

# Precision mass measurements for nuclear astro- and neutrino physics

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**Abstract.** Nuclear masses are indispensable ingredients in numerous physics applications ranging from nuclear structure physics, where, e.g., the shell closures and nucleon correlation energies can be studied by accurate mass measurements, via the nuclear astrophysics, where the masses of nuclei far from the valley of  $\beta$ -stability determine the pathways of, e.g.,  $r$ p- and  $r$ -processes of nucleosynthesis in stars, to tests of the standard model and fundamental interactions, where, e.g., the very-accurate masses of parent and superallowed  $\beta$ -decay daughter nuclei serve as one of inputs for the checking of the unitarity of the CKM quark-mixing matrix. In this review we focus on recent direct mass measurements conducted with storage rings and Penning trap mass spectrometry. Although these measurements have a broad impact, we restrict our discussion on two topics, namely nuclear astrophysics and neutrino physics.

## 1. Introduction

Atomic nuclei are unique many-body systems composed of two types of fermions, namely protons and neutrons, in which the binding energy is the result of the interplay of the strong, weak and electromagnetic fundamental interactions acting between the constituent nucleons. The complexity of these systems is, on one hand, a challenge for *ab-initio* theories but, on the other hand, they are natural laboratories for investigating these interactions.

Since the first experiments of J. J. Thomson about a century ago [1], which can be considered as a birth of mass spectrometry, mass measurements are one of the major boosters of physics research. In nuclear physics, the structure effects like, e.g., shell closures [2,3] or nucleon-nucleon pairing [4], have been discovered in the past as irregularities on the smooth nuclear mass surface. Also today, nuclear masses continue to be a very useful tool to study nuclear structure, like, e.g., phenomenological proton-neutron interaction,  $\delta V_{pn}$ , [5,6], onset of nuclear deformations [7], halo nuclei [8], shell structure [9], collective excitations [10], etc. Although, the masses of about 3000 nuclides are known experimentally, the knowledge of still unknown masses of very neutron-rich nuclei turns to be decisive in our understanding of the  $r$ -process on nucleosynthesis in stars, the process responsible for the production of about a half of all heavy elements above iron [11]. We note, however, that many of the nuclei on the  $r$ -process path will remain inaccessible even at the next generation radioactive beam facilities and, hence, their masses have to be calculated.

Thus, new mass measurements are also necessary for testing and constraining the modern nuclear theories (for details see recent reviews [12, 13]).

Modern methods for mass measurements of exotic and stable nuclei are based on the determination of revolution frequencies of the corresponding ions in magnetic fields. There are two highly complementary approaches, namely Storage Ring and Penning-Trap Mass Spectrometry [14]. While, the former one is superior in terms of the number of nuclides which can be measured simultaneously, the highest mass accuracy can be achieved with the latter. Since several exhaustive reviews have been published in the last few years covering a broad range of applications of the mass spectrometric data (see, e.g., Refs. [13–16]), we focus here on two topics which profited significantly from recent accurate Penning-trap and storage ring mass measurements, namely nuclear astrophysics and neutrino physics.

## 2. High-Precision Mass Spectrometry Methods

### 2.1. Storage Ring Mass Spectrometry

Presently there are two storage ring facilities worldwide performing direct mass measurements of radioactive nuclei [17]. These facilities are the Experimental Storage Ring ESR at GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany [18] and the Experimental Cooler-Storage Ring CSRe at the Institute for Modern Physics in Lanzhou, China [19]. There, the exotic nuclei are produced at relativistic energies employing the in-flight method, which provides fast and chemically independent separation. However, different to ISOL beams, commonly used in Penning-Trap Mass Spectrometry, the secondary beams are characterized by large phase-spaces [20].

In first-order approximation the revolution frequencies  $f$  of the stored ions can be related to their mass-to-charge ratios ( $m/q$ ) by the following expression [15, 21, 22]:

$$\frac{\Delta f}{f} = -\frac{1}{\gamma_t^2} \frac{\Delta(m/q)}{(m/q)} + \left(1 - \frac{\gamma}{\gamma_t^2}\right) \frac{\Delta v}{v}, \quad (1)$$

where  $\Delta v/v$  is the velocity spread of the ions,  $\gamma$  the relativistic Lorentz factor and  $\gamma_t$  the transition point of a storage ring, which is constant for a given ion-optical setting of the ring. Thus, in order to measure the  $m/q$  of stored ions, one has to measure the corresponding revolution frequency  $f$  and minimize the term containing  $\Delta v/v$ .

Two complementary techniques have been developed for in-ring mass measurements, namely Schottky (SMS) and Isochronous (IMS) Mass Spectrometry [15]. In the SMS, the velocity spread is reduced by stochastic and electron cooling of the stored ion beams. For intensities of below about a thousand stored ions,  $\Delta v/v \sim 5 \cdot 10^{-7}$  can be achieved. However, the cooling requires a few seconds and only nuclear species with longer half-lives can thus be investigated with the SMS. The measurement of the revolution frequencies is non-destructive and is based on the Fourier transform of the periodic signals induced by the circulating ions on a Schottky pick-up.

In the IMS, the ring is tuned into the so-called isochronous ion-optical mode [23]. In this mode  $\gamma_t \sim 1.41$  and 1.40 for ESR and CSRe, respectively. The ions are injected and stored in the ring with the corresponding  $\gamma = \gamma_t$ . In this mode, the velocity spread is compensated by the orbit lengths of the stored ions. The frequencies are measured with a dedicated time-of-flight detector. A thin (a few  $\mu\text{g}/\text{cm}^2$ ) foil is penetrated by swift ions thus releasing secondary electrons from the foil surface. The electrons are detected and the information on revolution frequencies can be extracted. No cooling is required in the IMS and nuclides with half-lives as short as a few ten  $\mu\text{s}$  (a few ten revolutions in the ring) can be addressed [24].

Both techniques are sensitive to single stored ions and can be applied to measure masses of nuclides with extremely small production yields of less than 1 ion per day. This has, e.g., been demonstrated by a recent ESR measurement on a single stored  $^{208}\text{Hg}^{79+}$  ion, which was produced only once during a two-week experiment [25].

## 2.2. Penning-Trap Mass Spectrometry

Currently there are several operating on-line Penning trap facilities in the world [14]. Unlike in storage rings, in Penning traps the ions are confined to very small volume of space. Irrespective of the production method of the ions of interest, they need to be decelerated and cooled down to below 1 keV of energy before they can be loaded into a trap for mass measurements.

A Penning trap consists of a strong homogenous magnetic field and a static quadrupolar electric potential [26]. The electric potential is formed with electrodes, either of the hyperbolic or cylindrical shape. The idea behind a Penning-trap mass measurement is to measure ion's mass  $m$  by measuring its free cyclotron frequency (for details see, e.g., Ref. [14])

$$\nu_c = \frac{1}{2\pi} \frac{q}{m} B, \quad (2)$$

where  $q$  is the charge of the ion and  $B$  the magnetic field. With on-line traps  $\nu_c$  is derived from the sideband frequency ( $\nu_+ + \nu_-$ ) by coupling the two radial eigenmotions with an azimuthal quadrupole electric field. So far every on-line trap facility has been using the time-of-flight ion-cyclotron (TOF-ICR) technique [27, 28] to determine the sideband frequency which corresponds to  $\nu_c$  with high precision [29]. Typically, relative mass accuracy of better than  $10^{-7}$  can be achieved in absolute mass measurements. Due to cancellation of systematic uncertainties, mass difference measurements of ions having same  $A/q$  have been measured even with  $10^{-9}$  precision [30]. Such high mass accuracies enable tests of fundamental symmetries (see, e.g., Ref. [14]) as well as can provide important information relevant for the neutrino research, one of the subjects discussed below.

## 3. Recent applications of high-precision mass data

### 3.1. Nuclear Astrophysics

In the past decade, nuclear astrophysics studies have tremendously benefitted from high-precision atomic mass measurements of short-living species. In recent years, masses near the astrophysical  $rp$ - and  $\nu p$ -processes have been covered with relative precision of better than  $10^{-7}$ . The results have been obtained by several Penning trap groups like ISOLTRAP at ISOLDE/CERN [31–35], JYFLTRAP at the University of Jyväskylä [36–40], SHIPTRAP at GSI [37, 41, 42], CPT at Argonne National laboratory [46–49] and LEBIT at MSU [50, 51]. The extensive coverage of these measurements is shown in Fig. 5 of Ref. [35]. Accurate knowledge of masses allows extraction of proton threshold energies which are used to derive *astrophysical reaction rates*. These are sensitive; a few-keV shift can actually change astrophysical reaction rates by orders of magnitude [40, 52].

Before the new Penning-trap mass measurements, calculations using previous mass data showed that the  $rp$ -process is terminated with the SnSbTe cycle [53]: Alpha decay channels in tellurium isotopes hinder the  $rp$  process to proceed further. The new mass values from JYFLTRAP [38] indicate considerable decrease of such recycling starting at  $^{105}\text{Sn}$  and thus the  $rp$ -process path moves closer to stability before the formation of the SnSbTe cycle. Another interesting result is a much lower  $\alpha$  separation energy in  $^{84}\text{Mo}$  [42]. This opens up a possibility for a previously unforeseen existence of ZrNb cycle, which would impose an upper temperature limit for the  $rp$ -process to synthesize nuclei beyond  $A = 84$ . Although a large region on the mass surfaces has been covered with high-precision mass spectrometry, more measurements are necessary along the  $rp$ -process path near and at the  $N = Z$  line.

First IMS measurements at the CSRe have provided accurate masses of short-lived  $rp$ -process nuclides  $^{63}\text{Ge}$ ,  $^{65}\text{As}$ ,  $^{67}\text{Se}$ , and  $^{71}\text{Kr}$  [43]. These measurements combined with the recent Penning-trap data have been implemented into the X-ray burst calculations. A breathtaking result is, that the previously considered waiting point  $^{64}\text{Ge}$  is *not* a significant  $rp$ -process waiting point for most relevant astrophysical conditions. This is particularly important, since  $^{64}\text{Ge}$  has a

long  $\beta$ -decay half-life of  $T_{1/2} = 64(3)$  s [44] and is the first waiting point encountered in the  $rp$ -process. By pinning down the role of  $^{64}\text{Ge}$  also the uncertainties in the final abundances of X-ray burst ashes above  $A > 64$  could be dramatically reduced. The measurements at CSRe have been recently extended to  $A = 2Z - 2$  nuclei, which will address the  $rp$ -process path in the  $A < 60$  region [45].

While the progress in mass measurements along the  $rp$ -process is tremendous, nuclides at the  $r$ -process stay mostly inaccessible at the present RIB facilities. The  $r$ -process abundances show pronounced peaks at  $A \sim 80, 130$ , and  $195$ , which are assigned to possible waiting point nuclei. JYFLTRAP and ISOLTRAP measurements of masses near  $N = 50$  have provided high-precision data for improved theoretical description of various separation energies [54, 55]. For example, masses of  $^{80-81}\text{Zn}$  have constrained the temperature vs. neutron-density conditions at which  $^{80}\text{Zn}$  is the  $r$ -process waiting point [55]. Beta-decay endpoint measurements on  $^{130}\text{Cd}$  have suggested, that  $N = 82$  shell closure might be quenched at  $Z = 48$  thus making  $^{130}\text{Cd}$  not be a waiting point [56]. The studies of isomeric decays of  $^{130}\text{Cd}$ , on the other hand, conclude that  $N = 82$  shell is not quenched at  $Z = 48$  [57]. It has been shown by the recent Penning trap measurements (see for example Refs. [58, 59]), that  $\beta$ -decay endpoint measurements may have systematic uncertainties and have to be verified if possible. Approaching  $^{130}\text{Cd}$  with direct mass measurements is still very complicated. Mass measurements of Cd isotopes only up to  $A = 128$  could be performed at ISOLTRAP [60]. Therefore, the present studies focus on nuclear structure developments in neutron-rich nuclides, for instance on the development of the  $N = 82$  and  $N = 126$  neutron shell closures [61]. Furthermore, ISOLTRAP measurements of  $^{132,134}\text{Sn}$  isotopes [62] and ESR measurements of neutron-rich Sb, Te and I isotopes [63] show significant deviations of the experimental mass values to the predictions of the ETFSI-Q mass model [64], in which the  $N = 82$  shell quenching is introduced explicitly.

### 3.2. Neutrino Physics

Neutrinos might be the most intriguing particles discovered so far. Postulated by Pauli in a letter to a local meeting on radioactivity at Tübingen, Germany, in 1930, they waited a quarter of a century for their discovery [65]. Until very recently neutrinos were considered to be massless, electrically neutral, weakly interacting, left-handed particles with spin 1/2. But the discovery of neutrino oscillations has finally placed neutrinos in the family of massive particles. Unfortunately, the oscillation experiments do not shed light on the rest mass of the neutrino, which has thus to be measured in independent experiments.

Penning traps due to their tremendous progress in the last decade [14] can provide experiments on the determination of the neutrino mass with invaluable information by measuring the masses of the involved nuclides with very high accuracy.

So far the most stringent limit of approximately 2 eV at 95% c.l. on the mass of the *electron antineutrino* has been derived from the investigation of the end point of the  $\beta^-$ -decay spectrum of tritium [66]. Here, the  $Q$ -value, which is the mass difference of tritium and  $^3\text{He}$ , enters into the fit of the decay spectrum as a free parameter. A cross-check of the  $Q$ -value obtained from the fit with that determined by direct Penning trap mass measurements can help shed light on possible undesirable systematic effects in these end point measurements. The most precise  $Q$ -value of  $18589.8(12)$  eV of tritium decay was obtained at SMILETRAP [67].

In the future experiment KATRIN, the upper limit on the mass of the *electron antineutrino* is aimed to be improved tenfold to about 0.2 eV (95% c.l.) [68]. This requires an improvement of our knowledge of the  $Q$ -value of tritium decay to a level of at least 100 meV. The THe-TRAP, which is under construction in Heidelberg, is called to achieve this goal [69].

Another project MARE aims to determine the *electron antineutrino* mass with an uncertainty of 0.2 eV (90% c.l.) from the  $\beta^-$ -decay of  $^{187}\text{Re}$  in a way very similar to KATRIN [70]. Here, the mass difference of  $^{187}\text{Re}$  and  $^{187}\text{Os}$  should be known with unprecedented relative uncertainty

of a few parts in  $10^{12}$ . This will be one of the goals of the PENTATRAP project [71, 72].

In contrast to the *electron antineutrino* mass, for the *electron neutrino* a much less stringent limit of only 225 eV was obtained from the investigation of orbital electron capture (EC) decay in  $^{163}\text{Ho}$ , the nuclide with the smallest known  $Q$ -value of the EC decay,  $Q_{\text{EC}}$ , [73]. At present, the most promising approach is based on combined efforts of cryogenic microcalorimetry and high-precision Penning-trap mass spectrometry. Calorimeters can measure the end point of the atomic de-excitation spectrum which follows the EC decay, whereas Penning traps account for the determination of the corresponding  $Q_{\text{EC}}$ -value. In order to push the upper limit on the *electron neutrino* mass to below 1 eV, the  $Q_{\text{EC}}$ -value in  $^{163}\text{Ho}$  must be determined with a similar uncertainty. Such a measurement will become feasible as soon as the PENTATRAP setup is taken into operation [71, 72].

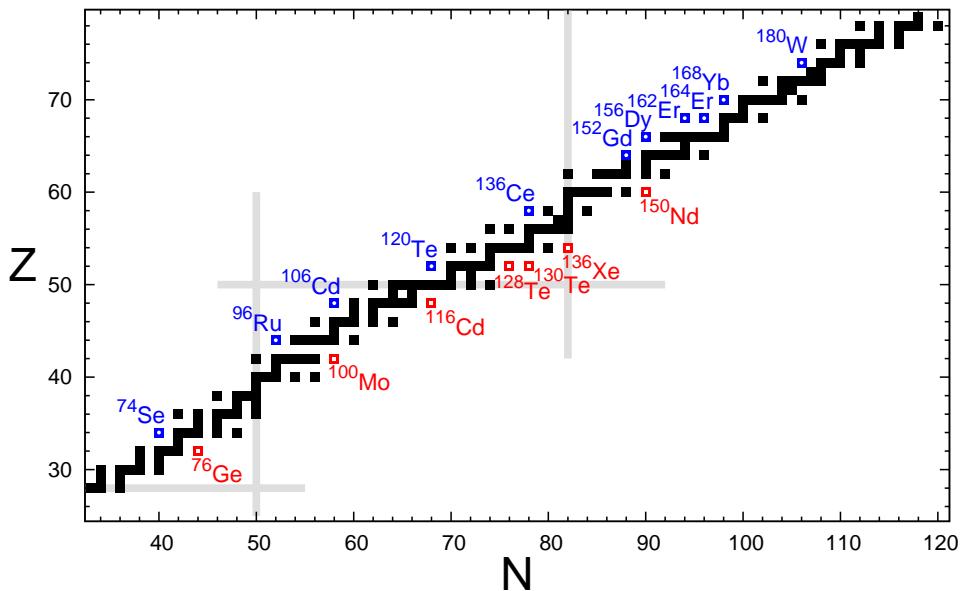
Besides  $^{163}\text{Ho}$ , there is a variety of nuclides with  $Q_{\text{EC}}$ -values below 100 keV, which are potentially suitable candidates for the determination of the *electron neutrino* mass from EC decay [74]. The choice of the best candidate among them is hampered by the imprecise knowledge of their  $Q_{\text{EC}}$ -values. Therefore, it was recently proposed to initiate a search for the best candidate among these nuclides by employing the high precision Penning trap spectrometry for measuring their masses. The  $Q_{\text{EC}}$ -value of one of such candidates, namely  $^{194}\text{Hg}$ , has already been measured at ISOLTRAP with an uncertainty of few keV [74]. This measurement has excluded  $^{194}\text{Hg}$  from the list of suitable candidates for the determination of the *electron neutrino* mass.

Since the neutrino is an electrically neutral particle, it can be a Dirac or a Majorana particle. Clarifying this ambiguity is essential. The best way to address this question is to search for the neutrinoless double  $\beta^-$ -decay,  $0\nu\beta^-\beta^-$ , or the neutrinoless double EC decay,  $0\nu2\text{EC}$ . These decays can only exist if the neutrino is its own antiparticle, *i.e.* a Majorana particle.

In Nature, there exist 35 nuclides which can undergo  $0\nu\beta^-\beta^-$ , but only eleven are of practical use for the search of  $0\nu\beta^-\beta^-$  due to their sufficiently large  $Q$ -values. An observation of a peak corresponding to the  $Q$ -value of the decay in a sum energy spectrum of the two emitted electrons would be a signature of the occurrence of the  $0\nu\beta^-\beta^-$ -decay. The width of the peak is approximately a few keV and is defined by the energy resolution of the electron detector. Unfortunately, the neutrinoless mode has to be observed against a huge background from the dominating “two-neutrino” mode. Thus, the knowledge of the  $Q$ -value of the decay with an uncertainty of below 1 keV is important in order to correctly interpret the results of the experiments searching the  $0\nu\beta^-\beta^-$ -decays. Here, the Penning-trap mass spectrometry has provided high-precision  $Q$ -values for the four most important double  $\beta^-$ -decay transitions used in large-scale experiments, *i.e.* the double  $\beta^-$ -decays of  $^{76}\text{Ge}$ ,  $^{100}\text{Mo}$ ,  $^{130}\text{Te}$  and  $^{136}\text{Xe}$  [75–81]. Although, the remaining candidates, *i.e.*  $^{48}\text{Ca}$ ,  $^{82}\text{Se}$ ,  $^{96}\text{Zr}$ ,  $^{110}\text{Pd}$ ,  $^{116}\text{Cd}$ ,  $^{124}\text{Sn}$  and  $^{150}\text{Nd}$ , are not yet considered in the present experiments, they might become attractive choices in future.  $Q$  value of  $^{116}\text{Cd}$  has been recently measured at JYFLTRAP [82]. The expected half-life is  $t_{1/2} = 10^{26}$  years (if assuming  $m_\nu = 20$  meV, a value covering the inverted mass hierarchy) and the decay of  $^{116}\text{Cd}$  could well be detected in the next generation experiments [83]. The  $Q$ -values of the other cases will be addressed by Penning traps soon.

In contrast to the neutrinoless double  $\beta^-$ -decay, the neutrinoless double EC-decay was long considered to be a virtually unobservable process due to the expected very long half-lives. However, in some special cases of  $0\nu2\text{EC}$ , when the initial and final states of the transition are degenerate in energy, *e.g.* when the  $Q$ -value is close to the energy of an excited state in the daughter atom, a strong resonant enhancement of the decay probability is expected [84]. The main uncertainty in the determination of the resonance conditions – a poor knowledge of the corresponding  $Q$ -values – can be overcome by high-precision mass measurements.

A search for resonantly enhanced  $0\nu2\text{EC}$  transitions was initiated at JYFLTRAP [85] with a  $Q$ -value measurement of  $^{112}\text{Sn}$  [86], which was then continued with measurements of  $^{74}\text{Se}$  and  $^{136}\text{Ce}$  [81, 87], out of which  $^{74}\text{Se}$  was measured also with the Penning-trap mass spectrometer



**Figure 1.** The double beta decay and double electron capture nuclei whose  $Q$ -values have been determined with Penning traps. Red symbols south-east from the valley of  $\beta$ -stability denote double beta decay emitters and blue symbols (north-west from beta stability) the measured double electron capture nuclei.

at FSU [76]. Very recently, a series of measurements has been done at SHIPTRAP [88], where the  $Q$ -values of  $0\nu 2\text{EC}$  in  $^{152}\text{Gd}$ ,  $^{164}\text{Er}$ ,  $^{180}\text{W}$ ,  $^{156}\text{Dy}$ ,  $^{106}\text{Cd}$ ,  $^{96}\text{Ru}$ ,  $^{162}\text{Er}$  and  $^{168}\text{Yb}$  [89–93] have been determined. In the case of  $^{152}\text{Gd}$  a resonant enhancement is observed, which pushes the half-life estimate down to  $10^{26}$  years when normalizing to the neutrino mass of 1 eV [89]. This is so far the only suitable candidate for a direct observation of  $0\nu 2\text{EC}$ . Furthermore, a unique phenomenon of a multiple resonant enhancement of  $0\nu 2\text{EC}$  has been found in  $^{156}\text{Dy}$  [91]. Unfortunately, the estimated half-life of  $^{156}\text{Dy}$  exceeds that of  $^{152}\text{Gd}$  by two orders of magnitude and thus does not allow considering the  $^{156}\text{Dy}$  nucleus as a suitable candidate for the search of the  $0\nu 2\text{EC}$ -decay.

The  $0\nu 2\text{EC}$  and double  $\beta^-$  emitters investigated so far with Penning traps are shown on a chart of nuclides in Fig. 1. Though several  $0\nu 2\text{EC}$   $Q$ -values have been measured with a high-precision in the past five years, the search for neutrinoless double EC decay is far from completion. Since a bunch of other potentially resonantly enhanced transitions exists, the compact and rather low-cost Penning trap setups are ideal devices to select the suitable cases.

#### 4. Summary and Outlook

In this work we reviewed the recent mass measurements relevant for nuclear astro- and neutrino physics. Although the masses of several ten nuclides are determined yearly for the former application, masses of a lot of important nuclides could still not be measured due to their small production yields and/or too short lifetimes. Therefore, new highly efficient and very fast experimental techniques have to be developed. One example of the ongoing technical developments is a new highly-sensitive resonant Schottky pick-up for the storage ring mass measurements which enabled SMS to address nuclides with half-lives down to a few 10 milliseconds. To increase the efficiency and the mass resolving power, installing of two time-of-flight detectors for in-ring velocity measurement is planned in one of the straight sections of the

CSRe in Lanzhou [94]. Development of the FT-ICR technique for the Penning trap spectrometry allows accurate mass determination from a single stored particle [95]. Storing of the reference and the investigated nuclides in the same magnetic field is a striking feature of the planned PENTATRAP facility [71, 72].

We like to conclude our review with the remark that mass spectrometry is a fast developing field of modern research: There are many new experiments planned worldwide at the existing facilities as well as mass spectrometric setups are planned at *all* next generation radioactive facilities, such as RIBF in RIKEN [96], FRIB at MSU [97], FAIR in Darmstadt [98, 99], Spiral-2 at GANIL [100], KoRIA in South Korea [101] and many others.

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