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Recent high-precision mass measurements with the Penning trap spectrometer ISOLTRAP

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Abstract. The Penning trap mass spectrometer ISOLTRAP has to date been used for the determination of close to 300 masses of radionuclides. A relative mass uncertainty of 10^{-8} can now be reached. Recent highlights were measurements of *rp*-process nuclides as for instance $^{72-74}$Kr or superallowed β emitters like ^{22}Mg, ^{74}Rb and ^{34}Ar. The heaviest nuclides measured so far with ISOLTRAP are neutron-rich radium and francium isotopes. An overview of ISOLTRAP mass measurements and details about the recent experiment on $^{229-232}$Ra and ^{230}Fr are presented.

PACS. 21.10.Dr Binding energies and masses – 82.80.Qx Ion cyclotron resonance mass spectrometry

1 Introduction

High-precision atomic mass measurements are important for many areas of science. For nuclear physics in general it is presently sufficient to determine the mass of a large number of nuclei with a relative uncertainty of roughly 10^{-6} since a few 100 keV is the current uncertainty level of global theoretical models [1]. Nevertheless, there are many cases where a lower experimental uncertainty and also a high resolving power is asked for. As an example, low-lying isomeric states may spoil the measurements and hence need to be resolved [2]. Furthermore, it is not always possible to assign the levels unambiguously from decay spectroscopy alone [3]. In such cases, high-resolution mass spectroscopy as performed with ISOLTRAP can be used to clarify and to discover yet unmeasured long-lived isomers [4].

Nuclear reaction rates, which scale with the Q-value, the mass difference between initial and final state, are important input parameters for nuclear astrophysics calculations. For two processes of nucleosynthesis, the *r*- and the *rp*-process there are especially important nuclei, so-called waiting points, that determine the path as well as the timescale of the process. In the neighborhood of these nuclei mass measurements are needed with a relative uncertainty of about 10^{-7} [5,6,7,8].

Beta-decaying nuclei can be used as a laboratory to investigate the weak interaction and the mass is a very important parameter in this context. According to the conserved-vector-current (CVC) hypothesis the weak force should not be affected by the strong force in the nuclear environment. A test is the comparison of superallowed $0^+ \rightarrow 0^+$ decays. After application of some theoretical corrections the comparative half-life Ft of these decays should be constant. Besides the decay half-life and branching ratio, the decay energy or Q-value is one of the experimentally accessible input parameters. However, since the statistical rate function f depends on Q^5 the required relative mass uncertainty is 10^{-8} and smaller [9,10,11,12].

During the last years the precision of ISOLTRAP mass measurements and the applicability to very short-lived and very rare nuclei has been improved considerably. It is now possible to reach a relative mass uncertainty better than 10^{-8} and to investigate nuclei that are produced at a rate of only 100 ions per second. The half-life limit for a nuclide subjected to precise mass measurements is well below 100 ms [11]. This gives new insights into nuclear

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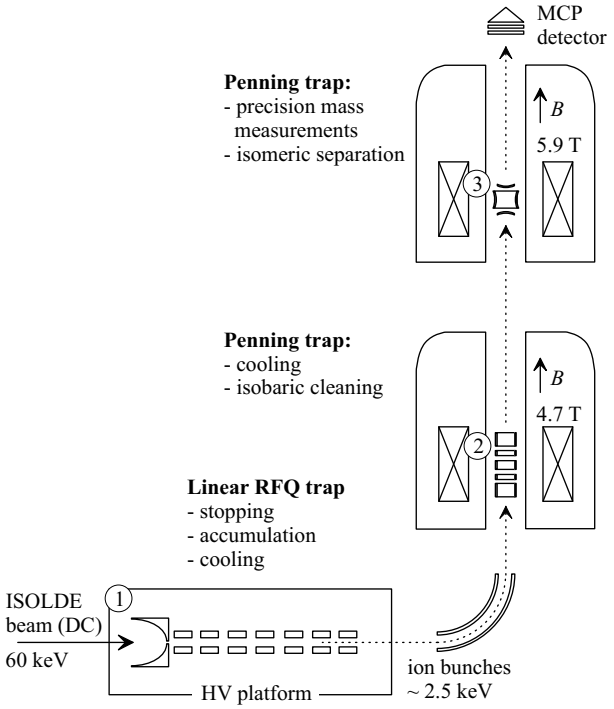

Fig. 1. Experimental setup of the ISOLTRAP Penning trap mass spectrometer. The three main parts are: 1) a gas-filled linear radio-frequency quadrupole (RFQ) trap for retardation of ions, accumulation, cooling and bunched ejection at low energy, 2) a gas-filled cylindrical Penning trap for further cooling and isobaric separation, and 3) an ultra-high-vacuum hyperboloidal Penning trap for the mass measurement. For this, the cyclotron frequency is determined by a measurement of the time of flight of the ions ejected out of the Penning trap to a micro-channel-plate (MCP) detector.

physics with a precision that has before only been possible for stable nuclei. Additionally, the very high resolving power of up to 10 million gives access to further interesting questions of physics [13,14,15].

2 The ISOLTRAP mass spectrometer

ISOLTRAP is a triple-trap mass-spectrometer setup at the on-line mass separator ISOLDE [16]. Radioactive nuclides are produced by bombarding a thick target with 1 or 1.4 GeV proton pulses with an average intensity of $2\,\mu$A. The produced atoms diffuse out of the target and are ionized either by a plasma discharge, surface ionization, or resonant laser ionization. The ions are accelerated to 60 keV and mass separated by a magnetic sector field of resolving power $R = m/\Delta m$ of up to 8000.

The ion beam is transported to the ISOLTRAP setup where it is efficiently stopped and cleaned from possibly remaining isobaric and isomeric contaminants before the mass of a radioactive nuclide can be measured. To this end the ISOLTRAP spectrometer consists of three main parts as shown in fig. 1: a gas-filled linear radio-frequency quadrupole (RFQ) trap, a gas-filled cylindrical Penning trap, and a high-vacuum hyperboloidal Penning trap.

The radioactive ion beam delivered from ISOLDE is accumulated, cooled, and bunched in the linear RFQ trap. The main task of this device is to transform the 60 keV continuous ISOLDE beam into ion bunches at low energy (2–3 keV) and low emittance ($\leq 10\,\pi$ mm mrad) [17]. These bunches can be efficiently transported to and captured in the first, cylindrical Penning trap, where a mass-selective buffer-gas cooling technique is employed. It allows the trap to operate as an isobar separator with a resolving power of up to $m/\Delta m = 10^5$ for ions with mass number $A \approx 140$ [18]. The ions are then transported to the second, hyperboloidal Penning trap [19]. This is the high-precision trap used for the mass measurements of the ions. It can also be used as an isomer separator with a resolving power of up to $m/\Delta m = 10^7$ [20]. The actual mass measurement is carried out via a determination of the cyclotron frequency $\nu_c = qB/(2\pi m)$ of an ion with mass m and charge q in a magnetic field of strength B. For this, an azimuthal radio-frequency field is applied to excite the ion motion. The duration T_{RF} of the excitation determines the mass resolving power $R = m/\delta m = \nu/\delta\nu$, $\delta\nu \approx 1/T_{RF}$. The energy gained from the excitation is detected by the corresponding decrease in time of flight when the ions are ejected from the trap to a detector. B is determined by measuring ν_c of a reference ion with a well-known mass [14].

The main contributions to the uncertainty of the frequency ratio $r = \nu_c^{ref}/\nu_c$ between the frequency of the reference ion ν_c^{ref} and that of the ion of interest ν_c are unnoticed magnetic-field changes and different orbits in the trap for reference ion and radioactive ion due to their mass difference. The magnitudes of these uncertainties have been investigated in a large number of carbon-cluster ions cross-reference measurements [14]. A residual deviation of only $\delta r/r = 8 \cdot 10^{-9}$ was found during these investigations [14].

The measured frequency ratio $r = \nu_c^{ref}/\nu_c$ is converted into the atomic mass value m for the measured nuclide by

$$m = r \cdot (m_{ref} - m_e) + m_e \qquad (1)$$

with the electron mass m_e and the atomic mass of the reference nuclide m_{ref}.

3 ISOLTRAP mass measurements

3.1 Overview and recent highlights

ISOLTRAP mass measurements span the whole range of physics presented in the introduction. Since they started in the late 1980s [21,22] close to 300 nuclides have been investigated as listed in table 1.

In the beginning, the technique of collecting the radioactive beam on a foil and releasing it by heating gave access only to surface ionizable elements. A large number of measurements of isotopes of alkali elements originate

Table 1. List of short-lived nuclei whose mass was measured with ISOLTRAP.

Element	Mass number	Reference
Ne	17	(b)
Ne	18, 19, 23, 24	[23]
Na	21, 22	[12]
Mg	22	[12]
K	35 ... 38, 43 ... 46	(b)
Ar	33, 34, 42, 43	[24]
	34	[13]
	32, 44, 45, 46	[25]
Cr	56, 57	[26]
Mn	56, 57	[26]
Ni	57, 65 ... 69	[27]
Cu	66 ... 74, 76	[27]
Ga	63 ... 65, 70, 72 ... 78	[27]
Se	70 ... 73	(a)
Br	72 ... 74	(a)
Kr	74 ... 78	[13,11]
	72 ... 74	[8]
	87 ... 95	(b)
Rb	75 ... 84, 86, 88 ... 94	[28,29]
	74	[13,11]
	82m	[26]
Sr	78 ... 83, 87, 91 ... 95	[28,29]
	76, 77	[30]
	92	[26]
Ag	98 ... 101, 103	(a)
Sn	128 ... 132	[30]
Xe	114 ... 123	[31]
Cs	117 ... 132, 134 ... 142	[32]
	124, 127	[26]
	145, 147	[33]
Ba	123 ... 128, 131, 139 ... 144	[32,34]
	130	[26]
Ce	132 ... 134	[35]
Pr	133 ... 137	[34]
Nd	130, 132, 134 ... 138	[34]
Pm	136 ... 141, 143	[34]
Sm	136 ... 143	[34]
Eu	139, 141 ... 149, 151, 153	[34,35]
Dy	148, 149, 154	[34,35]
Ho	150	[34]
Tm	165	[35]
Yb	158 ... 164	[35]
Hg	179 ... 195, 197	[4]
Tl	181, 183, 186m, 187, 196m	[33]
Pb	196, 198	[4]
	187, 197m	[33]
Bi	197	[4,33]
	190 ... 196, 215, 216	[33]
Po	198	[4]
At	203	[4]
Fr	209 ... 212, 221, 222	[36]
	203, 205, 229	[33]
	230	this work
Ra	214, 229, 230	[33]
	226, 230	[36,32]
	229 ... 232	this work

(a) Measured in 2002, to be published.
(b) Measured in 2004, to be published.

from this time [28,32,36]. The first important improvement was obtained when the first Penning trap was reconstructed and decoupled from the collection foil. This new trap could be used to purify the ISOLDE ion beam from isobaric contaminations very efficiently. This led to successful measurements in the rare-earths region of the chart of nuclei [34]. The next important step was the replacement of the collector foil by a large cylindrical Paul trap. This buffer-gas filled trap made it possible to stop and accumulate all elements produced at ISOLDE. A long chain of mercury isotopes and some heavier bismuth, polonium and astatine isotopes [4] have been measured after the installation of this device. The impact on the mass surface was considerable since there where many alpha-decay chains linked to the measured mercury isotopes. The main challenge was the resolution of low-lying, long-lived isomeric states in order to identify the ground state unambiguously. Later, to improve the efficiency, the large Paul trap has been replaced by a linear radio-frequency structure that is presently used [17]. The boost in efficiency enabled measurements on even more exotic nuclei as for instance the very short-lived ^{33}Ar [37]. Since then a large number of nuclides has been investigated and measured with ISOLTRAP (table 1).

A recent highlight is the measurement of the mass of ^{22}Mg and its reaction partners with unprecedented precision. These studies are motivated by both, nuclear astrophysics and a standard model test. Ten independent frequency ratios between ^{22,24}Mg, $^{21-23}$Na, and ^{37,39}K were measured with a relative uncertainty of 10^{-8}. A least-squares adjustment with input data from the Atomic Mass Evaluation 2003 [38] for ^{24}Mg, ^{23}Na, and ^{37,39}K determined the mass and β-decay Q values with an uncertainty of about 250 eV [12].

This allowed, together with the branching ratio and half-life measurements from [39] as well as the theoretical corrections from [40], to obtain a corrected Ft-value that is almost comparable to nine decays previously studied with high precision [41]. The uncertainty in Ft is still about 2.5 times higher than for the best cases but it is now mainly due to the branching-ratio uncertainty [41].

With the mass measurements of ^{74}Rb [11], the shortest-lived nucleus ever investigated in a Penning trap ($T_{1/2}$ = 65 ms), that of its daughter ^{74}Kr, and that of ^{34}Ar it became possible to add three nuclides to the investigation of superallowed β decays. Due to the recent ISOLTRAP experiments the precision of the comparative half-life Ft for these three decays is no longer limited by the mass uncertainty.

Another highlight shows the need of further mass measurements in the context of nuclear astrophysics, in particular of the rp-process above $Z = 32$. The mass of ^{72}Kr and other krypton isotopes has been measured with a relative uncertainty close to 10^{-7} [8]. Using these precise new mass values, the mass of rubidium and strontium nuclei (and hence the proton separation energies) could be determined via Coulomb energies with a rather high precision of about 100 keV. In combination with proton-capture rates, photo-disintegration rates and β-decay half-lives the

Table 2. Frequency ratios and mass excess values (ME) for radium and francium isotopes. ^{133}Cs was used for calibration of the magnetic field of the Penning trap. Literature values $\mathrm{ME_{lit}}$ are from [38]. The last column gives the difference between literature and ISOLTRAP values $\Delta = \mathrm{ME^{*}_{exp}} - \mathrm{ME_{lit}}$.

Nuclide	$T_{1/2}$	freq. ratio ν_c^{ref}/ν_c	$\mathrm{ME^{*}_{exp}}$ (keV)	$\mathrm{ME_{lit}}$ (keV)	Δ (keV)
^{229}Ra	4.0 min	1.723 295 23(20)	32542(24)	32563(19)	21
^{230}Ra	93 min	1.730 835 33(16)	34513(19)	34518(12)	5
^{231}Ra	103 s	1.738 389 50(16)	38226(19)	38400(300)$^{\#}$	174
^{232}Ra	250 s	1.745 932 03(10)	40498(12)	40650(280)$^{\#}$	152
^{230}Fr	19.1 s	1.730 875 61(24)	39500(29)	39600(450)$^{\#}$	100

* $m(^{133}\mathrm{Cs}) = 132.905\,451\,933(24)$ u [38];
1 u = 931.4940090(71) MeV/c^2 [42].
$^{\#}$ Value from extrapolation using experimental data trends [38].

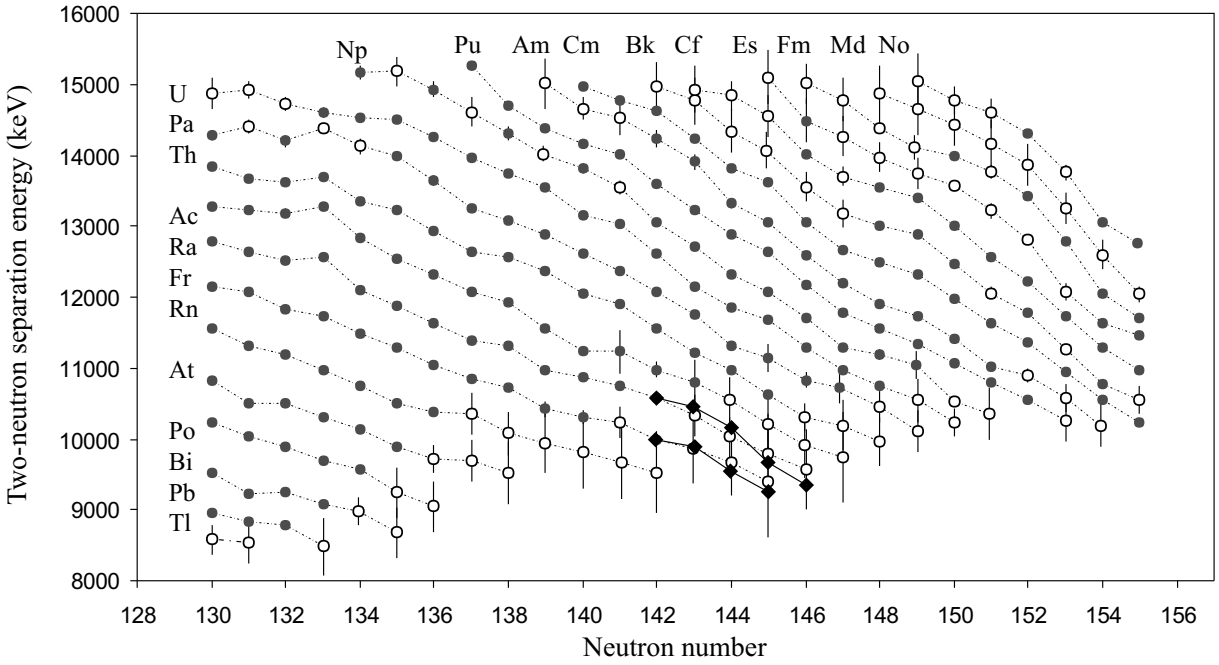

Fig. 2. Two-neutron separation energy plotted as a function of the neutron number. Full circles mark experimental values as cited in the Atomic Mass Evaluation 2003 [38]. Open circles result from an extrapolation [38]. Data that include one of the radium or francium nuclides presented in this work and in [33] are marked with diamonds and connected with a solid line. The error bars of these new data points are smaller than the symbols.

mass values allowed to perform detailed calculations in the vicinity of this possible waiting point. The result is the effective lifetime of ^{72}Kr in the stellar environment. It can be concluded that ^{72}Kr may indeed be a waiting point nucleus. However, the proton separation energy of ^{74}Sr changed due to the new mass values and consequently the rate of the $^{73}\mathrm{Rb}(p,\gamma)^{74}\mathrm{Sr}$ reaction has increased influence on the effective lifetime of ^{72}Kr. In particular a resonant state in ^{74}Sr close to the proton threshold could change the present picture considerably [8].

3.2 The masses of radium and francium isotopes

The masses of neutron-rich radium and francium nuclei, the heaviest masses investigated yet at ISOLTRAP, have been measured during a recent experiment. For the production of the radionuclides a uranium carbide target was bombarded about each second with approximately $3 \cdot 10^{13}$ protons per pulse at 1.4 GeV. After their diffusion out of the hot (≈ 2000 K) target container the atoms were surface-ionized in a tungsten ionizer cavity. The ions were accelerated to 60 keV, mass-separated by the high-resolution mass separator HRS at ISOLDE [16] and transferred to ISOLTRAP where the ratio of their cyclotron frequency was measured with respect to the cyclotron frequency of ^{133}Cs ions.

Ions of different mass (contaminations) simultaneously present in the precision trap would influence the cyclotron frequency of the ions of interest. Therefore, all measured cyclotron frequencies were evaluated as a function of the number of detected ions in each experimental cycle. This procedure allows an extrapolation to only a single ion stored in the trap. The extrapolation uncertainty can be considered as a reflection of possible, but unobserved contaminations [14].

The results of the mass measurements of very neutron-rich radium and francium nuclei are summarized in table 2. For comparison the mass excess values as given in the Atomic Mass Evaluation (AME) 2003 [38] have been added. The AME2003 values for ^{229}Ra and ^{230}Ra are based on previous ISOLTRAP measurements [33] and have been reproduced very well in the present measurements. The mass of three nuclides has been measured for the first time.

To visualize the effect of the new results on the mass landscape the two-neutron separation energy is plotted as a function of the neutron number before and after our measurements (fig. 2). The considerably lower uncertainty as well as the first ever measurement of ^{230}Fr mark a clear change in the trend of the two-neutron separation energy. While the extrapolation naturally delivered a smooth behavior, the experimental values do not follow this trend. A significant drop in the S_{2n} value is observed between $N = 144$ and 145 for radium and for francium between $N = 143$ and 144. Further investigations of the impact of these new and more precise mass values will follow in a detailed publication [33].

4 Summary and outlook

The wide range of physics interest in precise nuclear masses has triggered the development of many mass spectrometers. Penning trap spectrometers provide the most precise and reliable mass values. As an example the heaviest short-lived nuclides studied with ISOLTRAP have been presented —the neutron-rich radium and francium isotopes $^{229-232}$Ra and ^{230}Fr, three of which have been measured for the first time.

While the masses of close to 300 radioactive nuclides have already been determined with ISOLTRAP, the recent developments at this triple-trap mass spectrometer mark the present limit of measurements of short-lived radioactive nuclides. The relative mass uncertainty has reached 10^{-8} as verified by cross-reference measurements on carbon clusters. New experimental techniques are currently implemented that will improve the stability of the magnetic field, the sensitivity and give access to nuclides of elements not produced at ISOLDE [43]. Thus the range for possible applications will be further extended.

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