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Direct mass measurements of neutron-rich zirconium isotopes up to ^{104}Zr

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Atomic masses of radioactive zirconium isotopes from ^{96}Zr to ^{104}Zr have been measured with a relative accuracy of $\leqslant 5\times10^{-7}$ using a Penning trap coupled to the ion guide isotope separator on-line system. The obtained two-neutron separation energies show strong local correlation in relation to the shape change and shape coexistence between $N=58$ and 60.

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The mass of the ground state of a nucleus results from the structure of a complex quantum system. Therefore, an accurate determination of the nuclear mass surface can provide information additional to that obtained from excited states, on the underlying symmetries and microscopic features of nucleon systems such as charge symmetry of nuclear interaction, shell effects, coexisting structures, pairing effects, spin-orbit interaction, and so forth. For this to be successful, measurements and theory have to beable to probe fluctuations in the order of 1 to 100 keV. Global correlations are typically variations due to closed shells and broad areas of deformation where the required accuracies in mass measurements are typically of the order of 100 keV. However, detection of local correlations such as those due to the presence of closed shell discontinuities, the local zones of deformation or those due to configuration mixing or shape mixing require mass accuracies preferably of the order of 10 keV [1,2].

Zirconium isotopes are known to possess interesting and rapidly changing nuclear structure features (like shape) when the neutron number changes from 56 to 60. Extensive spectroscopic studies have been employed to understand this shape transition and its relation to structural dynamic symmetries as well as to underlying microscopic structures, proton-neutron interaction and to neutron-pairing. Experiments have so far investigated these nuclei via beta decays of yttrium parent nuclei [3], prompt fission fragment gamma-ray coincidence studies [4] as well as collinear laser spectroscopy [5]. These studies have changed the picture considerably such that the original description of the shape change [6] as a sharp phase transition from a spherical shape at $N=59$ to a strongly deformed ^{100}Zr is rather described as a gradual transition setting in already at $N=56$.

Despite very detailed spectroscopic studies of neutron-rich zirconium nuclei their masses are known rather poorly. The masses of ^{97}Zr and ^{98}Zr have been determined accurately by n-capture and (t,p) reactions, respectively. The binding energies of other neutron-rich zirconium isotopes have only been determined, if at all, by measuring the beta end-point energies of yttrium and zirconum as well as their daughter isotopes [7–9]. Q_β measurements often incur systematic errors due to inadequate knowledge of the decay schemes and the fact that the final mass determinations are relying on the long decay chains to the valley of stability.

In this article we report on the first direct nuclear mass measurements of radioactive zirconium ions employing the JYFLTRAP set up located at the IGISOL facility of the cyclotron laboratory of the University of Jyväskylä, see Fig. 1. The basic principle of mass determination by a Penning trap is based on the determination of the cyclotron frequency of an ion in a strong homogeneous magnetic field. In the presence of an axially symmetric quadrupole field ions perform characteristic motion consisting of three independent components. These are an axial motion with a frequency f_z and two radial motions, a slow magnetron motion (f_-) and a fast reduced cyclotron motion (f_+). The overall radial motion is a superposition of the two radial motions and is connected to the magnetic field via the relation

$$f_c = f_- + f_+ = \frac{1}{2\pi}\frac{q}{mB},$$

where f_c is the cyclotron frequency of an ion with a charge-to-mass ratio of q/m oscillating in the uniform magnetic field B. The field is determined using reference ions with well-known masses.

Ion trap technology was introduced to atomic mass measurements of radioactive isotopes at ISOLDE [10]. In the

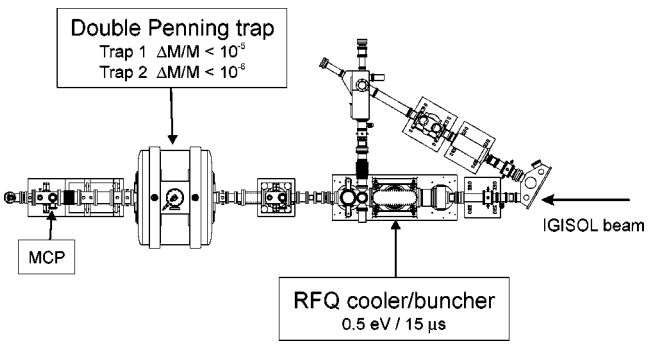

FIG. 1. JYFLTRAP setup.

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early days of these experiments, the lack of a fast and efficient injection into the trap posed serious limitation to the use of traps for exotic short-lived isotopes. This obstacle was recently solved by the introduction of a fast injection scheme based on the gas-filled linear Paul trap, see Ref. [11] for further reading.

At IGISOL—Ion Guide Isotope Separator Online—ions are produced in a thin target and recoil out into a fast flowing helium gas. After thermalization in the gas they are guided as singly charged ions by helium flow and electric fields through stages of differential pumping and finally accelerated to 30 keV. After acceleration the beam is mass selected by a dipole magnet, allowing separation of nuclei with different mass numbers. The IGISOL technique allows access to exotic short-lived isotopes of all chemical elements including the refractory ones [12] and is therefore ideal to supply radioactive ions for a trap system. As the energy spread of the IGISOL beam is rather large, up to 50–100 eV, the beam has to be cooled before it can be manipulated further. The cooling is accomplished in a buffer-gas filled radio frequency quadrupole (RFQ) [13]. This allows not only cooling but also bunching of ions leading to extraction of ion pulses with a duration of 15 μs and an energy spread of the order of 1 eV. These ions are injected into a Penning trap system which consists of two trapping regions, a purification trap with medium mass resolution and a precision trap with high mass resolution. The measurements reported in this paper were performed with the medium resolution purification trap. A superconducting magnet is used to create the magnetic field for the trap. This 7.0 T actively screened magnet system has two 1 cm^3 homogenous magnetic field regions along the beam axis, symmetrically 10 cm in both directions from the magnet center. In these areas the homogeneity of the magnetic field is below 10^{-6} and 10^{-7} in the purification and precision traps, respectively.

Ion bunches from the RFQ cooler are sent at low energy ($<$1 keV) into the purification trap where they are captured by a time-varying electric potential, thermalized and cooled in a buffer gas in the center of the potential. They are subsequently excited by applying successive dipole and quadrupole rf fields which lead to mass-selective cooling and centering according to $\omega_c = qB/m$, for more details see Refs. [14,15]. The ejection through a small diaphragm between the traps allows only those ions close to the trap axis to be extracted. These ions are then transferred to a micro channel plate (MCP) detector for their detection.

The first radioactive ions studied with the JYFLTRAP were produced in a ^{58}Ni$(p,n)^{58}$Cu reaction. In these experiments a mass resolving power of $M/\Delta M_{FWHM} = 145000$ for the purification trap, corresponding to $\Delta M = 400$ keV$/c^2$, was reached allowing determination of the (already well known) mass of ^{58}Cu with an overall uncertainty of about 10 keV. The second milestone was reached when neutron-rich nuclides produced in 30 MeV proton-induced fission of ^{238}U were successfully injected into JYFLTRAP. In the first experiments the main goal was to separate adjacent isobars ^{112}Rh and ^{112}Ru with a mass-energy difference of about 4 MeV. The mass resolving power determined from the full width at half maximum of the count rate versus frequency was approximately 60000. This corresponds to the width of the mass peak of about 1.5 MeV. Therefore, it was not sufficient to separate the isomeric state known to exist in ^{112}Rh from its ground state. On the other hand, in cases where only one long-living state, i.e., the ground state, is known to exist the current precision provided a possibility for accurate mass measurements employing the purification trap only. In such cases a typical accuracy of the order of 50 keV could be obtained for the peak position of the cyclotron resonance curve.

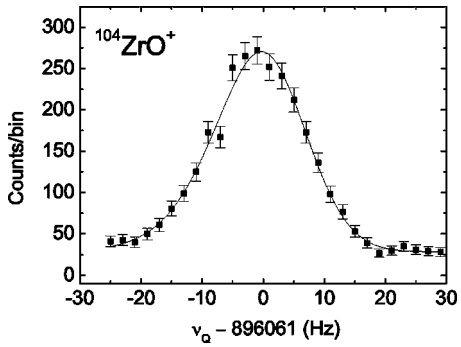

FIG. 2. Frequency scan for ^{104}Zr obtained with the purification trap of the JYFLTRAP setup.

Zirconium isotopes are available as primary beams at IGISOL with intensities of the order of 4000–200 ions/s from ^{98}Zr to ^{104}Zr, respectively. These isotopes are attractive for direct mass measurements only using the purification trap because decay [9] and laser spectroscopic [5] studies have indicated no long-living isomers that could survive the ion manipulation process at JYFLTRAP. The manipulation process consisted of a total cycle time of 450 ms within which the atomic zirconium ions from IGISOL were first injected for cooling and bunching into the RFQ cooler. They were ejected as a 15 μs long bunch into the purification trap, axially cooled for 330 ms, and subsequently excited into large radial orbits by applying a magnetron frequency in dipole mode for 15 ms. By applying a rf quadrupole field for 90 ms the orbits of those ions that corresponded to the resonant cyclotron frequency $\omega_c = qB/m$ were centered. During the course of these experiments it was discovered that zirconium ions reacted with impurities in the He buffer gas of both the cooler and trap, forming monoxide ions. No contaminants with a mass similar to ZrO were produced, thus mass measurements could be performed on isobarically clean zirconium monoxide ions. Figure 2 shows a cyclotron frequency spectrum measured for $^{104}ZrO^+$ ions using the measuring sequence described above. Similar measurements were performed for all zirconium isotopes between $A = 96$ and 104. Typically the measurement sample contained only few ions at a time. Therefore, no count rate dependent corrections for the observed frequencies were needed. The mass calibration was performed by using the well known mass of ^{97}Zr and cross-checked with the mass of ^{58}Ni created in the IGISOL gas cell by sputtering of the Ni-foil separating the uranium targets and the stopping gas cell of the ion guide.

To extract the cyclotron frequency the peak position of the distribution was determined. This was accomplished by fitting a function that included the possibility to account for asymmetries in the measured distributions. From the calcu-

TABLE I. The mass excess values of neutron-rich zirconium isotopes determined in the present experiment. The values from the recent Atomic Mass Evaluation [9] are also given.

Nuclide	Mass excess (keV)		AME03-value (keV)	
^{96}Zr	−85410	40	−85442.8	2.8
^{97}Zr	—	—	−82946.6	2.8
^{98}Zr	−81270	40	−81287	20
^{99}Zr	−77615	18	−77768	20
^{100}Zr	−76397	17	−76600	40
^{101}Zr	−73190	19	−73460	30
^{102}Zr	−71520	40	−71740	50
^{103}Zr	−67780	60	−68370	110
^{104}Zr	−65820	50	−66340	400

lated functions the peak position was determined numerically. As this procedure does not allow a direct determination of the statistical uncertainty of the cyclotron frequency, the bootstrap method (see [16]) has been employed. This method uses the original frequency scan to derive a number of synthetic data sets. These new sets are formed by adding a random number to the original data points. The distribution of this random number is given by a normal distribution, which has a mean of 0 and a standard deviation given by the uncertainty of the original data point (square root of the detected ions). By analyzing these synthetic data sets in the same way as the original frequency scan, a distribution of the peak frequencies around the original value can be determined. By calculation of the standard deviation of this artificially created distribution an estimate of the uncertainty of the determined cyclotron frequency can be derived. The overall uncertainty obtained via this procedure is the dominant part of the total uncertainty.

Results of the measurements are shown in Table I and in Fig. 3. The latter shows the mass difference between the recent Atomic Mass Evaluation AME03 [9] and this experiment. These results show that our measurements agree with

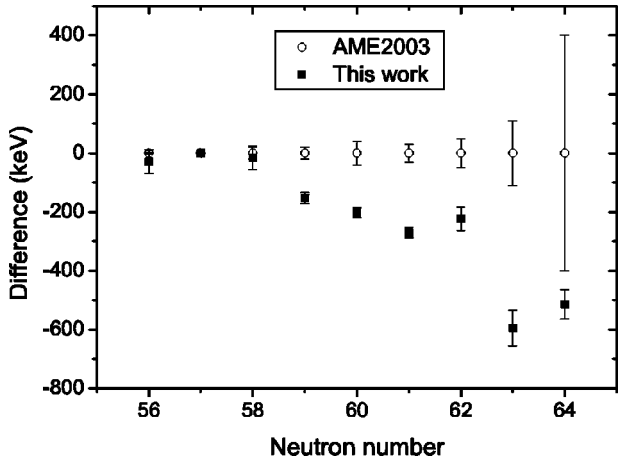

FIG. 3. Results of the atomic mass measurements of neutron-rich zirconium isotopes displayed as a difference between the 2003 Atomic Mass Evaluation [9] and the values measured in this work.

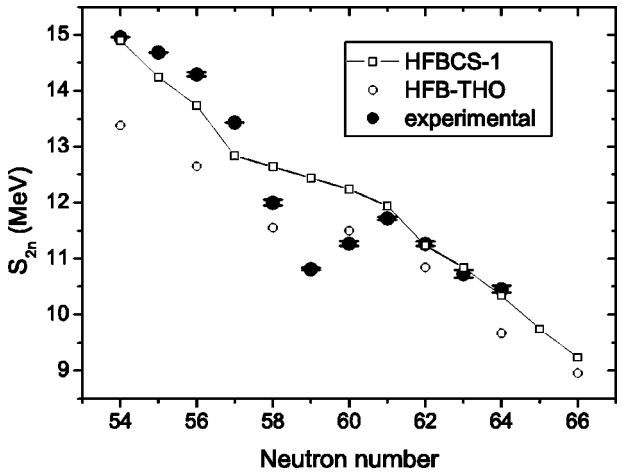

FIG. 4. Two-neutron separation energies of neutron-rich zirconium isotopes. Theoretical values are from the Hartree-Fock calculations of Goriely *et al.* [17] (open squares) and Stoitsov *et al.* [18] (open circles). See the text for more details.

the values from literature for the well known masses of ^{96}Zr and ^{98}Zr, but reveal significant deviations for the more neutron-rich isotopes—all of which have been previously determined either by beta end-point measurements or have not been measured at all (^{104}Zr).

The two-neutron separation energies are often used to trace changes in nuclear structure without complications of effects such as pairing. Therefore, in order to see possible local structure effects in the binding energy of zirconium isotopes in a way similar to excited states or charge radii we have plotted the two-neutron separation energy S_{2n} as a function of the neutron number in Fig. 4. In this presentation we see clearly a strong deviation from a smooth behavior developing at ^{99}Zr and ending at ^{101}Zr. This coincides with the neutron numbers where large changes in quadrupole deformation are known to occur based on the measurements of charge radii [5] and quadrupole moments of rotational bands [4].

As it is of significant interest to learn whether this effect is observed in the nuclear mean field we also show in Fig. 4 two-neutron binding energies from a recent calculation of Goriely *et al.* [17]. This calculation is based on the Hartree-Fock method with pairing correlations described by the BCS approximation using a delta-function pairing force and employing a Skyrme force, MSk7. The Skyrme and pairing parameters are determined by fitting to the data set of 1719 measured masses given in the 1995 compilation of Audi and Wapstra. In this calculation zirconium isotopes have a spherical ground state up to $N=55$, are only weakly deformed up to $N=57$, have an oblate ground state at $N=58$ and prolate shape with $\beta \approx 0.3-0.38$ for $N \geqslant 59$. It is observed that this calculation reproduces weakly a kink in the S_{2n} systematics at $N=57$ to 60. The calculation is in excellent agreement with the experimental data for heavy, well-deformed zirconium isotopes. A clearer indication of the kink is observed in a newer calculation based on the Hartree-Fock method by Stoitsov *et al.* [18]. Unfortunately, the calculation is done only for even-even nuclei. This calculation employs the Hartree-Fock-Bogolyubov approximation in the trans-

formed harmonic oscillator basis. The Skyrme force is SLy4 and pairing is of a mixed volume-surface type. Exact particle-number-projection is performed after finding the Lipkin-Nogami solution. The S_{2n} values obtained correspond to the minimum potential energy solution. In this calculation zirconium isotopes have a spherical ground state up to $N=56$ and a weakly deformed oblate ground state for $N=58$. The calculation gives a stable prolate shape with $\beta \approx 0.38$ between $N=60$ and $N=70$. Agreement with the experimental data for strongly deformed zirconiums is worse but could probably be improved with efforts to fit forces in this specific mass region. Also, significantly larger deformation of 0.42 has been deduced from laser spectroscopic measurements for ^{102}Zr [5]. In summary, it is obvious that at least partly the discontinuity in the two-neutron separation energies could be, at least partly, due to mean-field and pairing effects.

The observed deep minimum in two-neutron separation energies is inherently coinciding with the neutron numbers where different shapes coexist closely at low energy and consequently can mix substantially. It is clear that the shape transition in this case is very strongly exhibited in the binding energies of the ground states. Therefore, these nuclei would provide an interesting testing ground for similar theoretical calculations as applied for neutron-rich rare earth isotopes above the closed $N=82$ neutron shell that employed the interacting boson model, see Ref. [1].

In this work we have shown that accurate mass measurements are now possible for short-lived exotic isotopes of refractory elements. At IGISOL these measurements will be extended to more neutron-rich zirconium isotopes as well as to neighboring isotopes from Sr up to Pd in the near future. The precision trap under commissioning will soon allow mass measurements with 10 keV accuracy or even better. This will further enhance a quality of studying the connections between the mass surface and nuclear structure far from the valley of beta stability.

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