First Precision Mass Measurements of Refractory Fission Fragments

U. Hager,[1] T. Eronen,[1] J. Hakala,[1] A. Jokinen,[1,*] V. S. Kolhinen,[2] S. Kopecky,[1] I. Moore,[1] A. Nieminen,[1] M. Oinonen,[3] S. Rinta-Antila,[1] J. Szerypo,[2] and J. Äystö[1]

[1]*Department of Physics, University of Jyväskylä, P.O. Box 35 (YFL), FIN-40014, Finland*
[2]*Sektion Physik, University of Munich (LMU), Am Coulombwall 1, D-85748 Garching, Germany*
[3]*Helsinki Institute of Physics, University of Helsinki, P.O. Box 64, FIN-00014, Finland*

(Received 15 April 2005; published 1 February 2006; corrected 7 February 2006)

Atomic masses of $^{95-100}$Sr, $^{98-105}$Zr, and $^{102-110}$Mo have been measured with a precision of 10 keV employing a Penning trap setup at the IGISOL facility. Masses of ^{104,105}Zr and ^{109,110}Mo are measured for the first time. Our improved results indicate significant deviations from the previously published values deduced from beta end point measurements. The most neutron-rich studied isotopes are found to be significantly less bound (1 MeV) compared to the 2003 atomic mass evaluation. A strong correlation between nuclear deformation and the binding energy is observed in the two-neutron separation energy in all studied isotope chains.

DOI: 10.1103/PhysRevLett.96.042504 PACS numbers: 21.10.Dr, 07.75.+h, 27.60.+j

Accurate knowledge of masses of radioactive isotopes is important not only in nuclear physics but in several other areas of physics, such as the element synthesis in the Universe and studies of fundamental interactions and symmetries. Within nuclear physics, masses and binding energies are sensitive probes to explore the evolution of nuclei far from the valley of stability, including shell structure, pairing, and spin-orbit coupling. Until now there has been a significant lack of accurate and reliable data on masses of neutron-rich nuclei. The purpose of this Letter is to demonstrate that for the first time the accurate measurement of masses of exotic neutron-rich nuclei can be performed in a universal way employing a combination of the ion guide isotope separator on-line (IGISOL) method [1] and Penning trap technique.

In this Letter, we present results on the first precision measurement of the masses of neutron-rich Sr, Zr, and Mo isotopes. These studies are motivated by earlier gamma and laser spectroscopy experiments that observed a shape transition between $N = 58$ and 60. In our first, medium-resolution study of neutron-rich Zr isotopes with a purification Penning trap [2], this change of shape was observed to result in a large local increase in the two-neutron separation energy. Whether this behavior extends to the neighboring strontium and molybdenum isotopes was studied in this work with a significantly better mass resolution than previously available.

Earlier information of some masses measured in this work had been deduced from beta-decay end point energies; see discussion in Ref. [2,3]. As shown by this work those measurements lead to large systematic errors due to an inadequate knowledge of the decay schemes and complex spectra. Such erroneous data are serious problems in developing theoretical mass models for neutron-rich nuclei far from stability.

The mass measurements described in this Letter were performed at the IGISOL facility at the University of Jyväskylä [1]. Ions of interest were produced in a fission reaction of natural uranium induced by proton bombardment at 25 MeV. Yields of studied nuclei varied from a few thousands of ions/s down to ~100 ions/s for the most exotic ones. After acceleration to 30 keV and mass separation, ions are injected into a gas-filled radio frequency quadrupole cooler (RFQ) [4] where interactions between the ions and buffer gas atoms effectively cool the ion ensemble. In addition to cooling, the RFQ is used to collect and store the ions, which can be released as narrow bunches, typically of the order of 10–20 μs. A cooled and bunched ion cloud can be easily transported to the injection of a Penning trap.

The Penning trap system at IGISOL consists of a purification and a measurement trap. The two traps are housed within the warm bore of a 7 T superconducting magnet that has two homogeneous regions of 1 cm^3 volume separated by 20 cm in the center of the magnet. The first trap used for the isobaric purification has a homogeneity $\frac{\Delta B}{B}$ better than 10^{-6}, and the second trap, called a precision trap, better than 10^{-7}. Both traps are of a cylindrical type.

The trapping of ions is accomplished by a combination of a homogenous magnetic field and a static electric quadrupole field. Solving the equation of motion [5] for an ion in the trap results in three different eigenmotions: one axial and two radial motions, the latter corresponding to *magnetron* and *reduced cyclotron motions* with angular frequencies ω_- and ω_+, respectively. The sum of these two frequencies correspond to the true cyclotron frequency

$$\omega_c = \omega_+ + \omega_- = \frac{qB}{m}. \quad (1)$$

The purification trap uses a buffer gas cooling technique which employs helium as buffer gas with a pressure of 10^{-4} mbar. Isobaric purification is performed by first increasing the ions' magnetron radius beyond the 1 mm radius of a narrow channel located between the two traps.

This is done by applying a dipole electric field at the magnetron frequency. Following this, a mass-selective quadrupole excitation at the true cyclotron frequency is applied which converts the magnetron motion to reduced cyclotron motion thus increasing the kinetic energy of the excited ions. Because the first trap is filled with buffer gas, the fast reduced cyclotron motion is cooled via ion-atom collisions. Only those ions with the true cyclotron frequency are centered and extracted, resulting in an isotopically clean ion bunch. By choosing an appropriate excitation time and amplitude, it is possible to achieve a mass resolving power of up to 10^6 [2,6,7]. Figure 1 shows an example of an isobaric purification scan for $A = 101$ ions.

The isobarically clean sample is injected to the precision trap, which operates under vacuum. There the ions are first given some magnetron radius, typically 0.5 to 1 mm with a dipole excitation, using the phase-locking technique [8]. Following this, a quadrupole excitation with a duration of 400 ms was used. The radial energy is converted to axial energy when the ions leave the high magnetic field. Because of the large energy ratio, $\sim 10^6$, between the two radial motions, a significant reduction of the time of flight (TOF) to the ion detector is observed when the ions have been excited with their true cyclotron frequency. The detection of the resonance frequency is based on the time-of-flight technique [9].

The comparison between the cyclotron frequency of an unknown species with the frequency of a well-known reference ion will allow a precise mass determination via the following relation

$$m_{\mathrm{meas}} = (m_{\mathrm{ref}} - m_e)\frac{\omega_{c,\mathrm{meas}}}{\omega_{c,\mathrm{ref}}} + m_e. \qquad (2)$$

Figure 2 illustrates TOF spectra obtained for the reference ion ^{97}Zr and for the most neutron-rich isotopes measured, ^{100}Sr, ^{105}Zr, and ^{110}Mo.

During a week long measurement campaign, the masses of $^{98-105}$Zr, $^{95-100}$Sr, and $^{102-110}$Mo were measured. In all cases the reference frequency was obtained from ^{97}Zr [mass excess $ME = (-82946.6 \pm 2.8)$ keV, [3]] produced on-line in the fission reaction. For each frequency the time of flight was calculated from the average of the distribution. The resulting average time of flight versus frequency distribution was then fitted to obtain the true cyclotron frequency ω_c of the measured ion.

An empirical estimate for the statistical uncertainty did not exceed a few times 10^{-8} for each measurement. Therefore, the largest contribution to the total uncertainty originates from systematic effects. The uncertainty due to magnetic field fluctuations was deduced using all measurements of ^{97}Zr performed during the run. From this data set, the fluctuation of the cyclotron frequency due to magnetic field variations resulted in an uncertainty of about 4×10^{-8} of the frequency ratio $x = \frac{\omega_{c,\mathrm{ref}}}{\omega_{c,\mathrm{meas}}}$, corresponding to about ± 4 keV in this mass region.

Another possible source of error arising from the mass dependence was estimated from an off-line measurement by comparing the frequencies measured for ^{132}Xe and O_2 ions. The uncertainty was found to be $\frac{x_{\mathrm{exp}} - x_{\mathrm{AME}}}{x_{\mathrm{AME}}} = 7 \times 10^{-10}(m - m_{\mathrm{ref}})$, with x_{AME} being the frequency ratio calculated using the tabulated values from Ref. [3].

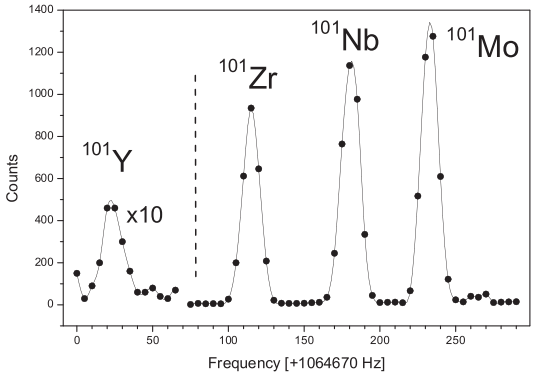

FIG. 1. Isobaric purification scan for fission products at mass $A = 101$.

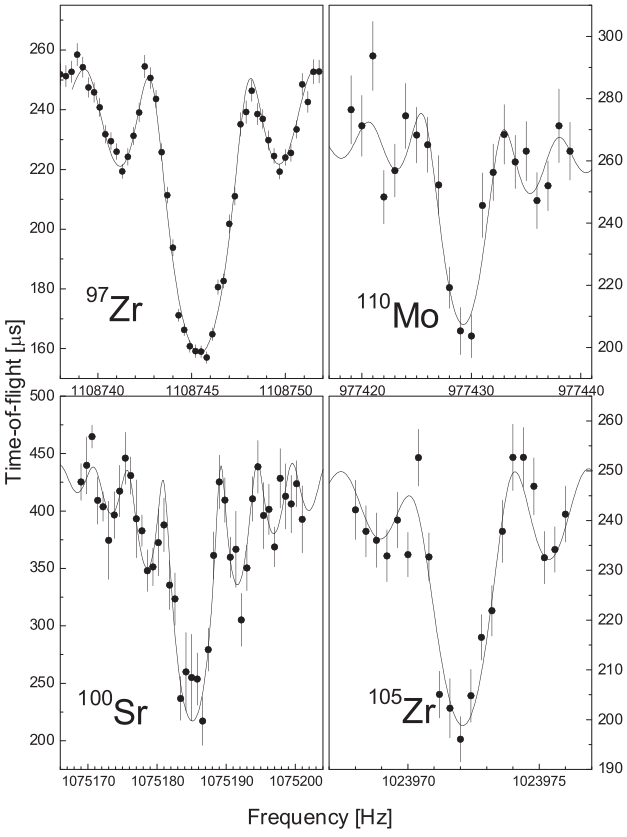

FIG. 2. TOF resonances measured for the reference ion ^{97}Zr and the most exotic isotope of each isotopic chain studied in this work. The modest asymmetries observed, and well reproduced in the fit, are due to a residual reduced cyclotron motion resulting from the incomplete radial cooling in the purification trap.

A large number of ions in the precision trap can cause the measured cyclotron frequency to shift due to ion-ion interactions in the trap. In order to reduce this effect, the count rate was kept below 20 ions per bunch after purification. When necessary, the beam intensity was reduced by inserting slits of variable opening before the RFQ. The count rate effect was carefully examined using ^{58}Ni ions. For the count rates used in this experiment, a maximum uncertainty of ± 0.1 Hz on the measured frequency was deduced, corresponding to 9×10^{-8} in the frequency ratio x.

The obtained frequency ratios x and the corresponding mass excess values are given in Table I. The uncertainty of the reference ion ^{97}Zr does not affect the uncertainty of the frequency ratio x, but only that of the resulting mass excess. For Zr isotopes the presented results up to ^{104}Zr agree reasonably well with our previous experiment that only employed the less accurate purification trap [2]. It is worth noticing that the mass excess of (-75123 ± 10) keV obtained for ^{95}Sr agrees well with the value of (-75117 ± 7) keV given in AME2003. This value in turn is primarily based on the high-accuracy measurement at ISOLTRAP [10].

The data in Table I represent the first direct mass measurement of these refractory-element isotopes. The experimental values for ^{104,105}Zr, as well as ^{109,110}Mo, are obtained for the first time. Published data with similar precision are available only for a very few other mass chains of neutron-rich nuclei. These are Rb and Sr isotopes studied at the ISOLTRAP facility of CERN [10]. Figure 3 displays the difference between the AME2003 values [3] and this experiment for all studied isotopes. With the exception of ^{99}Sr, the data show that neutron-rich Sr, Zr, and Mo nuclei are significantly less bound than the tabulated values in AME2003. The experimental values for the most neutron-rich isotopes studied differ by as much as 1 MeV from the extrapolated values in Ref. [3].

A detailed comparison of the new experimental values with the predictions of different mass models will be done in a forthcoming paper. For a recent evaluation, see Ref. [11]. Using the new data, it is instructive to study the evolution of the two-neutron separation energy as a function of the neutron number. The experimental data (Fig. 4) indicate a dramatic change in behavior for Sr and Zr isotopes between $N = 58$ and 60 which coincides with the known change of structure (deformation). The same effect is still observable in the Mo isotope chain, thanks to the current precise data. The very smooth, almost linear

TABLE I. Obtained frequency ratios x with statistical and systematic uncertainties and resulting mass excess values ME in keV. The systematic error is obtained as the quadratic sum of the count rate and the magnetic field related uncertainties.

Isotope	$x = \frac{\omega_{c,\mathrm{ref}}}{\omega_{c,\mathrm{meas}}}$	ME [keV]
^{102}Mo	$1.0515\,871\,4 \pm (2.2 \pm 9.9) \times 10^{-8}$	$-83\,570 \pm 10$
^{103}Mo	$1.061\,934\,81 \pm (2.3 \pm 9.9) \times 10^{-8}$	$-80\,965 \pm 10$
^{104}Mo	$1.072\,260\,38 \pm (1.7 \pm 9.9) \times 10^{-8}$	$-80\,354 \pm 10$
^{105}Mo	$1.082\,612\,57 \pm (1.9 \pm 9.9) \times 10^{-8}$	$-77\,341 \pm 10$
^{106}Mo	$1.092\,944\,70 \pm (2.0 \pm 9.9) \times 10^{-8}$	$-76\,139 \pm 10$
^{107}Mo	$1.103\,303\,20 \pm (2.3 \pm 9.9) \times 10^{-8}$	$-72\,556 \pm 10$
^{108}Mo	$1.113\,641\,90 \pm (2.4 \pm 9.9) \times 10^{-8}$	$-70\,760 \pm 10$
^{109}Mo	$1.124\,006\,02 \pm (7.5 \pm 9.9) \times 10^{-8}$	$-66\,670 \pm 12$
^{110}Mo	$1.134\,348\,35 \pm (24.9 \pm 9.9) \times 10^{-8}$	$-64\,547 \pm 24$
^{98}Zr	$1.010\,337\,15 \pm (2.9 \pm 9.9) \times 10^{-8}$	$-81\,291 \pm 10$
^{99}Zr	$1.020\,696\,54 \pm (10.6 \pm 9.9) \times 10^{-8}$	$-77\,627 \pm 13$
^{100}Zr	$1.031\,029\,22 \pm (3.0 \pm 9.9) \times 10^{-8}$	$-76\,376 \pm 10$
^{101}Zr	$1.041\,383\,61 \pm (3.3 \pm 9.9) \times 10^{-8}$	$-73\,164 \pm 10$
^{102}Zr	$1.051\,719\,85 \pm (2.1 \pm 9.9) \times 10^{-8}$	$-71\,590 \pm 10$
^{103}Zr	$1.062\,080\,43 \pm (2.5 \pm 9.9) \times 10^{-8}$	$-67\,819 \pm 10$
^{104}Zr	$1.072\,422\,41 \pm (4.3 \pm 9.9) \times 10^{-8}$	$-65\,728 \pm 10$
^{105}Zr	$1.082\,788\,40 \pm (9.0 \pm 9.9) \times 10^{-8}$	$-61\,469 \pm 12$
^{95}Sr	$0.979\,449\,05 \pm (3.1 \pm 9.9) \times 10^{-8}$	$-75\,123 \pm 10$
^{96}Sr	$0.989\,792\,20 \pm (3.0 \pm 9.9) \times 10^{-8}$	$-72\,926 \pm 10$
^{97}Sr	$1.000\,159\,07 \pm (2.9 \pm 9.9) \times 10^{-8}$	$-68\,587 \pm 10$
^{98}Sr	$1.010\,501\,76 \pm (2.9 \pm 9.9) \times 10^{-8}$	$-66\,431 \pm 10$
^{99}Sr	$1.020\,863\,85 \pm (2.2 \pm 7.3) \times 10^{-8}$	$-62\,524 \pm 7$
^{100}Sr	$1.031\,212\,53 \pm (4.5 \pm 9.3) \times 10^{-8}$	$-59\,828 \pm 10$

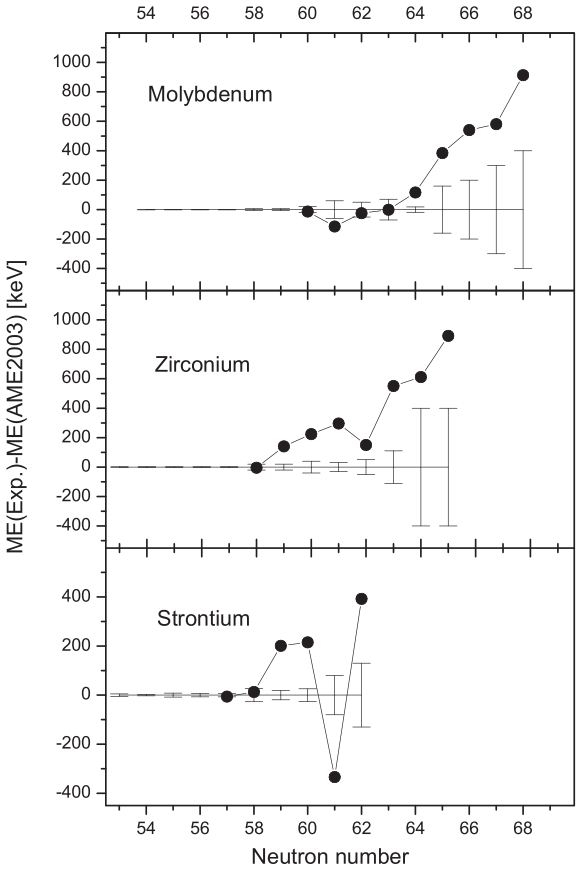

FIG. 3. Comparison of measured mass excess (ME) to AME2003 [3]. Error bars in the reference line correspond to AME2003 errors. Uncertainties in the experimental values are of the order of the size of the symbol; see Table I.

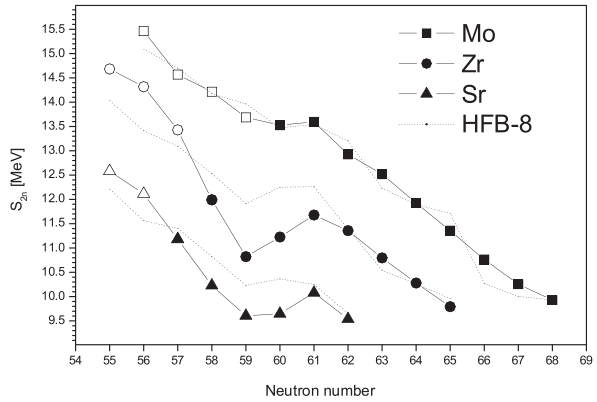

FIG. 4. Two-neutron separation energies. Filled symbols are from this measurement and open symbols are taken from AME2003 [3].

behavior of the experimental curves beyond $N = 60$ indicates stability in the structure of these nuclei. This is in accordance with the prediction in the liquid drop model context, where this behavior is produced by the asymmetry term only [12,13].

We compare our results with a recent successfully tested microscopic model, the Hartree-Fock-Bogoliubov (HFB) plus particle-number projection mass model [Ref. [14]]. The newest parameter set of this model (termed HFB-8) is determined by optimizing the fit to the full data set of the 2149 measured masses with $N, Z \geq 8$ (both spherical and deformed) of Ref. [3]. The corresponding root mean square error is 0.635 MeV for this data set. For the Sr, Zr, and Mo nuclei studied in this work the model produces an excellent agreement for isotopes with neutron numbers $N > 60$. Below this value, only Mo isotope masses agree well with the model, but Sr and Zr isotopes near the semimagic $Z = 40$, $N = 56$ are not in agreement with the data. In most models, above $N = 60$ these nuclei become prolate and well deformed. For Sr and Zr nuclei this is also confirmed by the experiments [15,16]. However, in the HFB-8 model at $N = 66$ and 68, Mo isotopes are already predicted to be oblate. This seems to lead to an underestimation of the two-neutron binding energy.

Any major change in nuclear structure would introduce a deviation from a smooth trend in two-neutron separation energies [12]. Therefore, it will be important to study these nuclei in another theoretical context which at the same time treats both excited structures as well as binding energies. This allows inclusion of mixing of different structures and intruder states. Such a development is underway for the Sr, Zr, and Mo isotopes employing the IBM approach [17].

In summary, precise data on the masses of 23 neutron-rich Sr, Zr, and Mo isotopes have been presented for the first time. A significant deviation has been observed in comparison with the recent mass evaluation. Taking into account the yield distribution of proton-induced fission and the universality of the IGISOL technique, more than 200 atomic masses of neutron-rich nuclei between Ni and Ce can be measured in the near future with a relative accuracy $\leq 10^{-7}$. This will provide an important contribution to the theoretical development of mass models and a pivotal role in providing data for *r*-process calculations. It will be particularly challenging to extend these experiments towards the neutron number $N = 70$ at which major changes in the nuclear structure are predicted to occur [18,19].

This work was has been supported by the European Union within the NIPNET RTD project under Contract No. HPRI-CT-2001-50034 and by the Academy of Finland under the Finnish Center of Excellence Programme 2000–2005 (Project No. 44875). A. J. is indebted to financial support from the Academy of Finland (Project No. 46351).

*Electronic address: ari.jokinen@phys.jyu.fi

[1] J. Äystö, Nucl. Phys. **A693**, 477 (2001).
[2] S. Rinta-Antila, S. Kopecky, V. S. Kolhinen, J. Hakala, J. Huikari, A. Jokinen, A. Nieminen, and J. Äystö, Phys. Rev. C **70**, 011301 (2004).
[3] G. Audi, O. Bersillon, J. Blachot, and A. H. Wapstra, Nucl. Phys. **A729**, 3 (2003).
[4] A. Nieminen, J. Huikari, A. Jokinen, J. Äystö, P. Campbell, and E. C. A. Cochrane, Nucl. Instrum. Methods Phys. Res., Sect. A **469**, 244 (2001).
[5] L. Brown and G. Gabrielse, Rev. Mod. Phys. **58**, 233 (1986).
[6] V. S. Kolhinen, T. Eronen, J. Hakala, A. Jokinen, S. Kopecky, S. Rinta-Antila, J. Szerypo, and J. Äystö, Nucl. Instrum. Methods Phys. Res., Sect. B **204**, 502 (2003).
[7] V. S. Kolhinen *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **528**, 776 (2004).
[8] K. Blaum, G. Bollen, F. Herfurth, A. Kellerbauer, H.-J. Kluge, M. Kuckein, S. Heinz, P. Schmidt, and L. Schweikhard, J. Phys. B **36**, 921 (2003).
[9] M. König, G. Bollen, H.-J. Kluge, T. Otto, and J. Szerypo, Int. J. Mass Spectrom. Ion Processes **142**, 95 (1995).
[10] H. Raimbault-Hartmann *et al.*, Nucl. Phys. **A706**, 3 (2002).
[11] D. Lunney, J. Pearson, and C. Thibault, Rev. Mod. Phys. **75**, 1021 (2003).
[12] R. Fossion, C. Coster, J. Garcia-Ramos, T. Werner, and K. Heyde, Nucl. Phys. **A697**, 702 (2002).
[13] P. Möller, J. Nix, W. Myers, and W. Swiatecki, At. Data Nucl. Data Tables **59**, 185 (1995).
[14] M. Samyn, S. Goriely, M. Bender, and J. Pearson, Phys. Rev. C (to be published).
[15] W. Urban *et al.*, Nucl. Phys. **A689**, 605 (2001).
[16] P. Campbell *et al.*, Phys. Rev. Lett. **89**, 082501 (2002).
[17] J. Garcia-Ramos, K. Heyde, R. Fossion, V. Hellemans, and S. D. Baerdemacker, Eur. Phys. J. A **26**, 221 (2005).
[18] N. Schunck, J. Dudek, A. Gozdz, and P. Regan, Phys. Rev. C **69**, 061305(R) (2004).
[19] F. Xu, P. Walker, and R. Wyss, Phys. Rev. C **65**, 021303(R) (2002).