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Direct mass measurements of exotic nuclei

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Abstract. The present status and recent results from direct mass measurements of exotic nuclei are presented. ISOL, in-flight, and combined facilities provide a wide variety of nuclides far-off stability covering a wide range of half-lives down to the sub-millisecond region. Modern direct mass measurements are carried out using frequency and time-of-flight techniques. The obtained accurate mass data point to nuclear-structure phenomena and serve as a basis for astrophysical and weak-interaction studies.

PACS. 21.10.Dr Binding energies and masses – 32.10.Bi Atomic masses, mass spectra, abundances, and isotopes

In this contribution the presently working experiments for direct mass measurements of exotic nuclei and their results as well as new developments are presented. For a general overview, in particular for an overview of other methods for mass determinations the reader is referred to review articles [1–4] and to ref. [5].

1 What can one learn from atomic masses of exotic nuclei

Besides a general interest in nuclear gross properties there are three pillars that serve as motivation for accurate mass measurements: nuclear astrophysics, weak-interaction studies, and —last but not least— basic nuclear and nuclear-structure physics. In the latter field many open questions wait for a decisive answer [6], for instance: Where are the driplines located? Where is the limit of stability for superheavy elements? Are there other decay modes? How does shell structure develop far away from the valley of stability? Are there new magic numbers, like, for instance, the new "halo-driven" magic numbers [7]? From atomic masses, separation energies and Q-values can be derived which are key parameters to answer these questions.

Of particular interest in nuclear astrophysics are atomic masses, reaction Q-values, and half-lives along the paths of nucleosynthesis, especially the r- and rp-process. Besides the precise knowledge of the astrophysical environment [8–10], these nuclear properties serve as input parameters for nuclear-reaction network calculations, which try to determine the true reaction path for the nucleosynthesis and to reproduce the observed elemental and isotopic abundances. Many of the relevant nuclei are still out of reach for a laboratory experiment, in particular those related to the r-process [8], and therefore the network calculations are mostly based on theoretical and/or extrapolated atomic masses. This, of course, introduces uncertainties to the results and therefore reliable experimental information on atomic masses is sought.

Possible limitations of the Electroweak Standard Model can be derived from nuclear beta-decays in superallowed $0^+ \to 0^+$ transitions, where the experimental ft-value is directly related to the weak vector coupling constant. Therefore, it is important to know accurately the half-life, branching ratios, and decay energy in these transitions. The ft-values of nine cases between ^{10}C and ^{54}Co are currently known with an uncertainty on the 0.1% level, which yield consistency with a unique value for the vector coupling constant. Further tests are expected to come from heavier masses among the $N = Z$ nuclei [11].

2 Frequency measurements

Three different devices, which take advantage of frequency measurements, are operational for mass measurements: Penning traps, the RF-spectrometer, and storage rings.

Penning traps are in routine operation for on-line mass measurements with unstable nuclei at ISOLTRAP [12] and many new systems are under construction (see below). The specific charge q/m of an ion is determined by the measurement of the cyclotron frequency $f_\mathrm{c} = \frac{1}{2\pi} \cdot B \cdot q/m$ in a magnetic field B, which is calibrated by using reference isotopes with well-known masses. Recently, clusters of ^{12}C, which defines the atomic mass unit, have been used

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at ISOLTRAP for this calibration, allowing for absolute mass measurements and, besides other advantages, minimizing possible mass-dependent systematic errors [13]. The mass-resolving power and the mass accuracy, which can be reached with a Penning-trap mass spectrometer, are limited by the Fourier limit, which connects the obtainable line width δf_c with the observation time T_obs according to $\delta f_\mathrm{c} \simeq 1/T_\mathrm{obs}$. This is presently the most severe accuracy limitation for unstable nuclei. One of the highlights of the recent work at ISOLTRAP was the measurement of short-lived nuclei with half-lives as small as 65 ms (^{74}Rb) and 174 ms (^{33}Ar) with a relative accuracy of $2.5 \cdot 10^{-7}$ and $1.3 \cdot 10^{-7}$ [14], respectively. Only several hundred up to a few thousand ions were detected. These measurements became possible after a capture and cooler device for the ISOLDE beam was installed [15], which extends the range of elements accessible with ISOLTRAP and which significantly improves the overall efficiency of the system due to its capability of beam accumulation and cooling. The measurement of the ^{33}Ar mass allows for a new test of the Isobaric Multiplet Mass Equation (IMME), which is widely used in its quadratic form to determine the unknown mass of a nuclide within an isospin multiplet simply as a function of the isospin projection T_Z [16]. The important result is that the quadratic form is no longer applicable if high mass accuracy is required. This may have severe consequences for experiments searching for scalar contributions to β-decay [17], where the Q-value is derived from the ^{32}Ar mass as obtained by the quadratic form of IMME. Thus, the direct mass measurement of this nuclide is needed and the required accuracy can presently be reached only with a Penning-trap system.

The MISTRAL experiment [18] is also installed at ISOLDE. It is based on a Smith-type radio-frequency spectrometer, where the injected ions perform two revolutions in a homogeneous magnetic field. If two different ions have the same trajectory in the same magnetic field their masses $(m(A), m(B))$ and the corresponding cyclotron frequencies $(f_\mathrm{c}(A), f_\mathrm{c}(B))$ fulfill the equation $f_\mathrm{c}(A) \cdot m(A) = f_\mathrm{c}(B) \cdot m(B)$. Ions undergo only two turns in the spectrometer and are transmitted only if their kinetic energy is appropriately modulated by an RF field, from which the cyclotron frequency is determined. The advantages of this spectrometer are short transit times (of the order of 100 μs), a high resolving power ($m/\Delta m \simeq 1 \cdot 10^5$, and a precision of the order of a few 10^{-7} given sufficient statistics. With the MISTRAL experiment short-lived neutron-rich Na, Ne, and Mg isotopes in the vicinity of the "island of inversion" near $N = 20$ [19] have been investigated with half-lives down to 30 ms (^{28}Na) [20]. The results indicate a pronounced weakening of the $N = 20$ shell closure. As a contribution to the test of of the CVC hypothesis and the unitarity of the CKM-matrix of the standard model, in a recent experiment, the mass of ^{74}Rb ($T_{1/2} = 65$ ms) has been determined with an uncertainty of 100 keV [21]. The result is in agreement with the value obtained by the ISOLTRAP experiment at about the same time with an uncertainty of 19 keV [22] and both values are consistent with the mass evaluation [23]. These results will allow for a test of calculations of the Coulomb correction to the ft-value in superallowed Fermi transitions, which accounts for nuclear-structure effects of the mother and daughter nuclei [11].

An example of a multi-turn mass spectrometer is the experimental storage ring ESR at GSI. Coupled to the in-flight separator FRS, where fragmentation and fission products of all elements can be produced and separated in the sub-μs range, it is a universal tool for precision mass measurements of exotic nuclei [24]. In Schottky Mass Spectrometry (SMS) the injected ions are cooled using an electron beam until they reach the same mean velocity. Their revolution frequency, which is then a direct measure for their specific charge, is determined from Schottky beam noise analysis and subsequent Fourier transformation. The masses are derived from the measured frequencies according to $\Delta f/f = -\alpha_p \cdot \Delta(m/q)/(m/q)$, where α_p is an ion-optical parameter of the ESR. In the latest experiment time-resolved Schottky analysis was used [25] allowing one to study dynamic effects in the ESR and to detect and to correct possible drifts and instabilities of the power supplies. Moreover, the cooling process and nuclear decay processes can be studied. A mass-resolving power of $7 \cdot 10^5$ is reached and the mass accuracy amounts typically 50–100 keV around $A = 180$. The required cooling and measurement time limits the accessible half-lives to approximately a few seconds. Concerning intensity, this technique reaches the ultimate sensitivity since one single ion yields a sufficiently high signal. Furthermore, the power of this experimental technique is evident from the following fact: in only two experiments, where neutron-deficient nuclei were produced by bismuth fragmentation [26,25], masses of almost 350 different nuclides were directly determined, more than 150 of them were previously unknown according to ref. [23]. This allows one to map the mass surface of large areas of the chart of nuclei rather than measuring single mass values. Some additional ninety new masses could be obtained by using the links of α chains, leading to one of the most important results of these experiments: the location of the proton dripline in the region of francium [27,28].

3 Time-of-flight measurements

Three devices are presently in use for time-of-flight mass measurements: the magnetic-rigidity spectrometer SPEG at GANIL, the coupled-cyclotron complex at GANIL, and the experimental storage ring ESR. The common feature is the production of the exotic nuclei in fusion, fragmentation, or fission reactions in "thin" targets (thin compared to the range of the primary ions in the target material) and the separation in-flight. This approach allows one to access nuclear ground-state masses and isomeric states with half-lives as short as the time of flight through these devices, which is less than 10 μs.

At SPEG [29] the exotic nuclei are produced in fragmentation reactions in the SISSI target and pre-separated with the α-spectrometer. The mass-to-charge ratio of each individual ion transmitted in SPEG is determined from its magnetic rigidity $B\rho$ (measured with a position-sensitive detector placed at the dispersive focal plane of SPEG) and its velocity v (determined from the time of flight for a 82 m long flight path) according to $B\rho = \gamma v \cdot m/q$. A mass resolution of 2–$4 \cdot 10^{-4}$ is obtained and the mass uncertainty ranges from typically 100 keV to 1 MeV around $A = 40$ depending on statistics. The method has several advantages: With a detector telescope located at the end of the flight path it is possible to identify each ion individually; this redundant information is a feature which presently no other technique can provide. Taking advantage of a special target consisting of several segments with different thicknesses and due to the kinematic focusing in fragmentation reactions, a large m/q range is accessible thus allowing for the simultaneous transmission of a broad spectrum of reference masses and many nuclides of interest, thus leading to the survey of a whole region, similar to SMS and IMS (see below). Also the sensitivity is comparable to those methods and experiments can be carried out with intensities as low as 0.01 ions per second. A possible difficulty arises from short-lived isomers having half-lives of the order of the flight time through the system, which cannot be resolved and may systematically shift the obtained mass values to higher masses. However, the NaI array at the final focus allows to detect delayed γ-rays and to determine isomeric ratios.

So far, the SPEG mass measurement program has aimed at the investigation of light to medium-heavy nuclei ($A =$ 10–80). Mass measurements along the $N = Z$ line have been carried out using a ^{78}Kr primary beam and dripline nuclei with extreme mass-to-charge ratios $m/q > 3$ have been studied [30,31] using neutron-rich Ca and Ni beams. Recently, improved and new masses were obtained for neutron-rich isotopes from Ne through Ar. Special attention was paid to the two-neutron separation energies near $N = 20$ and $N = 28$. For Ca, K, and Ar isotopes the typical behaviour of a shell closure around $N = 28$, whereas for the Cl, S, and P isotopes the typical change of slope occurs at $N = 26$ indicating a pseudoshell closure [19]. These findings can be explained by shape coexistence. Very recent studies [32] were devoted to extremely neutron-rich nuclei in the vicinity of the heaviest presently known one-neutron-halo nucleus ^{19}C up to ^{22}C.

The use of coupled cyclotrons for direct mass measurements (similar to SARA at Grenoble) was studied at GANIL several years ago [33] and had its greatest success in the first mass measurement of the doubly magic ^{100}Sn [34]. Several species, produced by fragmentation or fusion evaporation in a target placed between the two cyclotrons CSS1 and CSS2, are accelerated simultaneously and their relative time-of-flight difference in the cyclotron $\Delta t/t$ equals their relative m/q difference: $\Delta t/t = \Delta(m/q)/(m/q)$. Ions with small relative m/q differences (of the order of few 10^{-4}) can be transmitted simultaneously within transit times of the order of a few tens of μs. For light ions a mass resolution up to one million and for $A = 100$ isobars typically $3 \cdot 10^4$ has been reached. The mass accuracy in these experiments ranges from 100 keV to 900 keV (depending on statistics) [34]. In more recent experiments masses of $N = Z$ nuclei with $A = 68, 76, 80$ have been determined with uncertainties of the order of 100 keV [35].

The main motivation for these studies [31] comes from shell closures in doubly magic nuclei, deformations, isospin symmetry and the Wigner term [36]. The latter yields additional binding energy due to neutron-proton pairing and depends on the existence of spin-isospin independent forces and can be obtained from double binding-energy differences. The systematics on binding-energy differences could be significantly extended towards higher atomic numbers and confirm the decreasing Wigner effect with increasing mass number [35]. New mass values are expected from the new CIME cyclotron at the SPIRAL facility, which became operational recently [37]. The possibility to vary the RF will allow for a higher flexibility of the system and in particular for a broader m/q range, which can be transmitted with a fixed cyclotron setting.

The path length for the time-of-flight measurement of the ions is further increased in the ESR. The first mass measurements on short-lived nuclei have been carried out employing Isochronous Mass Spectrometry (IMS) [38,39] for neutron-deficient nuclei produced by projectile fragmentation from Kr and Cr beams. In contrast to SMS, ion cooling is not necessary because here the ESR is operated in an isochronous ion-optical mode (similar to TOFI [40] at Los Alamos), where the revolution time is to first order independent of the ion velocity and depends only on the specific charge of the ion. Typically the ions make several hundred turns in the ESR and at each revolution they pass through a fast time-pickup detector. From its signals the revolution time, which is of the order of 500 ns, is determined for each individual ion with a precision of the order of 1 ps. Similar to the measurements at SPEG, a broad m/q range can be transmitted simultaneously ($\pm 7.5\%$ have been observed in the case of Kr fragments [39]) thus yielding a broad spectrum of reference nuclei for mass calibration. The result is a mass-resolving power of $1.1 \cdot 10^5$ and for masses around $A = 70$, experimental uncertainties of 70–100 keV have been reached with the derived mass excess in agreement with the results of other experiments [39]. This method is ideally suited for the mass measurement of extremely short-lived nuclides and isomeric states with half-lives of the order of several tens of μs. So far, the shortest-lived observed nuclide is ^{45}Cr ($T_{1/2} = 50$ ms). Masses in this area play an important role in the modeling of the rp-process [9].

4 New developments

A variety of new setups for direct mass measurements is presently under construction, all of which employ Penning traps as mass spectrometers. The common objective of these experiments is the opportunity to benefit from the specific advantages of the various production and separation methods available for exotic nuclei. This partly requires the development of new techniques (see below).

Low-energy exotic nuclei will be provided by the new reactor in Munich for MAFF-TRAP [41] and by the IGISOL and beam cooler system in Jyväskylä for the JYFL-TRAP [42]. Both experiments will give access to neutron-rich fission fragments which will be available with high intensities.

An experimental challenge is the coupling of a Penning-trap system to an in-flight separator, a new approach which is presently pioneered at different laboratories. Exotic nuclei, which are produced at high energy in fusion or fragmentation reactions and separated in-flight (with the Enge spectrograph at ANL, the A1900 at MSU, and SHIP at GSI), will be stopped and thermalized in a gas-filled stopping cell, from which the ions can be extracted within a time scale of few milliseconds and transported into the trap with high efficiency. This approach combines the advantages of both in-flight separation and ISOL techniques and such systems are under construction at the CPT experiment at ANL [43], at LEBIT at MSU [44], and SHIPTRAP at GSI [45]. At CPT and SHIPTRAP this will allow one to perform mass measurements on fusion evaporation products, and of particular interest are of course candidates in the vicinity of ^{100}Sn, ground-state proton emitters, and elements heavier than uranium.

Finally, a new idea shall be mentioned: it is intended to use electrostatic time-of-flight mass spectrometers for direct mass measurements of short-lived nuclei [46]. These compact devices, in standard operation in molecular physics and in chemistry, are characterized by a high mass resolution and measurement times of the order of 100 μs. Latest improvements and recent investigations with stable lead ions have shown that a mass-resolving power $m/\Delta m = 24000$ and a relative mass accuracy of $\delta m/m = 1 \cdot 10^{-6}$ can be reached [47]. The system can be coupled to a buncher-cooler system like the one at SHIPTRAP and it is planned to use such a mass spectrometer for mass measurements of nuclides with half-lives of the order of few milliseconds. These measurements will ideally complement the Penning-trap mass program at SHIPTRAP.

5 Summary and conclusions

Six different experimental techniques are currently in use for direct mass measurements of exotic nuclei. The mass spectrometers are adapted to the various production mechanisms and separation techniques and employ frequency and time-of-flight measurements. Roughly speaking, frequency measurements deliver the most accurate mass values, whereas time-of-flight measurements allow to access the most short-lived nuclei. The common interest of all experimental efforts is to push forward the results at three frontiers: the precision frontier, the half-life frontier, and the isospin frontier. The common prerequisites to reach these goals are high-intensity secondary beams, selectivity of the mass separator and/or mass spectrometer, sensitivity of measurement and detection, and an unambigous particle identification. These challenges for future experiments will keep direct mass measurements a broad field of vivid interest and will lead to a better understanding of the strong force and effects in the nuclear medium.

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