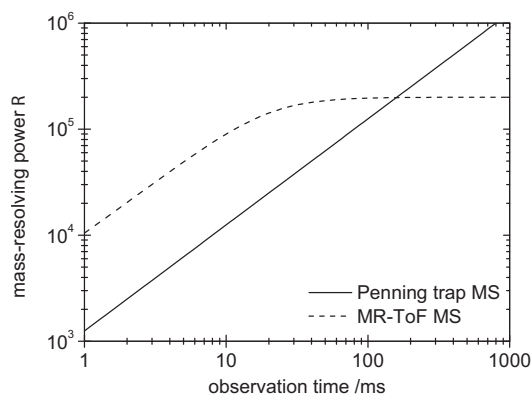


Fig. 11. Mass resolving power as a function of the observation time for ISOLTRAP's precision Penning trap ($B = 5.9 \text{ T}$) and MR-ToF MS for a typical ion of $A/z \approx 90$.

for a typical ion ($v_c = 1 \text{ MHz}$, e.g. $A/z \approx 90$). For isotopes with very short half-lives, i.e., below 100 ms, the mass resolving power of the MR-ToF MS is up to an order of magnitude higher than for the Penning trap. This favors the MR-ToF device as a mass separator and, regarding the statistical mass uncertainty, $\delta m/m \propto (R\sqrt{N})^{-1}$, also as a mass spectrometer, over the Penning trap [53]. However, recent developments in Penning-trap mass spectrometry have potentially extended the limits of the latter [91,92,93].

As already described in Section 3.1, unknown isotopes and molecules can be identified by performing a time-of-flight calibration of the instrument. The ion's time of flight is connected to its mass-over-charge ratio by the general relation

$$t = a \sqrt{\frac{m}{ze}} + b, \quad (2)$$

where a and b are device-specific parameters. The parameter a increases with the trapping time in the MR-ToF MS (number of revolutions the ions undergo in the device), while the parameter b is independent of it. To determine the two parameters a and b , at least two independent reference measurements have to be performed. This can involve either two calibrant ions with known masses or – if only one such reference ion is available – by performing measurements with at least two different numbers of turns in the MR-ToF device.

In addition, two types of calibrant ions can be distinguished, “internal” and “external”. Internal calibrants are the ones, which are recorded, in the same time-of-flight spectrum as the ion of interest. They are typically isobaric species delivered by the same ISOLDE ion beam. External calibrants are the ones which belong to different time-of-flight spectra than that of the ion of interest. They are typically ions from the off-line ion source. The two methods as well as a combination of internal and external calibrants are described briefly in the following.

The ideal situation for an MR-ToF mass measurement, from the point of view of the systematic errors, is the availability of a fully internal calibration with two or more simultaneously detected species with well-known masses. The advantage is that all species encounter the same fluctuations in time of flight, which may come from changes in the mirror potentials or other drifts in the setup. The fluctuations can be monitored through recording time-of-flight spectra in short measurement intervals, as shown in Fig. 12. Data

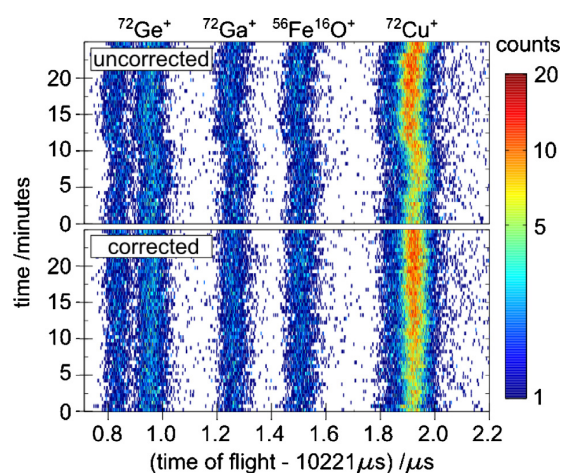


Fig. 12. 2D color-coded intensity plot of time-of-flight spectra of $A = 72$ isobars, i.e. the number of ion counts as a function of time of flight on abscissa and as a function of measurement time on the ordinate. The uncorrected time-of-flight data (top) can be corrected (bottom), in this particular case with respect to the $^{72}\text{Ge}^+$ signal, in order to sum up all events in one spectrum, as shown in Fig. 8 (see text for further explanations).

that was collected in this way can now be corrected for systematic fluctuations. The reference ions can act in each individual spectrum as anchor points from which the time-of-flight differences to the ion of interest can be evaluated. Therefore, the calibration is practically free of systematic errors originating from time-varying fluctuations and drifts of the spectrometer parameters. The data from Fig. 12 was used to create Fig. 8. Again, the increasing copper yield can be seen.

In the case that only one reference species accompanies the ion of interest, two methods can be applied to obtain the calibration and therefore the mass. Either the spectrum is measured at two different flight times, i.e. two different numbers of revolutions in the MR-ToF MS, or at least one additional ion species is employed as an external calibrant. The former method uses the fact that the parameters a and b can be eliminated from Eq. (2) by subtracting the time of flight times of two different revolution numbers. Thus, the mass of the ion of interest can be determined by only the time differences and the mass of the one internal reference ion. The alternative method uses an external calibrant, which has to be provided from, e.g., an off-line ion source. In all other respects, like the preparation in the RFQ, it is treated in the same way as the ion of interest. In particular, the external time-of-flight reference is measured at the same number of revolutions. This measurement is performed both immediately before and after the data of the ion of interest are taken.

The least convenient situation is the absence of internal calibrants. In this case, these have to be provided externally. As in all cases were a second time-of-flight spectrum is needed to complete a mass measurement, the spectra have to be measured in short alternating periods to reduce systematic drifts as much as possible.

Tests of these mass-measurement methods over a wide mass range have shown that – for the cases were at least one internal reference is available – the achievable relative uncertainty is in the ppm to sub-ppm region. First MR-ToF precision mass measurements of short-lived nuclides and a detailed discussion of the statistical and systematic uncertainties of the respective methods are beyond the scope of the present discussion and will be provided in future publications (ref. [94] and [95] respectively).

4. Conclusion and outlook

Three years after its implementation, the MR-ToF MS is a well-established enhancement of the ISOLTRAP mass spectrometer. Its performance as a mass purifier has increased the range of rare isotopes that is accessible for precision mass spectrometry. Due to the reduced time required to reach high mass resolving powers, the MR-ToF MS became the method of choice for the purification of short-lived isotopes, while the preparation Penning trap still serves as a specialized device for accumulation and cooling.

The MR-ToF MS application as a precision mass spectrometer of its own right for very short-lived species was recently tested successfully. It opens new regions of the chart of nuclides for future measurement campaigns as the experimental cycles are shorter and the required ion yields are smaller than that required for Penning-trap mass measurement. In addition, what used to be unwanted contaminants producing space-charge shifts in the case of ICR measurements are now exploited as mass markers, i.e. provide reference mass values. By monitoring the corresponding reference-ion signals a simultaneous calibration is performed which significantly reduces the uncertainty due to temporal fluctuations.

In addition to its use for beam purification as a preparatory step before the Penning-trap mass measurement and its application as a precision mass spectrometer itself, the MR-ToF MS can supply important information for the target and ion-source developments

at ISOLDE. Unambiguous identification of unknown species, e.g. molecular ions as isobaric contaminants of the ions of interest, and background-free yield analysis independent of decay spectroscopy, e.g. also suitable for long-lived species, will simplify and enhance the optimization of the on-line isotope-separator facility at CERN and the characterization of new developments.

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6.5 Plumbing Neutron Stars to New Depths with the Binding Energy of the Exotic Nuclide ^{82}Zn , Physical Review Letters 110, 041101 (2013)

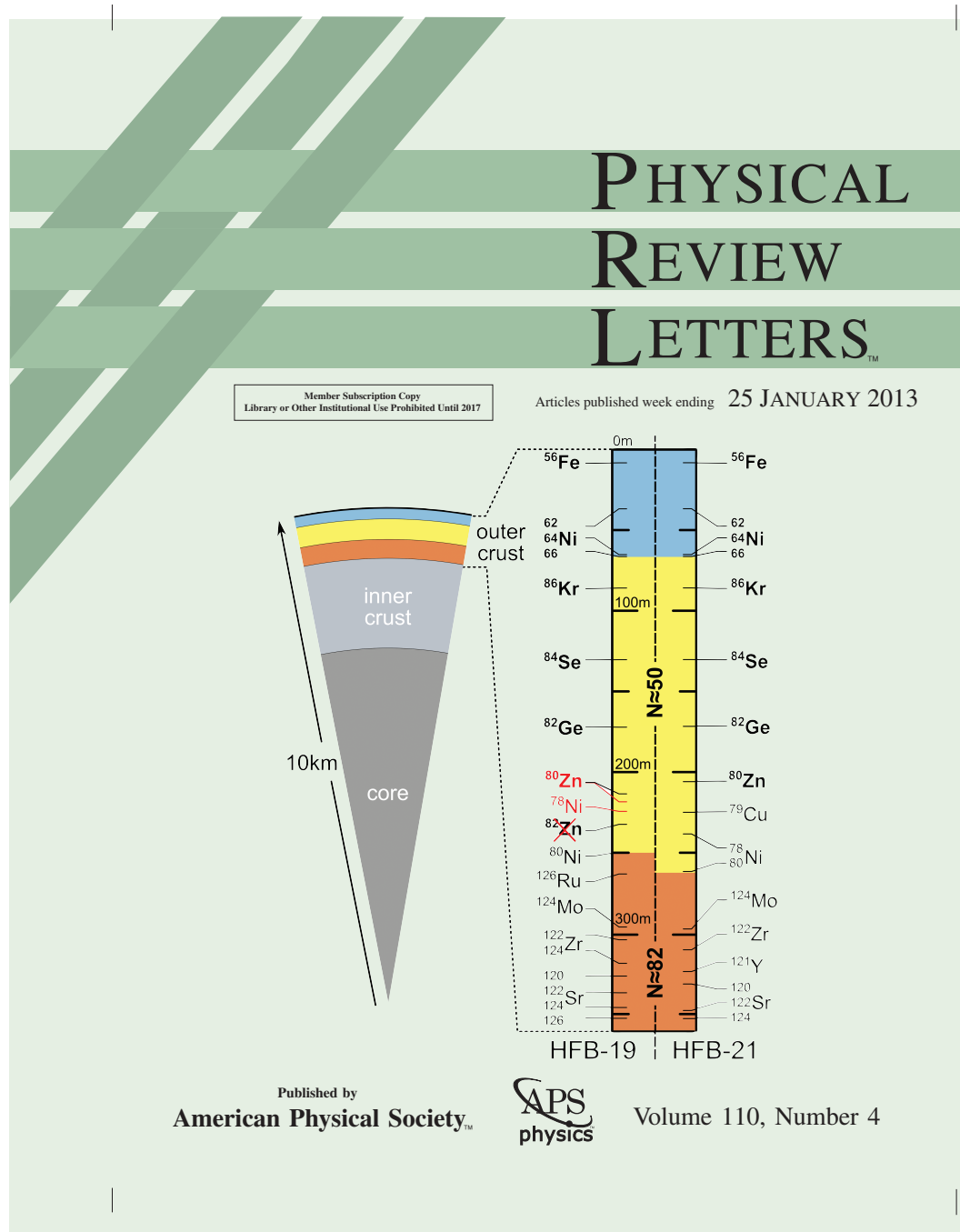


Figure 6.1: Figure 1 of the publication was chosen for the coverpicture of Physical Review Letters 110, Issue 4, 25 January 2013.

Plumbing Neutron Stars to New Depths with the Binding Energy of the Exotic Nuclide ^{82}Zn

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Modeling the composition of neutron-star crusts depends strongly on binding energies of neutron-rich nuclides near the $N = 50$ and $N = 82$ shell closures. Using a recent development of time-of-flight mass spectrometry for on-line purification of radioactive ion beams to access more exotic species, we have determined for the first time the mass of ^{82}Zn with the ISOLTRAP setup at the ISOLDE-CERN facility. With a robust neutron-star model based on nuclear energy-density-functional theory, we solve the general relativistic Tolman-Oppenheimer-Volkoff equations and calculate the neutron-star crust composition based on the new experimental mass. The composition profile is not only altered but now constrained by experimental data deeper into the crust than before.

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With the mass of the Sun compressed to the size of an average terrestrial city, neutron stars are among the densest objects known in the cosmos. While there are still many uncertainties, the details of the neutron-star crust composition are of particular interest in view of the possibility that neutron stars contribute to the abundance of lighter elements close to stability and heavier, neutron-rich nuclides that accumulate near shell closures as observed in our Solar System and Galaxy. The mechanism, rapid neutron capture (the r process), has been the subject of intense activity since its first suggestion, but the astrophysical site where it takes place remains a mystery (see review by Arnould *et al.* [1]). While (type-II) supernovae have been favored for a long time, many problems continue to thwart the correct modeling of an r process. Neutron stars offer a tantalizing alternative r -process site because of their large neutron content, a critical ingredient lacking in supernovae models [2,3]. As discussed recently [4], the decompression of neutron-star matter brought by tidal effects from a merger with a black hole or another neutron star allows an r process to occur as the ejected clump vaporizes into the interstellar medium. While the ejected mass per event is relatively low, it can still explain the total enrichment of r nuclei in the Galaxy. However, the astrophysical plausibility of this scenario requires a proper understanding of neutron stars, most importantly of the composition of their outer crusts.

As their name implies, these residues of core-collapse (type-II) supernova explosions are essentially composed of

neutrons. Three regions can be distinguished (Fig. 1): a locally homogeneous core and two concentric shells, characterized by different inhomogeneous phases [5]. The outermost shell, the so-called “outer crust,” consists of a crystal of ionized atoms coexisting with a quantum gas of electrons. While nuclei that are stable under terrestrial conditions are found at the star’s surface, those deeper are increasingly neutron-rich. The point where neutrons start to drip out marks the transition to the “inner crust,” an assembly of neutron-proton clusters immersed in a sea of unbound neutrons and electrons. Even deeper into the star, the crust dissolves into a uniform liquid of nucleons and leptons until the core is reached.

The conditions prevailing in the deep interior of a neutron star are so extreme that they cannot be reproduced in the laboratory. However, knowledge of specific nuclear binding energies for which high-precision mass measurements are indispensable, combined with neutron-star models, can place the composition of the outer crust on firm ground. Indeed, in analogy with ice cores, it is possible to “drill” into the neutron star and to determine the most abundant species in each layer. In their landmark paper, Baym *et al.* [6] showed that the only property of relevance (aside from the well-known electron and lattice energy) is the nuclear binding energy. Using known masses, the composition of the outer crust had already been robustly determined to a depth of about 212 m for a canonical neutron star of 1.4 solar mass and 10 km radius [7,8].

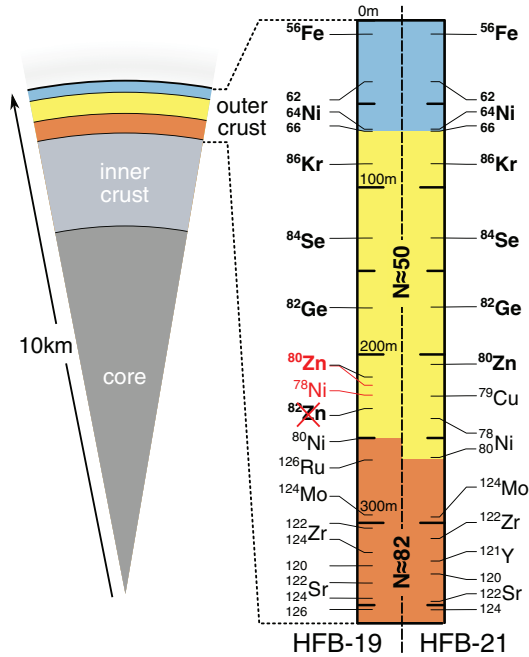


FIG. 1 (color online). The depth profile of a neutron star of 1.4 solar mass and 10 km radius. The scale on the right indicates the nuclide composition in the outer crust as predicted by the HFB-19 and HFB-21 mass models. Experimentally known nuclides are printed in bold. Including the new mass value for ^{82}Zn , the position of ^{80}Zn has changed and ^{78}Ni replaces ^{82}Zn (changes marked red).

In this Letter we report the first measurement of the mass of ^{82}Zn , which represents the frontier of knowledge for the $N = 50$ shell and as such the limit of knowledge for fathoming the neutron-star crust composition.

To plumb a neutron star, Einstein's equations of general relativity, which govern hydrostatic equilibrium, are solved as described by Tolman [9] as well as Oppenheimer and Volkoff [10]. The so-called TOV equations relate pressure and mass-energy density with neutron-star mass and radius. Stable and radioactive-beam facilities have already provided substantial information about a plethora of finite nuclei for the required equation of state (EOS) but even the most neutron-rich of these nuclei still have proton fractions of about 40%, i.e., far larger than the few percent in neutron-star cores or about 30% at the bottom of the outer crust. To make any statement about the composition of the deeper levels of the outer crust, one has to resort to theoretical mass models and extrapolate from the known masses to nuclei closer to the neutron drip line. However, different state-of-the-art microscopic mass models predict different compositions. They can only be tested by high-precision mass measurements on even more exotic species. The Brussels-Montreal microscopic atomic mass models based on the Hartree-Fock-Bogoliubov (HFB) method have been developed to simultaneously describe the binding energy of exotic nuclides, as well as the EOS of neutron matter resulting from many-body calculations with realistic two- and three-nucleon forces [11]. They can be applied to predict the properties of the inner crust and even of the liquid core, thus providing a consistent and unified treatment of all regions of a neutron star [12].

Mass measurements of exotic nuclides are a topic of intense research pursued at many laboratories worldwide [13–17] as they provide critical information about shell stability and other nuclear-structure effects. Because of nuclear shell effects, the exotic nuclides residing in neutron-star crusts accumulate around the magic neutron numbers $N = 50$ and $N = 82$, for the latter even at the vicinity of the neutron drip line (see Fig. 2). Binding

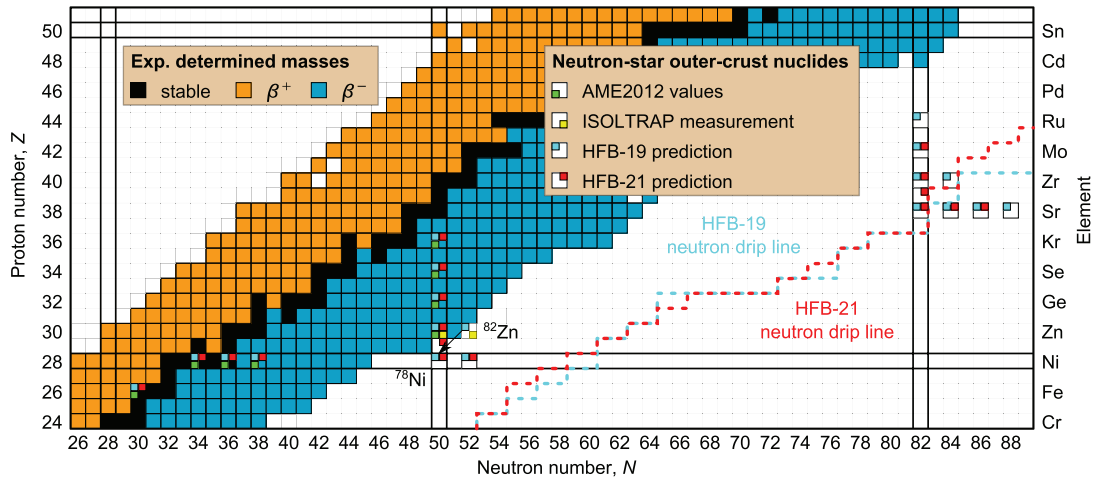


FIG. 2 (color online). The nuclear chart based on AME2012 [38], highlighting the present ISOLTRAP results and the nuclides that compose neutron-star outer crusts according to experiment and the HFB-19 and HFB-21 models.

energies for the $N = 50$ isotones were previously measured down to ^{81}Ga ($Z = 31$) [18], but this nuclide is not predicted to be present in the neutron-star crust (see Fig. 1). In a dedicated experiment that exploited the most advanced techniques for the production and handling of exotic radio-nuclides, we have measured the ($Z = 30$) ^{82}Zn mass. Having the most extreme neutron excess among the known $N = 52$ isotones, ^{82}Zn is of particular relevance for the persistence of the strength of the $N = 50$ shell—which has been questioned [13]—and thus of critical importance for the prediction of its presence in the neutron-star crust.

The ISOLTRAP Penning-trap mass spectrometer [19] (Fig. 3) at the radioactive-beam facility ISOLDE-CERN [20] has pioneered the art of on-line precision mass measurements. It uses electromagnetic fields to confine ions in an unperturbed environment and to measure their cyclotron frequency. The ISOLDE facility produces zinc isotopes by neutron-induced fission of uranium nuclei. To this end, instead of the uranium-carbide target itself, a tungsten converter [21] was bombarded by a 1.4-GeV proton beam. This technique reduced isobaric contamination, which would result from direct spallation reactions. In addition, a highly selective three-step laser excitation [22] to ionize zinc isotopes was applied as well as a temperature-controlled quartz transfer line [23] that prevented the copious surface-ionized rubidium isotopes from drowning the zinc ion beam. Despite these state-of-the-art precautions, over 6000 ions per second of ^{82}Rb were still present in the ^{82}Zn beam delivered to ISOLTRAP—to be compared to just a few ions of interest. Thus, the previous mass-measurement attempts only reached ^{81}Zn [24] but fell short of ^{82}Zn several times at different facilities, making it one of the most challenging nuclides for mass studies to date.

To succeed in measuring the ^{82}Zn mass, yet another type of ion trap was integrated into the ISOLTRAP

mass-spectrometer setup, previously consisting of a linear Paul trap (radio-frequency quadrupole [RFQ] cooler and buncher [25]) and two Penning traps: A multireflection time-of-flight mass separator (MR-ToF MS) allowed a separation of the residual $^{82}\text{Rb}^+$ contaminants from $^{82}\text{Zn}^+$ by repeated oscillations between electrostatic ion mirrors [26,27]. The decisive advantage compared to purification in Penning traps is a mass-resolving power $R_{\text{FWHM}} = m/\Delta m$ in excess of 100000 [28–30] that is obtained in only several tens of milliseconds compared to hundreds of milliseconds in Penning traps. This gain of an order of magnitude in time expands the frontiers of exotic nuclides accessible by ion-trap facilities, as for ^{82}Zn with a half-life of $t_{1/2} = 228(10)$ ms [31].

In this first on-line application, the $^{82}\text{Zn}^+$ ions were sent from the MR-ToF MS through a Bradbury-Nielsen gate [32], which deflected the contaminants. In particular, the radioactive ion beam from ISOLDE was accumulated for 100 ms after each proton pulse in the RFQ cooler and buncher and stored for additional 5 ms to thermalize the last incoming ions in a helium buffer-gas environment. The ensemble containing the mixture of $^{82}\text{Zn}^+$ and $^{82}\text{Rb}^+$ isotopes was then injected into the MR-ToF mass separator for 100 revolutions, equivalent to a flight time of about 2.5 ms. This was sufficient to separate the two species by multiple signal widths; i.e., the difference in time of flight was ≈ 500 ns and the individual signal widths ≈ 200 ns at the 1%-intensity level. This corresponds to a mass-resolving power of about $R_{1\%} = m/\Delta m = 6000$, adequate to separate the $^{82}\text{Rb}^+$ contamination. Subsequently, the $^{82}\text{Rb}^+$ ions were deflected from the beam-line axis by the Bradbury-Nielsen gate and thus removed from the bunch before injection into the next ion trap. This successful purification of an isobaric radioactive beam was decisive for the present measurement.

The isolated sample was transferred to the first of the two Penning traps situated in individual superconducting solenoids, where the ions were cooled in a helium buffer-gas environment as a preparation for the final mass measurement in the second, hyperbolic high-precision Penning trap. This preparation was accomplished in only 15 ms by applying a high helium pressure of about 10^{-3} mbar. The whole accumulation, purification, and preparation steps were performed in less than 25 ms, which is an order of magnitude faster compared to the commonly used mass-selective buffer-gas centering [33]. In the precision Penning trap, the ions' cyclotron frequency, $\nu_c = qB/(2\pi m)$ in the magnetic field, B , was measured via their flight time to a particle detector (time-of-flight ion-cyclotron resonance, ToF-ICR [34]) as a function of the frequency of a radio-frequency (rf) excitation (inset of Fig. 3). To determine the ions' mass $m = r(m_{\text{ref}} - m_e) + m_e$, the cyclotron-frequency ratio $r = \nu_c^{\text{ref}}/\nu_c$ of the ion of interest and a well-known reference isotope to calibrate the magnetic field, here $^{85}\text{Rb}^+$, has to be evaluated.

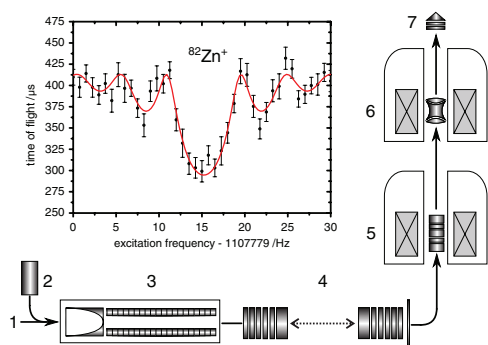


FIG. 3 (color online). Schematic ISOLTRAP overview and time-of-flight resonance of $^{82}\text{Zn}^+$. The main components relevant for this study are the incoming ISOLDE beam (1), reference ion source (2), RFQ cooler and buncher (3), MR-ToF mass separator (4), preparation Penning trap (5), precision Penning trap (6), and ToF detector (7). Top-left inset: Time-of-flight resonance of $^{82}\text{Zn}^+$ (see text).

In total, five measurements could be completed in 16 h with a total number of 1754 detected $^{82}\text{Zn}^+$ ions. These experiments included three conventional ToF-ICR measurements, one with 100 ms and two with 200 ms rf-excitation time, and two Ramsey-type ToF-ICR resonances [35] with an excitation-waiting-excitation scheme of 20 ms-160 ms-20 ms. The measurements resulted in a mean frequency ratio for $^{82}\text{Zn}^+$ and $^{85}\text{Rb}^+$ of $r = \nu_c^{\text{ref}}/\nu_c = 0.9651728601(391)$ and a mass-excess value of $\text{ME}(^{82}\text{Zn}) = m - A = -42.314(3) \text{ MeV}/c^2$ (where A is the mass number and u the unified atomic mass unit) with a relative mass uncertainty of $\delta m/m = 4 \times 10^{-8}$. The latter is dominated by the statistical uncertainty of the cyclotron frequency.

With this new ^{82}Zn mass value, calculations as described in Ref. [8] were performed to drill deeper down into the neutron-star crust. To this end the nuclides which minimize the Gibbs free energy per nucleon were determined as a function of the pressure in the neutron-star crust. By the use of the TOV equations, it was found to which depths in the crust these pressures correspond. The composition profile of a (cold, non-accreting and non-rotating) neutron star of 1.4 solar mass and 10 km radius was calculated with the new ^{82}Zn mass value and compared to the three most recent Brussels-Montreal mass tables HFB-19, HFB-20, and HFB-21. We have restricted our comparisons to these models because we also use their predictions of the equation of state of neutron-star matter for consistency (few mass models can provide such information). A more extensive study, involving a wide range of mass models, is beyond the scope of this Letter and will be the subject of a future publication. ^{82}Zn is considerably less bound than predicted by HFB-19 and HFB-20 ($\text{ME}_{\text{HFB-19,20}} = -42.96 \text{ MeV}/c^2$). From our measurement, ^{82}Zn is no longer present in the neutron-star crust (see Fig. 1). Moreover, the location of ^{80}Zn is limited to a deeper level (223 m) than predicted by HFB-19. This result has extended the knowledge of the neutron-star crust composition, literally, to new depths.

While HFB-19 (as well as HFB-20) did not correctly predict the new ^{82}Zn mass, the prediction of HFB-21 was close enough ($\text{ME}_{\text{HFB-21}} = -42.70 \text{ MeV}/c^2$) that the predicted neutron-star profile remains the same. Moreover, the fact that HFB-21 is slightly favored is significant in light of another recent mass measurement: that of the neutron star PSR J1614-2230. The measured value of 1.97(4) solar masses [36] excludes many theoretical models for the dense-matter EOS, in particular that of HFB-19, which predicts a lower maximum neutron-star mass [37]. As explained in Ref. [11], the three models HFB-19 to 21 were purposely constrained to reproduce three different representative realistic neutron-matter EOS (as obtained from many-body calculations using realistic two- and three-body forces) up to the highest densities prevailing in neutron stars. It is intriguing that a new mass

measurement from nuclear physics is consistent with a new mass from astronomy and that this advance in nuclear theory passes an important test of consistency and accuracy for astrophysical applications.

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7 Publications

7.1 Peer-reviewed publications

- New developments of the in-source spectroscopy method at RILIS/ISOLDE, B. A. Marsh, B. Andel, A. N. Andreyev, S. Antalic, D. Atanasov, A. E. Barzakh, B. Bastin, Ch. Borgmann, L. Capponi, T. E. Cocolios, T. Day Goodacre, M. Dehairs, X. Derkx, H. De Witte, D. V. Fedorov, V. N. Fedosseev, G. J. Focker, D. A. Fink, K. T. Flanagan, S. Franchoo, L. Ghys, M. Huyse, N. Imai, Z. Kalaninova, U. Köster, S. Kreim, N. Kesteloot, Y. Kudryavtsev, J. Lane, N. Lecesne, V. Liberati, D. Lunney, K. M. Lynch, V. Manea, P. L. Molkanov, T. Nicol, D. Pauwels, L. Popescu, D. Radulov, E. Rapisarda, M. Rosenbusch, R. E. Rossel, S. Rothe, L. Schweikhard, M. Seliverstov, S. Sels, A. M. Sjödin, V. Truesdale, C. Van Beveren, P. Van Duppen, K. Wendt, F. Wienholtz, R. N. Wolf, S. G. Zemlyanoy, [Nucl. Instr. and Meth. in Phys. Res. B](#), accepted for publication
- Recent developments of the mass spectrometer ISOLTRAP, S. Kreim, D. Beck, K. Blaum, Ch. Böhm, Ch. Borgmann, M. Breitenfeldt, T. E. Cocolios, D. Fink, S. George, F. Herfurth, A. Herlert, A. Kellerbauer, U. Köster, M. Kowalska, D. Lunney, V. Manea, E. Minaya-Ramirez, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, R. N. Wolf, K. Zuber, [Nucl. Instr. and Meth. in Phys. Res. B](#), accepted for publication
- ISOLTRAP's Multi-Reflection Time-of-Flight Mass Separator/Spectrometer, R. N. Wolf, F. Wienholtz, D. Atanasov, D. Beck, K. Blaum, Ch. Borgmann, F. Herfurth, M. Kowalska, S. Kreim, Y. Litvinov, D. Lunney, V. Manea, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, K. Zuber, [Int. J. Mass Spectrom.](#) **349-350**, 100 years of Mass Spectrometry, 123–133 (2013)
- Masses of exotic calcium isotopes pin down nuclear forces, F. Wienholtz, D. Beck, K. Blaum, Ch. Borgmann, M. Breitenfeldt, R.B. Cakirli, S. George, F. Herfurth, J.D. Holt, M. Kowalska, S. Kreim, D. Lunney, V. Manea, J. Menendez, D. Neidherr, M. Rosenbusch, L. Schweikhard, A. Schwenk, J. Simonis, J. Stanja, R. N. Wolf, K. Zuber, [Nature](#) **498**, 346-349 (2013)
- Towards systematic investigations of space-charge phenomena in multi-reflection ion traps, M. Rosenbusch, S. Kemnitz, R. Schneider, L. Schweikhard, R. Tschiersch, and R. N. Wolf, [AIP Conf. Proc.](#) **1521**, 53 (2013)
- Plumbing Neutron Stars to New Depths with the Binding Energy of the Exotic Nuclide ^{82}Zn , R. N. Wolf, D. Beck, K. Blaum, Ch. Böhm, Ch. Borgmann, M. Breitenfeldt, N.

- Chamel, S. Goriely, F. Herfurth, M. Kowalska, S. Kreim, D. Lunney, V. Manea, E. Minaya Ramirez, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, and K. Zuber, *Phys. Rev. Lett.* **110**, 041101 (2013)
- Buffer-gas-free mass-selective ion centering in Penning traps by simultaneous dipolar excitation of magnetron motion and quadrupolar excitation for interconversion between magnetron and cyclotron motion,
M. Rosenbusch, K. Blaum, Ch. Borgmann, S. Kreim, M. Kretzschmar, D. Lunney, L. Schweikhard, F. Wienholtz, and R. Wolf, *Int. J. Mass Spectrom.* **325-327**, 51-57 (2012)
 - Surveying the $N = 40$ island of inversion with new manganese masses,
S. Naimi, G. Audi, D. Beck, K. Blaum, Ch. Böhm, Ch. Borgmann, M. Breitenfeldt, S. George, F. Herfurth, A. Herlert, A. Kellerbauer, M. Kowalska, D. Lunney, E. Minaya Ramirez, D. Neidherr, M. Rosenbusch, L. Schweikhard, R. N. Wolf, and K. Zuber, *Phys. Rev. C* **86**, 014325 (2012)
 - On-line separation of short-lived nuclei by a multi-reflection time-of-flight device,
R. N. Wolf, D. Beck, K. Blaum, Ch. Böhm, Ch. Borgmann, M. Breitenfeldt, F. Herfurth, A. Herlert, M. Kowalska, S. Kreim, D. Lunney, S. Naimi, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, F. Wienholtz, K. Zuber, *Nucl. Instrum. Methods Phys. Res., Sect. A* **686**, 82-90 (2012)
 - Static-mirror ion capture and time focusing for electrostatic ion-beam traps and multi-reflection time-of-flight mass analyzers by use of an in-trap potential lift,
R. N. Wolf, G. Marx, M. Rosenbusch, L. Schweikhard, *Int. J. Mass Spectrom.* **313**, 8-14 (2012)
 - A multi-reflection time-of-flight mass separator for isobaric purification of radioactive ion beams,
R. N. Wolf, M. Eritt, G. Marx, L. Schweikhard, *Hyperfine Interact.* **199**, 115-122 (2011)

7.2 Miscellaneous, non-reviewed publications

- ISOLTRAP's MR-ToF mass separator to plumb a neutron star,
R. Wolf for the ISOLTRAP collaboration, CERN PH newsletter, 23.03.2013
<http://ph-news.web.cern.ch/tags/isoltrap>
- IS413: High-Precision Mass Measurements of n-rich Zinc Isotopes
R. Wolf for the ISOLTRAP Collaboration, CERN ISOLDE newsletter 2012
<http://isolde.web.cern.ch/sites/isolde.web.cern.ch/files/May%202012.pdf>
- IS498: A multi-reflection time-of-flight mass separator for isobaric purification at ISOLTRAP
R. N. Wolf and L. Schweikhard for the ISOLTRAP collaboration, CERN ISOLDE

newsletter 2011

<http://isolde.web.cern.ch/sites/isolde.web.cern.ch/files/April%202011.pdf>

- Aufbau und Test einer elektrostatischen Ionenstrahlfalle
Diploma Thesis, Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald
http://www6.physik.uni-greifswald.de/ClusterTrap/wolf/Diplom.Wolf_2008.pdf

8 Presentations

- ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer, Innovations in Mass Spectrometry Instrumentation Conference (INNMASSSPEC2013), Saint-Petersburg, Russia, 14.-18.07.2013, Poster
- ISOLTRAP's MR-ToF mass separator/spectrometer, 5th International Workshop on Electrostatic Storage Devices (ESD2013), Max-Planck Institute for Nuclear Physics Heidelberg, Germany, 17.-21.06.2013, Invited talk (in representation for Frank Wienholtz)
- ISOLTRAP's MR-ToF mass separator/spectrometer, 2nd EURISOL-NET (ENSAR/NA03) Working Group meeting, University of Jyväskylä, Finland, 28.-29.05.2013, Invited talk
- ISOLTRAP's MR-ToF mass separator/spectrometer, 530. WE-Heraeus-Seminar: Nuclear masses and nucleosynthesis, Bad Honnef, Germany, 23.-26.04.2013, Invited talk
- Mass separation with ISOLTRAP's MR-ToF, DPG spring meeting, Hannover, Germany, 18.-22.03.2013, Talk
- Mass separation with ISOLTRAP's MR-ToF, DPG spring meeting, Dresden, Germany, 04.-08.03.2013, Poster
- First on-line application of a multireflection electrostatic ion trap for isobaric purification of exotic beams, Fundamental Interactions with Atom & Ion Traps (FUNTRAP2012), 02.-06.12.2012, Weizmann Institute of Science, Rehovot, Israel, Invited talk
- The ISOLTRAP multi-reflection time-of-flight mass analyzer, 16th International Conference on Electromagnetic Isotope Separators and Techniques Related to their Applications (EMIS2012), 02.-07.12.2012, Matsue, Japan, Poster
- MR-ToF Isobar Separation at ISOLTRAP, EMMI Workshop: Physics Prospects at FLAIR - The Facility for Low-Energy Antiproton and Ion Research, 03.-04.05.2012, GSI, Darmstadt, Germany, Poster
- A multi-reflection time-of-flight mass separator for high-resolution beam purification at ISOLTRAP, 496. Wilhelm und Else Heraeus-Seminar: Astrophysics with modern small-scale accelerators, Bad Honnef, 06.-10.02.2012, Invited talk
- MR-TOF as an isobar filter at ISOLTRAP, Physics of Rare-RI Ring workshop (OMEG11), RIKEN Nishina Center, Hirosawa, Wako-shi, Saitama, Japan, 10.-12.11.2011, Talk

- The ISOLTRAP electrostatic MR-TOF mass separator, 4th International Workshop on Electrostatic Storage Devices, Gatlinburg, TN, USA, 08.-11.06.2011, Invited talk
- First application of a multi-reflection time-of-flight mass separator to radioactive ion beams, 59th ASMS Conference on Mass Spectrometry and Allied Topics, Denver, CO, USA, 05.-09.06.2011, Poster
- First application of a multi-reflection time-of-flight mass separator to radioactive ion beams, International Max Planck Research School Colloquium, Greifswald, Germany, 05.05.2011, Talk
- First application of a multi-reflection time-of-flight mass separator to radioactive beams, DPG spring meeting, Dresden, Germany, 13.-18.03.2011, Invited talk
- Implementation of a MR-ToF isobar separator at ISOLTRAP and first online results, ISOLDE workshop, CERN, Geneva, Switzerland, 08.-10.12.2010, Talk
- Implementation of a MR-ToF isobar separator at the on-line mass spectrometer ISOLTRAP, 1st European Conference on Trapped Ions, County Durham, UK, 19.-24.09.2011, Poster
- A multi-reflection time-of-flight mass separator for isobaric purification at ISOLTRAP, 2nd International European Radioactive Ion Beam Conference, Lamoura, France, 06.-11.06.2010, Poster
- A multi-reflection time-of-flight mass separator for isobaric purification at ISOLTRAP, 5th Conference on trapped charged particles and fundamental physics, Saariselkä, Finland, 12.-16.04.2010, Poster
- Implementation of a multi-reflection time-of-flight mass separator at ISOLTRAP, DPG spring meeting, Bonn, Germany, 15.-19.03.2011, Poster
- Development of a high resolution mass separator for ISOLTRAP, 18th International Mass Spectrometry Conference, Bremen, Germany, 30.08.-04.09.2011, Poster
- A high resolution multi-reflection time-of-flight mass separator for isobaric purification, 3rd International Workshop on Electrostatic Storage Devices, Aarhus, Denmark, 21.-25.06.2009, Oral presentation based on poster abstract
- A high resolution multi-reflection time-of-flight mass separator for isobaric purification, International Max Planck Research School Colloquium, Greifswald, Germany, 14.05.2009, Talk
- An electrostatic mass separator for ISOLTRAP, DPG spring meeting, Hamburg, Germany, 02.-05.03.2009, Poster
- An electrostatic storage device for mass separation and contamination removal at ISOLTRAP, DPG spring meeting, Darmstadt, Germany, 10.-14.03.2008, Poster
- An electrostatic storage device for mass separation and contamination removal at ISOLTRAP, DGMS annual meeting, Giessen, Germany, 02.-05.03.2008, Poster

9 Erklärung

Hiermit erkläre ich, dass diese Arbeit bisher von mir weder an der Mathematisch-Naturwissenschaftlichen Fakultät der Ernst-Moritz-Arndt-Universität Greifswald noch einer anderen wissenschaftlichen Einrichtung zum Zwecke der Promotion eingereicht wurde.

Ferner erkläre ich, daß ich diese Arbeit selbständig verfasst und keine anderen als die darin angegebenen Hilfsmittel und Hilfen benutzt und keine Textabschnitte eines Dritten ohne Kennzeichnung übernommen habe.

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