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IN TRAPS AND STORAGE RINGS**

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Mass Measurements of Short-Lived Isotopes in Traps and Storage Rings

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Abstract. High-accuracy mass measurements can be performed on short-lived radio-nuclides with half-lives $T_{1/2} \geq 1\text{ms}$ by storing them in ion traps or storage rings. The masses of about 80 isotopes have been determined with a resolving power of typically one million and an accuracy of $\delta m/m = 10^{-7}$ with the tandem Penning trap mass spectrometer ISOLTRAP installed at the on-line mass separator ISOLDE/CERN. Schottky and time-of-flight mass measurements will soon start at the storage ring ESR at GSI/Darmstadt on highly charged ions delivered at relativistic energies from the fragment separator. Further developments and projects are discussed.

INTRODUCTION

The mass of a quantum mechanical system, i.e. the sum of the rest masses of its constituents minus the binding energy is it's most fundamental property since all forces acting in such a system are involved in it. In nuclear physics, precise masses are especially important since an exact theory of strong interaction is still missing. A large variety of nuclear models has been developed in the course of the last decades. The comparison of the predictions of state of the art models with experimental masses has led to the development of collective models like the liquid-drop model or single-particle models like the (spherical) shell model and the (deformed) Nilsson model. More accurate mass values as well as the extension of the data set to those of isotopes far from the valley of nuclear stability have stimulated and still stimulate an on-going refinement of those models.

Today the masses of around 2000 isotopes are known: stable isotopes typically with an accuracy of about $\delta m/m = 10^{-8}$, deteriorating to about 10^{-7} for long-lived isotopes and to about 10^{-6} for short-lived ones. This decrease in accuracy is due to the fact that almost all the masses of radioisotopes are determined by Q-value measurements in nuclear decays or reactions.

Hence, the mass of an isotope far from stability is linked by many mass differences to the well known mass of a stable isotope. As a consequence, the errors in the mass differences accumulate. In addition, Q-value measurements require the knowledge of the initial and final nuclear state, quite often a difficult task in case of complex or unknown nuclear level schemes and a source of many wrong entries in the tables of nuclear masses.

About 4500 - 6000 nuclei are assumed to exist as bound systems between the hydrogen atom and the very heavy elements (with $Z=109$, meitnerium, being the last one detected) and the neutron and proton drip lines (where neutrons and protons become unbound, respectively). The masses of neutron-rich isotopes far from the valley of nuclear stability are especially important, since these nuclei were involved in the nucleosynthesis (r-process) of the elements and the one-neutron separation energy (S_{1n}) of these isotopes enters exponentially in the rate equations modelling the element cooking process. Because the r-process path runs so far away from known isotopes, nuclear models or mass formulae have to be used to calculate S_{1n} . The accuracy presently achieved for such models is $\delta m/m \simeq 10^{-5}$, corresponding to about 1 MeV or 10% of S_{1n} for medium heavy nuclei. In order to improve the predictive power of the models, high-accuracy mass data are required of radionuclides as far as possible from the valley of nuclear stability.

THE GOAL

Short-lived isotopes are produced by nuclear reactions of accelerated particles or neutrons with target atoms. The mass of the resulting radionuclide, quite often extremely rare, should be determined with

- high resolving power (in order to resolve the ground-state mass from that of the isomer),
- high accuracy ($\delta m/m \leq 10^{-6}$),
- in a short time period (at least comparable to the nuclear half-life $T_{1/2}$),
- with high efficiency and,
- in a direct or even absolute way (relative to the mass of a freely chosen reference isotope or relative to the mass of ^{12}C establishing the atomic mass unit on the microscopic scale).

In the following, two new approaches to accurate mass measurements of short-lived isotopes are described which both make use of storage techniques, i.e. the observation of the cyclotron frequency of ions confined in Penning traps or storage rings (Fig. 1).

MASS MEASUREMENT OF SHORT-LIVED ISOTOPES IN

ION TRAPS (ISOLTRAP / ISOLDE, CERN)

STORAGE RINGS (ESR / GSI, Darmstadt)

FIGURE 1. Principle of mass determinations of short-lived isotopes in a Penning trap and storage ring.

PENNING TRAP MASS SPECTROMETRY OF SHORT-LIVED ISOTOPES

Penning traps have been proven to be very effective instruments for high-accuracy mass determinations via the relation

$$\omega_c = \frac{qe}{m} B, \quad (1)$$

where ω_c is the cyclotron frequency, q the charge state and m the mass of the stored ion, e the elementary charge and B the magnetic field. With these devices a number of stable or long-lived particles, for example the electron and positron, electron and proton, proton and antiproton, helium-3 and tritium, and light ions and molecules have been determined to very high accuracy [1-6]. However, the current methods used in these experiments are not readily adapted to mass measurements of radioactive isotopes. In the following the special requirements which are imposed in the case of mass measurements of short-lived isotopes will therefore be discussed.

Resolving Power

Due to the superposition of an electrostatic quadrupole field and a homogeneous magnetic field the ion motion in a Penning trap is not a pure cyclotron motion with frequency ω_c according to (1) but consists of three independent harmonic eigen-motions [7]: a reduced cyclotron motion with frequency ω_+ modified from the pure cyclotron motion by the presence of the electric quadrupole field, an oscillation along the magnetic field lines with frequency ω_z , and a slow drift around the trap axis, called magnetron motion, with frequency ω_- . Generally,

$$\omega_+ \gg \omega_z \gg \omega_- . \quad (2)$$

The energy of an ion is therefore given as the sum of the energies of three harmonic oscillators by

$$E = \hbar\omega_+(n_+ + 1/2) + \hbar\omega_z(n_z + 1/2) - \hbar\omega_-(n_- + 1/2) . \quad (3)$$

Note that the magnetron motion is unstable. The eigenfrequencies ω_+ , ω_- , and ω_z can be determined by driving the corresponding motions with oscillating dipole fields ($n \rightarrow n + 1$) and detecting the change of the motional amplitudes $\varrho \sim \sqrt{n+1}$. However, the most direct approach is the direct measurement of the sum frequency [8]

$$\omega_c = \omega_+ + \omega_- . \quad (4)$$

The motion of a trapped ion can be directly excited at this frequency by an azimuthally quadrupolar RF-field [8] which leads to a simultaneous change in quantum numbers by

$$n_+ \rightarrow n_+ + 1; \quad n_- \rightarrow n_- - 1. \quad (5)$$

This excitation couples the magnetron and reduced cyclotron motion so that a periodic conversion from one motion into the other can be achieved. After a particular excitation time T_{RF} , an initially pure magnetron motion is transformed into a pure cyclotron motion which, according to (3), results at the same time in an increase of radial energy. The width of the cyclotron resonance curve $\Delta\nu_c$ is determined by the Fourier limit of the driving RF-field switched on for a time T_{RF} . From [8] the relationship is

$$\Delta\nu_c(\text{FWHM}) \simeq 0.8/T_{RF}. \quad (6)$$

FIGURE 2. Cyclotron resonances of both the isomeric and ground state of ^{84}Rb . The resolving power is about $R = 800000$. The mass difference measured by weighing corresponds to $\Delta E_{mass} = 464(7)\text{keV}$ in full agreement with the value of $\Delta E_\gamma = 463.7(1.0)\text{keV}$ as determined by γ -spectroscopy. Due to the limited statistics obtained in the on-line run, the data points were fitted by a Gaussian (full line) instead of the correct line profile (see Figure 3).

For example, a singly charged ion of mass number $A = 100$ in a field $B = 6\text{T}$ has $\nu_c \simeq 1\text{MHz}$. Exciting its motion for $T_{RF} = 800\text{ms}$ yields a line width of $\Delta\nu_c \simeq 1\text{Hz}$, corresponding to a resolving power of $R = 10^6$. This is more than sufficient to discriminate between different isotopes, more than adequate to distinguish between isobars further away from the valley of nuclear stability, and even suited to resolve ground and isomeric states of an $A \approx 100$ nucleus

with an excitation energy of $\Delta E \geq 100\text{keV}$. As an example, Fig. 2 shows the cyclotron resonance of both the ground state ($T_{1/2} = 32.9\text{d}$) and the isomeric state ($T_{1/2} = 20.5\text{min}$) of ^{84}Rb [9]. Note, that nuclei either in the ground or in the isomeric state can be eliminated by RF-excitation at the appropriate frequency ω_+ . In such a way an isomer separator can be realized resulting, for example, in an inverted population of a nuclear system.

Accuracy and Calibration

The line profile of the cyclotron resonance curve is the Fourier transform of the rectangular RF-pulse of duration T_{RF} modified by the periodic conversion of magnetron into cyclotron motion and by the transformation of radial energy into a change of time of flight from the trap to the detector when the ions are ejected (see below). As can be seen from Fig. 3, the resonance profile is very well understood. Hence, statistical errors well below $\delta\nu_c/\nu_c = 10^{-8}$ can be obtained. In order to determine the mass of the investigated isotope via (1), the magnetic field has to be known. This can be achieved by loading a stable isotope into the trap the mass of which is precisely known. In the future, it is planned to use ^{12}C cluster ions for calibrating the magnetic field which would enable to perform for the first time absolute mass measurements of short-lived isotopes.

FIGURE 3. Cyclotron resonance for ^{85}Rb ($T_{RF} = 3.6\text{s}$, $R = 4 \cdot 10^6$). The solid line represents a fit by the theoretical line shape.

THE ISOLTRAP TANDEM PENNING TRAP MASS SPECTROMETER

Figure 4 shows the experimental set-up of the ISOLTRAP spectrometer [10] at the on-line mass separator ISOLDE, a powerful facility for the production of radioactive isotopes. ISOLTRAP consists of two main parts, the lower Penning trap in an electro magnet acting as an ion beam accumulator, cooler and buncher (trap 1) and the upper high-precision trap (trap 2) in a superconducting coil in which the mass determination takes place.

FIGURE 4. The ISOLTRAP tandem Penning trap mass spectrometer installed at the on-line isotope separator ISOLDE at CERN/Geneva. The mass separated ion beam from ISOLDE enters from the bottom.

The continuous 60keV ISOLDE ion beam is collected on a rhenium foil mounted in the lower endcap electrode of trap 1. After a certain collection time the foil is rotated by 180° and heated so as to surface-ionize the accumulated material. The ions released from the foil are mass selectively cooled, and centered in trap 1 by He buffer gas collisions ($p_{He} = 10^{-3} - 10^{-4}$ mbar) which damp the axial as well as the cyclotron amplitudes. The increase in magnetron radius is counteracted by RF-excitation of the ω_c -resonance of the ion under investigation (see Eq. (5)). A fraction of about 10^{-3} to 10^{-4} of the ions accumulated on the foil is centered by this new cooling technique [11]. After cooling to room temperature, the ion cloud is ejected, accelerated to 1keV, retarded again in front of trap 2 and captured there in flight. The transfer between both traps can be realized with an efficiency close to 100% [12].

The high-precision trap is placed in the 5.7T field of the superconducting magnet. Deviations from the ideal quadrupole field are compensated by correction electrodes installed between the ring electrode and the end electrodes and at the holes in the endcaps required for injection and ejection of ions. As in trap 1 the ring electrode is split into four quadrants to create the azimuthal quadrupole field for ω_c -excitation.

A destructive technique is used to detect the increase in radial energy (Eqs. (3) and (5)) as a function of the applied RF-frequency [13]. After excitation for a period T_{RF} , the ions are gently pulled from the trap as short pulses, leaving the homogeneous part of the magnetic field and drifting along the magnetic field lines to an ion detector placed in the weak fringe field of the magnet. As they drift, the radial energy gained in resonance by the RF-excitation is converted into longitudinal energy in the inhomogeneous part of the magnetic field. Therefore, the measurement of the drift time as a function of the applied radio frequency yields a resonance curve with a minimum at $\omega_c = \omega_+ + \omega_-$ corresponding to a maximum in radial energy.

RESULTS

Up to the present, cyclotron resonances have been observed for about 80 different isotopes and isomers (Table 1). Three of these isotopes (^{39}K , ^{85}Rb , and ^{133}Cs) were used as reference isotopes. Results are published in Refs. [10,14-17].

Typically, the statistical accuracy in on-line runs is of the order of some parts in 10^8 . A systematic error of $1 \cdot 10^{-7}$ is added which takes care of possible calibration errors (in first order proportional to the mass difference of the isotope under investigation and the reference isotope), jumps in the magnetic field, and the accuracy with which the various parameters can be controlled under the condition of limited beam time.

One remarkable feature of the ISOLTRAP mass data is that the size of the error is almost constant being in a first approximation only proportional to the mass of the isotope under investigation (dashed lines in Fig. 5). The excellent agreement with earlier data [18] of superior accuracy in the mass region $84 \leq A \leq 88$ demonstrates the reliability of the trap technique. For isotopes far away from stability, Fig. 5 shows impressively the superiority of direct mass measurements over measurements of mass differences.

TABLE 1. Isotopes for which cyclotron resonances have been determined by use of the ISOLTRAP mass spectrometer.

Element	Mass Number
K	39
Rb	75-89, 90m, 91 - 94
Sr	78 - 83, 91 - 95
Cs	118 - 121, 122+122m, 122m, 123 - 140, 142
Ba	124, 126, 128, 138 - 144
Fr	209 - 212, 221, 222
Ra	226, 230

FIGURE 5. Rubidium masses as a function of mass number. Left: the difference is plotted between the Penning trap data (zero line with experimental errors given by the dashed line) and a mass adjustment of all masses known up to now except the trap data (squares). Right: difference between the Penning trap data (zero line) and a new adjustment of all known mass data.

FURTHER DEVELOPMENTS

Presently, the lower trap (Fig. 4) is being replaced by a cylindrical trap in a superconducting magnet. An increase of the overall efficiency by about two orders of magnitude to 10^{-2} is expected. In addition, the high resolving power of this trap of $R = 10^5$ under cooling conditions will enable to eliminate already in trap 1 contaminations like isobars before the ion cloud is transferred to the precision trap. Extension to isotopes of those elements not surface ionizable will be possible by laser desorption of the accumulated sample and resonance ionization. Incorporated with this system will be a laser desorption source for the creation of ionized carbon clusters over a wide mass range. These clusters will be used to calibrate the magnetic field for absolute mass measurements.

A further approach is to extend the tandem configuration to a triple trap system by addition of a Paul trap which collects the 60keV ISOLDE ion beam in-flight and cools the ions by buffer gas collisions [19]. Such a system is being developed at McGill, Montreal. This trap will have a very large phase space volume for ion collection allowing bunching efficiencies considerable in excess of $2 \cdot 10^{-3}$ obtained in a first test by use of a prototype [20].

THE EXPERIMENTAL STORAGE RING (ESR) AT GSI AS A MASS SPECTROMETER

Recently, the accelerator complex at GSI/Darmstadt has been extended by the heavy ion synchrotron SIS to a relativistic heavy-ion facility delivering ion beams up to uranium with energies in excess of 1GeV/u. Radioactive beams might be produced by use of the fragment separator FRS and injected into the storage ring ESR (Fig. 6).

Except for its size (e.g., circumference = 108m), such a storage ring has features very similar to those of an ion trap (Fig. 1). Cooling of the stored ions (to low momentum spread but not to rest like in traps!) is achieved by Coulomb interactions with “cold” electrons. These electrons are produced from a hot cathode in the electron cooler (Fig. 6) and accelerated to 170keV for 300MeV/u uranium ions. Since the energy spread ($\Delta E \approx 0.1\text{eV}$) is a constant of motion, the velocity spread of the electrons in longitudinal direction is reduced when thermal even at the velocity is increased to $\beta = v/c \approx 0.65$. For bare uranium ions at 300MeV/u, with an electron beam current of 200mA, and for an interaction region of 2m length, cooling times of the order of $\tau_{cool}(\text{U}^{92+}) = 0.1\text{ s}$ and an momentum spread of the stored ions of $\Delta p/p \leq 10^{-6}$ were obtained. Due to the small emittance of the cooled ion beam and the ultra-high vacuum ($\leq 10^{-10}\text{mbar}$), the storage time τ_{store} is as long as 1h for U^{92+} .

FIGURE 6. Experimental Storage Ring (ESR) at GSI/Darmstadt.

Since the velocity of the cooled ion beam is equal to the velocity of the electrons in the cooler which can easily be kept constant, a measurement of the ratio of the revolution frequencies of two ion species ($\nu_R = 1.7\text{MHz}$ for $300\text{MeV/u } U^{92+}$) can be used for the determination of the mass ratio of those species very similar to the Penning trap technique.

Mass Determination via the Schottky Signal

Pick-up electrodes mounted in the beam tube of the ESR are used to observe the image currents induced by the circulating beam particles and, after Fourier transformation from the time to the frequency domain, to determine the momentum spread of the stored ions. This tool for diagnosis, the so-called Schottky signal, can also be used to measure the revolution frequency with high precision [21]. In a pilot experiment [22], the mass ratio of the mass doublet $^{18}\text{F}^{9+}/^{20}\text{Ne}^{10+}$ was determined to agree with the known value within $3 \cdot 10^{-7}$ at a level of an accuracy of $4 \cdot 10^{-6}$.

Figure 7 shows the potential of this technique. In the course of the (successful) search for the bound β -decay of $^{163}\text{Dy}^{66+}$, an argon target in the ESR was used for detection of the β -decay daughter nucleus ^{163}Ho [23]. By nuclear (p;xp,yn)-reactions, neighbouring isotopes are produced which show up as extremely narrow peaks in the Schottky spectrum (Fig. 7). The widths of these signals correspond to a momentum spread of $\Delta p/p \approx 10^{-6}$.

FIGURE 7. Schottky spectrum of stored $^{163}\text{Dy}^{66+}$ and the signals of $^{163}\text{Ho}^{67}$ (bound β -decay) and other nuclides produced by nuclear reactions with an argon target.

Hence, a precision in frequency ratios of better than $\delta\nu_R/\nu_R = 10^{-6}$ is obtained [24]. How this transforms into accuracy of mass ratios is presently under investigation.

It should be noted that mass measurements at storage rings provide the mass of relativistic, highly charged or even fully stripped ions but not the mass of the neutral atom. The measured mass has to be corrected for relativistic effects, the mass of the stripped electrons and their binding energies. This is of no concern in the case of the anticipated accuracy of $\delta m/m \leq 10^{-6}$. On the contrary, measuring the revolution frequency of an ion in different charge states presents an excellent tool to search for non-linearities in the mass-frequency relation.

Isotopes with half-lives down to about $T_{1/2} \approx 1\text{ s}$ can be investigated with the Schottky technique. This is due to the time required to prepare a cooled ion beam after injection from the FRS. In the case of highly charged, heavy ions the detection limit is of the order of 1 to 10 ions of the same isotope and charge state stored at a time in the ESR.

Mass Determination via Time-of-Flight Measurement

Another approach to mass determinations in the ESR is the measurement of the time for one revolution in the ring [25,26]. In this case, the ESR is operated in an isochronous mode and the measurements are performed on "uncooled" ions immediately after injection from the FRS. Mass measurements of single ions with half lives of $T \geq 1\text{ ms}$ are planned.

The time-of-flight detector is similar to the one used in the TOFI-experiment at Los Alamos [27]. When the stored ion is transversing a thin carbon foil, secondary electrons are ejected. They are detected by a multichannel plate detector with a time resolution of $\delta t \approx 150\text{ ps}$. This corresponds to a time resolution per turn of $3 \cdot 10^{-4}$ for a typical revolution time of 500 ns. Since up to 100 revolutions should be observable per injected ion, the expected resolving power is of the order of $t/\Delta t = 3 \cdot 10^5$.

Still it has to be shown how to inject into a storage ring operated in the isochronous mode and how many turns an ion will survive. First mass measurements are planned for the second half of 1995.

Mass Measurements à la the RF Smith Spectrometer

There might be still other possibilities to perform mass measurements in a storage ring. Consider two resonators producing an electric field transversal ("stochastic") or longitudinal (RF-Smith) to the path of the ion in the storage ring. If those resonators are placed at exactly two opposite positions in the ring

and are out of phase by exactly 180° after a half turn of the ion in the storage ring, the transversal (longitudinal) energy picked up in the first resonator will exactly be compensated in the second one as long as the resonators are operated at the revolution frequency. Off resonance, the ion will pick up transversal (longitudinal) energy and get finally lost.

An ion stored in the ring will perform many turns in contrast to the two revolutions in the RF-Smith spectrometer. This will increase the resolving power. A further increase will be obtained by operating the resonators at higher harmonics just as in the Smith spectrometer.

Quantitative calculations have still to be performed but the proposed technique might have advantages in respect to resolution because it avoids the timing detector and possible changes in the charge state of the ion traversing the foil.

CONCLUSIONS

Powerful techniques for mass measurements of short-lived isotopes have been developed. A first Penning trap mass spectrometer (ISOLTRAP) is operational at the on-line mass separator ISOLDE/CERN which has a resolving power of about one million and an accuracy of $\delta m/m = 10^{-7}$. The half life limit at present is about $T_{1/2} = 1$ s. A similar spectrometer is under construction by a Chalk-River-Mannitoba-Montreal collaboration [28]. A RF-Smith spectrometer is presently prepared by an Orsay group for mass measurements on isotopes with $T_{1/2} < 1$ s [29]. Soon, mass measurements will also start at the storage ring ESR at GSI on highly charged ions. First, the Schottky technique will be applied for mass measurements of isotopes with $T_{1/2} \geq 1$ s. Later the time-of-flight method will be used for isotopes with half-lives in the region of less than one second down to 1 ms. At Stockholm, the SMILETRAP [30] is coupled to the electron beam ion source CRYISIS of the storage ring CRYRING. Here, mass measurements of stable, highly charged ions up to the completely stripped nucleus are measured with an accuracy of some parts in 10^9 .

Obviously, storing techniques have led to a breakthrough in mass spectrometry.

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