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# Recent developments in Penning-trap mass spectrometry

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#### ABSTRACT

Penning-trap mass spectrometry provides atomic masses with the highest precision. At accelerator-based on-line facilities it is applied to investigate exotic radionuclides in the context of tests of fundamental symmetries, nuclear structure studies, and nuclear astrophysics research. Recent progress in slowing down radioactive ion-beams in buffer-gas cells in combination with advanced ion-manipulation techniques has paved the way to reach nuclides ever-more far from stability. In this endeavor many efforts are underway to increase the sensitivity, the efficiency, and the precision of Penning-trap mass spectrometry. In this article some recent experimental developments are addressed with the focus on the phase-imaging ion-cyclotron-resonance technique and the Fourier transform ion-cyclotron-resonance technique.

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## 1. Introduction

Penning-trap mass spectrometry (PTMS) is a mature field that has been addressed in several review articles, see for example [1,2], and recently in a special issue on the occasion of the 100th anniversary of mass spectrometry [3]. The mass of an atom provides information on all interactions between its constituents. Nuclear structure information is revealed by the binding energy that is readily obtained from direct mass measurements. Such measurements do not require any knowledge of the nuclear level scheme of the investigated nuclide and are therefore well suited for studies of the nuclear structure evolution far off stability. Such studies provide indicators for changes in the shell structure resulting in different magic numbers [4] and they yield input data for nuclear astrophysical models of nucleosynthesis processes [5,6]. High-precision mass measurements of specific nuclides also contribute to tests of fundamental physics, for example the unitarity of the CKM matrix via the investigation of superallowed beta decays [7].

Since the first Penning trap was installed at an on-line facility about 25 years ago: ISOLTRAP at ISOLDE/CERN [8], PTMS has become a standard tool in nuclear physics studies [1,2]. Many PTMS setups have been installed at on-line facilities worldwide [9–16]. Because these facilities employ different production

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http://dx.doi.org/10.1016/j.nimb.2016.02.027 0168-583X/© 2016 Elsevier B.V. All rights reserved. schemes to produce the isotopes of interest, the various existing PTMS setups are complementary. Utilizing buffer-gas stopping and advanced ion-manipulation techniques, radioisotopes of essentially all elements are nowadays accessible by PTMS.

Many more exotic nuclides will be provided by next-generation radioactive beam facilities. For a bountiful harvest, fast and efficient preparation and measurement schemes are of major importance. Several improvements of PTMS are presently under development. Some of them are specific to the production mechanism such as improved gas catcher systems that will boost the efficiency of PTMS setups at in-flight facilities. To this end cryogenic gas catcher systems are utilized either in a linear geometry [17,18] or in the so-called cyclotron stopper at the NSCL [19]. The reach of PTMS towards nuclides far off stability is not only governed by the overall efficiency, but also by beam purity, and the required number of ions needed to perform an accurate mass measurement. Since many exotic nuclides of interest are rather shortlived, new developments for shorter measurement times without sacrificing precision are also called for.

In this paper two methods are discussed that will pave the way to mass measurements with higher precision, shorter measurement time, and fewer ions. These methods, the Fourier-transform ion-cyclotron-resonance (FT-ICR) [20,21] and the phase imaging ion-cyclotron-resonance (PI-ICR) technique [22], are briefly introduced in Section 2. In Section 3 selected recent experiments are addressed. Section 4 concludes with future perspectives for online mass measurements of radionuclides.





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## 2. Mass measurement techniques

Mass measurements of radionuclides at on-line facilities typically utilize two techniques: mass measurements based on frequency measurements, performed by PTMS and in storage rings [23], and time-of-flight mass spectrometry [24]. While the latter reaches far off stability due to short measurement times, the achievable precision is typically limited to a level of  $\delta m/m \approx 10^{-6}$  for conventional spectrographs [24] and below  $10^{-7}$  (for stable nuclides) in the case of multi-reflection time-offlight systems [25]. PTMS provides masses with a relative precision on the order of  $\delta m/m \approx 10^{-9}$  for radionuclides [26] and as low as  $\delta m/m \approx 10^{-11}$  for stable nuclides [27]. PTMS is versatile and can be applied to isotopes of practically all elements ranging from short-lived ones to stable isotopes. The shortest-lived nuclide measured by PTMS to date is <sup>11</sup>Li [28]. In the following some of the basics for high-precision mass measurements in Penning traps are introduced. More details can be found in review articles and textbooks [3,29,30]. In a Penning trap an ion with mass m and charge q is confined by electromagnetic fields that lead to three independent harmonic eigenmotions of the trapped ion, in the ideal case of a single ion in a perfect quadrupolar electric field. The radial eigenmotions are related to the free cyclotron frequency  $v_c = qB/(2\pi m)$  [31] from which the mass *m* of a trapped ion is obtained using appropriate reference ions for calibration. A detailed description of the ion motion in Penning traps is given in the literature [32–35].

For a measurement of the cyclotron motion in the case of radionuclides the most common technique employed is the socalled time-of-flight ion-cyclotron-resonance (ToF-ICR) technique, a versatile and robust method that has been introduced about 25 years ago [36] based on the method by Gräff [37]. An ion, injected with magnetron and cyclotron radii as close to zero as possible, is excited by an external dipolar RF field at the magnetron frequency  $v_{-}$ . Afterwards the ion is excited by an quadrupolar RF field at the cyclotron frequency  $v_c$ . This leads to a coupling of the two initially independent radial motions and results in a conversion from a pure magnetron motion into a pure cyclotron motion. The conversion is controlled by the amplitude, the duration, and the phase of the external field. The parameters are usually chosen such that a full conversion is achieved after a given time. This process is accompanied by an increase of the (radial) kinetic energy of the ion. This is probed via the acceleration the ion experiences in the gradient field of the superconducting magnet after extraction from the trap, by measuring its flight time to a microchannel plate detector. The accelerating force depends on the magnetic moment of the ion and is proportional to its frequency. Scanning the frequency of this external field around the expected cyclotron frequency results in a resonance curve from which the cyclotron frequency is obtained. However, such a scan takes time and at least about 30 detected ions are required. In the case of very rare isotopes this results in very long measurements times. For example, a single resonance of <sup>256</sup>Lr recorded at SHIPTRAP took about four days [38]. This is very challenging since multiple parameters such as the trapping fields have to be kept constant. Single-ion mass measurement techniques are thus crucial to access more exotic nuclides with even lower yield.

The statistical mass uncertainty  $\delta m$  depends on a statistical factor based on the number of detected ions *N*, the line-width of the resonance curve, which is Fourier-limited and thus depends on the excitation time  $T_{rf}$ , and the cyclotron frequency according to [33,39]

$$\frac{\delta m}{m} \propto \frac{1}{v_c T_{rf} \sqrt{N}} = \frac{2\pi m}{q B T_{rf} \sqrt{N}}.$$
(1)

A higher precision can be achieved by using high magnetic field strength like in the case of LEBIT at the NSCL [12] and by using higher charge states as utilized for example at the TITAN facility at TRIUMF [13]. Further options are the use of a Ramsey-type excitation scheme [40-43] that improves the precision by a factor of about two to three and the use of an octupolar excitation instead of the conventional quadrupolar excitation. The latter increases the mass resolving power by at least one order of magnitude for the same measurement time [44,45]. Such a high mass resolving power has allowed resolving the isobar pair <sup>164</sup>Er-<sup>164</sup>Ho in a mixed sample for an excitation time of two seconds corresponding to  $m/\Delta m \approx 10^7$  [46]. However, a high stability of the confining fields and a careful ion preparation, in particular finding the proper excitation amplitudes, are crucial for reaching the highest resolving power [44,45]. Thus this technique has not been applied to online mass measurements yet.

An interesting alternative to the versatile ToF-ICR technique is the novel phase-imaging ion-cyclotron-resonance (PI-ICR) method. It has been recently introduced at SHIPTRAP and found to be about 25 times faster than ToF-ICR for comparable precision [22]. It provides forty times higher mass resolving power and features the possibility to resolve isomeric states of short-lived nuclides. In the PI-ICR technique the radial eigenfrequencies of the trapped ion are determined based on a phase measurement. To this end the radial ion motion is projected onto a position-sensitive ion detector after extraction from the trap (see Fig. 1). The phase of the ion motion is measured for a given trapping time that determines the number of revolutions.

From the phase difference for two different revolution times (after excitation) the frequency of a given eigenmotion is obtained according to the relation [22,47]

$$v = \frac{\phi + 2\pi n}{2\pi t},\tag{2}$$

where  $\phi$  is the phase difference of the ion motion under investigation, *t* is the phase accumulation time, and *n* is the number of full revolutions an ion performs in time *t*. One of the advantages using the PI-ICR technique is that the uncertainty  $\delta v$  depends on the ratio between the ion radius after excitation and the radial spread  $R/\Delta R$ [22,47] according to

$$\delta v \approx \frac{\Delta R}{\pi t R}.$$
(3)

The precision can be increased without increasing the measurement time by changing this ratio in a certain range so that high precision is reached even for short-lived nuclides (or in shorter measurement time). The maximum radius is ultimately limited



Fig. 1. Photo of the SHIPTRAP PI-ICR detector (Roentdek DLD 40).

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