ION TRAP MASS SPECTROMETRY OF RARE ISOTOPES*

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Electromagnetic ion traps have become essential instruments for the precise and accurate mass determination of rare isotopes. Penning trap mass spectrometers are now successfully operated at several radioactive beam facilities in the world. Accurate masses values obtained with these devices provide key data for the test of fundamental interactions, the modeling of the nuclear synthesis of the elements, and for improving our understanding of nuclear structure far from stability. This paper gives an overview of the basic techniques, the specific challenges, and the status of Penning trap mass spectrometry of rare isotopes.

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1. Introduction

The further development of mass measurement techniques for the study of nuclear binding far from the valley of stability remains important. The systematic exploration of masses allows us to directly observe nuclear structure effects like the location of shell and subshell closures, pairing, or the onset of deformation. Masses play an important role in the understanding of nuclear astrophysical processes [1]. Many important nuclei in these processes are still not accessible in the lab and mass prediction by models and mass formula have to be employed [2–4]. Only accurate experimental data far from stability can provide a stringent test of these theoretical predictions. For a general exploration of nuclear structure effects an accuracy of 100 keV is often sufficient while for the study of more subtle effects and for key nuclei in nuclear astrophysical processes an accuracy of the order of

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10 keV is often desirable. There are special cases where the mass measurement accuracy must go even further. This is for example the case for testing the standard model via a precise study of super-allowed β emitters [5, 6]. Mass measurements with an accuracy of 1 keV or less are typically required.

A variety of new techniques for the direct measurement of nuclear masses [7–10] have been developed during the past two decades. Those that are presently in use are either based on time-of-flight or frequency measurements. Frequency measurements are carried out with storage rings, transmission spectrometers, and Penning traps. The latter have proven to deliver unprecedented accuracy even for very exotic nuclides.

2. Rare isotope Penning trap mass spectrometry (RI-PTMS)

In a Penning trap a strong homogeneous magnetic field with strength B and an axial symmetric electric quadrupole potential are employed for the storage of a charged particle [11]. Electrode configurations like those shown in Fig. 1 are used to create the electric potential. The motion of an ion in a Penning trap is a superposition of three independent harmonic eigen motions. They are an axial motion parallel to the magnetic field lines and a radial motion. The radial motion itself is a superposition of a slow circular drift motion (magnetron motion) with frequency ν_- and a (reduced) cyclotron motion with the frequency ν_+. Several techniques have been developed for the detection of the frequencies of the eigen motions of the ions. In most RI-PTMS experiments a direct observation of the sum of magnetron and reduced cyclotron frequencies $\nu_+ + \nu_- = \nu_c$ is used [12, 13] to determine the cyclotron frequency

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

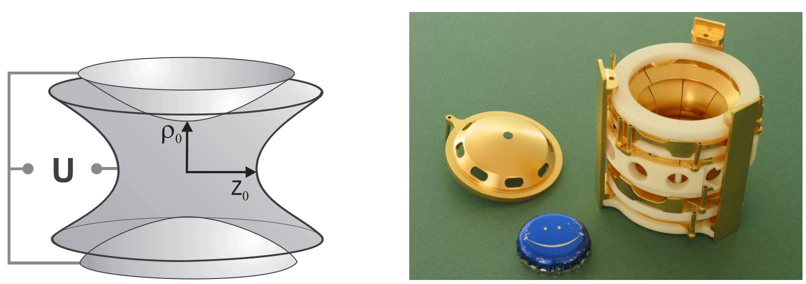

Fig. 1. Left: Basic electrode configuration of a Penning trap. Right: The electrodes of the Penning trap of the LEBIT facility at MSU. The ring electrode is segmented for the creation of azimuthal multipole RF fields for the excitation of the motion of the trapped ions.

of an ion with charge-to-mass ratio q/m. From this sum frequency the mass m of the ion can be obtained, if its charge state is known. The value of the magnetic field B required for the mass determination can be determined from the cyclotron frequency ν_c of an ion with well-known mass. The resolving power depends on ν_c and the time of observation T_{obs} of the ion motion

$$R = \frac{m}{\Delta m} \approx 1 \cdot \nu_c \cdot T_{\text{obs}} \,. \tag{2}$$

On one hand this means that the resolving power can be increased by enlarging the observation time of the ions. On the other hand it shows that the half-life of an nuclide poses a limit on the maximum resolving power that can be achieved. The statistical uncertainty $\delta m/m$ with which the cyclotron frequency can be determined is inverse proportional to both the resolving power R and to the square root of the number N_{ion} of detected ions. The fore factor depends to some extent on the detection scheme in the experiment but is typically close to unity resulting in

$$\left(\frac{\delta m}{m}\right)_{\text{stat}} \approx 1 \cdot R^{-1} \cdot N_{\text{ion}}^{-1/2} \,. \tag{3}$$

Using the above relations the accuracy and sensitivity of RI-PTMS can be evaluated for several scenarios. This is illustrated in Table I for nuclides with different mass and half-life. It is assumed that the maximum storage time is twice the half-life of the investigated nuclide. The magnetic field is taken to be 9.4 T, which corresponds to the field strength available in the LEBIT project discussed below. Case 1 (^{19}C) illustrates a measurement on a short-lived halo nucleus. With only two hundred detected ions a statistical uncertainty of a few keV can be achieved, sufficient to help understanding the "size" of the neutron halo. Cases 2 and 3 describe the situation of a high-accuracy mass measurement on a short-lived nuclide like the super-allowed Fermi-emitter ^{62}Ga. In order to achieve a statistical accuracy as required for a meaningful test of the Conserved-Vector-Current (CVC) hypothesis several ten thousand ions will have to be detected. This number can be drastically reduced if highly-charged ions are used. Case 4 describes the situation of a nuclide close to doubly-magic (^{132}Sn). The long half-life allows high resolving powers to be achieved, sufficient to resolve ground and long-lived isomeric states. The last case illustrates the situation of a mass measurement of a super-heavy nucleus. Of course, the total accuracy of the mass values has to include possible systematic errors. The design of present ion trap systems for nuclear mass measurements is such that systematic errors due to field imperfections and magnetic field instabilities are typically below $\delta m/m < 1 \times 10^{-8}$. Some care has to be taken to avoid systematic errors due to Coulomb interaction between ions of different mass stored simultaneously [14]. Such effects can be avoided if the measurements are performed with only a single ion trapped at a time.

The ability to resolve nuclear isomeric and ground states is an important feature of RI-PTMS in particular for ensuring an accurate determination of ground state masses. Nearly one third of the known nuclides have long-lived isomeric states with (in many cases unknown) excitation energies. Only in a few cases information about the production ratio exists which may vary drastically depending on the spins, the half-lives and on the parameters of rare nuclide production. Therefore, the resolution of nuclides in their ground or isomeric state is essential for an unambiguous determination of the mass of the nuclide in one or the other state (see for example [14–17]).

TABLE I

RI-PTMS on various rare nuclides in a 9.4 Tesla Penning trap (see text).

	$T_{1/2}$	$T_{\rm obs}$	$\nu_{\rm c}$ [MHz]	R [10^6]	$N_{\rm ion}$	$\delta m/m$	δm [keV]
^{19}C$^+$	46 ms	92 ms	7.6	0.7	200	1×10^{-7}	1.8
^{62}Ga$^+$	116 ms	232 ms	2.3	5.4	40 000	1×10^{-8}	0.5
^{62}Ga^{10+}	116 ms	232 ms	2.3	5.4	400	1×10^{-8}	0.5
^{131}Sn$^+$	56 s	10 s	1.1	11.0	100	1×10^{-8}	1.1
^{269}Hs$^+$	9 s	2 s	0.5	1.1	100	1×10^{-7}	23

3. Specific challenges in RI-PTMS

Penning traps have since long proven to be very accurate mass spectrometers. A large variety of mass measurements with an accuracy down to and even below 10^{-10} have been performed on stable, mostly light particles (see for example [18–22]. In most of these experiments the ions have been created inside the trap. For stable nuclides a high efficiency in ion production (and ion capture if performed at all) is not important and a fast measurement technique is not required. This is different in the case of RI-PTMS where specific challenges have to be met: The ions are delivered from external sources; the beam energy can range from several tens of keV to several GeV depending on the rare isotope production schemes; the most exotic species are only created in minute quantities; the half-life of the rare isotope to be investigated can be as short as a few tens of milliseconds. This means that very fast and efficient techniques for beam handling, cooling and trapping are required as well as fast measurement techniques are required.

3.1. Rare isotope beam production and preparation

An important parameter in RI-PTMS are the properties of the rare isotope beam which are strongly depending on the rare isotope production scheme. Rare isotope production via projectile fragmentation and fission of high energy primary beams ($> 50\,\mathrm{MeV}/u$) is a powerful approach since it is fast and allows isotopes of practically all elements lighter than Uranium to be produced. Fast-beam facilities are for example installed at NSCL at MSU, GSI in Darmstadt, and at RIKEN in Japan. Another example for in-flight separation at lower energies is the velocity filter SHIP at GSI where superheavy and other elements can be produced via fusion-evaporation reactions [23]. The ISOL (Isotope Separation On-Line) technique, which has a long tradition, delivers low-energy beams (a few tens of keV) of rare isotopes produced in thick targets bombarded with light ions or neutrons. Examples of facilities are ISOLDE at CERN, ISAC at TRIUMF and HRIBF at ORNL. A variation of the ISOL approach are ion-guide ISOL (IGISOL) systems [24].

In all production scenarios a major challenge is the apparent mismatch between the beam properties (*i.e.* high energy, large emittance) and the requirements of trap-type experiments (*i.e.* cold ions confined in small volumes). The basic principles adopted by all projects for the preparation of the rare isotope beams for their capture in ion traps are illustrated in Fig. 2. In the case of ISOL-type beams the ions are decelerated electrostatically to low energy before they enter a gas-filled radio-frequency ion trap [25]. Here the ions are accumulated and cooled, and finally released as short, cooled ion bunches. In the case of high-energy in-flight separated beams electrostatic deceleration is not possible. Instead, a slowing down and thermalization procedure is employed which is based on the usage of solid degraders and a stopping cell filled with helium at a pressure ranging from 0.1 to 1 bar [26–30].

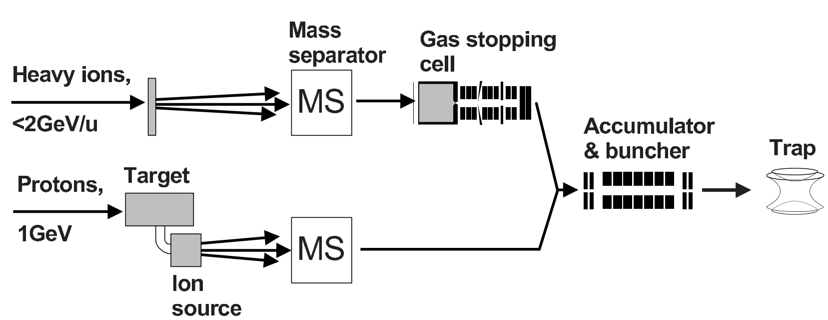

Fig. 2. Beam preparation for RI-PTMS in the cases of rare isotope production via the ISOL approach and by in-flight separation.

3.2. Precision mass measurements with RI-PTMS

A time-of-flight technique has turned out to be advantageous for the determination of the ion's cyclotron frequency ν_c of most rare isotope ions [31]. An ion is dynamically captured in the trap. A radio-frequency (rf) field is applied for a period T_{obs}. Subsequently, the ion is ejected out of the trap and its time of flight to a detector outside the strong trapping field is measured. The trap is refilled and this procedure is repeated for different rf frequencies ν_{rf}. Due to an adiabatic conversion of the ions' radial energy into axial energy, a reduction of time of flight is measured if $\nu_{rf} = \nu_c$. Hence, measuring the time of flight as a function of ν_{rf} yields a resonance curve from which ν_c can be determined (for examples see Fig. 4). For the efficient study of masses of many different rare isotopes this measurement technique has several advantages. Tuning of resonant circuits is not required allowing very fast switching from one ion species to another. Single-ion sensitivity can be achieved and a few tens or hundreds detected ions are often sufficient for a mass measurement. However, for rare isotopes with extremely low production rates like those of superheavy elements non-destructive techniques are mandatory; narrow-band image-current resonance detection techniques are presently under development [32].

4. PTMS at ISOL-type facilities

ISOLTRAP [31] at ISOLDE/CERN was the first system to demonstrate that Penning traps can be applied to short-lived radioactive ions. ISOLTRAP is a triple trap spectrometer. An RFQ trap ion beam cooler and buncher [33] stops the 60 keV ISOLDE beam and prepares it for efficient transfer into a buffer-gas filled Penning trap [34] which has the task of accumulating and purifying ions. This is achieved with a mass selective buffer gas cooling technique [12, 13, 35]. The ions are then transported to a 6 Tesla precision trap [36] for the determination of their cyclotron frequency. More than 300 masses have been investigated with ISOLTRAP with results important for example for the test of fundamental interactions [37–40], nuclear astrophysics [41], or nuclear structure studies [42–45]. JYFLTRAP [46], a Penning trap system for precision mass measurements and decay studies is installed at the IGISOL facility at the cyclotron laboratory in Jyväskylä. Similar to ISOLTRAP ions are delivered to an RFQ ion cooler and buncher and then transferred into a tandem 7 Tesla Penning trap system. A number of high precision mass measurements have been carried out, in particular on proton-rich isotopes along the $N = Z$ line [47] and on fission products [48, 49]. TITAN [50] at the high-power ISOL facility ISAC at TRIUMF in Vancouver is the youngest Penning trap mass spectrometer to come into operation. First measurements of singly-charged ions of neutron-rich Lithium isotopes have been reported recently [51]. In its final stage, TITAN will use highly charged ion for the mass measurements, which will be of advantage for the study of very short-lived isotopes with high accuracy (see Table I). The highly-charged ions will be obtained by coupling the spectrometer to a charge breeder based on an electron beam ion trap (EBIT).

5. PTMS with in-flight separated low-energy beams

Fusion-evaporation reactions at MeV/u energies offer cross-section advantages for the production of rare isotopes in certain regions of the nuclear chart. In order to use these isotopes for Penning trap mass spectrometry it is necessary to stop an thermalize them as discussed above. The Canadian Penning Trap CPT [52] was the first ion trap project that made use of thermalized radioactive products from such low-energy nuclear reactions for mass measurements. After in-flight separation in an Enge-type separator the reaction products are stopped in a low-pressure (100 mbar He) gas cell from where they are extracted as a low-energy ion beam. Following a scheme similar to ISOLTRAP the ions pass an isobar separator before being captured in the measurement Penning trap. On-line mass measurements with CPT have concentrated on proton-rich isotopes [53–55]. Furthermore, measurements were made on fission products obtained from a fission source [56]. SHIPTRAP [57] is installed at the velocity filter SHIP at GSI. The ultimate goal of this project is the precise study of properties of transactinides and superheavies. The SHIP reaction products (typical energy several MeV/u) are converted into a low-energy high-quality pulsed beam using a gas cell coupled to an RFQ ions guide and buncher system. The actual Penning trap spectrometer is a sister system to that at JYFLTRAP and uses two traps in a single magnet system, one for isobar separation, the other for the mass measurements. SHIPTRAP has been used very successfully for measurements of neutron-deficient isotopes produced by fusion evaporation reactions [32,58] and first experiments on transactinide isotopes are on their way [59]. A Penning trap mass spectrometer similar to SHIPTRAP is presently under construction at MLL in Garching. MLLTRAP [60] is eventually planned to be installed at the SPIRAL-2 facility at GANIL.

6. PTMS with high-energy rare isotope beams — LEBIT at MSU

The Low-Energy Beam and Ion Trap facility LEBIT at the NSCL at MSU was the first to demonstrate that rare isotopes produced by fast-beam fragmentation can be slowed down and prepared for precision experiments with low-energy beams.

Fig. 3 presents a schematic overview of the LEBIT facility. The main components of LEBIT are a gas stopping station for the high-energy fragmentation beams, a cryogenic RFQ trap ion cooler and buncher, and a 9.4 Tesla Penning trap mass spectrometer. Rare isotopes with an energy in the order of 100 MeV/u are produced at the NSCL's Coupled Cyclotron Facility, separated in-flight by the A1900 fragment separator [61], and delivered to the LEBIT facility. The particular difficulty to be overcome in thermalizing such projectile fragments is the high energy of the order of 100 MeV/u and the large energy spread. Compared to beam stopping at lower energies this requires either a very large gas stopping cell or a very high gas pressure. The latter option is presently pursued at LEBIT.

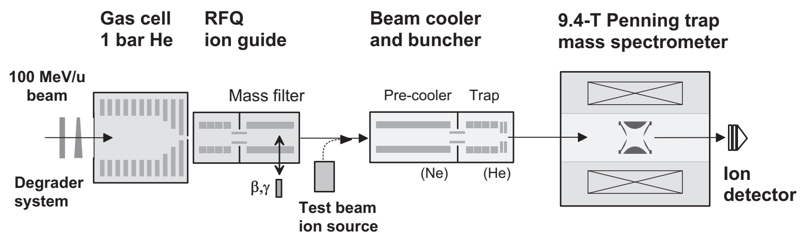

Fig. 3. Schematics of the LEBIT facility at the NSCL. The main components of LEBIT are a gas stopping station for the high-energy fragmentation beams, a cryogenic RFQ trap ion cooler and buncher, and a 9.4 Tesla Penning trap mass spectrometer.

The rare isotope ions delivered by the A1900 fragment separator pass degraders for momentum compression and energy reduction before they enter the gas cell through a thin beryllium window. The 50 cm long gas cell [30] is operated with high-purity helium gas at pressures of up to 1 bar. After stopping and thermalization, the ions are transported and extracted by a combination of DC electric fields and gas flow. An RFQ ion guide transports the ions into high vacuum. The last section of the ion guide is operated as a mass filter to select ions according to their mass-to-charge ratio [62]. The ions are then injected into a buffer-gas filled RFQ cooler and buncher [63]. The RFQ cooler and buncher has a novel electrode design and is cryogenically cooled to improve the cleanliness and to reduce the emittance of the extracted ion beam. Ion pulses with about 100 ns length are extracted for efficient capture in the LEBIT Penning trap [64]. The Penning trap (Fig. 1) is located inside an actively shielded superconducting solenoid. The LEBIT Penning trap mass spectrometer has been optimized for measurements of short-lived rare isotopes with half-lives on the order of 10 ms and low production rates of less than 100 ions/s. Features which contribute to this end are the employment of a 9.4 T magnetic field, a high-precision electrode system, the fast preparation of ions with a "Lorentz steerer" [65], and an active stabilization of the magnetic field. The 9.4 T field provides resolving powers of about 10^6 even for short ion trapping times and hence, for short-lived nuclei. In comparison to common 6–7 T systems the use of a 9.4 T magnet system allows the same mass uncertainty to be reached in about half the measurement time.

With LEBIT mass measurements of about thirty rare isotopes on both sides of the valley of β-stability have been performed since 2005. Mass uncertainties below 10^{-8} have been achieved and mass measurements of short-lived isotopes with half-lives below 100 ms have been demonstrated.

The first successful physics experiments concentrated on the neutron-deficient, short-lived calcium isotopes ^{37}Ca ($T_{1/2}=180$ ms) and ^{38}Ca ($T_{1/2}=$ 440 ms) [66]. The mass value of the super-allowed beta emitter ^{38}Ca was measured with an uncertainty of $\delta m=280$ eV ($\delta m/m=8\times10^{-9}$) [67]. The achieved accuracy made this isotope a new candidate for the test of the Conserved-Vector-Current (CVC) hypothesis, which is presently confirmed at a level of 1.3×10^{-4} [6]. The mass value of ^{37}Ca, now known with an uncertainty of less than 2 keV, could contribute to a test of the isobaric multiplet mass equation (IMME) [68] in the $A=37, T=3/2, J^{\pi}=3/2^{+}$ quartet. The uncertainty of the isobaric analog state of 37Ar is presently too large for such a test, but assuming its validity IMME can be employed to make a 6-fold improved prediction of the excited state energy of ^{37}Ar.

Mass measurements of neutron-deficient rare isotopes have been performed around $N=Z=33$–35. These masses are in particular relevant for the astrophysical rp process [69]. With LEBIT the masses of ^{63,64}Ga, $^{64-66}$Ge, $^{66-68}$As, $^{68-70}$Se, and ^{70m,71}Br were measured, most of them with uncertainties well below 10 keV. ^{66}As with a half-life of 96 ms is one of the shortest-lived rare isotopes ever measured in a Penning trap. The mass of the drip line nucleus ^{70}Br was determined experimentally for the first time by combining the measured mass of the isomer ^{70}Br and the well-known excitation energy. The mass values for the important waiting point nuclei ^{64}Ge and ^{68}Se were determined with uncertainties of 3.8 keV and 580 eV, respectively [70, 71]. With the improved mass values reaction network calculation were performed and results obtained that are important for a better understanding of the rp process. For example, it was found that ^{64}Ge constitutes less of a waiting point of the rp process [70].

Extending the applicability of Penning trap mass measurements into regions previously not accessible is important. Masses of neutron-rich sulfur isotopes up to ^{44}S ($N=28$) and of neutron-rich Fe and Co isotopes near $N=40$ have been measured recently. Isotopes of these elements are difficult or impossible to obtain at ISOL facilities and most mass measurements on light neutron-rich rare isotopes have so far been carried out via time-of-flight measurements, reaching far from stability but providing limited accuracy. Fig. 4 shows a cyclotron resonance for ^{44}S, which has a half-life of only $T_{1/2} = 123$ ms. These and future high-accuracy mass measurements in these regions will help to shed light on the evolution of shell structure in these regions, where despite substantial experimental and theoretical effort the situation remains unclear. For example, a weak two proton knockout cross section for ^{44}S had been interpreted as a strong indication of a magic spherical nucleus ^{42}Si [72] with a large $Z = 14$ shell gap while recent results from γ-spectroscopy [73] indicate the disappearance of the $Z = 14$ and $N = 28$ spherical shell closures.

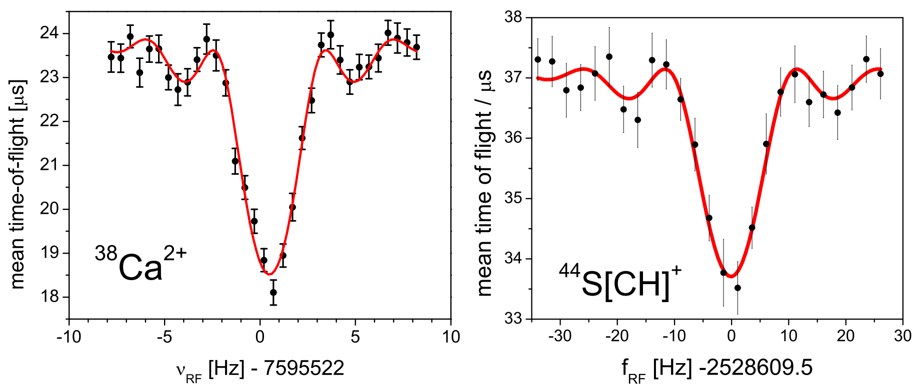

Fig. 4. Cyclotron resonance curves for $^{38}Ca^{2+}$ and for $^{44}S[CH]^{+}$ obtained with the LEBIT Penning trap.

7. Conclusion

Penning trap spectrometry for high-accuracy mass measurements of short-lived rare isotopes has been very successful. New developments and efforts in the manipulation of rare isotope beams by gas-stopping and trapping techniques have extended the applicability of Penning trap mass spectrometry from ISOL-type beams to fast rare isotope beams. The results will continue to contribute to a better understanding of nuclear structure, nuclear astrophysical processes, and fundamental interactions.

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